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CdTexSe1-x: A potential candidate for room-temperature radiation detector applications

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$CdTe_xSe_{1-x}$: A potential candidate for room-temperature radiation detector applications

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Introduction: Why do we search for an alternate material to CdZnTe?

CdZnTe suffers from three major detrimental defects that limit the charge transport properties and their non-uniformity

- **Non-unity segregation coefficient of Zn (~1.35) in CdTe matrix results in a compositional gradient along the ingot, hence limiting the thickness of detectors and lowering the production yield of useful detector material.**
- **Presence of a large concentration of sub-grain boundaries and their networks. These defects also lower the μτ product, as well as impose a spatial non-uniformity in the charge transport properties that reduces the energy resolution of the device.**
- **Presence of large amounts of Te inclusions/precipitates (on the order of 2-8x10^5 /cm^3 with a diameter larger than ~2 μm). These secondary phases have a deleterious effect on the μτ product and create non-uniformities in the charge transport properties and electric field inside the devices that cannot be corrected electronically.**

Comparison of CdTeSe and CdZnTe

Material / Properties	Segregation coefficient	Te-inclusions /precipitations (/cm^3)	Sub-grain boundary network	Charge transport	Etchpit density (/cm^2)
CdZnTe	~ 1.35 for Zn	2-8x10^5	High density	High μτ	~ 10^5
CdTeSe	~0.98 for Se	--------	Almost free (solution hardening effect of Se is more effective)	Effective electron mass is lower than CdTe: higher μτ is expected	~10^4

From the stand point of segregation coefficient and defect densities, CdTeSe has tremendous potential for room-temperature semiconductor detectors as an lower-cost replacement for CZT.

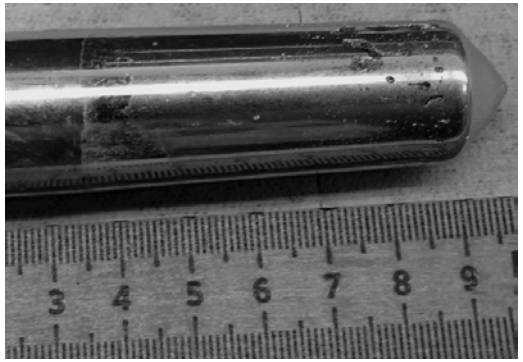
Growth of CdTeSe

- 6N purity CdSe and CdTe were taken for the initial charge
- The compound was synthesized and purified by sublimation
- In was used as the compensating dopant for achieving high resistivity
- Ampoule diameter used was 22-mm ID

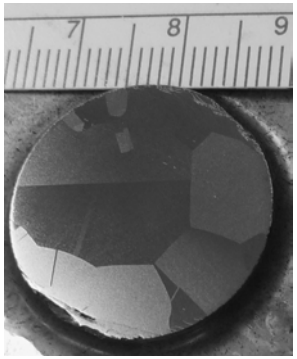
Various crystals were grown by the vertical Bridgman as well as THM technique with 5, 7 and 10% Se composition.

- For THM, the growth temperature and growth rate were 850 ^{0}C and ~3 mm/day, respectively.
- For Bridgman growth the growth rate used was ~3 cm/day

