Nuclear Science and Security Consortium
Virtual Scholar Showcase 2020

Radiation Hardness Characterization of LKH-5 Scintillating Glass

June 3, 2020

Mairead Montague
University of Tennessee, Knoxville

NSSC
NUCLEAR SCIENCE and SECURITY CONSORTIUM

June 2 - 3, 2020

Introduction

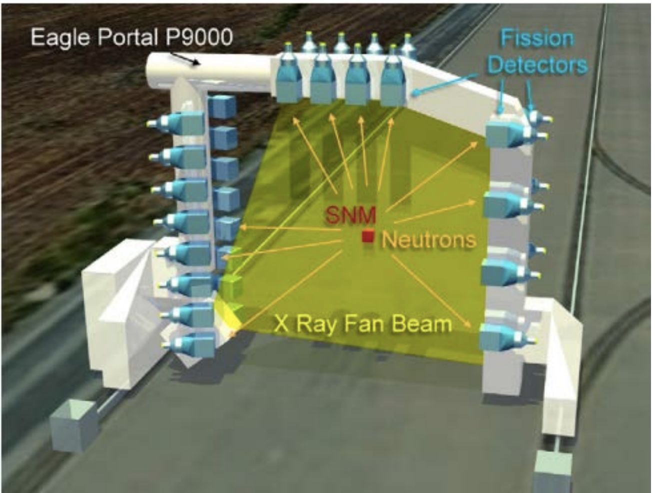
- **Academic Advisor: Jason Hayward**
  - Department of Nuclear Engineering
- **Lab Mentor: Paul Hausladen**
  - Radiation Detection and Imaging

Radiation Detection and Instrumentation

- Glass Scintillator Characterization for High Energy Cargo Scanning
- Fast Neutron Tomography for Spent Nuclear Fuel Verification

Cargo Scanning for Non-Proliferation

- **High Energy X-Ray Radiography**
  - Linear accelerator to image cargo containers
  - Search for SNM through detection of signatures of photonuclear reactions
  - Use both scintillator detectors and neutron detectors
- **Radiation Hardness**
  - Radiation damage: decrease light transmission by ionizing electrons within the scintillator and creating myriad defects within the crystal
  - Scanning system lifetime is dependent on scintillator lifetime
    - Scintillator lifetime dependent on radiation resistance
    - Important in order to make associated systems field-ready



[1]

Quantifying a scintillator's reductions in light transmission after irradiation results in information on its expected lifetime and therefore the expected lifetime of the scanning system.

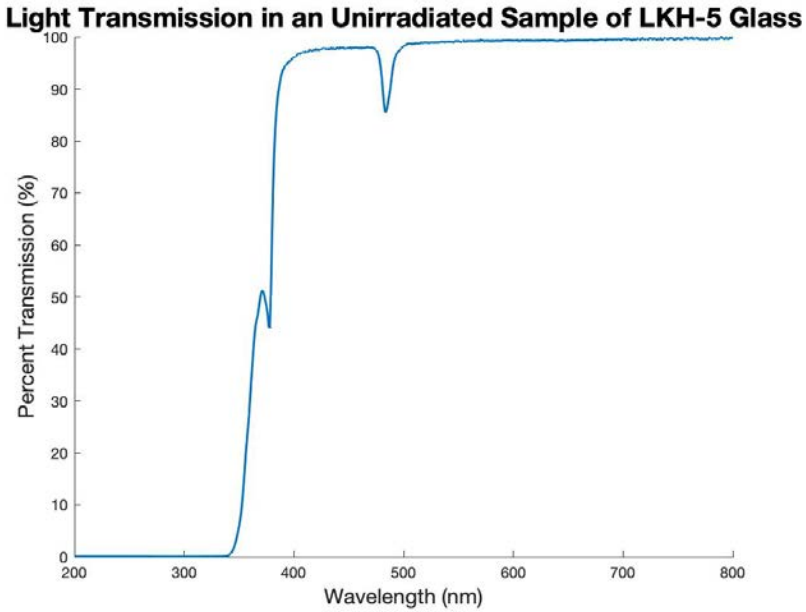
Scintillator Detectors for Cargo Scanning

