U.S. DEPARTMENT OF
ENERGY

PNNL-22869

Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

Annual Report: Property Improvement in CZT via Modeling and Processing Innovations

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September 2013

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Annual Report

Property Improvement in CZT via Modeling and Processing Innovations

PL10-CZTproc-PD05

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Date:	**September 24, 2013**

Property Improvement in CZT via Modeling and Processing Innovations

PL10-CZTproc-PD05

1. AIM AND SIGNIFICANCE

The objective of this project is to develop growth models of CZT crystals from the melt using vertical gradient freeze (VGF) as a typical process. Further, the project will perform critical experiments to validate growth models and to provide detailed data for modeling and simulation. Ideally, the project will develop growth models that will provide, for the first time, choices for optimal CZT single crystal growth from the melt based on model input. In our view this depends on 1) understanding crystal growth processes, including annealing and cool-down processing, and 2) understanding the role of defects on detector response since it is not possible, yet, to produce defect-free materials. Models of defect structure and formation are addressed. Validated models and experiments on reducing defects in melt-grown crystals are used to guide our understanding of growth processes and in-furnace annealing plus cool-down.

This work has demonstrated that CZT growth occurs naturally along the <110>-direction, which results in facetted growth fronts, higher Te-particle content, and higher dislocation density. Since dislocations appear to seed Te-particle nucleation and growth this is not optimal. Rather, *we find that <111>-direction appears to be the optimal growth direction resulting in lower Te-particle content and reduced dislocation densities*, but that this growth direction requires seeded growth.

2. TECHNICAL APPROACH

We created a multiscale-modeling framework for simulations of CZT growth from the melt and validated these models using data from a VGF growth of CZT at PNNL and from a variety of CdTe laboratory-scale growths. The multiscale approach ultimately ran into issues at the atomistic-scale with an inability to create an accurate interatomic potential for CdTe or CZT. However, valuable information has been generated by this project using molecular dynamics of solidification that impact CZT growth choices. Other modeling tools were developed at the mesoscale level that were more successful in linking theory to experiment using phase field methods and experimental data.

This project made good use of new and modern characterization methods to quantify Te-particle orientations in CZT, to study the size and spatial distributions of Te-particles in CZT, and to study the growth of CdTe from the melt. Electron backscatter diffraction (EBSD) and transmitted IR (TIR) imaging with 3D mapping tools provided us with new and important data for CZT melt growth physics. Growths of Te-rich CdTe as a function of cooling rates in laboratory-scale growths revealed some important growth mechanisms for the first time.

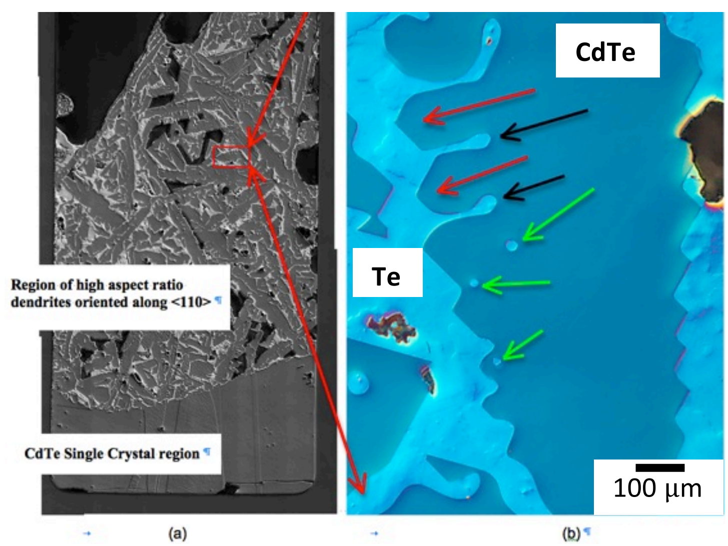

Figure 1. CdTe laboratory-scale growth. In (a) is entire freeze with <110> dendrites. In (b) is high resolution image showing characteristic <110> growth fingers and Te-particle capture.

3. SCIENTIFIC FINDINGS

We developed a phase field model describing the diffusional growth and crystallography of large Te-particles in CZT. We found that the particles occupy a (111)-tetrahedron within the CZT crystal with the

(111) surface energy of the CZT determining the minimum energy morphology at equilibrium. Molecular dynamics (MD) results (see Figure 2) reveal that a (111) growth interface is preferred for smooth interfaces and low defect densities. However, since the <110> or <112> growth direction is self-selected by CZT this will require (111) seeded growth to achieve.

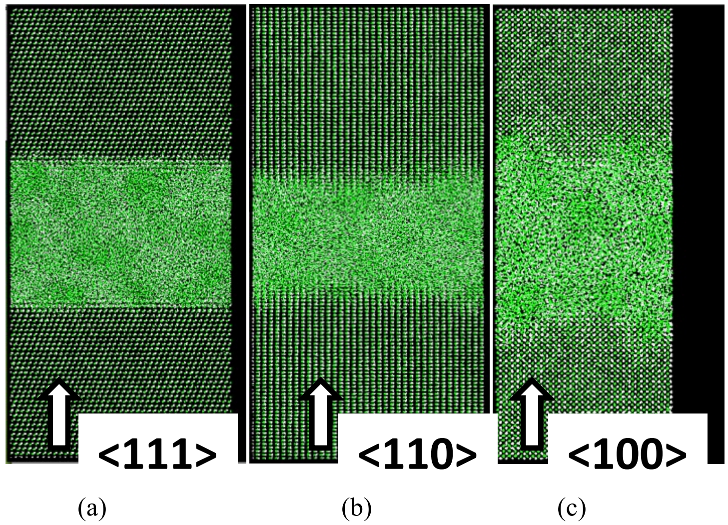

Figure 2. Atomic models using PNNL-developed potential for CdTe showing (a) <111>, (b) <110>, and (c) <100> growth directions. The <110> direction is fastest growing and self-selected but is rough as is the <100> growth interface. In contrast the <111> is slower via a ledge growth mechanism and is much smoother leading to lower defect densities. This can only be chosen via seeded growth, however.

