Evaluation of THM-Grown CdZnTe Material for Large-Volume Gamma-Ray Detector Applications

Mark Amman, Julie S. Lee, Paul N. Luke, Henry Chen, Salah A. Awadalla, Robert Redden, and Glenn Bindley

***Abstract*—Over 25 1-cm^3 CdZnTe crystals produced using the Traveling Heater Method at Redlen Technologies have been characterized. The charge carrier mobility and lifetime, and charge carrier transport uniformity of each crystal were measured using alpha particles. Some of the crystals were made into coplanar-grid detectors and their performance characterized using 662 keV gamma rays. The average electron mobility-lifetime product for these crystals was found to be a factor of about five times greater than that measured from crystals obtained over the last decade from two other commercial crystal growers. The coplanar-grid detectors produced from the material typically achieved an energy resolution at 662 keV near 2% FWHM when operated at room temperature. This is comparable to the best coplanar-grid detectors commercially produced today.**

***Index Terms*—CdZnTe detectors, Gamma-ray spectroscopy detectors, Charge carrier lifetime, Charge carrier mobility**

I. Introduction

The spectroscopic detection of gamma rays with excellent energy resolution is commonly accomplished with high-purity Ge (HPGe) based detectors. Unfortunately, the small bandgap energy of Ge requires that these detectors be cryogenically cooled (typically to about 100 K) in order to eliminate the resolution degradation caused by thermally induced leakage currents. This cooling requirement necessitates a costly, bulky detector system with either a large power requirement (mechanically cooled system) or the need for cryogenic coolant (liquid nitrogen cooled system). Applications in the areas of homeland security, medical and industrial measurement, and basic science would benefit greatly from a detector technology that operates at room temperature yet provides the efficiency and spectroscopic performance of HPGe-based detectors. Detectors based on wide-bandgap compound semiconductors with high densities and large atomic numbers are logical choices to fulfill this technology need. Compared to other candidate compound semiconductors, CdZnTe is the most well developed as indicated by its good electron collection properties, lack of room temperature polarization effects, high electrical resistivity, and long term stability [1].

Currently, the widespread use of CdZnTe is hampered by the lack of readily available crystals that are large volume, inexpensive, and of the quality required for high-resolution gamma spectroscopy. Redlen Technologies is addressing this problem through the commercial-scale production of CdZnTe using the Traveling Heater Method (THM). With this method, a large boule is grown, the boule is then cut into slices, and finally the slices are thermally treated to obtain the desired material properties. The optimum conditions of the post-growth treatment depend on the slice thickness. Previous characterization studies of Redlen CdZnTe material were primarily of samples cut from slices approximately 5 mm thick [2][3]. Many applications require detectors much thicker than this for the efficient absorption of high-energy gamma rays. For this reason, Redlen has recently begun producing 10 mm thick material [4]. The subject of this paper is the characterization of samples cut from slices 10 mm thick. We present charge transport and gamma-ray detection measurements made on 29 samples of the 10 mm thick material. These measurements are compared with those from CdZnTe samples obtained over the last decade from two other commercial crystal growers.

II. Characterization Procedure

Efficient and uniform charge carrier transport is critical to the spectroscopic performance of semiconductor-based gamma-ray detectors. The mobility-lifetime products for electrons and holes of a material are a measure of the average efficiency of the material's charge transport. We have used alpha-particle response measurements to determine the charge carrier mobility and lifetime of the CdZnTe crystals. The details of the crystal processing and the measurement technique are given elsewhere [5] and are only briefly summarized here. Each crystal is first fabricated into a simple planar detector by thermally depositing Au electrodes onto

Manuscript received June 30, 2008. This work was supported by the Director, Office of Science, of the U.S. Department of Energy under Contract Number DE-AC02-05CH11231 and by the U.S. Department of Homeland Security.

M. Amman, J. S. Lee, and P. N. Luke are with the Lawrence Berkeley National Laboratory, Berkeley, CA 94720 USA (e-mail: Mark_Amman@lbl.gov; PNLuke@lbl.gov).

H. Chen, S. A. Awadalla, R. Redden, and G. Bindley are with Redlen Technologies, Sidney, British Columbia, Canada (e-mail: henry.chen@redlen.com).

