Transient Behavior in TlBr Gamma-Ray Detectors and Its Analysis Using 3-D Position Sensing

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Abstract-TIBr detectors have been investigated as room-temperature or close to ambient temperature operational semiconductor gamma-ray spectrometers. The three-dimensional position-sensitive single-polarity charge sensing technique, successfully used on CdZnTe gamma-ray imaging spectrometers, has been applied to 5-mm thick TIBr detectors and has resulted in energy resolutions as good as 0.73% FWHM and 0.97% FWHM at 662 keV on the best anode pixel and from all nine pixelated anodes respectively. The application of the three-dimensional position-sensing readout technology has significantly improved the spectroscopic performance of TIBr detectors, and has also enabled the study of transient behavior immediately following the application of cathode bias as a function of three-dimensional location within the TIBr detector material. This work presents the latest spectroscopic performance and characteristic initial transient behavior observed at -20° C on a number of 5-mm thick TIBr detectors manufactured by Radiation Monitoring Devices. Characterizing the initial transient behavior observed in TIBr detectors results in reconstruction technique improvement and may also lead to device fabrication improvements such that stable operation occurs immediately following bias for practical applications.

Index Terms—Room-temperature semiconductor, thallium-bromide.

I. INTRODUCTION

R ESEARCHERS have studied TlBr for room-temperature gamma-ray measurements due to its favorable material properties; however, some TlBr detectors must undergo a conditioning phase before stable operation is possible, similar to HgI₂ detectors [1]. During this conditioning phase, transient behavior is observed in the signal gain and the electron drift velocity which ultimately affects the spectroscopic performance. After conditioning, 5-mm thick TlBr detectors have demonstrated good spectroscopic performance of 0.97% FWHM at 662 keV. Furthermore, three-dimensional position-sensing technology has been applied to TlBr detectors, making this a

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viable alternative material to CdZnTe as a gamma-ray imaging spectrometer.

In order to avoid polarization, TIBr detectors must be cooled to suppress ionic conductivity [2]. TIBr detectors can achieve long-term stability at -20° C [3], [4], and work has been ongoing toward characterizing and improving the stability and performance at room temperature [5]–[12]. Detectors tested in this work also demonstrate stability at -20° C after the conditioning phase, with no evidence of polarization. Low charge carrier trapping is also experimentally demonstrated, indicating good detector material quality. Good energy and depth resolution demonstrated in this work further illustrates the potential for TIBr detectors to be used as Compton-imaging spectrometers.

II. METHODS

A. Experimental Setup

Detectors tested in this work were manufactured by Radiation Monitoring Devices. The crystals were grown via the traveling molten zone method. At least 100 zone refining passes were completed at a rate of 2.5 mm/h. An evaporator was used to apply chromium then gold for both electrodes. All measurements were performed in a Thermotron S-1.2-3200 environmental chamber to maintain a constant ambient temperature of -20° C. Digital pulse waveforms from nine anode pixels and a planar cathode connected to eV Products charge sensitive preamplifiers were recorded using a 14-bit GaGe Octopus CompuScope PCI bus on a personal computer. Each recorded waveform has 512 data points sampled every 100 ns. Pulse amplitudes were evaluated using a digital $CR - (RC)^1$ filter with 10 μs and 20 μs shaping times for the anode and cathode respectively. Gamma-ray measurements were performed by irradiating the entire detector volume with ¹³⁷Cs. Alpha particle measurements were performed by irradiating the cathode side with ²⁴¹Am. All measurements were performed immediately after -1000 V cathode bias was applied, and data were acquired continuously for days.

B. Gamma-Ray Data Analysis

The TIBr devices tested in this work are pixelated, such that the anode signal is uniform at all depths while the cathode signal changes linearly with depth. Consequently, the cathode-to-anode signal ratio (CAR) [13] can provide depth-dependent data which are used to study material properties. For example, the energy resolution and photopeak centroid channels as functions of depth reveal trapping behavior within a device. Furthermore, depth-dependent photopeak electron drift

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Fig. 1. Collecting pixel (with positive slopes near time zero) and the cathode (with negative slopes during $-10 \ \mu s \le t < 0 \ \mu s$) photopeak waveforms measured from ¹³⁷Cs irradiating all depths. The slow rise in the cathode waveforms, which are plotted negative to differentiate from anode pulses, indicates hole collection.