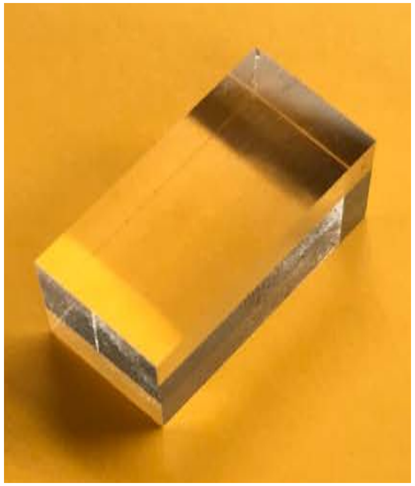
Cadmium Tungstate[1,2]	Cesium Iodide[3]	LKH-5 Glass[4,5,6]
High density, High spatial resolution, No afterglow	High optical yield	Low manufacturing cost; Well coupled with conventional a-Si TFT array sensitivity
Difficult to manufacture, Performance variability, Expensive	Hygroscopic, Visible afterglow	Radiation hardness must be investigated

1.J. Stevenson et. al, "Linac Based Photofission Inspection System Employing Novel Detection Concepts," NIMA 2011
2.M. Kobayashi et. al "Cadmium Tungstate Scintillators with Excellent Radiation Hardness and Low Background," NIMA 1994
3.C. L. Woody, et. al, "Radiation damage in undoped CsI and CsI(Tl),"IEEE Trans. 1992
4.Y.-H. Hu et. Al , "Characterizing a Novel Scintillating Glass for Application to Megavoltage Cone-beam Computed Tomography," Medical Physics, 2019
5.L. Ur et. al, "Terbium-Doped Heavy Metal Glasses for Green Luminescence," Journal of Rare Earths, 2011.
6.R. Weisfield, "Amorphous Silicon TFT X-Ray Image Sensors," IEEE 1999

Transmission Measurements

- **36 Samples: 10 mm x 10 mm x 20 mm**
  - All 6 sides polished
  - All close in starting color
  - Minimal aberrations from glass pouring
- **Transmission Measurements**
  - Spectrophotometer with wavelengths from 800 nm to 200 nm
  - Baseline 100% transmission with an unobstructed beam
  - Relative transmission with the sample in the beam path
  - Baseline 0% transmission with completely obstructed beam

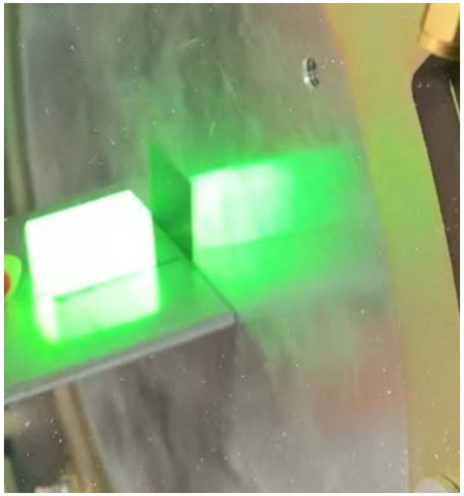




Experimental Procedure

- **Irradiation**
  - 9MV Linear Accelerator
  - Doses from 11 kRad–5 MRad
  - Dose Rate at 150 mm: 133.3 kRad/min
  - Placed 150 mm from target with the 10 mm x 10 mm face centered in the beam path
- **Activation**
  - Some activation seen after irradiation
  - Geant4 simulation to determine source of activation
  - Most likely: Neutron capture with 161Gd (t1/2 = 3.66 min)

Sample #	Actual time (s)	Dose (krads)
1, 2, 3	5	11.1
4, 5, 6	23	51.1
7, 8, 9	45	100.0
10, 11, 12	123	273.3
13, 14, 15	225	500.0
16, 17, 18	338	751.1
19, 20, 21	450	1000.0
22, 23, 24	900	2000.0
25, 26, 27	1350	3000.0
28, 29, 30	1800	4000.0
31, 32, 33	2250	5000.0
34, 35, 36	0	0.0