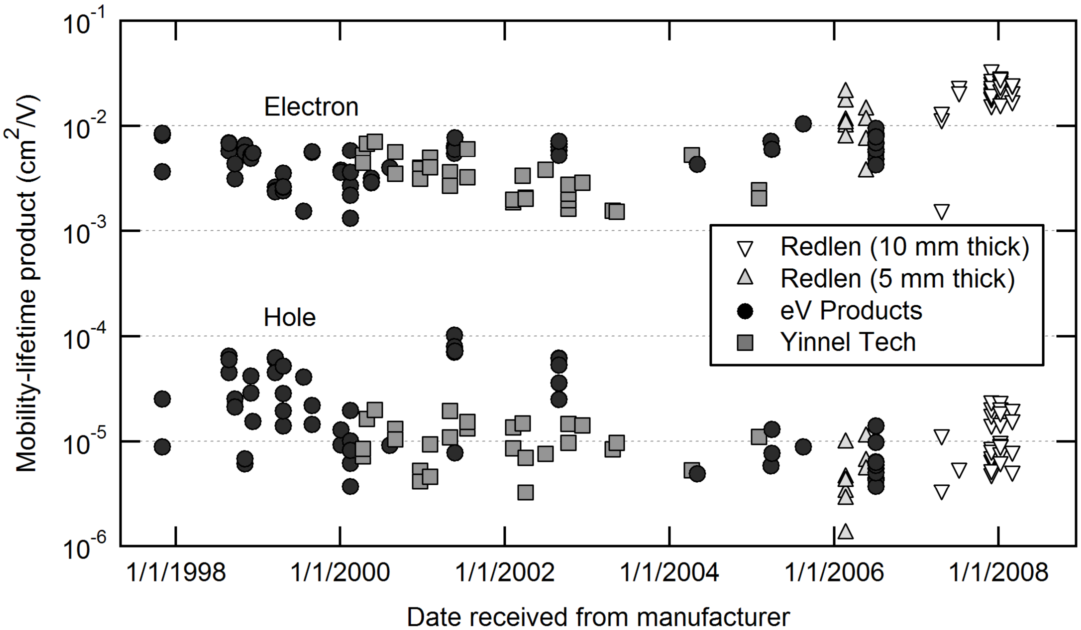

Fig. 1. Charge carrier mobility-lifetime product for electrons and holes measured from CdZnTe samples obtained from three different manufacturers. The values are plotted against the date the sample was received from the manufacturer.

two opposing crystal surfaces. The completed detector is then mounted into a vacuum chamber where one of the detector electrodes is illuminated with alpha particles from an ^{241}Am source. A bias voltage is applied across the detector to cause the collection of the electrons and holes generated by the alpha-particles within the detector. The induced charge pulses from the detector are measured with a charge-sensitive preamplifier and standard pulse-processing electronics chain. With the alpha-particles incident on the detector cathode, the electron mobility and lifetime for the crystal are extracted by measuring rise times and step heights of the induced charge pulses as a function of bias. The mobility is calculated from the rise time, and the lifetime determined by a fit of the rise time and pulse height data to the Hecht equation. Likewise the hole transport properties are determined from pulses obtained with the alphas incident on the detector anode.

Beyond the need for efficient charge carrier transport is the requirement of spatially uniform charge carrier generation and transport. Previously, we have demonstrated the use of alpha-particle response measurements to quantify and analyze uniformity [5][6]. A simple measure of electron generation and transport uniformity is obtained by uniformly illuminating the full cathode area of the planar detector with an uncollimated ^{241}Am alpha-particle source, and measuring the resultant pulse-height spectrum. Spatial variation in the electron generation or transport will lead to variation in the measured pulse heights. The spectral line shape and width, and any background that may be present will then provide an indication of uniformity. A narrow peak width and little background signify good uniformity.

The definitive test of CdZnTe material quality for gamma-ray detection is to evaluate the material as a gamma-ray detector. Since CdZnTe has poor hole transport and moderate electron trapping, good gamma-ray spectroscopic performance will not be obtained with large CdZnTe crystals at high energies using standard detection techniques. However, electron-only detection techniques such as the coplanar-grid [7] and pixels combined with depth-sensing [8] substantially eliminate these problems, and excellent spectroscopic performance can be achieved with crystals that have good spatial uniformity. We have used the coplanar-grid detector configuration to evaluate the gamma-ray detection performance of select samples. The detector fabrication is similar to that of the planar detector, except that the full-area anode is replaced with a coplanar-grid electrode structure that is defined by performing the Au evaporation through a shadow mask. The completed coplanar-grid detector is placed inside a test chamber where it is illuminated with gamma rays from a ^{137}Cs source and operated as a differential-gain coplanar-grid detector [9]. The operating conditions of the detector are then optimized in order to produce the best possible 662 keV gamma-ray peak resolution.

