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Low temperature photoluminescence (PL) and high-resolution X-ray diffraction studies of travelling heater method (THM) grown $Cd_{0.9}Zn_{0.1}Te_{0.98}Se_{0.02}$ crystals

Utpal N. Roy[1], Jonathon N. Baker[1], Giuseppe S. Camarda[2], Yonggang Cui[2], Ge Yang[3] and Ralph B. James[1]

[1]Savannah River National Laboratory, Aiken, SC 29808, USA.

[2]Brookhaven National Laboratory, Upton, NY 11973, USA.

[3]North Carolina State University, Raleigh, NC 27695, USA.

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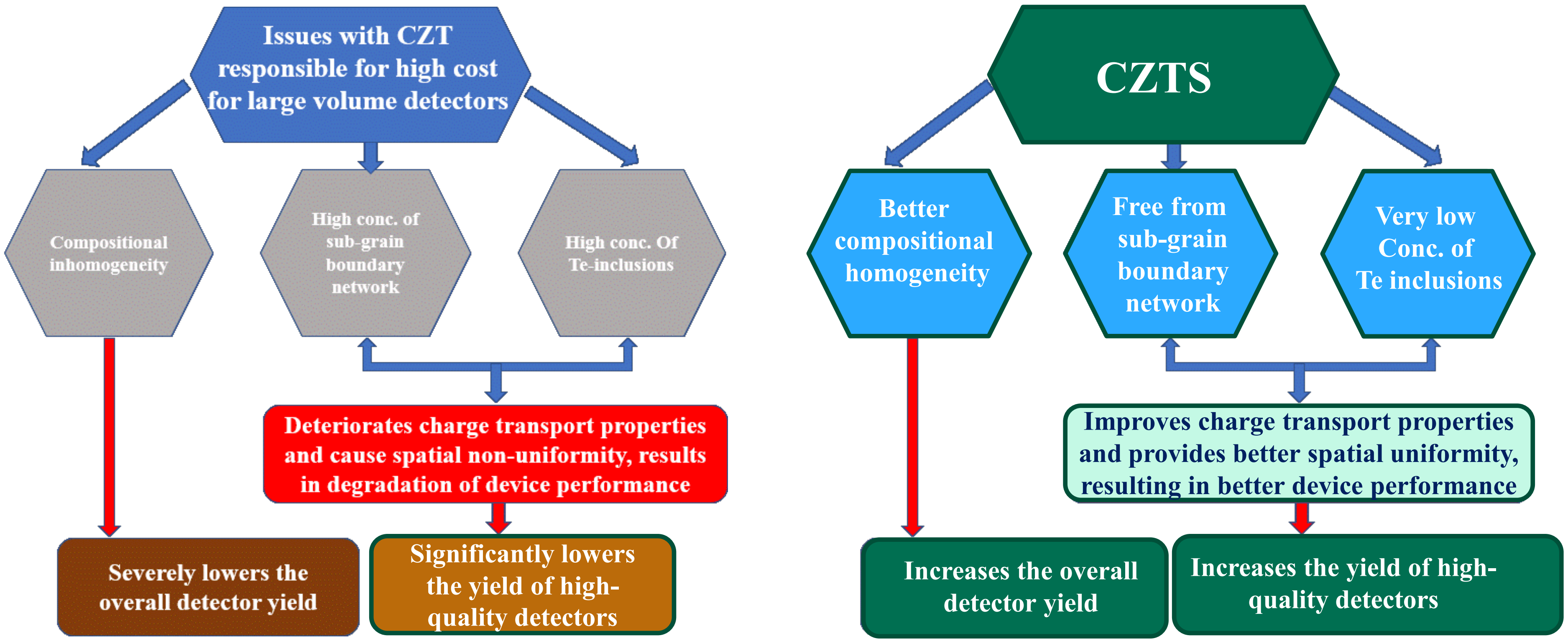
Advantages of CdZnTeSe in the present form



Benefits of Se in CdZnTeSe matrix

- Strong influence in modifying the Zn segregation coefficient: better compositional homogeneity (over about 90% of the ingot length) for THM-grown ingots.

