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Final Report
Planetary Instrument Definition and Design Program
(PIDDP) Support Project

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Abstract

The results of Sandia National Laboratories' participation in the NASA Planetary Definition and Design Program are summarized. Areas reported include the characterization of large area cadmium zinc telluride spectrometers and the application of simulation techniques to the prediction of device performance. Also investigated was the response of mercuric iodide devices in the region from 1 to 100 KeV. A literature study to determine the status of radiation damage measurements in room temperature semiconductor devices is also reported.

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Final Report
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INTRODUCTION

Wide band gap semiconductor detectors capable of room temperature operation offer distinct advantages over conventional scintillator-photocell and cryogenic semiconductor based systems. They are substantially lighter than either scintillator or cryogenic systems, offer energy resolution significantly better than scintillators (although inferior to cryogenic systems) and unlike cryogenic systems are maintenance free. The most advanced of the wide band gap materials are cadmium zinc telluride, mercuric iodide, and cadmium telluride. While in their present stage of development they are suitable for many astrophysics applications, further development, particularly relative to detector size, is required before the technology can be fully exploited for space science.

As a participant in the Planetary Instrument Definition and Design Program (PIDDP), Sandia National Laboratories was tasked to investigate the technical issues currently limiting the application of these devices in space applications. Although areas of potential astrophysical application are numerous, this PIDDP project was concerned primarily with applications involving light-weight, low- power, gamma ray spectrometers for energies up to 10 MeV as well as high resolution x-ray spectrometers for the region below 10 KeV, and the region of K-alpha x-rays from the heavy elements.

Specifically, Sandia tasking involved the evaluation of large volume cadmium zinc telluride (CZT) gamma -ray spectrometers and the computer simulation of detector response based on currently available transport data (using Sandia developed response function codes). Sandia was also tasked to fabricate and evaluate a small volume mercuric iodide (HgI₂) x-ray detector. Further tasking included a survey of the status of radiation damage measurements in the leading room temperature semiconductor materials and the preparation a report.

The present report summarizes Sandia activities in support of the PIDDP during the current contract period. Details of these activities are contained in Appendix A where copies of pertinent Sandia publications are attached.

DETECTOR STUDIES

Cadmium Zinc Telluride

A 1.8x 1.8 x .5 cm (1.62 CM³) CZT spectrometer was fabricated from high resistivity material. Spectrometer performance was evaluated using both digital rise time and dual time constant sampling to mitigate charge collection problems (Lund et al. 1996). Using rise time compensation 1.7% (FWHM) resolution was obtained at 662 KeV. (see Figure 1) While slightly poorer results were obtained using dual time constant sampling, the peaks were more symmetric. In addition, the dual time constant method can be implemented with commercially available nuclear electronics. Development of additional single charge collection procedures were continued in an effort to more efficiently deal with hole trapping. Included was the development of a novel electrode configuration (Lee et al. 1998) which simulates the action of a Frisch grid without the electronic complexity (and noise) associated with coplanar Frisch grids being developed elsewhere (Luke 1994) (He et al. 1996). Energy resolution of 1.4 % at 662 KeV was obtained with 10x10x2 mm³ detector. Figure (2).

As a means of achieving increased detector volume, coaxial detectors were fabricated and tested as alternatives to the conventional planar designs (Lund et al. 1996). The coaxial devices produced full energy peaks from Cs -137 and barium -133 although the performance was limited by poor hole transport.

Efforts were continued to better understand factors affecting carrier mobility and lifetime which currently limit the performance of large volume CZT devices. Material properties which affect carrier transport include crystallographic defects (inclusions, cracks, pipes, etc), impurities and chemical inhomogenities. Techniques employed for their study included infrared transmission microscopy (ITM), particle induced x-ray emission (PIXE), tri-axial x-ray diffraction (TAXRD), photoluminescence (PL) ,and mass spectroscopy (ICPMS).

Mercuric Iodide

A number of mercuric iodide x-ray spectrometers with areas up to 25 square millimeters were fabricated from high resistivity material grown by the vapor transport method (Van Scyoc et al. 1997). Spectrometers of this size yielded energy resolution of 1% at 59.5 KeV (599 eV FWHM) without cooling either the device or the associated preamplifier input stage. With modest cooling (provided by a thermoelectric device) exceptional performance was achieved in the low energy x-ray region. As illustrated in Figure 3, the K- alpha x-ray of potassium at 3.31 KeV was detected with a resolution of 8% (261 eV FWHM), well resolved from the K-alpha x-ray from chlorine at 2.62 KeV. The lack of system noise down to 1 KeV evident in that figure indicates that the K-alpha x-ray from sodium at 1.04 KeV as well as the 1.25 KeV K-alpha x-ray from magnesium should be readily detected.

Detectors of this type were used in the construction of an active, portable x-ray fluorescence analysis instrument. Stimulation of the fluorescence was provided by three isotopic sources: Cd-109 , Fe-55, and Am -241. Access to elements sodium (Z=11) through uranium (Z=92) was demonstrated in field test of the instrument.

Simulations

Sandia has been actively involved in the development of simulation techniques to predict the performance of semiconductor detectors over a range of carrier transport properties and under various incident radiations. These techniques have been reduced to a code that can be executed on a desktop computer. As input parameters the code uses carrier mobilities, lifetimes, electric field conditions, gamma ray absorption coefficients, and detector dimensions to produce an ideal (noise free response) spectral response. This ideal response is then convoluted with random noise generated by the detector and electronics to yield results consistently in good agreement with experimental data.

Using these codes Sandia investigated the performance of numerous detector configurations (Lee et al. 1998). An example of the results are shown in Figure 2. These calculations are of particular value in both system design where performance can be examined from the vantage point of transport properties and device geometries. Simulations are also of value in material science where the efficacy of various research strategies can be assessed, thereby avoiding certain trial and error laboratory investigations.

RADIATION DAMAGE

An extensive literature search was undertaken to determine the status of radiation damage measurements on room temperature semiconductor detectors. The survey was carried out using the computer databases INSPEC, CALPLUS, and COMPENDEX, together with DOE archives. In all, more than 300 abstracts were reviewed. The survey was confined to the materials cadmium zinc telluride, cadmium telluride and mercuric iodide.

Although the response of these materials to ionizing radiation is far from complete a number of significant features have emerged. Cadmium zinc telluride shows resolution degradation beginning at about 10^8 p/cm² from 200 MeV protons but no degradation from moderated fission spectrum neutrons at fluences up to 10^{10} n/cm² although activation is evident under those conditions. Similar results were reported for cadmium telluride although proton exposures were limited to intermediate energies (33 MeV). Over the exposure region reported to date (up to 10^{12} p/cm² intermediate energy protons and 1.2×10^8 p/cm² 1.5 GeV protons), mercuric iodide appears to be free of resolution degradation. We note that these data are based on a very limited sampling, often from a single detector. In addition, the fluence range frequently does not cover that expected for many space missions and rate effects have yet to be investigated. The results of this investigation were reported in SAND98-8237 (Franks et al. 1998) and in the open literature (Franks et al. 1999).

Figure 1: Pulse height spectrum obtained by irradiating a 1.8 cm x 1.8 cm x 0.5 cm $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ detector with a ^{137}Cs source and applying pulse height compensation. The width of the 662 keV photopeak is 1.7% FWHM. Approximately 30% of the volume of the detector was sampled in the experiment.

Figure 2: The simulated pulse height spectrum (solid line) and the experimental data (circles) for the unipolar γ -ray detector taken from the anode with cathode triggering. The energy resolution is excellent ($\sim 1.4\%$ FWHM). For the simulation of the pulse height spectrum, finer binning was done to match the experimental pulse height channel bin size, and the overall count per minute (cpm) was scaled to the experimental photoelectric peak.

Figure 3: Spectrum of a KCl salt sample, taken with a 5 x 5 x 0.5 mm³ HgI₂ detector.

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DEVICE SIMULATION OF AN UNIPOLAR GAMMA-RAY DETECTOR

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ABSTRACT

The pulse height spectra from a new kind of unipolar gamma-ray detectors were predicted using a new three-dimensional simulation program developed at Sandia National Laboratories. The detectors were fabricated at Sandia and RMD Inc., and tested at Sandia. They were fabricated from $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ crystals and they were electron-transport-only devices. For the simulation, a successive overrelaxation method was used to determine the three-dimensional internal electric field within a detector, and to find the weighting potentials for the anode and the cathode. Uniform irradiation and ionization from a ^{137}Cs source was assumed, and the charge transport and the signal induction within the detector were numerically computed using the appropriate materials and design parameters. The simulation gave excellent agreement with experimental pulse height spectra, and it demonstrated the power of such a simulation to correlate the materials parameters and the device design to the actual detector performance.

INTRODUCTION

Room temperature gamma-ray detectors are rapidly gaining wide-spread use, due to their superior energy resolution and portability [1]. Recent advances have taken place both in their materials properties and design. One of the most exciting advances in this field is the advent of the unipolar devices [2-3]. Due to the larger mobilities, longer lifetimes, and smaller effective masses of electrons over holes, all of such unipolar devices are electron-transport-only devices, and these terms are used interchangeably. In these devices, the signal is determined by the motion of the electrons only, resulting in superior energy resolution over the existing planar detectors. However, due to the relatively complex three-dimensional (3-D) structures of these devices, detailed understanding of their behavior remains illusive. Analytical simulation of such a device is, in general, difficult to do properly, except in simplest cases where a high degree of symmetry exists.

In this manuscript, a 3-D numerical simulation of a new kind of unipolar device is described. Pulse height spectra were simulated using realistic materials parameters and device dimensions, and the simulated pulse height spectra are in excellent agreement with the experiment. The unipolar devices were fabricated at Sandia National Laboratories, California, and at RMD Inc. of Watertown, Massachusetts, and their design is schematically depicted in Fig. 1. (Ref. 4) On a parallelepiped CdZnTe (CZT) crystal, the cathode and the anode are placed at the opposite ends, as is usual for a conventional planar detector. However, in this unipolar device, a Au grid is evaporated at some short distance away from the anode, all the way around the detector, and it is grounded. The grid functions to shield the electric field of the cathode from the anode, while allowing the carrier transport of the electrons across the grid toward the anode. A detailed discussion of other unipolar devices can be found in Ref. 3 and other references therein. A further description of Sandia's unipolar device will be given elsewhere [5].

In this paper, a model of carrier transport and electrical signal generation within an unipolar device will be presented, followed by a description of the computational method used to predict the detector performance. Next, simulations for various grid positions will be discussed, relying on charge induction maps for the anode and the cathode to relate the features in the pulse height spectra to specific design parameters of the unipolar device. Then a comparison of an experimental pulse height spectrum from a unipolar device to a simulation will be made. Lastly, the effect of the electron lifetime on the simulated pulse height spectrum shape will be discussed.

Fig. 1. An unipolar γ -ray detector design. $+V_1$ is the dc anode bias and $-V_2$ is the dc cathode bias. The signal is measured from the anode in the single pulse mode, and, optionally, the cathode signal is used as a trigger for the anode signal, to improve the detector energy resolution.

MODEL OF CHARGE CARRIER TRANSPORT AND SIGNAL GENERATION IN A SEMICONDUCTOR RADIATION DETECTOR

When ionizing radiation interacts within a semiconductor radiation detector, it gives rise to an equal number of free electrons and holes near the point of interaction, and the electrons and the holes start to move under the influence of the electric field within the detector. If $Q_e(0)$ electrons are created at time $t=0$ and position $\mathbf{r} = \mathbf{r}_0$, the trapping of the charge carriers causes an exponential decay of the free charge carriers.

$$Q_e(t) = Q_e(0) \exp(-t/\tau_e), \quad (1)$$

where τ_e is the lifetime of the electrons. A similar expression can be written for the holes. In the following equations, we will omit the subscript e for the electrons, keeping in mind that the equations apply for both the electrons and the holes.

The motion of the free carriers can be characterized by their velocity, $\mathbf{v}$. In the typical low field condition, the velocity is a linear function of the electric field.

$$\mathbf{v}(\mathbf{r}(\mathbf{r}_0, t)) = \mu \mathbf{E}(\mathbf{r}(\mathbf{r}_0, t)), \quad (2)$$

where μ is the mobility and $\mathbf{E}(\mathbf{r}(\mathbf{r}_0, t))$ is the electrostatic field along the trajectory of the charges at the position $\mathbf{r}(\mathbf{r}_0, t)$ and time t . During their motion, the carriers experience an electrostatic force $\mathbf{F}$, given by

$$\mathbf{F}(\mathbf{r}(\mathbf{r}_0, t)) = Q(t) \mathbf{E}(\mathbf{r}(\mathbf{r}_0, t)). \quad (3)$$

Based on the principle of superposition, each electrode exerts a force that, summed together with the forces by all the other electrodes, gives the total force on the charge carriers. The force that the i th electrode exerts is

$$\mathbf{F}_i(\mathbf{r}(\mathbf{r}_0, t)) = Q(t) \mathbf{E}_i(\mathbf{r}(\mathbf{r}_0, t)), \quad (4)$$

where $\mathbf{E}_i(\mathbf{r}(\mathbf{r}_0, t))$ is the electric field in the configuration where the i th electrode is set at the biasing voltage, V_i , but all the other electrodes are grounded. It is convenient to introduce

$$\mathbf{E}_i^w(\mathbf{r}(\mathbf{r}_0, t)) = \mathbf{E}_i(\mathbf{r}(\mathbf{r}_0, t)) / V_i. \quad (5)$$

In this case, $\mathbf{E}_i^w(\mathbf{r})$ is the electric field when the i th electrode is set at 1 Volt and all the other electrodes are grounded. This electric field is called the weighting field, and the potential field associated with it is called the weighting potential [3]. The power expended by the moving particle

on the i th electrode is $F_i \cdot v$, and this must equal the power induced on the external circuit, $i \times V_i$, where i is the current. Typically, V_i is kept constant, and hence

$$i(r_0, t) = \mu Q(t) E_i^w(r(r_0, t)) \cdot E(r(r_0, t)). \quad (6)$$

The function that is the total charge, $q_i(r_0)$ induced on the i th electrode by a radiation quantum interacting at the position r_0 is called the charge induction map, and it is

$$q_i(r_0) = \int_0^T i(r_0, t) dt, \quad (7)$$

where T is the integration time of the measurement. In typical nuclear electronics, T is approximately equal to the shaping time of the spectroscopy amplifier. Equations (1) and (2) can be used to simulate the charge carrier transport, and Equations (5), (6), and (7) can be used to calculate the charge induced on each electrode. The pulse height spectrum for the detector can be obtained from the charge induction map. In a multichannel analyzer, the counts in the channel collecting charges between q_j and $q_j + \Delta q$ are determined by the equation

$$N_j = \int q(r_0) \vartheta(q(r_0) - q_j) \vartheta(q_j + \Delta q - q(r_0)) dr_0, \quad (8)$$

where ϑ is the Heaviside step function, and the integration is over the entire detector volume.

When combined with the solutions for the weighting fields and the total electric field, these equations allow for the simulation of the pulse height spectrum produced by the detector. A simulated pulse height spectrum can be directly compared to the experimental spectrum, and the charge induction map and the weighting fields can give insights into the operation of the device, by giving information on the spatially resolved response of the device to incident ionizing radiation.

The simulation can be applied to conventional planar detectors, but its real power is in simulating multiple electrode 3-D geometries, such as an unipolar device, that can not be readily treated by analytical models. In an ideal unipolar device, the charge induction map for the anode would be relatively large and uniform between the grid and the cathode, and it would be zero between the grid and the anode.

COMPUTATIONAL METHOD

The detector simulation divides naturally into two parts. The first part is the determination of the electrostatic field within the detector, and the second part is the calculation of the charge transport within the detector, along with the corresponding signal induced on the electrodes. Except in the simplest approximations, both parts require detailed 3-D numerical computations.

For our simulation, the electrostatic field within the detector was solved by applying a finite-element method called successive overrelaxation (SOR) to the Poisson equation [6]. In this method, the field is first discretized and a trial potential field is selected, then SOR is applied to the lattice space. Successive iterations result in a lowering of the total electrostatic energy of the field and convergence of the field. We defined a lattice space of $120 \times 120 \times 40$ and set the lattice spacing at 0.1 mm. Typically, 1000 iterations gave 1 part in 10^5 convergence of both the total electrostatic energy and the potential field at all points of the lattice. Periodic boundary conditions were assumed, and the detector was placed inside the boundary. For the results to be discussed here, $10 \times 10 \times 2$ mm³ detectors with various electrode geometries and voltages were placed 1 mm away from all the border faces.

The charge transport was numerically computed using Equations (1) and (2), and the corresponding charge induction maps were also numerically calculated using Equations (5) through (9). The transport parameters for the simulation were typical for commercially available CZT [7]. The CZT electron and hole mobilities are taken to be 1000 and 100 cm²/V/sec, respectively, and the electron and hole lifetimes to be 3 and 1 μ sec, respectively. The dielectric constant of the detector was assumed to be 10 (cgs units) and that for the surrounding medium was

assumed to be 1, i.e. the value for air. For the simulation of the pulse height spectra, a shaping time of 1 μ sec and uniform irradiation and ionization by 662 keV γ -rays from ^{137}Cs was assumed.

COMPUTATIONAL RESULTS

For the detector schematically shown in Fig.1, one of the important design parameters is the grid-to-anode spacing, Δs . One should minimize Δs to obtain the largest active detector volume, while allowing minimal sacrifice to the detector resolution. Four grid spacings of 1, 3, 5, and 7 mm from the cathode were modeled. In Fig. 2, the weighting-potential fields for the 3 mm case are shown. The anode, grid, and cathode biases are 275, 0, -1000 V, respectively. The weighting potentials show that the grid shields the anode and the cathode from each other, but there are distortions near the grid. Note that the equipotential lines push through the center region at the plane of the grid. This effect becomes more pronounced as Δs becomes smaller.

For the same boundary conditions as Fig. 2, Fig. 3 (a) and (b) show the charge induction maps for the anode, resolved of the electron and the hole contributions. The charge induction map for the electron can be seen to be fairly uniform between the cathode and the grid, characteristic of an unipolar device. However, near the grid, distortions can be seen, and $q(r_0)$ drops to zero nearly linearly between the grid and the anode. The charge induction map for the hole is nearly zero between the grid and the cathode, and it decreases nonlinearly from the anode to the grid, due to hole trapping.

Fig. 2(a) A contour map of the weighting potential for the anode, examined in the plane in the middle of the $10 \times 10 \times 2 \text{ mm}^3$ CZT detector. Each pair of contour lines separates 25 mV, the anode being at 1 Volt and the other electrodes being grounded. The potential is depicted in the lattice space, where 1 lattice unit is 0.1 mm. The grid is located 3 mm away from the anode and it is 0.1 mm thick. (b) The weighting potential for the cathode. Each contour line separates 25 mV.

Fig. 3(a) The charge induction map on the cathode due to the electron transport only, examined in the plane in the middle of the detector. (b) The charge induction map on the anode due to the hole transport only, scaled up for ease of viewing. The absolute magnitude of the charge induced by the holes is much smaller than for the electrons.

Recalling that $q(r_0)$ is the charge induced at an electrode, a detailed examination of the induction maps shown in Fig.3 indicates that, even though $q(r_0)$ for the anode is fairly uniform between the grid and the cathode, it has a local maximum near the grid, due to the shorter distance to the anode and hence shorter transit time. Because of electron trapping, shorter transit times mean larger signals.

Some simulated pulse height spectra for various grid-to-anode spacings, Δs , are shown in Fig. 4. For $\Delta s = 1$ and 3 mm, the electron peaks are seen to have sharp main peaks surrounded by broader tails. It was found by detailed examination of the charge induction maps that the sharp main peaks are due to electrons originating from the region near the plane of the grid. More specifically, for $\Delta s = 1$ mm, the signal originates mostly from the region between ~ 2 and 4 mm away from the grid, on the cathode side of the detector. This region is relatively far away from the grid, because the close proximity of the grid to the anode causes the anode weighting potential to extend significantly beyond the grid plane. For $\Delta s = 3$ mm, it originates from the region between ~ 1 and 3 mm away from the grid, on the cathode side.

The broader tails in both the $\Delta s = 1$ and 3 mm cases originate from the regions near the grid and near the cathode. The larger inhomogeneity of the electric field near the grid causes the 1 mm case to have a much broader tail.

In the 5 mm case, a qualitatively different shape is seen. The inhomogeneity in the field that gave the sharp feature is no longer there, and this results in a relatively uniform charge induction map between the grid and the cathode. However, this is at the price of detection efficiency and decreased charge collection. The count under the electron peak in this case is only 24% compared to the 1 mm spacing case. Note that this is worse than one would expect based purely on detection volume considerations, which would predict a decrease to only 56%. The additional degradation is mostly due to increased electron trapping between the grid and the anode.

Fig. 4. The simulated pulse height spectra of the unipolar device described in the text with grid-to-anode spacings between 1 and 7 mm. The signal is taken at the anode. The peaks are due to photoelectric interaction of 662 keV γ -rays. A real spectrum would have additional features due to Compton scattering and electronic noise of the nuclear electronics.

In the 7 mm case, the electron peak broadens again, and the peak position moves to lower channels, both effects being mostly due to the electron trapping.

A comparison of the simulation and the experimental data is shown in Fig. 5. The experimental spectrum is discussed in detail in Ref. 5. For the experiment, a $10 \times 10 \times 2$ mm³ CdZnTe crystal was used. Two Au electrodes were evaporated on opposite 10×2 mm² faces as the cathode and the anode. The grid was 2 mm thick Au film located 1.5 mm away from the anode. The cathode was biased at -1301 V and the anode at +220 V. The anode signal was triggered off the cathode to select ionization events occurring between the grid and the cathode, to optimize the energy resolution.

Fig. 5. The simulated pulse height spectrum (solid line) and the experimental data (circles) for the unipolar γ -ray detector described in the text, taken from the anode with cathode triggering. The energy resolution is excellent, being $\sim 1.4\%$ FWHM. For the simulation of the pulse height spectrum, finer binning was done to match the experimental pulse height channel bin size, and the overall count per minute (cpm) was scaled to the experimental photoelectric peak.

The simulation was performed using the exact geometry, the typical CZT materials parameters, and the cathode triggering used in the experiment. It can be seen that the photoelectric peak is fit very well, even though there are no fitting parameters. The tail in the experimental spectrum may be lower due to a larger electron lifetime or a higher electron mobility.

To see the effect of the materials parameters on the pulse height spectrum, additional simulations were done. Fig. 6 shows a comparison of two simulated pulse height spectra, with the typical electron lifetimes of 3 μsec and a shorter lifetime of 1 μsec . Cathode triggering was not used in these cases. Lower lifetime resulted in increased tailing and broadening of the photoelectric peak. It is clear that, by varying the materials parameters, one can get an even better fit than shown in Fig. 5.

Fig. 6. The simulated pulse height spectra for electron lifetimes of 3 μsec (dashed line) and 1 μsec (solid line). The shorter lifetime results in a longer tail and degraded energy resolution. Cathode triggering was not included in these cases.

CONCLUSION

In conclusion, simulation of Sandia's unipolar CdZnTe γ -ray detectors were performed. The simulated pulse height spectra for the electron-transport-only devices were in very good agreement with the experimental results. Detectors with various grid-to-anode spacings were simulated and found to have very different pulse height spectra. In addition, the electron lifetime was varied and also found to have a significant effect on the pulse height spectra, demonstrating the importance of the device design and the materials properties on the performance of the electron-transport-only devices. In the future, simulations will be completed on other device structures, and detailed comparisons between the simulations and the experimental data will be used to optimize the device performance.

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Modeling and Simulation of Uniformity Effects in $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ Gamma-Ray Spectrometers

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Abstract—Monte Carlo simulations of pulse height spectra for $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ detectors are used to investigate the effect of variations in alloy composition and carrier drift lengths on energy resolution. The results, which are based on a simple phenomenological model, show that these nonuniformities can have significant detrimental effects on spectrometer performance. For the case of Bridgman-grown material, the orientation of the growth axis relative to the detector axis is shown to be an important consideration, especially for crystals which come from the beel end of a boule, where the composition gradient due to zinc segregation is large. Other effects which we have simulated include growth striations, zinc segregation at grain boundaries, and trapping by inclusions and grain boundaries; each of these effects is detrimental to energy resolution. We conclude that material nonuniformity is a major obstacle to achieving statistically limited energy resolution in cadmium zinc telluride detectors.

Index Terms—Cadmium zinc telluride, material nonuniformity, Monte Carlo simulation, radiation detectors.

I. INTRODUCTION

IN PREVIOUS papers we and others have demonstrated a link between material uniformity and detector performance in $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ nuclear spectrometers [1]–[4]. This work was strictly empirical, directly correlating uniformity measures based on material characterization techniques with energy resolution of pulse height spectra. In this paper we propose simple models to account for this correlation and use Monte Carlo simulations to obtain approximate, quantitative dependence of energy resolution on the degree of uniformity. Two types of nonuniformity are considered: variation in zinc fraction, x , and variation in the electron and hole drift lengths, $(\mu\tau)_e E$ and $(\mu\tau)_h E$, where μ and τ are the mobility and trapping lifetime, respectively, and E is the applied electric field.

II. BASIC MODELS FOR EFFECT OF UNIFORMITY ON ENERGY RESOLUTION

A. Variation in Composition

The simplest mechanism by which composition variation affects detector performance is through the electron-hole pair

production energy ϵ . It is established that ϵ , the average energy required to produce an electron-hole pair, varies linearly with bandgap E_g , although there is some disagreement about the precise form of the linear equation [5], [6]. The bandgap, in turn, varies quadratically with zinc fraction x , but again there are numerous published relations which differ regarding the "bowing parameter," the coefficient of the quadratic term in $E_g(x)$. In this paper we use the following relations:

$$E_g(x) = 1.5 + 0.5x + 0.2x^2 \text{ eV} \quad (1)$$

$$\epsilon = 2.7E_g + 0.43 \text{ eV} \quad (2)$$

We choose the former equation because it is consistent with our photoluminescence measurements [7] and the latter because it gives values that compare reasonably well with the known value of ϵ for CdTe and the estimated value for ZnTe [8].

For the case in which the drift lengths are taken to be uniform along the path of a given charge carrier, we calculate the charge collection as a function of interaction depth using the well-known Hecht relation [9]

$$Q(x_0) = \frac{qNE}{d} \left\{ (\mu\tau)_e \left\{ 1 - \exp \left[\frac{-(d-x_0)}{(\mu\tau)_e E} \right] \right\} + (\mu\tau)_h \left\{ 1 - \exp \left[\frac{-x_0}{(\mu\tau)_h E} \right] \right\} \right\} \quad (3)$$

where x_0 is the distance from the cathode at which the interaction occurs, d is the thickness of the detector, q is the magnitude of the electron charge, and N is the number of electron hole pairs created. The case in which a charge carrier experiences a nonuniform drift length will be discussed below. Equation (3) is strictly valid only for parallel-plate detectors when the shaping time is longer than the hole transit time, so in this paper we treat only that case. This equation also assumes that all charge from a single interaction is generated at a single point, an assumption which is somewhat weak at higher gamma-ray energies. For this reason we consider only energies of 1 MeV or less.

Using (1)–(3) one can perform a quick estimate of how much a given degree of composition variation affects detector resolution. Since Q is linearly proportional to N , it is clear that $\delta Q/Q = \delta N/N$ so that the fractional width of the photopeak is the same as the fractional variation in N . Fortunately, the fractional variation in N is generally much smaller than the fractional variation in x . Recalling that N is related to the zinc fraction by (1) and (2), we have for photoelectric absorption

$$N = h\nu/[2.7(1.5 + 0.5x + 0.2x^2) + 0.43 \text{ eV}] \quad (4)$$

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Fig. 1. Ratio of fractional variation in number of electron-hole pairs created (N) to fractional variation in zinc content (x) as a function of x . For commercially available material $x = 0.1$ or 0.2 so that the variation in N is an order of magnitude lower than the variation in x .

where h is Planck's constant and ν is the frequency of the incident photon. Differentiating (4) yields

$$\frac{\delta N}{N} = \frac{\delta x}{x} \left[\frac{-2.7x(0.5 + 0.4x)}{2.7(1.5 + 0.5x + 0.2x^2) + 0.43 \text{ eV}} \right]. \quad (5)$$

Since x is typically 0.1 or 0.2 in commercially available material, the factor of x in the numerator on the right-hand side of (5) is fortuitous—it means that for the widely used alloy compositions, the fractional variation in N is much less than the fractional variation in x . Fig. 1 shows a plot of $(\delta N/N)/(\delta x/x)$ versus x . Notice that as x becomes larger, a given degree of composition variation has a greater impact on detector resolution, but $\delta N/N$ is always less than $\delta x/x$. In the remainder of this paper we will use $x = 0.2$, for which $\delta N/N = 0.07\delta x/x$.

B. Variation in Transport Properties

The other form of nonuniformity which we consider is a variation in the electron and hole drift lengths, $(\mu\tau)_e E$ and $(\mu\tau)_h E$. Because the mobility, lifetime, and electric field appear as a product, it is impossible to separate the effects of the three parameters in the model.

In the case that the drift lengths vary only transversely to the detector axis, a reasonable approximation for thin detectors (3) can again be used as the basis for modeling the effect. In this case it is slightly more difficult to see intuitively how much a given degree of variation in $\mu\tau E$ affects the energy resolution, because $\mu\tau E$ appears both as a multiplying factor and in the exponent. Differentiating (3) with respect to $(\mu\tau)_e E$ and ignoring the hole contribution gives

$$\frac{\delta Q}{Q} = \frac{\delta(\mu\tau)_e E}{\mu\tau E} \left[1 - \frac{d}{\mu\tau E} \frac{1}{(e^{d/(\mu\tau E)} - 1)} \right] \quad (6)$$

where the subscript e has been omitted. Equation (6) states that a given degree of variation in $\mu\tau E$ transverse to the detector axis affects energy resolution somewhat less than the same degree of variation in N , the effect increasing as the ratio

Fig. 2. Ratio of fractional variation in charge collection (Q) to fractional variation in electron drift length $[(\mu\tau)_e E]$ as a function of detector thickness (d) divided by drift length. The better the electron transport in the material, the less spatial variations in transport matter. This curve applies only to variations in $(\mu\tau)_e E$ perpendicular to the detector axis.

Fig. 3. Simulated pulse height spectrum for 300-KeV radiation incident on a 1.0×0.5 cm $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ detector with $(\mu\tau)_e E = 5$ cm and $(\mu\tau)_h E = 0.1$ cm. The channel number has no direct meaning; it is simply a convenient way of scaling the axis for plotting. The discontinuity around channel 200 is caused by the lower cutoff of charge collection for photoelectric absorption.

$d/\mu\tau E$. Hence the longer the electron drift length relative to the detector thickness, the less important are variations in the drift length. Fig. 2 shows a plot of $(\delta Q/Q)/(\delta(\mu\tau)_e E)/\mu\tau E$ versus $d/\mu\tau E$. Most of the simulations in this paper are carried out using $d = 0.5$ cm and $\mu\tau E = 5.0$ cm, for which $\delta Q/Q = 0.05 \delta(\mu\tau)_e E/\mu\tau E$.

It is expected that variations in the hole drift length will have a negligible impact on performance as long as $(\mu\tau)_e \gg (\mu\tau)_h$, as is always the case for $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$.

For the case in which $(\mu\tau)_e E$ varies along the detector axis, (3) is not applicable, and the induced charge must be integrated over the path of the charge carriers

$$Q = \frac{qEN_0}{d} \left[\mu_e \int_0^{t_{ee}} \exp\left(-\frac{t}{\tau_e}\right) dt + \mu_h \int_0^{t_{ch}} \exp\left(-\frac{t}{\tau_h}\right) dt \right] \quad (7)$$

where t_{ee} and t_{ch} are the electron and hole collection times, respectively. Since the variation in material parameters is known (or modeled) as a function of position, the time

Fig. 4. Determination of mobility-lifetime products from bias dependence of alpha particle pulse height for a spectrometer-grade detector.

Fig. 5. Top—Composition map for a boule sliced along the growth axis determined by room-temperature photoluminescence. Darker areas are regions of higher zinc content. Bottom—Predicted variation of zinc concentration in $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ based on the normal freeze equation using a nominal x value of 0.1 and a segregation coefficient of 1.35.

integrals in (7) must be converted to spatial integrals using the variable substitution:

$$dt = \pm \frac{dx}{\mu_{e/h} E}. \quad (8)$$

The result is

$$Q = \frac{qN_0}{d} \left\{ \int_{x_0}^d \exp \left[- \int_{x_0}^x \frac{dx'}{(\mu\tau)_e E} \right] dx + \int_0^{x_0} \exp \left[- \int_x^{x_0} \frac{dx'}{(\mu\tau)_h E} \right] dx \right\}. \quad (9)$$

Here are the factors of μE before the outer integrals have canceled. It appears that the effects of random variations in drift lengths along the detector axis are eliminated by the integrations in (9), and this intuition is borne out by simulations in Section IV-D.

III. MONTE CARLO SIMULATION METHOD

The simulations in this paper use a simple, phenomenological approach which incorporates the minimum necessary set of physical effects, allowing good computational speed. Photoelectric absorption and single and multiple Compton scattering are included, but X-ray escape and backscatter peaks are omitted. It is assumed that the charge is generated at a single point for each interaction. Much more sophisticated simulations of pulse height spectra for $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ detectors have been carried out [10], but we are the first to incorporate material nonuniformity effects.

Only two spatial dimensions are modeled in our simulations, as they are sufficient to examine all interesting effects. Uniform illumination across the detector width is assumed, and Gaussian charge generation statistical fluctuations are added based on a Fano factor of 0.14 [11]. The minimum obtainable Gaussian noise broadening is also added based on the band gap of the material and the applied electric field [12]. A

representative simulation for 300-KeV photons is shown in Fig. 3.

Key parameters which must be chosen at the outset are the electron and hole drift lengths and the detector dimensions. Because we are focusing on uniformity effects rather than hole tailing, we choose $\mu\tau E$ values which are relatively high, but realistic. The highest quality $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ material can have $(\mu\tau)_e \cong 6 \times 10^{-3} \text{ cm}^2/\text{V}$ and $(\mu\tau)_h \cong 3 \times 10^{-4} \text{ cm}^2/\text{V}$. (Fig. 4 shows determination of these parameters from plots of α particle response pulse heights versus bias voltage.) Applying an electric field of 1000 V/cm gives maximum drift lengths of 6 cm and 3 mm, respectively. For the remainder of this paper we choose relatively conservative values of $(\mu\tau)_e E = 5.0 \text{ cm}$ and $(\mu\tau)_h E = 0.1 \text{ cm}$. For detector dimensions we choose a width of 1.0 cm and a thickness of 0.5 cm.

IV. RESULTS AND DISCUSSION

A. Effect of Zinc Segregation Along the Growth Axis

The most obvious form of composition variation in Bridgman-grown $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ is the segregation of zinc along the growth axis. Since Zn in CdTe has a segregation coefficient greater than one, x decreases as the growth proceeds according to the normal freeze equation

$$x = x_{\text{nom}} K(1 - w)^{K-1} \quad (10)$$

where x_{nom} is the nominal zinc fraction, w is the molar fraction of starting material which has solidified, and K is the segregation coefficient. We use a value of 1.35 for K [13] and assume a perfectly cylindrical boule so that w can be replaced by y/L , with y being the distance from the tip and L being the boule length. Fig. 5 shows a plot of (10) along with a room-temperature photoluminescence composition map for a longitudinal slice of a boule.

Since boules are often cut parallel to the growth axis into thin slices before being diced into detectors, a portion of the composition gradient shown in Fig. 5 appears across the face of the detector. Since this gradient becomes steeper as one approaches the heel (last to freeze) end of the boule, detectors which are cut from near the heel have a substantial, unavoidable nonuniformity.

The first question which we address with our simulations is how much this composition gradient degrades energy resolution for the case in which the detector axis is perpendicular to the growth axis. Fig. 6 shows simulated spectra for 300-KeV radiation for detectors cut from various positions along the growth axis and compares them with a completely uniform detector. It is clear that the energy resolution of the detector from the tip (first to freeze) is nearly the same as for the uniform detector, while the resolution of the detector from the heel is significantly poorer. Fig. 7 shows a plot of the width-full width at half-maximum (FWHM) of the photopeak as a function of position along the growth axis for 150-, 300-, and 600-KeV radiation. The variation in energy resolution along the growth axis is greater for the higher energy photons, since at lower energies the leakage current noise and statistical fluctuations are dominant. The composition gradient broadens the photopeak by as much as 1%. As anticipated in Section II-

Fig. 6. Simulated photopeaks for 300-KeV radiation as a function of position along the growth axis when the detector axis is perpendicular to the growth axis. Same detector parameters as in Fig. 3.

Fig. 7. Maximum obtainable energy resolution for three photon energies as a function of position along the growth axis with the detector axis perpendicular to the growth axis. Same detector parameters as in Fig. 3.

Fig. 8. Simulated photopeaks for 300-KeV radiation with the detector axis perpendicular to the growth axis for (a) 1.0-cm-wide and (b) 5.0-cm-wide detectors. All other parameters the same as in Fig. 3. As the detector width increases, the effect of the composition gradient is more pronounced.

A, this effect is rather small but still significant compared to the statistical limit. Furthermore, the effect is more pronounced for larger area detectors. (See Fig. 8, which shows 300-KeV spectra for a 1-cm-wide and a 5-cm-wide detector.)

The effect of the composition gradient can be largely eliminated in principle by slicing the crystal perpendicular

Fig. 9. Simulated photopeaks for 300-KeV radiation as a function of position along the growth axis when the detector axis is parallel to the growth axis. Same detector parameters as in Fig. 3.

