BNL-99912-2013-CP

Growth and Characterization of CdTeSe for Room-Temperature Radiation Detector Applications

Utpal Roy

Presented at the SPIE Optics and Photonics 2013

September 2013

Nonproliferation and National Security Department

Brookhaven National Laboratory

P.O. Box 5000

Upton, New York 11973

www.bnl.gov

Notice: This manuscript has been authored by employees of Brookhaven Science Associates, LLC under Contract No. DE-AC02-98CH10886 with the U.S. Department of Energy. The publisher by accepting the manuscript for publication acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

This preprint is intended for publication in a journal or proceedings. Since changes may be made before publication, it may not be cited or reproduced without the author's permission.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or any third party's use or the results of such use of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof or its contractors or subcontractors. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Growth and characterization of CdTeSe for room-temperature radiation detector applications

U. N. Roy¹, A. E. Bolotnikov¹, G. S. Camarda¹, Y. Cui¹, A. Hossain¹, G. Yang¹, R. B. James¹, A. Fauler², M. Fiederle², M. Sowinska³, G. Hennard³ and P. Siffert³

¹Brookhaven National Laboratory, Upton, NY, USA

²Freiburger Material for schungszentrum FMF, Stefan-Meier-Strasse 21, 79104 Freiburg, Germany

³Eurorad S.A., Rue Ettore Bugatti, 67201 Eckbolsheim, France

ABSTRACT

The near-unity segregation coefficient of Se in a CdTe matrix ensures the compositional homogeneity, both axial- and radial, of the CdTeSe ternary compound, so making it a material of choice for room- temperature radiation detectors. In this study, we grew CdTeSe crystals by the Traveling Heater Method (THM), using Te as the solvent, and characterized the crystals by IR transmission microscopy, white-beam X-ray diffraction topography, and low-temperature photoluminescence. The total average concentration of the secondary phases obtained for the CdTeSe sample was about $7 \times 10^4 \text{ cm}^{-3}$ for crystals grown at two different laboratories. The best resistivity registered was $5 \times 10^9 \text{ ohm-cm}$, and the estimated $\mu\tau$ product for the electrons was $3\text{-}4 \times 10^{-3} \text{ cm}^2/\text{V}$.

1. INTRODUCTION

The ternary compound $\text{CdTe}_x\text{Se}_{1-x}$ (CTS) could be a good choice for usage in room-temperature radiation detectors. It has several advantages over CdZnTe (CZT), particularly the near-unity segregation coefficient of Se in CdTe that ensures the axial- and radial-compositional uniformity of the grown ingots^{1,2}. The ability to obtain larger, more uniform detector-grade crystals than those presently available potentially could drastically reduce the cost of producing such detectors compared to those made from CdZnTe. Here, CTS offers another advantage, viz., that of having a lower electron effective-mass compared to CdTe³. Furthermore, selenium assures effective solution hardening, judged against the Zn in the CdTe matrix^{1,4} so drastically reducing the sub-grain boundary network. The presence of sub-grain boundaries in present-day CZT crystals are known to be the most detrimental factor for the device's performance⁵. Therefore, we consider that CST offers many advantages over CZT. Indeed, during the early developmental stages of CZT as detector material in 1994, it was demonstrated that CTS was more promising than CZT and CdTe⁶. Hence, we considered that it was worth reconsidering the ternary compound $\text{CdTe}_x\text{Se}_{1-x}$ for radiation-detectors. In this paper, we discuss our growth and characterization of $\text{CdTe}_x\text{Se}_{1-x}$ crystals, and detail some preliminary results on the properties of devices fabricated from it.

2. EXPERIMENTAL

We grew the $\text{CdTe}_x\text{Se}_{1-x}$ samples that we characterized for our research by the Traveling Heater Method (THM) with a 10% nominal concentration of Se. Herein, we also discuss some of the results with THM-grown CTS samples generated by the collaborations of M. Fiederle and the Eurorad group; furthermore, we include the preliminary characterization of samples grown at Brookhaven National Laboratory. All our samples were grown from a Te-rich solution at the rate of $\sim 3 \text{ mm/day}$. The samples were cut and polished to a mirror-like finish by a final polishing using $0.05\text{-}\mu\text{m}$ alumina with a felt pad. Samples were characterized by IR transmission microscopy, and white beam X-ray diffraction topography in the reflection mode using the NSLS facility at BNL's X19C beam line. To explore the crystalline quality of the samples, we used low-temperature photoluminescence (PL) at 4.2 K. High-resolution charge-collection mapping was carried out at BNL's beam line, X27B. Finally, we fabricated planar devices from samples measuring $10 \times 10 \times 1 \text{ mm}^3$ using chemically deposited gold as the electrode. Their resistivity and charge-transport characteristics were evaluated by

measuring their I-V characteristics; we plotted the efficiency of charge collection versus applied bias to derive the $\mu\tau$ product for electrons at room temperature. Finally, we assessed the characteristics at room temperature for our planar detectors using a ^{241}Am gamma source.