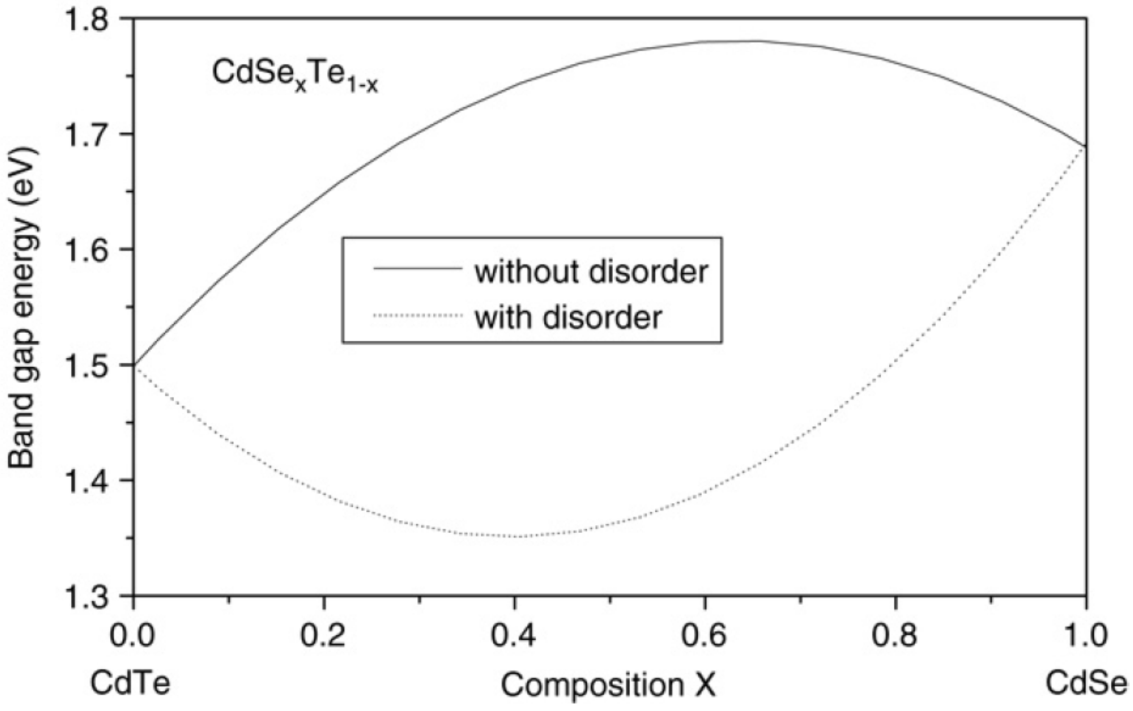
- Effective solution hardening in arresting sub-grain boundaries and their network (free from sub-grain boundary network).

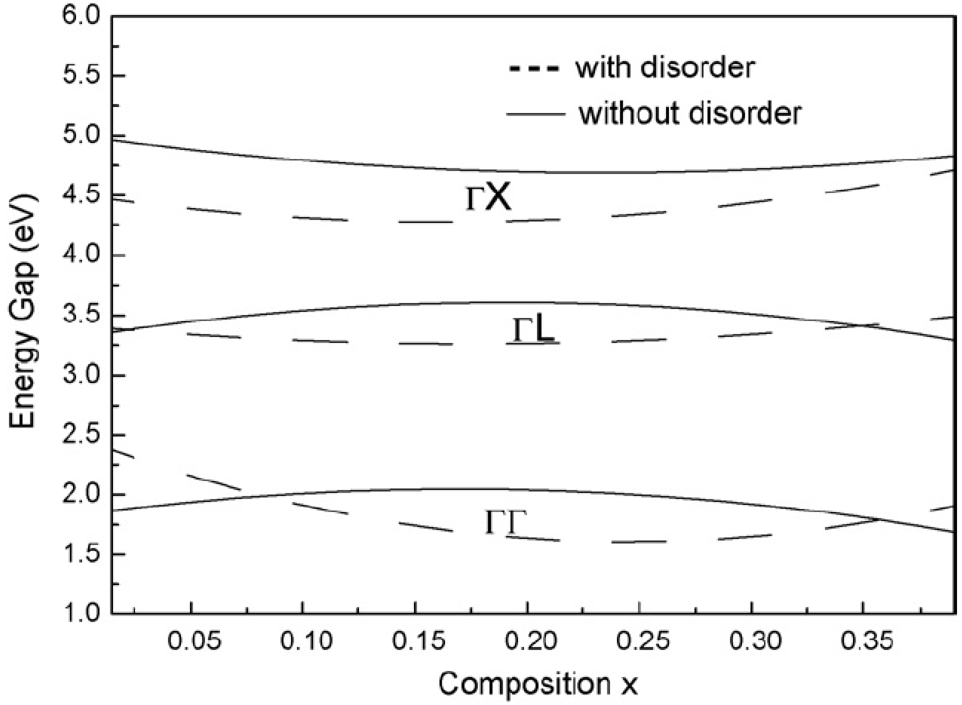
- Decreased concentrations of Te inclusions/precipitates.