III. Results

We have performed the characterization measurements as described in the previous section on 29 CdZnTe crystals approximately 10 mm x 10 mm x 10 mm in size obtained from Redlen Technologies. All 29 samples were characterized for carrier transport. Eight of these samples were then fabricated into coplanar-grid detectors and characterized through their gamma response. This subset of samples was chosen such that the entire range of material uniformities existing in the total set of 29 crystals was representatively sampled (that is, samples with both large and small alpha peak widths were selected). Similar measurements were also made previously on thinner samples from Redlen (approximately 5 mm thick)

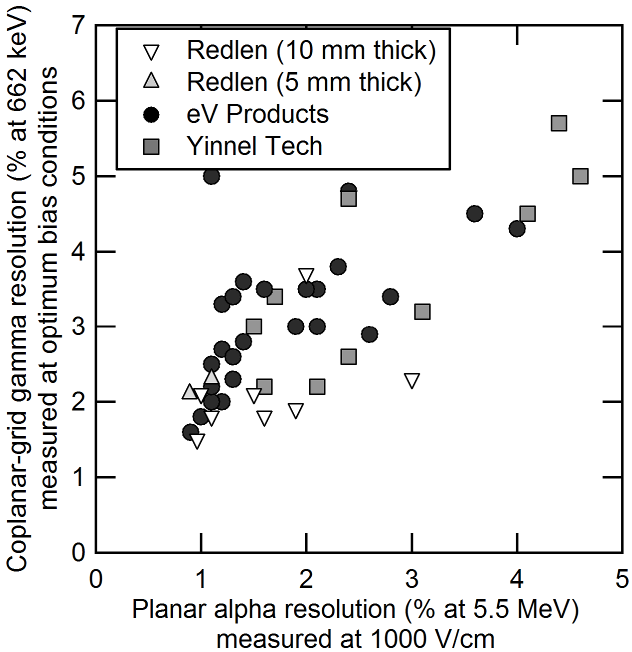

Fig. 2. Measured coplanar-grid gamma-ray energy resolution at 662 keV plotted against the planar detector alpha-particle energy resolution at 5.5 MeV for CdZnTe samples obtained from three different manufacturers. The alpha-particle measurements were made at a cathode bias of -1000 V per 10 mm thickness, whereas the gamma-ray measurements were made at biases that gave the best energy resolution. All measurements were made at room temperature.

and on samples obtained from eV Products (high-pressure Bridgman material) and Yinnel Tech (conventional Bridgman material). The samples from Redlen were typically oriented, whereas the samples from the other manufacturers were typically of an unknown crystalline orientation.

A comparison of the average charge transport for electrons and holes is given in Fig. 1. The extracted mobility-lifetime products are plotted in this figure as a function of the date the crystal was received from the manufacturer. Good electron transport and poor hole transport are consistently obtained and demonstrate the necessity of electron-only detection for achieving good spectroscopic performance with large-volume detectors. Furthermore, the 1-cm^3 crystals recently obtained from Redlen have electron transport that is higher than that of earlier samples from the other manufacturers with an average mobility-lifetime product of 2.2×10^{-2} cm^2/V as compared to 5.1×10^{-3} cm^2/V for eV Products and 3.4×10^{-3} cm^2/V for Yinnel Tech.

Another comparison between the different manufacturers is given in Fig. 2. Here the coplanar-grid detector gamma-ray energy resolution obtained from the CdZnTe samples is plotted against the planar detector alpha-particle energy resolution obtained previously with these same samples. There is a rough correlation between the two measurements since good spatial uniformity as indicated by a small alpha-particle peak width is a necessary requirement for achieving good gamma-ray spectral performance. A perfect correlation is not expected, though, since material non-uniformity is only one of several physical mechanisms leading to degraded gamma resolution. The alpha resolution provides a lower limit on the best possible gamma resolution but does not set an upper limit since other broadening mechanisms such as electronic noise may dominate. From the Fig. 2 plot, it is clear that coplanar-grid detectors produced from the 1-cm^3 Redlen crystals perform as well as the best crystals obtained earlier from the other manufacturers and typically achieve a 662 keV energy resolution near 2% FWHM.