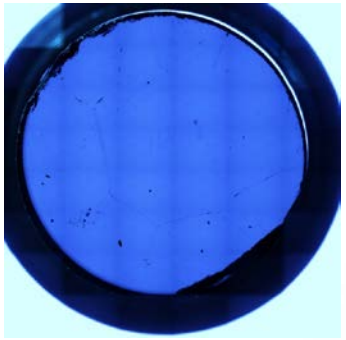
CTS ingots grown by Bridgman and THM



CTS ingot grown by THM

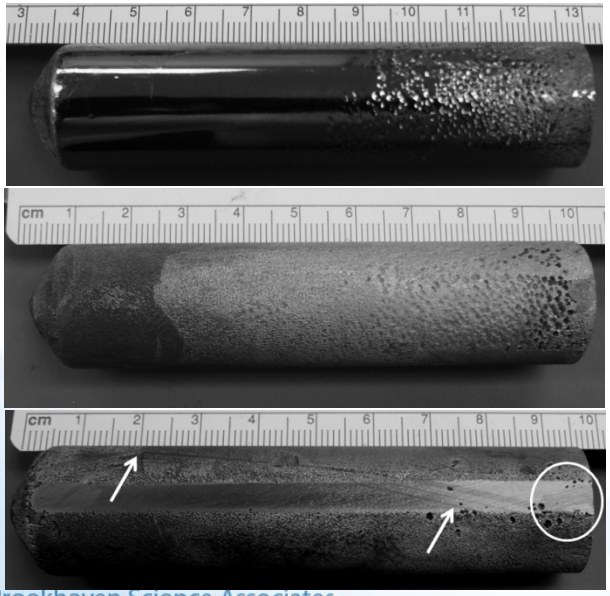


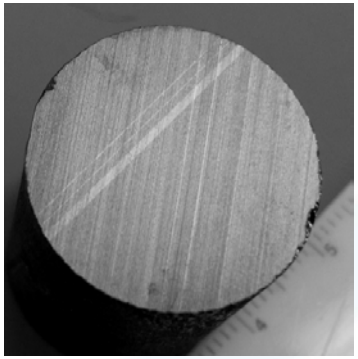
Lapped wafer



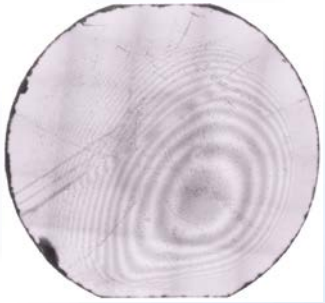
IR transmission image

Typical THM grown CTS ingot





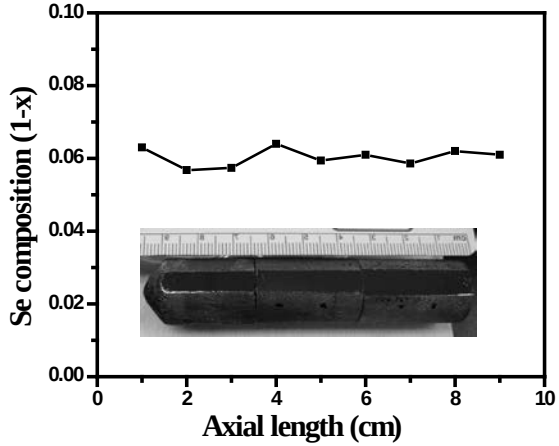
Lapped wafer



IR transmission image

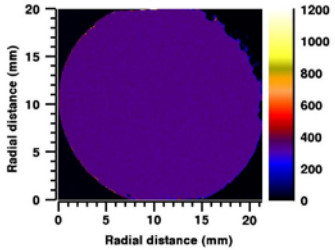
Typical Bridgman grown CTS ingot

Compositional homogeneity (Bridgman grown CTS)

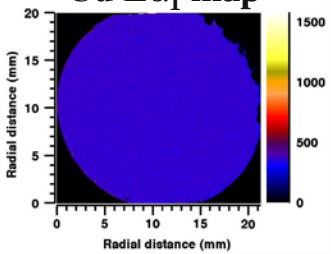


Se composition measured by EDS for typical CTS ingot with nominal concentration of 5% Se grown by the Bridgman technique

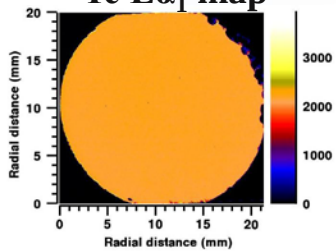
Very high compositional homogeneity for the CTS ingots, similar radial compositional homogeneity was also observed for THM grown CTS ingots



Cd Lα_1 map



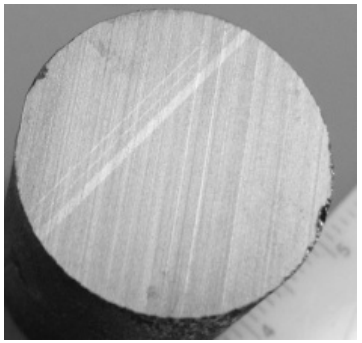
Te Lα_1 map



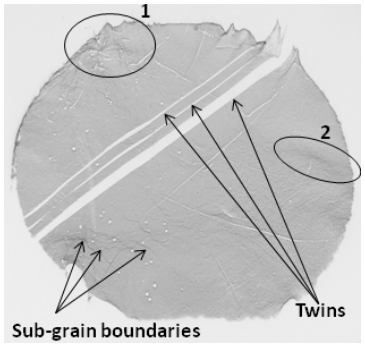
Se Kα map

X-ray fluorescence elemental map for Cd Lα_1, Te Lα_1 and Se Kα line of Bridgman grown $CdTe_{0.95}Se_{0.05}$ wafer