- Reduced Cd vacancies.

- Reduced deep-level trap density.

However, CdTeSe/CdZnTeSe suffers from compositional/lattice disorder





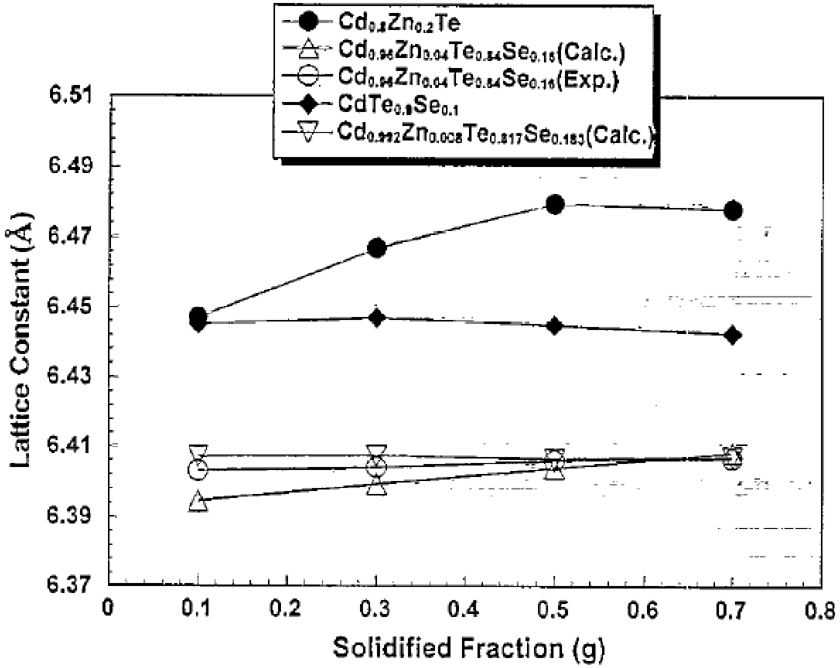
Direct band gap-energy ($E^{\Gamma}{}_{\Gamma}$) in $CdSe_xTe_{1-x}$ mixed crystals as a function of Se concentration. *(Taken from L. Hannachi et al., Superlattices and Microstructure 44 (2008) 794.)*

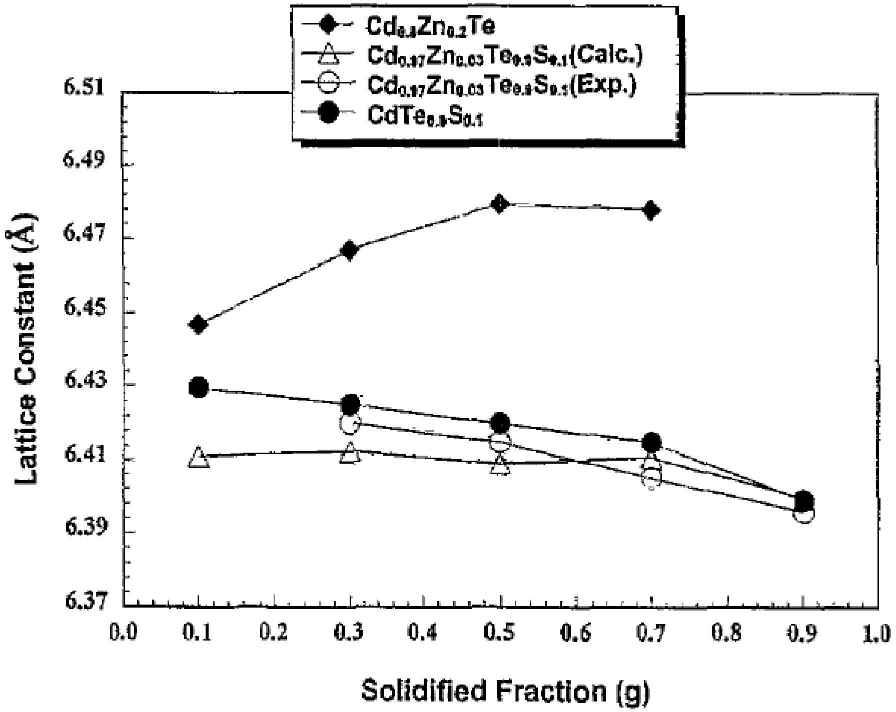
Variation of direct gap at Γ and indirect gap at X and L as a function of Se concentration in $Zn_yCd_{1-y}Se_xTe_{1-x}$ crystals. *(Taken from L. Hannachi et al., Superlattices and Microstructure 44 (2008) 794.)*