Plotted in Figs. 3 and 4 are representative alpha-particle and gamma-ray spectra measured from four different 1-cm^3 Redlen samples. All the measurements were made at room temperature. As explained before, the alpha spectral response should provide a measure of electron generation and transport uniformity. Peak widths near or less than 1 % FWHM indicate a level of uniformity that is required for high-resolution gamma-ray spectroscopy. We have observed that some of the samples exhibit a strong detector bias dependence in their alpha response. For example (see Sample 16 in Fig. 3c), the peak obtained at -1000 V can be relatively broad while that measured at -2000 V will be below 1 % FWHM. Some amount of peak width reduction with increasing detector bias is observed in nearly all samples. A reduction in peak width with increasing bias is expected based on the following argument. The larger bias causes more rapid electron collection, and as a consequence a smaller amount of electron trapping takes place. Since this average amount of electron trapping is less, it is likely that the variation in the amount of electron trapping from event to event will also be less. This then would produce a reduced peak width. Some of the Redlen samples, however, exhibit a peak width bias dependence much larger than that typically observed in samples obtained from eV Products or Yinnel Tech. At this point, the physical mechanism leading to this strong dependence is unknown. The existence of this effect in the Redlen material will necessitate a more careful analysis of the alpha response measurements in order to assess material uniformity.

The ^{137}Cs gamma spectra shown in Fig. 4 were acquired with the samples fabricated into coplanar-grid detectors. The good energy resolution, symmetric peak shape, and high peak-valley and peak-Compton ratios obtained with most of the crystals demonstrate the suitability of the Redlen CdZnTe for large-volume gamma-ray spectrometers.

Table I and Fig. 5 summarize the data from the complete set of 1-cm^3 Redlen samples. The alpha particle measurements obtained at both -1000 V and -2000 V are included to show the bias sensitivity of the results. In contrast to what we have observed previously with eV Products material [5], the alpha peak widths from the Redlen samples at -1000 V do not appear to provide a good indication of coplanar-grid gamma resolution. Sample 16 illustrates this point in that its -1000 V peak width of 3 % would predict only a mediocre coplanar-grid gamma performance. Despite this, the sample gave a reasonably good gamma resolution of 2.3 %. A likely reason for this better than expected performance is the strong bias