Fig. 10. Simulated photopeaks for 1.0-MeV radiation, using a detector taken from near the heel of the boule with the detector axis (a) parallel and (b) perpendicular to the growth axis. Same detector parameters as in Fig. 3 except that $d = 1.0$ cm.

to the growth axis rather than parallel. However, one then runs the risk of shorting the detector electrodes through the pipe defects which run along the growth axis [14]. Fig. 9 shows simulated spectra for 300-KeV radiation for detectors from various positions along the growth axis but with the detector axis parallel to the growth axis. The degradation of resolution along the growth axis which appeared in Fig. 7 is largely eliminated. This improvement has three causes. First, since the thickness of the detector typically is not as great as the width, a detector with this orientation spans a smaller fraction of the boule length than the previous geometry. This advantage does not apply to cube-shaped detectors, however. Second, because of the exponentially decreasing probability distribution for the interaction depth, the incident radiation samples the composition gradient less strongly in this orientation. This effect is weak for higher energy gamma-rays, which illuminate the detector almost uniformly throughout its volume. Lastly, there is a compensation effect, in which the variation in the average number of electron hole pairs created at different positions along the growth axis tends to counteract the hole tailing in a small region near the photopeak. This last effect is subtle but does make a substantial difference. Fig. 10 shows simulated spectra for 1-MeV radiation using a

Fig. 11. Maximum obtainable energy resolution in the presence of random variations in zinc concentration.

Fig. 12. Simulated photopeaks for 300-KeV radiation at various positions along the growth axis, assuming that the electron lifetime is limited by an impurity which segregates toward the heel with a segregation coefficient of 0.75. In these simulations the zinc segregation has been eliminated. (a) Detector axis perpendicular to growth axis. (b) Detector axis parallel to growth axis.

1-cm $\times$ 1-cm detector from the heel of the boule, in each of the two orientations. Since in this case the detector width and thickness are equal and the penetration depth of the radiation is much greater than the detector thickness, the great difference between the two spectra can only be due to a charge generation versus charge collection compensation effect.

B. Random Composition Fluctuations

In addition to the systematic decrease in zinc content along the growth axis, we have observed random composition fluctuations by room temperature photoluminescence mapping [1]–[3], [7]. To model this effect we divide the detector into cells and generate a map of random composition variation

Fig. 13. Simulated photopeaks for 300-KeV radiation showing effect of random variation in electron and hole drift lengths. If $(\mu\tau)_e E \gg d$, as in (a) and (c), there is no observable effect. If $(\mu\tau)_e E \approx d$, as in (b) and (d), then large variations in $(\mu\tau)_e E$ can have a noticeable effect, but variations in $(\mu\tau)_h E$ are still of no consequence.

having a Gaussian distribution. We use 200×200 cells so that the cell dimension is 25 to 50 μm , comparable to the step size which we use for most of our photoluminescence (PL) mapping experiments. We use as our figure of merit for uniformity the standard deviation of the zinc fraction divided by the mean $\sigma/\langle x \rangle$. Experimentally we typically observe values of $\sigma/\langle x \rangle$ between 1 and 3%, with occasional values above or below this range.

We have simulated the effects of random composition variation on detector performance. Energy resolution versus $\sigma/\langle x \rangle$ for 300- and 600-KeV radiation is shown in Fig. 11. At $\sigma/\langle x \rangle = 5\%$ the nonuniformity broadens the photopeak by about 1%, which is close to the prediction of (5).

C. Impurity Segregation Along the Growth Axis

We begin our consideration of drift length variations by supposing that the carrier lifetimes are controlled by a dominant impurity which has a segregation coefficient less than one and thus segregates toward the heel. This is not an accurate description of currently available material but will become more realistic as improved crystal growth reduces the importance of structural defects. The mean trapping lifetime is then

$$\tau = \frac{1}{\sigma_c N_T v_{th}} \quad (11)$$

where σ_c is the capture cross section, N_T is the trap density, and v_{th} is the thermal velocity. We assume that N_T follows the normal freeze equation (10) so that the drift length varies as

$$\mu\tau E(y) = \mu\tau E_{nom} / [K(1 - y/L)^{K-1}] \quad (12)$$

where y is the distance along the growth axis and L is the boule length. We assume that the mobility and electric field are uniform.

Fig. 14. Combined effects of zinc and impurity segregation on energy resolution with detector axis perpendicular to growth axis. There is a region near the middle where the two effects tend to cancel.

We assume the electron drift length varies according to (12) and that the hole drift length remains constant. As in the case of zinc segregation, we obtain progressively worse energy resolution as we approach the heel of the boule, as shown in Fig. 12(a). Also as with zinc segregation, the problem can be reduced by orienting the detector axis parallel to the growth axis, as in Fig. 12(b). This fact confirms our statement in connection with (9) that the effect of variations in transport along the detector axis tends to be eliminated by the integration along the path of the charge carriers.

D. Random Variation in Drift Lengths

It has been observed by alpha particle mapping experiments that there are local variations in $\mu\tau E$ products which do not correspond to systematic segregation of an impurity along the growth axis [4]. We model this effect by assuming random fluctuations about the nominal value. Fig. 13(a) shows that if

Fig. 15. Room-temperature photoluminescence composition map showing striations (dark bands at right) and a discontinuity in zinc concentration (middle).

the electron transport is good, $[(\mu\tau)_e E \gg d]$, then even a relatively large variation of 10% in the electron drift length has no observable effect on energy resolution because of the integration over the path of charge carriers in (9). If, however, the electron transport is poor, $[(\mu\tau)_e E \approx d]$, as in Fig. 13(b), then there is a noticeable degradation of energy resolution with large variations in $(\mu\tau)_e E$. These conclusions also apply to the case of fluctuations in the electric field, as have been reported based on electro-optical measurements [15]. On the other hand, local random variations in the hole transport are unimportant. Fig. 13(c) and (d) show that a 10% random variation in $(\mu\tau)_h E$ has no observable effect.

E. Combination of Zinc and Impurity Segregation

When zinc and impurity segregation are simulated simultaneously, the energy resolution along the growth axis is as shown in Fig. 14. Note that there is a region near the middle of the boule where the two segregation effects tend to cancel each other, since as one moves toward the heel the charge collection efficiency decreases while the number of electron-hole pairs generated increases. This result is based on assumptions that are too simple to accurately describe the current experimental situation, but it represents the best obtainable result under ideal conditions.

The important observation is that segregation alone can limit energy resolution to twice that obtainable with a perfectly uniform detector. This broadening is certainly significant, given that there are special techniques which can produce energy resolution below 2% at 662 KeV with good quality material [16], [17]. Indeed these uniformity effects may help to explain why electron-only devices sometimes achieve 2% resolution and sometimes only 3–5% [18]. On the other hand, the gradual variations simulated here are inadequate to explain the strong experimental correlation between uniformity

Fig. 16. Simulated pulse height spectra for 600-KeV radiation in the presence of striations such as shown in Fig. 15 with (a) only leakage current noise and (b) 2-KeV rms of excess noise in addition to leakage current noise.

measures and detector performance. We are therefore led to consider defects which introduce much stronger variations in composition or transport properties.

F. Modeling of Specific Defects

In this section we simulate the effects of several forms of nonuniformity which we and others have observed in real material, including growth striations, zinc segregation at a grain boundary, increased band gap near an inclusion, and trapping at a grain boundary.

In room temperature PL composition maps we have seen alternating bands of higher and lower zinc, as well as a composition discontinuity, as shown in Fig. 15. Such striations can be caused by temperature fluctuations at the growth interface due to convection [19], while the discontinuity may be segregation at a grain boundary. Each of these effects will lead, in principle, to a double photopeak in the pulse

Fig. 17. Room-temperature photoluminescence composition map showing regions of zero PL intensity (white areas) surrounded by regions of higher band gap (dark areas). We postulate that these are Te or other inclusions which have increased the band gap in the region around them either by gettering zinc or through strain.

height spectrum if the detector axis is perpendicular to the plane of the PL maps in Fig. 15. It is unlikely that such a double peak would actually be observed, as noise would cause the two peaks to blend together. The result would be a greatly broadened photopeak, as the simulation shown in Fig. 16 demonstrates.

Another feature which we see often in PL maps is a region of near zero intensity, surrounded by a region in which the spectrum is shifted to higher energy (see Fig. 17). We postulate that the defect responsible is an inclusion, probably of tellurium, and that the region of increased band gap around it is due to either strain or gettering of zinc. It has not yet been demonstrated experimentally that these inclusions have an effect on transport properties, but to investigate the possibility that they may have an impact on detector performance we model each inclusion as a central point in which the electron drift length is reduced by a factor of 100, surrounded by points in which the zinc fraction is increased by 7% of the nominal value.

The effect of a distribution of these inclusions is simulated in Fig. 18. We introduced an average of one inclusion per 0.5 mm, consistent with our experimental observations, distributed randomly throughout the detector. The result is a reduction of the photopeak height and introduction of a "hump" below the photopeak, due to the areas of reduced charge collection. If the trapping effect is not included, the inclusions have no observable effect on the spectrum. Hence we conclude that inclusions can have a significant impact on detector performance only if they affect the carrier transport properties.

The last effect which we simulate is strong trapping at a grain boundary, as reported by Luke and Eissler [20]. We allow the grain boundary to run diagonally across the detector

Fig. 18. (a) Simulated 600-KeV photopeak in the presence of inclusions of the type shown in Fig. 17, assuming that the inclusion acts as a trapping center and (b) simulated spectrum from a uniform detector for comparison. If the trapping effect is omitted and only the zinc variation is taken into consideration, there is no observable effect on the spectrum. Same detector parameters as in Fig. 3.

and assume that the $\mu\tau$ product for electrons decreases by a factor of 100 at the grain boundary, since this gives results which are consistent with the observations of Luke and Eissler. The result of the simulation is shown in Fig. 19. If the grain boundary is taken to span the entire detector, the degradation of the spectrum is extreme, with the photopeak being completely replaced by a broad hump. If the grain boundary spans only part of the detector, the effect is relatively minor, with a hump appearing below the photopeak, similar to that in Fig. 18.

V. CONCLUSIONS

We have used simple models to show that variations in alloy composition and carrier transport properties can have a

Fig. 19. Simulated 600-KeV pulse height spectrum in the presence of a grain boundary that runs diagonally across (a) part or (b) all of the detector width. Same detector parameters as in Fig. 3.

significant effect on energy resolution of $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ gamma-ray spectrometers. Systematic segregation effects alone can increase the photopeak width to as much as twice that for a perfectly uniform detector, assuming a detector width of 1 cm and a nominal zinc fraction of 0.2. These effects may be partly responsible for limiting the performance of high-resolution, electron-only devices. Furthermore, the impact of uniformity on detector performance is greatly increased if a larger detector width or higher zinc concentration is used. However, we have shown that the impact of zinc and impurity segregation on detector performance can be greatly reduced by orienting the detector axis parallel to the growth axis.

The strong experimental correlation between measures of uniformity and detector performance can be explained by the existence of structural defects which act as trapping centers, but further experimental investigation of the impact of inclusions on transport properties is needed.

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EXTRACTING TRAP PARAMETERS FROM PICTS SPECTRA IN CADMIUM ZINC TELLURIDE RADIATION DETECTOR MATERIAL

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ABSTRACT

We demonstrate that the regularization method due to Weese can resolve closely-spaced peaks in PICTS spectra for cadmium zinc telluride. We also show that electron and hole traps can be distinguished from each other by the bias dependence of the spectrum when using an excitation source that is primarily infrared but which contains a small component of visible light. Lastly we show that there are qualitative differences between PICTS spectra taken with infrared excitation and those taken with visible excitation. We attribute the surface-related levels to diffusion into the detector material of gold from electroless deposition of contacts.

INTRODUCTION

While great progress has been made in recent years in qualitative understanding of the properties of CdZnTe radiation detector material, detailed understanding of the behavior of specific impurities and defects is still insufficient. Photo-induced current transient spectroscopy (PICTS) is a promising technique for characterizing defect levels, but accurate extraction of energy levels can be problematic, especially in alloys, where the energy spectrum is broadened by microscopic and macroscopic composition variation. In this paper we discuss some problems of data collection and analysis and potential solutions.

ANALYSIS ISSUES

The basic procedure of a PICTS experiment is to subject the sample under bias to millisecond pulses of light and observe the turn-off transient of the photocurrent at various temperatures. The rate of decay of the photocurrent is controlled by the emission rate of the dominant trap, which is strongly temperature dependent. The evolution of the turn-off transient with temperature therefore contains information about trap levels and capture cross sections, but extracting that information is not always simple.

Theory of PICTS Experiments

The classic assumption in PICTS analysis is that the turn-off photocurrent transient in the presence of a single trap is given as a function of time by:

$$i(t) \propto \frac{1}{\tau} e^{-t/\tau} \quad (1)$$

where τ , the relaxation time, is equal to one over the emission rate of the trap and depends on temperature as:

$$\frac{1}{\tau} \propto T^2 e^{-\Delta E/(kT)} \quad (2)$$

where ΔE is the depth of the trap. If equations (1) and (2) are valid, then a plot of i vs. T for fixed t will have a maximum at the temperature at which $1/t$ equals the emission rate. If the peak temperature, T_m , is measured for a number of times t , then a plot of $(T_m^2 t)$ vs. $1/kT_m$ should be a

straight line with slope ΔE . The y-intercept of the line can be used to calculate the capture cross-section, σ . Further discussion can be found in Bube [1].

Methods of Data Analysis

In practice the PICTS signal is taken as the difference between currents at two times, t_1 and t_2 . Fundamentally the purpose of the subtraction is to remove the dark current, so that one would like to make t_2 as long as possible. In practice, however, it is found that better resolution between peaks is obtained by using a fixed, and relatively small, ratio of t_2/t_1 [2]. However the smaller the t_2/t_1 ratio, the greater noise problems become, since one is subtracting two currents which are not very different, and plotting changes in this difference against small temperature changes.

A further difficulty is that equation (1) is valid only under the condition that the excitation is high enough to saturate the traps, but such high excitation levels may lead to space-charge limiting of the photocurrent. An alternative analysis method which eliminates this problem is the four-gate technique [3], but this method also involves subtracting currents at closely spaced times, requiring a very low noise level.

A radically different analysis method is to regard the turn-off transient waveform as having a relaxation time spectrum, $h(\tau)$, such that:

$$i(t) = i_D + \int_0^{\infty} h(\tau) e^{-t/\tau} d\tau \quad (3)$$

where i_D is the dark current. Weese has produced a program to solve equation (3) for $h(\tau)$ using the technique of fast Tikhonov regularization [4], and in the following section we illustrate the use of this program to resolve closely spaced trap levels in the presence of alloy broadening.

Application of the Regularization Method to CdZnTe Spectra

Figure 1 shows PICTS spectra for two similar CdZnTe samples. More information about the samples and discussion of the possible origin of the bands are given in a separate paper [5]. In each spectrum there are two dominant bands, one in the 180-200 K range (band I), and one in the 230-280 K range (band II). The peaks shift to lower τ with increasing T as expected. In the spectrum on the left (sample 1), each band has the appearance of a single peak, while in the spectrum on the right (sample 2) each band appears to have at least two components. The relaxation time spectra for band I are shown in Figure 2 and confirm the presence of two levels. (The third peak for sample 2 at 186K and 192 K seems to be spurious, since it does not appear in spectra at lower or higher temperatures.) For sample 2 the two peaks are comparable in amplitude, while for sample 1 the second peak is extremely weak, which explains why it is not evident in the PICTS spectrum. An interesting feature of all of the relaxation time spectra is the rather large breadth of the bands compared to results reported for CdTe [6]. We attribute this broadening to microscopic and macroscopic composition fluctuations.

Figure 3 shows the use of Arrhenius plots to determine the trap levels and cross-sections. The levels b_1 and a_2 have approximately the same energy (0.31 eV) and cross-section (10^{-16} cm^2) and can be identified as the same defect. Levels b_2 (0.52 eV, 10^{-10} cm^2) and a_1 (0.18 eV, 10^{-20} cm^2) are clearly distinct, however. The Arrhenius plots tend to be rather scattered, so that the extracted values must be regarded as rough estimates. The regularization method is therefore most useful in cases where even estimates for the trap levels cannot be obtained from the traditional

Figure 1 : PICTS spectra for two CdZnTe samples from the same boule. Bias = 20V, peak excitation wavelength = 875 nm, $t_2/t_1 = 8$.

Figure 2 : Relaxation time spectra for the two CdZnTe samples.

Figure 3 : Arrhenius plots extracted from the relaxation time spectra of Figure 2.

A similar analysis for band II yields two levels for each spectrum. For sample 1 the levels and cross sections are c_1 : 0.76 eV, 10⁻¹¹ cm² and d_1 : 0.22 eV, 10⁻²⁰ cm²; for sample 2 they are c_2 : 0.59 eV, 10⁻¹⁴ cm² and d_2 : 0.34 eV, 10⁻¹⁸ cm². The parameters for d_1 are curiously close to those for a_1 , but the two series of peaks in the relaxation time spectra are distinct, so that they must be regarded as separate levels.

Intrinsic vs. Extrinsic Excitation

It is sometimes stated in the literature that PICTS measurements should (or must) be carried out with below-bandgap excitation in order to fill trap levels uniformly throughout the sample volume and avoid space-charge effects. On the other hand, in one of the earliest PICTS papers [7] Martin and Bois used visible light to selectively excite electron or hole traps depending on bias polarity. For most of our measurements we have taken a middle-ground approach of using an infrared LED with a peak wavelength of 875 nm - below the band gap of CdZnTe - but with a slight tail into the visible region. In this way we obtain excitation throughout the volume, but with a small component of intrinsic excitation that gives sufficient polarity dependence to distinguish between electron and hole traps in some cases (see Figure 4).

On the other hand, we have found that by using strictly above-bandgap excitation we sometimes obtain qualitatively different results than with IR excitation. Figure 5 shows PICTS spectra for a CdZnTe sample taken with visible (left) and infrared excitation. The "electron" and "hole" spectra with visible excitation are nearly the same, while the spectrum taken with IR excitation is qualitatively different. These facts may imply that bands III and IV are surface-related, while band V is due to a bulk defect. The energy level and cross-section for band III extracted by the two-gate method are 0.27 eV and 10⁻¹⁴ cm², which agree precisely with values reported for a

gold-related level in CdTe [8]. We conclude that gold from the electroless contacts has diffused into the material, creating a defect-rich region.

Figure 4 : PICTS spectrum for a CdZnTe detector with illuminated contact biased positive (solid) and negative (dashed). Because the signal is stronger in the former case, both bands I and II can be identified as hole traps.

Figure 5 : Comparison of PICTS spectra with visible (left) and infrared excitation.

CONCLUSIONS

We have shown that Weese's regularization method can be used to resolve closely-spaced trap levels in the presence of alloy broadening. The Arrhenius plots for determining the energy levels tend to be rather scattered, however, so that the extracted values must be considered as a rough estimate.

We have also shown that electron and hole traps can be distinguished from one another by the polarity dependence of the spectrum using an excitation source that is primarily infrared but which has a small visible component. By using a strictly visible source, on the other hand, we have shown that surface-related levels, such as those due to diffusion of gold from electrodeless contacts into the detector material, can be detected.

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ON THE ACTIVE VOLUME OF CADMIUM ZINC TELLURIDE GAMMA-RAY SPECTROMETERS

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ABSTRACT

In this paper we develop quantitative models to predict the active volume of cadmium zinc telluride (CZT) detectors operated as gamma-ray pulse height spectrometers. Three cases are considered: a conventional planar detector, a unipolar device, and a detector in which electronic signal processing has been applied to correct for charge trapping effects. We find that existing detectors are very limited in their maximum attainable active volume, but unipolar devices with charge correction show promise for producing large active volume devices.

INTRODUCTION

Cadmium Zinc Telluride gamma ray detectors are being used with greater frequency as crystals of this material become more available. When considering the use of a CZT detector in a new application, it is often desirable to know the efficiency of the detector for the radiation to be detected. The first step in determining the efficiency of a CZT detector towards penetrating gamma-rays is to determine the active volume of the detector. Often, it is assumed that the active volume of the detector is the same as its physical volume. However, when CZT detectors are used as pulse height spectrometers the active volume of the detector is often much less than the physical volume of the detector. The difference between the active volume and the physical volume is due to charge trapping effects.

In this paper we compute the expected efficiency of the CZT gamma ray spectrometers under various circumstances. In particular we compute the active volume of a planar detector as a function of material parameters and operating conditions. Next we compute the active volume of a unipolar charge sensing or "electron only" type of device. Finally we compute the expected efficiency of a device employing electronic methods to compensate for charge trapping.

Before we begin our discussion of the efficiency of a CZT detector we wish to define precisely what we mean by "active volume" in a spectroscopic detector. Consider the hypothetical pulse height spectrum shown in Figure 1 due to monoenergetic photons interacting uniformly throughout the detector and producing Q_0 of charge with each interaction. The pulse height spectrum arising from this hypothetical interaction may be thought of as some function $p(Q)$, which represents the probability of measuring a given charge on the contact of the detector. Note that for a perfect detector $p(Q)$ would be a delta function at Q_0 . The active volume (Vol_{eff}) of the detector, from a pulse height spectroscopy point of

Figure 1: Hypothetical pulse height spectrum resulting from a monoenergetic photon interaction producing Q_0 of charge. The peak width is denoted as Γ .

view, is proportional to the ratio of events under the peak to the total number of interactions occurring in the detector. More rigorously,

$$Vol_{eff} = Vol_{physical} \int_{Q_o - \Gamma}^{Q_o + \Gamma} p(Q) dQ, \quad (1)$$

where $Vol_{physical}$ is the physical volume of the device, and Γ is the width of the peak. Using this simple result we can now compute the effective volume of a detector under various circumstances.

Planar Detector

The simplest type of CZT radiation detector is the planar detector; formed by the application of contacts on opposite parallel faces of a CZT sample. In a planar detector of thickness d , the charge collection efficiency, η , as a function of position in the detector, x , is given by [1],

$$\eta(x) = \frac{\lambda_e}{d} \left(1 - \exp\left(-\frac{d-x}{\lambda_e}\right) \right) + \frac{\lambda_h}{d} \left(1 - \exp\left(-\frac{x}{\lambda_h}\right) \right), \quad (2)$$

where λ_e and λ_h are the drift length of electrons and holes respectively. If we assume that the electric field in the detector is a constant and extends across the entire detector then the drift length of electrons is given by

$$\lambda_e = \frac{\mu_e \tau_e V}{d}, \quad (3)$$

where V is the potential difference across the detector, μ_e is the drift mobility and τ_e is the trapping time of electrons. A similar expression may be written for holes.

With a knowledge of the charge collection efficiency as a function of position in the detector, we can compute the active volume of the detector. For any well behaved charge collection distribution, we would expect the most probable value of charge collection to be equal to the maximum charge collection value. Thus computing the active volume of a planar detector reduces to finding the position in the detector where the charge collection is at a maximum and finding the distance from this point to where the charge collection falls to $Q_{max} - \Gamma$ of its value (Figure 2). For a planar

Figure 2: Charge collection versus position in detector (left), and corresponding noise-free pulse height spectrum (right). Note: $Q(x) = \eta(x)Q_o$

detector the maximum value of the charge collection efficiency occurs at, x_{max} (4)

$$x_{max} = \frac{\lambda_h}{\lambda_e + \lambda_h} d, \quad (4)$$

$$\eta_{max} = \eta(x_{max}) = \frac{\lambda_e + \lambda_h}{d} \exp\left(-\frac{d}{\lambda_e + \lambda_h}\right). \quad (5)$$

The active thickness of the detector for the planar case can be computed by solving the following equation for x over the range $\{0 < x < d\}$.

$$\eta(x) = \eta_{max} - \Gamma \quad (6)$$

If equation(6) has two solutions, (on either side of the maximum) and we denote these solutions x_1 and x_2 then the active thickness of the detector is simply.

$$x_{active} = |x_1 - x_2| \quad (7)$$

Conversely, if equation (6) has only one solution, then that solution is the active thickness. Of course, for a planar detector, once the active thickness is known the active volume can be computed from it by simply multiplying by the area of the device.

Using the above formalism, and assuming a constant electric field in the detector, we have computed the active volumes of some typical planar CZT detectors and some results are shown in Figure 3. Note that the active volume of typical devices is much smaller than the physical volume. Note too that thin devices have a greater relative active volume, as should be expected since - in the constant field case - the ratio of the drift length to the device thickness goes as V/d^2 . Thus two stacked detectors can often give more active volume than a single element device of greater physical thickness. When computing the results shown in Figure 3 we truncated the solution to exist only if the field in the detector was less than 2000 V/cm. Our laboratory experience has shown that is very difficult to build devices that can withstand a field greater than 2 kV/cm with current technology.

It has been noted [2] that when the drift length of both carriers becomes much less than the thickness of the device, relatively uniform charge collection results. In principle this would appear to offer a method for producing more active volume in a detector. In practice, however, when the effects of photon transport and noise are considered, it becomes apparent that reducing the drift length of the carriers is not a practical solution to increasing the active volume. For instance, consider a CZT detector used to perform pulse height spectroscopy on ^{137}Cs (662 keV photons). If the bias on the detector were reduced to where the charge collection is somewhat uniform throughout the detector, the maximum signal would be reduced to about 1/10 of the value of full

Figure 3: Active detector thickness as a function of bias for three different device thickness.

charge collection. Assuming the total noise of the detector was 14 keV FWHM of noise referred to full charge collection (2% relative energy resolution), the noise would become 20% of the 662 keV peak at the reduced bias needed for uniform charge collection. At 20% energy resolution, the CZT detector would have no performance advantage over a scintillator based system. To compute the proper effects of reducing carrier drift length it is necessary to convolve the expected charge pulse height spectrum with the Gaussian electronic noise expected in the detection system. However, for simplicity in estimation purposes, we recommend constraining the solutions of equation (6) such that the charge collection efficiency is always greater than about 0.6; under these conditions the convolution can be avoided and equation (6) can be used directly to estimate the active thickness of the detector. For the case of a detector operating with a constant electric field, an approximate solution to the active volume of the detector is given by the following expression

$$x_{active} = x_{max} + (\eta_{max} + \Gamma)d \exp\left(\frac{d - x_{max}}{2\lambda_e}\right) \quad (8)$$

This expression was obtained by approximating the charge collection efficiency as a function of detector position as a line between η_{max} , x_{max} and $\eta(d)$, d . Equation (8) may be used to predict the active volume of a detector based on measured charge carrier transport characteristics.

Unipolar Device

There has been a great deal of recent interest in the use of CZT gamma-ray spectrometers based on unipolar or "electron-only" designs. Such devices promise to have a much larger active volume because only the trapping effects of electrons (not holes) diminish the performance of the device as a pulse height spectrometer.

Consider a somewhat idealized unipolar device where the weighting potential [3] of the anode is localized very close to the anode (under these circumstances charge is only induced on the anode when electrons travel very close to it). In this hypothetical unipolar device, trapping of electrons induces a distortion of the pulse height spectrum and corresponding decrease in spectroscopic efficiency. For the idealized unipolar device, the charge collection efficiency as a function of distance from the anode is simply given by,

$$\eta(x) = \exp\left(-\frac{x}{\lambda_e}\right), \quad (9)$$

where, x is the distance from the cathode and, as before, λ_e is the drift length of electrons. Since the charge collection is simply a monotonic decreasing function of distance from the anode, the active thickness of the device at a given resolution is given by,

$$x_{active} = -\ln\left(\frac{1 - \Gamma}{\lambda_e}\right). \quad (10)$$

Note that for a spectroscopic device with good energy resolution (Γ small), the active volume is simply $\Gamma\lambda_e$. Thus, for an electron only device made with present day material ($\lambda_{max} \sim 4$ cm), good energy resolution ($\Gamma < 0.1$) limits the active thickness to only about 4 mm maximum. However, as we discuss in the next section of this paper, electron only devices show much greater promise for large volume devices when electronic means are used to compensate for the electron trapping.

A variety of methods have been proposed over the years to electronically compensate for the effects of charge carrier trapping [4-7]. Such methods offer the possibility of substantially improving the active volume of CZT gamma-ray detectors by "moving" pulses (with reduced amplitude from charge trapping) back into the full-energy peak. Most of the charge compensation methods developed to date rely on correcting the amplitude of the pulse based on its shape in the time domain. Implicit in the reasoning behind the charge compensation methods based on pulse shape is that there is a one-to-one correspondence between the position of interaction in the detector and the amount of charge compensation required. However, multiple interaction events (such as Compton scattering followed by photo-electric absorption) do not have a single position of interaction in the detector and thus cannot be properly corrected using charge compensation methods. Once detectors become substantially larger than 1 cm³, multiple interaction events become an important (even dominant) contribution to the full energy peak in a gamma-ray spectrometer. Thus, the deficiencies of conventional charge compensation techniques preclude their use in developing CZT detectors that could compete with germanium or scintillation detectors for efficiency. Nonetheless, conventional charge compensation techniques would be expected to boost the active volume of small detectors whose gamma-ray efficiency is dominated by photo-electric interactions. The active (photo-electric only) volume for a hypothetical charge compensation method can be derived by assuming that all of the trapped charge is placed back in the full-energy peak. At first glance this would seem to indicate that charge compensation would produce detectors whose entire physical volume is active. However when the additional noise broadening of the corrected charge is considered, the efficiency of a compensated device is diminished. In particular, the full energy peak in a compensated device may be thought of as composed of the sum of a series of Gaussian peaks with the same centroid but different standard deviations. Note that, in general, this sum of Gaussians peak would not itself be a Gaussian. The standard deviation due to each point in the detector is simply equal to the noise width (equivalent noise charge (ENC) divided by the charge collection efficiency at that point in the detector). In the limit, this sum of Gaussians becomes an integral,

$$V_{fraction} = \int_{Q_o - \Gamma}^{Q_o + \Gamma} \int_0^d \frac{\eta(x)}{ENC \sqrt{2\pi}} \exp\left(-\left(\frac{(Q - Q_o)\eta(x)}{2ENC}\right)^2\right) dx dQ. \quad (11)$$

In other words, charge compensation can correct the amplitude of a peak but it cannot change its relative width.

Charge trapping compensation methods can also be applied to unipolar devices as first suggested by He [8]. In the case of unipolar devices, it may also be possible to correct for multiple interaction Compton events. This possibility arises because multiple scatter events can be distinguished in unipolar devices. In an ideal unipolar device, no charge is induced on the anode until electrons drift into close proximity. Thus, multiple scatter events are distinguishable by the distinct (but close) arrival times of the induced charge pulses on the anode. Such a compensating system for multiple scatter events would be difficult to implement however, because it would necessitate short amplifier integration times to distinguish the distinct arrival of the pulses (tens of ns assuming an electron mobility of 1000 cm²/Vs and a average electric field of 1000 V/cm). At these short integration times the series white noise [3] of the amplifier would be rather large, and the energy resolution would become dominated by noise. Despite these technical difficulties, a unipolar device capable of compensating multiple interaction events would offer the possibility of CZT detectors with active thickness of several centimeters, many times the active thickness achievable using other methods

SUMMARY

We have computed the expected spectroscopic efficiency in CZT detectors under various circumstances. We found that conventional planar detectors available today have very small active volumes compared to their physical size. Planar detectors that are commercially available are limited in their active thickness to about 2 mm by hole trapping effects, irrespective of the physical thickness of the detector. Unipolar devices offer an increase of active volume but these devices too are also limited, in this case by electron trapping, to active thickness of less than about 5 mm. Of course, if the charge carrier transport properties of CZT crystals were to improve this would lead to an immediate improvement in achievable active volumes. The most promising approach to CZT detectors that could attain large active volumes would seem to be a unipolar device with some form of correction for electron trapping. If such a compensation mechanism could be implemented, and it was capable of correctly processing multiple interaction events, we would see CZT detector with truly large active volumes ($> 10 \text{ cm}^3$).

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Performance of a coaxial geometry $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ detector

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Abstract

For many years, coaxial geometry detectors have been the preferred configuration for large volume high purity germanium detectors. Recently, developments in $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ crystal growth and detector fabrication have enabled the construction of coaxial geometry $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ detectors for the first time. We report on the performance of a coaxial geometry $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ detector operated as a gamma ray spectrometer. The pulse height spectra obtained from this detector were analyzed using a theory developed by Sakai for use with germanium detectors and good agreement was obtained between the predictions of Sakai's theory and the results measured in our laboratory. We also comment on some possible improvements that could be made to future coaxial geometry $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ detectors.

1. Introduction

In the last few years $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ (CZT) has emerged as one of the leading materials for the construction of room temperature semiconductor gamma ray spectrometers. $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ crystals with zinc mole fraction (x) in the range of 0.1 to 0.3 can now be routinely produced in large volumes ($>1\text{ cm}^3$) by the high pressure Bridgman technique. The CZT crystals produced by the high pressure Bridgman method have higher electrical resistivity ($10^{11}\ \Omega\text{ cm}$) than the more commonly used CdTe crystals ($10^9\ \Omega\text{ cm}$). Because the electrical resistivity of CZT crystals is typically two orders of magnitude larger than CdTe crystals, it is possible to construct CZT detectors with an order of magnitude greater area than a CdTe detector, operate them at the same electric field, and obtain the same "shot noise" due to detector leakage currents. To date all CdZnTe detectors reported have been of cylindrical (with contacts on two ends of the cylinder) or of rectangular parallelepiped geometry [1]. However, the availability of suitable crystals, and the development of more advanced fabrication methods, has allowed the manufacturers of CZT detectors to explore the use of alternative device geometries. Motivated by the success of coaxial geometry germanium detectors, one CZT manufacturer, eV Products of Saxonburg, PA, USA has produced detectors with a coaxial geometry. We report on the performance of one of these detectors.

2. Detector design

The coaxial geometry CZT detector we analyzed (eV Products #A1353) had an inside diameter (r_1) of 0.3 cm, an outside diameter (r_2) of 0.625 cm, and a length of 0.7 cm. Fig. 1 is a diagram illustrating the geometry of the detector. The detector was housed in a cylindrical aluminum package with a BNC connector attached. The material used to construct the detector was grown by the high pressure Bridgman technique, and estimated by the manufacturer to contain a zinc fraction of approximately 20 mole%.

Fig. 1. Diagram illustrating the geometry of the coaxial detector used in this study.

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3. Detector testing

Conventional pulse height analysis instrumentation was used to measure the signals produced by the coaxial detector: a charge sensitive preamplifier, a shaping amplifier and a pulse height analyzer. The charge sensitive preamplifier used was a Tennelec TC-170 which was AC coupled to the detector. Before conducting the studies of the coaxial CZT detector, the pulse height apparatus was calibrated by affixing a silicon detector to the preamplifier and irradiating the silicon detector with X-ray photons of known energy. Using the average energy required to

produce an electron-hole pair in silicon (3.65 eV) [2] was then possible to calibrate the pulse height analysis system in absolute charge units (electrons/channel).

The coaxial CZT detector was then affixed to the preamplifier, biased with a high voltage power supply, and irradiated with isotopic gamma ray sources. In this paper we use the convention of a positive bias (+HV) to represent the potential of the inner contact to be more positive than the outer contact.

Typical pulse height spectra obtained by irradiating the detector with isotopic sources are shown in Fig. 2. The ^{133}Ba and ^{137}Cs sources used for testing were approximately 10 μCi in strength and were placed approximately 10 cm from the detector, and perpendicular to the central axis of the device. Qualitatively, the performance of the detector as a gamma ray spectrometer was somewhat worse than a planar geometry CZT detector of similar volume. Note in Fig. 2 that the 662 keV photopeak of the ^{137}Cs is clearly resolved, however an asymmetrical distortion of the peak is also clearly evident. This asymmetrical distortion of high energy gamma ray peaks is also observed in CZT detectors of a cubic geometry and is known to arise from charge collection effects [3]. In particular, if the drift length of holes is less than the distance between the detector contacts, "hole tailing" (asymmetrical peak formation) is observed. This "hole tailing" arises from the fact that the charge collected efficiency varies depending on the depth of interaction of the gamma within the crystal [4]. A more quantitative analysis of the shapes of the peaks in the pulse height spectra is discussed in the next section of this paper.

Fig. 2. Pulse height spectra obtained by irradiating a coaxial geometry CZT detector with gamma rays from: (upper fig.) ^{133}Ba , and (lower fig.) ^{137}Cs . Note: upper fig. has logarithmic vertical axis and lower fig. includes the peak produced by an injected electronic pulser.

4. Analysis

The pulse height spectra obtained from the coaxial CZT detector were analyzed using a theory developed by Sakai for use with germanium detectors [5]. Sakai derived an expression for the charge collected from a coaxial geometry detector as a function of the position of interaction of the gamma ray within the detector (r_0):

$$Q(V, r_0) = \frac{Q_0}{\ln\left(\frac{r_2}{r_1}\right)} \cdot \left\{ \int_{r_0}^{r_2} \frac{1}{r} \exp\left[\frac{-\ln\left(\frac{r_2}{r_1}\right)}{2\mu\tau_h V} (r^2 - r_0^2) \right] dr + \int_{r_1}^{r_0} \frac{1}{r} \exp\left[\frac{-\ln\left(\frac{r_2}{r_1}\right)}{2\mu\tau_e V} (r_0^2 - r^2) \right] dr \right\}, \quad (1)$$

where Q_0 is the charge created by the gamma interaction, $\mu\tau_e$ and $\mu\tau_h$ are the product of the mobility and drift lifetime for electrons and hole respectively in CZT, V is

the potential difference between contacts at r_1 and r_2 (see Fig. 1), and Q is the measured charge. Sakai's expression assumes that the mobility and trapping lifetime are not dependent on the electric field, thus Eq. (1) would not be accurate when the electric field in the detector becomes high and "hot electron" effects become significant (velocity of electrons and holes no longer proportional to field strength).

Eq. (1) can be used to predict two types of experimental measurements. First, the maximum pulse height observed from the detectors (Q) can be predicted as a function of the bias applied to the detector (V). Eq. (1) can also be used to estimate the shape of the photo-peak produced by gamma ray interaction in the detector. For deeply penetrating photons (such as the 662 keV photon of ^{137}Cs), the photo-electric gamma ray interactions from a large number of photons may be approximated by a uniform distribution of point like energy depositions (all of magnitude E_0 , the energy of the photo-electron) throughout the thickness of the detector. Thus, by sampling Eq. (1) at regular intervals over r_0 (from r_1 to r_2), and taking a histogram of the sampled amplitudes, a noise-free approximation to the pulse height spectrum of a high energy photo-peak can be obtained.