Brill et al. reported the empirical formula, $E_g(x,y) = 1.511 - 0.54x + 0.6y$ ($x,y \leq 0.10$), for $Cd_{1-y}Zn_ySe_xTe_{1-x}$ (G. Brill, JEM 34 (2005) 655.)

Band gap bowing is due to the compositional/lattice disorder.

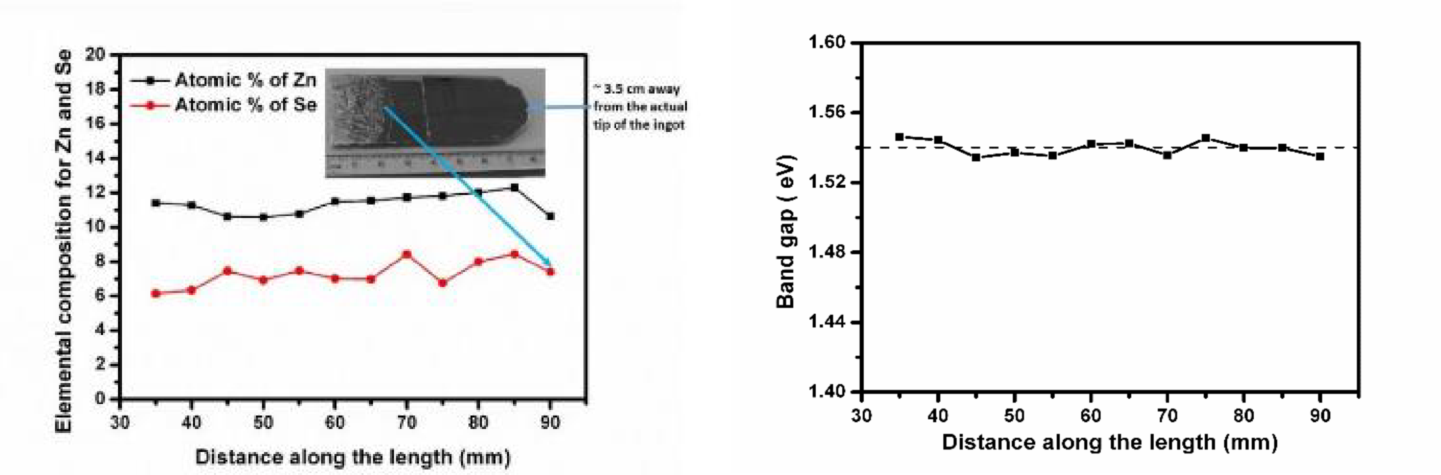
CdTeSe/CdZnTeSe macroscopic composition variation (Bridgman growth)



