Mobility-Lifetime Product Characterization of Long CdZnTe Room-Temperature Semiconductor Gamma-ray Detectors

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Abstract:

Cadmium zinc telluride ($Cd_{1-x}Zn_xTe$ or CZT), a ternary semiconductor material is well suited and has been the material of choice for good charge collection efficiency and high energy resolution room temperature x- and gamma-ray radiation detectors. In addition, these detectors can be small in size, have fast timing characteristics, and the effective thickness can be varied. CZT detector fabrication, and the characterization and analysis of the mobility-lifetime ($\mu\tau$) product in long (thick) CTZ detectors was performed in this study using large area CZT detectors. Four 2 cm long CZT detectors were fabricated and pulse height energy spectra collected using a ^{241}Am source of high energy photons. All the long CZT detectors achieved fair energy resolutions from 8 to 11%, at different applied biases, requiring further improvement. Charge Collection Efficiency (CCE), the measured collected charges in the long CZT detectors saturate at higher applied bias. The deposited charges agree well with the expected amount of charge at the higher applied bias, leading to a conclusion that these long CZT detectors do not show measurable electron trapping, and charge collection is feasible in long detectors, but improved energy resolution is required for high energy x- and gamma-ray detection.

Keywords: CdZnTe, radiation detectors, mobility-lifetime product

1. Introduction

The proliferation of weapons of mass destruction and radioactive materials present a threat to national security. The ability to detect high energy radiation such as x-rays and gamma-rays is important to the Department of Homeland Security and the Department of Defense. Radiation detection is also important in medical imaging, nuclear particle physics, environmental safety and scientific research. Ionizing radiation is invisible and exposure can result in damage to living tissue. Radiation detection instruments are required. Research and development is ongoing to enhance the instruments and techniques for the detection of ionizing radiation, and in new materials that can help detectors achieve greater efficiencies and better energy resolutions. The use of a semiconductor material as a detection medium is of great advantage. The best energy resolution can be achieved from semiconductor detectors. In addition, these detectors can be small in size, have fast timing characteristics, and the effective thickness can be varied. The main advantage of semiconductor detectors is the small values of the ionization energy. The ionization energy is temperature and material dependent, with its value increasing with decreasing temperature. Some radiation detectors require cryogenic cooling for high energy resolution and operate with cryostats at a liquid nitrogen temperature. Semiconductor materials presently used in solid-state high energy detectors include silicon (Si), germanium (Ge), mercury iodide (HgI_2), silver chloride (AgCl), and cadmium telluride (CdTe).

2. Materials and Methods

Key semiconductor material properties required for high efficiency and high resolution high energy radiation detectors operable at room temperature are a high atomic number, ideal bandgap for high resistivity and low leakage current, high carrier mobility-lifetime ($\mu\tau$) product to ensure complete charge collection, and high-purity, homogenous, and defect-free. Cadmium zinc telluride ($Cd_{1-x}Zn_xTe$ or CZT), a ternary semiconductor material is well suited and has been the material of choice for high efficiency and high resolution room temperature X- and γ-ray radiation detectors. CZT is recognized as one of the leading materials for fabrication of room temperature X- and γ-ray radiation detectors [1, 2]. Research is ongoing to improve quality, compositional homogeneity, and the charge-transport characteristics of CZT crystals, to increase the yield and lower the cost. However, production of large-volume of CZT crystals with these properties remain a challenge requiring further research in the techniques used for growing large CZT single crystal volumes. The performance of CZT crystals varies greatly due to the growing process and other factors. Different methods have been used by others to grow CZT crystals including high pressure Bridgman (HPB) [3, 4], low pressure Bridgman (LPB) [5], and the traveling heater method (THM) [6-8]. The basic Bridgman method involves the movement of a crucible containing the melt through a horizontal or vertical furnace designed to provide a suitable temperature profile. The crucible can be stationary, with a moving heater, or the crucible can be transported through a stationary heater [9]. The THM system consists of a quartz ampoule, with a CZT seed crystal and a polycrystalline, high-purity CZT charge. The quartz ampoule is lowered or the heater is raised upward [6].