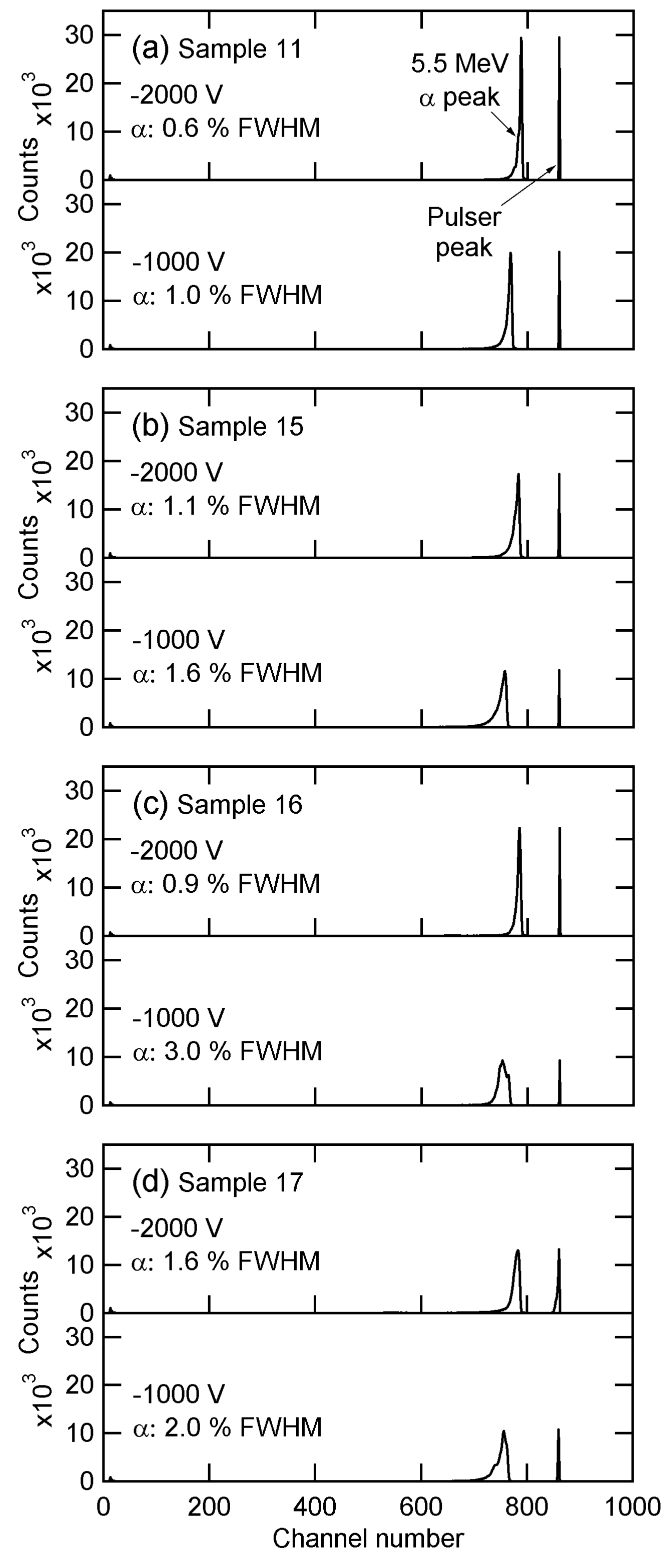

Fig. 3. ^{241}Am alpha-particle pulse-height spectra obtained with planar detectors fabricated from Redlen Technologies CdZnTe samples. The alpha-particle spectrum for each sample was measured at cathode biases of -1000 V and -2000 V. The alpha source was uncollimated and illuminated the full cathode area of the sample. All measurements were made at room temperature.