To test if Eq. (1) was useful in predicting the performance of the coaxial CZT detector, a series of ^{137}Cs spectra were taken at various detector bias voltages (Fig. 3). The position of the peaks due to the 662 keV photons obtained at the different bias voltages was also recorded (Fig. 4). A linear amplifier time constant of 10 μs was used to acquire all of the data shown in Figs. 3 and 4 to prevent any ballistic deficit effects. Also plotted in Fig. 4

Fig. 3. Pulse height spectra obtained from the coaxial CZT detector at various bias voltages. A ^{137}Cs source was used to irradiate the detector, and only the regions of the pulse height spectra in the vicinity of the 662 keV photo-peak are shown.

Fig. 4. Maximum pulse height observed when irradiating the coaxial detector as a function of bias applied to the detector. A ^{137}Cs source was used to irradiate the detector. Square symbols represent experimental data; circles represent the predictions of the theory discussed in the text.

are the maximum values of Q from Eq. (1) (over all r_0 from r_1 to r_2) for various detector bias voltages. Values of $\mu\tau_e = 6 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1}$ and $\mu\tau_h = 9 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1}$ were used in the calculations; these mobility-lifetime estimates were obtained experimentally from measurements taken on a planar geometry detector constructed of CZT and similar to that used to construct the coaxial device. The value of Q_0 used in Fig. 4 was obtained by assuming a mean value of the energy to create an electron-hole pair, ϵ_{pair} , of 4.9 eV. Thus, we would expect 1.35×10^5 electrons of charge to be generated when a 662 keV photon undergoes a photoelectric interaction in the detector.

Figs. 5 and 6 illustrate the expected pulse height spectra—using the aforementioned uniform sampling method—along with some experimental data; good agreement is obtained between the predictions and the experimental values.

Using this same technique of sampling and histogramming Eq. (1), it is possible to estimate the charge collection distortion from future detectors: Figs. 7 and 8 illustrate the use of this predictive technique. Fig. 7 indicates that improving the quality of the CZT used to manufacture the coaxial detector would lead to substantially improved performance. In particular, we estimate that increasing the trapping lifetime of holes by one order of magnitude would have a very beneficial effect on coaxial detector performance. The results shown in Fig. 8 indicate that some improvement in coaxial detector performance could also be achieved by altering the device geometry, even with existing quality CZT.

Fig. 5. Comparison of experimental ^{137}Cs pulse height spectrum and prediction using theory discussed in the text. A detector bias of 390 V was used in the measurement.

Fig. 6. Comparison of experimental ^{137}Cs pulse height spectrum and prediction using theory discussed in the text. A detector bias of 245 V was used.

5. Summary

A $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ detector of coaxial geometry was tested as a gamma ray spectrometer. The detector produced useful pulse height spectra of energetic sources, but the photo-peaks of deeply penetrating photons were distorted because of the poor hole transport in the material. We found that the response of the coaxial CZT detector could be predicted—to good approximation—using a theory developed by Sakai for use with coaxial Ge detectors. Using this theory, we were also able to predict that

Fig. 7. Estimate of pulse height spectrum that would be obtained by irradiating a coaxial geometry CZT detector with ^{137}Cs photons if the mobility–lifetime product of holes in the material were 10 times greater than in existing material (solid circles). For comparison, the estimated performance of an existing detector under the same operating conditions (detector bias = 390 V) is also shown (solid squares). The geometry of the detector assumed in the calculations was the same as that of the device measured in the laboratory ($r_1 = 0.3$ cm, $r_2 = 0.625$ cm).

Fig. 8. Estimate of pulse height spectra that would be obtained by irradiating a coaxial geometry CZT detector with ^{137}Cs photons. Three geometries are assumed: 1) thick detector ($r_1 = 0.2$ cm, $r_2 = 0.625$ cm), 2) existing detector ($r_1 = 0.3$ cm, $r_2 = 0.625$ cm), and 3) thin detector ($r_1 = 0.5$ cm, $r_2 = 0.625$ cm). A detector bias of 390 V was assumed for all calculations.

substantial improvements in coaxial CZT detector performance could be obtained through altering the geometry of the device or improving the lifetime of holes in the material.

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MATERIAL INHOMOGENEITIES IN $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ AND THEIR EFFECTS ON LARGE VOLUME GAMMA-RAY DETECTORS

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Abstract

Cadmium zinc telluride ($\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ or CZT) has shown great promise as a material for room-temperature x-ray and gamma-ray detectors. In particular, polycrystalline material grown by the High Pressure Bridgman method with nominal Zn fraction (x) from 0.1 to 0.2 has been used to fabricate high resolution gamma-ray spectrometers with resolution approaching that of cooled high-purity Ge. For increased sensitivity, large areas ($> 1 \text{ cm}^2$) are required, and for good sensitivity to high energy gamma photons, thick detectors (on the order of 1 cm) are required. Thus there has been a push for the development of CZT detectors with a volume greater than 1 cm^3 . However, nonuniformities in the material over this scale degrade the performance of the detectors. Variations in the zinc fraction, and thus the bandgap, and changes in the impurity distributions, both of which arise from the selective segregation of elements during crystal growth, result in spectral distortions. In this work several materials characterization techniques were combined with detector evaluations to determine the materials properties limiting detector performance. Materials measurements were performed on detectors found to have differing performance. Measurements conducted include infrared transmission (IR), particle induced x-ray emission (PIXE), photoluminescence (PL), and triaxial x-ray diffraction (TAXRD). To varying degrees, these measurements reveal that "poor-performance" detectors exhibit higher nonuniformities than "spectrometer-grade" detectors. This is reasonable, as regions of CZT material with different properties will give different localized spectral responses, which combine to result in a degraded spectrum for the total device.

Introduction

Cadmium zinc telluride ($\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ or CZT) has been developed for use in room-temperature x-ray and gamma-ray detectors only since 1992 [1,2]. During the few years of development since, however, there has been tremendous improvement such that polycrystalline material grown by the High Pressure Bridgman (HPB) method with nominal Zn fraction (x) from 0.1 to 0.2 has been used to fabricate high resolution gamma-ray spectrometers with resolution approaching that of cooled high-purity Ge. In many applications the available radiation level is low, requiring as high a sensitivity as possible. This means both a need for large active areas of more than 1 cm^2 , and for the detection of higher energy gamma photons, an active thickness on the order of 1 cm or greater. Thus there has been a push for the development of CZT detectors with a volume greater than 1 cm^3 . So far, detectors as large as $1.5 \text{ cm} \times 1.5 \text{ cm} \times 1.5 \text{ cm}$ have been fabricated and evaluated [3,4].

However, nonuniformities in the material at this scale degrade the performance of the detectors. For example, variations in the zinc fraction (and thus the bandgap), and changes in the impurity distributions, both of which arise from the selective segregation of elements during crystal growth, result in spectral distortions. This is particularly a problem in the dimension of a detector in the crystal growth direction. In addition, there is an inherent problem with the poor carrier transport properties of holes as compared to those of electrons. The location of interaction of a gamma event within the detector affects the distances traveled by the electron and hole in reaching the positive and negative electrodes, and thus the pulse response measured. Finally, there are variations in the materials in directions perpendicular to the growth axis because of growth variations.

In this work several materials characterization techniques were combined with detector characterizations to determine the materials properties limiting detector performance. Given the relative effect of composition on bandgap, a one percentage point change in zinc fraction (e.g., from $x=0.095$ to $x=0.105$) results in a change in the energy required to produce an electron-hole pair (E_{pair}) of one-half percent, which means that the amount of charge created, and thus the pulse height measured, changes by $1/2 \%$ across the detector. This leads to a broadening of the

photopeak and a loss in resolution. Thus the inhomogeneity in the zinc composition in a CZT detector leads to a degradation of the spectral response. In addition, in thicker detectors there is also a problem with incomplete charge collection, as the drift length for holes can be less than the distance from the point of photon interaction to the negative contact. This gives rise to "hole-tailing" wherein the low energy side of the photopeak has a long decay down to background. Fig. 1 shows a ^{137}Cs spectrum from a 1 cm^3 CZT detector exhibiting hole-tailing. By mapping out the zinc composition and electrically-active defects, one can determine how large a detector crystal can be "mined" out of a boule without leading to an excessive loss of spectral resolution. There are signal processing approaches being pursued to improve the performance of current large volume CZT spectrometers in the near term by compensating for the poor hole collection problems electronically [5].

Materials Characterizations

Samples Studied

For the materials characterizations two nominal $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ $1\text{ cm} \times 1\text{ cm} \times 0.2\text{ cm}$ detectors from eV Products were used. After detector characterizations the gold contacts were removed to allow for the various materials measurements. One detector, the "spectrometer-grade" device, had good spectral response, while the other, the "poor-performance" device, had little spectral resolution, acting only as a counter.

Infrared Transmission Microscopy

A relatively quick and simple qualitative measurement of CZT material quality is infrared transmission microscopy. While the CZT itself is transparent to the IR, precipitates and inclusions like metallic tellurium or cadmium, and other imperfections are opaque to the IR and show up as dark spots. For the work here, a simple IR emitting LED was shone on one side of the samples and the transmitted light collected with lenses and detected with a simple CCD camera with its IR filter removed. Fig. 2a shows the images obtained for both the spectrometer-grade and poor-performance materials. These are negative images, meaning that darker regions indicate higher

transmission. As mentioned above, IR generally highlights the metallic inclusions and precipitates in CZT. The spectrometer-grade material appears to be more uniform than the poor-performance, in that the dark regions are found in uniformly distributed small dots, as opposed to the larger clumps seen in the poor-performance sample. In addition, the spectrometer grade material exhibits diagonal lines of high transmission regions.

One concern in these types of measurements is the presence of surface irregularities, as these will strongly affect the transmission. These samples were polished initially, although it is unclear how the subsequent removal of the contacts affected the surface. However, by focusing through the sample with the collection lens system, it was found that the structure exhibited in the images shown here was in the bulk of the material.

The interpretation of these results is not yet complete. However, some explanations can be attempted. It may be that the predominant mechanism here is the effects of compositional variations. As the zinc fraction, x , increases in CZT the bandgap increases, from 1.44 eV for $x=0$, or CdTe, to 2.26 eV for $x=1$, or ZnTe. Regions of higher zinc, and thus larger bandgap, will be more transmitting to the broadband IR from the LED.

Particle Induced X-ray Emission (PIXE)

Particle (or proton) induced x-ray emission is a useful technique for probing the elemental composition of the near surface region of samples. In PIXE a focused ion beam is used to excite the elements in a sample, and the characteristic x-rays emitted during relaxation are measured. In this work, 3 MeV protons from a 10 MV tandem accelerator provided the excitation. These protons have a range on the order of 50 μm in the CZT. The x-rays of interest have half-thicknesses on the order of 10 μm , which sets the depth of material probed. A Si(Li) detector was used to characterize the x-ray emissions. Figure 2b shows the PIXE results next to their corresponding IR images. The darkness of a pixel indicates the ratio of Zn to Cd and Te fluorescence x-rays detected, and thus a darker pixel indicates a higher local amount of Zn. As can be seen in the figure, the spectrometer-grade sample exhibits more uniformity than the poor-performance detector. Although the system was not calibrated to determine the absolute

composition, the relative changes in composition across the region were computed. There was a $\pm 20\%$ variation about the mean for the spectrometer-grade detector versus a $\pm 30\%$ variation in the poor-performance sample. Consistent with the IR results, the PIXE maps show smaller clumps of higher zinc concentration for the spectrometer-grade sample, and larger clumps for the poor-performance sample, along with a left-to-right gradation. Further qualitative information is not apparent.

Photoluminescence (PL)

Photoluminescence (PL) is a useful tool for analyzing semiconductor materials. In photoluminescence the radiative recombination processes are viewed via optical excitation. In this work the excitation source was the 488 nm line of an argon ion laser. The luminescence was detected with a double-pass spectrometer coupled to a photomultiplier tube. Two sets of experiments were conducted. In the first set the samples were cooled in a liquid helium cryostat to around 4.2 K and the excitation not focused to give the low temperature PL response averaged over a large portion of the crystal. In the second set of experiments, the sample was held at room temperature on a precision translation stage and the beam focused to about 3 μm to give a detailed map of the room temperature PL.

Several peaks are visible in the near band-edge PL spectrum for CZT samples including free excitons, donor-bound excitons, and acceptor-bound excitons [6,7]. Fig. 3 shows a small region of the PL spectra near the bandgap from the spectrometer-grade and poor-performance detectors. A strong neutral-donor-bound exciton (D0-X) peak and a weaker neutral-acceptor-bound exciton (A0-X) peak are evident. The D0-X peak is much narrower in the spectrometer-grade sample than the poor-performance sample, and other features can be resolved. In terms of photon energy, the widths of the D0-X peaks, $\Delta E/E_0$, are 1.8×10^{-3} and 3.3×10^{-3} respectively. In general, the sharpness of the peaks in PL spectra relates to the "quality" of the crystal. Variations in the crystal structure, electrically-active impurity distributions, or other properties lead to broadening of the features. A particularly effective mode in CZT would be the composition variations. Since the Zn fraction (x) has a significant effect on the bandgap, changes in x result in shifts in the PL spectrum.

When looking at the integrated PL spectrum from a large area, these shifted responses combine to give the broadened features measured.

There have been several attempts to relate a measured material property with the exact zinc fraction, x . From PL it is possible to measure the bandgap to a high precision and accuracy and to detect small changes in the bandgap to the meV level. The difficulty has been in relating the bandgap to the zinc composition. There are several different fits reported by various researchers. As many factors affect the relation, it is difficult to determine which is universally "correct", as the fit depends on many properties of the particular material being studied. By correlating the PL measurements with the x-ray diffraction work discussed later in this paper, a fit that best represents the material studied here has been developed. Reference 6 discusses this fitting process in more detail.

Given the large uncertainty in the absolute value of the zinc composition, but the high precision in determining relative changes in x , the variation in x is more meaningful. In the second set of experiments, high resolution spatial maps of the relative zinc composition were measured for the two samples. Fig. 4 shows two such maps, taken over small (sub-mm) regions at small step sizes of $10\text{ }\mu\text{m}$. The spectrometer-grade detector has more uniformity in the zinc concentration than the poor-performance detector, as expected. The poor-performance one has a higher degree of mottling, or larger localized changes, as well as an overall gradual change from one side to the other.

Tri-axial X-ray Diffraction (TAXRD)

X-ray diffraction is a useful method for studying the crystallinity and lattice parameters of materials. In this work a high-resolution, tri-axial x-ray diffraction system was used. From the lattice parameters calculated from the XRD data, one can estimate the zinc composition. Here again, however, there is some uncertainty in the quantitative relation of this parameter to zinc composition.

For these measurements, a different set of comparable CZT detectors was used. Fig. 5 shows the XRD-measured zinc fraction at three points for the two samples. As can be seen, once again the poor-performance sample exhibits more zinc variation on the scale of the detector dimensions.

The tri-axial x-ray diffraction method allows for the separation of the diffraction peak broadening effects due to strain from changes in lattice parameter from those due to tilt from mosaicity. One can form the reciprocal space maps (RSM) of the samples which give a contour map of the intensity versus strain and tilt. Fig. 6 shows the RSMs of the (111) reflection for these two detectors. As can be seen, the poor-performance detector has a much higher amount of tilt than the spectrometer-grade detector. However, the spectrometer-grade detector does seem to exhibit a larger amount of strain, which would seem to indicate some level of compositional inhomogeneities. It appears that in this case the poor-performance detector is more limited by crystallinity problems.

Summary

In this work several materials characterization methods were performed on "good" and "bad" detectors to gain an understanding of the properties that limit the performance of large volume cadmium zinc telluride gamma-ray detectors. In particular, by using infrared transmission microscopy, particle-induced x-ray emission, photoluminescence, and tri-axial x-ray diffraction techniques it was found that poor-performance material exhibited larger variations in zinc composition and crystallinity than spectrometer-grade crystals. Efforts must be made to reduce these inhomogeneities if one wants to have high-quality large volume CZT gamma-ray spectrometers.

Acknowledgments

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Figure Captions

1. Spectrum of ^{137}Cs taken with a $1\text{ cm}^3\text{ Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ detector, showing the "hole-tailing" effect.
2. Infrared transmission microscopy images (a) and proton-induced x-ray emission maps (b) on the same $2.5\text{ mm} \times 2.5\text{ mm}$ region for spectrometer-grade and poor-performance CZT detectors. The IR images have been inverted, such that the dark regions indicate places of high transmission, while the PIXE image pixels are darker for a higher $\text{Zn}/(\text{Cd}+\text{Te})$ ratio.
3. Near band-edge low temperature (4.2K) photoluminescence spectra of the spectrometer-grade and poor-performance CZT detectors, and the width of the neutral-donor-bound exciton peak (D0-X) for each.
4. Maps of the relative zinc compositional changes in the spectrometer-grade and poor-performance detectors, as measured with room-temperature photoluminescence. The step size was $10\text{ }\mu\text{m}$ in the two dimensions.
5. Variation of the zinc fraction for the spectrometer-grade and poor-performance detectors, as determined from x-ray diffraction. The measurement points were spaced about 3 mm apart.
6. Reciprocal space maps of the spectrometer-grade and poor-performance detectors taken with tri-axial x-ray diffraction. The maps give a contour of the intensity versus the strain (change in lattice parameter from zinc variations) and the tilt (mosaicity in the crystal structure).

Spectrometer-Grade

(a)

Poor-Performance

(b)

Spectrometer-Grade
 $\Delta E/E_0 = 1.8 \times 10^{-3}$

Poor-Performance
 $\Delta E/E_0 = 3.3 \times 10^{-3}$

Spectrometer-Grade

Poor-Performance

Spectrometer-Grade

Poor-Performance

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Performances of CdTe and $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ nuclear radiation detectors at elevated temperatures

Abstract

This study of the performances of CdTe and $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ detectors at elevated temperatures, 25 - 70°C, shows that $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ has better performance as gamma-ray detector. However, ^{133}Ba spectra show that the quality factor, for the 81 keV peak, for $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ varies largely as temperature increases (decreasing from 1.12 at 25°C to 0.57 at 60°C), and CdTe has smaller variation (increasing from 0.19 at 30°C to 0.25 at 50°C and decreasing to 0.18 at 70°C). The polarization in peak position is much less in spectra from $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ than in spectra from CdTe.

I. Introduction

Cadmium telluride (CdTe) and cadmium zinc telluride ($\text{Cd}_{1-x}\text{Zn}_x\text{Te}$) are among materials being developed for several years now by researchers, as room-temperature semiconductor nuclear radiation detectors. These developments are being made to eliminate the inconveniences and cost of cooling earlier detectors, such as Ge and Ge(Li), to liquid nitrogen temperature. Thus the new detectors being developed must combine the advantage of room temperature operation and the efficiency of scintillators with the energy resolution of Ge detectors.

CdTe is the first material to have been developed as a room-temperature semiconductor detector.^{1,2} Its high Z (48 and 52 for Cd and Te respectively) is relatively large to provide a high stopping power for gamma and x-rays,³ and gives a higher gamma radiation detection efficiency than either Si or Ge.¹ It has a room temperature band gap of 1.45 eV that is wide enough to ensure low

leakage current under conditions of intrinsic resistivity, and the charge-carrier mobilities of about $1000 \text{ cm}^2/\text{Vs}$ for electrons and about $90 \text{ cm}^2/\text{Vs}$ for holes⁴ are high enough to achieve efficient charge collection.⁵

In recent times, researchers alloy CdTe with certain other materials to produce ternary compounds of higher band gaps and increase energy of defect formation.⁶ This is expected to lead to solid-state detectors with improved performance. In fact, Butler *et al.* recently reported⁷ that nuclear radiation detectors fabricated from $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ crystals grown by a high-pressure Bridgeman method exhibited significantly better performance than those fabricated from CdTe in terms of energy resolution and some other important operating parameters. There are several studies going on in our laboratory to obtain basic informations about this important new semiconductor radiation detector material. Results reported⁵ recently from our laboratory show that the room-temperature hole mobilities in $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ are on the average about 50% larger than hole mobilities in CdTe. The present study is aimed at investigating and comparing the performances of these two detectors, as gamma-ray (and x-ray) spectrometers, at elevated temperatures up to 70°C . These temperatures may be encountered in industrial applications, and in desert regions.

II. Experiment

CdTe crystals of sizes $2 \text{ mm} \times 2 \text{ mm} \times 2 \text{ mm}$ packaged in BNC connector housing were obtained from Radiation Monitoring Devices, Inc. (RMD) in Watertown, Massachusetts, USA. Polished $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ crystals (grown by a high pressure Bridgeman method) of sizes $2 \text{ mm} \times 2 \text{ mm} \times 1.5 \text{ mm}$ were obtained from Aurora Technologies Corporation (ATC) in San Diego, California, USA.

Contacts on the $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ crystals were made of graphite by painting the surfaces with a

graphite suspension in water ("aquadag" by Acheson, Inc.). Pt leads, 0.01 in. in diameter were attached to the contacts also by graphite suspension. The contacts with the attached parts of the Pt leads were covered with a protective coating ("humiseal" by Chase Corp.). This coating provides mechanical stability and is also transparent to gamma and x-rays. The CdTe detectors were test ready-made. The RMD's standard BNC connector provides protective housing for the CdTe crystals.

Figure 1 shows the schematic diagram of the experimental set-up. The small oven (6.5 cm x 3.6 cm x 4.0 cm) in which each detector is placed and irradiated by gamma-rays from ^{133}Ba , is made of aluminum and boron nitrate. It has electrical lead outlets which is connected directly, with the aid of BNC connectors, to an EG&G pre-amplifier just 3.5 cm away. The boron nitrate serves as a good electrical insulator as well as a good conductor of heat from the heater ("Minko" brand) to the detector.

The temperature of the oven at various stages of the experiment was monitored with copper-constantin thermocouple connected to a thermocouple thermometer ("Digi-Sense" Model No. 8528-20, of Cole-Parmer Instrument Company). The electrical signals generated from the exposure of the detectors to gamma radiation pass through the EG&G pre-amplifier and an Tennelect TC242 amplifier where the signals are amplified before they go on to the multichannel analyzer in the computer.

III. Results and Discussion

The spectra of ^{133}Ba obtained from the CdTe detector at 50 volts bias and the $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ detector at 350 volts bias are shown in Figs. 2 and 3 respectively, for various temperatures. As expected, the $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ detector has better energy resolution than the CdTe detector. This is evident from the values of the full-width-at-half-maximum (FWHM). Another indication of the

performance of gamma-ray spectrometer used in the comparison of these detectors is the quality factor, defined to be,

$$\text{quality factor} = [(\text{peak/valley ratio})/(\%FWHM)]^{1/2}$$

As shown in Fig. 5, the $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ detector has higher quality factor than the CdTe detector. However, the variation in quality factor with temperature changes is higher in $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ than in CdTe. The quality factor decreased from 1.12 at 25°C to 0.57 at 60°C in $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$. In CdTe, the quality factor increased from 0.19 at 30°C to 0.25 at 50°C and decreased to 0.18 at 70°C.

There is evidence of polarization in both detectors, resulting to shifts in energy peak positions, as shown in Fig. 6. The cause of this is mainly due to temperature changes. The changes in peak positions as temperature increases are shown in Table 1. The peak shift over the temperature range of 30 - 60°C is within 9 channels in CdTe and 2 channels in $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$. Thus, the $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ detector exhibit very small polarization compared to the CdTe detector. In fact, the peak position was steady for the $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ detector at temperatures 30 - 50°C.

IV. Conclusion

This experimental study of the performances of CdTe and $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ detectors has shown that the $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ detector has better energy resolution. Although the variation in quality factor is larger in $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ as temperature increases, it exhibits much smaller polarization in peak position than the CdTe detector.

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Table 1. Energy peak position at various temperatures.

Temperature (°C)	CdTe detector		Cd _{0.8} Zn _{0.2} Te detector	
	Counts/Channel	Channel Number	Counts/Channel	Channel Number
25	—	—	3068	64
30	980	68	2952	65
40	1243	71	2740	65
50	1315	78	2276	65
60	1282	76	1894	66
70	1444	73	—	—

Fig. 1.

Fig. 2: Spectra of Ba133 from CdTe

Fig. 3: Spectra of Ba133 from $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$

Fig. 4: Variation of FWHM with temperature
(Ba133 spectra)

Fig. 5: Variation of quality factor with temperature
(Ba133 spectra)

Fig. 6: Polarization in peak position
(a) Ba1 33 spectra from CdTe

(b) Ba1 33 spectra from $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$

Composition and performance mapping of CdZnTe nuclear spectrometers

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ABSTRACT

We have applied several techniques, including photoluminescence (PL), proton-induced x-ray emission (PIXE), photocurrent, and alpha particle response mapping, for mapping micron- and millimeter-scale variations in cadmium zinc telluride. We have correlated the degree of inhomogeneity determined by these techniques with performance of gamma-ray spectrometers fabricated from the material.

Keywords: photoluminescence, photocurrent, spatial mapping, alpha particle mapping, PIXE, CdZnTe, composition variation, gamma-ray spectrometers, nuclear radiation detectors

1. INTRODUCTION

Composition inhomogeneity may be a limiting factor in the performance of cadmium zinc telluride for nuclear spectrometer applications. In previous publications¹⁻³ we have discussed several mapping techniques for characterizing the spatial variation of composition and performance in $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ detectors and shown a marked contrast in all measures of uniformity between spectrometer-grade and poor-performance detectors. In this paper we discuss the application of these techniques to a larger set of detectors and show a correlation between the degree of inhomogeneity and detector performance.

2. EXPERIMENTAL

Experimental details for low-temperature photoluminescence, room-temperature PL mapping and photocurrent mapping have been described in references 1-3, and the alpha particle mapping system will be described in a future publication. Only a few relevant facts are repeated here.

The crystal was grown by high-pressure Bridgman with a nominal zinc fraction of 0.1. For most of the photocurrent and PL mapping, the laser beam was focused to a spot diameter of approximately 25 μm , and the maps were obtained in either 25 or 50 μm increments. In one case, described in section 3.1, the beam was focused to 3 μm , and the map was obtained in 10 μm increments. For low-temperature PL, the laser spot was not focused, so that the spectrum was averaged over a substantial fraction of the sample area. The detector thickness was approximately 2.5 mm, and the contact area was between 10 and 30 mm^2 . For alpha-particle mapping, the source was collimated to a spot size of 1 mm. Pulse-height spectra for ²⁴¹Am were collected using a charge-sensitive preamp and a shaping amp with shaping time of 1 μs .

3. RESULTS AND DISCUSSION

3.1 PL mapping results

We have obtained room-temperature PL maps of composition variation in increments as small as 10 μm . Figure 1 shows an example in which the high spatial and spectral resolution reveal features that are not detected by simple intensity mapping or by large-scale composition mapping. Figure 1(a) is a composition map showing minute growth striations; Figure 1(b) is a simultaneously obtained intensity map of the same area. The striations are not obvious in the intensity map; further, the intensity map is affected by scratches and other surface features that are not related to the underlying material quality. Note that the dark bands (higher zinc regions) are only about 20 μm wide - high spatial resolution is necessary to detect them. In addition, the maximum composition variation is only about 10% of the mean value - good spectral resolution is also necessary.

Figure 1: Room-temperature PL maps of (a) composition and (b) maximum intensity. The composition map clearly shows minute striations that are not apparent in the intensity map. The maps were obtained simultaneously in 10 μm increments.

3.2 Correlation of PL measurements with detector performance

To minimize variation in material parameters among samples, we selected 8 samples from two adjacent columns of a lengthwise slice of a boule and performed low-temperature PL and room-temperature PL mapping on them. We then sputter-deposited gold contacts, measured I-V characteristics and collected gamma-ray spectra using an ^{241}Am source. The low-temperature PL spectra, room-temperature PL maps, I-V curves and pulse-height spectra for 5 samples from a single column are shown in Figures 2 (a) through (d). Although the samples came from virtually the same position along the growth axis of the boule and have nearly equal zinc concentrations as indicated by the position of the donor-bound-exciton (D^0X) peak, the degree of composition variation as indicated by the room temperature PL maps and by the width of the (D^0X) peak varies substantially, as does the detector performance. The I-V curves are similar, with all of the detectors exhibiting increasing resistance with increasing voltage, a characteristic which has been shown to be favorable⁴. One reason for the generally poor quality of the pulse-height spectra is that the contacts covered only a fraction of the sample area, and the source was not collimated, so that charge carriers generated near the edge of the contact were subject to a weaker electric field than those that were generated near the center.

Table 1 summarizes the results of these measurements on all 8 samples. The first column gives the width of the (D^0X) peak in the 4.2K PL spectrum. This peak width is dominated by alloy broadening in CdZnTe and so can be taken as a measure of composition variation over the area probed by the laser spot. The second column gives the standard deviation of the zinc fraction determined by room-temperature PL mapping, as a percentage of the mean. The third column gives the peak-to-valley ratio for the 59.5 KeV photo peak in the ^{241}Am gamma-ray spectrum.

Table 1: PL Results and Detector Performance for Samples from Two Adjacent Columns of the Boule Slice

Sample ID	4.2K PL Peak Width	300K PL Variation	59.5 KeV Peak/Valley
10D2	0.76	1.26	3.08
10A2	0.77	1.81	2.76
10D1	0.92	2.35	1.96
11C1	1.02	1.79	3.08
11B2	1.06	4.49	1.98
11C2	1.09	2.68	1.65
11B1	1.19	2.58	1.34
11D1a	1.17	3.43	~1

Figure 2: (a) Low-temperature PL spectra, (b) room-temperature PL maps, and (c) I-V curves for 5 samples from a single column of the boule slice.

Figure 2 (d) : Pulse-height spectra for 5 samples from a single column of the boule slice.

The principal question which this paper addresses is whether the data of columns 1 and 2 in Table 1 can be used as predictors of detector performance. Figures 3 (a) and (b) show plots of detector performance vs. these two predictors; with the exception of a single outlying point in each case, there is clearly a direct relation.

Figure 3 : Detector performance vs. composition variation. The dashed lines are only to guide the eye.

3.2 PHOTOCURRENT MAPS

Photocurrent maps using visible light for three of the detectors are shown in Figure 4. In all cases the lower contact was biased negative, so the photocurrent is greatest when the sample is illuminated near the bottom. The map of Figure 4 (a) most closely resembles the ideal situation of horizontal contours, suggesting a relatively uniform electric field. Not surprisingly this detector had the best performance of the three. Figure 4(b) exhibits a decreasing electric field as one moves from left to right, due to reduction in the electric field near the edge of the contact. Figure 4(c) has "hot spots" or high-photocurrent regions near the negative contact. We have observed this phenomenon frequently and believe that it is related to space charge concentrations.

Figure 4: Photocurrent maps for three detectors from one column of the boule slice. (a) shows the nearly-horizontal contours that one would expect for a uniform electric field. (b) shows a reduced electric field as one moves left to right. (c) shows hot spots near the negative contact, possibly due to space charge.

We have shown in a separate paper⁵ that it is possible in some cases to extract an estimate for the electron mobility-lifetime product from one-dimensional sections through these two-dimensional maps. The $\mu\tau$ values obtained are in reasonable agreement with typical published values for CdZnTe.

3.3 ALPHA PARTICLE RESPONSE MAPS

While the alpha particle mapping technique is rather new, we have produced preliminary results which suggest the potential of the technique to study material inhomogeneities. Figure 5 shows two coarse (1 mm steps) 5x5 maps for separate detectors from the same column of the boule slice. These maps do not have sufficient spatial resolution to provide much information about variation of material parameters, but the effect of the reduced electric field near the edges of the circular contact is clearly visible. It is significant that the detector on the left, which performed better than the one on the right (see Table 1), has a lower overall average rise time and less variation, even near the center of the contact.

Figure 5: Alpha particle maps for two detectors from one column of the boule slice. The effect of the reduced electric field near the edges of the circular contact is apparent. (bias = 100V, 1 mm steps, 50 pulses averaged)

3.4 Correlation between PIXE and PL maps

We have attempted to demonstrate a correlation between composition maps determined by PIXE and those determined by photoluminescence. Aligning the two maps is in general an exceedingly difficult problem; one case in which it has proved manageable is for a sample containing Te precipitates, which serve as alignment markers. A PL single-wavelength intensity map for a sample containing Te precipitates is shown in Figure 6. The dark cross-hairs are metal wires which were attached to the sample to provide a point of reference. The dark, winding islands are Te precipitates, which are also visible in the PIXE map of tellurium concentration shown in Figure 7. The two maps are of the same sample, but not necessarily the same area. More work is needed to correlate PL composition maps with PIXE maps.

Figure 6: PL Intensity map showing Te precipitates

Figure 7: PIXE map showing Te precipitates

Uniformity of $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ grown by high-pressure Bridgman

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Abstract

We have employed both low-temperature photoluminescence (PL) spectroscopy and high-spatial-resolution, room-temperature PL mapping to determine composition variation in $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ grown by high-pressure Bridgman. Composition variations Δx of approximately 5–10% are observed between tip and heel of an approximately 13 cm long boule, while fluctuations of 1–2% are observed within a single, detector-scale sample. We also show that there are great discrepancies in the calculated zinc concentration, depending on which expression for $E_g(x)$ is chosen from the literature. We have performed high-resolution, triaxial X-ray analysis on selected samples to determine which of the published relations is most accurate for our material. We have also examined the relationship between the low-temperature PL spectrum and detector performance, and found that the line width of the donor-bound-exciton peak can be used as a predictor of performance.

1. Introduction

Cadmium zinc telluride shows promise for room temperature X-ray and γ -ray detection and spectroscopy in a variety of applications. It possesses some distinct advantages over CdTe, notably a wider bandgap, which reduces the leakage current and a lower density of vacancies, due to the lower vapor pressure of Zn relative to Cd [1]. However, the alloy nature of the material raises concerns about possible detrimental effects on detector performance due to composition nonuniformity. In this paper we investigate the variation of zinc concentration in $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ grown by high-pressure Bridgman, both on a large scale using low-temperature photoluminescence spectroscopy and on a small scale using room-temperature PL mapping. We discuss the correlation between the composition variation as determined from the PL spectra and detector performance.

2. Experimental

The crystal was grown by a high-pressure Bridgman method with a nominal zinc fraction of 0.1. A section of the ingot approximately 2 mm thick was cut along the boule axis; this slice was then cut into strips approximately 1 cm wide. Each strip was divided into samples approxi-

mately 1–2 cm² in area. Each sample was etched for 7 min in a 0.5% bromine–methanol solution.

For low-temperature photoluminescence measurements, samples were cooled with liquid helium in a variable temperature cryostat and illuminated with the 488 nm line of an argon ion laser. The beam was not focused, so that the laser spot excited an area of several square millimeters; hence the low-temperature spectra are effectively averaged over a substantial fraction of the sample area. The emitted radiation was detected using a 0.85 m double-pass spectrometer coupled to a photomultiplier tube with an S20 response. The signal was either chopped at 750 Hz and analyzed with a lock-in amplifier, or analyzed with a photon counter.

The instrumentation for room-temperature photoluminescence was identical to that described above. To provide a two-dimensional mapping capability, the sample was attached to an X–Y translation stage with 0.1 mm stepping capability. The laser beam was focused to a spot diameter of approximately 3 μm using a microscope objective, which also collected the emitted radiation.

3. Results and discussion

Fig. 1 shows a high-resolution spectrum of the near band edge region for a $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ sample of particularly high quality. The peaks have been identified [2] as due to free excitons in their ground ($X_{n=1}$) and first excited

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Fig. 1. High-resolution, 4.2K PL spectrum (near band edge region) for a very high-quality Cd_{1-x}Zn_xTe sample.

(X_{n-2}) states, upper polariton states (X_{up}) and excitons bound to neutral donors (D^0-X) and neutral acceptors (A^0-X). The narrow lines observed in this spectrum, showing well-resolved X_{n-1} and X_{n-2} peaks, permit calculation of the free exciton binding energy. Assuming hydrogen-like energy levels:

$$E(X_n) = E_{gap} - \frac{E_1}{n^2}$$

where E_1 is the ground-state free-exciton binding energy, yields a value of 10.8 meV for E_1 , which agrees well with previously reported results [3]. The dissociation energy for an exciton bound to a neutral donor is determined from Fig. 1 to be 2.2 meV; thus the D^0-X peak lies 13.0 meV below the bandgap.

The width of the D^0-X peak in the low-temperature PL spectrum can be taken as a predictor of detector performance. Fig. 2 shows a comparison of the near band edge spectra for material of poor, spectrometer grade and excellent quality. There is a clear relation between the narrowness of this peak and detector performance. The broadening of the peak in the poor performance sample may be due to small-scale variations in the composition, as discussed further below.

Fig. 3 shows the position of the D^0-X peak vs. distance from the boule tip (first to freeze region) for samples near

poor performance $\Delta E/E_0 = 3.3 \times 10^{-3}$ spectrometer grade $\Delta E/E_0 = 1.8 \times 10^{-3}$ high performance $\Delta E/E_0 = 0.78 \times 10^{-3}$

Fig. 2. Near band edge region of low-temperature PL spectra for samples with varying performance as radiation detectors

Fig. 3. Variation of D^0-X peak position with distance from boule tip.

the boule axis. Since this peak closely tracks the bandgap, it is clear that the bandgap of the material and hence the zinc concentration, decreases with distance from the tip. This fact is a consequence of the segregation coefficient for Zn in CdTe being greater than one, that is, the zinc concentration in the solid phase exceeds that in the melt, leading to depletion of zinc from the melt and progressively lower zinc concentration in the crystal.