3. RESULTS AND DISCUSSION

Figure 1 represents a typical optical photograph of a CTS sample of $10\times 10\times 1\text{ mm}^3$, wherein the grain boundaries are visible. The sample was etched in a 2% bromine-methanol solution. Figure 2 is the X-ray diffraction topographic image of the large grain taken in the reflection mode. The bending on the topographic image is the signature of stress associated

Figure 1. Optical photograph of the CTS sample
(dimensions: $10\times 10\times 1\text{ mm}^3$)

Figure 2. White-beam X-ray topographic
image of the CTS sample

with the sample. In addition, large sub-grain boundaries are visible as dark lines in the topographic image. The IR transmission image of the sample is shown in figure 3, while an enlarged image of the rectangle therein is presented in figure 4; the area of the image is $\sim 2.5\times 2\text{ mm}^2$. The presence of the secondary phases is clearly visible. To investigate the concentration and size distribution of the total average secondary phases in the CTS samples, we studied a sample, 5-mm thick, to obtain better statistics. Figure 5 is its typical histogram, illustrating the concentration and size-distribution of the secondary phase present within it. We explored the distribution of the inclusions over the volume of $1.1\times 1.5\times 5\text{ mm}^3$, as presented in Fig. 4. The concentrations of the secondary phase were measured at different depths over the sample's entire thickness, viz., 5 mm in the present case. Figure 5 is the result of superimposing all the IR transmission images taken at different depths; we show the image as negative, so that the inclusions appear as white spots, instead

Figure 3. IR-transmission image of the CTS
sample in Fig. 1. (dimensions: $10\times 10\text{ mm}^2$)

Figure 4. Enlarged view of the marked portion
in fig. 3. (dimensions: $2.5\times 2.0\text{ mm}^2$)

of black ones as in a normal image. We measured the total average concentration of the secondary phases in the CTS sample as $7\times 10^4\text{ cm}^{-3}$, a value that is 5-8 times lower than that in the standard CdZnTe available today⁷. The histogram

of size distribution extends up to 17-18 μm , with the peak at about 7-8 μm . We consider that the lower concentration of the secondary phases is very encouraging compared to that in CZT; further, we expect the charge-transport characteristics and their uniformity will be superior to those in the presently available CZT. Similar behavior was observed for the crystals grown at BNL by the THM technique. The total average concentration of the secondary phases obtained for BNL's CTS sample was $6.6 \times 10^4 \text{ cm}^{-3}$, i.e., strikingly similar to the value for CTS sample provided by M. Fiederle and the Eurorad group. Although making a definitive statement is premature, seemingly the lower concentration of the secondary phase, compared to CZT, possibly is an intrinsic property of the CTS ternary compound. Our low-temperature photoluminescence study at 4.2 K showed sharp well-resolved excitonic peaks that are the signature of the sample's good crystallinity.

Figure 5. Concentration and size distribution of the secondary phases in the CTS sample. The volume investigated was $1.1 \times 1.5 \times 5 \text{ mm}^3$.

Figure 6. Superimposed IR transmission image over in a 5 mm-thick crystal. The image area is $1.1 \times 1.5 \text{ mm}^2$.

Figure 7. Room- temperature ^{241}Am gamma response for a CTS planar detector.

Figure 8. Charge collection versus applied bias for a CTS sample using 59.6-keV gamma line of ^{241}Am .

The resistivity of the sample was $5 \times 10^9 \text{ ohm-cm}$. The pulse height spectrum for a planar detector of $10 \times 10 \times 1 \text{ mm}^3$ is shown in figure 7; it was obtained from an ^{241}Am gamma source under an applied bias of 25 V. We note that even at very low applied bias of 5 V across a 1-mm-thick sample a well-resolved gamma peak was also revealed, which is very unusual for a similar sized standard CZT sample. This finding implies that its charge- transport properties are very good. To evaluate the mobility-lifetime product for electrons, we plotted charge collection versus applied-bias voltage for the detector in planar geometry using ^{241}Am gamma line and fitted with the Hecht's relation; this is illustrated in figure 8.

The $\mu\tau$ product for an electron estimated from the Hecht's equation was $3.5 \times 10^{-3} \text{ cm}^2/\text{V}$. This result is very encouraging for our early growth experiments; we expect to enhance the $\mu\tau$ product by optimizing the growth parameters and purifying the starting materials.

4. SUMMARY

Reasonably good spectroscopic CdTeSe (CTS) samples were grown by the THM technique. The average concentration of the secondary phases present in the CTS crystals grown from two different sources are encouragingly similar; the value obtained was $7 \times 10^4 \text{ cm}^{-3}$, a level that is 5-8 times lower than in commercial CZT presently available. For our early grown samples, the $\mu\tau$ value for electrons was $3.5 \times 10^{-3} \text{ cm}^2/\text{V}$, which also is very promising. Higher resistivity and $\mu\tau$ values may be realized after we purify the material and optimize the growth parameters. Overall, we consider that CTS is very promising for radiation-detector applications, and it has remarkable potential for replacing CZT.

5. ACKNOWLEDGEMENT

This work was supported by the U.S. Department of Energy, Defense Nuclear Nonproliferation R&D.

6. REFERENCES

- [1] C. J. Johnson, SPIE 1106 (1989) 56.
- [2] K. H. Kim, J. K. Hong and S. U. Kim, Journal of Crystal Growth 310 (2008) 91.
- [3] L. Hannachi and N. Bouarissa, Superlattices and Microstructures 44 (2008) 794.
- [4] P. Rudolph, Prog. Crystal Growth and Charact. 29 (1994) 275.
- [5] A. E. Bolotnikov, S. Babalola, G. S. Camarda et al., IEEE Trans. Nucl. Sci. 58 (2011) 1972.
- [6] M. Fiederle, D. Ebling, C. Eiche et al., Journal of Crystal Growth 138 (1994) 529.
- [7] A. E. Bolotnikov, S. Babalola, G. S. Camarda et al., IEEE Trans. on Nucl. Sci. 57 (2010) 910.