time data may be used to calculate the electron drift velocity as a function of depth. Leading edge time pick-off methods were implemented without concern for amplitude walk because only single-interaction ¹³⁷Cs photopeak events were considered. Using these data, the mobility may then be calculated from (1) which is derived from (1), (2), and (3) assuming a constant mobility, and where V_{bias} is the applied voltage, E_x is the electric field along the detector depth, and d is the detector thickness. Note that (2) is distinguished from (3) to explicitly show the assumption that mobility is constant. The electric field may then be estimated as a function of depth from (3).

$$V_{bias} = \int_{0}^{d} E_x(x) dx \tag{1}$$

$$v_x(x) = E_x(x)\mu(x) \tag{2}$$

$$E_x(x) = \frac{v_x(x)}{\mu} \tag{3}$$

$$\mu = \frac{1}{V_{bias}} \int_{0}^{a} v_x(x) dx \tag{4}$$

The relationship between the CAR reconstructed depth and the true depth was studied using a mechanical tungsten collimator. Measurements of 230 μ Ci of ¹³⁷Cs collimated through a 50 μ m slit were made starting at the cathode side and moving to the anode side in 0.635 mm increments. The cathode side was established by determining the position at which source counts were first detected.

Some TIBr detectors have high hole mobility-lifetime products $(\mu_h \tau_h)$, on the order of $10^{-4} \text{ cm}^2/\text{V}$ [14], relative to their electron mobility-lifetime products $(\mu_e \tau_e)$, demonstrated by the slow rise in the cathode waveforms in Fig. 1. As a result, the CAR is overestimated at most depths which distorts the CAR depth-based energy correction. To overcome this problem, the energy correction was calculated based on the electron drift time for detectors with high $\mu_h \tau_h$.



Fig. 2. From left to right: the raw alpha waveforms after filtering, the normalized alpha waveforms, and the aligned waveforms after applying a baseline shift. In each figure, the x-axis represents time and the y-axis represents the pulse amplitude.



Fig. 3. Detector 935-16B1R CAR depth distributions for various collimated depths. The same measurement time was used for each distribution. Because this detector has relatively low $\mu_h \tau_h$, the true depth has a one-to-one relationship with the CAR depth.

C. Alpha Waveform Analysis

In order to visualize trends present in the alpha-particle data, raw waveforms were filtered and adjusted based on techniques previously applied to CdZnTe detectors [15], as illustrated in Fig. 2. Using filters based on peak amplitudes and peaking times, the gamma-ray and pileup events were eliminated. Due to attenuation in air or source material or a possible thin inactive region on the cathode, the remaining alpha waveforms have a range of peak amplitudes. However, all alpha-particle-induced electron clouds are produced only at the cathode side and drift through the entire bulk of the crystal, so the waveforms were normalized. Finally, a baseline shift was applied to align all of the waveforms. The drift velocity was determined at each depth for each pixel by calculating an average waveform from thousands of waveforms and assuming a change in amplitude is proportional to a change in depth.

III. RESULTS

A. Depth Reconstruction

Accurate depth reconstruction is necessary in order to correct for the effects of charge carrier trapping and the weighting potential in pixelated detectors. The efficacy of CAR depth reconstruction in TlBr detectors was studied using the collimator procedure described in Section II-B. CAR depth distributions at various collimated depths are shown in Fig. 3. The centroid of the CAR reconstructed depth distribution matched each true depth, and the FWHM for each distribution was approximately 300 μ m for this detector. These data show the CAR properly



Fig. 4. Depth calculated from the CAR parameter as a function of true depth for detectors 935-16B1R and 44B2L. Having relatively high $\mu_h \tau_h$, the slope approaches unity (from 0.84 to 0.95) and the offset approaches zero (1.2 to 0.64) for detector 44B2L as the cathode shaping time decreases. Data from the cathode side were not included in the linear fits due to the similarity in cathode pulse amplitudes for the collimator position at the cathode and the collimator position 0.635 mm away from the cathode. For comparison, the slope and offset for detector a detector with low $\mu_h \tau_h$ (935-16B1R) are 0.98 and 0.03 respectively.

reconstructs depth in a pixelated TlBr detector with relatively low $\mu_h \tau_h$.