X-ray diffraction topography of CTS

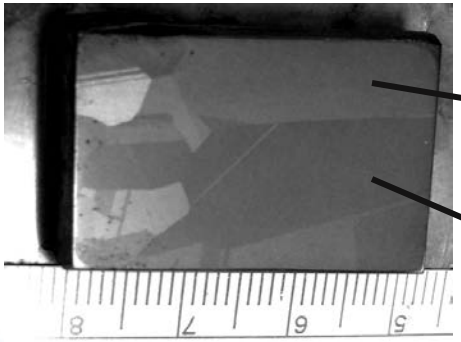


Lapped wafer

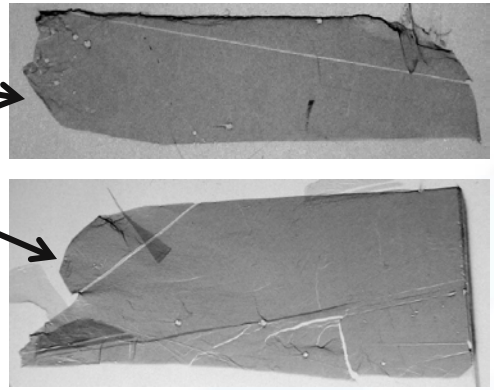


X-ray diffraction topography of the wafer

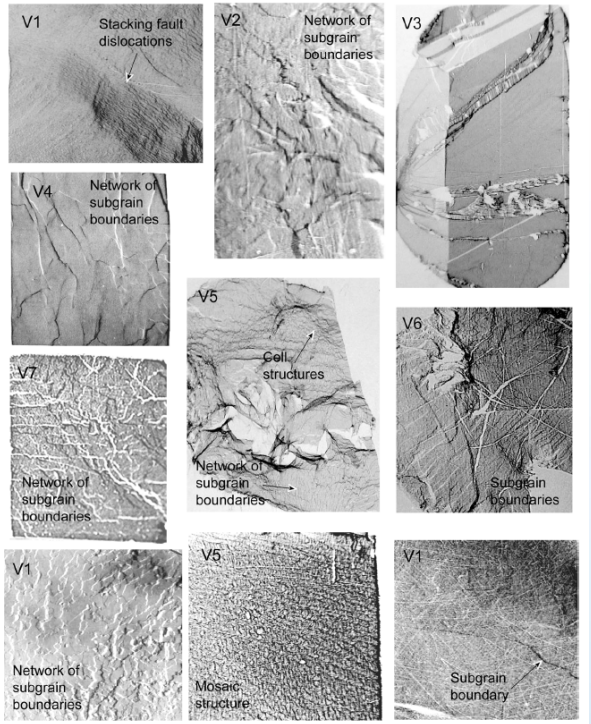
Bridgman grown $CdTe_{0.95}Se_{0.05}$ wafer



Lapped wafer along the length of the ingot



X-ray diffraction topography of the grains shown by the arrows



Diffraction topographs showing ~1 cm^2 areas of detector-grade CZT samples supplied by seven different vendors.

