New Scintillators for Nuclear Security

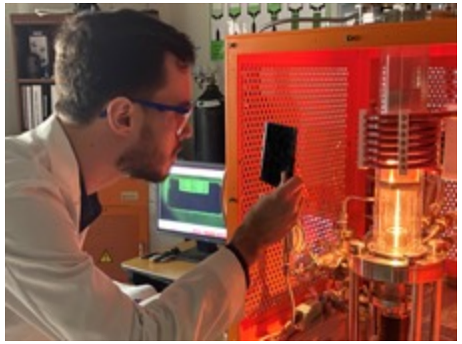
Chuck Melcher

Scintillation Materials Research Center

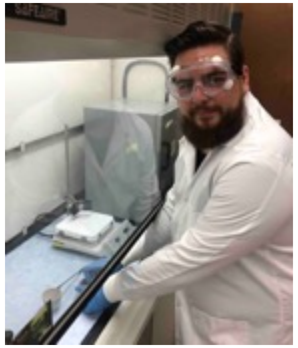
University of Tennessee

What is the Scintillation Materials Research Center?

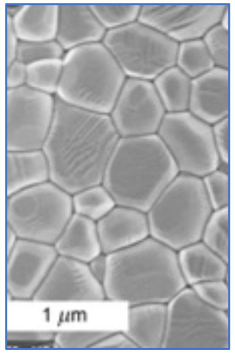
CRYSTAL GROWTH

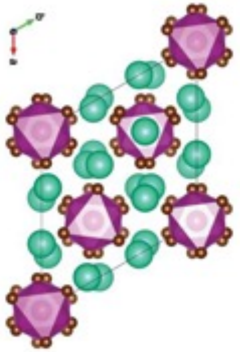


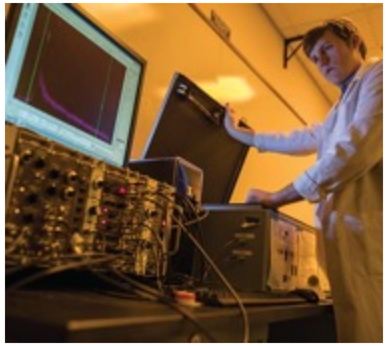
CERAMIC SYNTHESIS



MATERIALS AND SCINTILLATION CHARACTERIZATION







Started in 2006 as an academic-industrial collaboration for nuclear medicine.

$4M grant from Siemens Medical Imaging.

Broadened scope in ~2008 to include nuclear security.

Who is the Scintillation Materials Research Center

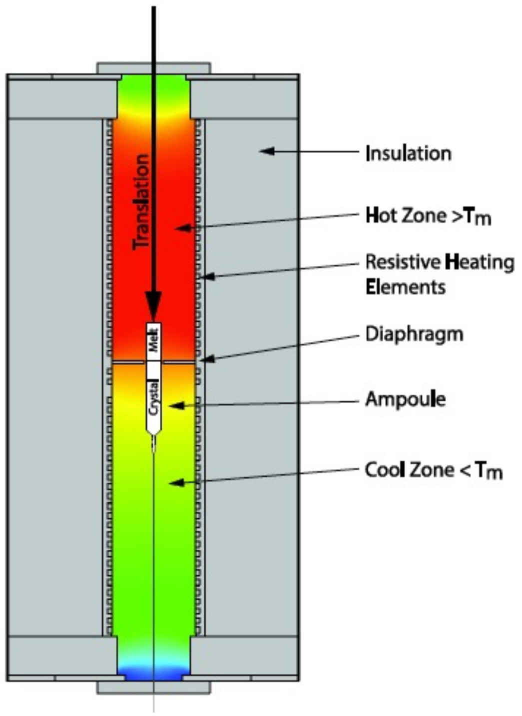
Located in the College of Engineering: personnel from Departments of Materials Science and Nuclear Engineering