CZT crystals are usually characterized with non-homogeneities and defects including impurities, secondary phases, voids, dislocations, grain boundaries, pipes, wires, cracks, tellurium inclusions and precipitates, and twin boundaries. Cracks are formed in CZT ingots during crystal growth and cool down. Cracks are also formed during crystal slicing and dicing. Cracks are the major source of material loss and their elimination would provide the most significant yield improvement for detector fabrication [3]. Pipes are hollow tubular structures running intermittently and parallel to the growth axis of HPB CZT ingots. Pipes are formed by the trapping of gas bubbles at the gas interface and are related to changes in the growth conditions (temperature, pressure, growth rate) during the solidification process [3]. Pipe formation may be reduced by a lower temperature gradient in the melt and by improved process control [3]. Non-homogeneities and defects have adverse affects on the electrical properties, the yield and the material cost of CZT crystals used for the fabrication of room temperature radiation detectors. Material homogeneity is the most critical parameter in achieving high performance in many semiconductor detectors [1, 10-12]. Tellurium (Te) inclusions, dislocations, twin and grain boundaries affect the energy resolution and the efficiency of the device [13]. Te precipitates originate during the cooling process and have an average diameter of 10 – 30 nm, while Te inclusions originate from morphological instabilities at the growth interface as Te-rich melt droplets are captured from the boundary layer ahead of the interface [14]. Te inclusions increase leakage current and degrade the energy resolution of the device [3]. Te inclusions can be eliminated from CZT by controlling the Cd partial pressure during crystal growth or by post growth thermal annealing in Cd vapor [14]. Impurities and secondary phases accumulate around these defects trapping charge carriers and inhibiting charge carrier transport [15].

The research objectives in this project included the study of CZT detector fabrication, and the characterization and analysis of the mobility-lifetime ($\mu\tau$) product in long (thick) CTZ detectors. The typical thickness of CZT for detecting energy less than 200 keV is usually 6 mm or less, which is inadequate for gamma-ray spectroscopy applications for Homeland Security. Commercially available detectors average dimensions ranging from 5 x 5 x 1 mm^3 to 10 x 10 x 2 mm^3. Large area CZT detectors with thicknesses of 10 mm or more, and high energy resolution, operating with high efficiency at room temperature are required. The energy of these CZT detectors can be improved when techniques are applied that increase the sensitivity of the induced charge [16-18].

3. Results

3.1 CZT Room Temperature x- and γ-ray High Energy Detector Fabrication

The fabrication of semiconductor CZT high energy detectors involves a number of critical steps, including the growth of high resistivity material, slicing and polishing of the device volume, the application of metal contacts, surface passivation to limit surface leakage currents, and lastly packaging and bonding to the external circuitry. The primary goal is the production of defect free, high-resistivity material, in large quantities to ensure high yield and low cost.

CZT crystals were grown in a process that yielded high resistivity crystals. High resistivity is a requirement for CZT crystals for high energy resolutions detectors to help reduce leakage current. CZT crystals were sliced, polished and processed for simple device geometries. Contact material was deposited onto the crystal under optimal processing conditions. Surfaces were passivated to limit surface leakage currents, and detectors packaged. Devices were tested for performance including spectral resolution and counting efficiency.

Under operating conditions, a high voltage (on the order of 1000 V/cm) is typically applied across the device, and incident high energy photons interact in the volume of the detector. Voltages ranging from 200 V to 4000 V were applied across 2 cm long devices, CZT detectors, and the most common calibration source for alpha particles ^{241}Am was used as the source of high energy photons. The charges created by the absorption or interaction of the high energy photons and the CZT detector material drift across the CZT detector volume inducing a current in the external in the external circuit. This current was integrated by an external circuit with the total charge collected. A histogram of the pulse heights detected was created, corresponding to the energies of the incident photons. A summary of the results are presented, showing the ability of long (thick – 2 cm) CZT detectors can achieve good energy resolution, with further improvements required.

Four 2 cm long CZT detectors were fabricated and pulse height energy spectra collected using an ^{241}Am source of high energy photons, shown in Figure 1. All four long CZT detectors achieved fair energy resolutions from 8 to 11%, at different applied biases, requiring further improvement.