A. E. Bolotnikov et al. J. Cryst. Growth 379 (2013) 46.

THM grown $CdTe_{0.9}Se_{0.1}$ wafer cut along the length of the ingot

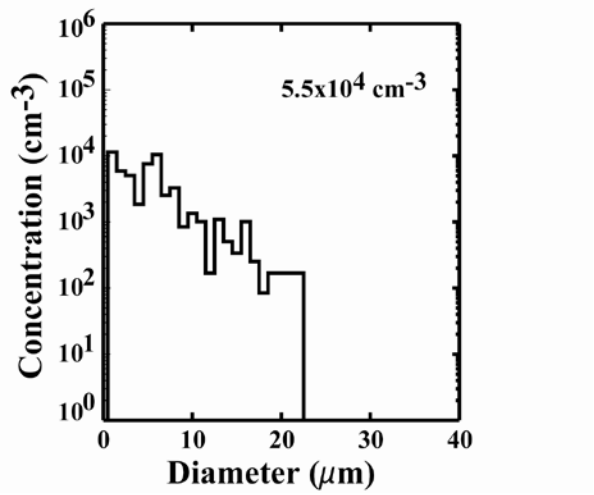
X-ray diffraction topography of CTS

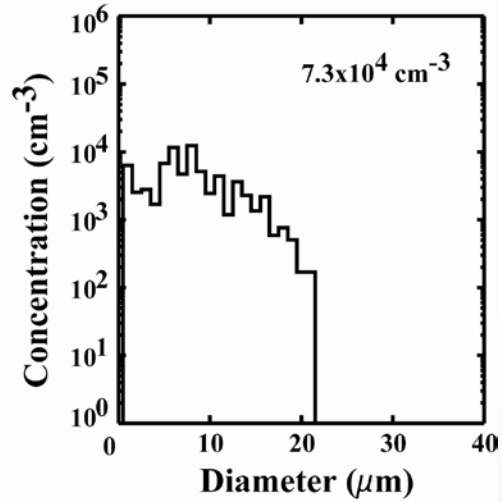
- **Highly reduced sub-grain boundary**
- **Almost stress-free ingot**
- **No sub-grain boundary network**
- **No mosaic structure**
- **No cell structures**

Our etch-pit study also showed absence of sub-grain boundary network

Se in CTS is indeed is very effective in solid solution hardening in arresting the formation of sub-grain boundary networks compared to Zn in CZT.

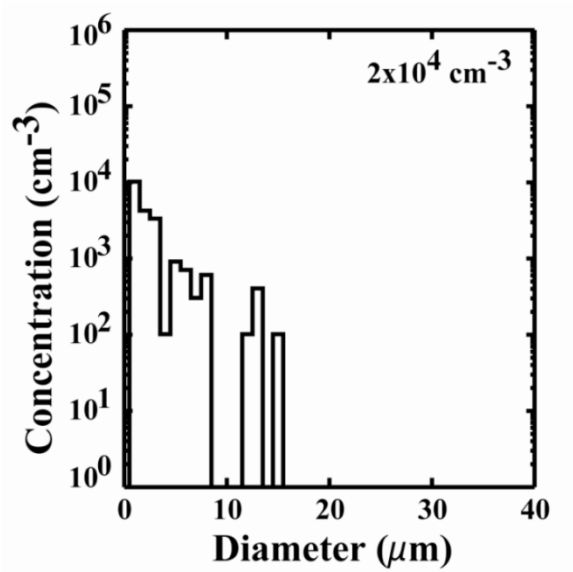
Concentration and size distribution of secondary phase in BM grown CTS

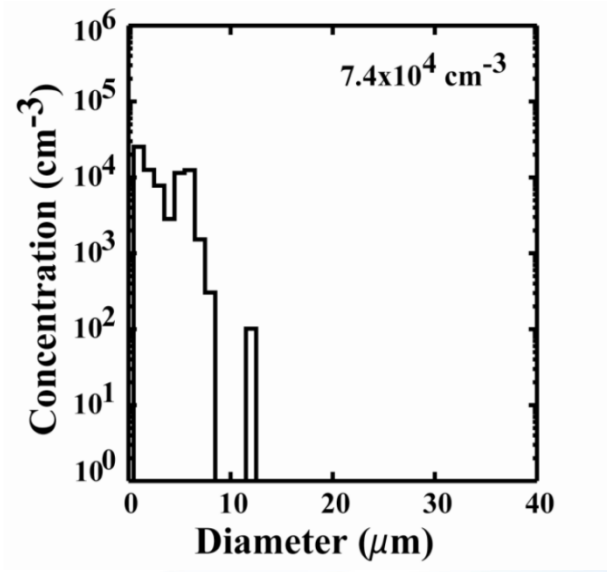




Size distribution of secondary phases over 1.1x1.5x8mm^3 of Bridgman grown $CdTe_{0.9}Se_{0.1}$. Total average concentration is 3-7 x10^4 cm^{-3}.

Concentration and size distribution of secondary phases in THM as-grown CTS

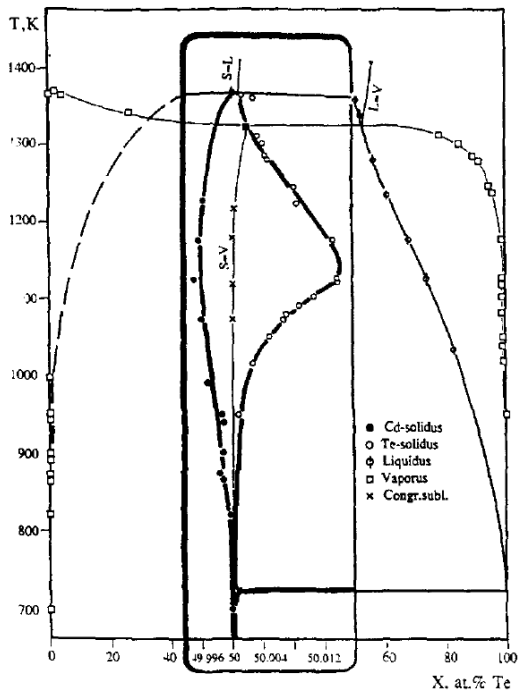




Size distribution of secondary phases over 1.1x1.5x6.65mm^3 of as-grown $CdTe_{0.9}Se_{0.1}$ by THM. Total average concentration is 2-7 x10^4 cm^{-3}.