2 faculty
2 post-docs
1 research associate
7 grad students
4 undergrads

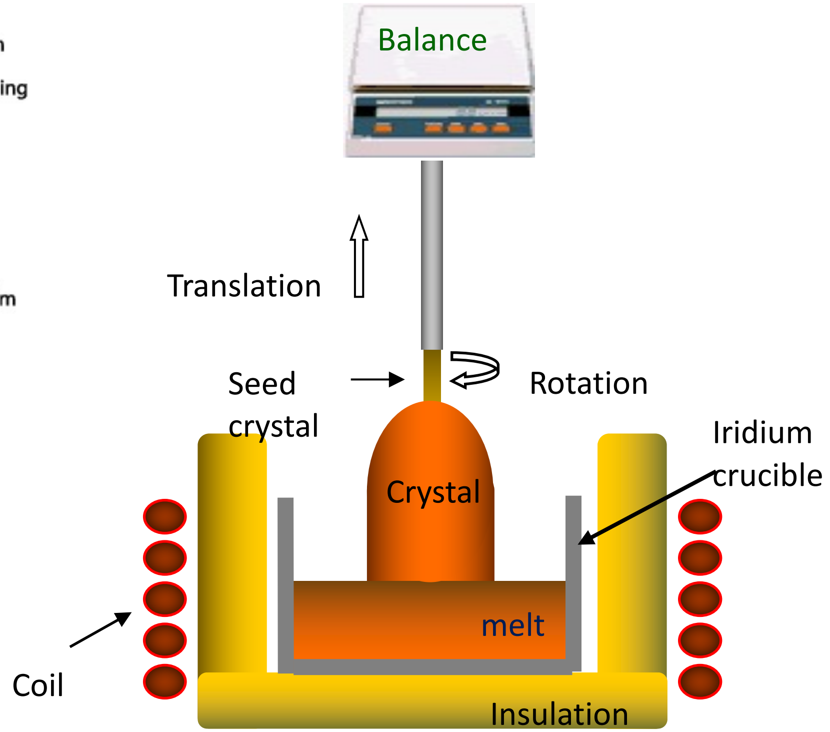
Collaborators from academia, industry, national labs