Sample glowing brightly within the beam path

Annealing Process

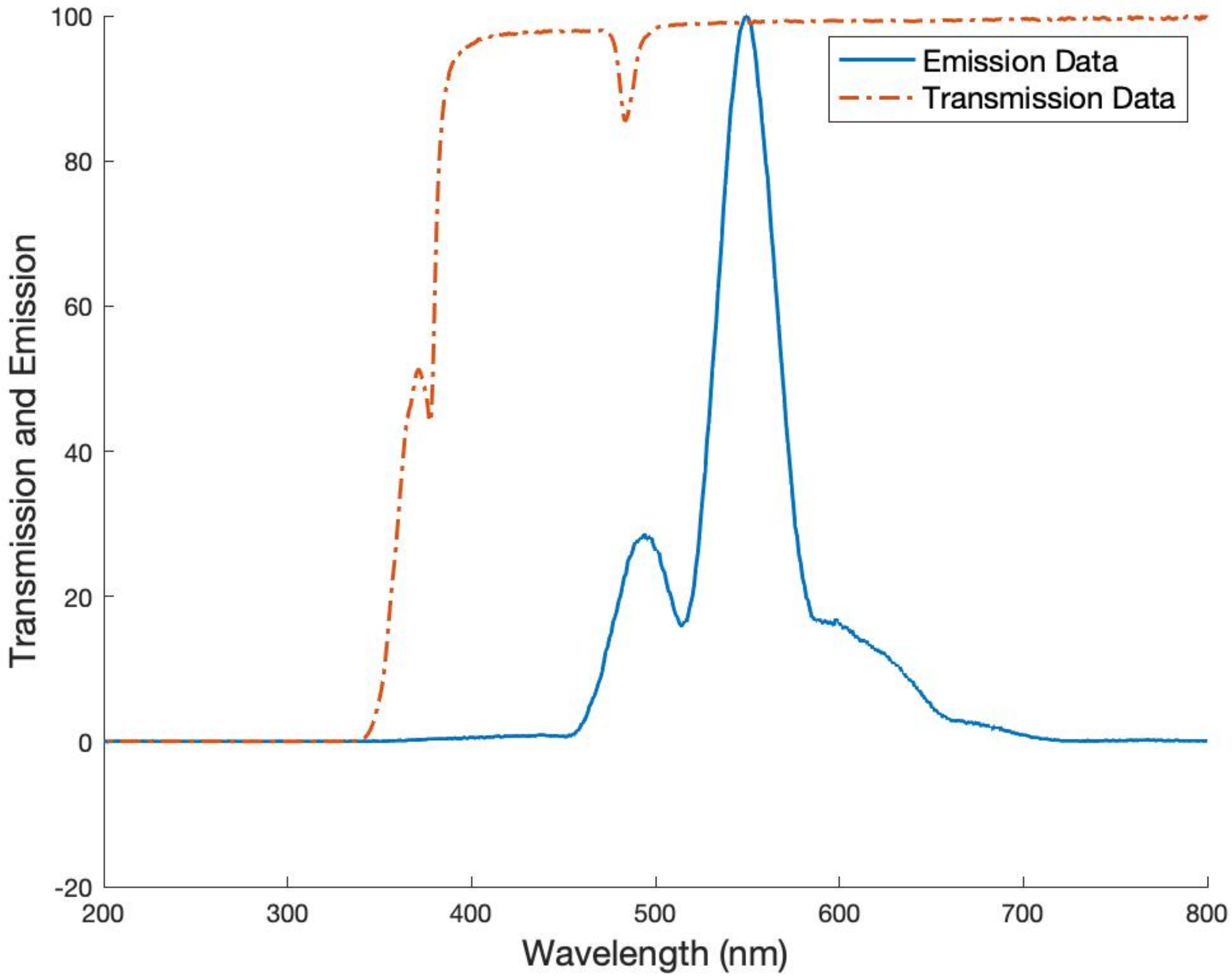
- **Annealing**
  - 12 samples annealed
    - 1 sample from each of the dose levels
  - Manufacturer recommended annealing method
    - Heat furnace to 585°C at a rate of 10°C per minute ~1hr
    - Hold at 585°C ~30 minutes.
    - Lower temperature to 550°C at a rate of 1° per minute ~35 min
    - Lower temperature to 450°C at a rate of 2° per minute ~50 min
    - Lowers to room temperature at a maximum rate of 11° per minute ~40 min