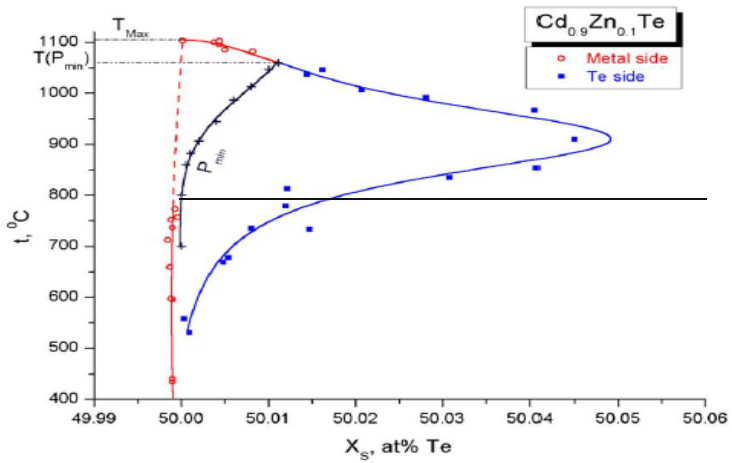
Average concentration of secondary phases is roughly one order of magnitude lass than conventional CZT

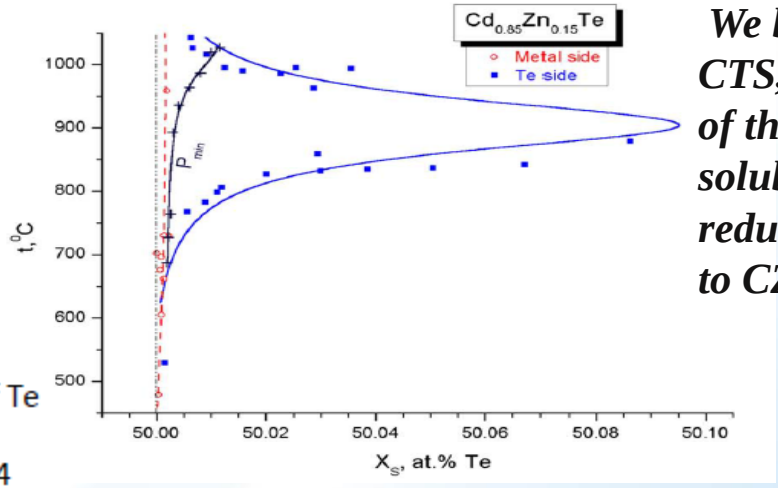
Phase diagram of CdTe and CZT



T-X projection of CdTe solidus

Taken from Greenberg, J. Cryst. Growth 161 (1996) 1.





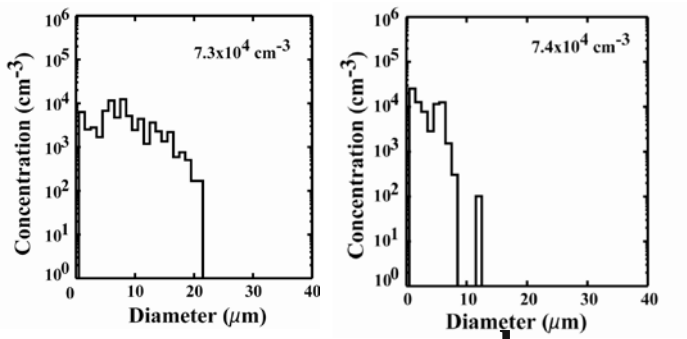
We believe that in CTS, the bulging of the retrograde solubility curve is reduced compared to CZT

Material	Temperature (°C)	x_s at% of Te
CdTe	~ 900	~ 50.014
$Cd_{0.9}Zn_{0.1}Te$	~ 900	~ 50.052
$Cd_{0.85}Zn_{0.15}Te$	~ 900	~ 50.1

T-X projection of $Cd_{0.9}Zn_{0.1}Te$ and $Cd_{0.85}Zn_{0.15}Te$ solidus

Taken from Greenberg and Guskov, J. Cryst. Growth 289 (2006) 552.