Not a “user facility” but a collaboration platform for funded projects

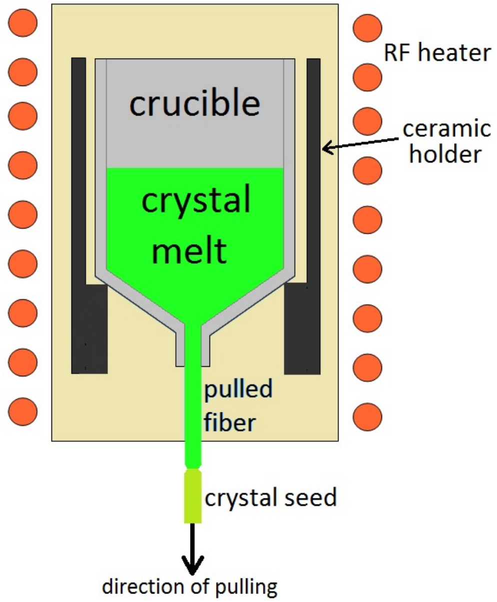
Synthesis techniques



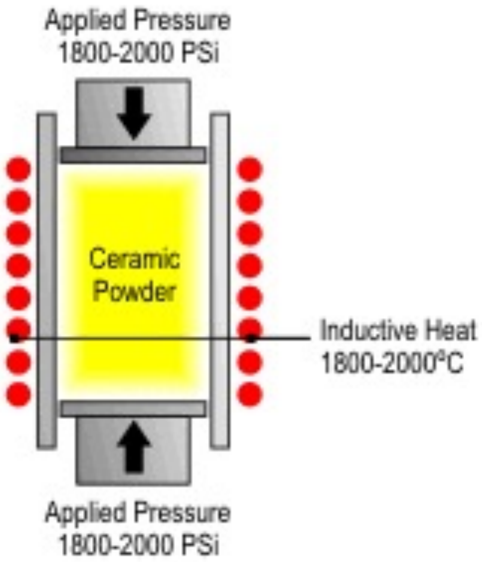
Bridgman crystal growth



Czochralski crystal growth



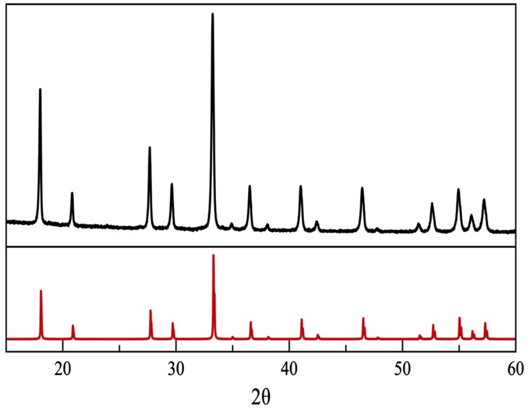
Micro-pulling-down



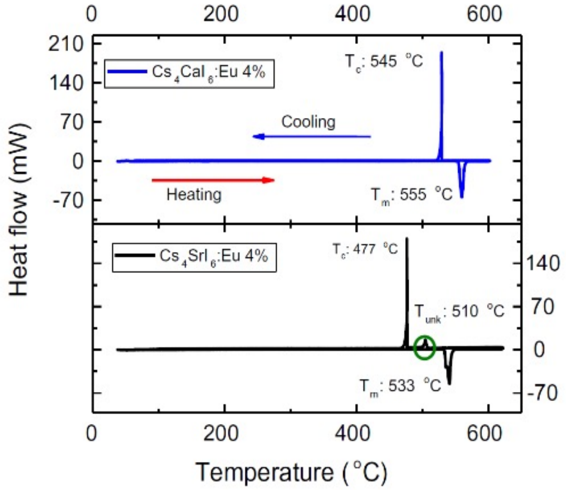
Hot-pressing ceramics

Characterization

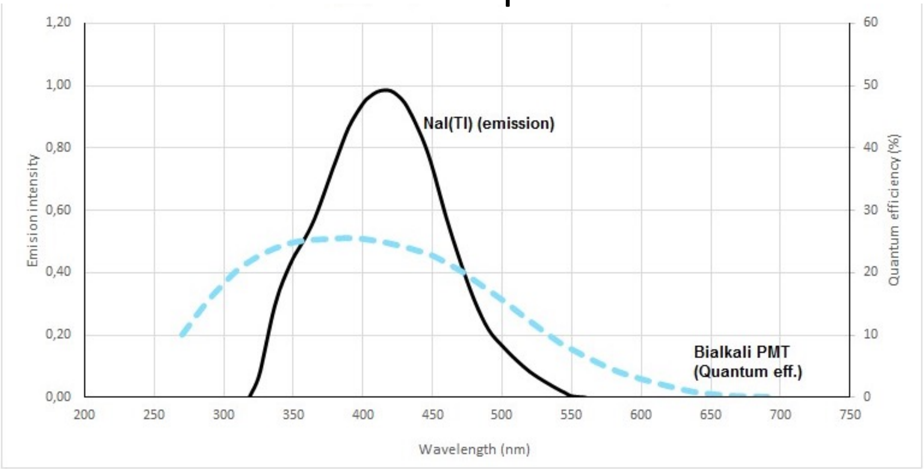
X-ray Diffraction



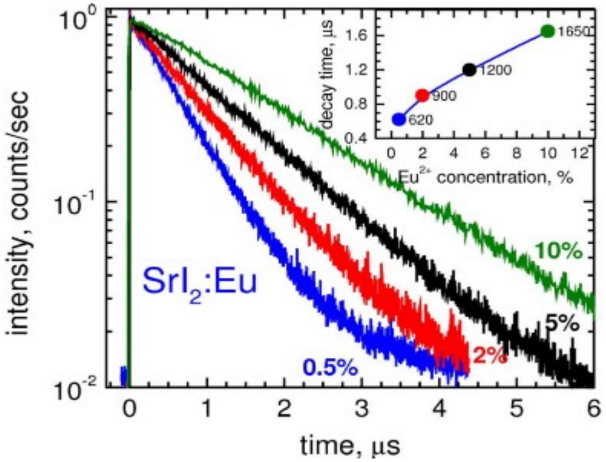
Differential Scanning Calorimetry



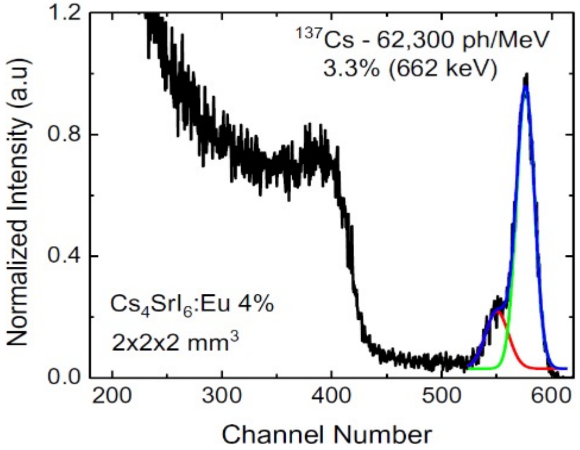
Emission spectrum



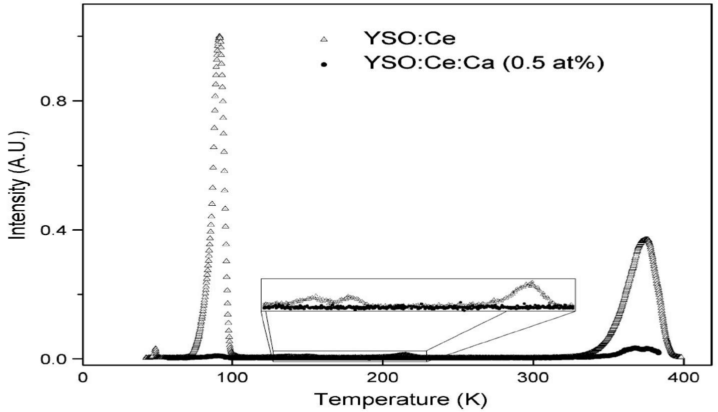
Scintillation time profile



Gamma-ray response



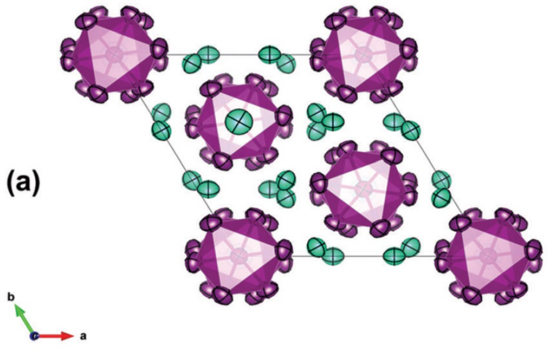
Thermoluminescence