Before Irradiation

After Irradiation

After Annealing

Radioluminescence and Transmission



Irradiation and Annealing Results

400 nm Data

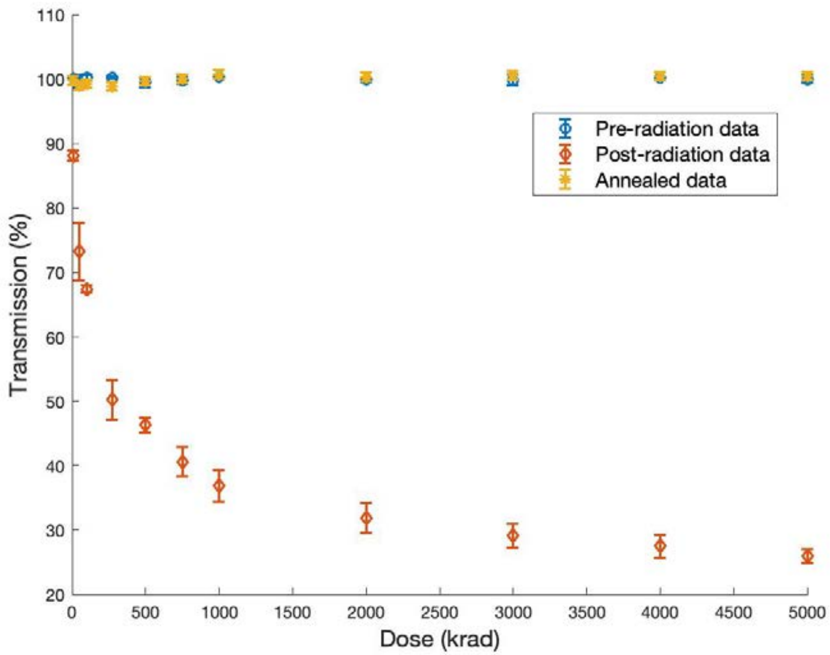
27% change in transmission at 5Mrad

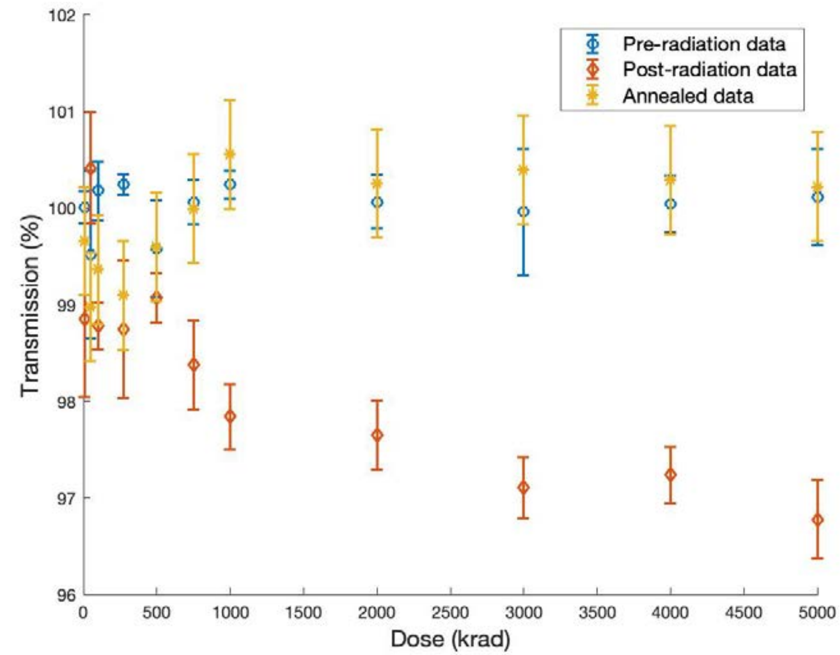
Demonstrates annealing process

550 nm Data

Less than 3% change in transmission

Demonstrates radiation hardness





LKH-5 Comparison to Conventional Scintillators

LKH-5 Pros

- Low Cost[4]
- Potential use in conventional a-Si TFT arrays[6]
- Comparable signal-to-noise to CWO through many inches of steel with increasing glass thickness

LKH-5 Drawbacks[4]

- Modest light yield
- Moderate density

Radiation Hardness

- **LKH-5 comparable to CWO** at their respective emission peaks and 5 MRad of dose[2]
- **LKH-5 better than CsI(Tl)** at 4.2 MRad of dose at 550 nm[3]