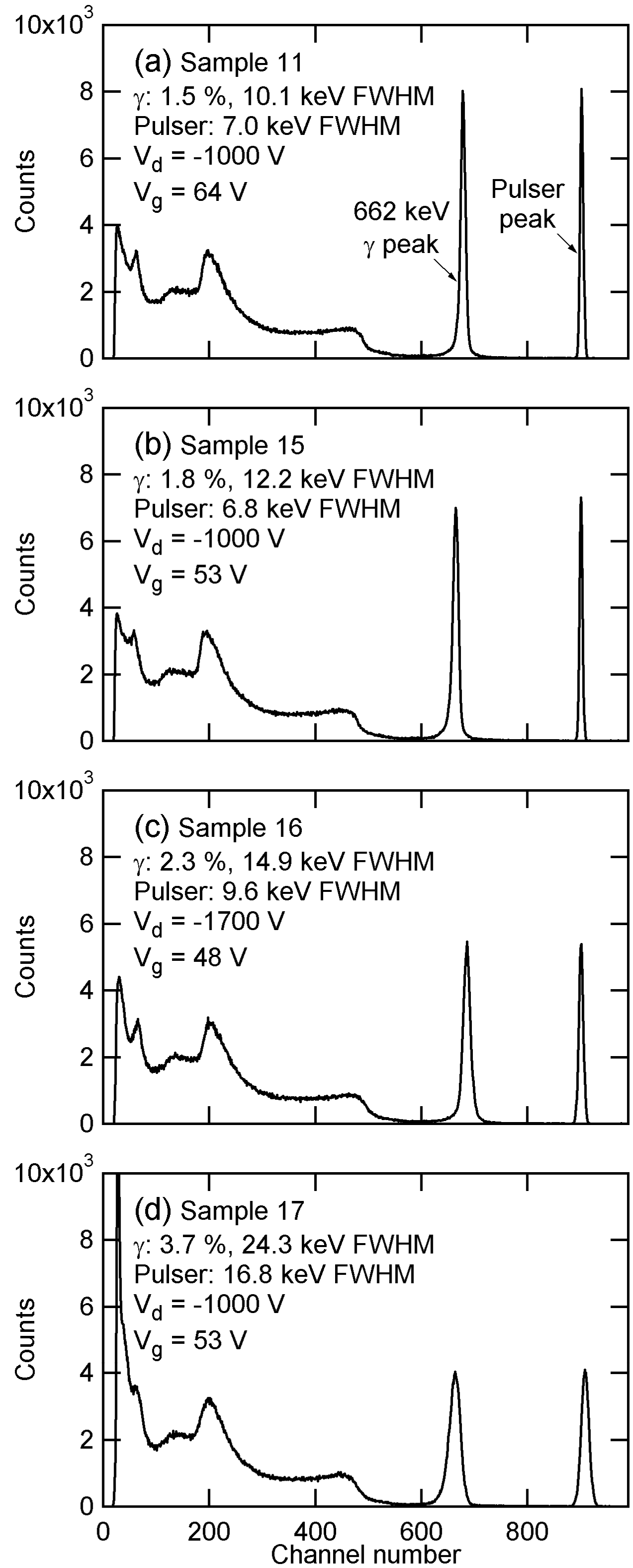

Fig. 4. ^{137}Cs gamma-ray pulse-height spectra obtained with coplanar-grid detectors fabricated from Redlen Technologies CdZnTe samples. The spectra were acquired at biases that gave the best energy resolution. All measurements were made at room temperature.

TABLE I
SUMMARY OF MEASUREMENTS FROM REDLEN SAMPLES

Sample	$\mu\tau_e$ (10^{-3} cm²/V)	$\mu\tau_h$ (10^{-6} cm²/V)	Alpha (%) -1 kV	Alpha (%) -2 kV	Gamma (%)
1	1.6	*	5.2	3	
2	12	3.4	1.5	1.2	2.1
3	13	11	1.1	1.2	1.8
4	24	5.5	1.9	0.93	1.9
5	21	3.5	3	1.9	
6	16	6.6	1.8	0.77	
7	18	7.1	1.3	0.65	
8	26	4.8	1.6	1.3	
9	28	7.7	1.5	0.78	
10	24	9.9	1.1	0.63	
11	29	9.3	0.96	0.56	1.5
12	20	8.7	1.1	0.65	
13	34	14	1.1	0.88	
14	22	18	1.6	0.73	
15	19	5.3	1.6	1.1	1.8
16	27	21	3	0.89	2.3
17	23	24	2	1.6	3.7
18	26	24	1.8	1.3	
19	27	6.3	1.5	1.2	
20	22	9.1	1	0.57	2.1
21	24	15	1.9	0.99	
22	20	8.1	1.4	0.86	
23	16	20	2	1.7	
24	**	**	**	**	
25	28	19	1.6	1.2	
26	17	7.9	1.6	0.86	
27	21	20	1.5	0.88	
28	25	5.1	1.3	0.78	
29	25	16	***	***	

Summary of the measurements made on the CdZnTe samples obtained from Redlen Technologies. The alpha numbers are planar detector alpha-particle energy resolutions at 5.5 MeV measured at cathode biases of -1000 V and -2000 V. The gamma numbers are the coplanar-grid gamma-ray energy resolutions at 662 keV. All measurements were made at room temperature.
* Signals too small to extract meaningful values.
** Too noisy to evaluate.
*** Multiple peaks in spectrum.

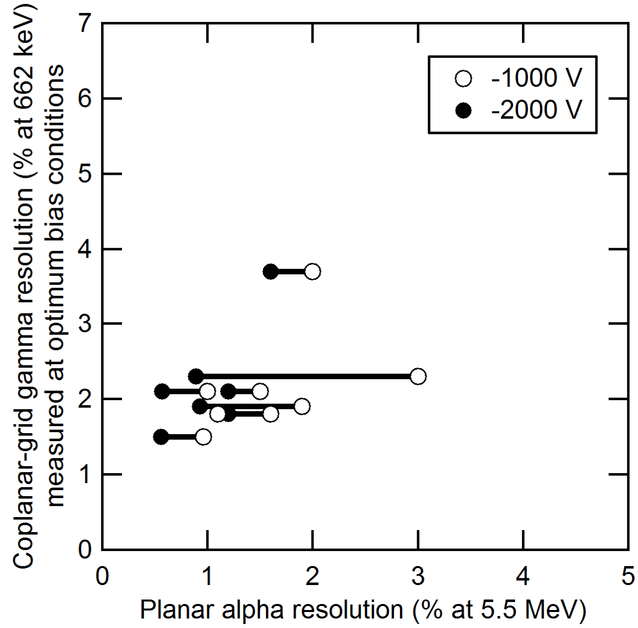

Fig. 5. Measured coplanar-grid gamma-ray energy resolution at 662 keV plotted against the planar detector alpha-particle energy resolution at 5.5 MeV for CdZnTe samples obtained from Redlen Technologies. The alpha-particle measurement on each sample was made at cathode biases of -1000 V and -2000 V, whereas the gamma-ray measurement was made at biases that gave the best energy resolution. All measurements were made at room temperature.

