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$\text{Cs}_2\text{LiCeCl}_6$: An intrinsic scintillator for dual gamma and neutron detector applications

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70 YEARS OF
DISCOVERY

A CENTURY OF SERVICE



SRNL-STI-2017-00527

Introduction

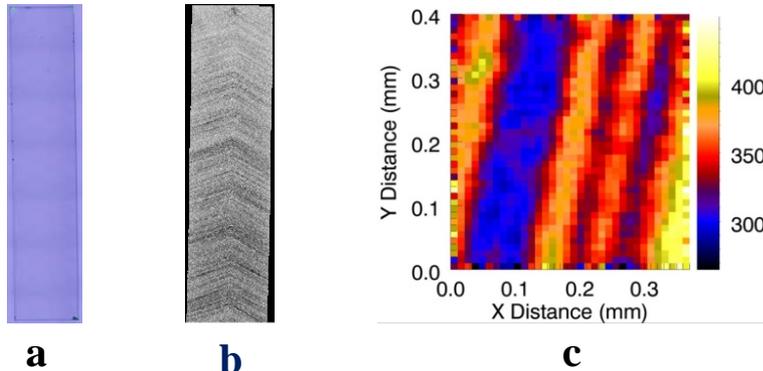
Why are intrinsic materials preferred ?

Most of the scintillators are activated by foreign dopants

e.g.; NaI:Tl, LaBr₃:Ce and SrI₂:Eu

Disadvantages: i) Segregation of dopants
ii) Growth striations

The segregation and striations can severely degrade the uniformity and energy resolution of large-volume devices.



a) Optical transmission picture of a LSO:Ce sample of dimensions $\sim 20 \times 6 \times 6$ mm³, b) X-ray response map, and (c) high magnification fluorescence map of Ce atoms (dopant).

Advantage of intrinsic scintillators:
Intrinsic compounds will ensure homogeneity of the material throughout the grown ingot, and thus a uniform detector response can be achieved at substantially lower cost for a large volume detector. The energy resolution is expected to be independent of detector volume.

Motivation

$\text{Cs}_2\text{LiYCl}_6:\text{Ce}$ (CLYC) is the most well known dual gamma-neutron- detector material. In our present study $\text{Cs}_2\text{LiCeCl}_6$ (*CLCC*) was chosen as the intrinsic scintillator as a potential replacement for CLYC.

This material is from the Elpasolite family; it has a cubic crystal structure and a density of ~ 3.4 gm/cc.

Advantage of $\text{Cs}_2\text{LiCeCl}_6$:

Intrinsic material; energy resolution is expected to be independent of the volume of the detector.

Growth of $\text{Cs}_2\text{LiCeCl}_6$

Crystals were grown by the vertical Bridgman technique.

The compound was synthesized from



99.999% pure anhydrous CsCl
> 99.999% pure anhydrous CeCl_3
> 99% pure anhydrous LiCl

Same ampoule was used for synthesis and growth.

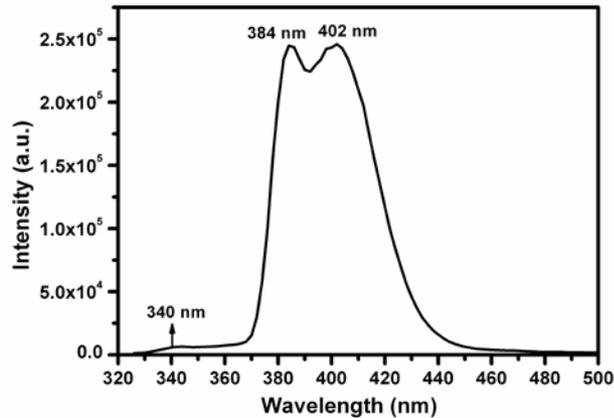
Loaded material in the quartz ampoule was heated to $\sim 180^\circ\text{C}$ for 24 hrs under a dynamic vacuum.