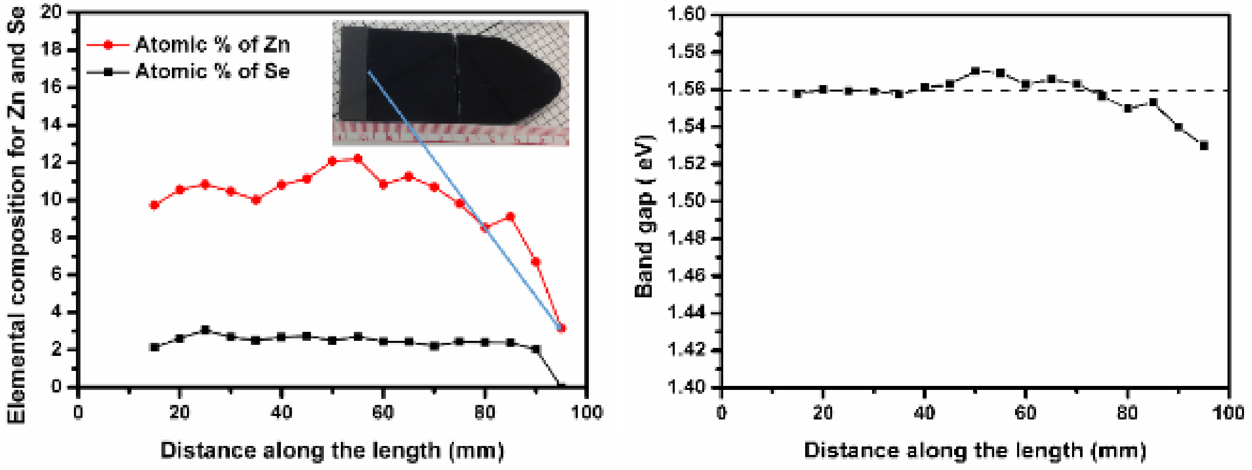


Lattice constant versus solidified fraction of the ingots of CdTe alloyed with Zn and Se, and with Zn and S. Bridgman Growth technique used. *(Taken from C.Y. Chang and B.H. Tseng, Mat. Sc. and Engr. B49 (1997) 1).*

CdTeSe/CdZnTeSe macroscopic composition variation (THM growth)



$Cd_{0.9}Zn_{0.1}Te_{0.93}Se_{0.07}$ *(Taken from U.N. Roy et al., Sc. Rep. 9 (2019) 1620).*



$Cd_{0.9}Zn_{0.1}Te_{0.98}Se_{0.02}$ *(Taken from U.N. Roy et al., Sc. Rep. 9 (2019) 7303).*

The compositional/lattice disorder is expected to affect the overall crystalline quality

DCRC data of CdTe crystals alloyed with Zn, Se and S

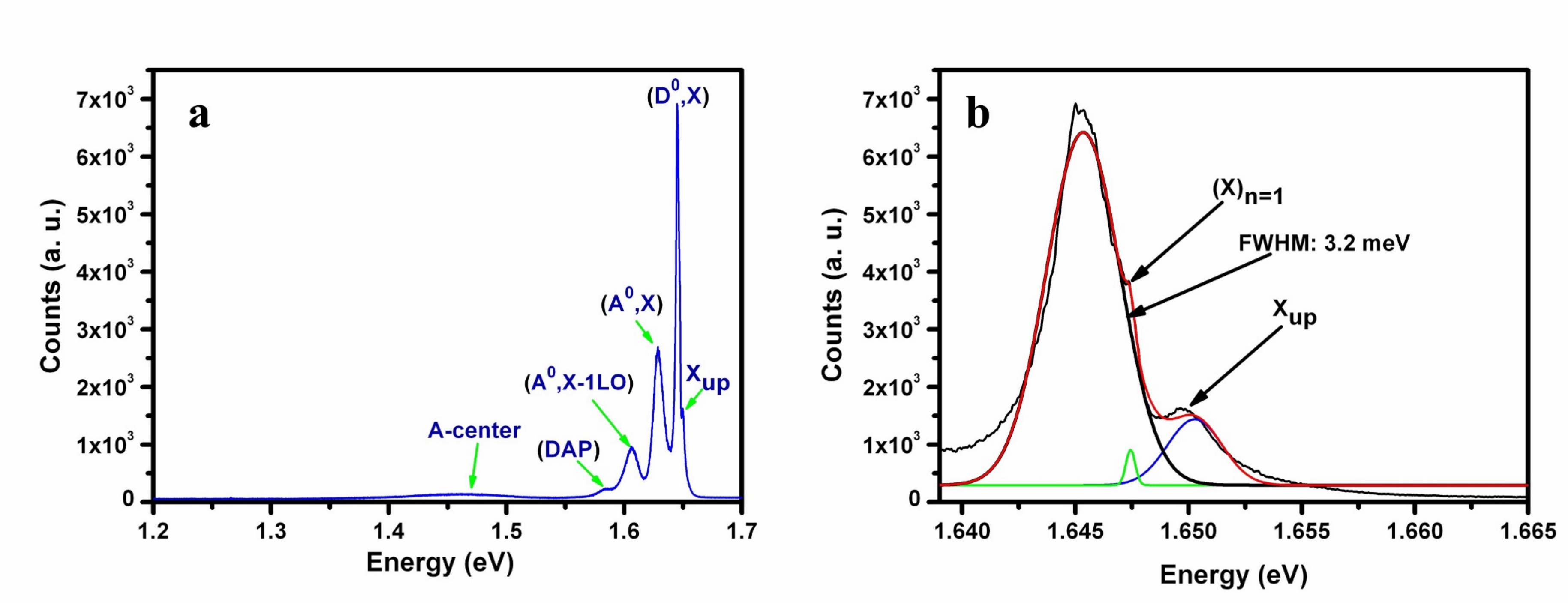
Alloy composition	Growth method[a]	Sampling position[b]	FWHM (arc s)
$Cd_{0.96}Zn_{0.04}Te$	VB	0.5	70
$CdTe_{0.96}Se_{0.04}$	VD	0.5	80
$CdTe_{0.9}Se_{0.1}$	VB	0.5	85
$\boxed{CdTe_{0.9}S_{0.1}}$	VB	0.5	93
$Cd_{0.97}Zn_{0.03}Te_{0.9}S_{0.1}$	VB	0.7	103
$Cd_{0.96}Zn_{0.04}Te_{0.84}Se_{0.16}$	VB	0.5	107