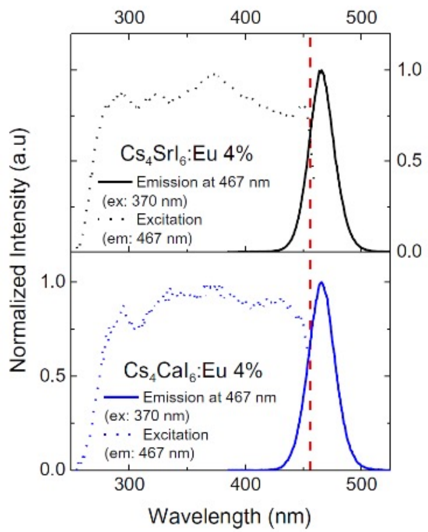


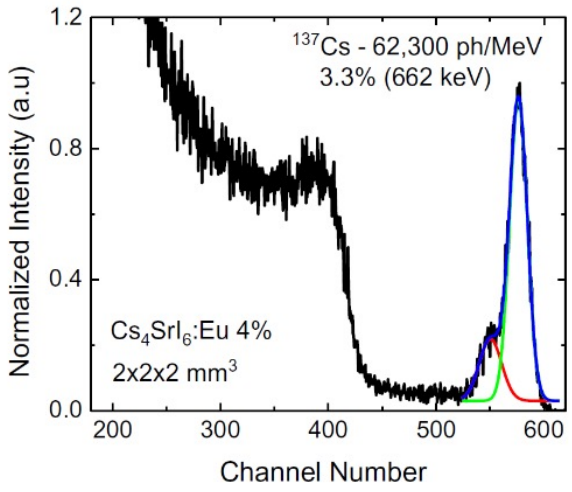
Compositional engineering of A_4BX_6:RE scintillators

- Cs_4SrI_6 and Cs_4CaI_6 discovered at SMRC at University of Tennessee
- First publication by L. Stand *et al.*
- Prelim. results: <4% ER at 662 keV (for small 2x2x2 mm^3 crystals)
- Demonstrated ability to be grown from the melt
- Properties superior to traditional scintillators

(a)







Properties compared with existing commercially available scintillators

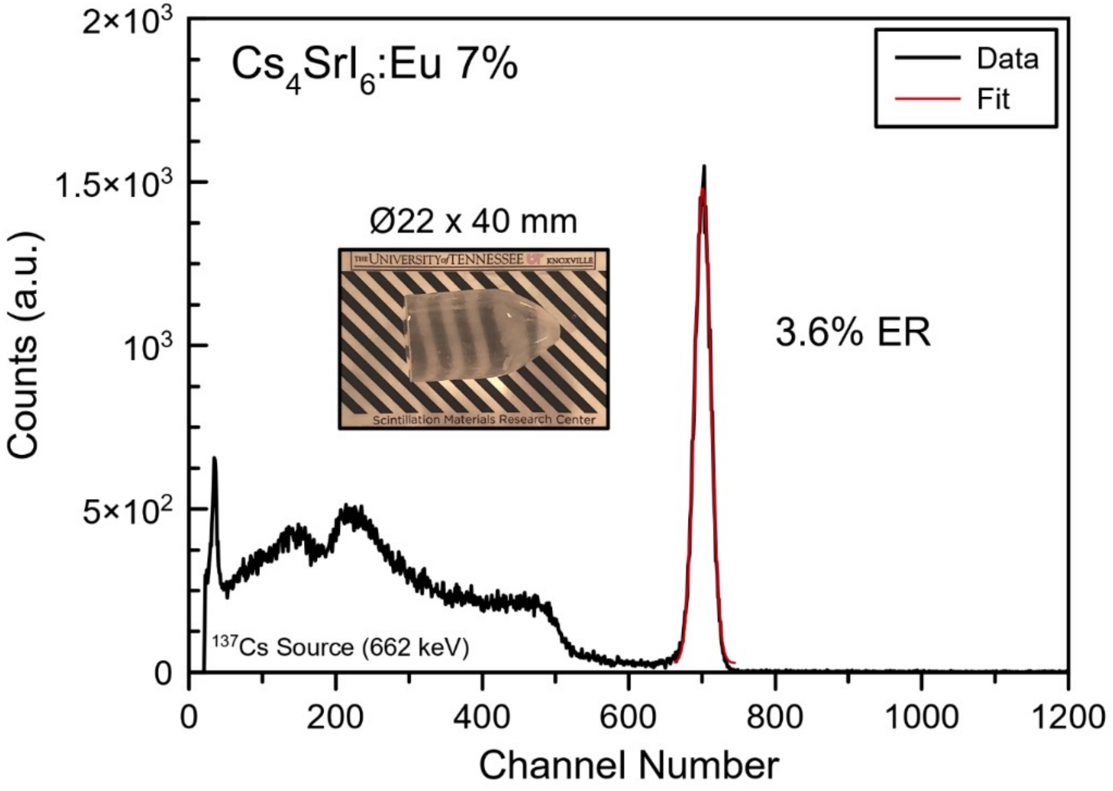
	ER at 662 keV (%)	Light Yield (ph/MeV)	Emission (nm)	Z_{eff}
NaI:Tl	7%	38,000	415	51
CsI:Tl	6%	54,000	550	54
Cs_4SrI_6:Eu 4%	3.3%	62,300	460	53.5
Cs_4CaI_6:Eu 4%	3.6%	51,800	462	53.4