Scintillator Property	CdWO	CsI(Tl)	LKH-5
Density (g/cm^3)	7.9	4.5	3.8
Optical Yield (photons/MeV)	15,000	54,000	4,000
Cost ($\$/cm^3$)	\$35	\$9	~\$1
Emission Peak (nm)	470	550	550
Rad Hardness Discrepancy	2%	19%	3%

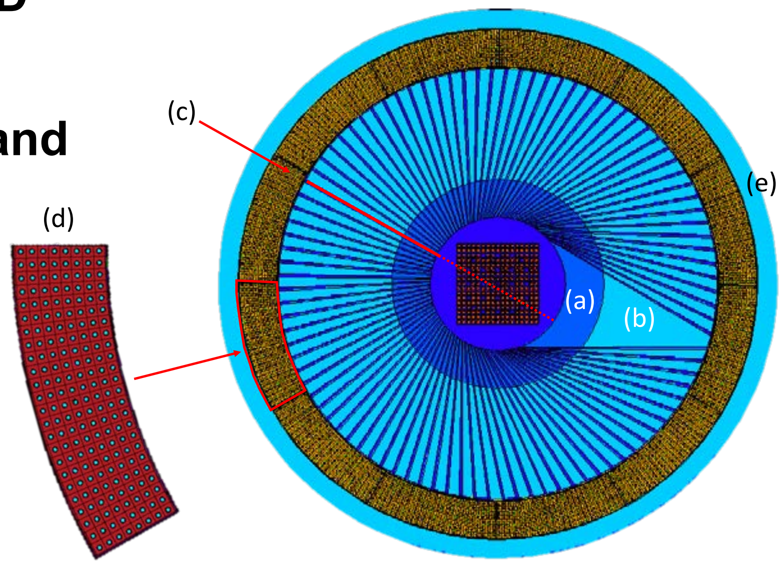
2. M. Kobayashi et. al "Cadmium Tungstate Scintillators with Excellent Radiation Hardness and Low Background," NIMA 1994
3. C. L. Woody, et. al, "Radiation damage in undoped CsI and CsI(Tl),"IEEE Trans. 1992
4. Y.-H. Hu et. Al , "Characterizing a Novel Scintillating Glass for Application to Megavoltage Cone-beam Computed Tomography," Medical Physics, 2019
6. R. Weisfield, "Amorphous Silicon TFT X-Ray Image Sensors," IEEE 1999