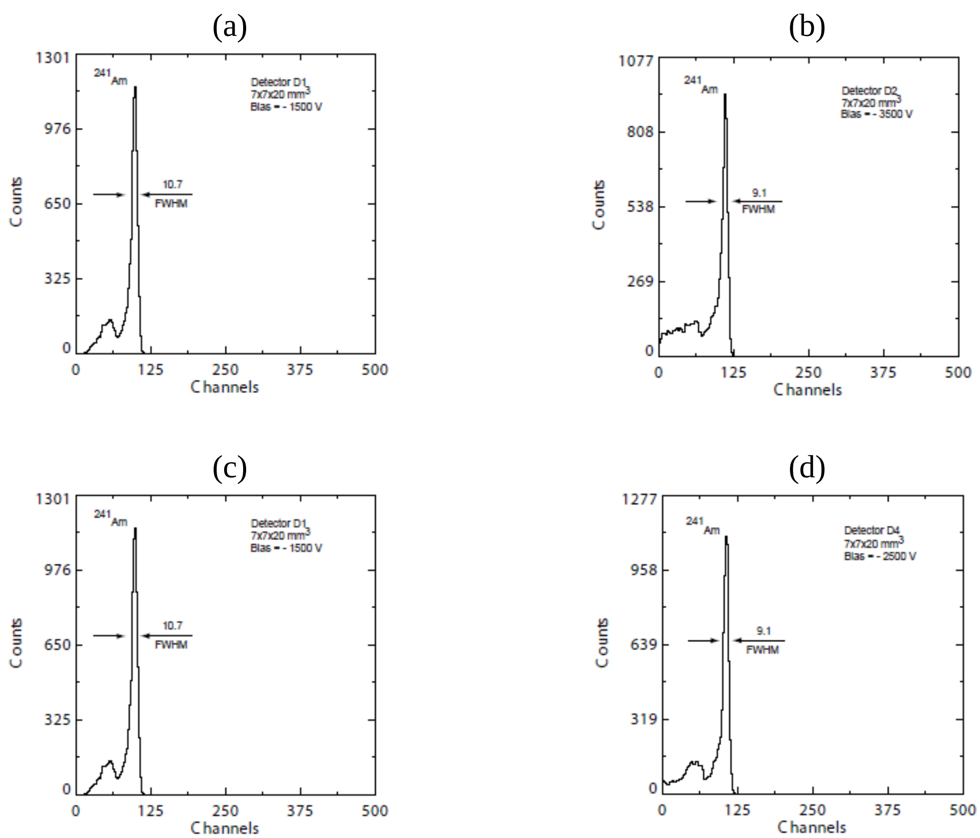

Figure 1. Pulse-height spectra obtained for four different 2 cm long CZT detectors, (a) D1: 10.5% energy resolution with -1500 V, (b) D2: 8.2% energy resolution with -3500 V, (c) D3: 9.5% energy resolution with -1500 V, and (d) D4: 8.6% energy resolution with -3500 V. All CZT detectors achieved fair energy resolutions from 8 to 11%, with different applied biases.

3.2 Mobility-Lifetime (μτ) Product Characterization and Analysis

Charge transport is characterized by two parameters for both electrons and holes: mobility, μ, and lifetime, τ. The mean drift length is directly related to CZT detector performance is the product of $\mu\tau E$, where E is the electric field. One of two methods used in this research to study the charge transport properties in the CZT detectors is based on the response to alpha particles of an ^{241}Am source placed in close proximity (~2 cm) to the CZT detector. In the first method, the single-particle Hecht relation in equation (1) was modeled using Interactive Data Language (IDL) software:

$$Q(V) = \frac{eN_o(\mu\tau)_e V}{d^2}\left[1 - \exp\left(-\frac{d^2}{(\mu\tau)_e V}\right)\right] \qquad (1)$$