Our most important finding may be the experimental work that we have done using TIR imaging and 3D particle mapping to determine the Te-particle size and spatial distributions within an as-grown CZT boule. The boule slice is shown in Figure 3 and the spatial distribution of Te-particles from a large volume within the boule slice is shown in Figure 4. The key result is the Te-particles form a network that greatly resembles a dislocation cellular network implying that the Te-particles in CZT may indeed coexist with dislocations. This is significant in helping determine optimal growth strategies.

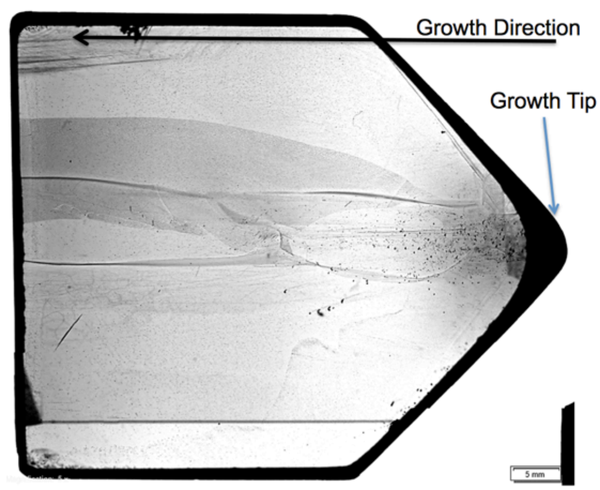

Figure 4. Transmitted IR image of CZT boule slice grown at PNNL. Speckles in the image are Te-particles.

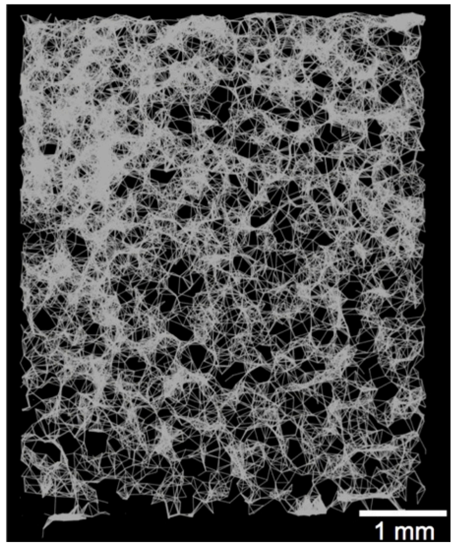

Figure 5. Cellular network model of Te-particles in CZT from Fig. 4. Stick bonds up to 175 μm long connect the Te-particles. The cell size is about 150 μm, which corresponds to the dislocation network size in CZT.

4. PATH FORWARD

This research has identified a need for determining the effects of as-grown CZT microstructures and Te-particle distributions on energy loss and energy resolution. Currently, there is no adequate theory of energy resolution degradation for semiconductor radiation detectors with as-grown defects. We recommend such a study, since without such a theory it is difficult to envision improved crystal growth models or optimal growth process developments.

5. PUBLICATIONS AND INVITED PRESENTATIONS

CH Henager, Jr., DJ Edwards, AL Schemer-Kohrn, SK Sundaram, BJ Riley, and M Bliss, "Electron Backscatter Diffraction of a Ge Growth Tip from a VGF Furnace", J. Cryst. Growth (2008) 311(1):10-14.

SY Hu and CH Henager, Jr., "Phase-field Simulations of Te-Precipitate Morphology and Evolution Kinetics in Te-Rich CdTe Crystals", J. Cryst. Growth (2009) 311(11):3184-3194.

CH Henager, Jr., DJ Edwards, AL Schemer-Kohrn, M Bliss, and JE Jaffe, "Preferential orientation of Te precipitates in melt-grown CZT", J. Cryst. Growth (2009) 311(9):2641–2647.

CH Henager, Jr. and JR Morris, "Atomistic simulation of CdTe solid-liquid coexistence equilibria", Physical Review B (2009) 80(24):Article Number: 245309.

SK Sundaram, CH Henager, Jr., DJ Edwards, AL Schemer-Kohrn, M Bliss, and BJ Riley, "EBSD Analysis of a CZT Growth Tip from a VGF Furnace", J. Cryst. Growth (2011) 329(1):12-19.

SY Hu, CH Henager, Jr., and LQ Chen, "Simulations of Stress-induced Twinning and De-twinning: a Phase-field Model", Acta Materialia (2010) 58(19):6554-6564.

G Lin, J Bao, AM Tartakovsky, and CH Henager, Jr., "A phase-field model coupled with lattice kinetics solver for modeling crystal growth in furnaces", Commun. Comput. Phys. (2012) 15 (2014), pp. 76-92.

CH Henager, Jr., "Crystal Growth Modeling: Atoms to Continuum", Presented by Chuck Henager, Jr. (Invited Speaker) at NA-22 Theory and Modeling Workshop, Berkeley, CA on October 30, 2008.

CH Henager, Jr., and JR Morris, "CdTe-interfaces", Presented by James R Morris (Invited Speaker) at CMSN workshop, San Francisco, CA on February 19, 2009.

CH Henager, Jr., "Multiscale Modeling of Crystal Growth and Microstructural Evolution of CdZnTe", Presented by Chuck Henager, Jr. (Invited Speaker) at APS March Meeting (American Physical Society), Baltimore, MD on March 21, 2013.

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