Some TIBr detectors have shown a relatively high $\mu_h \tau_h$. For these detectors, the CAR is overestimated due to the hole contribution in the cathode signal. Furthermore, high $\mu_h \tau_h$ causes nonlinearity in the CAR near the cathode side due to complete electron and hole collection occurring within the cathode shaping time. This region of nonlinear CAR will increase as $\mu_h \tau_h$ increases. Fig. 4 shows the CAR depth as a function of true depth for a detector with relatively high $\mu_h \tau_h$ using 24 μ s and 8 μ s cathode shaping times. Perfect depth reconstruction would produce a one-to-one relationship between the CAR depth and the true depth, but Fig. 4 shows the CAR is overestimated at each depth. The slope approaches unity and the offset approaches zero as the shorter cathode shaping time excludes portions of the hole movement; however, ballistic deficit causes portions of the electron component to be excluded at depths near the cathode as the cathode shaping time decreases.

To achieve optimal spectroscopic performance the CAR does not need to be a direct indicator of depth; however, each depth must be associated with a single CAR value in order to properly reconstruct the deposited energy. Contrariwise, accurate depth reconstruction is essential for Compton imaging because images are created from the three-dimensional interaction locations. Because the cathode shaping time may not be perfectly optimized for each depth, we should instead isolate the electron components of cathode waveforms with digital signal processing. For the purposes of this work, it was sufficient to perform the energy correction using the electron drift time instead of the CAR.

B. Initial Transient Behavior and Conditioning

During the conditioning phase, a TIBr detector experiences a change in its signal gain and electron drift velocity resulting in a change in spectroscopic performance which is illustrated in Figs. 5, 7, 8, and 9. Detector 935-16B1R, shown in Fig. 5, demonstrated the most dramatic spectroscopic improvement after conditioning (4.32% to 0.97% FWHM at 662 keV for the overall corrected spectrum shown in Fig. 5(a)) and the best



Fig. 5. (a) The overall corrected spectrum energy resolution for detector 935-16B1R improved from 4.32% to 0.97% FWHM at 662 keV after stabilizing. The best performing pixel (the central pixel) improved from 2.12% to 0.73% FWHM at 662 keV, illustrated in the top figure. The y-axis scales are not the same for each measurement. (b) The average raw photopeak counts and photopeak position, illustrated in the bottom figure, increase as the signal gain increases during the conditioning phase.



Fig. 6. The electric field for the center pixel after detector 935-16B1R stabilized, with the error bars representing experimental error.

overall performance of all the detectors that were tested. The average photopeak counts and position for this detector generally increase over time as shown in Fig. 5(b), indicating the signal gain increases during the conditioning phase resulting in more counts in the photopeak region. Due to geometry limitations, cathode-side alpha irradiation was not physically





Fig. 7. (a) The overall corrected spectrum energy resolution for detector 70BA1L improved from 2.61% to 1.76% FWHM at 662 keV after stabilizing. The best performing pixel (the pixel in the upper-right) improved from 2.68% to 1.22% FWHM at 662 keV, illustrated in the top figure. The y-axis scales are not the same for each measurement. (b) Weaker drift velocity is initially observed in the central depths of the detector. During the conditioning phase, the drift velocity increases in the central depths and generally becomes more uniform over all depths.

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stabilized. The detectors in Figs. 7 and 8 also show improvement in the overall corrected energy resolution (2.61% to 1.76% and 1.33% to 1.19% FWHM at 662 keV respectively). The low-energy spectral features change similarly in each of these detectors, indicating that the increase in signal gain causes more counts to appear above the readout threshold. The drift velocity increases

Fig. 6 only shows the calculated electric field after the detector

Fig. 8. (a) The overall corrected spectrum energy resolution for detector 70BA1R improved from 1.33% to 1.19% FWHM at 662 keV after stabilizing. The best performing pixel (the pixel in the lower-right) improved from 1.13% to 1.05% FWHM at 662 keV, illustrated in the top figure. The y-axis scales are not the same for each measurement. (b) Similar behavior to detector 70BA1L is observed in the drift velocity.

in the central region of the detectors in Figs. 7 and 8, generally becoming more uniform as the devices stabilize. The electron drift velocity also increases overall for the detector in Fig. 7, possibly due to a change in electric field or mobility. The detector in Fig. 9 showed insignificant changes in both spectroscopic performance and drift velocity indicating not all TIBr detectors necessitate a conditioning phase before stable operation is possible.