where N_o is the number of charge carriers created by the source, Q is the total charge collected, d is the distance between the anode and the cathode, e is the electronic charge, and V is the applied bias. For the four CZT detectors, d was 2 cm, and the applied bias varied from 200 to 4000 V. The charge collection was measured with a long shaping time as a function of applied bias and the resulting data was fitted to the Hecht relation in equation (1). Using the Q versus V data, curve fits were generated for the four CZT detectors and the mobility-lifetime $(\mu\tau)_e$ product extracted. The same Q versus V data was also fitted to a second model based on an alternative method to extract the $\mu\tau$ product for both electrons and holes, and the results are listed in Table I. The obtained $(\mu\tau)_e$ values ~ $2\text{x}10^{-2}$ cm^2/V are consistent with typical values for standard/small, 5 mm thick CZT detectors, which makes this an exceptional level for the long 2 cm CZT detectors. The energy resolution, R%, is 8 to 11% and is shown in Table 1. The long CZT detectors require additional improvement to achieve an energy resolution below 1% for the long CZT detectors. The full width half maximum (FWHM) is ~9-10 for the 4 detectors, within the applied bias range of 600 to 1500 V, shown in Figure 2. Detector D1, noticeably experiences deterioration with applied biases of 2000 V and higher. Semiconductor detectors are typically operated with sufficient reverse bias voltage so that the depletion region extends the full thickness of the crystal, creating a fully depleted detector. The active volume of a fully depleted detector is no longer a function of the applied bias, as saturation is reached, as seen in Figure 3. The thickness of the detector that can be fully depleted using voltages short of catastrophic breakdown depends on the purity of the semiconductor crystal.

TABLE I

Mobility-lifetime (μτ), charge collection efficiency (CCE%), and energy resolutions (R%) of four CZT detectors measured.

Detector ID	**$(\mu\tau)_e$ (cm^2/V)**	**$(\mu\tau)_h$ (cm^2/V)**	**CCE (%)**	**R (%)**
D1	0.0235	0.00200	97.8	10.5
D2	0.0200	0.00181	97.5	8.2
D3	0.0169	0.00270	96.9	9.5
D4	0.0200	0.00162	97.5	8.6

The measured collected charge or energy can be smaller than the alpha-particle energy, due to charge trapping. As seen in Figure 3, Charge Collection Efficiency (CCE), the measured collected charges in all long CZT detectors saturate at higher applied bias. The deposited charges agree well with the expected amount of charge at the higher applied bias, leading to a conclusion that these long CZT detectors do not show measurable electron trapping, and charge collection is feasible in long detectors, but improved energy resolution is required.

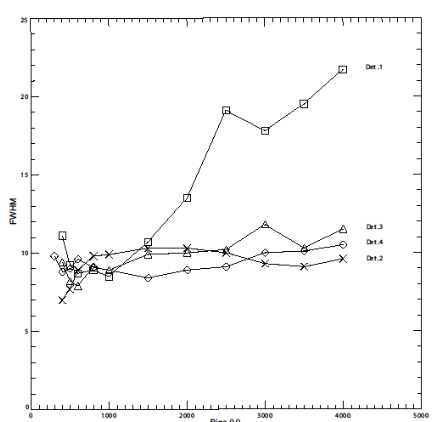

Figure 2. FHWM as a function of applied bias for all four long CZT detectors.

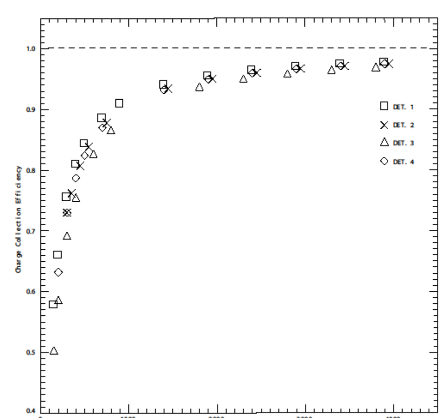

Figure 3. Collected charge efficiency as a function of applied bias for all detectors.

4. **Conclusions**

Productions of high quality, large-volume CZT for room temperature semiconductor detectors remain a challenge requiring additional research and development. Growth, electronics and processing techniques developed should result in CZT crystals that exceed the typical material properties for CZT radiation detectors, increase the yield through improved crystal quality, improve the energy resolution and increase the charge collection efficiency. The four long CZT detectors fabricated and characterized in this research project demonstrated that charge collection is feasible in long CZT detectors, but improved energy resolution is required for high energy x- and gamma-ray detection.

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