Several groups have published correlations between the low-temperature PL spectrum and composition of Cd_{1-x}Zn_xTe [4–8]. These relations are of the form [9]:

$$E(x) = E_0 + (\Delta E - a)x + ax^2$$

where ΔE is the difference in bandgap between the two alloyed materials and a , which is sometimes called the “bowing parameter”, indicates the degree of nonlinearity in the dependence of the bandgap on composition. The reported values of the bowing parameter for Cd_{1-x}Zn_xTe vary from 0.166 eV [6] to 0.463 eV [5]. Since there are many possible reasons for this discrepancy and no obvious reason to favor one result over another, we have performed measurements on selected samples to determine which relation is most accurate for our material. We have performed high-resolution, tri-axial X-ray diffraction analysis to determine the lattice constant and determined the zinc fraction of the samples from Vegard’s law:

$$a(x) = a_1 + (a_2 - a_1)x$$

where $a(x)$ is the lattice constant of the alloy and a_2 and a_1 are the lattice constants of the two constituent materials. We have plotted the zinc fraction determined in this way vs. the D^0-X PL peak position; our results (see Fig. 4) suggest that the relation of Magnea et al. [4] is most appropriate for our samples.

We have also compared our results for the molar zinc fraction as a function of distance along the boule axis with that expected from the normal freeze equation [10]:

Fig. 4. D0-X peak position vs. molar zinc fraction according to various relations from the literature with experimental points.

$$C = C_0 K(1 - g)^K - 1$$

Here C is the zinc concentration in the solid, C_0 is the initial zinc concentration in the melt, K is the segregation coefficient and g is the solidified mole fraction. Tanaka et al. [11] have determined the value of K to be 1.35. Using this result along with $C_0 = 0.1$ yields the curve shown in Fig. 5, which again leads us to believe that Magnea's relation is most accurate for our material.

The near-band-edge, room-temperature PL spectrum for $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ consists of a single broad band. The shape of this spectrum has been shown to be due only partially to band-band recombination, with some contribution from excitonic transitions and their phonon replicas [12].

Other researchers have demonstrated the efficacy of photoluminescence mapping to examine composition variations in $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ [13–15], but they have concentrated on wafer-scale mapping, using relatively large spatial increments. We have performed one- and two-dimensional mapping of detector-scale samples, using increments as small as $10\ \mu\text{m}$. The mapping procedure entails determining the room-temperature PL peak position at each point

Fig. 5. Variation of zinc concentration from tip to heel - theoretical vs. experimental using various relations from the literature.

Fig. 6. Small-scale variation of zinc concentration for a sample near the heel of the boule. In the two-dimensional map, dark areas correspond to regions of high zinc concentration.

and calculating the zinc concentration using the relation [8]:

$$E_{\text{peak}} = 1.51 + 0.606x + 0.139x^2 \text{ eV}$$

Since it is known that the peak energy depends not only on the composition but also on the type of material and the recombination processes occurring, this relation may not be correct in determining zinc concentration in our material. Thus we have expressed the data in terms of variation of zinc concentration to reduce the sensitivity to inaccuracy in the expression for $E(x)$.

Fig. 6 shows a one-dimensional plot of Δx vs. position for a sample near the heel of the boule. In addition to the underlying systematic decrease, as in Fig. 5 above, there is a small-scale variation showing "islands" of high zinc concentration. Fig. 6 also shows a two-dimensional composition map for the same sample, which exhibits the same small-scale composition variation and rules out the possibility that the fluctuations in the one-dimensional plot are due to growth striations.

4. Conclusions

We have characterized the composition variation in high-pressure Bridgman-grown cadmium zinc telluride using low-temperature and room-temperature photoluminescence mapping. Composition variations Δx of approximately 5–10% are observed between tip and heel of a boule, while smaller composition fluctuations are

observed across the cross-section of the boule. Fluctuations of 1–2% are observed within a single, detector-scale sample; this variation may have undesirable consequences, especially in imaging applications. While both techniques track composition variations precisely, knowledge of the absolute zinc concentration requires an accurate relation between the molar zinc fraction, x , the band gap energy, E_g , and the lattice parameters. We have used high-resolution, triaxial X-ray diffraction to determine that the relationship published by Magnea et. al. [4] is relatively accurate for our material. By comparing the low-temperature PL spectra of samples with varying performance as radiation detectors, we have found that the narrowness of the donor-bound-exciton peak in the low-temperature PL spectrum can be used as a predictor of detector performance.

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The Use of Pulse Processing Techniques to Improve the Performance of $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ Gamma-Ray Spectrometers

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Abstract

$\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ (CZT) has recently shown great promise for use as a room temperature gamma-ray detector material. The availability of large volume ($> 1 \text{ cm}^3$) high resistivity CZT crystals has allowed the demonstration of detectors much larger than can be built with the similar material CdTe. However, CZT — like many other room-temperature materials — suffers from the poor transport properties of holes. The poor hole drift properties of CZT cause the characteristic “hole tailing” in gamma-ray pulse height spectra. We have applied pulse processing methods to reduce the hole tailing effects and improve the energy resolution of CZT detectors. We have used two signal processing methods to reduce hole tailing in CZT detectors: *digital rise-time compensation* and *dual time-constant sampling*. We discuss the implementation of these techniques, demonstrate results obtained in the laboratory, and compare the performance obtained with other detector systems.

I. INTRODUCTION

For many years room-temperature semiconductor detectors have shown great promise for use in gamma-ray spectrometers because of their relatively good energy resolution compared with scintillation spectrometer systems. However, the good energy resolution that has been obtained with room-temperature semiconductor devices was obtained from small devices. Furthermore, analysis of the charge collection behavior of these devices, and experimental measurements, indicates that the *effective volume* of the devices is typically much smaller than their physical volume (usually by more than a factor of ten) due to “hole-tailing” effects. For instance, a typical room-temperature detector might be reported as having an energy resolution of 7 keV at 662 keV but further analysis of the pulse height spectrum will reveal that only about 5 % of the “photo-peak” events actually lie within a 7 keV window centered at 662 keV. For many applications it is not only important to have better energy resolution than scintillators but also to have substantial active detector volume (to improve gamma-ray sensitivity through both photoelectric and multiple Compton interactions).

For applications requiring good sensitivity and energy resolution for energetic photons ($> 200 \text{ keV}$), room temperature semiconductors do not often fare well. The situation is illustrated graphically in Fig. 1. The aim of the

work described in this paper is to employ methods for increasing the energy resolution of room-temperature semiconductor detectors while maintaining as large an active volume as possible. To date, our best results have been obtained using pulse processing methods applied to CdZnTe detectors, but we recognize that other approaches also appear very promising [1]. The details of the results we have obtained using two different pulse processing methods applied to large volume CdZnTe detectors are described below.

Fig. 1. Range of detector volumes and energy resolutions obtainable with various gamma-ray spectrometers.

II. DIGITAL RISE-TIME COMPENSATION

For some time [2] it has been recognized that there is a correlation between the rise-time of a signal from room-temperature semiconductor materials and the deficit in its pulse height due to charge loss. Using this correlation it is possible to identify pulses which correspond to less than complete charge collection. Once incomplete charge pulses are identified, they may either be rejected (pulse height discrimination) or gain corrected and added to the pulse height spectrum (pulse height compensation). Generally, a

combination of these techniques is used with a trade-off obtained between the energy resolution and efficiency of the detector.

The apparatus we have used to implement rise-time compensation is diagrammed in Fig. 2. Our apparatus differs from the technique implemented by Richter [3] in that all discrimination and compensation is performed digitally. Furthermore, our implementation differs somewhat from the digital systems described recently [4] in that we use two different shaping amplifiers. Two amplifiers were used because the optimal shaping time (in terms of signal to noise) is not necessarily the same for the rise-time measurement as it is for the amplitude determination.

Fig. 2. Apparatus used to perform rise-time compensation experiments. The results produced by analog to digital converter #1 (ADC1) were polled in coincidence with ADC2.

Some data obtained using this apparatus is shown in Fig. 3. Compensation was implemented by empirically fitting a polynomial function to the measured relationship between pulse height and rise-time; the pulse height spectrum obtained at each rise-time was then shifted according to this empirical relation. Fig. 4 illustrates an amplitude versus rise-time data set after it has been corrected; subsequently summing the shifted data produces a compensated pulse height spectrum. Simultaneous discrimination and compensation can be implemented by limiting the number of pulse height spectra (at different rise times) that are summed. Figs. 5 and 6 illustrate some results using this combination of compensation and discrimination methods; a trade-off is obtained between the efficiency of the detector and the energy resolution that is obtained. However, this tradeoff is always far more desirable than the results obtained with no pulse processing. Fig. 7 illustrates a ^{137}Cs pulse height spectrum that would be obtained by summing all of the data in Fig. 3 without any compensation. Fig. 8 summarizes the performance we obtained with digital rise-time compensation with the 1.62 cm^3 detector described earlier.

Fig. 3. Typical data set obtained from the rise-time apparatus. The plot depicts a series of pulse height spectra obtained at different rise-time values. A 1.8 cm x 1.8 cm x 0.5 cm $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ detector operated at 1000V bias and irradiated with a ^{137}Cs source was used.

Fig. 4. Rise-time data set of Fig. 3 after a rise-time compensation algorithm has been applied to it. An empirical polynomial fit was made to the photo-peak height versus rise-time data from Fig. 3. The polynomial was then used to shift the data from longer rise-times.

Fig. 5. Pulse height spectrum obtained by irradiating a 1.8 cm x 1.8 cm x 0.5 cm $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ detector with a ^{137}Cs source and applying pulse height compensation. The width of the 662 keV photopeak is 1.7 % FWHM and 30 % of the volume of the detector was sampled in the experiment.

Fig. 7. Pulse height spectrum obtained by irradiating a 1.8 cm x 1.8 cm x 0.5 cm $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ detector with a ^{137}Cs source with no pulse height compensation. The detector was operated at a bias of 1000 V.

Fig. 6. Pulse height spectrum obtained by irradiating a 1.8 cm x 1.8 cm x 0.5 cm $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ detector with a ^{133}Ba source and applying pulse height compensation. Approximately 20 % of the volume of the detector was sampled. Note that the algorithm used to compensate the data (which was calibrated at 662 keV) remains linear; all of the ^{133}Ba peaks are in their correct relative position.

Fig. 8. Summary of the performance of the rise time compensation method applied to a 1.62 cm³ $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ detector. The fraction of the detector volume used was estimated by integrating the pulse height spectrum from above the backscatter peak and comparing that integral with the uncompensated spectrum. FW0.1M refers to the full width of the 662 keV photo-peak at one tenth of its maximum amplitude.

III. DUAL TIME-CONSTANT SAMPLING

We have recently implemented a pulse processing system with much simpler hardware requirements than rise-time compensation. The technique, which we call dual time-constant sampling, relies on simultaneously digitizing the peak of pulses shaped with both a "short" and a "long" integration time. A diagram of the apparatus we use to implement this technique is shown in Fig. 9.

Qualitatively, the dual time-constant apparatus may be viewed as a system that extracts information about the electron component of the signal through the short time-constant amplifier, and extracts information about both the electron and hole motion through the long time-constant amplifier. Thus, with a knowledge of the pulse height from both the short and long term signals it is possible to deduce the degree of trapping associated with a given pulse. A related technique (but using different methods to sample the signal) has been tried with some success with HgI_2 detectors [5]. Apparently the related method was not widely adopted for use with HgI_2 detectors because it only worked with a few specific detectors and was not generally applicable.

Very recently Heanue, et al. [6] have used hardware similar to the type described here to process signals from a CZT detector. However, Heanue, et al. apparently did not find this technique practical for use in their particular application (SPECT).

Fig. 10. illustrates a typical data set obtained from our dual time-constant apparatus from a CdZnTe detector irradiated with a ^{137}Cs source. The detector was the same as described previously (1.8 cm. x 1.8 cm x 0.5 cm) and operated at 1000 V. The data in Fig. 10 were obtained using a short shaping time of 0.25 μs and a long time constant of 3 μs . The short time constant was chosen as slightly greater than the estimated maximum drift time of electrons in the device (0.23 μs). The long time constant was chosen to be greater than the estimated hole trapping time (1 μs). The data shown in Fig. 10 clearly shows that the trapped signals have a different ratio of short/long amplitudes. Pulses that have the same amplitude at both short and long time constants correspond to full charge collection events. Conversely, events that give rise to pulses with a different amplitude at short and long time constants, exhibit incomplete charge collection.

An algorithm was developed that transformed the data obtained from the dual time-constant experiment into a form analogous to the data obtained from the rise-time experiment. Once these data were corrected they could be summed (or selectively summed) in a manner similar to the method used with the rise-time compensation experiment. Some results obtained using this method are shown in Fig. 11 and Fig. 12. It should be noted that the algorithm we used to compensate the dual time-constant data would be simple to implement for real-time use in hardware; a simple digital circuit that determined the ratio of the output of the slow channel and fast channel ADCs would be used to provide an address to increment memory (as opposed to a conventional ADC circuit where the ADC output itself is used to increment memory).

A summary of the data obtained using the dual time-constant method is shown in Fig. 13. Although the best energy resolution obtainable with the dual time-constant technique is slightly worse than that obtainable with the rise-time compensation method; the method seems to give more symmetric peaks (as evidenced by the nearly constant ratio of FWHM to FW0.1M for much of the data). Furthermore, the dual time-constant technique is much simpler to implement in hardware and would be a better choice for a portable system.

Fig. 9. Apparatus used to perform dual time-constant sampling experiments. The results produced by analog to digital converter #1 (ADC1) were polled in coincidence with ADC2.

Fig. 10. Contour plot of typical data set obtained from the dual time constant sampling experiment.

Fig. 11. ¹³⁷Cs pulse height spectrum produced using the dual time-constant method compared with unprocessed pulse height spectra from the fast (0.25 μ s) semi-Gaussian shaping amplifier and the slow (3 μ s) shaping amplifier channels. The dual time-constant processed spectrum used 54 % of the counts from the detector. Note that the effective photopeak efficiency in the processed data is actually greater than in the unprocessed spectra, despite the fact that approximately one half of the events were rejected.

Fig. 12. ¹³⁷Cs pulse height spectra obtained using the dual time-constant technique for three different values of the event fraction. With this method the event fraction corresponds approximately to the effective volume of the detector.

Fig. 13. Summary of the performance of the dual time-constant method applied to a 1.62 cm³ Cd_{0.9}Zn_{0.1}Te detector.

IV. SUMMARY

We have investigated the use of pulse processing circuits to improve the performance of large volume (> 1 cm³) CdZnTe spectrometers. Substantial improvements in the performance of CZT devices were made (in terms of energy resolution and lack of spectrum distortion) using these techniques and at little expense in terms of device efficiency.

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Large volume room temperature gamma-ray spectrometers from $\text{Cd}_x\text{Zn}_{1-x}\text{Te}$

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Abstract

Recent developments in $\text{Cd}_x\text{Zn}_{1-x}\text{Te}$ (CZT) crystal growth technology have enabled the production of large samples of detector grade material. In this paper we discuss the technical issues associated with building large volume CZT detectors ($> 10 \text{ cm}^3$) for use in gamma-ray spectroscopy. In particular, an analysis of the electronic noise in CZT devices and how it scales with detector size is presented, along with a discussion of methods to reduce noise in large CZT detector systems. The degradation of energy resolution caused by charge collection problems is also discussed and the use of electronic signal processing and improved device design to minimize charge collection effects are described.

1. Introduction

Gamma-ray spectrometers are widely used in many applications. The most common gamma-ray spectrometer detector systems used in industry and research are scintillation detectors coupled to photo-multiplier tubes, and cryogenically cooled high purity germanium detectors (HPGe). HPGe detectors have excellent resolution and good efficiency but cannot be used in some applications due to the inconvenience of their cryogenic cooling. Scintillation spectrometers are currently the most widely used detectors when room temperature operation is critical, but their poor energy resolution limits their utility in many applications.

We are investigating the use of CZT detectors as a replacement for scintillation spectrometers. To be useful in a range of applications, a CZT spectrometer should have better energy resolution than a scintillation spectrometer ($\sim 1\%$ FWHM at 662 keV), yet have reasonable efficiency (10 cm^3 effective detector volume). Such performance should be achievable with incremental improvements over existing systems. In this paper we discuss the performance of existing CZT detectors, how their performance scales with detector volume and what improvements might be expected from these detectors.

The energy resolution of existing CZT detectors is currently limited by two factors: electronic noise and charge collection effects, and both of these effects are discussed in detail below. At the current level of CZT technology, the broadening of gamma-ray spectra due to

the statistics of charge collection (and modified by the Fano factor) is not an important factor in determining the energy resolution of CZT detectors.

2. Electronic noise

As with all other room-temperature semiconductors, electronic noise can be an important factor in determining the energy resolution of CZT gamma-ray spectrometers. The dominant electronic noise sources in a CZT detector system are the parallel white noise [1] due to the dark current flowing through the detector and the series white noise originating in the JFET of the readout preamplifier (but controlled by the capacitance of the detector). For the present discussion, we will ignore the contribution of “ $1/f$ ” noise sources in our detector system; suffice it to say that we expect the contribution of $1/f$ sources to be less than 10 electrons/pF of detector capacitance, and this contribution will be small compared to other noise sources.

The parallel white noise arising from dark currents in the detector may be written as [1]:

$$\text{ENC}_p = \sqrt{\frac{I_d \tau}{q}} \quad (1)$$

where ENC is the equivalent noise charge in units of electrons, I_d is the dark current in the detector, and τ is the integration time of the linear amplifier. This expression may be modified slightly depending on the transfer function of the linear amplifier (e.g.: triangular or semi-Gaussian). The above expression for parallel noise is only valid for pulsed reset systems. In continuous feedback charge preamplifiers the parallel noise component is increased by

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a factor of $\sqrt{2}$ due to the continuous reset current applied to the gate of the FET (the fluctuations in the feedback current are uncorrelated with detector leakage current and thus must be added in quadrature to the shot noise). The expression for parallel noise is more convenient if it is expressed in conventional spectroscopy units (keV FWHM) by multiplying by ϵ_{pair} , the mean energy required to create an electron hole pair, and a numerical factor:

$$\Gamma_p = \text{ENC}_p \cdot \epsilon_{\text{pair}} \cdot 2.35 \quad (2)$$

where Γ_p is the broadening due to parallel noise in units of keV FWHM. If we assume that the CZT detector is of a planar geometry, and exhibits "Ohmic" behavior, then the dark current is easily predicted:

$$I_d = \frac{VA}{\rho d} \quad (3)$$

where d and A are the thickness and area of the detector, V is the potential difference applied to the (parallel) contacts of the detector, and ρ is the resistivity of the $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ used to construct the device. Eq. (3) may be substituted into Eq. (1) to predict the parallel noise as a function of detector dimensions and operating bias.

The other major electronic noise source in a typical CZT detector system is the series white noise contribution. The series noise originates as Johnson noise in the channel of the field effect transistor (FET) used to read out the detector. FETs are typically used in the charge sensitive preamplifier that reads out the detector because they are the lowest noise amplifying device [1] available. Although this parallel noise source arises in the FET, its influence on detector energy resolution is affected strongly by the capacitance of the detector. The magnitude of the FET series noise may be expressed as:

$$\text{ENC}_{\text{series}} = (C_g + C_d + C_{\text{stray}}) \sqrt{\frac{4kT}{\tau g_m}} \quad (4)$$

where C_g is the capacitance of the gate of the input FET, C_d is the capacitance of the detector, C_{stray} is the stray capacitance associated with the connection of the amplifier to the detector, g_m is the transconductance of the readout FET and τ is the integration time of the amplifier. The series noise would be expected to increase with detector area as the capacitance of the detector also increases with area. However, if large capacitance detectors are used, larger transconductance FETs (or parallel FETs) may be used to reduce the series noise somewhat.

The total resolution broadening due to noise should be (ignoring $1/f$ sources) the quadratic sum of the series and parallel sources. For a planar geometry CZT ohmic detector operated at a fixed bias, the total noise increases with the square root of the detector area, but decreases with the square root of the detector thickness. If there were no additional constraints, noise calculations alone would

lead one to believe that it is desirable to make detectors very thick relative to their contact area. However charge collection effects tend to degrade rapidly with detector thickness (approximately with the square of the detector thickness). If we assume that reasonable charge collection can be achieved with a 1 cm thick CZT device operated at 1000 V, we can constrain the thickness parameter and predict the amplitude of the noise we would expect as a function of CZT detector volume.

Fig. 1 shows the estimated electronic noise contribution to energy resolution broadening of three CZT detectors of 1, 10 and 100 cm^3 volume as a function of readout integration time. To compute the results shown in Fig. 1 we assumed that all detectors were 1 cm thick and operated at a bias of 1000 V, we also assumed that the resistivity of the material was $2.5 \times 10^{11} \Omega \text{cm}$ and the mean energy required to create a charge pair was 5.0 eV; these values are consistent with measurements obtained in our laboratories as well as those reported by others [2] for $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$. In computing the results shown in Fig. 1 it was also assumed that conventional readout electronics were used: a g_m of 6 mS and an input capacitance of 2.5 pF were assumed for the readout FET (typical of a commercial 2N4416 type device), and a stray capacitance of 10 pF was assumed in the calculations to account for typical interconnections. Also shown in Fig. 1 are the maximum electron and hole transit times (based on electron mobility of $1350 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$ and a hole mobility of $120 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$ [2]); these numbers are important because they provide an estimate of how long the signal must be integrated to collect all of the charge. If we use an electron-only device design, an integration time at least 0.73 μs will be required. If the device design requires that both the electron and hole components are collected, a

Fig. 1. Estimated electronic noise versus amplifier integration time for CZT detectors of three different volumes. It was assumed that all detectors were 1 cm thick and operated at a bias of 1000 V.

collection time of 8.3 μs would be needed to collect all of the charge from every region of the detector.

If noise alone were the only source of resolution broadening in CZT detectors, relatively high performance gamma-ray spectrometers could be built from this material. For instance, examination of Fig. 1 indicates that a 10 cm^3 device could have approximately 1% energy resolution (7 keV FWHM) for ^{137}Cs (662 keV). Although the resolution of existing small volume CZT detectors ($\approx 1\text{ cm}^3$) are not limited by electronic noise, it is useful to consider methods to reduce the electronic noise for future large volume devices. The series noise could be reduced somewhat when reading out large area detectors by using FETs that are matched to the detector's capacitance and also reducing the stray capacitance. Fig. 2 illustrates the effects such improvements might make on a 10 cm^3 device (10 cm^2 area by 1 cm thick). The calculations used to generate Fig. 2 assumed that the FET input capacitance was the same as the detector's capacitance, the stray capacitance was zero, and the ratio of the transconductance to the input capacitance of the FET were the same as a 2N4416 device (2.4 mS/pF). Fig. 2 indicates that some reduction in overall noise level would result from an optimized readout system, however it is also clear from the figure that the dominant noise source is the parallel noise arising from the leakage current in the detector. It might be possible to reduce the leakage current in the device by utilizing a blocking contact structure. If nearly perfect diodes could be constructed, the leakage current could be reduced to a level where it is dominated by generation currents from the volume of the depletion region. In such a perfect diode, we estimate the leakage current from a reverse biased CZT

Fig. 2. Estimated electronic noise versus amplifier integration time for a CZT detector of 10 cm^3 volume (1 cm thick and operated at a bias of 1000 V). The line marked "total" is the quadratic sum of the parallel and series noise components for a typical readout system. The line marked "optimized total" is an estimate of the noise broadening if an optimized FET, with zero stray capacitance were used to readout the detector.

device would be less than 1 pA cm^{-3} of depleted detector volume (assuming a generation-recombination life-time of 1 μs) [3]; and such a device would produce little parallel noise. However, it is not clear that diodes of this type could be constructed from CZT. If high resistivity materials always contains a large trap density, it may not be possible to develop good blocking contacts because of injection phenomena that occur between the contacts and trap states in the detector crystal [4].

Another method of reducing the noise level in large volume CZT detector systems would be to simply use a number of smaller detector elements each with an individual readout amplifier. The recent development of multiple channel monolithic preamplifiers and monolithic multiplexer circuits would greatly facilitate such an approach.

In summary, electronic noise is currently not the leading factor in determining the energy resolution of CZT detector systems. We anticipate that electronic noise will become a factor when the volume of devices exceed about 10 cm^3 . However, for large volume devices there exist several methods for reducing the electronic noise.

3. Charge collection effects

Fig. 3 illustrates a portion of a typical pulse height spectrum obtained by irradiating a CZT detector with a ^{137}Cs source. An obvious feature in Fig. 3 is the asymmetrical distortion of the 662 keV photo-peak due to incomplete charge collection throughout the volume of the detector. This distortion in the spectrum arises from the

Fig. 3. Portion of the pulse height spectrum obtained when a $1\times 1\times 1\text{ cm}^3$ $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ detector is irradiated with a ^{137}Cs isotopic source (open circles). The solid line is a computer simulation of the expected photo-peak response based on the measured electrical properties of the device. Note the strongly asymmetric "hole tail" on the photo-peak characteristic of poor hole drift length.

fact that the drift length of holes in CZT is less than the thickness of the detector under typical operating conditions. The degree of distortion produced by charge trapping depends on the mobility–lifetime product of holes in CZT, the thickness of the detector and the electric field within the device. In general, the distortion due to charge collection problems is a more severe problem than noise in limiting the performance of present-day CZT detectors, but it is also more difficult to simply describe in a quantitative manner. The broadening produced by electronic noise discussed earlier results in a purely Gaussian distortion in the pulse height spectrum, and the characteristics of the Gaussian broadening can be defined by a single parameter (FWHM). However, the asymmetrical distortion due to charge collection problem can not be described by a single parameter. For instance, it is possible for a CZT device to have a FWHM of 2% at 662 keV, yet have a full width ten maximum (FWTM) of 100%. Often only the FWHM of asymmetrical peaks is reported resulting in an overly optimistic view of the detector's performance.

4. Improved material properties

The most obvious solution to the charge collection problem in CZT detectors is to improve the quality of the CZT crystals until the lifetime of holes in the crystal is substantially increased. We estimate that the lifetime of holes would have to be improved by at least one order of magnitude (to greater than 2×10^{-5} μ s) in order to obtain better than 1% energy resolution at 662 keV. Although such an improvement in the quality of CZT crystals might be possible – through improvements in crystal growth and material engineering – it might take a few years to attain sufficiently improved CZT crystals. Fortunately, there exist other methods for minimizing charge collection distortion that do not require improvements in the average electron and hole transport properties and which can be applied to detectors made from existing materials; these methods are discussed below.

5. Electronic signal processing

Several methods of electronic signal processing have been developed to correct the charge collection problem in other semiconductor materials and are applicable to CZT detectors. Rise-time compensation has been investigated by several groups [5–7] for use with CdTe detectors, and a variation of this method for use with CZT is currently under development in our laboratory. Another signal processing method, related to rise-time compensation, simultaneously samples the detector at short and long integration times [8] and may be useful for correcting poor charge collection in CZT. Delayed pulsed sampling [9] has been applied with some success to HgI₂ detectors and may

Fig. 4. ^{137}Cs pulse height spectra as a function of pulse rise-time acquired with a multi-parameter data acquisition system. These data were acquired from a $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ detector $1.8 \times 1.8 \times 0.2$ cm thick and operated at 1000 V. Note that the "hole tailing" is resolved as a series of peaks whose mean amplitude decreases with longer signal rise-time.

be applicable to CZT as well. We do not have space to treat each method in detail in this paper, and at the present time, it is not clear which of these methods is the best for application to CZT. In general, most of these methods require the sampling of many of the signal pulses under conditions of incomplete charge collection. Thus the relative contribution of electronic noise will be increased for the incomplete charge pulses. We intend to discuss this problem in greater detail in a subsequent publication [10].

An illustration of the application of one of these signal processing methods – rise-time compensation – is illustrated in Figs. 4–7. Note that with rise-time compensation (and all other electronic signal processing methods) the trade-off between detector efficiency and energy resolution can be altered. This trade-off enables advantages in applications which favor one parameter over the other (energy resolution over efficiency). However, the ability to trade-off energy resolution for efficiency complicates a simple identification of the characteristics of the detector system.

6. Device design

Another method for reducing the effects of charge collection broadening in CZT detectors is through alteration of the electric field within the detector or through the design of the readout contacts. An interesting device

Fig. 5. The same pulse-height/rise-time data set shown in Fig. 4 after being transformed by a rise-time correction algorithm. Note that the projection of the data onto the pulse amplitude axis would now produce much less hole-tailing than in Fig. 4.

design, known as a Coplanar-Grid, has recently been demonstrated by Luke [11,12]. The coplanar grid concept was inspired by the Frisch Grid designs used in gas detectors [13,14]. Unlike a true Frisch Grid detector the coplanar design does not require conductors to be placed inside the detector (a challenging prospect in a semiconductor device). Instead, the Coplanar-Grid approach makes use of two sets of contact strips (at slightly different potentials) on one face of a planar detector. The coplanar grid technique utilizes two readout amplifiers to interrogate the charge deposited on the two different sets of contact strips. The signals from the two charge preamplifiers are then subtracted. Because the coplanar grid technique relies

Fig. 6. ^{137}Cs pulse height spectrum produced using a rise-time compensation algorithm. These data were acquired from a $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ detector $1.8 \times 1.8 \times 0.2$ cm thick and operated at 1000 V. Approximately 40% of the original counts acquired were used to create this pulse height spectrum.

Fig. 7. ^{137}Cs pulse height spectrum obtained with same detector used to acquire the data shown in Fig. 6, but without using rise-time compensation methods. The detector was operated under the same conditions as in Fig. 6, and for the same acquisition time.

on the sampling of incomplete charge signals it suffers from the same deficiency as the purely electronic signal processing methods; it must process signals of small amplitude with degraded signal to noise ratios. A linear combination of signals with reduced signal to noise ratio is always degraded more than one signal sampled at full amplitude (even if the combined signals sum to full amplitude). Thus, the coplanar design, as well as the purely electronic signal processing methods, can never achieve the same resolution (at full counting efficiency) as a detector with complete charge collection efficiency (in which hole and electron drift lengths are greater than detector thickness).

It is worth noting that the shape of the pulse height peaks produced by a coplanar device, and from any of the signal processing schemes discussed earlier, are not Gaussian in shape; although they are symmetric. Both the coplanar detector and the electronic processing methods rely on the combination of signals of different noise widths into a single peak, thus both of these techniques produce peaks which are the superposition of many Gaussians of different widths. This superposition is not itself a Gaussian peak and care must be used in describing the properties of photo-peaks produced by these detectors (or signal processing methods). Unlike a true Gaussian, a single parameter (such as FWHM) cannot be used to describe the shape of the peaks produced by coplanar devices or electronic signal processing methods. To avoid ambiguity when communicating spectral performance to potential detector users, two or more parameters should be used to describe a photo-peak in a gamma-ray spectrum (e.g. FWHM and FWTM).

In addition to the coplanar device described by Luke, other device configurations have been tried or are under consideration. Industry has developed the technology to manufacture CZT detectors in a co-axial geometry and devices employing this topology with its non-constant electric fields may prove useful [15]. If the manufacturing technology evolves sufficiently, it might be possible to

build CZT detectors with a true Frisch Grid topology (electrodes buried in the semiconductor); such a device would offer the advantage that a full charge signal would be sampled, simultaneously minimizing charge collection and noise problems.

7. Summary

Recent advances in CZT crystal growth methods have enabled the construction of large volume detectors (10 cm^3) from this material. The two factors limiting the energy resolution of large volume CZT detectors are electronic noise and distortion in the pulse height spectrum due to incomplete charge collection. Electronic noise in large volume CZT detectors is dominated by shot noise due to leakage currents through the detector, but owing to the high resistivity of CZT crystals, detectors up to about 10 cm^3 can be made before noise becomes a major problem. Charge collection distortions are the biggest problems associated with large volume CZT detectors, and arise because of the poor drift length of holes relative to detector dimensions. However, many recent advances have been made in electronic signal processing and improved detector design which reduce the magnitude of the charge collection problem. Together these advances promise gamma-ray spectrometers utilizing CZT detectors that have much better energy resolution than scintillation spectrometers, but reasonable gamma-ray stopping efficiencies due to their large volume.

Acknowledgments

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Correlation of Nuclear Spectrometer Performance with Uniformity and Resistivity in Cadmium Zinc Telluride

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Abstract

We have used low-temperature photoluminescence (PL) spectroscopy and room-temperature photoluminescence mapping as a measure of composition variation in cadmium zinc telluride grown by high-pressure Bridgman. We have correlated the uniformity, as measured by the line width of the bound exciton peak in the low-temperature spectrum and the degree of variation in the peak position in the room-temperature map, with the peak-to-valley ratio of the 59.5 keV photopeak in the pulse-height spectrum of ²⁴¹Am. For detectors having active areas of 10-30 mm², the PL measurements can be combined with resistivity measurements to give a strong predictor of detector performance. For arrays of 1 mm diameter detectors, the correlation between these material parameters and detector performance is much weaker. Our measurements show that material parameters and detector performance can vary substantially between points on a 1 cm² sample.

I. INTRODUCTION

It has been demonstrated informally by a variety of techniques that uniformity of composition is a key factor affecting performance of gamma ray spectrometers fabricated from cadmium zinc telluride [1]. In a previous paper [2] we have shown a correlation between uniformity of composition, as measured by low-temperature photoluminescence line widths and room-temperature PL maps, and detector performance for 2.4 mm thick Cd_{0.9}Zn_{0.1}Te detectors having active areas of 10 - 30 mm². In this paper we extend these results by including the effect of resistivity, and by applying the analysis to arrays of 1 mm diameter detectors.

II. EXPERIMENTAL

The details of the low-temperature PL measurements and room-temperature PL mapping have been described in a previous publication [3]. Only a few key facts will be repeated here.

The crystal was grown by high-pressure Bridgman with a nominal zinc fraction of 0.1. The surfaces were chemically polished in an orbital shaker using a 0.5% bromine-methanol/ethylene glycol solution. For the photoluminescence mapping, the laser beam was focused to a spot diameter of approximately 25 μ m, and the maps were obtained with a step size of 25 or 50 μ m. For low-temperature PL, the laser spot was not focused, so that the spectrum was averaged over a large portion of the sam-

ple area. Pulse height spectra for ²⁴¹Am were collected using a charge-sensitive preamp and a shaping amp with a shaping time of 1 μ s. Gold contacts approximately 400 Å thick were applied by RF sputtering.

III. RESULTS AND DISCUSSION

To determine the relationship between composition uniformity and detector performance, we performed the following experiment using a number of samples from two adjacent columns of a single boule slice: After the sample surface was polished, the PL spectrum at 4.2K was measured with the laser beam illuminating a large fraction of the sample area. Next room-temperature PL mapping was performed on a 1 mm² portion of the same sample area. Gold contacts of area 10 to 30 mm², depending on the sample size and shape, were deposited and the I-V curve of the detector measured. Lastly a pulse-height spectrum was obtained with the detector using an ²⁴¹Am gamma ray source.

For low-temperature PL, the fractional width of the donor-bound exciton (D⁰X) peak was used as a measure of sample uniformity. For room-temperature PL mapping, the peak position of the spectrum at each spatial point was converted to zinc fraction, x , using an empirical relationship from the literature [4]. The data are expressed as fractional deviation from the mean, $\Delta x/x_0$. The standard deviation of x divided by the mean was used as a measure of uniformity over the scanned area. The I-V data were expressed in terms of equivalent resistivity, given by:

$$\rho = VA/Id \quad (1)$$

where V is the bias voltage at which the pulse height spectrum was later obtained, I is the measured current at that voltage, A is the contact area, and d is the thickness of the sample. The peak-to-valley ratio of the 59.5 keV photopeak in the ²⁴¹Am spectrum was used as a measure of detector performance.

Figure 1 shows a representative low-temperature PL spectrum, room-temperature PL map, I-V curve and pulse height spectra. The PL spectrum is dominated by the near band edge (excitonic) features, indicating good crystallinity. The room temperature PL composition map shows some systematic reduction in zinc content as one approaches the heel of the boule (toward left in the figure), plus a background of random variation which is partly an artifact of fluctuations in the measurement system. We have quantified the random measurement error by disconnecting the translation actuators and repeatedly

Figure 1: Low temperature PL spectrum (a), room-temperature PL composition map (b), I-V curve (c), and ^{241}Am pulse height spectrum without collimator (d), and with collimator (e) for $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ detector with sputtered gold contacts.

measuring the spectrum at a single point. Under these conditions we obtained a σ/x_0 value of 0.52%, less than one-half of the minimum value for any of the detectors and well below the median value of 2.5%. Thus the random variation does not have a significant impact on the correlation.

The I-V curve has an asymmetry which we have seen consistently with sputtered contacts: the resistivity is higher when the contact that was sputtered first is biased negative. The pulse height spectrum exhibits relatively little hole tailing in this case - the broadening of the 59.5 KeV photopeak is partly due to fringing of the electric field near the edges of the contacts, which did not extend to the edges of the crystal. This problem can be reduced by collimating the source (compare 1(d) with 1(e)), but in order to study the effect of crystal uniformity on detector performance, we allowed the source to irradiate the entire surface.

Plots of detector performance vs. equivalent resistivity and the two measures of sample uniformity are given in Figures 2 and 3. The size and darkness of the circle used to represent a data point indicate the performance of that detector. (The largest circle represents peak/valley ~ 3.1 , while the smallest is ~ 1.1 .) One would expect the best performing detectors to lie in the upper left corner of the graph (higher resistivity, higher uniformity), with the worst performing detectors in the lower right, and the data are largely consistent with this rule. For these detectors the degree of uniformity as measured by photoluminescence can be combined with resistivity to give a strong predictor of detector performance.

The experiment described above has two important limitations: first, unmeasured material parameters, such as mobility-lifetime products, may vary significantly from sample to sample. Second, because of throughput limitations the room-temperature PL mapping was performed on only a fraction of the

Figure 2: Detector performance vs. equivalent resistivity and low-temperature PL (D^0X) peak width for larger-area detectors. Size of circle indicates peak/valley ratio of 59.5 KeV photopeak.

Figure 3: Detector performance vs. equivalent resistivity and variation in the room-temperature PL map for larger-area detectors.

sample area, which may not be representative of the entire sample. To reduce variations in other material parameters and ensure that the area probed by the PL techniques was precisely the same as the detector area, we repeated the above experiments using multiple, 1 mm diameter detectors on a single 1 cm^2 sample. The PL measurements were made through 1 mm holes in a mask and the front contacts deposited through the same holes, as illustrated in Figure 4. The rear side was completely metallized. Two samples of known quality were used, one having been previously tested as a detector and exhibiting good performance, and the other coming from a lot of material which produced consistently poor detectors. These two arrays will be referred to as the "good" and "poor" arrays, respectively.