N. Cherepy et al., IEEE Transactions on Nuclear Science, **56** (2009) 873-880
L. Stand et al., Journal of Crystal Growth, **486** (2018)

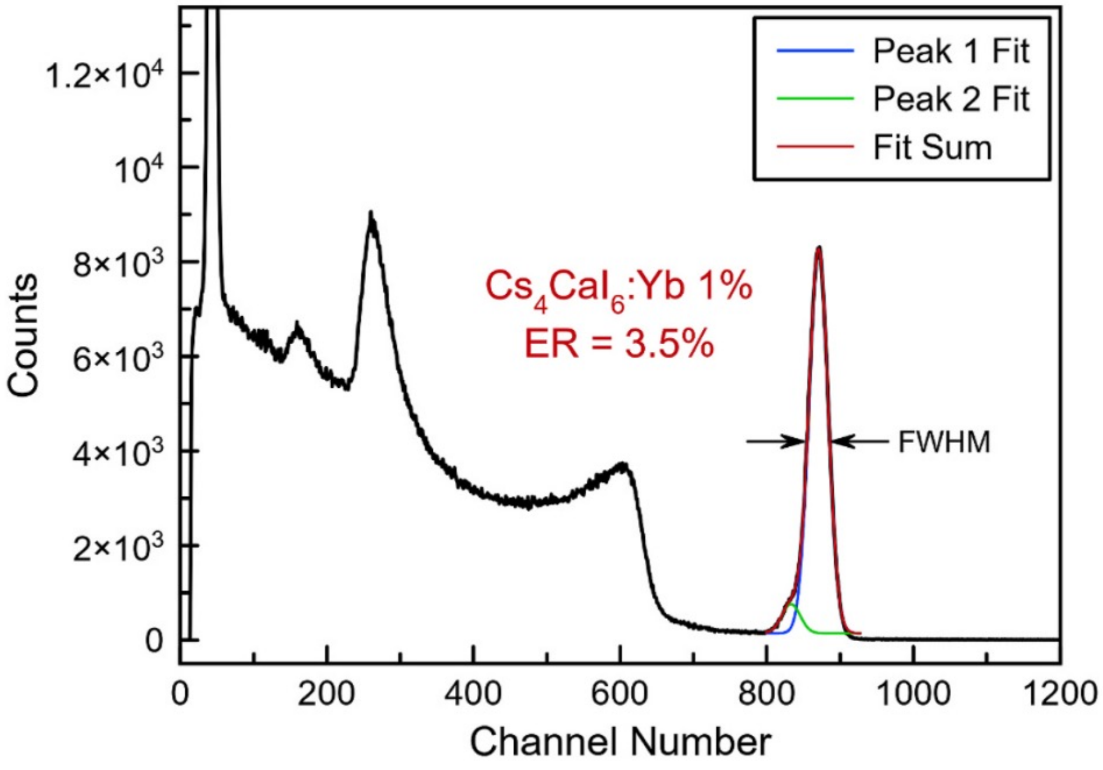
Comparison of Eu^{2+} and Yb^{2+} luminescent activators