Theoretical prediction of Te inclusions on device performance and our measured values

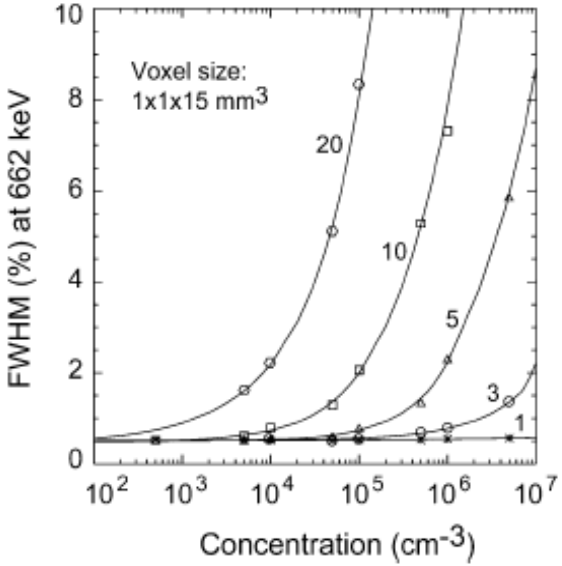


a b

Concentration and size distribution of secondary phase in CTS (10% Se) grown by a) Bridgman and b) THM technique

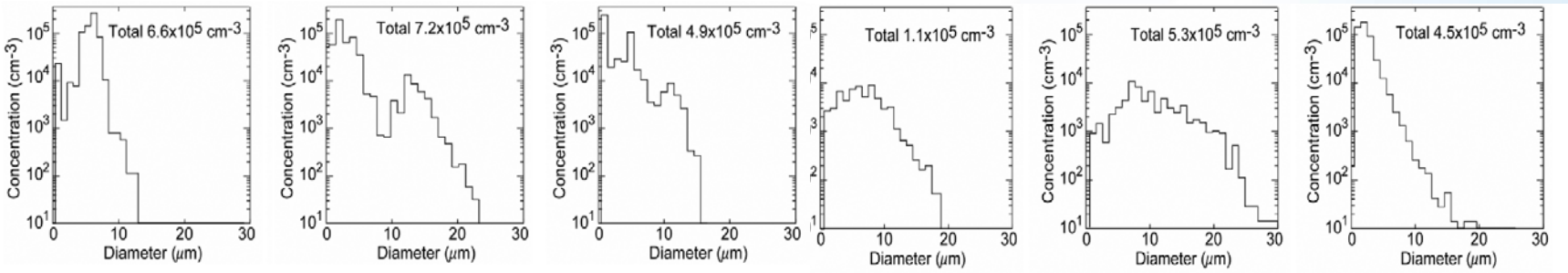
Important Point: The distribution (size and concentration) of inclusions in CTS may have no adverse effect on detector performance.

The average concentration is roughly one order of magnitude lower than commercial CZT available today.



FWHM (%) at 662 keV, of the cumulative effect of Te inclusions to energy resolution (after bi-parametric correction) versus their concentration calculated for 1-, 3-, 5-, 10-, and 20- μm inclusions. The detector's length is 15 mm.

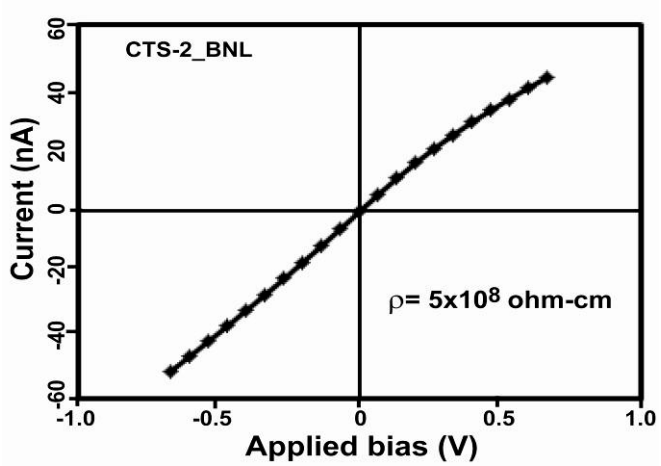
A. E. Bolotnikov et al., IEEE Trans. on Nucl. Science 54, 821 (2007).