[a] VB, vertical Bridgman method.
[b] *g*, solidified fraction along the ingot.

Increase in Se concentration degrades the crystalline quality. *(Taken from C.Y. Chang and B.H. Tseng, Mat. Sc. and Engr. B49 (1997) 1.)*

In connection to effective Zn segregation and crystalline quality, Se is preferable compared to S.

Low-temperature photoluminescence of THM-grown $Cd_{0.9}Zn_{0.1}Te_{0.98}Se_{0.02}$

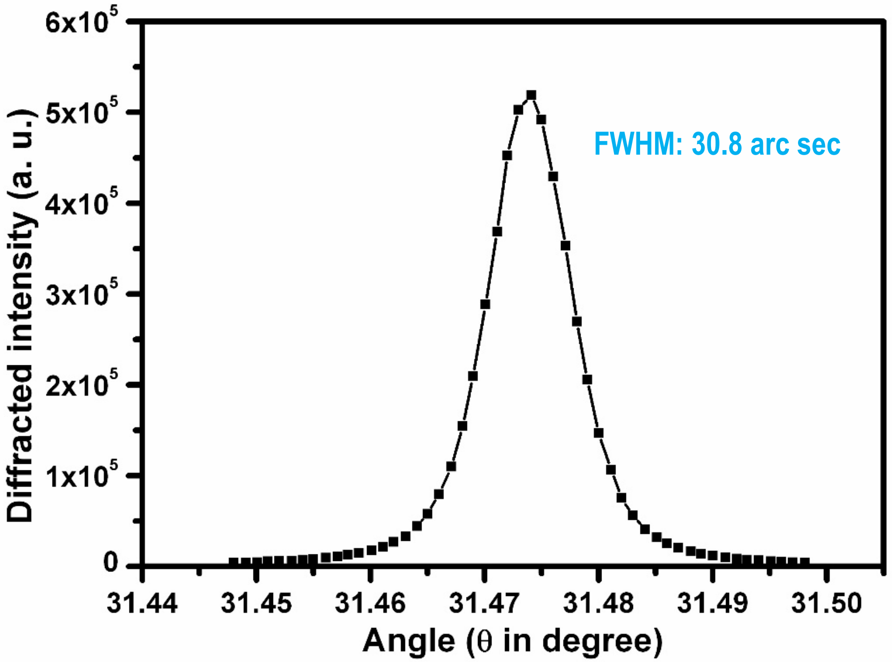


(a) Low-temperature (4.2 K) photoluminescence spectrum of the as-grown $Cd_{0.9}Zn_{0.1}Te_{0.98}Se_{0.02}$ sample and (b) enlarged version of the (D^0, X) peak.