^{137}Cs pulse height spectra

Eu^{2+}

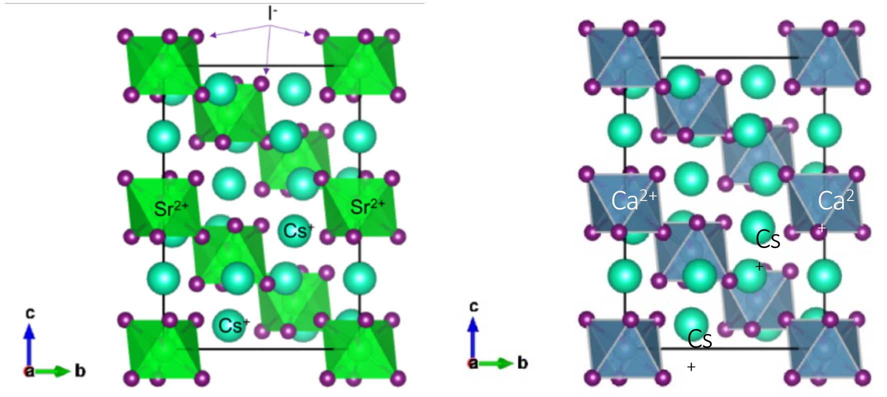


Yb^{2+}

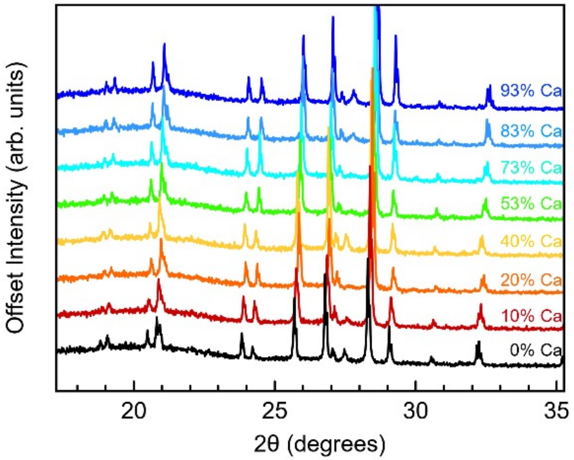


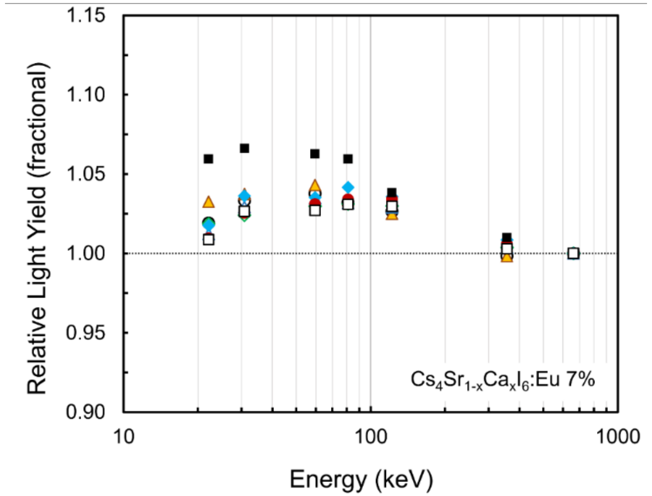
Optimize Sr:Ca ratio on divalent site in $Cs_4Sr_{1-x}Ca_xI_6$

- Goal
  - Improve ER and LY by substitution of the divalent cation
- Cs_4SrI_6 and Cs_4CaI_6 both have trigonal crystal structure, space group *R*-3*c*
  - Expected to form solid solution
- Sr^{2+} and Ca^{2+}
  - Occupy same site
  - Slight ionic radii mismatch
    - Sr^{2+} = 1.18 Å
    - Ca^{2+} = 1.01 Å



Cs_4SrI_6 Cs_4CaI_6



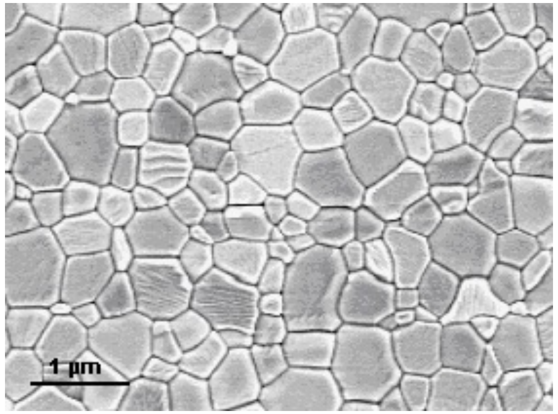


*Structural models generated using single crystal X-ray diffraction data collected at ORNL