dependence exhibited by this sample. The alpha peak width dropped substantially from 3 to 0.89 % when the bias was increased from -1000 V to -2000 V. Consequently, we would expect to obtain a good gamma resolution if the coplanar-grid detector made from this sample was operated at a higher bias. For this sample, the optimum coplanar-grid detector bias was -1700 V, and, as a result, the reasonably good gamma resolution is consistent with that predicted by the alpha resolution if one takes into account the variation in the alpha peak width between -1000 V and -2000 V.

The optimum bias for a coplanar-grid detector is dictated by a trade-off between an increased bias reducing broadening from material and device non-uniformity and a decreased bias reducing electronic noise. The optimum bias is therefore not known in advance of the actual coplanar-grid detector testing. This does not impact the use of the planar detector alpha response method for screening material as long as there is little variation in the alpha peak width over the bias range used for the coplanar-grid detectors. Since this is not always the case with the Redlen material, it is necessary to use a slightly more complex screening process in which the alpha peak width is measured at, for example, the limits of the expected operating bias (or simply at the high bias limit). Nonetheless it appears that, when carefully applied, such measurements can be used as the basis to screen the Redlen materials for high resolution gamma-ray detector fabrication.

In summary, our analysis of 10-mm thick CdZnTe material grown by Redlen Technologies using the Traveling Heater Method indicates that the material has electron mobility-lifetime products that are on average higher than that measured previously on materials from two other manufacturers and spatial uniformity in electron generation and transport that is comparable to some of the better materials. As expected based on the electron transport measurements, coplanar-grid detectors fabricated from the Redlen samples exhibit good gamma-ray spectroscopic performance with a typical energy resolution at 662 keV near 2% FWHM when operated at room temperature. Overall, we conclude that the material holds great promise for large-volume gamma-ray detector applications.

REFERENCES

[1] C. Szeles and M. C. Driver, "Growth and properties of semi-insulating CdZnTe for radiation detector applications," Proceedings of SPIE, vol. 3446, 1998, pp. 2-9.

[2] H. Chen, S. A. Awadalla, J. Mackenzie, et al., "Characterization of traveling heater method (THM) grown $Cd_{0.9}Zn_{0.1}Te$ crystals," IEEE Trans. Nucl. Sci., vol. 54, 2007, pp. 811-816.

[3] H. Chen, S. A. Awadalla, F. Harris, et al., "Spectral response of THM grown CdZnTe crystals," IEEE Trans. Nucl. Sci., vol. 55, 2008, pp. 1567-1572.

[4] H. Chen, S. A. Awadalla, K. Iniewski, et al., “Large-volume, high-resolution cadmium zinc telluride radiation detectors: recent developments,” Proceedings of SPIE, vol. 6706, 2007, pp. 670602-1-14.

[5] M. Amman, P. N. Luke, and J. S. Lee, “CdZnTe material uniformity and coplanar-grid gamma-ray detector performance,” IEEE Trans. Nucl. Sci., vol. 47, 2000, pp. 760-767.

[6] M. Amman, J. S. Lee, and P. N. Luke, “Electron trapping nonuniformity in high-pressure-Bridgman-grown CdZnTe,” J. Appl. Phys., vol. 92, 2002, pp. 3198-3206.

[7] P. N. Luke, “Unipolar charge sensing with coplanar electrodes – application to semiconductor detectors,” IEEE Trans. Nucl. Sci., vol. 42, 1995, pp. 207-213.

[8] Z. He, W. Li, G. F. Knoll, et al., “3-d position sensitive CdZnTe gamma-ray spectrometers,” Nucl. Instr. and Meth. A, vol. 422, 1999, pp. 173-178.

[9] M. Amman and P. N. Luke, “Optimization criteria for coplanar-grid detectors,” IEEE Trans. Nucl. Sci., vol. 46, 1999, pp. 205-212.