#### Contract No:

This document was prepared in conjunction with work accomplished under Contract No. 89303321CEM000080 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

#### Disclaimer:

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U.S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied:

- 1) warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or
- 2) representation that such use or results of such use would not infringe privately owned rights; or
- 3) endorsement or recommendation of any specifically identified commercial product, process, or service.

Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.

#### SRNL-STI-2021-00584

# Comparative Studies of CdZnTe, CdMnTe, and CdZnTeSe Materials for Room-Temperature Nuclear Detection Applications

STEPHEN U. EGARIEVWE,<sup>1,2</sup> STEPHAN D. SOTO,<sup>1</sup> SIMEON W. SYKES,<sup>1</sup> LESLIE J. FULLER,<sup>1</sup> QUENTIN J. ALSBROOKS,<sup>1</sup> MOHAMMAD A. ALIM,<sup>1</sup> UTPAL N. ROY,<sup>2,3</sup> EZEKIEL O. AGBALAGBA,<sup>4</sup> MEBOUGNA L. DRABO,<sup>1</sup> AND RALPH B. JAMES<sup>3</sup>

> <sup>1</sup>Alabama A&M University, Huntsville, Alabama, USA <sup>2</sup>Brookhaven National Laboratory, Upton, New York, USA <sup>3</sup>Savannah River National Laboratory, Aiken, South Carolina, USA <sup>4</sup>Federal University of Petroleum Resources, Effurun, Delta State, Nigeria



VITUAL 2021 IEEE NUCLEAR SCIENCE SYMPOSIUM AND MEDICAL IMAGING CONFERENCE

28<sup>th</sup> International Symposium on Room-Temperature Semiconductor Detectors



16 - 23 October 2021











#### VITUAL 2021 IEEE NUCLEAR SCIENCE SYMPOSIUM AND MEDICAL IMAGING CONFERENCE

### **Presentation** Outline

• Goals: Review of Our Recent Studies on CdTe-Based Detectors

16 - 23 October 2021

- Operation of CdTe-Based Detectors
- Device Engineering Design Constraints
- Constant-Temperature Annealing of CdMnTe Detector
- Temperature-Gradient Annealing of CdMnTe Detector
- XPS Study of Passivated CdZnTe Surfaces
- Planar Detector Fabrication
- Electrical Instrumentation for Current-Voltage Measurement
- Surface Chemical Treatments of CdZnTe Detector
- Chemical Passivation of CZTS
- Summary







- Cadmium telluride (CdTe) and its ternary and quaternary compound have found applications in the development of X-ray and gamma-ray detectors used in medical imaging and in the detection of radiological and nuclear threats.
  - Examples: CdZnTe (CZT), CdMnTe (CMT), and CdZnTeSe (CZTS).
- These detectors can operate at room temperature without cryogenic cooling.
- The goal of this presentation is to review our recent studies of studies of CdTe-based detectors and the effects of chemical treatments on their electrical properties and radiation detection performances.
- The properties to be studied include detector surface species, resistivity, and energy resolution.
- Surface treatment studies include chemo-mechanical polishing and passivation using KOH and NH<sub>4</sub>F solutions.



VITUAL 2021 IEEE NUCLEAR SCIENCE SYMPOSIUM AND MEDICAL IMAGING CONFERENCE

MIC 28th International Symposium on Room-Temperature Semiconductor Detectors 16 - 23 October 2021



### Operation of CdTe-Based Detectors

©ieee NPSS



- This creates electrical signal.
- The signal is amplified before the MCA.
- MCA displays the spectrum on the computer screen.

Meaning of Symbols: e<sup>-</sup>: Electron (o). h<sup>+</sup>: Hole (•). V: Applied voltage. E: Electric field direction. Amp: Amplifier. MCA: Multichannel analyzer.





# Device Engineering Design Constraints

- High resistivity >  $10^8 \Omega$ -cm.
  - Implies low current. High current could damage the pre-amplifier and amplifier.
- Low surface current (nano-amps). This is required to reduce electronic noise in the generated signal.
- Chemical passivation must result in reduced surface current.
- High charge-carrier mobility-lifetime product.
- Free of defects that affect charge collection and uniformity.
- Low production cost.