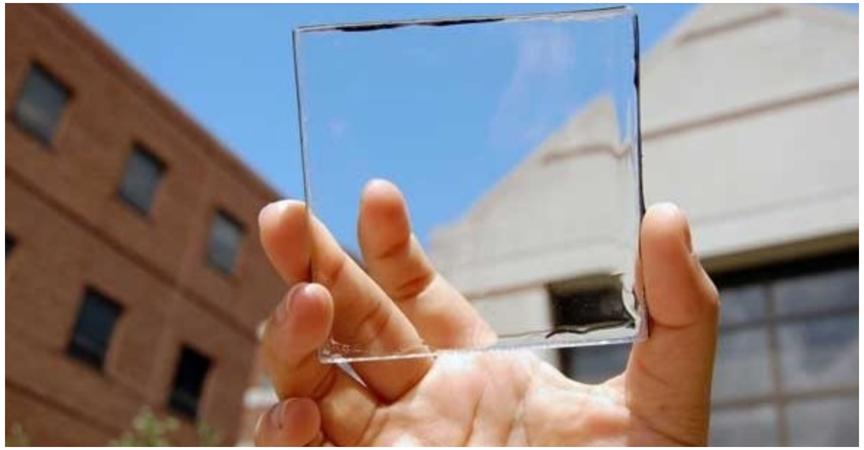
Polycrystalline ceramic synthesis of $Li_5La_3Ta_2O_{12}$

- Radiation detection advantages
  - High density and atomic number for gamma rays
  - High lithium content for neutrons (^{6}Li option)
  - Environmentally inert
  - Cubic crystal structure
- Advantages of ceramic synthesis
  - Faster than crystal growth
  - Lower temperature
  - Control of lithium content
  - Scalable process; can produce near net shapes
- Scintillation advantages
  - Trivalent site for Ce3+ activation
  - Fast decay time
  - No optical absorption
  - Potential intrinsic emission