Plots of detector performance vs. leakage current (since the detectors all have the same dimensions there is no need to

calculate equivalent resistivity) and 4.2K PL peak width for the two arrays are shown in Figures 5 and 6. The same general trend (best detectors in the lower left corner of the grid) can be seen, but with more scatter than in the case of the larger area detectors. We attribute the greater degree of scatter to unmeasured material parameters which vary from detector to detector in each array, but whose effects averaged out in the case of the larger area detectors. A likely candidate is the presence of traps which limit hole transport properties, but attempts to measure hole $\mu\tau$ products by standard alpha particle techniques were unsuccessful with these devices, because of inadequate charge collection.

While the data for the arrays have more scatter than the correlation between uniformity and detector performance seen with the larger detectors, they do illustrate some important points. First, the resistivity of the good array varies by an order of magnitude from one detector to another, and as a consequence the detector performance varies from quite good (peak-to-valley ratio >2) to poor (peak-to-valley ratio barely >1). Second, the best detectors in the poor array are better than the worst ones in the good array. Hence the fact that a 1 cm^2 sample is of good overall quality does not guarantee that there will not be significant differences in performance among small detectors fabricated on the sample. Figure 7 shows the best and worst pulse height spectra for each array, illustrating the variation of performance within each array.

The room temperature PL maps for the arrays showed no obvious correlation with detector performance. In addition to the effect of unmeasured material parameters mentioned above, there are two other reasons for this failure. One is the presence of inclusions of unknown nature which increase the variation in the PL map but do not significantly harm detector resolution. While the inclusion itself produces no PL intensity and is not counted in the statistics, we have observed in a sepa-

Figure 4: Experimental procedure for fabricating and testing 4×4 arrays of 1 mm diameter detectors.

Figure 5 : Detector performance vs. leakage current and low-temperature PL (D^0, X) peak width for "good" array of 1 mm diameter detectors.

Figure 6 : Detector performance vs. leakage current and low-temperature PL (D^0, X) peak width for "poor" array.

Figure 7 : Best (solid) and worst (dashed) pulse height spectra for "good" (left) and "poor" arrays. Channel number has been shifted by 150 for spectra on the right.

rate experiment that the area around the inclusion consistently shows a shift to higher energy in the PL spectrum, which shows up in the PL map as a large, apparent composition variation. This phenomenon is also present in at least one of the larger area detectors and is responsible for the extreme outlying point in Figure 3 (see Figure 8). We are still investigating the nature and properties of these inclusions and will report our results in a future paper.

Another reason for the failure of the room temperature PL maps to correlate with detector performance in the case of the 1 mm detectors is an edge effect caused by the metal mask. As can be seen in Figure 9, the PL spectrum shifts to higher energy near the edge where the mask contacts the sample. We

have observed that the room temperature PL spectrum shifts to lower energy with increased laser power due to local heating, and we believe that the mask reduces this effect near the edge by blocking a portion of the incident light. We attempted to account for the edge effect by discarding points more than 0.4 mm from the center of the 0.5 mm radius circular opening. This procedure resulted in the exclusion of more than one-third of the data, causing the map to be less representative of the detector area, and a meaningful correlation was not obtained.

Figure 8: Room temperature PL map for a sample containing large inclusions of unknown type (black areas with white borders).

IV. CONCLUSIONS

We have shown that for $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ detectors having active areas of $10\text{-}30\text{ mm}^2$, the degree of uniformity as measured by low-temperature photoluminescence and room-temperature PL mapping can be combined with resistivity measurements to give a strong predictor of detector performance. For arrays of 1 mm diameter detectors these predictors are less successful, a fact which we attribute to variation in an unknown material parameter whose effects averaged out in the larger area detectors. The PL mapping results for the arrays were also adversely affected by the presence of inclusions in many of the detectors and by an edge effect related to the mask.

Figure 9: Room temperature PL map for one of the array detectors. The PL spectrum shifts to higher energy near the edge of the circular opening in the mask.

We have also shown that material parameters and detector performance can vary substantially among points on a 1 cm^2 sample. The implication of this observation is that some crystals which perform well as single detectors may not perform well as imaging arrays.

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Orthogonal Strip Gamma Ray Imaging System for Use with HgI_2 and $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ Detectors

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Abstract

We have designed and constructed an orthogonal strip imaging system for use with room temperature semiconductor strip detectors. The system has been tested with both HgI_2 and $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ (CZT) detector elements. Our first system consists of complete readout electronics and software for spectroscopy and imaging with 8×8 orthogonal strip detectors. The readout electronics consist of 16 channels of hybrid charge sensitive preamplifiers, and 16 channels of parallel discriminators, shaping amplifiers, and a 16 channel ADC implemented in CAMAC and NIM. The software used to readout the instrument is capable of performing intensity measurements as well as spectroscopy on all 64 pixels of the device. In this paper we describe measurements to determine the factors limiting the performance of this system.

Keywords: Cadmium Zinc Telluride, Mercuric Iodide, Orthogonal Strip Detector, Gamma-Ray Imaging

1. Introduction

The application for which this gamma camera was developed is the monitoring of radioactive waste, stored nuclear materials, and medical applications (such as a miniature gamma camera for intraoperative use). However, the camera described in this paper does not have performance sufficient for these applications, instead our intention was to use this device as a platform to aid in the development of more sophisticated gamma cameras to be developed at Sandia National Laboratories.

2. Description of Instrument

Overview

The primary factor that determined the design of our gamma camera was the nature of the position sensitive radiation detector used as the basis of the system. We elected to use an orthogonal strip design as the position sensitive detector in our camera. The orthogonal strip design was first described by Gerber et al¹, and has received much recent attention for applications in astrophysics where high pixel counts are needed in coded aperture gamma-ray telescopes^{2,3,4}. The orthogonal strip design is attractive because it requires only $2 \times N$ channels of electronics to readout an N^2 array of pixels; thus minimizing the cost and complexity of the read-out electronics. Of course, for high count rate applications with a large number of pixels, the orthogonal strip design starts to lose these advantages. In these cases, integrated circuits can readily provide the needed channel density for the readout of a pixellated detector without limiting the count rate to one event per read cycle⁴.

The system we constructed was designed such that detectors could be readily inserted into the front-end read-out electronics as a distinct mechanical module using a standard electronic package (24 pin dual in-line package), as opposed to wire bonding the detector directly to the input of the preamplifiers. The disadvantage of this modular approach is that additional noise may be added by the interconnections and longer circuit foil traces between detector and preamplifier. The primary advantage of this design is that it allows a variety of detectors to be tested relatively easily without re-fabricating the front-end of the instrument each time a detector is changed. Thus, this instrument is particularly useful for testing various detector designs. The details of the design and construction of the hardware and software that comprise this instrument are described in another paper⁵. In this paper we wish to focus on the performance characteristics of this instrument; nonetheless, a brief review of the hardware and software that comprise this instrument is given here so that the measurements performed with this instrument can be more clearly understood.

The detectors were 8×8 orthogonal strip devices fabricated on HgI_2 and CZT substrates approximately 2.0 mm thick design with an active area of approximately 1 cm^2 . These devices were placed on chip carriers that allowed them to be plugged directly into the readout electronics. Detectors were fabricated by evaporating metal contacts on to the surface of etched

crystal substrates using a shadow mask to define the strip pattern. A total of four detectors were fabricated: three from CZT and one from HgI_2 . The detectors were mounted on standard 24 pin alumina dual in-line packages (DIPs) commonly used for hybrid electronic circuits. Fine gold wires (25 μm diameter) were bonded to the metal contact strips using silver epoxy, the other end of the wire was then bonded to a metal foil pattern on the alumina substrate of the dual inline package. The packaged detectors could then be inserted directly into a socket on the front-end readout circuit board.

The electronic system used to readout the detectors consisted of 8 channel hybrid preamplifiers (Lecroy HQV 820), system-by NIM and CAMAC readout amplifiers and ADCs. The performance of the readout system is determined largely by the hybrid preamplifiers which first amplify the signals from the 8×8 detector array; the rated performance of the preamplifiers translates to about 5 keV FWHM of noise referenced to a CZT gamma spectrum.

Software was written to readout the coincident signal from the strip detector, decode the pixel position, and visualize the gamma ray intensity distribution at the detector plane. The software interrogated the ADCs and discriminators in the CAMAC crate, decoded the position of interaction on the orthogonal strip detector, and created pulse height spectra of the interactions that occurred at each pixel. Another function of the software was to provide real time feedback on the operation of the camera, and diagnostics of various camera functions (such as cross-talk between channels). The software was written in the high level control language "Kmax" to minimize development and reduce the amount of time spent writing low level CAMAC control routines.

3. Performance of Camera

After we had assembled and debugged the camera hardware and software, we began testing the ability of the camera to measure the energy distribution of isotopic sources using pulse height spectroscopy and to form an image of a radioactive source using a pin-hole collimator to project the image onto the detector plane.

Image Formation

A pin-hole collimator was placed between gamma-ray photon sources and the detector and images acquired with the system. Such an image is shown in Figure 1. The ability to discriminate sources of a particular energy was verified and some results are also shown in Figure 1.

Figure 1. Pinhole images of a ^{135}Ba point source while the detector was flood illuminated by an ^{241}Am source from behind. B is the same as A except with an energy window of greater than 70 keV applied.

Signal Propagation

Additional measurements were performed with the camera system to determine if the signals produced by the detector matched our expectations based on the design of the detector elements and readout electronics. Figure 2 show some typical waveforms from adjacent strips observed at the output of the analog signal processing chain (preamplifier and shaping amplifier) with a digital oscilloscope. This cursory examination of the signals with an oscilloscope indicated that electronic "cross-talk" was not a significant problem. A more careful study of charge sharing between adjacent strips was performed with the apparatus shown in Figure 3. Some results obtained with this apparatus are shown in Figure 4; analysis of these data indicated that the ratio of events shared between strips (with flood-field irradiation of the strip detector) is consistent with what would be expected based on the fraction of metallization on the surface of the strip detector.

Figure 2. Waveforms observed with a digital oscilloscope when observing the output of the shaping amplifiers from three adjacent strips on an 2 mm thick 8x8 CZT detector with a strip pitch of 1.0 mm. The detector was being irradiated with a ^{133}Ba source. Plot A is the most common waveform observed at the output; all of the signal appears localized to one strip. The output in Plot B is observed much less commonly and may arise from an event generating electrons on the opposite of the detector and drifting toward the strips being observed. Initially, the same polarity current would be induced on both strips, but when the electrons near one strip, we would expect a current of opposite polarity to be generated on the adjacent strip. Plot C is a waveform that probably corresponds to an event occurring somewhere between two adjacent strips with current being induced on both strips.

Figure 3. Apparatus used to measure the simultaneous pulse height spectra from two adjacent strips. The ADCs used to measure pulse heights were triggered on a signal from either of the two strips.

Figure 4. Contour plot of pulse height spectra acquired simultaneously on two adjacent strips of a CZT strip detector irradiated with photons from a ^{133}Ba source. The ADCs used to measure pulse heights were triggered on a signal from either of the two strips.

Energy Resolution

The applications we are interested in require the gamma camera to simultaneously estimate the position of gamma ray emissions as well as measure the energy distribution of the sources; thus the energy resolution of the detector system is of great interest. A typical pulse height spectrum obtained from a single strip of one of our CZT strip detectors is shown in Figure 5. Examination of Figure 5 reveals that two factors degrade the pulse height resolution in the strip detectors: Gaussian broadening due to electronic noise and - for higher energy photons- an asymmetric distortion of the peaks due to incomplete charge collection effects. We have measured the contribution of both noise and charge collection broadening and report on it below.

Figure 5. Pulse height spectra obtained by irradiating an 8x8 CZT orthogonal strip detector with photons from a ^{133}Ba source. Plot A shows a typical pulse height spectrum obtained by interrogating one strip in the detector. The pulse height spectrum shown in Plot B was obtained by summing the pulse height spectra from all eight rows in the detector. Some degradation in energy resolution occurs when multiple rows (or columns) are summed because: 1. the noise broadening is

determined by the noisiest strip, and 2. the system software does not always correct perfectly for the fluctuations in the gain of each readout channel.

Electronic Noise Contribution

The relative contribution of the electronic noise to the pulse height spectrum broadening was measured by injecting a fixed amplitude charge pulse to the input of the preamplifier and measuring the width of the peak produced by the charge pulses in the pulse height spectrum under various conditions. Figure 6. shows a plot of the measured electronic noise as a function of linear amplifier time constant (semi-Gaussian shaping). Because there was no convenient way to inject charge pulse of known absolute amplitude into the front end of the preamplifier, a CZT pulse height spectrum of ^{241}Am (59.5 keV peak) was used to calibrate the charge gain of the system. A value for the average charge pair creation energy in CZT of 5.0 eV was assumed in the calculations. Noise measurements were also performed on the same detector strip using different readout electronics and the results are also shown in Figure 6. Using the conventional methods for analyzing the properties of detector system noise sources⁷, the results in Figure 6 indicate that the dominant noise source in the detector system is a parallel source. The fact that the parallel noise source is present even when there is no bias applied to the detector, indicates that the parallel noise source is not due to leakage current in the detector (shot noise), more likely, the parallel noise contribution is from the resistive feedback in the charge amplifier array.

Figure 6. Equivalent noise charge (ENC) versus amplifier shaping time (semi-Gaussian shaping) for a single strip of a CZT 8x8 orthogonal strip detector. Two different sets of readout electronics were used: 1. a single channel of the LeCroy 8 channel hybrid preamplifier used in our imaging camera prototype, and 2. a standard single channel charge sensitive preamplifier (Tennelec TC-170). The single channel preamplifier contributed much less noise than the 8 channel unit used in our camera. The general increase in the noise magnitude with increasing shaping time indicates that the noise broadening in the pulse height spectrum was dominated by $1/f$ and parallel white components. With the LeCroy preamplifier the parallel noise source was probably the feedback resistor in the charge sensitive preamplifier, as shot noise had no measurable contribution to the noise (no difference was measured in the noise with the bias on and off in the detector). Note that 1000 electrons rms of ENC corresponds to approximately 12 keV FWHM in the pulse height spectrum of a CZT detector.

Charge Collection Broadening

When the ratio of the strip pitch to the detector thickness is greater than one, we leave the "small pixel" regime⁸ (both the electron and hole motion contribute to the induced charge on each contact strip). Under these conditions, an orthogonal strip detector behaves much like a conventional large area parallel contact detector. A common characteristic of pulse height spectra produced by conventional detectors made from room-temperature semiconductor detectors is the presence of "hole

tailing" in the gamma-ray photo-peaks. This asymmetrical broadening of the peaks toward lower pulse heights has been studied extensively^{9,10} and arises from the short drift length of holes relative to the detector thickness. Because the holes drift only a fraction of the distance between the contacts, the charge collected on the contact depends on the depth of interaction of the gamma-ray photon in the crystal. The shapes of the peaks in the pulse height spectra that we observed were dominated by hole tailing effects at energies greater than about 100 keV; consistent with the penetration distances of photons in CZT, and the poor hole drift length in this material. These effect can be seen in the pulse height spectra shown in Figure 5.

4. Summary

We have designed, built and tested a small gamma camera based on an orthogonal strip detector design. The gamma camera utilizes "8x8" strip detectors made from either CZT or HgI₂. The camera is able to measure the position of interaction of single gamma ray photons (of energy greater than ~ 10 keV) on the detector plane and estimate the energy distribution of gamma-rays interacting with the detector using pulse height spectroscopy methods. We examined the factors that limit both the spatial resolution of this system and the energy resolution when operated as a pulse height spectrometer.

The position resolution of the detector was limited largely by the small number of pixels such that position resolution was only one in eight in each direction. The basic design and construction techniques used in the camera were measured to be sound however, as little or no cross-talk was observed between strips in the detector, and these same methods could be used to build devices with a higher density of strips and better relative position encoding.

The energy resolution of the system was also examined when operated as a pulse height spectrometer. It was found that the energy resolution was limited at lower photon energies (< 150 keV) by Gaussian broadening due to electronic noise, and at higher photon energies to charge collection effects. The electronic noise was dominated by the readout amplifiers and not controlled by leakage current in the detector. Thus, a significant improvement in the performance of the system could be realized (particularly at lower energies) through improvements in the electronic readout system.

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Recent CdZnTe Detector Fabrication Developments

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ABSTRACT

High resistivity CdZnTe has attracted extensive interests for its use as X-ray and gamma ray detector operated at room and elevated temperature due to its superior electro-optical and structural properties compared to other materials. The post-growth surface annealing under constituent over pressure, passivation, oxidation and chemical etching in terms of restoring stoichiometry, reducing surface damage and charge carrier trapping centers have been investigated by low temperature photoluminescence, x-ray photoelectron spectroscopy and atomic force microscopy. The choice of contacting metal and deposition method also plays a important role in lowering surface leakage current and increasing the ohmic behavior between contact and detector, and subsequently directly affects the detector performance. The data revealed by spectroscopic and microscopic measurements were correlated with current-voltage and x-ray spectrum results to give an improved detector fabrication process based on CdZnTe detector. These results will be presented and discussed.

Keywords: CdZnTe, Room-Temperature X-ray detector, surface leakage current, detector fabrication.

1. INTRODUCTION

During the past years, the development of Cadmium Zinc Telluride ($\text{Cd}_{1-x}\text{Zn}_x\text{Te}$) technology has attracted a considerable interest due to its high potential for many room temperature as well as elevated temperatures X-ray and gamma ray detector applications¹⁻⁵.

The widespread research and commercial applications of CdZnTe detectors in medical imaging, environmental monitoring, and industrial process control are actively being pursued at a variety of companies, universities, and government laboratories in the United States and around the world. CdZnTe's high efficiency, good energy resolution, excellent imaging resolution, and low bias voltages combined with the room temperature operation make CdZnTe an excellent technology for imaging and spectroscopy from 10 - 200 keV. In addition, the material growth technology is advancing such that large area detector grade material is becoming available at a reasonable cost. CdZnTe detector technology will enable large area arrays to be built with coded apertures masks for arcsecond positioning of gamma-ray burst sources and for wide-field hard x-ray astronomy. In addition, small CdZnTe detector arrays can be used at the foci of multilayer mirrors for high-sensitivity hard x-ray astronomy. Finally, CdZnTe detectors could be developed into remote sensing x-ray and gamma-ray spectrometers for flyby, orbital, and penetrator missions^{6,7}.

Surface preparation (polishing, chemical etching), metal contact deposition and surface passivation of CdZnTe are crucial in exploiting the full potential of CdZnTe detectors. Building large arrays of CdZnTe will require careful control of the entire fabrication process to achieve the optimum performance of the array. We have already demonstrated at Fisk University some of the potential advantages of ZnCdTe and proved that improvements in the device performance can be achieved through the optimization of the surface preparation of CdZnTe. The crystals used in the Fisk studies were grown by the high pressure Bridgman method and were obtained from Digirad, Inc. and eV Products, Inc. For the detector grade material superior crystallinity and high resistivity bulk is required; nevertheless, like many other semiconductor devices, the surface preparation during the detector fabrication plays a vital role in determining the contact characteristics and the surface leakage current factors determining the performance of a final $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ detector.

A limited number of reports⁸⁻¹¹ on the selection of electrode materials, contact deposition methods and surface treatments of $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ detector performance exists. In this paper, we present a review of recent developments in the area of detector fabrication aimed at lowering the dark leakage current and coupled with an analysis on the impact on the detector performances.

2. CHEMICAL ETCHING

Similar to CdTe , the crystal surface of $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ needs to be chemically etched during the device fabrication process. Yet, to date there are very few publications addressing the impact of surface treatment on the detector performance of $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$. Even for CdTe , despite the fact that various etching solutions exist, only a few are suitable for nuclear detector surface treatment with the Br -methanol solution being the most standard one used today. Several papers on chemical etching of CdTe have been reported¹²⁻¹⁷

Chemical etchants do not act uniformly on all the components of materials, generally leaving behind regions of nonstoichiometric material. In the case of a CZT crystal, this nonstoichiometric material is believed to be a tellurium-rich surface layer having a conductivity that is greater than the underlying stoichiometric CZT material. It is therefore desirable to remove nonstoichiometric material from the crystal surface, thereby reducing the conductivity of the crystal surface layer. It would be very beneficial to establish more comprehensive correlations between detector parameters such as resistivity, ohmicity, energy resolution and surface recombination of detector grade $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ for crystals that have been treated with different chemical etchants. A comparative study of $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ detectors for crystals that have been treated by different chemical etchants was conducted where AFM was used to examine surface roughnesses due to these chemical treatments. The best surface roughness (1.25 nm) was obtained with the BMLB treatment (5% Br in methanol) followed by (20% lactic acid in ethylene glycol + 2% Br), compared with the conventional bromine in methanol etching or the BM treatment. The AFM images are shown in Figure 2 (a) and 2(b). One might then assume that successive use of lactic acid and bromine, such as the BMLB treatment, could combine the advantages of each etchant and consequently generates the best results. In fact, low temperature photoluminescence spectra of the sample with BMLB etching reveal the largest $I(D^0, X) / I_{\text{def}}$ value and the narrowest full width-at-half-maximum of the (D^0, X) peak, in addition to a very good $I(D^0, X) / I_{\text{DAP}}$. This indicates the BMLB treatment results in the lowest defect concentration and better crystalline quality of the near surface region. We could then expect that this treatment should yield a good detector performance. Furthermore, only this treatment results in a surface for which we can resolve the X_{DAP} peak from the (D^0, X) peak, thus supporting the idea that this treatment should lead to a better surface preparation.

By comparing the low temperature photoluminescence spectra of our crystal under five different treatments, we conclude that the best surface preparation for detector grade $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ ($x = 0.1$) is the BMLB treatment, i.e., etching by the standard 5% bromine in methanol solution prior to Br -added (2%)-20% lactic acid in ethylene glycol. The results have demonstrated that a careful chemical etching is a necessary procedure to ensure the quality and smoothness of the crystal surface and that a cleaner and smoother surface correlates with a better detector performance. Figure 3 shows the lowering in the leakage current as the result of the BMLB treatment and a spectrum of ^{55}Fe could be obtained with this detector at room temperature and is shown in Figure 4.

3. METAL CONTACTING

There are four basic techniques of metallization : (i) electroless method¹⁸ (from AuCl_3 solution) which is the method most often used commercially, (ii) thermal vacuum evaporation from tungsten boats, (iii) sputtered Pt deposition¹⁹ and (iv) sputtered Au deposition.

In a recent study the above four ways of detector contact fabrication were compared²⁰. In order to minimize the surface oxidation contacts the metals were deposited onto the crystal surface immediately after etching. Typical sputtering conditions were 50 W of RF power delivered to the Au (Pt) target. The Ar gas pressure was kept constant at 5×10^{-2} Torr. The sputtering deposition time was 5 minutes. The thermal evaporation was carried out at 10^{-6} Torr and the deposition time was 2 minutes. The best performance was obtained with thermally evaporated Au contacts.

Figure 1 Spectral response of Cd_{0.9}Zn_{0.1}Te detector treated by conventional and new etchants (room temperature, contact area = 0.16 cm², thickness = 0.16 cm, 100 V bias, 100 s accumulation time, 4 μ s peaking time) : (a) 5% Br in methanol (BM), (b) (5% Br in methanol) + (20% lactic acid in ethylene glycol + 2% Br) (BMLB),

Figure 2. Atomic force microscopy images of BM (a) and BMLB (b) treated surfaces , 3-D scans of 1 μ mX1 μ m

Figure 3. Current-voltage curves obtained with the standard BM treatment and with the improved BMLB treatment

Figure 4. The spectral response obtained with a CZT detector fabricated using the BMLB treatment

One possible explanation for the poorer behavior of the samples where contacts were deposited by sputtering is that the more energized Pt and Au atoms might have damaged the crystal surface more violently compared to thermal evaporated Au atoms. Accordingly, the sputtered atoms might have generated more additional crystal defects in the near surface region which lead to more surface recombination and injecting contacts. And, an increased leakage current due to injecting contacts can further degrade the performance of the device. On the other hand, one possible reason that thermal Au evaporation is superior to the electroless method is that the latter method might have left some chemical byproducts on the detector surface. In addition, this method is also known to lack reproducibility and depend on the person performing the processing step²¹.

4. SURFACE PASSIVATION

Additional improvements in detector performance can be obtained by minimizing surface leakage currents. The process is called passivation and one commonly used approach is to find chemical treatments of the region between the contacts (lateral sides of the planar detector) that will stabilize its chemical composition. Based on the equilibrium phase diagram of the Cd-Te-O system CdTeO₃ was proposed²² as the most stable phases having a high resistivity.

A wet chemical oxidation process for CZT, based on hydrogen peroxide, was confirmed²³ to form an oxide layer with a thickness in the range of 21 - 44 Å, and a saturation stage being reached after approximately 5 minutes of oxidation. The surface leakage current was decreased by 37% and can be related to the oxide layers. One concern in developing surface passivation schemes is to avoid heating the sample, since post growth heat treatments of the crystals above 150 °C was shown to lead to irreversible deterioration of the detection properties. The hydrogen peroxide process had the advantage of being performed at room temperature, but other low temperature passivation processes, such as bombardment with atomic oxygen could also be developed.

5. SUMMARY

The necessity for improved CZT detector fabrication procedures is becoming increasingly important as additional applications and novel detector configurations need to be implemented. The efforts in this area will have to be performed in parallel to the progress in the crystal growth activity to ensure a future steady expansion of the technology. The existing fabrication procedures will have to be refined and sensitive surface characterization techniques, such as measurements surface recombination velocities, will have to be integrated into the research for a better understanding of the limitations in the detector performance.

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Gamma Ray Imaging and Spectroscopy System Using Room-Temperature Semiconductor Detector Elements

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Abstract

We report on the design, construction, and testing of a gamma-ray imaging system with spectroscopic capabilities. The imaging system consists of an orthogonal strip detector made from either HgI₂ or CdZnTe crystals. The detectors utilize an 8x8 orthogonal strip configuration with 64 effective pixels. Both HgI₂ or CdZnTe detectors are 1 cm² devices with a strip pitch of approximately 1.2 mm (producing pixels of 1.2 mm x 1.2mm). The readout electronics consist of parallel channels of preamplifier, shaping amplifier, discriminators, and peak sensing ADC. The preamplifiers are configured in hybrid technology, and the rest of the electronics are implemented in NIM and CAMAC with control via a Power Macintosh computer. The software used to readout the instrument is capable of performing intensity measurements as well as spectroscopy on all 64 pixels of the device. We report on the performance of the system imaging gamma-rays in the 20-500 keV energy range and using a pin-hole collimator to form the image.

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Introduction

Devices for imaging the spatial distribution of gamma-ray emitting isotopes- gamma cameras- have been in wide use in nuclear medicine for many years and currently represent a commercial annual market of several billion US dollars. Gamma cameras are potentially useful in non-medical applications as well; in fields such as non destructive evaluation of dense materials, and imaging of stored high-level radioactive waste. The conventional gamma camera employed in a hospital today consists of an Anger camera¹; a device consisting of an array of photomultiplier tubes (PMTs) attached to a large (approximately 1.0 meter in diameter) crystal of inorganic scintillating material such as NaI(Tl) or BGO. When a gamma-ray photon interacts with the scintillating crystal in a gamma-camera, it produces an energetic electron via a photo-electric or Compton scattering process. The energetic electron produced by gamma ray interaction subsequently deposits its energy within a millimeter or so of the gamma-ray interaction site. When the electron loses its energy it produces scintillating photons which are detectable by the PMTs. By determining the position centroid (Anger logic) of the light detected from several PMTs, the position of interaction of the photon can be determined. The scintillators used in conventional gamma cameras require some tens of eV of energy deposited to produce a single scintillating photon. Thus only a few tens of optical photons are produced per keV of energy deposited by the gamma ray. This low number of photons- or more accurately the statistical fluctuation in this number of photons- ultimately determines both the energy and position resolution of the detector used in existing gamma camera systems. The resolution with which a gamma-camera can encode position on the detector is not the same as the spatial resolution with which the gamma camera system can estimate the position of the gamma ray emitter. The position resolution of the collimator is also an important factor; in general, the position resolution in the resultant image is the convolution of the spatial response function of

the collimator and the spatial response function of the detector. In modern large gamma cameras, the position resolution of the system is usually determined by the collimator. However, in a portable system employing a high resolution collimator (such as a pinhole collimator), the system resolution would be determined by the position resolution of the detector plane. Under these circumstances (i.e. portable gamma-cameras) the position resolution of the gamma ray imager may be limited by the scintillator /PMT detector. Furthermore, if one seeks to determine the isotope that is producing gamma ray emission, the ability to resolve isotopes is limited by the energy resolution of a scintillator based detector system.

An obvious method for overcoming the limitations of scintillator based systems is to use a semiconductor detector based gamma camera. In a semiconductor detector, only a few eV are required to produce an electron-hole pair. Furthermore, the generation of the charge pairs in a semiconductor detector is a sub-Poissonian process (fluctuation is less than a random process), resulting in a lower theoretical limit for the energy resolution of the detector, and potentially better position resolution as well. Indeed, semiconductor gamma ray spectrometers made from germanium long ago displaced scintillation systems as the best detection technique for energy spectroscopy. However, because of the cryogenic cooling involved, it is often not practical to use a germanium detector particularly for field use. Instead, one solution to producing improved miniature gamma-cameras is to use room temperature semiconductor materials (such as HgI₂ and CZT) to build the detectors. In this paper we describe the design, construction and testing of such a semiconductor gamma camera.

System Design

There are several approaches that might be tried to produce a semiconductor detector with imaging capabilities. The most obvious solution is to configure a square array of individual

detector elements into the desired image plane. However, since a typical imager could require a square array of dimensions 32×32 elements (1024 total elements), it is not economical to fabricate and assemble into an array such a large number of individual elements.

The next most obvious approach is to fabricate a monolithic array of individual detector elements on a single semiconductor substrate. If lithographic approaches to fabrication are used, it becomes practical to fabricate monolithic arrays with a suitable number of elements, however reading out the signal from such an array would require very complex electronics. In general, to readout an $N \times N$ array of individual pixels requires N^2 separate channels of readout electronics; for arrays larger than about 4×4 this becomes a very complex solution, particularly since each channel of electronics must be very high performance (low noise) to take advantage of the benefits of semiconductor detectors.

An alternative approach to reading out a semiconductor detector array was first proposed by Gerber et al² and is known as an orthogonal strip design. Such an approach is illustrated in Figure 1 and consists of rows of parallel electrical contacts (strips) placed at right angles to each other on opposite sides of the detector. By making use of the temporal coincidence between events recorded on both sides of the detector, it is possible to readout an effective array of N^2 effective detector elements using only $2N$ channels of readout electronics. The orthogonal strip approach was used for all of the detectors described in this paper.

Detector Design and Fabrication

Detectors were designed and built using both HgI_2 and CZT materials as substrates. Our first detectors were 8×8 devices fabricated on HgI_2 and CZT substrates approximately 2.0 mm thick. These 8×8 orthogonal strip detectors resulted in devices with 64 effective pixels with each pixel a

square of approximately $0.125 \times 0.125 \text{ cm}^2$ dimensions. These devices were placed on chip carriers (see Figure 2) that allowed them to be plugged directly into the readout electronics.

Detectors were fabricated by evaporating metal contacts on to the surface of etched crystal substrates using a shadow mask to define the strip pattern. A total of four detectors were fabricated: three from CZT and one from HgI_2 . The detectors were mounted on standard 24 pin alumina dual in-line packages (DIPs) commonly used for hybrid electronic circuits. Fine gold wires ($25 \text{ }\mu\text{m}$ diameter) were bonded to the metal contact strips using silver epoxy, the other end of the wire was then bonded to a metal foil pattern on the alumina substrate of the dual inline package. The packaged detectors could then be inserted directly into a socket on the front-end readout circuit board. Subsequent testing of the detectors indicated that all of the detectors operated correctly as spectroscopic imaging devices, however, in three of the four devices (1 HgI_2 , and 2 CZT) there was one strip that did not function. As of this writing it is not clear if the cause of the strip failure was in the detector or in the bonding and interconnection used to connect the detector to the readout electronics. The remaining CZT detector was fully functional (all rows and column strips functioned).

Electronic Readout

The system- used to readout the 8×8 detector arrays made use of hybrid preamplifiers followed by NIM and CAMAC readout amplifiers and ADCs. A block diagram of the electronics used to readout the 8×8 detector array is shown in Figure 3. The performance of the readout system is determined largely by the hybrid preamplifiers which first amplify the signals from the 8×8 detector array. We chose to use a commercially available preamplifier array in our first prototype gamma camera. The preamplifiers (Lecroy HQV 820) are constructed in thick film hybrid technology and contain 8 channels of preamplifier in a single wide 24 pin dual inline

package (DIP). The rated performance of the preamplifiers translates to about 5 keV FWHM of noise referenced to a CZT gamma spectrum. We chose the HQV 820 preamplifiers because they were the only multi-channel units available commercially.

A circuit board was designed and built that accommodated two of the eight channel hybrid preamplifiers as well as the miscellaneous passive components to couple the detector to the preamplifier unit. The "front-end" circuit board containing the preamplifiers also contained a socket for holding an 8x8 detector. The additional capacitance and conductor trace length needed to socket the detectors would degrade the noise performance of the system somewhat, but we decided that the tradeoff of being able to test several detector designs with the same readout circuit outweighed the slight performance degradation that socketing the detector induced. The front end circuit board was constructed of a Teflon derivative to minimize "1/f" noise³ that would be induced by a conventional electronic circuit board. A photograph of the assembled circuit board is shown in Figure 4 with an 8x8 CZT detector attached.

The front-end electronics were housed in a separate chassis which comprised the "camera head" and was connected via coaxial cables to the remaining readout electronics. The remaining electronics were housed in NIM bins and a CAMAC crate and interfaced to the readout computer with a SCSI CAMAC controller. A form of sparse readout- implemented in hardware- was used to limit the amount of data the computer must obtain from the comparators and ADCs in the CAMAC crate. Signals from all 16 channels (8 columns and 8 rows, 64 pixels) were fed to a 16 channel comparator; when at least two comparators had fired, a master gate signal was triggered initiating the readout sequence. Additional event selection was also performed in software.

Software

In order to readout the coincident signal from the strip detector, decode the pixel position, and visualize the gamma ray intensity distribution at the detector plane it was necessary to write software to control the gamma camera. The software interrogated the ADCs and discriminators in the CAMAC crate, decoded the position of interaction on the orthogonal strip detector, and created pulse height spectra of the interactions that occurred at each pixel. Another function of the software was to provide real time feedback on the operation of the camera, and diagnostics of various camera functions (such as cross-talk between channels). The software was written in the high level control language "Kmax" to minimize development and reduce the amount of time spent writing low level CAMAC control routines. Figure 5 illustrates the graphical user interface (GUI) presented to the user when operating the software. The software, which executes on a Power Macintosh computer, also contains many more windows and dialog boxes than are shown in Figure 5, and can be called up for various diagnostic functions.

Results

To demonstrate the capabilities of imaging with room temperature semiconductor detectors, the gamma camera system described in the previous sections of this report was tested with isotopic sources. Three general types of experiments were performed with both HgI₂ and CZT detectors mounted in the gamma camera: flood field images of the detector plane to determine the uniformity of its response, imaging of objects with the aid of a pinhole collimator, and gamma-ray pulse height spectroscopy of isotopic sources.

Flood field images were obtained by mounting a ¹³³Ba source a few cm in front of the detector plane and recording the count rate at each pixel location as well as the energy spectrum of the source. Imaging studies were performed by mounting a pinhole collimator on the front of the gamma camera assembly and irradiating the collimator with isotopic sources of small active

diameter. It was then possible to measure the position resolution on the detector by recording images of the source as it was moved in a plane parallel to the detector plane. Some results from such an experiment are shown in Figure 6.

Pulse height spectra were also acquired in both flood field mode and with the pinhole collimator. A typical pulse height spectrum taken with a CZT detector is shown in Figure 7. Two components were identified in the broadening of the pulse height spectra: Gaussian broadening due to random electronic noise³ and an asymmetrical distortion of the gamma-ray peaks due to charge trapping effects^{4,5}.

Discussion

An instrument which imaged and performed spectroscopy on gamma-rays was designed built and tested. The imaging system used a room temperature semiconductor detector made from either CZT or HgI_2 . All detectors fabricated for this study were approximately 1.0 cm² in active area and were of an 8x8 orthogonal strip design (64 effective pixels). Reliability problems with the detectors were experienced; only one of four detector units was fully functional, but all detector units were partly functional (15 out of 16 strips were functional on the remaining three devices). It is not clear- as of this writing- the cause of the non-functional strips.

Simple tests of the position resolution of the detector elements indicated that the CZT and HgI_2 strip orthogonal strip detectors were capable of resolving the movement of a point source on the imaging plane with a resolution of better than 1.0 mm. Further testing will be required to better quantify the spatial resolving capabilities of this instrument. The ability of the imaging detectors to resolve the energy of the detected gamma rays was also measured. It was found that energy resolution- as measured by pulse height spectroscopy- was limited at low energies by electronic noise at approximately 15 keV FWHM. At higher gamma ray energies, the energy resolution was

limited by hole trapping and extensive "hole-tailing" was observed on the low energy sides of the peaks. This behavior was expected, and is consistent with the performance of single element detectors made from these materials.

Despite the deficiencies observed in the performance of our first system, we found our results (and those obtained by others using similar methods) to be quite encouraging. Orthogonal strip detectors made from room-temperature semiconductor detector materials appear to be a viable method for imaging the distribution of radionuclides with high spatial resolution and good energy resolution; particularly if the hole trapping effects in these detector materials can be minimized.

We are currently working on improving the performance of our gamma-ray imaging devices by making use of electron-only device designs (analogous to the gridded ion chambers used with gas detectors^{6,7}) to minimize hole tailing, and using an custom integrated circuit readout chip to reduce the electronic noise and increase the number of readout channels.