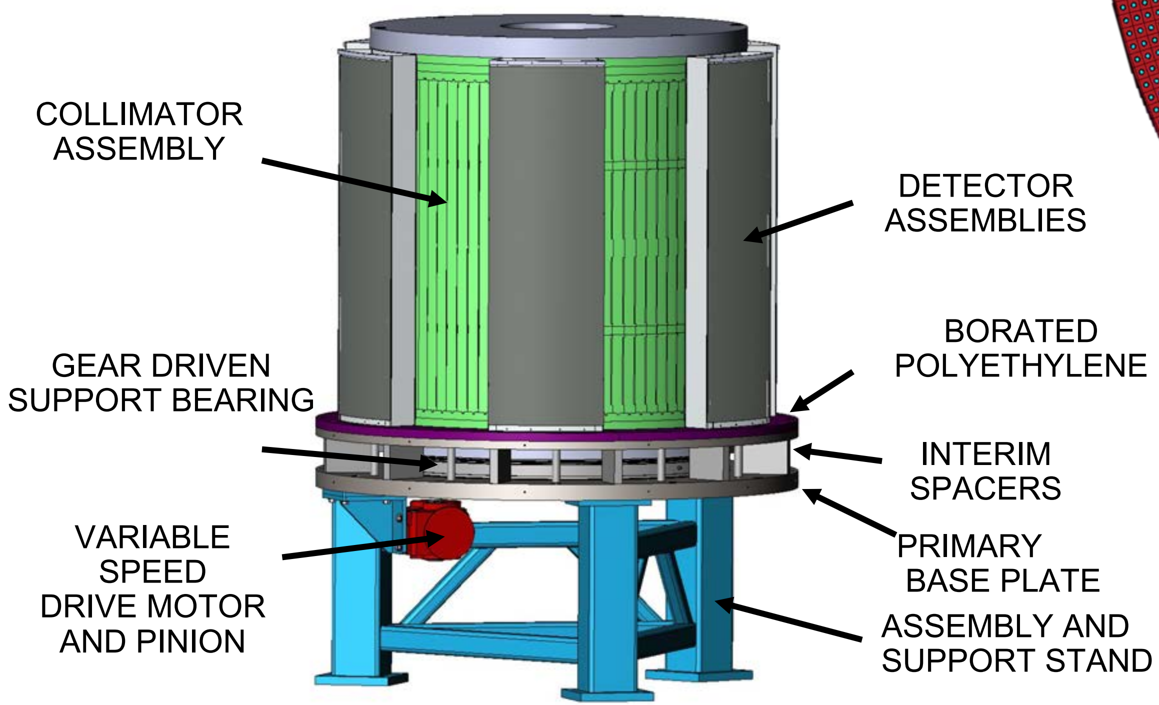
Conclusion

- **Light transmission at the emission peak decreases by 3% at 5 MRad of dose**
  - Indicates that radiation will have little effect on scintillator performance
  - Indicates long scintillator lifetime, which could lead to long cargo scanning system lifetime and low replacement detector costs
  - Glass annealing improves transmission and this process can prolong scintillator lifetimes
- **Appealing alternative if a detector design based upon LKH-5 glass can be consistently engineered to yield similar performance**
  - Comparable radiation hardness to CWO and low cost
  - Same emission peak as CsI(Tl) and lower cost
- **A journal paper on this work is currently under review, and plans for transition to commercial production by Varex Imaging are in progress**

Fast Neutron Tomography for Spent Fuel Verification

- **Modified parallel slit collimator for 3D imaging**
- **Used to verify spent fuel quantities and composition**





Collimator annulus
(a) inner stainless steel
(b) outer borated polyethylene ring
(c) 96 slits - define lines of response across the field of view.
Neutron detection
(d) 12 detector modules - 24 rows of 8 boron straws each
(e) 5-cm-thick ring of borated polyethylene shielding

NSSC Experience

Nuclear and Particle Physics
- 2017-2018

Radiation Detection and Instrumentation
- 2019-2020

2017: NSSC Undergrad Affiliate

5/2018: B.S. Nuclear Eng, UC Berkeley

6/2018: ORAU Summer Fellowship

5/2019: ORNL Research

12/2019: M.S. Nuclear Eng, UTK

2020: NSSC Fellow

Acknowledgements

2019 NSSC Fall Workshop at LLNL

This material is based upon work supported by the Department of Energy National Nuclear Security Administration under Award Number DE-NA0003180.

Resources

1. J. Stevenson, T. Gozani, M. Elsalim, Cathie Condron and C. Brown, “Linac Based Photofission Inspection System Employing Novel Detection Concepts,” Nucl. Instruments and Methods in Physics Research A, 652, 2011, 124-128.
2. M. Kobayashi, M. Ishii, Y. Usuki, and H. Yahagi, “Cadmium Tungstate Scintillators with Excellent Radiation Hardness and Low Background,” Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, vol. 349, no. 2, pp. 407 – 411, 1994.
3. C. L. Woody, J. A. Kierstead, P. W. Levy, and S. Stoll, “Radiation damage in undoped CsI and CsI(TI),”IEEE Trans. Nucl. Sci., vol. 39, pp. 524–531, 1992
4. Y.-H. Hu, D. Shedlock, A. Wang, J. Rottmann, P. Baturin, M. Myronakis, P. Huber, R. Fueglistaller, M. Shi, D. Morf, J. Star-Lack, and R. I. Berbeco, “Characterizing a Novel Scintillating Glass for Application to Megavoltage Cone-beam Computed Tomography,” Medical Physics, vol. 46, no. 3, pp. 1323–1330, 2019
5. L. ur, J. Pisarska, and W. Pisarski, “Terbium-Doped Heavy Metal Glasses for Green Luminescence,” Journal of Rare Earths, vol. 29, pp. 1198–1200, 12 2011.
6. R. L. Weisfield, "Amorphous silicon TFT X-ray image sensors," International Electron Devices Meeting 1998. Technical Digest (Cat. No.98CH36217), San Francisco, CA, USA, 1998, pp. 21-24, doi: 10.1109/IEDM.1998.746237.