The ampoule was sealed under dynamic vacuum of 2×10^{-6} torr.

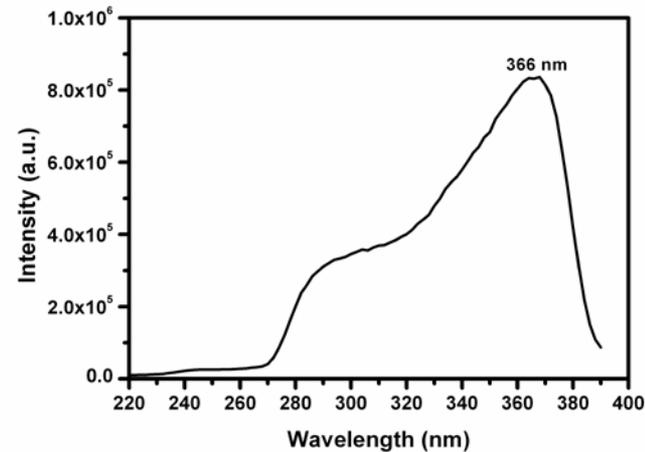
Growth rate: ~ 1.4 cm/day.

Post growth cooling rate: 4°C/hr .

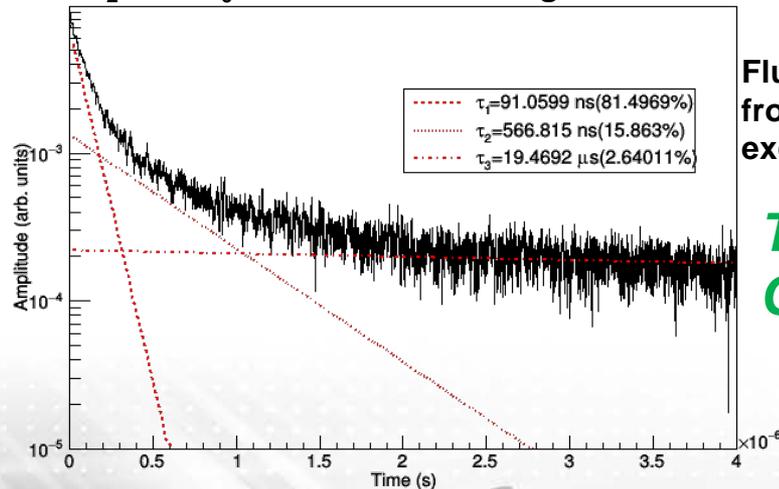
Emission and excitation spectra of $\text{Cs}_2\text{LiCeCl}_6$ and fluorescence decay



Optically pumped emission spectrum of $\text{Cs}_2\text{LiCeCl}_6$. Excitation wavelength: 280 nm