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Figure 1. Diagrammatic view of an "8x8" orthogonal strip detector and front-end readout electronics. The metal contact strips are deposited on opposite sides of a square piece of semiconductor wafer. Event localization on the detector plane is determined by scoring a coincidence event between a column and a row. Using this method reduces the complexity of the readout electronics considerably. In general, to readout an array of N^2 effective pixels only requires $2 \times N$ channels of readout electronics, as opposed to N^2 channels of readout required for a detector consisting of an array of individual pixels.

Figure 2. Photograph of an 8x8 orthogonal strip CZT detector mounted on a ceramic 24 pin DIP package.

Figure 3. Block diagram of the electronic readout system used in the 8x8 gamma camera. The system may be viewed as consisting of two branches: one for counting hits (discriminator) and the other branch for spectroscopy (ADC). Pulses from the Lecroy HQV820 preamps in the camera head are shaped and amplified by a CAEN 16 channel spectroscopy amplifier, these pulses are then fed to both a 16 channel discriminator and to a 16 channel ADC. Triggering of a read cycle is initiated in hardware whenever at least two channels are above the discriminator threshold, subsequent event selection is then performed in software after the CAMAC discriminators and ADCs have been read.

Figure 4. Photograph of the "front-end" circuit board used in the 8x8 gamma camera. An 8x8 CZT detector module mounted in a DIP socket is visible in the center of the photograph. The white cylindrical objects on either side of the detector module are the decoupling capacitors (an AC coupled configuration was used to connect the detector strips to the preamp). The black rectangular objects on either end of the circuit board are the eight channel preamplifier arrays. The circuit board was constructed from Duroid- a Teflon derivative- to minimize the "1/f" noise produced by the circuit traces between the detector and the preamplifier inputs.

Figure 5. Printout of Graphical User Interface (GUI) of software for controlling the operation of the "8x8" gamma camera. A large number of windows and dialog boxes are available to the operator for controlling and monitoring the operation of the camera. The window titled "EVENT-HIST" is a display of the total number of counts at each pixel. The window titled "ENERGY_WIN" is the display of intensity of hits that fall within a range of pulse height values selected on a master histogram of pulse height intensities.

Figure 6. Contour plots of the intensity distribution of the gamma-ray count rate on a CZT orthogonal strip detector. These images were formed by a pin-hole lead collimator placed between the detector and the ^{133}Ba source. The images on the right are composed of raw data sorted into spatial bins by the data collection software. The images on the left are filtered versions of the raw data; a simple cut-off filter was used (at 50% of maximum image intensity) to reject counts below the cut-off. The source is located at an initial position in the uppermost images, the image of the source on the detector plane source was then moved to the right 3.75 mm and the middle images were produced. Finally, the source was moved again (up 1.25 mm) and the lowermost images obtained. These data imply that the spatial resolution at the detector plane is better than 1.0 mm.

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Orthogonal Strip Gamma Ray Imaging System for Use with HgI_2 and $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ Detectors

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Abstract

We have designed and constructed an orthogonal strip imaging system for use with room temperature semiconductor strip detectors. The system has been tested with both HgI_2 and $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ (CZT) detector elements. Our first system consists of complete readout electronics and software for spectroscopy and imaging with 8×8 orthogonal strip detectors. The readout electronics consist of 16 channels of hybrid charge sensitive preamplifiers, and 16 channels of parallel discriminators, shaping amplifiers, and a 16 channel ADC implemented in CAMAC and NIM. The software used to readout the instrument is capable of performing intensity measurements as well as spectroscopy on all 64 pixels of the device. In this paper we describe measurements to determine the factors limiting the performance of this system.

Keywords: Cadmium Zinc Telluride, Mercuric Iodide, Orthogonal Strip Detector, Gamma-Ray Imaging

1. Introduction

The application for which this gamma camera was developed is the monitoring of radioactive waste, stored nuclear materials, and medical applications (such as a miniature gamma camera for intraoperative use). However, the camera described in this paper does not have performance sufficient for these applications, instead our intention was to use this device as a platform to aid in the development of more sophisticated gamma cameras to be developed at Sandia National Laboratories.

2. Description of Instrument

Overview

The primary factor that determined the design of our gamma camera was the nature of the position sensitive radiation detector used as the basis of the system. We elected to use an orthogonal strip design as the position sensitive detector in our camera. The orthogonal strip design was first described by Gerber et al¹, and has received much recent attention for applications in astrophysics where high pixel counts are needed in coded aperture gamma-ray telescopes^{2,3,4}. The orthogonal strip design is attractive because it requires only $2 \times N$ channels of electronics to readout an N^2 array of pixels; thus minimizing the cost and complexity of the read-out electronics. Of course, for high count rate applications with a large number of pixels, the orthogonal strip design starts to lose these advantages. In these cases, integrated circuits can readily provide the needed channel density for the readout of a pixellated detector without limiting the count rate to one event per read cycle⁴.

The system we constructed was designed such that detectors could be readily inserted into the front-end read-out electronics as a distinct mechanical module using a standard electronic package (24 pin dual in-line package), as opposed to wire bonding the detector directly to the input of the preamplifiers. The disadvantage of this modular approach is that additional noise may be added by the interconnections and longer circuit foil traces between detector and preamplifier. The primary advantage of this design is that it allows a variety of detectors to be tested relatively easily without re-fabricating the front-end of the instrument each time a detector is changed. Thus, this instrument is particularly useful for testing various detector designs. The details of the design and construction of the hardware and software that comprise this instrument are described in another paper⁵. In this paper we wish to focus on the performance characteristics of this instrument; nonetheless, a brief review of the hardware and software that comprise this instrument is given here so that the measurements performed with this instrument can be more clearly understood.

The detectors were 8×8 orthogonal strip devices fabricated on HgI_2 and CZT substrates approximately 2.0 mm thick design with an active area of approximately 1 cm^2 . These devices were placed on chip carriers that allowed them to be plugged directly into the readout electronics. Detectors were fabricated by evaporating metal contacts on to the surface of etched

crystal substrates using a shadow mask to define the strip pattern. A total of four detectors were fabricated: three from CZT and one from HgI_2 . The detectors were mounted on standard 24 pin alumina dual in-line packages (DIPs) commonly used for hybrid electronic circuits. Fine gold wires (25 μm diameter) were bonded to the metal contact strips using silver epoxy, the other end of the wire was then bonded to a metal foil pattern on the alumina substrate of the dual inline package. The packaged detectors could then be inserted directly into a socket on the front-end readout circuit board.

The electronic system used to readout the detectors consisted of 8 channel hybrid preamplifiers (Lecroy HQV 820), system-by-NIM and CAMAC readout amplifiers and ADCs. The performance of the readout system is determined largely by the hybrid preamplifiers which first amplify the signals from the 8x8 detector array; the rated performance of the preamplifiers translates to about 5 keV FWHM of noise referenced to a CZT gamma spectrum.

Software was written to readout the coincident signal from the strip detector, decode the pixel position, and visualize the gamma ray intensity distribution at the detector plane. The software interrogated the ADCs and discriminators in the CAMAC crate, decoded the position of interaction on the orthogonal strip detector, and created pulse height spectra of the interactions that occurred at each pixel. Another function of the software was to provide real time feedback on the operation of the camera, and diagnostics of various camera functions (such as cross-talk between channels). The software was written in the high level control language "Kmax" to minimize development and reduce the amount of time spent writing low level CAMAC control routines.

3. Performance of Camera

After we had assembled and debugged the camera hardware and software, we began testing the ability of the camera to measure the energy distribution of isotopic sources using pulse height spectroscopy and to form an image of a radioactive source using a pin-hole collimator to project the image onto the detector plane.

Image Formation

A pin-hole collimator was placed between gamma-ray photon sources and the detector and images acquired with the system. Such an image is shown in Figure 1. The ability to discriminate sources of a particular energy was verified and some results are also shown in Figure 1.

Figure 1. Pinhole images of a ^{133}Ba point source while the detector was flood illuminated by an ^{241}Am source from behind. B is the same as A except with an energy window of greater than 70 keV applied.

Signal Propagation

Additional measurements were performed with the camera system to determine if the signals produced by the detector matched our expectations based on the design of the detector elements and readout electronics. Figure 2 show some typical waveforms from adjacent strips observed at the output of the analog signal processing chain (preamplifier and shaping amplifier) with a digital oscilloscope. This cursory examination of the signals with an oscilloscope indicated that electronic "cross-talk" was not a significant problem. A more careful study of charge sharing between adjacent strips was performed with the apparatus shown in Figure 3. Some results obtained with this apparatus are shown in Figure 4; analysis of these data indicated that the ratio of events shared between strips (with flood-field irradiation of the strip detector) is consistent with what would be expected based on the fraction of metallization on the surface of the strip detector.

Figure 2. Waveforms observed with a digital oscilloscope when observing the output of the shaping amplifiers from three adjacent strips on a 2 mm thick 8x8 CZT detector with a strip pitch of 1.0 mm. The detector was being irradiated with a ^{133}Ba source. Plot A is the most common waveform observed at the output; all of the signal appears localized to one strip. The output in Plot B is observed much less commonly and may arise from an event generating electrons on the opposite of the detector and drifting toward the strips being observed. Initially, the same polarity current would be induced on both strips, but when the electrons near one strip, we would expect a current of opposite polarity to be generated on the adjacent strip. Plot C is a waveform that probably corresponds to an event occurring somewhere between two adjacent strips with current being induced on both strips.

Figure 3. Apparatus used to measure the simultaneous pulse height spectra from two adjacent strips. The ADCs used to measure pulse heights were triggered on a signal from either of the two strips.

Figure 4. Contour plot of pulse height spectra acquired simultaneously on two adjacent strips of a CZT strip detector irradiated with photons from a ^{133}Ba source. The ADCs used to measure pulse heights were triggered on a signal from either of the two strips.

Energy Resolution

The applications we are interested in require the gamma camera to simultaneously estimate the position of gamma ray emissions as well as measure the energy distribution of the sources; thus the energy resolution of the detector system is of great interest. A typical pulse height spectrum obtained from a single strip of one of our CZT strip detectors is shown in Figure 5. Examination of Figure 5 reveals that two factors degrade the pulse height resolution in the strip detectors: Gaussian broadening due to electronic noise and - for higher energy photons - an asymmetric distortion of the peaks due to incomplete charge collection effects. We have measured the contribution of both noise and charge collection broadening and report on it below.

Figure 5. Pulse height spectra obtained by irradiating an 8x8 CZT orthogonal strip detector with photons from a ^{133}Ba source. Plot A shows a typical pulse height spectrum obtained by interrogating one strip in the detector. The pulse height spectrum shown in Plot B was obtained by summing the pulse height spectra from all eight rows in the detector. Some degradation in energy resolution occurs when multiple rows (or columns) are summed because: 1. the noise broadening is

determined by the noisiest strip, and 2. the system software does not always correct perfectly for the fluctuations in the gain of each readout channel.

Electronic Noise Contribution

The relative contribution of the electronic noise to the pulse height spectrum broadening was measured by injecting a fixed amplitude charge pulse to the input of the preamplifier and measuring the width of the peak produced by the charge pulses in the pulse height spectrum under various conditions. Figure 6. shows a plot of the measured electronic noise as a function of linear amplifier time constant (semi-Gaussian shaping). Because there was no convenient way to inject charge pulse of known absolute amplitude into the front end of the preamplifier, a CZT pulse height spectrum of ^{241}Am (59.5 keV peak) was used to calibrate the charge gain of the system. A value for the average charge pair creation energy in CZT of 5.0 eV was assumed in the calculations. Noise measurements were also performed on the same detector strip using different readout electronics and the results are also shown in Figure 6. Using the conventional methods for analyzing the properties of detector system noise sources⁷, the results in Figure 6 indicate that the dominant noise source in the detector system is a parallel source. The fact that the parallel noise source is present even when there is no bias applied to the detector, indicates that the parallel noise source is not due to leakage current in the detector (shot noise), more likely, the parallel noise contribution is from the resistive feedback in the charge amplifier array.

Figure 6. Equivalent noise charge (ENC) versus amplifier shaping time (semi-Gaussian shaping) for a single strip of a CZT 8x8 orthogonal strip detector. Two different sets of readout electronics were used: 1. a single channel of the LeCroy 8 channel hybrid preamplifier used in our imaging camera prototype, and 2. a standard single channel charge sensitive preamplifier (Tennelec TC-170). The single channel preamplifier contributed much less noise than the 8 channel unit used in our camera. The general increase in the noise magnitude with increasing shaping time indicates that the noise broadening in the pulse height spectrum was dominated by 1/f and parallel white components. With the LeCroy preamplifier the parallel noise source was probably the feedback resistor in the charge sensitive preamplifier, as shot noise had no measurable contribution to the noise (no difference was measured in the noise with the bias on and off in the detector). Note that 1000 electrons rms of ENC corresponds to approximately 12 keV FWHM in the pulse height spectrum of a CZT detector.

Charge Collection Broadening

When the ratio of the strip pitch to the detector thickness is greater than one, we leave the "small pixel" regime⁸ (both the electron and hole motion contribute to the induced charge on each contact strip). Under these conditions, an orthogonal strip detector behaves much like a conventional large area parallel contact detector. A common characteristic of pulse height spectra produced by conventional detectors made from room-temperature semiconductor detectors is the presence of "hole

tailing" in the gamma-ray photo-peaks. This asymmetrical broadening of the peaks toward lower pulse heights has been studied extensively^{9,10} and arises from the short drift length of holes relative to the detector thickness. Because the holes drift only a fraction of the distance between the contacts, the charge collected on the contact depends on the depth of interaction of the gamma-ray photon in the crystal. The shapes of the peaks in the pulse height spectra that we observed were dominated by hole tailing effects at energies greater than about 100 keV; consistent with the penetration distances of photons in CZT, and the poor hole drift length in this material. These effect can be seen in the pulse height spectra shown in Figure 5.

4. Summary

We have designed, built and tested a small gamma camera based on an orthogonal strip detector design. The gamma camera utilizes "8x8" strip detectors made from either CZT or HgI₂. The camera is able to measure the position of interaction of single gamma ray photons (of energy greater than ~ 10 keV) on the detector plane and estimate the energy distribution of gamma-rays interacting with the detector using pulse height spectroscopy methods. We examined the factors that limit both the spatial resolution of this system and the energy resolution when operated as a pulse height spectrometer.

The position resolution of the detector was limited largely by the small number of pixels such that position resolution was only one in eight in each direction. The basic design and construction techniques used in the camera were measured to be sound however, as little or no cross-talk was observed between strips in the detector, and these same methods could be used to build devices with a higher density of strips and better relative position encoding.

The energy resolution of the system was also examined when operated as a pulse height spectrometer. It was found that the energy resolution was limited at lower photon energies (< 150 keV) by Gaussian broadening due to electronic noise, and at higher photon energies to charge collection effects. The electronic noise was dominated by the readout amplifiers and not controlled by leakage current in the detector. Thus, a significant improvement in the performance of the system could be realized (particularly at lower energies) through improvements in the electronic readout system.

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Status of Radiation Damage Measurements in Room Temperature Semiconductor Radiation Detectors

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STATUS OF RADIATION DAMAGE MEASUREMENTS IN ROOM TEMPERATURE SEMICONDUCTOR RADIATION DETECTORS

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Abstract

The literature of radiation damage measurements on cadmium zinc telluride (CZT), cadmium telluride (CT), and mercuric iodide (HgI_2) is reviewed for the purpose of determining their applicability to space applications. CZT strip detectors exposed to intermediate energy (1.3 MeV) proton fluences exhibit increased interstrip leakage after 10^{10} p/cm² and significant bulk leakage after 10^{12} p/cm². CZT exposed to 200 MeV protons shows a two-fold loss in energy resolution after a fluence of 5×10^9 p/cm² in thick (3 mm) planar devices but little effect in 2 mm devices. No energy resolution effects were noted from moderated fission spectrum neutrons after fluences up to 10^{10} n/cm², although activation was evident. CT detectors show resolution losses after fluences of 3×10^9 p/cm² at 33 MeV for chlorine-doped detectors. Indium doped material may be more resistant. Neutron exposures (8 MeV) caused resolution losses after fluences of 2×10^{10} n/cm². Mercuric iodide has been studied with intermediate energy protons (10 to 33 MeV) at fluences up to 10^{12} p/cm² and with 1.5 GeV protons at fluences up to 1.2×10^8 p/cm². Neutron exposures at 8 MeV have been reported at fluences up to 10^{15} n/cm². No radiation damage was found under these irradiation conditions.

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Status of Radiation Damage Measurement in Room Temperature Semiconductor Radiation Detectors

Introduction

As part of its FY-97 PIDDP work scope, Sandia National Laboratories, California, was tasked to assess the status of radiation damage measurements for a group of wide-bandgap semiconductor materials being developed for use as x-ray and gamma-ray spectrometers. Interest in radiation damage in these materials stems from the growing interest in their application to space science and from the known susceptibility of cryogenic germanium and silicon to damage at relatively modest fluence levels.

Radiation damage in semiconductor radiation detectors is manifest in a number of ways including changes in energy resolution, leakage current, and peak position. Under certain conditions, activation may also occur. While the space environment contains a wide variety of high energy particles, damage to semiconductor detectors is due primarily to electrons, protons, and neutrons. Parameters affecting the response of materials to radiation fields include fluence level, flux, incident energy, detector bias, and detector temperature. Additionally, detector dimensions and impurities can also be of importance.

Methods

Radiation effects data were obtained through a literature search using the computer data bases INSPEC, CALPLUS, and COMPENDEX, together with DOE archives. In all, more than 300 abstracts were reviewed. The review was confined to the materials cadmium zinc telluride, (CZT), cadmium telluride (CT), and mercuric iodide (HgI_2), the most promising of the wide-band gap materials now under development.

Results

The radiation effect reported most frequently was the change in energy resolution although changes in the leakage current and peak position shifts were sometimes cited. Energy resolution (ΔE) was determined in most cases by recording the spectrum of a monochromatic photon or alpha particle before and after exposure. Detector energy resolution is reported in terms of the

full width of a given spectral line at its half intensity point (FWHM). In general, neither the detector bias conditions nor the detector temperature were reported. Where available, this information is noted in this report. The results of this investigation are summarized in Tables 1-3. References are indicated in the final column of each table and listed in Appendix A.

Before discussing results of the current study, it is useful to note that significant energy resolution losses in silicon occur at about 10^{12} n/cm² for fast neutrons (Ewan, 1975) and leakage current and pulse height changes after 5×10^{11} protons/cm². Planar germanium detectors exhibit significant resolution losses after 10^9 n/cm². Coaxial (n-type) are significantly more resistant to neutrons (Pehl, 1979) than p-type. Coaxial (p-type) germanium detectors exhibit resolution losses at about 2×10^7 protons/cm² while in n-type coaxial (reverse electrode) detectors, resolution losses appear at about 2×10^8 protons/cm². Despite the susceptibility of germanium to radiation damage, thermal annealing methods have been developed to mitigate, if not reverse, the symptoms of high fluence exposures.

Mercuric Iodide

The results of this investigation are summarized in Table 1. Proton exposures at 10, 10.7, 33, and 1500 MeV have been reported. (Iwanczyk, 1996; Becchetti, 1983; Patt, 1990, and Nakano, 1976). Fluences ranged from 1.2×10^8 protons/cm² (1500 MeV) to 10^{12} protons/cm² (10.7 MeV). Also reported were 8 MeV neutron exposures at fluences up to 10^{15} n/cm². No evidence of radiation-induced damage was reported in HgI₂ under the exposure conditions cited. We note, however, that the 1500 MeV data did not extend beyond a fluence of 1.2×10^8 protons/cm² which is equivalent to only about one year in earth orbit.

TABLE 1.
Radiation Damage in HgI₂ Room Temperature Semi-Conductor Detectors

Material	Radiation	Effects	Reference
HgI ₂	Proton/10.7 MeV	No ΔE loss @ 5.9 KeV after 10^{12} p/cm ²	1
	10.0 MeV	No effect on 5.5 MeV alpha pulse height up to 10^{10} p/cm ²	2
	1500 MeV	No ΔE loss @ 5.9 KeV up to 1.2×10^8 p/cm ²	3
	33 MeV	No ΔE loss @ 59.6 and 122 KeV up to 2.5×10^{10} p/cm ²	4
	Neutron/8 MeV	Little ΔE effect up to 10^{15} n/cm ² in 5.3 MeV alpha spectrum.	2
	Photon	No data	--

Cadmium Telluride

Damage studies were found for proton, neutron, and photon irradiations. The results are summarized in Table 2. Proton exposures at 33 MeV were made on chlorine and indium-doped material. This energy was selected so that the beam passed through the thickest samples (2.8 mm) and thus contributed no Bragg peak in the test samples. In chlorine-doped samples, the energy resolution degraded sharply after about 3×10^9 protons/cm² at photon energies of 59.6 and 122 KeV. There was some evidence of a slight resolution improvement at lower fluences. Evidence of a detector thickness dependence in the resolution response was also noted. While quantitative data were not obtained for the single indium-doped device tested, the data suggests a higher degree of radiation resistance than with the chlorine-doped material. Neutron irradiations were reported at an energy of 8 MeV. Significant reductions in energy resolution were reported after about 2×10^{10} n/cm² in 5.5 MeV alpha spectra. Photon irradiations were reported using Co-60 (1.17 and 1.33 MeV) with the resolution monitored at 662 and 59.6 KeV. Marked changes in peak shape were reported at both energies after exposure of several times 10^5 R. No data were found at intermediate and low exposure levels.

TABLE 2.

Radiation Damage in CdTe Room Temperature Semiconductor Detectors

Material	Radiation/Energy	Effects	Reference
CdTe	Proton/33 MeV	Chlorine doped — ΔE degradation after $\sim 3 \times 10^9 \text{ p/cm}^2$ @ 59.6 and 122 KeV in 0.9 mm thick sample Onset of ΔE loss in 1.75 mm sample @ 59.6 KeV near $3 \times 10^8 \text{ p/cm}^2$ Indium doped — evidence of greater radiation resistance, no quantitative data	4
	Neutron/8 MeV	p-type — increasing ΔE loss (5.5 MeV alpha) @ $> 2 \times 10^{10} \text{ n/cm}^2$	5
	Photon/1.25 MeV	Substantial ΔE loss @ 59.6 KeV after 10^5 R	6

Cadmium Zinc Telluride

Radiation damage studies on CZT have been conducted using both protons (1.3 and 200 MeV) and neutrons (moderated fission spectrum). The results of these studies are shown in Table 3. With 1.3 MeV protons, the bulk leakage was found to increase significantly after 10^{12} p/cm^2 in unbiased strip detectors (10 x 10 x 2 mm). The interstrip leakage increased significantly after about 10^{10} p/cm^2 . No energy resolution data were reported (Bartlett, 1996).

Considerable work has been reported in the region of 200 MeV. Varnell (1996) irradiated 2 and 3 mm thick planar detectors with up to $5 \times 10^9 \text{ p/cm}^2$ and found resolution losses in the thicker devices (3 mm). The initial (unirradiated) FWHM values at 59.6 and 122 KeV of 3.2 KeV and 3.9 KeV, respectively, degraded to 4.1 and 4.3 after 10^9 p/cm^2 and to 6.2 and 9.2 KeV after $5 \times 10^9 \text{ p/cm}^2$. The thinner device (2 mm) showed no degradation at either energy after like exposures, however. A downward shift in peak channel with increasing fluence was reported for both thick and thin detectors. Varnell attributes the resolution degradation to increased electron trapping.

In a similar study, Bartlett (unpublished data), exposed both strip and planar detectors to 200 MeV protons. The strip detectors (15 x 15 x 2 mm) were exposed under bias to fluences from 10^8 to $5 \times 10^9 \text{ p/cm}^2$. A small gain shift (3%) was noted after a fluence of $1 \times 10^9 \text{ p/cm}^2$ and a significant shift (>25 %) after $5 \times 10^9 \text{ p/cm}^2$. No consistent pattern of resolution degradation was

found. The resolution of a single detector exposed to 5×10^9 p/cm² was unchanged at a photon energy of 59.6 KeV and significantly degraded at 122 KeV while small losses (and gains) were found at 1×10^8 and 1×10^9 p/cm². In this study, the outputs of three strips in each detector were summed; one detector was used for each fluence level (1, 10, and 50×10^8 p/cm²). Two planar devices (10 x 10 x 2 mm and 15 x 15 x 2 mm) were exposed to a fluence of 5×10^9 p/cm², one under bias the other unbiased. Gain shift and energy resolution were measured at photon energies of 14.4, 17.8, 59.6, and 122 KeV. Gain shifts were found at all energies in both biased and unbiased cases. Significant (>45%) resolution losses were found at 59.6 and 122 KeV in the unbiased device. Minor changes (both positive and negative) were reported for the biased case.

Neutron irradiations with a moderated fission spectrum source (CF-252) at fluences up to about 10^{11} n/cm² on a single detector were reported (Bartlett, 1996). The detector (10 x 10 x 2 mm) was biased during exposure. No resolution degradation was found at photon energies of 14.4, 26.3, 59.6, and 122 KeV for fluences up to 10^{10} n/cm². Significant resolution losses were found after 7×10^{10} n/cm², however. It is interesting to note that the resolution losses were largely recovered after 12 weeks of annealing at room temperature. Evidence of neutron activation, in the form of gamma-ray lines from cadmium and tellurium isotopes, was apparent at fluences beyond about 10^{10} n/cm².

TABLE 3.

Radiation Damage in Cadmium Zinc Telluride (CZT) Room Temperature
Radiation Detectors

Material	Radiation/Energy	Effects	Reference
CZT	Protons/199 MeV	<u>3 mm thick detector</u> — ΔE @ 59.6 and 122 KeV degrades starting at 10^8 p/cm ² , 2 fold increase in ΔE @ 5×10^9 p/cm ² -- 2 mm thick detector — little ΔE change up to 5×10 p/cm ² -- Downward peak shift proportional to fluence in 2 mm and 3 mm devices -- Effects attributed to increased e-trapping	7
	Protons/200 MeV	<u>Strip detector (biased)</u> — <u>2 mm thick</u> -- Significant (>25%) gain shift @ 59.6 and 122 KeV from 5×10^9 p/cm ² -- ΔE loss @ 122 KeV after 5×10^9 p/cm ² — no ΔE effect @ 59.6 KeV <u>Planar (biased)</u> — <u>2 mm thick</u> -- Small ΔE effects, both positive and negative found @ 14.4, 59.6, and 122 KeV <u>Planar (unbiased)</u> — <u>2 mm thick</u> -- Large (>45%) ΔE losses at 59.6 and 122 KeV following 5×10^9 p/cm ² exposure -- Gain shifts in both biased and unbiased detectors at 14.4, 17.8, 59.6, and 122 KeV	8
	Proton/1.3 MeV	<u>2 mm thick strip detector (unbiased)</u> -- Bulk leakage increases significantly after 10^{12} p/cm ² -- Interstrip leakage increases after 10^{10} p/cm ² -- No spectral data	9

TABLE 3. (continued)

Radiation Damage in Cadmium Zinc Telluride (CZT) Room Temperature
Radiation Detectors

Material	Radiation/Energy	Effects	Reference
CZT	Neutron/Moderated Fission Spectrum	<u>Planar detection — (10 x 10 x 2 mm)</u> — No ΔE effects @ 59.6 or 122 KeV for fluence up to 10^{10} n/cm ² — Measurable ΔE loss @ 10^{11} n/cm ² — Activation emission (γ) from cadmium and tellurium isotopes appear at 10^{10} n/cm ² — Thermal annealing reverses most of ΔE loss in 12 weeks	10

Conclusions

Knowledge of the radiation susceptibility of the leading room temperature semiconductor detectors CZT, CdTe, and HgI₂, is at best fragmentary. Factors known to be of significance in semiconductor radiation damage such as rate effects (flux), incident energy, and device temperature have not yet been examined. Moreover, the available data are from a very small sampling of detectors (sometimes a single device) and do not, in general, cover the complete fluence range of interest.

Despite these shortcomings, several interesting features emerge from the existing data. The most apparent is that HgI₂ appears to be relatively immune to proton and neutron-induced radiation damage. No resolution degradation was found from intermediate energy protons at fluences up to 10^{12} p/cm². Similarly, no degradation was found from high energy protons although the effects of fluences significantly beyond 10^8 p/cm² have not been investigated and accordingly the suitability of HgI₂ for long-term space mission remains in question. Additionally, the material is apparently not susceptible to damage from intermediate energy neutrons.

The situation for cadmium telluride is less clear. No data were found for effects of high energy protons although the results at intermediate energy suggest vulnerability beginning in the region of 10^8 p/cm². Neutron data are also incomplete although at intermediate energies (8 MeV) the damage threshold for resolution degradation is relatively high (10^{10} n/cm²). While no evidence of activation was reported, effects similar to those in CZT can be expected.

The radiation susceptibility of CZT is also in question. There is evidence of resolution degradation from 200 MeV protons beginning in the region of 10^9 p/cm² as well as a downward shift in peak channel proportional to the proton fluence. However, the resolution degradation was apparent only in a 3 mm thick device and not a 2 mm detector. There is also evidence that the resolution degradation is dependent on bias conditions although this is based on the results from a single detector. Detector response changes following high energy proton irradiation are consistent with increased electron trapping and the associated decreases in the mobility-lifetime product. With intermediate energy protons (1.3 MeV), bulk and interstrip leakage was evident but only at high fluence levels. Damage for moderated fission neutrons is evident only after 10^{10} n/cm². Neutron activation lines from cadmium and tellurium isotopes appear after about 10^{10} n/cm². It is interesting to note that annealing at room temperature was very effective in restoring resolution losses.

Recommendations

A substantial amount of work remains before an understanding of the radiation susceptibility of room-temperature, semiconductor detectors emerges. Because of the great interest in deploying CZT-based instruments (and the apparent resistance of HgI₂) it is suggested that initial efforts be confined to developing a working knowledge of CZT related effects. Several areas are in particular need of near-term attention. Of particular interest are high energy proton-induced resolution losses in the fluence region beyond 10^8 p/cm² where a larger number of detectors encompassing a wider range of detector types is required. Questions concerning the effects of device thickness and bias conditions also need to be resolved. Further measurements at low and intermediate proton energies should be carried out to determine the vulnerability of the contact-CZT interface and validate the promising results obtained on a single device. Because of the potential for resolution degradation at relatively modest fluence levels, a practical annealing procedure should be developed.

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HOMOGENEITY OF CdZnTe DETECTORS

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We describe the current state of nuclear radiation detectors produced from single crystals of $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ (CZT), with $0.04 < x < 2.4$, grown by the vertical high pressure Bridgman (VHPB) method. The crystals investigated were grown commercially both in the USA and at the Institute of Solid State Physics, Chernogolovka, Russia. The CZT was evaluated by Sandia National Laboratories and the UCLA and CMU groups using proton-induced X-ray emission (PIXE), X-ray diffraction (XRD), photoluminescence (PL), infrared (IR) transmission microscopy, leakage current measurements and response to nuclear radiation. We discuss the homogeneity of the various CZT crystals based on the results from these measurement techniques.

1 Introduction

The addition of Zn, which has a lower vapor pressure than Cd, into the melt of Cd and Te during crystal growth helps to reduce the dislocation density [1]. For example, Zn has been used to successfully produce higher quality substrate material for epitaxial growth of HgCdTe infra red detectors [2]. Hence, $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ with $0.04 < x < 2.4$ was found to have relatively high electrical resistivity (10^{10} - 10^{11} ohm-cm) particularly if the material is produced through vertical high pressure Bridgman (VHPB) growth [3-9]. Therefore, CZT is used also for nuclear radiation detector material with much better results than reported for CdTe [10-17]. The energy band gap dependency for $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ reported by Olega et al. [18-19] is $E_g(x) = 1.510 + 0.606x + 0.139x^2$ eV at 300 K. The band gap energy of $\text{Cd}_{0.96}\text{Zn}_{0.04}\text{Te}$ is near 1.534 eV while the band gap

energy for $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ is near 1.637 eV. Elevated temperature tests on CdZnTe

energy for $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ is near 1.637 eV. Elevated temperature tests on CdZnTe detectors yield an activation energy of 0.8 eV, indicating that the material is most likely compensated by a balance between deep and shallow levels [20]. The charge carrier mobilities for $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ have been measured to be $120 \text{ cm}^2/\text{V-s}$ for holes and $1350 \text{ cm}^2/\text{V-s}$ for electrons [21]; the measured charge carrier mean free drift times are 200 ns for holes and 100 ns for electrons, thus producing a case in which position dependent charge collection can degrade energy resolution. Optical measurements also indicate that the electric fields in CZT devices with Schottky contact junctions are non-uniform, which can also effect detector performance [22].

We have evaluated CZT detector crystals grown in the USA and in Russia by VHPB [8-10]. The difference between USA and Russian CZT is the USA material has always been prepared with an excess of Cd+Zn and labeled "n-type" CZT. On the other hand, Russian CZT has been prepared in both n- and p-type (excess Te) forms. The p-type has proved to be much more uniform than any of the (Russian and/or USA) n-type material. However, the crystallinity and nuclear radiation response of the p-type CZT was much poorer than the US-produced material. The present paper describes some recent homogeneity and stoichiometry results obtained on USA and Russian CZT using various analytical methods to evaluate these materials.

2 Experimental

2.1 n- and p-type CZT

As mentioned above, we define n- or p-type material according to the excess of either Cd+Zn or Te, respectively. Determination of the dominant electrical carrier (electrons for n-type and holes for p-type) is difficult for the CZT crystals because one cannot easily perform Hall effect measurements on high resistivity semiconductors. Thus, we define p-type CZT as material whose atomic ratio $r = \text{Te}/(\text{Cd}+\text{Zn})$ is greater than 1 and whose photoluminescence (PL) spectrum is dominated by acceptor bound exciton recombination. When $r < 1$ and the donor bound exciton is significantly larger than the acceptor bound exciton, we refer to the material as n-type.

The Russian p-type CZT was prepared using pre-synthesized ZnTe and CdTe mixed in the desired proportion of 5N purity Zn, Cd ("ELMA", Russia) and Te ("Pure metal plant", Ukraine). An excess of 1% tellurium was added to the ZnTe and CdTe mixture synthesized at 980°C and 1100°C respectively. The crystals were then grown using the high pressure vertical Bridgman method [8] under 8 MPa argon pressure and a crucible pulling rate of 2 mm/h. The Russian n-type CZT was prepared in a similar way to the USA n-type material by directly mixing the elements

Cd, Zn and Te. Some of the results obtained on both n-type and p-type Russian CZT are published elsewhere [8,9].

2.2 X-ray Analysis

Triple axis x-ray diffraction (TAD) techniques [23] have been used. The composition was determined by measuring the absolute lattice parameter of the $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ alloy and assuming Vegard's law between CdTe and ZnTe. The crystallinity was assessed through TAD rocking curve measurements. A Bede D3 high resolution x-ray diffractometer was used with a copper tube, and the beam conditioning optics included a Si (111) channel cut collimator, a Si (111) monochromator, and a Si (111) or Si (220) analyzer crystal. The compositional variations along the axial direction of the boules for the p-type $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ samples with $x = 0.2$ were studied, and the results showed a high degree of homogeneity [8,9]. The compositional variations were on the order of 1-2% throughout a large region (70-90%) which are indicative of excellent uniformity. By comparison, both USA and Russian grown n-type CZT showed very large variations, e.g., an axial variation of Zn from $.05 > x > 0.13$ for a nominal composition of $x = 0.1$

(a)

(b)

Figure 1. Comparison of the TAD results of (a) compositional homogeneity and (b) mosaicity of p-type Russian and n-type USA produced CdZnTe. In (a), the error bars represent typical variations within a single boule. In (b), the typical ranges of mosaic spread for representative good crystalline quality materials are shown in terms of full width at 1% maximum of TAD rocking curves.

Figure 1 compares the TAD homogeneity results of the axial distribution of the Zn for representative Russian p-type CZT and USA n-type CZT. The TAD measurements confirmed that only small variations could be found (mostly within 10% of the intended composition), in the p-type compared to ~70% of its nominal value found in USA n-type CZT crystals, in which the zinc segregates approximately according to the normal freezing conditions. The nonhomogeneity of the Russian n-type CZT behaves in a similar way. Unfortunately, the crystallinity of the p-type samples examined by TAD was found to be significantly worse than that of the n-type crystals grown in the USA. These results are shown in Figure 1(b), in which the mosaic spread values (in terms of full width at 1% maximum of TAD rocking curves) for representative good crystal quality samples are compared. The mosaic spread and the typical range are significantly smaller for the USA produced CZT, indicating better crystal quality.

2.3 Photoluminescence studies

PL spectra were taken on both p- and n-type CZT. The sample is suspended above a liquid helium reservoir at 4.2K. The excitation was provided by the 514.5 nm line of an argon-ion laser. The laser is focused to a spot of approximately 1 mm diameter and the total power incident on the sample was approximately 15 mW. The luminescence was collected into a 0.85 m double-pass spectrometer and detected with a high-purity Ge PIN diode detector. The light was chopped at 750 Hz and the signal was processed with a lock-in amplifier.

PL spectra were taken at several points on the sample; all points were qualitatively similar for the p-type CZT. Figure 2 shows a representative comparative spectrum of a p- and n-type CZT. It can be seen that the PL spectrum is dominated by acceptor bound exciton recombination for p-type and the donor bound exciton PL is significantly larger than the acceptor bound exciton PL for n-type CZT. The spectrum also reveals that p-type material has a relatively low quality, as evidenced by the weakness of the excitonic bands as compared to donor-acceptor and defect-related bands. Further, in the n-type CZT there was significant variation in the peak position of the donor-bound exciton (D0,X) peak, indicating substantial composition variation.

Fig. 2: PL spectra at 4.2K of a p-type Russian and n-type CZT material

2.4 PIXE studies

Proton-induced x-ray emission (PIXE) stoichiometric maps were obtained on Russian p- and n-type crystals using a 3 MeV proton beam focused to 100 microns [23]. Nondestructive PIXE analysis simultaneously records the characteristic elemental x-rays emitted from a sample as the accelerated and focused ions interact locally with the target atom electrons.