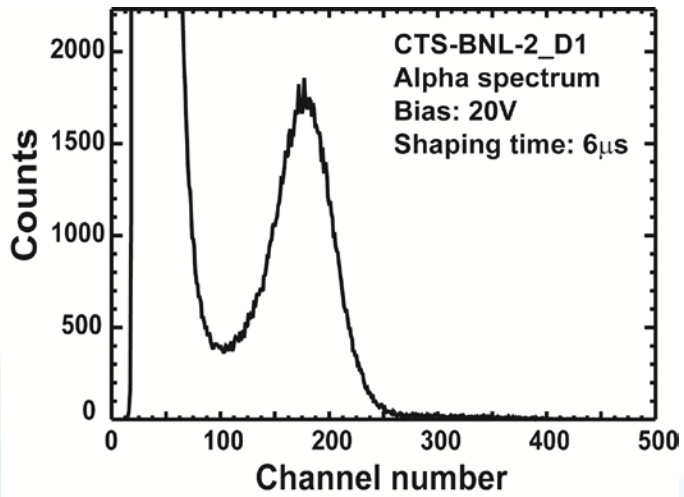
Size distribution and concentration of secondary phases for standard CZT from different vendors.

A. E. Bolotnikov et al., IEEE Trans. on Nucl. Science 57 (2010) 910.

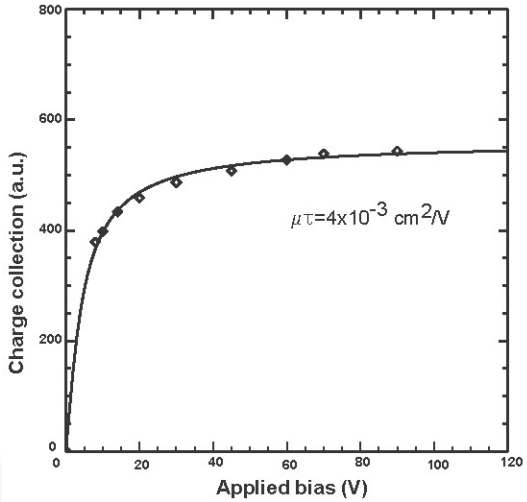
Charge transport and device characteristics of THM grown CTS



I-V characteristics of CTS (10%Se) as-grown sample

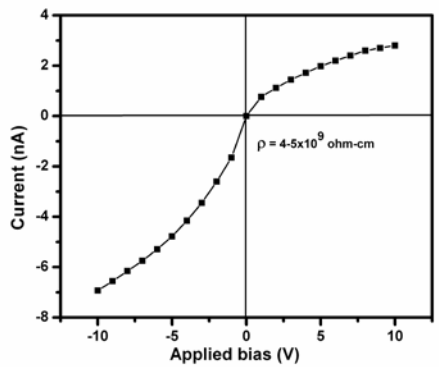


Alpha response from an Am-241 source for CTS (10% Se) sample grown at BNL. Sample size: 5.5x5.5x2 mm^3

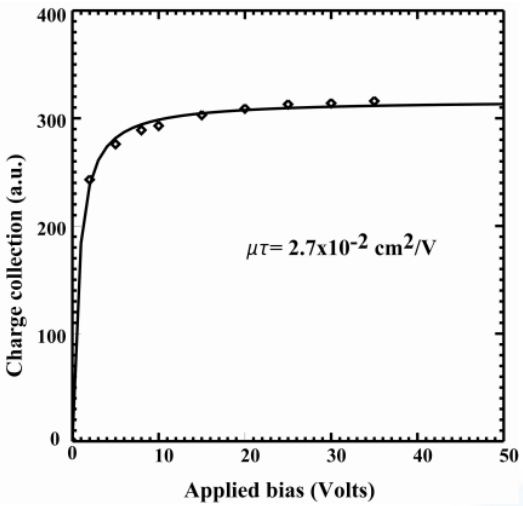


Mobility-lifetime plot for CTS (10%Se) sample
Sample size: 5.5x5.5x2 mm^3

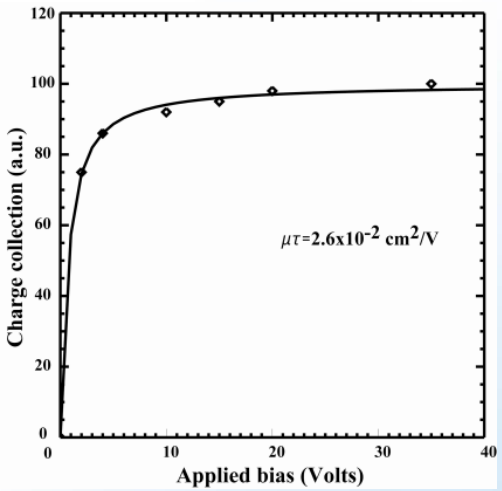
Charge transport and device characteristics of THM grown CTS