C. Promising Performance

The energy resolution and photopeak centroid were binned as a function of depth in the CAR depth correction process, making it possible to evaluate the bulk detector material quality. Surface



Fig. 9. (a) The spectrum characteristics remained relatively constant for detector 47AR(R). The overall corrected energy resolution degraded slightly from 1.75% to 1.89% FWHM at 662 keV. The y-axis scales are not the same for each measurement. (b) The drift velocity remained relatively uniform throughout the duration of the measurement.

preparation and electrode quality will cause systematic performance degradation at all depths, therefore depth-dependent data are not useful for characterizing these effects. Fig. 10 shows detector 935-16B1R has little or no performance degradation due to charge carrier trapping in the best-performing pixels (refer to Fig. 5(a)). Uniformity in the energy resolution at each depth indicates uniform material, i.e. no concentration of trapping centers, whereas poor electron transport would cause the energy resolution to degrade from the anode to the cathode. The photopeak centroid is affected by both weighting potential and trapping (e.g. Pixel (3,3) in Fig. 10(b)). In the absence of electron trapping, the photopeak centroid amplitude would remain uniform at most depths and decrease near the anode side as a result of the weighting potential (e.g. Pixel (1,3) in Fig. 10(a)). Assuming uniform bulk trapping the photopeak centroid amplitude



Fig. 10. (a) The energy resolution as a function of depth is relatively uniform for the best-performing pixels, indicating low electron trapping. (b) The weighting potential effect is apparent in the photopeak centroid channel as a function of depth plot, but there is little effect due to charge trapping.

should increase as a function of depth from the cathode side to the anode side (e.g. Pixel (3,3) in Fig. 10(a)). A trapping center at a given depth would cause a sharp decrease in photopeak centroid and increase in energy resolution at that depth (e.g. Pixel (3,3) in Fig. 10).

Despite current limitations of TlBr, good energy resolution has been achieved. Table I summarizes the FWHM at 662 keV for multiple TlBr detectors, each approximately 5 mm thick with nine anode pixels and a planar cathode. The spectroscopic performances of these devices are relatively consistent, maintaining an overall energy resolution near 1% at 662 keV. Sub-1% energy resolution at 662 keV was observed on a single pixel in two detectors.

IV. CONCLUSION

Transient behavior was observed in some TIBr detectors immediately following bias. However, one TIBr detector tested in this work demonstrated stable behavior for the duration of its operation. These observations show both the current need to condition some TIBr detectors before stable operation is possible and the potential to ultimately fabricate devices in way that eliminates the initial transient behavior.

Hole movement in some detectors distorts the CAR, but this obstacle may be overcome by isolating electron components

TABLE I TIBr Spectroscopic Performance

Detector	Date	Overall	Best Pixel
		Resolution	Resolution
58A3R	June 2010	1.32%	0.93%
58A4L	May 2010	1.98%	1.04%
48A2R	March 2011	4.26%	3.16%
935-16B1R	May 2011	0.97%	0.73%
935-16B1L	June 2011	1.45%	1.07%
44B2L	September 2011	2.64%	1.81%
70BA1L	December 2011	1.76%	1.22%
70BA1R	December 2011	1.19%	1.05%
47AR	April 2012	1.89%	1.09%

of cathode waveforms with digital signal processing. The energy correction may be executed using the drift time rather than the CAR; however, the CAR is less sensitive to a non-uniform electric field. Furthermore, the CAR is the most accurate depth correction process for many detectors and creates the ability to evaluate bulk detector material quality from gamma-ray data. Good depth resolution was demonstrated for a 5-mm thick TIBr detector with low $\mu_h \tau_h$. Overall energy resolution of 1.01% FWHM at 662 keV and 0.78% at 662 keV on a single pixel was demonstrated. These results indicate that TIBr is a promising material for room-temperature gamma-ray spectroscopy.

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