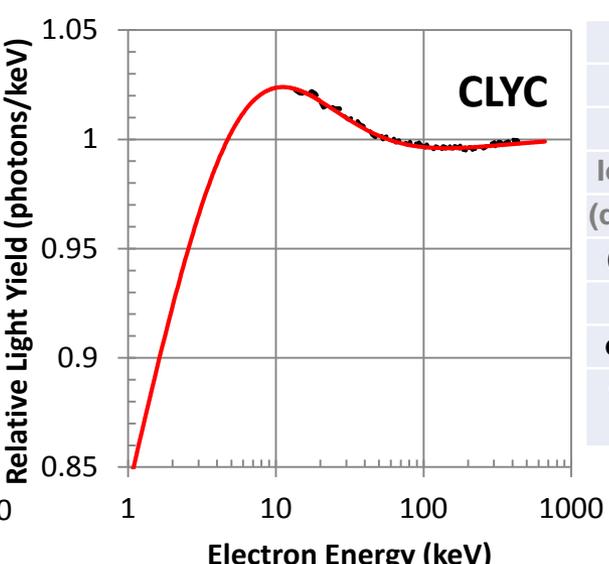
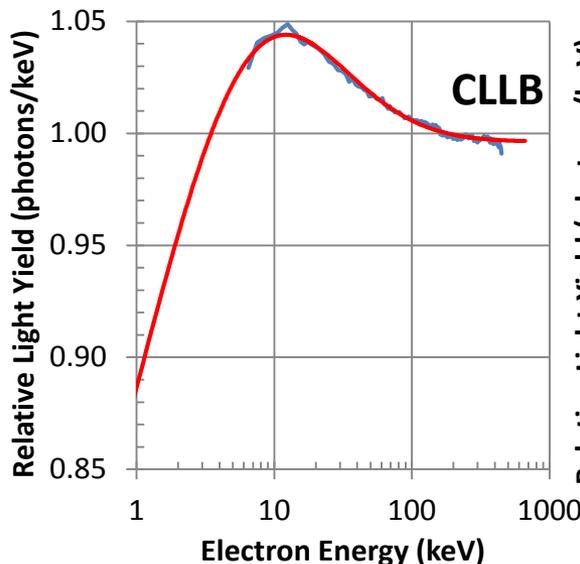
Excitation spectrum of $\text{Cs}_2\text{LiCeCl}_6$.



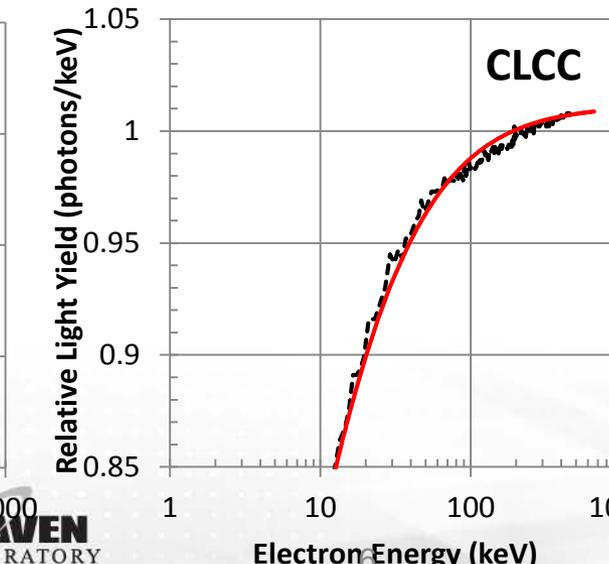
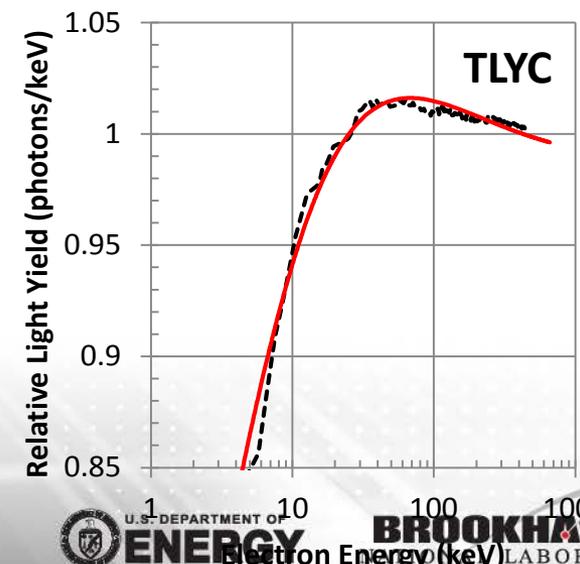
Fluorescence decay obtained from $\text{Cs}_2\text{LiCeCl}_6$ using 662-keV excitation from a ^{137}Cs source.

The response of CLCC is faster than CLYC.