VIFTUAL 2021 IEEE NUCLEAR SCIENCE SYMPOSIUM AND MEDICAL IMAGING CONFERENCE

ternational Symposium on Room-Temperature Semiconductor Detectors



# Constant-Temperature Annealing of CdMnTe Detector

- Thermal annealing is used to remove defects caused by Te inclusions in the detector wafer.
- Te inclusions and related defects limit the detector's ability to detect X-rays and gamma-rays.
- <u>Process</u>:
  - The detector wafer is placed together with Cd in a quartz ampoule and sealed in vacuum using a vacuum pump.
  - The ampoule is placed in the furnace region at 720°C for 12 hours.
  - Cd reacts with the excess Te to form a crystalline structure in the wafer, thus reducing the Te inclusions and related defects.



VIRTUAL 2021 IEEE NUCLEAR SCIENCE SYMPOSIUM AND MEDICAL IMAGING CONFERENCE

ISS MIC 28th International Symposium on Room-Temperature Semiconductor Detectors 16 - 23 October 2021



#### Constant-Temperature Annealing of CdMnTe Detector





(a) Before annealing

(b) After annealing

Infrared images (256  $\mu$ m x 367  $\mu$ m) of the same region of the CdMnTe wafer before and after annealing in Cd vapor.



Three-zone annealing furnace (shown on left-side) and vacuum pump (shown on right-side) for removing air when sealing CdMnTe wafers in ampoules.



VIRCUAL 2021 IEEE NUCLEAR SCIENCE SYMPOSIUM AND MEDICAL IMAGING CONFERENCE

ISS MIC 28th International Symposium on Room-Temperature Semiconductor Detectors 16 - 23 October 2021



### Constant-Temperature Annealing of CdMnTe Detector



(a) Before annealing.

(b) After annealing.

Te inclusions eliminated from a CdMnTe wafer after being annealed at  $570^{\circ}$ C for 26 hours in Cd vapor.

Source: S. U. Egarievwe, G. Yang, A. A. Egarievwe, A. A., I. O. Okwechime, J. Gray, Z. M. Hales, . . . James, R. B. (2015). Post-growth annealing of Bridgman-grown CdZnTe and CdMnTe crystals for room-temperature nuclear radiation detectors. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 784, 51-55. doi: 10.1016/j.nima.2015.02.006.



# Temperature-Gradient Annealing of CdMnTe Detector



Furnace temperature profile for temperature-gradient annealing.



VITUAL 2021 IEEE NUCLEAR SCIENCE SYMPOSIUM AND MEDICAL IMAGING CONFERENCE

5 MIC 28th International Symposium on Room-Temperature Semiconductor Detectors 16 - 23 October 2021

# Temperature-Gradient Annealing of CdMnTe Detector

Size reduction and elimination of Te inclusions in CdMnTe wafer after it was annealed at  $730^{\circ}$  C for 18 hours at  $18^{\circ}$ C/cm temperature gradient.









(b) After annealing.

10

**Source:** S. U. Egarievwe, G. Yang, A. A. Egarievwe, A. A., I. O. Okwechime, J. Gray, Z. M. Hales, . . . James, R. B. (2015). Post-growth annealing of Bridgman-grown CdZnTe and CdMnTe crystals for room-temperature nuclear radiation detectors. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 784, 51-55. doi: 10.1016/j.nima.2015.02.006.



VIETUAL 2021 IEEE NUCLEAR SCIENCE SYMPOSIUM AND MEDICAL IMAGING CONFERENCE

# XPS Study of Passivated CdZnTe Surfaces

16 - 23 October 2021

- Passivation helps to reduce the surface current and improve shelf-life of the device.
- <u>Process</u>: The detector wafer is dipped in the passivation solution and dried with compressed air.
- <u>Chemicals used in Our Studies</u>:
  - A mixture of 0.1-g potassium hydroxide (KOH) and 10-ml 30% hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) aqueous solution.
  - A mixture of ammonium fluoride and 10% hydrogen peroxide in distilled water ( $NH_4F + H_2O_2 + H_2O$ ).