FWHM of D^0X peak is 3.2 meV. The corresponding value for CZT grown using the same growth conditions was 2.2 meV.

The observed higher FWHM might result from the lattice disorder effect.

High resolution XRD of THM-grown $Cd_{0.9}Zn_{0.1}Te_{0.98}Se_{0.02}$



High resolution x-ray diffraction curve of an as-grown $Cd_{0.9}Zn_{0.1}Te_{0.98}Se_{0.02}$ sample grown by the THM.

The high-resolution x-ray diffraction (HRXRD) study was carried out at NSLSII beam line 4-ID at Brookhaven National Laboratory (BNL). The energy of the monochromatic beam used for the diffraction measurements was 7.11 keV with a beam size of 0.50 x 0.05 mm^2.

Density functional theory (DFT) used to estimate the bond length/lattice disorder in CdZnTeSe

Table 1: Summary of representative bond length statistics from a subset of the SQS* simulations

		Bond length (Å)		Ideal (110) spacing (Å)	
Composition	Bond type	Mean	Stnd. dev	Mean	Stnd. dev
$Cd_{.9074}Zn_{.0926}Te$	Zn-Te	2.7119	0.0077	4.4285	0.0098
	Cd-Te	2.8657	0.0077	4.6797	0.0098
$Cd_{.9074}Zn_{.0926}Te_{.9815}Se_{.0185}$	Zn-Te	2.7123	0.0114	4.4291	0.0145
	Cd-Te	2.8651	0.0094	4.6786	0.0120
	Cd-Se	2.7267	0.0020	4.4527	0.0026

Standard deviation of Te bond lengths/interplanar spacing increases upon addition of Se. The alloy disorder was also found to be more effective with increasing Se concentration.

* Special Quasi-random Structure

Does lattice disorder affect the device performance?

No adverse effect was observed on device performance due to the presence of lattice disorder in CZTS.

- Higher charge collection efficiency was reported for CdTeSe with respect to CdTe/CZT (Fiederle et al.).
- Minority carrier lifetime reported to enhance many-fold for CdTeSe compared to CdTe due to reduced mid-bandgap defect states (Guo et al.).
- Very high resolution CZTS based Frisch grid reported to have the lowest energy resolution of ~0.77% at 662 keV. (Roy et al.).
- CdZnTeSe reported to perform better than CdZnTe under high flux for medical imaging applications. (Yakimov et al.).

1. M. Fiederle et al., J. Cryst. Growth 138 (1994) 529.
2. J. Guo et al., Appl. Phys. Lett. 115 (2019) 153902.
3. U. N. Roy et al., Scientific Rep.11 (2021) 10338.
4. A. Yakimov et al., Proc. SPIE 11114 (2019) 111141N.

Analysis of THM-grown CdZnTeSe shows higher levels for some impurities

6N purity CZT (raw material)

CZT 1

Element	Concentration [ppb at]
Cr	<3
Fe	34
Ni	<5
Cu	<15
Sn	<45
Pb	<2

CZT 2

Element	Concentration [ppb at]
Cr	<3
Fe	110
Ni	<4
Cu	<8
Sn	<30
Pb	<2

6N purity $Cd_{0.9}Zn_{0.1}Te_{0.98}Se_{0.02}$ grown by THM

Ingot #1

Element	Concentration [ppb at]
Cr	<20
Fe	42
Ni	<4
Cu	22
Sn	<100
Pb	10

Ingot #2

Element	Concentration [ppb at]
Cr	36
Fe	42
Ni	16
Cu	<4
Sn	<100
Pb	11

The impurities present in THM-grown $Cd_{0.9}Zn_{0.1}Te_{0.98}Se_{0.02}$ are 3-8 times higher compared to CZT raw material.

For commercial THM-grown CZT (ppb at):
Cr-ND, Fe-22, Ni-ND, Cu-ND

ND- Not Detected

J.J. McCoy et al., J. Electronic Materials 48, 4226 (2019).

Most of the impurities are expected to originate from the CdSe raw material.

Much room to improve the detector performance of CdZnTeSe

We strongly believe the detector performance can further be improved by using higher purity CdSe starting material.

Acknowledgement

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Thank you for your kind attention!