Desired grain structure

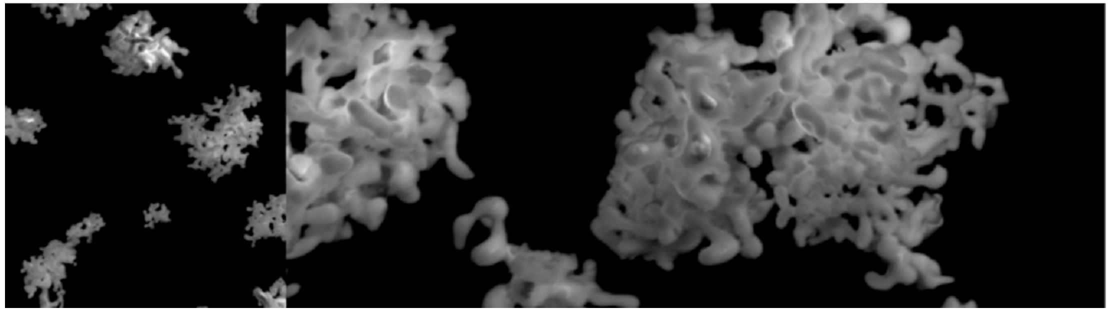


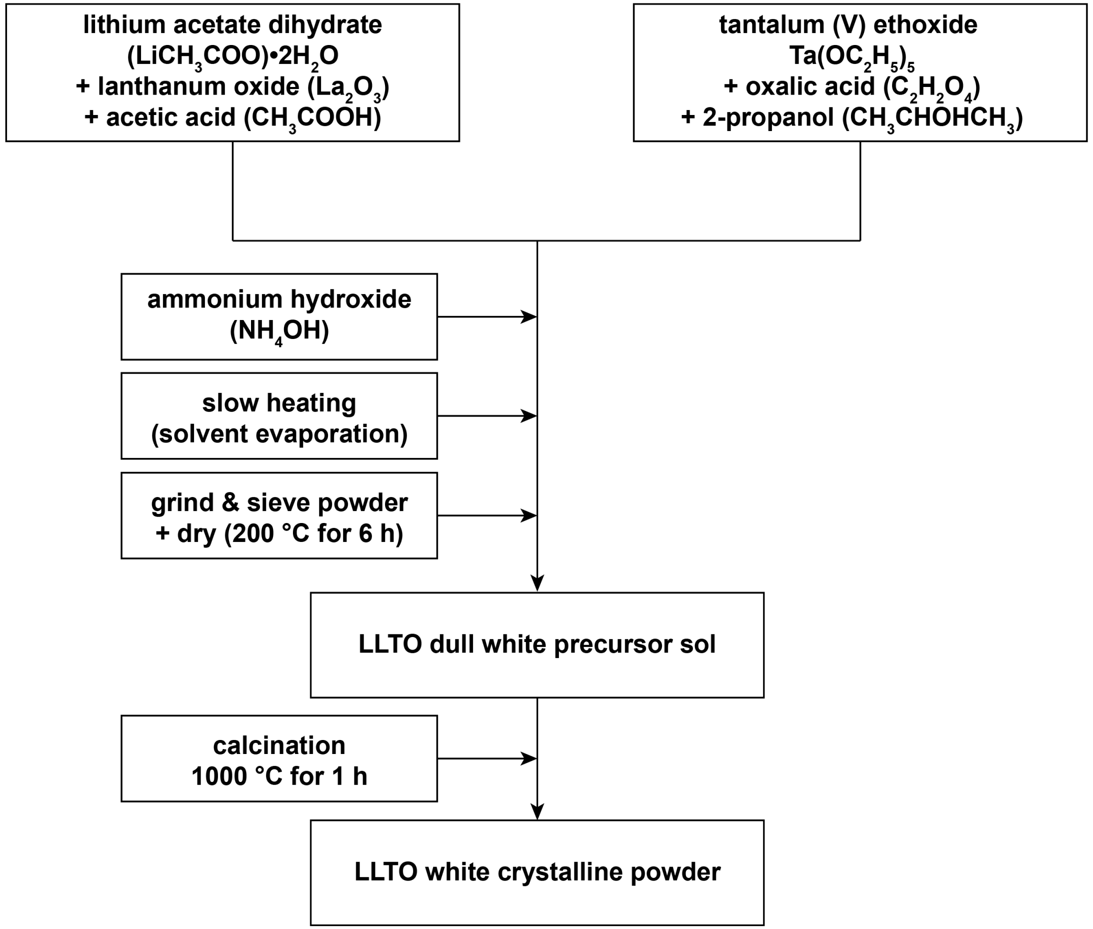
Desired transparency



Synthesis of precursor powder

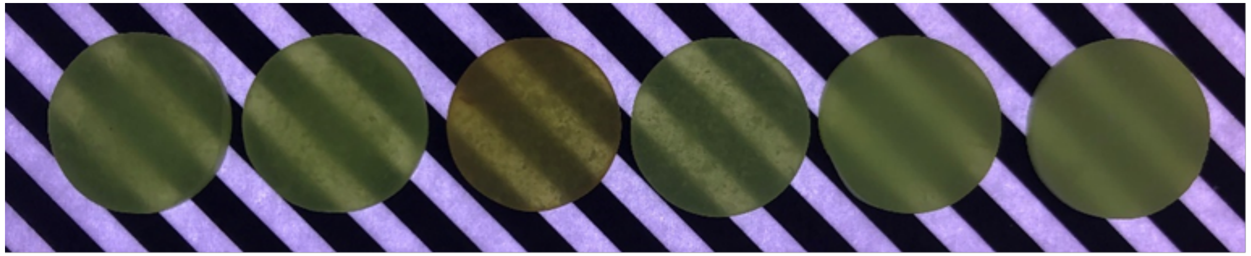
- Flexible alkoxide sol-gel method for synthesis of LLTO
    - Process reaction:
        - Hydrolysis and polycondensation of Ta-ethoxide
    - Chelating agent:
        - Oxalate (bidentate ligand) and Acetate





Effect of hot-pressing parameters on transparency

Decreasing Applied Pressure

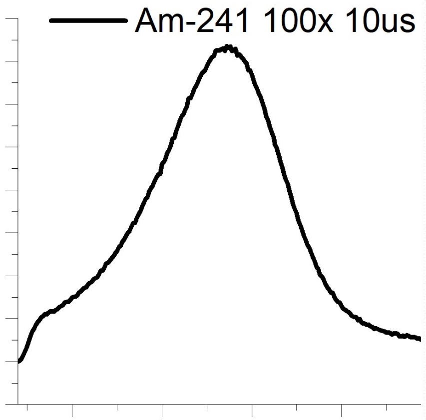


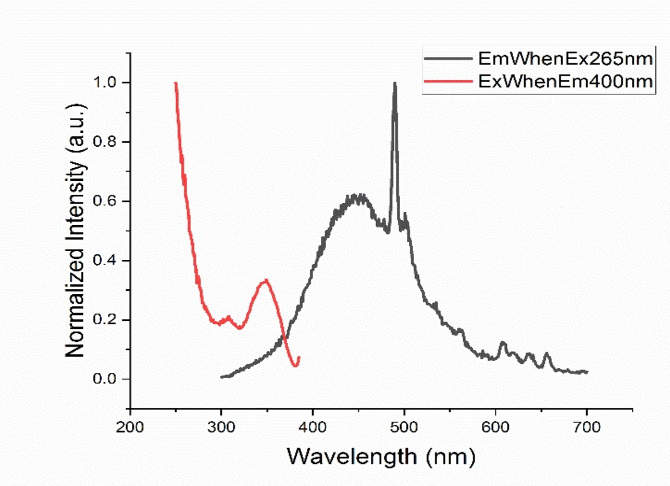
1200°C 67 MPa	1200 °C 46 MPa	1200 °C 33 MPa 1000 °C Air Anneal	1200 °C 33 MPa	1200 °C 33 MPa 1 wt% $LiCO_3$	1100 °C 33 MPa

Increasing Temperature

1150 °C 1200 °C 1250 °C

First observation of radiation response





Acknowledgments