Non-proportionality of CLCC and other elpasolites (activated by cerium) for comparison

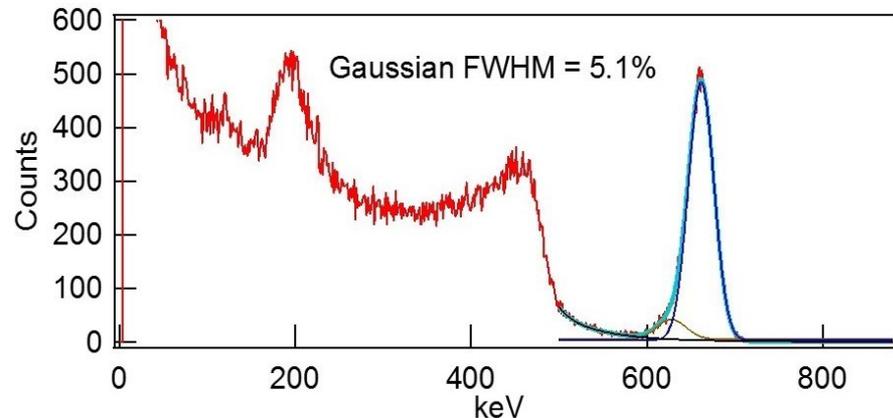


Parameter	Elpasolites			
	CLLB	CLYC	TLYC	CLCC
Density	4.2 g/cm ³	3.3	4.5	3.52
Ioniz. Energy	0.45 keV	0.33	0.47	0.36
(dE/dx) _{ONSAGER}	36.4 MeV/cm	36.4	36.4	36.4
(dE/dx) _{BIRKS}	730 MeV/cm	(520)	300	155
(dE/dx) _{TRAP}	30 MeV/cm	40	0	0
e/h creation	19%	22	25	23
Calc'd Res. @ 662 keV	1.53%	(1.7?)	2.90	3.46



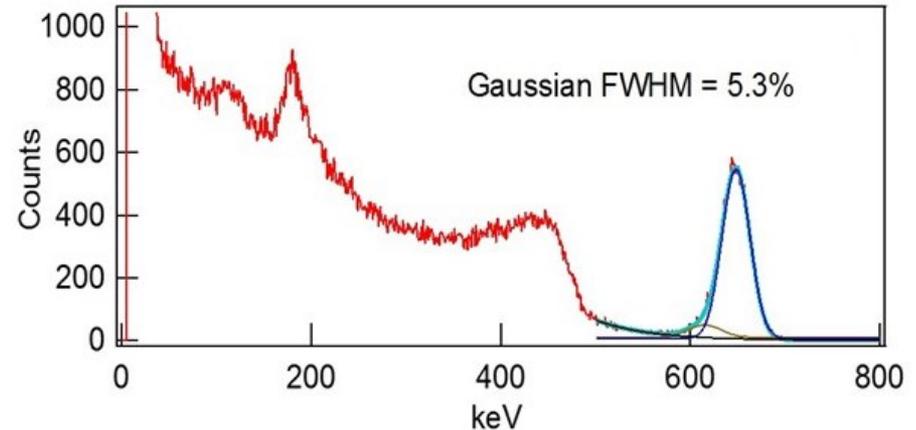
- All scintillators have similar percent of free electrons/holes created
- Bromide (CLLB) has least Birks exciton annihilation and best predicted resolution, CLCC has the most
- CLYC & CLLB have large amount of trapping
- TLYC & CLCC have little trapping, perhaps because TI⁺ and Ce³⁺ (respectively) reduce the bandgap and circumvent impact of native shallow traps