Two-dimensional maps of $Zn/(Cd+Te)$ ratios are formed by analyzing the measured X-ray yields (proportional to concentration) in each pixel of the region scanned on the samples. Figure 3 shows the PIXE mapping of the $Zn/(Cd+Te)$ variation in the Russian p- and n-type samples. The average value over the 5 mm x 5 mm region (100 micron pixels) scanned on the p-type material was 0.133 (the corresponding average

Table 1: (Zn+Cd)/Te ratio measured in various CZT samples by PIXE

CZT Sample	(Cd+Zn)/Te ratio
US n-type	1.070
US n-type	1.075
Russian p-type	0.889
Russian n-type	1.023

(Zn+Cd)/Te ratio over this same region is 0.889 as given in Table 1 below). For the n-type CZT, the average Zn/(Cd+Te) ratio was 0.123 over the 1 mm x 1 mm scanned area analyzed by PIXE. The average composition of the p-type Russian sample was determined to be $\text{Cd}_{0.71}\text{Zn}_{0.23}\text{Te}_{1.06}$ which means there is an excess of 6% Te. In comparison, the composition of the Russian n-type was measured to be $\text{Cd}_{0.79}\text{Zn}_{0.22}\text{Te}_{0.99}$. Table 1 gives the (Cd+Zn)/Te ratios for p- and n-type Russian CZT material, and n-type CZT material received from two commercial US vendors. One can see clearly that n-type has an excess of (Cd+Zn), whereas p-type has an excess of Te.

Figure 3. Two-dimensional particle-induced x-ray emission (PIXE) spatial maps of the Zn/(Cd+Te) ratios in p- and n-type Russian CZT samples. The data were collected using 3 MeV protons focused to a spot size of 100 microns. The variation in stoichiometric ratio is about 8% (p-type) and 6% (n-type) between the light and dark pixels of each respective image.

2.5 IR Microscopy, Leakage Current and Charge Transport Mapping

The main defects identified in IR micrographs of Russian p- and n-type crystals are pipes and bubble arrays. Fig. 4 shows an IR micrograph of such pipes of 5-10 mm length and 30-50 micron wide. The pipes may contain precipitates or may be voids formed in the material by volatilization of Ar, Cd or other high vapor pressure impurities that could

Fig. 4: IR micrograph of n-type Russian CZT detector showing five different 30-50 micron wide and 5-10 mm long tubular voids.

released during cooling. The US detector grade material has much smaller round shaped precipitates of about 10 microns diameter and which are less elongated or pipe shaped. It is possible that the elongated tubular precipitates found in the Russian CZT stem from non equilibrium cooling and a relatively lower HPVB gas overpressure, as compared with the US material. IR transmission microscopy was also used for mapping the homogeneity of n-type USA produced CZT. Figure 5 shows an example of one of these mapping results performed on a $\sim 100 \text{ cm}^2$ center slice CZT wafer. Shown are also: (a) leakage current mapping, (b) maximum alpha particle pulse height mapping, and (c) infrared transmission image prior to preparing the sample as a detector. The leakage current varies by 3 orders of magnitude within the detector. Obvious correlation exists, as excessive leakage currents are measured along the dark regions in the infrared transmission image. These dark regions are places where microcracks or pipes run through the material, as can also be inspected visually. Common feature to these microcracks is that they propagate from the outer edge of the wafer, i.e., from the crystal-to-crucible wall interface. This is an indication that the stress built up at this interface during crystal cool down period might is

relieved through mechanical cracking. In (b), the scale is given as the maximum alpha particle pulse height (peak channel). The data can be converted to the electron mobility and life time $\mu\tau$ product, and the low and high values correspond to $0.5 \times 10^{-3} \text{ cm}^2/\text{V}$ and $6.4 \times 10^{-3} \text{ cm}^2/\text{V}$, respectively. The greater part of the entire wafer area, as can be seen from the figure, exhibits low electron $\mu\tau$, unacceptable for spectrometer grade detectors. Also, regions of high leakage currents correspond to regions of low $\mu\tau$ values, indicating that noise is limiting the detector performance. Thus, the IR data correlate also to detector performance results.

Figure 5. Comparison of (a) leakage current mapping, (b) maximum alpha particle pulse height mapping, and (c) infrared transmission image of a $\sim 100 \text{ cm}^2$ center slice $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ wafer.

2.6 Nuclear Spectroscopic Response

This section will briefly review some results reported by the Sandia group and elsewhere with n-type CZT devices. For example, 5 mm x 5 mm x 1.5 mm detectors in which RT energy resolution was 2.89 keV FWHM for 60 keV gamma-rays and 6.9 keV FWHM for 122 keV gamma-rays have been reported. Thicker detectors generally suffer from incomplete hole collection, thus leading to gamma ray pulse height spectra skewed towards lower energies.

Several methods have been proposed to correct and reduce position dependent charge collection effects, the most promising being pulse processing techniques [10,26,27] and single charge carrier sensing devices [28]. Presently, pulse processing techniques have been used with a 18 mm x 18 mm x 2 mm $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ device to demonstrate RT energy resolution of 9.3 keV FWHM for 662 keV gamma rays [10], and with a 18 mm x 18 mm x 5 mm $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ detector to demonstrate RT energy resolution of 11.25 keV FWHM for 662 keV gamma rays [26]. Similar pulse processing techniques used for a 0.6 mm thick and 4 mm² $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ detector at reduced temperature (-25°C) yielded energy resolution of 2.6 keV FWHM for 662 keV gamma rays [27].

Fig. 6: Spectrum of alpha particles emitted by ^{241}Am showing the 5 MeV alpha photopeak on a 3.5 thick n-type CZT detector, with a FWHM resolution of 8%.

Single charge carrier sensing devices have been successfully used with a $5 \times 5 \times 5 \text{ mm}^3$ CdZnTe detector to demonstrate RT energy resolution less than 16.0 keV FWHM for 662 keV gamma rays [28]. The gamma-ray detection efficiency can be greatly enhanced by coupling multiple detectors in a stack [29]. However, the charge collection efficiency for each device is slightly different which gives a separate full energy peak in the pulse height spectrum from each detector for the same gamma ray energy. The problem is easily corrected with a shifting circuit which allows the pulses from all of the detectors to be summed through a single preamplifier without the appearance of multiple peaks for any single energy [29]. Room temperature operated $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ detectors have recently been used to detect the emission lines from the ^{113}Cd and ^{114}Cd which were clearly identified [30]. The $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ device demonstrated thermal neutron sensitivity of $3.7 \pm 1.9 \%$ for the 558.6 keV prompt gamma-ray emissions. Recent studies indicate that CdZnTe is capable of withstanding fast neutron fluent up to 10^{10} n/cm^2 without suffering significant damage [31]. However, activation gamma-ray spectra and resolution degradation become apparent at fluent above 10^{10} n/cm^2 .

3 Conclusions

The p-type CZT grown in Russia is more compositionally homogeneous than n-type CZT produced in the USA. However, their electrical charge transport properties are much worse, probably due to much shorter carrier lifetimes and sometimes to higher leakage currents, both of which prevent the fabrication of good quality spectrometers. It may be that defects such as long tubular pipes, bubbles, and precipitates deteriorate the electrical properties by providing pathways for electrical conductivity and therefore more leakage current under bias. Further work will try to achieve both uniform homogeneity and improved charge transport properties to produce large volume nuclear detectors from CZT.

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Development of a Portable X-ray and Gamma-ray Detector Instrument and Imaging Camera for Use in Radioactive and Hazardous Materials Management

J. M. Van Scyoc¹, R. B. James¹, R. J. Anderson¹, A. J. Antolak¹, X. J. Bao⁸, G. S. Bench⁶, D. Boehme¹, B. A. Brunett³, G. Buffleben¹, A. Burger⁴, Y.-C. Chang¹¹, K.-T. Chen⁴, E. Cross¹, M. DeVries¹², T. S. Gilbert³, M. S. Goorsky¹⁰, H. Hermon¹, N. R. Hilton⁹, J. S. Iwanczyk¹³, L. A. Lim¹², J. C. Lund¹, J. M. Markakis¹, D. McArthur², D. S. McGregor¹, J. E. McKisson⁷, D. L. Medlin¹, D. H. Morse¹, M. Natarajan⁸, R. W. Olsen¹, B. E. Patt¹³, C. Perrino¹, A. E. Pontau¹, M. Roth⁵, L. Salary⁴, M. Schieber¹, T. E. Schlesinger³, H.-K. Sim¹¹, R. H. Stulen¹, E. Soria¹, J. E. Toney³, J. O. Weeks¹, H. Yao¹², H. Yoon¹⁰

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Abstract

The overall goal of this LDRD project was to develop instruments for use in the management of radioactive and hazardous wastes. Devices for identifying and imaging such wastes are critical to developing environmental remediation strategies. Field portable units are required to enable the on-site analysis of solids, liquids, and gas effluents. Red mercuric iodide (α -HgI₂) is a semiconductor material that can be operated as a high-energy-resolution radiation detector at ambient temperatures. This property provides the needed performance of conventional germanium- and silicon-based devices, while eliminating the need for the cryogenic cooling of such instruments. The first year of this project focused on improving the materials properties of the mercuric iodide to enable the new sensor technology; in particular the charge carrier traps limiting device performance were determined and eliminated. The second year involved the development of a field portable x-ray fluorescence analyzer for compositional analyses. The third and final year of the project focused on the development of imaging sensors to provide the capability for mapping the composition of waste masses. This project resulted in instruments useful not only for managing hazardous and radioactive wastes, but also in a variety of industrial and national security applications.

Acknowledgment

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Preface

The ability to detect, identify, and image various forms of hazardous, radioactive, and mixed wastes is critical for the development and implementation of environmental remediation programs. Detailed information about elemental and radionuclide content, concentration, and distribution are required to effectively localize and process these wastes. One of the most common methods for elemental analyses is x-ray fluorescence spectroscopy (XRF), which can be used to identify and quantify elements from a large portion of the periodic table to the part-per-thousand level. For the identification of radionuclides, energy spectroscopy is used. While both of these techniques are readily available in any standard analytical laboratory, they have been traditionally either unavailable or impractical for field use. In order to obtain the high energy resolution required for these applications, semiconductor detectors are required. For XRF, the conventional x-ray spectrometer used is lithium-drifted silicon (Si:Li), while for the energy spectroscopy of radionuclides high-purity germanium (HPGe) is the choice. Both of these semiconductor detectors require cryogenic cooling (liquid nitrogen at 77K) to operate, as the thermal leakage currents in these devices at ambient temperatures overwhelm the signal currents. In addition to the requirement of cooling during operation for both Si:Li and HPGe, Si:Li detectors also generally require continuous cooling during storage to avoid device degradation. These cryogenic cooling requirements introduce weight, size, maintenance, and preparation-time issues to the associated instruments, which tend to limit their use in the field.

In recent years there has been a push to develop alternative semiconductor materials that eliminate these problems. Of these room-temperature semiconductor detector materials, red alpha-phase mercuric iodide (α -HgI₂) has shown the greatest promise, particularly for the demands of XRF on resolving low-energy x-rays. In addition, mercuric iodide has the capability to efficiently detect and identify higher energy gamma photons from radionuclides. An instrument built around a mercuric iodide sensor element could be used for XRF and gamma spectroscopy in the field in the desired "any-time, any-where" mode of operation.

However, the main obstacle to the implementation of such systems has been the availability of high quality mercuric iodide devices. The trapping of electrical charge carriers in the bulk crystal reduces the energy resolution and low energy spectroscopy capabilities of the detector. In addition, the effects of mobile impurity-related defects in the device leads to polarization phenomena, which are the changes in device response versus time. Therefore, there have been significant efforts expended on identifying and eliminating the impurities and other charge-trapping defects.

Once the material had reached a sufficient development level, it was possible to fabricate prototype detectors and instruments. The first instrument developed was a portable XRF unit for the field analysis of toxics in various sample forms. The XRF instrument was designed to be carried and operated by a single technician for a day. The only maintenance requirements are the recharging of the batteries at the end of the workday. Once this instrument was implemented the focus shifted to imaging devices that could be used to determine the distribution of radionuclides in the environment. Because of the semiconductor operation of mercuric iodide detectors, it is possible to use lithographic fabrication procedures to yield a detector device with high spatial resolution as well as good energy spectrum resolution.

The work to improve mercuric iodide materials and develop prototype portable XRF analyzers and imaging instruments is discussed in this report.

Summary

The end goal of this project was to provide a capability in the characterization of hazardous and radioactive wastes that is currently unavailable. New instruments based on mercuric iodide nuclear radiation spectrometers that can operate at ambient temperatures were developed in order to provide detailed analytical capabilities in the field. The primary enabling technology for such instruments is the mercuric iodide sensor itself. Therefore, the first year of this project focused on improving the yield and performance of mercuric iodide detectors by reducing the number of charge trapping defects. The second year focused on developing and building a prototype portable x-ray fluorescence spectroscopy analyzer for use in the elemental characterization of toxics in the environment. The third and final year of the project focused on developing and building a prototype gamma-ray imaging camera to be used in determining the spatial distribution of radioactive wastes in a given region. The imaging system consists of an orthogonal strip detector made from HgI_2 crystals. The detectors utilize an 8×8 orthogonal strip configuration with 64 effective pixels. The detector arrays are 1 cm^2 devices with a strip pitch of approximately 1.2 mm (producing pixels of $1.2 \text{ mm} \times 1.2 \text{ mm}$). The readout electronics consist of parallel channels of preamplifier, shaping amplifier, discriminators, and peak sensing ADC. The preamplifiers are configured in hybrid technology, and the rest of the electronics are implemented in NIM and CAMAC with control via a Power Macintosh computer. The software used to readout the instrument is capable of performing intensity measurements as well as spectroscopy on all 64 pixels of the device. We report on the performance of the system imaging gamma-rays in the 20-500 keV energy range and using a pin-hole collimator to form the image. Taken together, these instruments provide new tools that can be provided to workers responsible for characterizing and remediating hazardous and radioactive wastes.

Nomenclature

ADC	analog to digital converter
AES	Auger electron spectroscopy
AFM	atomic force microscopy
ASIC	application specific integrated circuit
α -HgI ₂	alpha-phase mercuric iodide
CAMAC	Computer Automated Measurement and Control
CMOS	complementary metal-oxide-semiconductor
DIP	dual in-line package
DSC	differential scanning calorimetry
EDP	electrodrift purification
GUI	graphical user interface
HPGe	high purity germanium
ICP-AES	inductively-coupled atomic emission spectroscopy
ICP-MS	inductively-coupled mass spectroscopy
MCA	multichannel analyzer
NEST	Nuclear Emergency Search Team
NIM	Nuclear Instrumentation Module
PICTS	photo-induced current transient spectroscopy
PIXE	proton-induced x-ray emission
PL	photoluminescence
PMT	photomultiplier tube
PR	photoresponse mapping
RSM	reciprocal space map
Si:Li	lithium-drifted silicon
SNM	special nuclear material
TAXRD	triple axis x-ray diffraction
TCT	transient charge technique
TEM	transmission electron microscopy
TSC	thermally stimulated currents
VASE	variable angle spectroscopic ellipsometry
XRF	x-ray fluorescence spectroscopy
XRDT	x-ray diffraction topography
Z	atomic number

Development of a Portable X-ray and Gamma-ray Detector Instrument and Imaging Camera for Use in Radioactive and Hazardous Materials Management

Introduction

There are many applications in hazardous and radioactive materials management where the ability to characterize in real-time the elemental and radiological composition of a waste mass can greatly enhance the effectiveness and efficiency of an operation. Instruments conventionally available for detailed elemental and radiological identification and quantification are generally too bulky and maintenance-intensive for practical use in the field. The conventional field portable devices have too poor of performance for the detailed analyses required. Therefore, the goal of this project was to develop new tools based on the room-temperature nuclear radiation spectrometer mercuric iodide. The two tools most of use are a portable x-ray fluorescence (XRF) analyzer and a radionuclide imaging camera, both of which have been developed.

Background

There are many types of devices available for detecting x-rays and gamma-rays over a wide energy range. Gas-filled detectors generally have too low a sensitivity density for anything other than very low energy x-rays and can be damaged by rough handling; therefore, solid-state devices are most appropriate for portable applications. While scintillators can have a high detection efficiency, their energy resolution is too poor for the spectroscopy capabilities required. Energy resolution is most obviously important for separating closely spaced emissions. As Figure 1 shows, however, it is also important for detecting weak signals in a noisy background, as may be found in the complicated environment of hazardous and radioactive wastes measurements. Semiconductor detectors are then the device of choice for applications requiring energy spectroscopy. The conventional semiconductors of choice have become lithium-drifted silicon (Si:Li) for x-rays and high-purity germanium (HPGe) for gamma-rays. While these two materials provide for devices with ultra-high energy resolution, they require continual cryogenic (liquid nitrogen at 77K) cooling during operation. This limits their applicability for field portable units.¹

Given this, there has been considerable interest in developing semiconductor detectors that operate at room temperature. Several materials have been considered, but only a select few have shown sufficient promise to warrant advancement to the point where they can be applied. Mercuric iodide (HgI_2) has many optimal properties which have lead to it being the leading candidate material for many applications.²⁻⁴ HgI_2 has a large bandgap of 2.13eV at room-temperature which leads to a high electrical resistivity on the order of $10^{13}\Omega\cdot\text{cm}$.⁵ This high resistivity leads to very low dark currents in the detector, which reduces the amount of noise degradation in the output spectrum. This is particularly critical for XRF where high energy resolution is required for low energy x-ray photons down to a few keV. Mercuric iodide also has a high average atomic number (Z), which yields a high efficiency for stopping higher energy gamma-photons. This is useful for the spectroscopy and imaging of radioactive materials which can emit photons with energies up

to several hundred keV. Therefore, mercuric iodide has been known as an excellent possibility for use in spectrometers designed to operate anywhere in the x-ray and gamma-ray range for many years.⁶

However, mercuric iodide has several problems that limit the immediate implementation of the desired characterization tools. The material itself is mechanically soft and has aging problems associated with the sublimation of material from surfaces exposed to air. In addition, the material is significantly degraded if heated much above room-temperature, and cannot be grown from the melt. These problems are merely issues that complicate the growth and processing of the device, but that have been addressed. The more pressing issue, however, is the electrical performance of the semiconductor. Figure 2 shows the basic operation of a mercuric iodide detector. An incoming photon creates a photoelectron which rapidly passes its energy into the creation of electron-hole pairs. These mobile charge carriers are then swept in opposite directions by the applied electric field. The motion of these carriers gives rise to a current signal in the device leads and external circuit, which is then the signal measured and quantified. Therefore, the critical step in the operation of the device is the successful transport of the carriers from their creation point through the bulk of the mercuric iodide to their respect electrodes. In mercuric iodide, there are charge carrier traps that remove the carriers from their transport, thus degrading the signal measured. These traps can be caused by crystallinity defects, impurities, etc. The key to providing mercuric iodide-based instruments, then, is the improvement of the material to reduce these charge-causing defects to an acceptable level.

Figure 1. Simulated gamma-ray spectra showing the effect of detector energy resolution on the detectability of minority constituents in a strong background. (From Reference 1)

Figure 2. Schematic of mercuric iodide nuclear radiation detector operation.

Improvement of Mercuric Iodide Materials

As discussed above, charge trapping has been the primary problem limiting the application of mercuric iodide to radiation detector purposes. The effects of trapping can be seen clearly in the spectral performance of the devices for the radioisotope americium-241 (^{241}Am), as shown in Figure 3. ^{241}Am emits gamma photons of one midrange photon energy, 59.54keV, and several lower energy x-ray peaks from the de-excitation of daughter products. The spectral peaks, which optimally would be very sharp and symmetric Gaussian-shaped, are broadened and asymmetric. This is most clearly seen at the 60keV line, where a low-channel-side "hole-tailing" occurs. The hole-tailing results from some of the detected events having decreased pulse height due to the trapping of the transporting holes.

Figure 3. Spectrum of ^{241}Am from a $1 \times 1 \times 0.1 \text{ cm}^3 \text{ HgI}_2$ detector exhibiting hole tailing.

It is possible, however, to produce mercuric iodide detectors which exhibit excellent spectral properties. Figure 4 shows the spectrum from the same radioisotope, but with a detector actually used in one of the instruments developed for this project. Here the peaks are much sharper and more symmetric. The hole-tailing is still visible, but it is now at a level that does not prevent the usage of the detector for high resolution spectroscopy. A new "peak" does show up, but this is related to Compton scattering processes, and is intrinsic to the geometry of the measurement.

The key for providing the desired instrumentation is the improvement of the charge transport properties in the mercuric iodide. This is achieved by improving the materials properties of bulk mercuric iodide. The approach for doing this is to couple basic materials measurements with detector evaluations to determine the materials properties most negatively affecting device performance. A wide variety of materials measurements were applied to this problem. These tools included high-resolution triple-axis x-ray diffraction (TAXRD) and x-ray diffraction topography (XRDT), transmission electron microscopy (TEM), atomic force microscopy (AFM), proton-induced x-ray emission (PIXE), quantitative chemical analyses such as inductively-coupled atomic emission spectroscopy or mass spectroscopy (ICP-AES and ICP-MS), photoluminescence spectroscopy (PL), differential scanning calorimetry (DSC), transient charge technique (TCT), thermally stimulated currents (TSC), photo-induced current transients spectroscopy (PICTS), Auger electron

spectroscopy (AES), and variable angle spectroscopic ellipsometry (VASE). The primary device evaluation tools included current-voltage curves (I-V), photoresponse mapping (PR), and nuclear spectroscopic response. The following paragraphs highlight a few examples of these measurements.

Figure 4. Spectrum of ^{241}Am from a $5 \times 5 \times 0.5 \text{ mm}^3$ HgI_2 detector, demonstrating the excellent energy resolution possible in these devices (600eV at 60keV, or 1.0%).

One origin for charge-trapping defects is in the imperfection of the crystal structure of the bulk mercuric iodide. Any perturbations in the regularity of the atomic structure and its associated potential field can lead to the formation of trapping centers. These defects can range from simple point defects such as vacancies or interstitials through line defects such as dislocations to planar and three-dimensional defects such as grain boundaries. Changes in local lattice parameter lead to strains which also can introduce traps. Triple axis x-ray diffraction (TAXRD) is a method that can give very detailed information on the localized crystalline perfection. It can separately quantify the amount of lattice strain, which is changes in atomic spacing, and lattice mosaicity, which can be viewed as the variation in tilts for small sub-crystals. Figure 5 shows TAXRD-measured reciprocal space maps (RSM) for good and poor performing mercuric iodide detectors.

Another potential origin of charge traps is from impurities in the mercuric iodide. A wide range of elements and molecules can end up incorporated into the mercuric iodide during growth or subsequent processing. Certain metallic elements, such as copper (Cu) and silver (Ag) are particularly problematic, as they introduce device-killing traps and have a high diffusivity in mercuric iodide, even at room temperature. Any Cu or Ag that comes in contact with the mercuric iodide will readily diffuse into the bulk. One place that these impurities can be introduced is during the chemical processing required to fabricate detectors. A simple, yet powerful characterization tool that has been widely applied to mercuric iodide is low-temperature photoluminescence (PL). In PL the cryogenically cooled (liquid helium at 4.2K) sample is excited by a laser beam to produce a large number of free carriers; as these carriers recombine they re-emit photons of different energies or wavelengths whose spectrum is captured. The wavelength of these emissions gives the energy level of the associated radiative trap, and the intensity is related to the trap level density. Figure 6 shows the PL spectrum from mercuric iodide before and after exposure to a Ag-doped etchant solution. The appearance of a new peak, whose wavelength is that of the trap level

associated with silver impurities, indicates that the silver from the etchant has been incorporated into the bulk mercuric iodide.

Figure 5. Triple axis x-ray diffraction (TAXRD) analysis of HgI₂ revealing differences between "good" (spectrometer) and "bad" (counter) detectors. The reciprocal space maps (RSM) separate the diffraction broadening due to mosaicity (ω axis) and strain ($\omega-2\theta$ axis), with the good detector having a much sharper, more intense peak. The RSM on the left (a) shows both the good and bad devices, while the RSM on the right (b) is a blowup of just the bad device. The angular FWHM (on the ω axis) for the good detector is 4 arcseconds, while it is 170 arcseconds for the bad one.

Figure 6. Photoluminescence analysis of HgI₂ revealing introduction of Ag from the etchant.

The problem of impurities introduced during processing can be reduced by using very high purity processing materials. However, there are also impurities introduced into the original mercuric iodide starting materials that

are incorporated during crystal growth. It is most critical to remove these impurities prior to the growth of the actual detector crystal, not only for the elimination of the impurity itself from the detector, but also to remove the impurity's impact on the growth process. A new method for removing certain electrically active and highly detrimental impurities has been developed and patented.⁷ This method relies on the electrically charged and mobile nature of these impurities in the bulk mercuric iodide; these impurities are "swept" from the mercuric iodide center section into sacrificial end regions. A useful method for quantifying the impurities in mercuric iodide has been inductively-coupled mass spectroscopy (ICP-MS). Figure 7 shows the results of an electrodrift purified mass of mercuric iodide, as measured by ICP-MS, demonstrating purification by a factor of 100.

Figure 7. Chemical analysis (ICP-MS) results quantifying the electrodrift purification removal of Ag from a bulk polycrystalline sample of HgI_2 .

These and other types of measurements done on mercuric iodide are discussed further in Reference 8.

Development of a Mercuric Iodide Portable X-ray Fluorescence Analyzer

X-ray fluorescence spectroscopy (XRF) is a very commonly used technique for elemental analyses of various forms of materials in the laboratory. XRF involves the excitation of the inner shell electrons of the atoms in the sample by means of an incident x-ray beam, generally from an electrical x-ray tube. As these electrons return to their ground (unexcited) state, they emit their excess energy in the form of electromagnetic photons in the x-ray range. Because the electrons can only exist in finite, quantized energy states, the energy of these emitted x-rays is a constant for the de-excitation of a given electron state. Because the energies of these states are different for each element, the energies of the emitted x-rays are characteristic for each element. Each element will emit a set of x-rays at a well-defined set of energies. In general, the lower two series of these sets of emissions -- the K and L shell level emissions -- will be readily excitable and detectable. Within each of these series are several energies. The analysis of XRF, then, is essentially the pattern matching of an unknown, complicated spectrum with the underlying elemental spectra. In order to do this, the x-ray detector must be sensitive to a wide range of x-ray energies (1keV to 100keV), and have very high energy resolution (a few 100s of eV) in order to separate closely spaced peaks.

For a field portable unit, an electrical x-ray tube excitation source is impractical, so radioisotopes are used. With the use of three different radioisotopes, a large fraction of the periodic table is accessible, from sodium (Na, $Z = 11$) to Uranium (U, $Z = 92$). Iron-55 (^{55}Fe), with emissions around 6keV is useful for the low atomic number elements, cadmium-109 (^{109}Cd) is useful for the mid-range (transition metal) elements, and americium-241 (^{241}Am) covers the high atomic elements. A measurement sequence involves taking a separate spectrum with each of these three sources for a completely unknown sample, or fewer of the sources if only a few elements in the sample are of interest.

The detector is a mercuric iodide spectrometer, that is cooled slightly (along with the preamplifier input stage) with a low power thermoelectric cooler to further improve energy resolution. The detector is kept under a continuous bias of about 1000V (1000 to 2000V per mm of detector thickness) at all times to avoid polarization problems. Because of the very small dark currents in mercuric iodide detectors, the battery used for this biasing lasts about six months. The thermoelectric cooler and signal chain electronics -- preamplifier, shaping electronics, analog to digital converter (ADC), and multichannel analyzer (MCA), along with the digital processing electronics and computer are powered by rechargeable nickel-cadmium batteries designed for an eight-hour use before recharge. The entire unit can be readily carried by a field worker, with the computer, ADC, and MCA in a shoulder-strapped box, and the detector, preamplifier, and sources in a hand-held probe head. Figure 8 shows a schematic of the instrument configuration.

Once the portable XRF instrument had been assembled, several samples were studied to characterize its capabilities. Figure 9 shows the spectrum obtained with the ^{55}Fe source on a sample of potassium chloride (KCl) salt. As can be seen, the K and Cl K_{α} peaks are readily separable. The low channel noise peak is just starting to rise at 1keV, indicating that the detector could be used to detect the 1.04keV K_{α} line of Na. This demonstrates the capabilities at the low end of the periodic table.

Figure 8. Schematic of the design of a mercuric iodide portable XRF analyzer.

Figure 9. Spectrum of a KCl salt sample, taken with a $5 \times 5 \times 0.5 \text{ mm}^3$ HgI₂ detector.

Figure 10 demonstrates the midrange capabilities of the instrument with the spectrum of a gallium arsenide (GaAs) sample excited with the ¹⁰⁹Cd source. The K α and K β lines from the Ga and the As are visible and mostly separable.

Figure 10. Example x-ray fluorescence (XRF) spectrum taken with the field portable instrument based on a HgI_2 high-resolution spectrometer. This spectrum shows the toxic elements Ga and As as found in GaAs.

Figure 11 shows the spectrum from a more complicated sample, a piece of old lead-based paint. Here the emissions from the various elements are again well separated.

Figure 11. X-ray fluorescence spectrum from an old paint sample illuminated with ^{109}Cd x-rays.

Finally, Figure 12 shows the spectrum of higher atomic number elements from a complicated geological samples. The mercuric iodide detector provides the previously unavailable field capability of separating out for identification and quantification the uranium (U) content.

Figure 12. Spectrum of a geological sample containing several elements, including U, Cd, Sn, Cs, and Ba, taken with a $5 \times 5 \times 0.5 \text{ mm}^3$ HgI_2 detector.

Development of a Mercuric Iodide Imaging Camera

Devices for imaging the spatial distribution of gamma-ray emitting isotopes have been in wide use for many years, particularly in nuclear medicine. Gamma cameras are also critical in the imaging of radioactive waste stored in a variety of different containers. The conventional gamma imaging instrument employed today consists of an Anger camera⁹; a device consisting of an array of photomultiplier tubes (PMTs) attached to a large (approximately 1.0 meter in diameter) crystal of inorganic scintillating material such as NaI(Tl) or BGO. When a gamma-ray photon interacts with the scintillating crystal in a gamma-camera, it produces an energetic electron via a photo-electric or Compton scattering process. The energetic electron produced by gamma ray interaction subsequently deposits its energy within a millimeter or so of the gamma-ray interaction site. When the electron loses its energy, it produces scintillating photons which are detectable by the PMTs. By determining the position centroid (Anger logic) of the light detected from several PMTs, the position of interaction of the photon can be determined. The scintillators used in conventional gamma cameras require some tens of eV of energy deposited to produce a single scintillating photon. Thus only a few tens of optical photons are produced per keV of energy deposited by the gamma ray. This low number of photons -- or more accurately the statistical fluctuation in this number of photons -- ultimately determines both the energy and position resolution of the detector used in existing gamma camera systems. The resolution with which a gamma-camera can encode position on the detector is not the same as the spatial resolution with which the gamma camera system can estimate the position of the gamma ray emitter. The position resolution of the collimator is also an important factor; in general, the position resolution in the resultant image is the convolution of the spatial response function of the collimator and the spatial response function of the detector. In modern large gamma cameras, the position resolution of the system is usually determined by the collimator. However, in a portable system employing a high resolution collimator (such as a pinhole collimator), the system resolution would be determined by the position resolution of the detector plane. Under these circumstances (i.e., portable gamma-cameras), the position resolution of the gamma ray imager may be limited by the scintillator/PMT detector. Furthermore, if one seeks to determine the isotope that is producing gamma ray emission, the ability to resolve isotopes is limited by the energy resolution of a scintillator based detector system.

An obvious method for overcoming the limitations of scintillator based systems is to use a semiconductor detector based gamma-camera. In a semiconductor detector, only a few eV are required to produce an electron-hole pair, yielding a large reduction in the statistical fluctuations compared to the scintillator-based systems. Indeed, semiconductor gamma-ray spectrometers using cryogenic germanium long ago displaced scintillation systems as the best detection technique for energy spectroscopy. However, because of the cooling involved, it is often not practical to use a germanium detector, particularly for field use. Instead, one solution to producing improved miniature gamma-cameras is to use a room temperature semiconductor material (such as HgI₂) to build the detectors.

There are several approaches that might be tried to produce a semiconductor detector with imaging capabilities. The most obvious solution is to configure a square array of individual detector elements into the desired image plane. However, since a typical imager could require a square array of dimensions 32 x 32 elements (1024 total elements), it is not economical to fabricate and assemble into an array such a large number of individual elements.

The next most obvious approach is to fabricate a monolithic array of individual detector elements on a single semiconductor substrate. If lithographic approaches to fabrication are used, it becomes practical to fabricate monolithic arrays with a suitable number of elements; however, reading out the signal from such an array would require very complex electronics. In general, to readout an $N \times N$ array of individual pixels requires N^2 separate channels of readout electronics; for arrays larger than about 4×4 this becomes a very complex solution, particularly since each channel of electronics must be very high performance (low noise) to take advantage of the benefits of semiconductor detectors.

An alternative approach to reading out a semiconductor detector array was first proposed by Gerber et al.¹⁰ and is known as an orthogonal strip design. Such an approach is illustrated in Figure 13 and consists of rows of parallel electrical contacts (strips) placed at right angles to each other on opposite sides of the detector. By making use of the temporal coincidence between events recorded on both sides of the detector, it is possible to readout an effective array of N^2 effective detector elements using only $2N$ channels of readout electronics. The orthogonal strip approach was used for all of the imagers described in this report.

Figure 13. Diagrammatic view of an "8x8" orthogonal strip detector and front-end readout electronics. The metal contact strips are deposited on opposite sides of a square piece of semiconductor wafer. Event localization on the detector plane is determined by scoring a coincidence event between a column and a row. Using this method reduces the complexity of the readout electronics considerably. In general, to readout an array of N^2 effective pixels only requires $2 \times N$ channels of readout electronics, as opposed to N^2 channels of readout required for a detector consisting of an array of individual pixels.

Detectors were designed and built using HgI_2 crystals as substrates. Our first detectors were 8×8 devices with an approximate thickness of 2.0 mm (see

Figure 14). These 8x8 orthogonal strip detectors resulted in devices with 64 effective pixels with each pixel a square of approximately $0.125 \times 0.125 \text{ cm}^2$ dimensions. These devices were placed on chip carriers (see Figure 15) that allowed them to be plugged directly into the readout electronics.

Figure 14. Photograph of an 8x8 HgI_2 orthogonal strip detector, shown at five times actual size.

Figure 15. Photograph of an orthogonal strip detector mounted on a ceramic 24 pin DIP package.

Detectors were fabricated by evaporating metal contacts on to the surface of etched crystal substrates using a shadow mask to define the strip pattern. The detectors were mounted on standard 24 pin alumina dual in-line packages (DIPs) commonly used for hybrid electronic circuits. Fine gold wires (25 μm diameter) were bonded to the metal contact strips using silver epoxy, the other end of the wire was then bonded to a metal foil pattern on the alumina substrate of the dual inline package. The packaged detectors could then be inserted directly into a socket on the front-end readout circuit board. Subsequent testing of the detectors indicated that all of the detectors operated correctly as spectroscopic imaging devices, however, there was one strip that did not function. As of this writing it is not clear if the cause of the strip

failure was in the detector or in the bonding and interconnection used to connect the detector to the readout electronics.

The system used to readout the 8x8 detector arrays made use of hybrid preamplifiers followed by NIM and CAMAC readout amplifiers and ADCs. A block diagram of the electronics used to readout the 8x8 detector array is shown in Figure 16. The performance of the readout system is determined largely by the hybrid preamplifiers which first amplify the signals from the 8x8 detector array. We chose to use a commercially available preamplifier array in our first prototype gamma camera. The preamplifiers (Lecroy HQV 820) are constructed in thick film hybrid technology and contain 8 channels of preamplifier in a single wide 24 pin dual inline package (DIP). The rated performance of the preamplifiers translates to about 5 keV FWHM of noise referenced to a gamma spectrum. We chose the HQV 820 preamplifiers because they were the only multi-channel units available commercially.

A circuit board was designed and built that accommodated two of the eight channel hybrid preamplifiers as well as the miscellaneous passive components to couple the detector to the preamplifier unit. The "front-end" circuit board containing the preamplifiers also contained a socket for holding an 8x8 detector. Although the additional capacitance and conductor trace length needed to socket the detectors would degrade the noise performance of the system somewhat, we decided that the tradeoff of being able to test several detectors designs with the same readout circuit outweighed the slight performance degradation that socketing the detector induced. The front end circuit board was constructed of a Teflon derivative to minimize "1/f" noise¹¹ that would be induced by a conventional electronic circuit board. A photograph of the assembled circuit board is shown in Figure 17.

The front-end electronics were housed in a separate chassis which comprised the "camera head" and was connected via coaxial cables to the remaining readout electronics. The remaining electronics were housed in NIM bins and a CAMAC crate and interfaced to the readout computer with a SCSI CAMAC controller. A form of sparse readout -- implemented in hardware -- was used to limit the amount of data the computer must obtain from the comparators and ADCs in the CAMAC crate. Signals from all 16 channels (8 columns and 8 rows, 64 pixels) were fed to a 16 channel comparator; when at least two comparators had fired, a master gate signal was triggered initiating the readout sequence. Additional event selection was also performed in software.

In order to readout the coincident signal from the strip detector, decode the pixel position, and visualize the gamma-ray intensity distribution at the detector plane, it was necessary to write software to control the gamma camera. The software interrogated the ADCs and discriminators in the CAMAC crate, decoded the position of interaction on the orthogonal strip detector, and created pulse height spectra of the interactions that occurred at each pixel. Another function of the software was to provide real-time feedback on the operation of the camera, and diagnostics of various camera functions (such as cross-talk between channels). The software was written in the high level control language "Kmax" to minimize development and reduce the amount of time spent writing low level CAMAC control routines. Figure 18 illustrates the graphical user interface (GUI) presented to the user when operating the software. The software, which executes on a Power Macintosh computer, also contains many more windows and dialog boxes than are shown in Figure 18, and can be called up for various diagnostic functions.