High-performance XPS Surface Analysis System by Thermo Fisher

11



# XPS Spectra of CdZnTe Surfaces: Cadmium (Cd) Peaks





# XPS Spectra of CdZnTe Surfaces: Te and TeO<sub>2</sub> Peaks



<u>Source</u>: S. U. Egarievwe, A. Hossain, I. Okwechime, A. Egarievwe, D. Jones, Dominique U. Roy, and R. B. James, (2016). Effects of Chemical Treatments on CdZnTe X-Ray and Gamma-Ray Detectors. IEEE Transactions on Nuclear Science. 63. 1091-1098. 10.1109/TNS.2016.2527779.

13



virtual 2021 IEEE NUCLEAR SCIENCE SYMPOSIUM AND MEDICAL IMAGING CONFERENCE

SS MIC 28th International Symposium on Room-Temperature Semiconductor Detectors 16 - 23 October 2021



14

# XPS Spectra of CdZnTe Surfaces: Te and TeO<sub>2</sub> Peaks

Process	Te3d <sub>3/2</sub> O <sub>2</sub> /Te3d <sub>3/2</sub>	Te3d <sub>5/2</sub> O <sub>2</sub> /Te3d <sub>5/2</sub>
Before Passivation	0.49	0.46
After passivation in KOH.	1.25	1.19
After passivation in NH <sub>4</sub> F.	4.90	5.34

- $TeO_2$  peaks increased after passivation. This shows the formation more  $TeO_2$ .
- Passivation in NH4F produces more than passivation in KOH.
- Thus,  $NH_4F$  is better passivation chemical than KOH.



VIFTUAL 2021 IEEE NUCLEAR SCIENCE SYMPOSIUM AND MEDICAL IMAGING CONFERENCE

onal Symposium on Room-Temperature Semiconductor Detectors



15

**Planar Detector Fabrication** 

16 - 23 October 202

- Involves four major steps.
  - <u>Wafer Cutting</u>:
    - Cuts detector wafer to desired dimensions using a special cutting machine.
  - <u>Mechanical Polishing</u>:
    - Remove surface defects and residue using silicon-carbide abrasive papers.
  - Chemical Etching and Drying:
    - Used to remove any fine residual damage caused by mechanical polishing.
  - Deposition of Electrical Contacts:
    - Use electroless contact deposition method to deposit electrical contact on two opposite planner surface of the wafer.



# Electrical Instrumentation for Current-Voltage Measurement

Keithley Picoammeter/Voltage Source



Aluminum box where sample is mounted

16

Current-Voltage measurement system at Brookhaven National Laboratory.



VITUAL 2021 IEEE NUCLEAR SCIENCE SYMPOSIUM AND MEDICAL IMAGING CONFERENCE

28<sup>th</sup> International Symposium on Room-Temperature Semiconductor Detectors

Alabama A&M UNIVERSITY



17

# Surface Chemical Treatments of CdZnTe Detector

S NPSS

16 - 23 October 2021



Current-voltage plots for different surface chemical treatments of CdZnTe.



# Chemical Passivation of CZTS



Current-Voltage plots that show passivation results after different days.

18

**Source**: S. U. Egarievwe, U. N. Roy, C. A. Goree, B. A. Harrison, J. Jones, and R. B. James, (2019). Ammonium fluoride passivation of CdZnTeSe sensors for applications in nuclear detection and medical imaging. Sensors, 19(15), 3271.



# Chemical Passivation of CZTS



Voltage regions with reduced current 28 days after passivation.

19



# Chemical Passivation of CZTS

CZTS detector response from a <sup>241</sup>Am source at -200V before passivation and 28 days after passivation.







Thank You



- Thermal annealing reduces the sizes and concentrations of Te inclusions.
- XPS studies of passivated samples showed that:
  - TeO<sub>2</sub> formation increased after passivation.
  - $NH_4F$  is better passivation chemical agent than KOH.
- Passivation in  $NH_4F$  improves energy resolution.

Acknowledgements: This work was supported in part by the National Science Foundation (NSF) Major Research Instrumentation (MRI) through award number 1726901; in part by U.S. Department of Energy (DOE), Office of Defense Nuclear Nonproliferation Research and Development, the DNN R&D (NA-22), and DOE NNSA MSIPP award number DE-NA0003980; in part by the U.S. Nuclear Regulatory Commission (NRC) through award 31310018M0035; in part by the U.S. Department of Homeland Security, Domestic Nuclear Detection Office through award number 2012-DN-077-ARI065-05; and in part by the NSF HBCU-UP Program through award number 1818732.