I-V characteristics of CTS (5%) as-grown sample



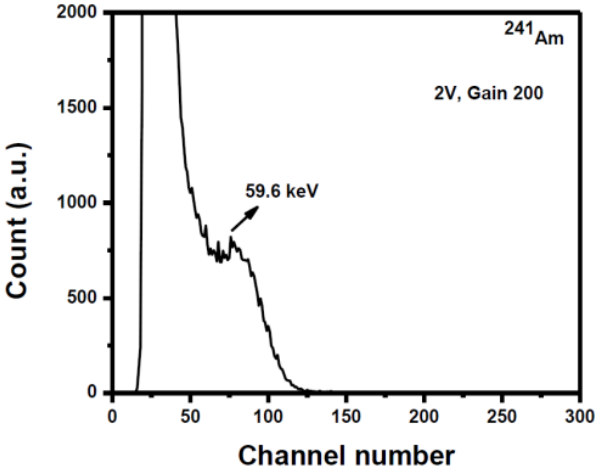
a

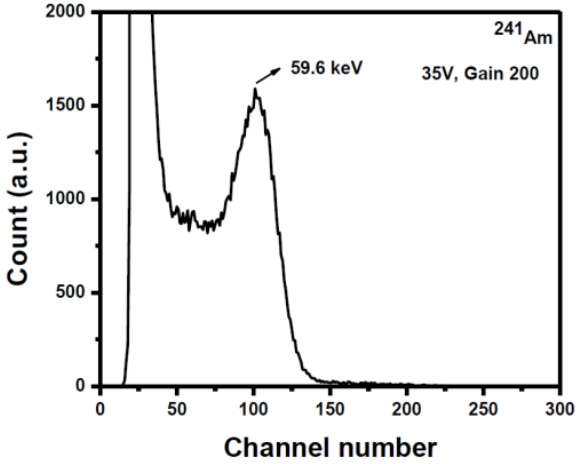


b

Mobility-lifetime plot for CTS (5%) sample. a) For alpha-particle line, and b) for gamma-ray line of ^{241}Am source. Sample size: 6.5x6x1.8 mm^3

Charge transport and device characteristics of THM-grown CTS (continued)





Room temperature detector response of the same planar detector for the gamma lines of ^{241}Am source. The resistivity of the sample is 4-5x10^{9} ohm-cm.

Comparison of CdTeSe and CdZnTe: Present status

Material / Properties	Segregation coefficient	Te-inclusions /precipitations (/cm^3)	Sub-grain boundary network	Charge transport	Etchpit density (/cm^2)	Resistivity (ohm-cm)
CdZnTe	~ 1.35 for Zn	2-8x10^5	High density	High μτ	~ 10^5	2-4x10^{10}
CdTeSe	~0.98 for Se	--------	Almost free (solution hardening effect of Se is more effective)	Effective electron mass is lower than CdTe: higher μτ is expected	~10^4	~ 7x10^9
CdTeSe (our result)	≈ 1	2-7x10^4	Almost free from sub-grain boundary network	Fairly high μτ	10^4 -10^5	~ 4-5x10^9

CdTeSe has tremendous potential for room-temperature semiconductor detectors as a lower-cost replacement for CZT, however more effort to increase the resistivity is required.

Summary of observation

- **The effective segregation coefficient of Se is ≈1: very high compositional uniformity throughout the ingot.**
- **Se solid solution hardening is efficient in arresting the sub-grain boundary and their network.**
- **Se has a strong effect in reducing the dislocation density (etch-pit density), which might be due to effective solid solution hardening.**
- **Se is possibly responsible in reducing the bulging of the retrograde solidus line near stoichiometry and effective in reducing the concentration of secondary phases by an order of magnitude compared to conventional CZT.**
- **Obtained fairly high μτ value for as-grown CTS by THM.**
- **Effort is being made to increase the resistivity of as-grown CTS.**

Very high compositional uniformity, absence of sub-grain boundary network, and one order of magnitude less concentration of secondary phases are very encouraging for the future of CTS for both substrate and detector applications.

Acknowledgement

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

Thank you for your attention!