To demonstrate the capabilities of imaging with room temperature semiconductor detectors, the gamma camera system described above was tested with isotopic sources. Three general types of experiments were performed with the HgI_2 detector array mounted in the gamma camera: flood field images of the detector plane to determine the uniformity of its response, imaging of

objects with the aid of a pinhole collimator, and gamma-ray pulse height spectroscopy of isotopic sources.

Figure 16. Block diagram of the electronic readout system used in the 8x8 gamma camera. The system may be viewed as consisting of two branches: one for counting hits (discriminator) and the other branch for spectroscopy (ADC). Pulses from the Lecroy HQV 820 preamps in the camera head are shaped and amplified by a CAEN 16 channel spectroscopy amplifier, these pulses are then fed to both a 16 channel discriminator and to a 16 channel ADC. Triggering of a read cycle is initiated in hardware whenever at least two channels are above the discriminator threshold, subsequent event selection is then performed in software after the CAMAC discriminators and ADCs have been read.

Figure 17. Photograph of the "front-end" circuit board used in the 8x8 gamma camera. An 8x8 detector module mounted in a DIP socket is visible in the center of the photograph. The white cylindrical objects on either side of the detector module are the decoupling capacitors (an AC coupled configuration was used to connect the detector strips to the preamp). The black rectangular objects on either end of the circuit board are the eight channel preamplifier arrays. The circuit board was constructed from Duroid -- a Teflon derivative -- to minimize the "1/f" noise produced by the circuit traces between the detector and the preamplifier inputs.

Figure 18. Printout of Graphical User Interface (GUI) of software for controlling the operation of the "8x8" gamma camera. A large number of windows and dialog boxes are available to the operator for controlling and monitoring the operation of the camera. The windows titled "EVENT_HIST" is a display of the total number of counts at each pixel. The window titled "ENERGY_WIN" is the display of intensity of hits that fall within a range of pulse height values selected on a master histogram of pulse height intensities.

Flood field images were obtained by mounting a ^{133}Ba source a few cm in front of the detector plane and recording the count rate at each pixel location as well as the energy spectrum of the source. Imaging studies were performed by mounting a pinhole collimator on the front of the gamma camera assembly and irradiating the collimator with isotopic sources of small active diameter. It was then possible to measure the position resolution on the detector by recording images of the source as it was moved in a plane parallel to the detector plane.

Pulse height spectra were also acquired in both flood field mode and with the pinhole collimator. Two components were identified in the broadening of the pulse height spectra: Gaussian broadening due to random electronic noise¹¹ and an asymmetrical distortion of the gamma-ray peaks due to charge trapping effects^{12,13}.

Conclusions

In this project, new instruments were developed for use in hazardous and radioactive materials management. These tools provide a previously unavailable capability for the detailed characterization of hazardous, radioactive, and mixed materials in the field, *in situ*. With these tools, it will be possible to more effectively and efficiently perform environmental remediation efforts in a variety of scenarios.

The fundamental enabling technology for these new instruments is the novel mercuric iodide high-resolution nuclear radiation spectrometer that is capable of operation at ambient temperatures and with minimal support and maintenance requirements. While mercuric iodide has been considered for many years as the semiconductor of choice for such applications, it is only through the improvement of the materials properties achieved in this project that the needed performance and yield of high quality detectors have been reached. In particular, the reduction of charge-trapping defects to a level sufficient to allow for the effective collection of all of the charge created by a photon interaction event has been achieved by coupling materials measurements with device performance evaluations in order to determine the particular materials properties most degrading performance, and eliminating their source. With these improvements, mercuric iodide devices with high energy resolution over the range of x-ray and gamma-ray photon energies of interest (1keV to 1000keV) can be reliably produced.

The first instrument developed was a field portable XRF instrument, for detailed, quantitative elemental analyses of any form of sample. The instrument developed can quantify elements ranging from Na to U to the part-per-thousand by using each of its three radioisotope excitation sources. The unit consists of a shoulder-strap-borne computer processing unit and a hand-held probe head unit that can be readily carried by a field worker for an eight-hour shift with minimal notice -- the unit can be used in an anytime, anywhere mode. The unit was demonstrated to have sufficient energy resolution to easily separate and identify the individual elements in complicated samples.

Finally, an instrument which imaged and performed spectroscopy on gamma-rays was designed, built, and tested. The imaging system used a room-temperature semiconductor detector made from HgI_2 . All detectors fabricated for this study were approximately 1.0cm^2 in active area and were of an 8×8 orthogonal strip design (64 effective pixels). Simple tests of the position resolution of the detector elements indicated that the HgI_2 strip orthogonal strip detectors were capable of resolving the movement of a point source on the imaging plane with a resolution of better than 1.0mm . Further testing will be required to better quantify the spatial resolving capabilities of this instrument. The ability of the imaging detectors to resolve the energy of the detected gamma rays was also measured. It was found that energy resolution -- as measured by pulse height spectroscopy -- was limited at low energies by electronic noise at approximately 15keV FWHM. At higher gamma-ray energies, the energy resolution was limited by hole trapping and extensive "hole-tailing" was observed on the low energy sides of the peaks. This behavior was expected, and is consistent with the performance of single element detectors made from these materials. Despite the deficiencies observed in the performance of our first system, we found our results (and those obtained by others using similar methods) to be quite encouraging. Orthogonal strip detectors made from room-temperature semiconductor detector materials appear to be a viable method for imaging the distribution of radionuclides with high spatial resolution and good energy resolution, particularly if the hole trapping effects in these detector materials can be

minimized. We are currently working on improving the performance of our gamma-ray imaging devices by making use of electron-only device designs (analogous to the gridded ion chambers used with gas detectors^{1,14}) to minimize hole tailing, and using a custom integrated circuit readout chip to reduce the electronic noise and increase the number of readout channels.

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RADIATION DAMAGE MEASUREMENTS IN ROOM TEMPERATURE SEMICONDUCTOR RADIATION DETECTORS

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Abstract

The literature of radiation damage measurements on cadmium zinc telluride (CZT), cadmium telluride (CT), and mercuric iodide (HgI_2) is reviewed and in the case of CZT supplemented by new alpha particle data. CZT strip detectors exposed to intermediate energy (1.3 MeV) proton fluences exhibit increased interstrip leakage after 10^{10} p/cm² and significant bulk leakage after 10^{12} p/cm². CZT exposed to 200 MeV protons shows a two-fold loss in energy resolution after a fluence of 5×10^9 p/cm² in thick (3 mm) planar devices but little effect in 2 mm devices. No energy resolution effects were noted from a moderated fission spectrum of neutrons after fluences up to 10^{10} n/cm², although activation was evident. Exposures of CZT to 5 MeV alpha particles at fluences up to 1.5×10^{10} α /cm² produced a near linear decrease in peak position with fluence and increases in FWHM beginning at about 7.5×10^9 α /cm². CT detectors show resolution losses after fluences of 3×10^9 p/cm² at 33 MeV for chlorine-doped detectors. Indium doped material may be more resistant. Neutron exposures (8 MeV) caused resolution losses after fluences of 2×10^{10} n/cm². Mercuric iodide has been studied with intermediate energy protons (10 to 33 MeV) at fluences up to 10^{12} p/cm² and with 1.5 GeV protons at fluences up to 1.2×10^8 p/cm². Neutron exposures at 8 MeV have been reported at fluences up to 10^{15} n/cm². No radiation damage was reported under these irradiation conditions.

Introduction

The well documented radiation damage in cryogenic germanium and silicon detectors (refs 1,2) at relatively modest charge particle fluences has prompted interest in the response of the room temperature semiconductor materials now under development. Radiation damage in semiconductor detectors causes degradation of energy resolution, increased leakage current, and reduction of the peak position. Depending on the elemental constituents, activation may also occur in the case of neutron exposures. Exposure parameters affecting the response of materials to radiation fields include the fluence level, flux, incident energy, detector bias, and detector temperature. In addition to the base constituents, trace contaminants as well as detector dimensions can also affect radiation response.

In the case of cryogenic silicon, significant energy resolution losses occur at about 10^{12} n/cm² (ref 1) with leakage current and pulse height changes after 5×10^{11} protons/cm². Planar germanium detectors exhibit significant resolution losses after 10^9 n/cm². Coaxial n-type detectors are appreciably more resistant to neutrons than p-type (ref 2). Coaxial p-type germanium detectors begin to show resolution losses at about 2×10^7 p/cm², while in n-type coaxial (reverse electrodes) detectors, resolution losses appear at about 2×10^8 p/cm². Despite the susceptibility of germanium to radiation damage, thermal annealing methods have been developed to mitigate, if not reverse, the symptoms of high fluence exposures.

In this paper we report the results of a survey of radiation damage studies in the room temperature semiconductor materials: cadmium zinc telluride (CZT), cadmium telluride (CT), and mercuric iodide (HgI₂). Also reported are new 5 MeV alpha particle data on CZT including α response maps showing material inhomogeneities. The data are useful in the selection of room-temperature semiconductor radiation detectors for use in high radiation environments, such as long-term space missions and monitoring of high-level radioactive materials.

Methods

Survey data were obtained through a literature search using the computer databases INSPEC and CALPLUS together with DOE archives. In all, more than 300 abstracts were reviewed. The review was confined to the materials cadmium zinc telluride, (CZT), cadmium telluride (CT), and mercuric iodide (HgI₂), the most promising of the wide-band gap materials now under development.

Alpha particle exposures were made with He²⁺ ions from the Sandia Tandem Van de Graaff. The flux was maintained at 2.8×10^7 α /cm²-sec and measured by means of a scaler connected to the output of the detector under test. The beam spot size was 1 micron in diameter. Exposures were made in vacuum and carried out by scanning the beam over the surface of the detector. This procedure provided both radiation exposure effects data as well as homogeneity maps of the

detector surface. Gross positioning was accomplished with a manually adjusted x-y stage; fine adjustments, over a 250 micron by 250 micron area, were made with the accelerator's beam handling system.

Results

The radiation exposure effect reported most frequently was the change in energy resolution, although changes in the leakage current and peak position shifts were sometimes cited. Energy resolution (ΔE) was determined in most cases by recording the spectrum of a monochromatic photon or alpha particle before and after exposure. Detector energy resolution is reported here in terms of the full width of a given spectral line at its half intensity points (FWHM). In general, neither the detector bias conditions nor the detector temperature were reported. Where available, this information is noted in this report. The results including the recently obtained α particle data are summarized in Tables 1-3.

Mercuric Iodide

The results of this investigation for mercuric iodide are summarized in Table 1. Proton exposures at 10, 10.7, 33, and 1500 MeV have been reported at fluences ranging from 1.2×10^8 protons/cm² (1500 MeV) to 10^{12} protons/cm² (10.7 MeV). Also reported were 8 MeV neutron exposures at fluences up to 10^{15} n/cm².

For 10.7 MeV proton exposures, no losses in energy resolution were found in the 5.9 keV Mn - K α line in a group of 6 detectors following fluences up to 10^{12} p/cm² (ref 3). Similar results were reported (ref 4) for a single detector 0.4 mm thick in 5.5 MeV α peaks for 10 MeV protons at fluences up to 10^{10} p/cm²; the relative pulse height decreased by approximately 20% following exposure of 10^{11} p/cm², however. No resolution losses were found in a group of 3 detectors from 33 MeV proton exposures in 59.5 and 122 keV photon peaks following fluences of up to 2.5×10^{10} p/cm² (ref 5).

A group of 4 detectors with an average active area of 6.4 mm² were exposed to a flux of 10^5 protons at 1.5 GeV (ref 6). The detectors, initially unbiased, exhibited anomalous leakage current after the first exposure of 2×10^6 protons/cm². Subsequent exposures were made under bias and the leakage remained stable. No loss in resolution was found in 5.9 keV photon peaks following exposures up to 1.2×10^8 p/cm². Little effect was found in a single 50 mm² detector (active thickness 0.26 mm) from 8 MeV neutron exposures in 5.3 MeV α peaks after fluences up to 10^{15} n/cm² (ref 4). No data on effects from high-energy photon exposures were found.

TABLE 1.
Radiation Damage in HgI₂ Room Temperature Semiconductor Detectors

Material	Radiation	Effects	Refs.
HgI ₂	Proton-10.7 MeV	No ΔE loss up to 10^{12} p/cm ²	3
	10.0 MeV	No effect up to 10^{10} p/cm ²	4
	33 MeV	No ΔE loss up to 2.5×10^{10} p/cm ²	5
	1.5 GeV	No ΔE loss to 1.2×10^8 p/cm ²	6
	Neutron- 8 MeV	Little ΔE effect up to 10^{15} n/cm ²	4
	Photon	No reported data	

Cadmium Telluride

Damage studies were found for proton, neutron, and photon irradiation. The results are summarized in Table 2. Proton exposures at 33 MeV were made on chlorine-doped and indium-doped material (ref 5). This energy was selected so that the beam passed through the thickest samples (2.8 mm) and thus contributed no Bragg peak in the test samples. In chlorine-doped samples, the energy resolution in 59.5 and 122 keV peaks degraded sharply after about 3×10^9 protons/cm². There was some evidence of a slight resolution improvement at lower fluences. Evidence of detector thickness dependence in the resolution response was also noted. While quantitative data were not obtained for the single indium-doped device tested, the data suggests a higher degree of radiation resistance than with the chlorine-doped material. Neutron irradiations were reported at a mean energy of about 3 MeV (ref 7). Significant reductions in energy resolution were reported after about 2×10^{10} n/cm² in 5.5 MeV alpha spectra using a flux of 10^8 n/cm²-s. Photon irradiations were reported using Co-60 (1.17 and 1.33 MeV) with the resolution monitored at 662 and 59.6 keV (ref 8). Marked changes in peak shape were reported at both energies after exposure of several times 10^5 R. No data were found at intermediate and low exposure levels.

TABLE 2.

Radiation Damage in CdTe Room Temperature Semiconductor Detectors

Material	Radiation/Energy	Effects	Refs.
CdTe	Proton 33 MeV	<u>Chlorine doped</u> -- ΔE degradation after $\sim 3 \times 10^9 \text{ p/cm}^2$ <u>Indium doped</u> -- evidence of greater radiation resistance, no quantitative data	5
	Neutron 3 MeV	<u>p-type</u> -- increasing ΔE loss in 5.5 MeV α peaks at $2 \times 10^{10} \text{ n/cm}^2$	7
	Photon 1.17 and 1.33 MeV	Substantial ΔE loss @ 59.5 and 662 keV after 10^5 R	8

Cadmium Zinc Telluride

Radiation damage studies on CZT have been carried out with protons (1.3 and 200 MeV) and neutrons (moderated fission spectrum). The results of these studies are shown in Table 3. In a single detector with 1.3 MeV protons, the bulk leakage was found to increase significantly after 10^{12} p/cm^2 in a single unbiased $10 \times 10 \times 2 \text{ mm}^3$ strip detector (ref 9). The interstrip leakage increased significantly after about 10^{10} p/cm^2 . No energy resolution data were reported.

Considerable work has been reported in the region of 200 MeV. In one study (ref 10) 2 and 3 mm thick planar detectors were irradiated with up to $5 \times 10^9 \text{ p/cm}^2$ and resolution losses were found in the thicker devices (3 mm). The initial (unirradiated) FWHM values at 59.5 and 122 keV of 3.2 keV and 3.9 keV, respectively, degraded to 4.1 and 4.3 after 10^9 p/cm^2 and to 6.2 and 9.2 keV after $5 \times 10^9 \text{ p/cm}^2$. The thinner device (2 mm) showed no degradation at either energy after like exposures, however. A downward shift in peak channel with increasing fluence was reported for both thick and thin detectors. The degradation in energy resolution is attributed to radiation induced electron trapping.

In a similar study, (ref 9), both strip and planar detectors were exposed to 200 MeV protons. The strip detectors ($15 \times 15 \times 2 \text{ mm}^3$) were exposed under bias to fluences from 10^8 to $5 \times 10^9 \text{ p/cm}^2$. A small gain shift (3%) was noted after a fluence of $1 \times 10^9 \text{ p/cm}^2$ and a significant shift (>25 %) after $5 \times 10^9 \text{ p/cm}^2$. No consistent pattern of resolution degradation was found. The resolution of a single detector exposed to $5 \times 10^9 \text{ p/cm}^2$ was unchanged at 59.5 keV but significantly degraded at 122 keV while small losses (and gains) were found at 1×10^8 and $1 \times 10^9 \text{ p/cm}^2$. In this study, the outputs of three strips in each detector were summed; one detector was

used for each fluence level (1, 10, and 50×10^8 p/cm²). Two planar devices ($10 \times 10 \times 2$ mm³ and $15 \times 15 \times 2$ mm³) were exposed to a fluence of 5×10^9 p/cm², one under bias the other unbiased. Gain shift and energy resolution were measured at photon energies of 14.4, 17.8, 59.5, and 122 keV. Gain shifts were found at all energies in both biased and unbiased cases. Significant (>45%) resolution losses were found at 59.5 and 122 keV in the unbiased device. Minor changes (both positive and negative) were reported for the biased case.

Neutron irradiation with a moderated fission spectrum source (Cf-252) at fluences up to about 10^{11} n/cm² have been reported (ref 11) on a single detector. The $10 \times 10 \times 2$ mm³ detector was biased during exposure. No resolution degradation was found at photon energies of 14.4, 26.3, 59.5, and 122 keV for fluences up to 10^{10} n/cm². Significant resolution losses were found after 7×10^{10} n/cm², however. It is interesting to note that the resolution losses were largely recovered after 12 weeks of annealing at room temperature. Evidence of neutron activation, in the form of gamma-ray lines from cadmium and tellurium isotopes, was apparent at fluences beyond about 10^{10} n/cm². Although not reported, similar effects can be expected in CdTe.

TABLE 3.

Radiation Damage in Cadmium Zinc Telluride (CZT) Room Temperature Detectors

Material	Radiation/Energy	Effects	Refs.
CZT	Proton 199 MeV	-- onset of ΔE loss at 10^9 p/cm ² ; 2 fold increase in ΔE @ 5×10^9 p/cm ² (3mm thick detector), -- little ΔE change in 2mm thick device -- Downward peak shift proportional to fluence in 2 mm and 3 mm devices	10
	Protons 200 MeV	<u>Strip detector (biased)</u> -- >25% gain shift following 5×10^9 p/cm ² <u>Planar (biased)</u> -- Small ΔE effects, both positive and negative found @ 14.4, 59.5, and 122 keV <u>Planar (unbiased)</u> -- Large (>45%) ΔE losses following 5×10^9 p/cm ² exposure -- Gain shifts in both biased and unbiased detectors at 14.4, 17.8, 59.5, and 122 keV	9
	Proton 1.3 MeV	<u>2 mm thick strip detector (unbiased)</u> -- Bulk leakage increases after 10^{12} p/cm ² -- Interstrip leakage increases after 10^{10} p/cm ²	9

TABLE 3. (continued)

Radiation Damage in Cadmium Zinc Telluride (CZT) Room Temperature Detectors

Material	Radiation/Energy	Effects	Refs.
CZT	Neutron Moderated Fission Spectrum	<u>Planar</u> -- No ΔE effects up to 10^{10} n/cm ² -- Measurable ΔE loss @ 7×10^{10} n/cm ² -- Activation emission (γ) at 10^{10} n/cm ²	11
	Alpha 5 MeV	-- linear decrease in peak position with exposure beginning at 2.5×10^9 α /cm ² -- ΔE increases by 60% at 1.5×10^{10} α /cm ²	11

The results of this review were supplemented with alpha particle irradiation. The exposures were made with 5 MeV He²⁺ ions produced in the Sandia Tandem van de Graaff and made on a single 3 x 3 x 2 mm³ CZT detector. The exposures were in vacuum over the region from 2.5×10^9 α /cm² to 1.5×10^{10} α /cm² by scanning the 1 micron diameter beam spot over the surface of the detector. The flux was maintained at approximately 2.8×10^7 α /cm²-sec by monitoring a scaler connected to the output of the detector. Alpha particle spectra obtained by integrating the pulses from a small irradiated area are shown in Figure 1. Plots of the resultant FWHM and the peak position are shown in figure 2. The data of Figure 2 show a near linear decrease in the peak position with fluence. The FWHM, after decreasing initially, increases by more than 60% after an exposure of 1.5×10^{10} α /cm². Variation in the response of the detector as measured by the α counts recorded as the detector was scanned are shown in figure 3. One notes a number of low responding elements distributed throughout the sample including a band in the lower left. The cause of this effect is currently under investigation.

Summary And Conclusions

Knowledge of the radiation susceptibility of the leading room temperature semiconductor detectors CZT, CdTe, and HgI_2 , is incomplete. Factors known to be of significance in semiconductor radiation damage such as rate effects (flux), incident energy, and device temperature have not yet been fully examined. Moreover, the available data are from a very small sampling of detectors (sometimes a single device) and do not, in general, cover the complete fluence range of interest.

Despite these shortcomings, several interesting features emerge from the existing data. The most apparent is that HgI_2 appears to be relatively immune to proton and neutron-induced radiation damage. No resolution degradation was found from intermediate energy protons at fluences up to 10^{12} p/cm². Similarly, no degradation was found from high-energy protons although the effects of fluences significantly beyond 10^8 p/cm² have not been investigated and accordingly the suitability of HgI_2 for long-term space missions remains in question. Additionally, the material is apparently not susceptible to damage from intermediate energy neutrons.

The situation for cadmium telluride is less clear. No data were found for effects of high-energy protons although the results at intermediate energy suggest vulnerability beginning in the region of 10^8 p/cm². Neutron data are also incomplete although at intermediate energies (8 MeV) the damage threshold for resolution degradation is relatively high (10^{10} n/cm²). While no evidence of activation was reported, effects similar to those in CZT can be expected.

The radiation susceptibility of CZT is also in question. There is evidence of resolution degradation from 200 MeV protons beginning in the region of 10^9 p/cm² as well as a downward shift in peak channel proportional to the proton fluence. However, the resolution degradation was apparent only in a 3 mm thick device and not a 2 mm detector. There is also evidence that the resolution degradation is dependent on bias conditions although this is based on the results from a single detector. Detector response changes following high-energy proton irradiation are consistent with increased electron trapping and the associated decreases in the mobility-lifetime product. With intermediate energy protons (1.3 MeV), bulk and interstrip leakage was evident, but only at high fluence levels. Damage for moderated fission neutrons is evident only after 7×10^{10} n/cm². Neutron activation lines from cadmium and tellurium isotopes appear after about 10^{10} n/cm². It is interesting to note that annealing at room temperature was very effective in restoring resolution losses. Resolution degradation and the downward trend in peak position from 5 MeV α particles, while based results from a single detector, are generally consistent with the proton data.

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Figures and Captions:

Figure 1. Pulse height spectra of 5 MeV alpha particles obtained from a small irradiated area. The alpha flux is $2.8 \times 10^7 \alpha/\text{cm}^2\text{-s}$.

Figure 2. Peak position and full width at half maximum as a function of exposure time for α irradiated CZT detector. The alpha flux is $2.8 \times 10^7 \alpha/\text{cm}^2\text{-s}$.

Figure 3. Alpha particle response map of detector surface as measured by deviations in counts in the photopeak channel. Dark areas indicate regions with lower response.

Ion Microbeam Studies of Cadmium Zinc Telluride Radiation Detectors by IBICC*

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Abstract

Ion Beam Induced Charge Collection (IBICC) and Time Resolved IBICC (TRIBICC) techniques were used for imaging electronic properties of Cadmium Zinc Telluride (CZT) room temperature radiation detectors. The detectors were bombarded with a scanned 5.4 MeV He microbeam and the detector response was analyzed at each point. The electron mobility (μ_e) and lifetime (τ_e), and charge collection efficiency maps were calculated from the data. In order to determine the radiation damage to the detectors, the signal deterioration was measured as the function of dose.

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1. Introduction

In recent years CZT has become over other semiconductors a leading candidate as a radiation detector material. CZT has many advantages, such as high intrinsic resistivity ($\sim 10^{10} \Omega \cdot \text{cm}$), high absorption cross section for X and γ rays, and it operates at room temperature. This latter feature makes CZT very attractive. Without the expensive and cumbersome cryogenic system, CZT spectrometers can be lightweight, even hand-held, and operated unattended for an extended period of time. Using small CZT detectors (a few mm^2), gamma ray imaging arrays can be built with spectroscopic capabilities. The availability of inexpensive, portable X -ray and γ -ray spectrometers would allow for a large range of new medical, industrial, environmental, and scientific applications. However, the price of presently available CZT detectors is quite high due to the low manufacturing yield. This low manufacturing yield is due to the presence of impurities and defects introduced during the crystal growth and at various stages during detector fabrication. Therefore, it is important to understand how these defects and impurities affect the performance of these detectors. In addition, identifying regions on the wafer that are suitable for detector fabrication prior to fabrication would lower the cost.

Several groups have tested CZT detectors using high energy α particles from radioactive sources. In a recent experiment Brunett et al. [1] mapped the charge collection efficiency and leakage current of CZT detectors by moving the detector under a collimated α particle beam. These data allowed them to calculate electron mobility, lifetime and resistivity maps. Using a nuclear microbeam, the same measurements can be accomplished with much higher resolution and perhaps provide more insight into defect structures.

IBICC was originally developed to study damage to microelectronic devices [2, 3, 4, 5] and TRIBICC [6] was developed to extend the capabilities of IBICC in the same field. However, a semiconductor detector is just a simple microelectronic device. Its response to radiation – which is its main function – can be studied the same way as in integrated circuits. By performing IBICC and TRIBICC measurements, the spatial distribution of the electronic transport properties (such as charge carrier mobility and lifetime) of the detector can be calculated. Recently Bogdanovic et al. [7] studied collection efficiency, incomplete charge collection and polarization effects in different semiconductor detectors using IBICC with a proton microbeam.

2. Experimental methods

We used the Sandia microbeam [8] facility to characterize CZT detectors. This facility is still based upon the same magnetic quadrupole doublet lens (of the Martin design) with electrically isolated pole tips for electrostatic scanning of the beam spot on target, but has undergone significant upgrades in other areas. The entire beam line and target chamber are now mounted on a vibration isolation table, the object to aperture slit distance has been increased from 1.4 to 4 meters and a new system of apertures and slits was installed. The new system routinely achieves submicron beam spot diameters for IBICC applications.

The CZT detectors (3 mm x 3 mm x 2 mm spectroscopy and counter grade crystals from eV Products) were irradiated with 5.4 MeV α and 4 MeV proton microbeams of $<1\ \mu\text{m}$ diameter, scanned over $\sim 10,000$ - $40,000\ \mu\text{m}^2$ areas with 512 step resolution in both x and y directions. Because of the low number of detected ions per pixel the data were compressed to 64×64 maps. Beam currents were maintained between 200 and 1000 particles per second for the IBICC measurements. The charge induced by each individual ion strike was collected at a detector electrode, integrated by a charge-sensitive preamplifier, and sent through a pulse-shaping amplifier into our PC-based multi-channel analyzer system. This system has been described elsewhere [9, 10]. These IBICC pulse height spectra were used to determine spatial distribution of the charge collection efficiency, the $\mu\tau$ (mobility-lifetime) product of electrons, and to study the damage to the detectors as the function of dose.

The electron lifetime and transit time distributions were determined from the TRIBICC measurements. The Sandia system for time-resolved IBICC is described more fully elsewhere [6]; only a brief description will be given here. Since the bandwidth of the charge sensitive preamplifier is large, the output of the preamplifier is a close approximation to the induced charge on the contacts of the detector as a function of time [1]. In our TRIBICC measurements on these detectors we digitize and store the current transient output of the preamplifier for each individual ion strike using a fast, computer-controlled Tektronix 680B digital oscilloscope (1 GHz analog equivalent). For these particular measurements, we digitized and stored 500ns of the transient with 1ns time resolution. Beam damage effects and multiple ion-strikes are virtually eliminated by the use of a high-speed ($<200\ \text{ns}$) on-demand beam deflector to remove the beam from the target during the relatively long period ($\sim 0.1\text{s}$) of time the system takes to process and store each transient.

Thus each recorded transient is the result of a single ion strike at that particular point in the scan. The TRIBICC scans were done with at most 64x64 point spatial resolution due to the slower data acquisition.

3. Results

3.1 Charge collection efficiency measurements

The detectors used in these measurements were planar detectors having gold contacts on the opposite sides of the CZT crystal. The induced charge on the contacts is given by the Hecht equation [11, 12]

$$Q(x) = N_0 e \left\{ \frac{\lambda_e}{d} \left[1 - e^{-\frac{x_e}{\lambda_e}} \right] + \frac{\lambda_h}{d} \left[1 - e^{-\frac{x_h}{\lambda_h}} \right] \right\} \quad (1)$$

where N_0 is the number of electron-hole pairs created, e is the elementary charge, d is the distance between the contacts of the detector (detector thickness), λ_e and λ_h are the electron and hole drift lengths, and x_e and x_h are the distances the electrons and holes have to travel. In this experiment 5.4 MeV alpha particles were impinging on the negative contact of the detector. Since the range of these alpha particles is very short, most of the electron-hole pairs were generated very close to the negative electrode (in the top 20 μm layer). Therefore, the distance the holes have to travel is practically zero, while the electrons have to travel the whole width of the detector. Under these conditions Eq. (1) reduces to the single carrier Hecht equation [11]:

$$Q_m = N_0 e \frac{\lambda_e}{d} \left[1 - e^{-\frac{d}{\lambda_e}} \right] \quad (2)$$

Using equation (2) we can calculate the electron drift length for each point on the sample. Furthermore, if we assume a constant electric field throughout the sample and locally homogenous material, the carrier drift lengths can be expressed by the transport parameters of the material, and the applied electric field:

$$\lambda_i = \frac{(\mu \cdot \tau)_i \cdot V}{d} \quad (3)$$

where μ is the carrier mobility, τ is the carrier lifetime (or average drift time), V is the applied bias voltage, and i refers to either electrons or holes. Substituting (3) into (2) the bias dependence of the collected charge is:

$$Q(V) = N_0 e \frac{(\mu \cdot \tau)_e \cdot V}{d^2} \left[1 - e^{-\frac{d^2}{(\mu \tau)_e \cdot V}} \right] \quad (4)$$

Figure 1a shows the collected charge dependence on the bias voltage for a spectroscopy grade detector. Eq. (3) was fitted to the data points using $(\mu\tau)_e$ and a multiplication constant (that converts $N_0 e$ to ADC channels) as fitting parameters. For every bias voltage the electron drift length was calculated, then using a linear fit we obtained the value of $(\mu\tau)_e$. The two values, $(\mu\tau)_e = (6.50 \pm 0.16) \times 10^{-4} \text{ cm}^2/\text{V}$ from the nonlinear fit and $(\mu\tau)_e = (6.72 \pm 0.08) \times 10^{-4} \text{ cm}^2/\text{V}$ from the linear fit agree within error and are in the same range as other measurements, although they are somewhat lower than average. Figure 1b shows the $(\mu\tau)_e$ map of a $90 \times 90 \mu\text{m}^2$ scanned area. Both fits (peak channel vs. bias voltage and drift length vs. bias voltage) were applied to the maps. The results were the same within errors. Figure 1b shows the peak channel vs. bias fit. $(\mu\tau)_e$ seems more or less uniform apart from somewhat higher values in the lower left corner and a "hole" in the bottom of the scanned area. The surface of the detector seemed uniform under the microscope, therefore, we have to assume that some imperfection in the crystal caused the "hole" in the $(\mu\tau)_e$ distribution. On counter grade (poorer) detectors we usually observed more non-uniformity.

3.2 Transition time and lifetime determination from the TRIBICC measurements

The peak channel vs. bias curve allows us to calculate the $(\mu\tau)_e$ product, but it is not possible to calculate the carrier mobility and lifetime independently from these data. Analyzing the time dependence of the output of a charge sensitive preamplifier signal will allow us to calculate the carrier mobility and lifetime independently. Ignoring the effects of charge detrapping and the contribution of the holes, the voltage on the output of the preamplifier can be described by the following equation [13]:

$$V(t) = \begin{cases} V_0 & t < t_0 \\ V_0 + \frac{V_m \cdot \tau}{T_r} \left[1 - e^{-\frac{t-t_0}{\tau}} \right] & t_0 \leq t < T_r + t_0 \\ V_0 + \frac{V_m \cdot \tau}{T_r} \left[1 - e^{-\frac{T_r}{\tau}} \right] & T_r + t_0 \leq t \end{cases} \quad (5)$$

where V_0 is the offset voltage of the preamplifier, V_m is proportional to the measured charge, t_0 is the time of interaction, τ is the electron lifetime, and T_r is the transition time of electrons. Since the beam is impinging on the negative contact, the collection of holes occurs very quickly, and only contributes very little to the beginning of the signal. By fitting the preamplifier signal, the lifetime and transit time can be calculated. Figure 2a shows a typical TRIBICC signal from a CZT detector with its fit. Figure 2b and 2c show the 2D lifetime and transit time maps for 4 MeV proton beam. Mobility can be calculated from the transit time knowing the detector thickness and bias voltage assuming a constant electric field and homogenous materials properties. The average lifetime from the map is $\tau=55\pm 4$ ns and the average transit time is $T_r=118\pm 3$ ns. The applied bias was 200 V and the detector thickness was 2 mm. The calculated average mobility is $\mu=1695\pm 44$ cm²/Vs, which gives $\mu\tau=(0.93\pm 0.07) \times 10^{-4}$ cm²/V. This value is considerably smaller than the one calculated from the bias dependence, but it is still in the acceptable range. The reason for this discrepancy is not clear yet and is subject to further investigation.

3.4 Damage studies

There have been several radiation damage studies carried out for CZT detectors. A recent overview of these studies can be found in [14]. In the reviewed experiments the detectors were irradiated with 1-200 MeV protons, fission neutrons, and 5 MeV α particles. Significant performance degradation was found above 10^{10} particles/cm² dose for protons and α particles. In our experiment we irradiated a counter grade and a spectroscopy grade detector with a 5.4 MeV α particle beam scanned over a 90×90 μm^2 area. Figure 3a shows the averaged peak position and the standard deviation dependence on the accumulated dose for the spectroscopy grade detector. After a slower decrease of the peak height the decrease becomes almost linear. At 4×10^{10} α/cm^2 the loss in peak height is about 28%. The width of the peak (standard deviation) surprisingly decreases at first, then quickly increases with the accumulated dose reaching 500% of the initial peak width. Figure 3b shows the median peak height map after 4×10^{10} α/cm^2 dose. From the figure it is obvious that the scanned area was not uniformly damaged. The damage is uniform in a larger central square, but there is much less damage on the edges, specially on the lower and upper edges. That explains the large increase in the peak width. The qualitative explanation of the increased damage is as follows: the

beam spot size was about $0.5 \times 0.5 \mu\text{m}^2$ and the scanned area was $90 \times 90 \mu\text{m}^2$ using 512×512 scans. Obviously the scans overlapped except for small bands on the sides. Therefore, the damage is less at the edges. The scan overlaps does not explain completely the effect, since the scan overlap would produce only a $0.5 \mu\text{m}$ wide band (with $0.5 \mu\text{m}$ beam spot) while the non uniformly damaged region extends as far as $12 \mu\text{m}$ as shown in Figure 3c. This phenomenon is currently under investigation and a detailed analysis will be published elsewhere [15].

The same experiment was carried out on a counter grade detector. The results are shown in Figure 3d. While the spectroscopy grade detector suffered only 13% peak height loss after $3 \times 10^{10} \alpha/\text{cm}^2$ dose, the peak height loss of the counter grade detector is more, about 30%. Presumably, this is a function of the number of defects in the detector material.

4. Summary

We have demonstrated that the nuclear microbeam is a useful tool to study the transport properties of semiconductor radiation detectors. Using IBICC and TRIBICC measurements the materials properties of these detectors can be measured and high resolution maps of these properties can be obtained. Performance degradation of the detectors was found for larger than $10^{10} \alpha/\text{cm}^2$ doses. It was shown that the increase of the peak width is partially due to the non-uniform damage of the scanned area. Counter grade detectors were found to be more easily damageable than spectroscopy grade detectors. In the future we will try to correlate insufficient charge collection areas to crystal imperfection determined by other methods.

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Figure captions

Fig. 1

(a) Peak height and drift length vs. bias voltage. Solid triangles ($\blacktriangle$) represent the pulse peak height (median), and solid squares ($\blacksquare$) represent the corresponding drift length calculated from the position independent Hecht equation. The solid and the dashed lines are the fits to the pulse height and drift length, respectively. The calculated values of $(\mu\tau)_e$ are $(6.50 \pm 0.16) \times 10^{-4} \text{ cm}^2/\text{V}$ for the pulse height fit and $(6.72 \pm 0.08) \times 10^{-4} \text{ cm}^2/\text{V}$ for the drift length fit. (b) $(\mu\tau)_e$ distribution in cm^2/V across the scanned area ($90 \times 90 \mu\text{m}^2$) calculated from the peak height fit for each point (64×64)

Fig. 2

(a) An example TRIBICC signal (o) with its fit (solid line). (b) 2D lifetime map across the scanned area. (c) 2D transit time map of the scanned area. The mobility can be calculated from the transit time using $\mu = d^2/VT_r$.

Fig. 3

(a) Peak position and peak width vs. accumulated dose. Solid triangles ($\blacktriangle$) represent the pulse height (median) and solid squares ($\blacksquare$) represent the peak width (standard deviation). The lines are drawn to guide the eye. (b) 2D map of the peak height (median) after $4 \times 10^{10} \text{ } \alpha/\text{cm}^2$ dose. The peak shift is significantly greater in the central area than at the edges. (c) Projection of the 2D map of peak height to Y axis at several doses. The damage (peak height shift) increases starting from the edges to about $12 \mu\text{m}$ and then remains constant in the central area. (d) Relative peak height position vs. accumulated dose for a spectroscopy ($\blacktriangle$) and a counter grade detector ($\blacksquare$). The rate of peak shift is significantly larger for the counter grade detector than for the spectroscopy grade detector.

Figures 1a and 1b

(a)

(b)

Figures 2a-2c

(a)

(b)

(c)

Figures 3a-b

(a)

(b)

Figures 3c-d

(c)

(d)

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