Detector response of $\text{Cs}_2\text{LiCeCl}_6$ for gamma radiation



After polishing, before packaging

- LY = 34,000 Ph/MeV
- R(662 keV) = 5.1%

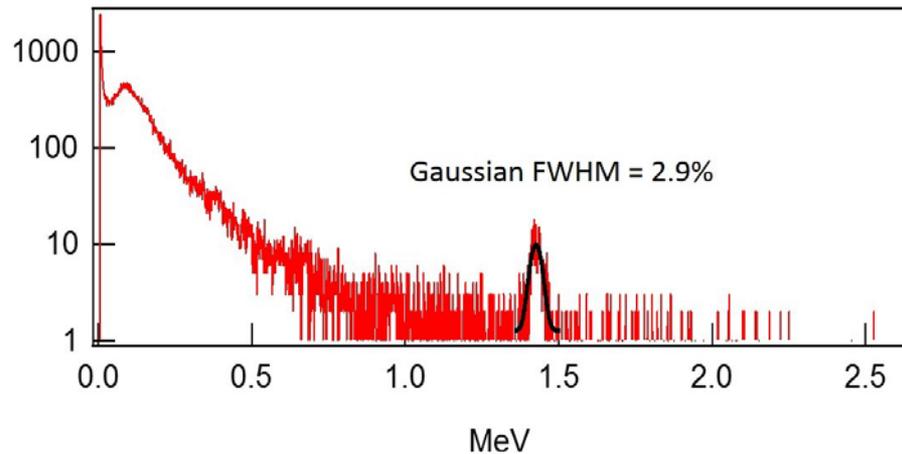


Crystal after packaging

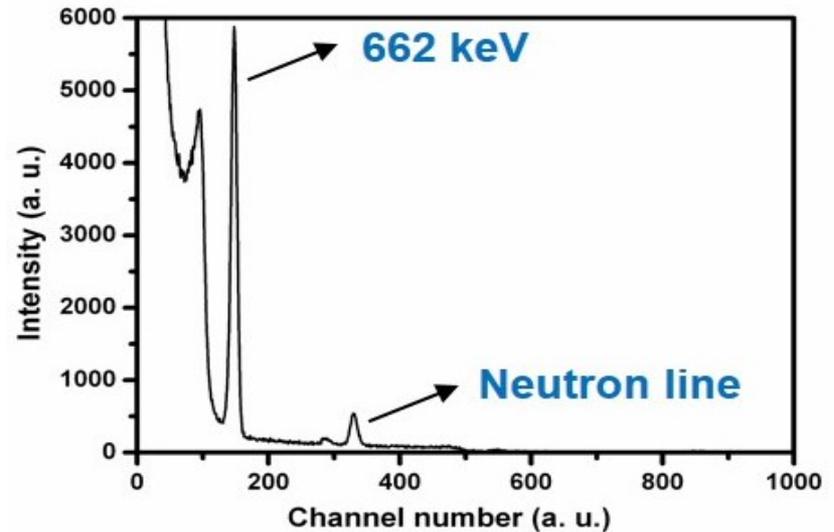
- LY = 30,000 Ph/MeV
- R(662 keV) = 5.3%

CLCC is *brighter* than CLYC. Light output of CLCC is ~34,000 ph/MeV, CLYC is ~22,000 ph/MeV

Detector response of $\text{Cs}_2\text{LiCeCl}_6$ to thermal neutron & gamma radiation



Neutron capture peak at 1.4 MeV



Pulse height spectrum of Am-Be thermal-neutron and ^{137}Cs gamma source, measured at BNL.
Resolution ~4%.

Neutron capture peak at 1.48 MeV

Summary

- *CLCC crystals can be grown in large volumes due to its cubic structure.*
- *CLCC is perhaps the only intrinsic scintillator capable of dual gamma- and neutron-detection.*
- *Because of intrinsic nature, the energy resolution of CLCC is expected to be independent of detector volume.*
- *CLCC has less trapping, and is capable of detecting lower-energy gamma rays compared to CLYC.*
- *CLCC is faster than CLYC.*
- *CLCC: brighter than CLYC.*

CLCC appears to be very promising and has tremendous potential to compete with CLYC, especially for large-volume detectors.

There is enough room for further improvement of energy resolution after successive purification by zone refining of the starting material.

Acknowledgement

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Thank you all for your kind attention