Experimental Investigation of a Machine Learning Model for Scintillator Discovery

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Affiliated lab: LANL

NSSC3 Kickoff Meeting and Advisory Board Review
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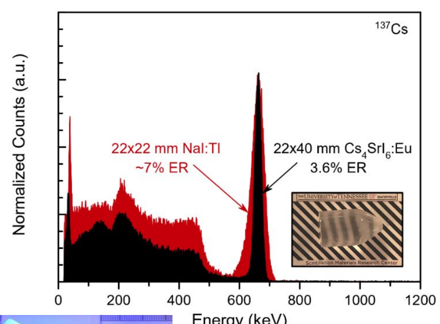
Introduction

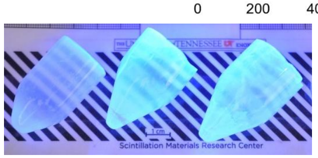
Department and University: Materials Science and Engineering Department, University of Tennessee
Academic Advisor: Dr. Mariya Zhuravleva
NSSC Research Focus Areas: Radiation Detection and Instrumentation
Planned Graduation Date: Dec. 2023

Lab Mentor/Partner Laboratory: Dr. Kenneth McClellan (LANL, MST-8)

Research Description: Discovery and development of novel scintillator materials

High Energy Resolution – Cs_4SrI_6:Eu

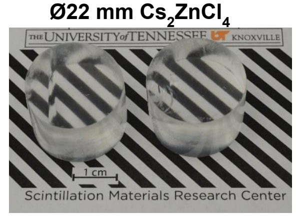


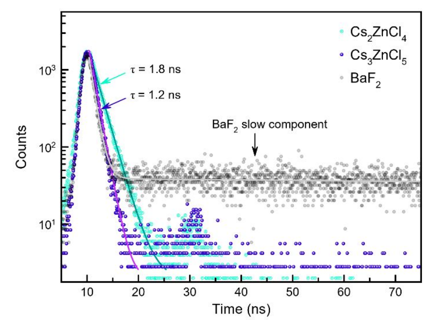


- 3.0% ER at 662 keV for small crystals
- Comparable performance to SrI_2:Eu
- Able to grow in large sizes

Fast timing – Cs_2ZnCl_4 and Cs_3ZnCl_5

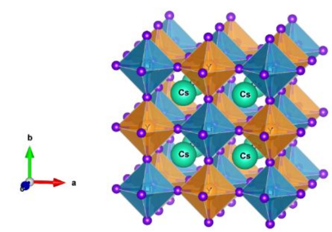
Ø22 mm Cs_2ZnCl_4





- 1-2 ns decay times
- Coincidence time resolution (CTR) as good as 137 ps FWHM
- Able to grow in large sizes

ML Prediction Model – Double Perovskites



- **LANL internship**
- **Topic of today's talk**

Relevance to NSSC / NNSA Mission

Mission: reduce global nuclear security threats through the innovation of technical capabilities to *detect*, *identify*, and *characterize* illicit diversion of special nuclear materials

The focus of this research is on developing scintillators with better capabilities (energy resolution, decay time, light yield, etc.) for nuclear safeguards and nonproliferation applications

- Ex. – use in handheld RIIDs, X-ray imaging systems, etc.

Experimental Investigation of Scintillator Prediction Model

LANL Internship Project

- Machine learning (ML) model has been developed for high-throughput screening of potential new scintillator materials □

Physics-informed machine learning for inorganic scintillator discovery

Cite as: J. Chem. Phys. **148**, 241729 (2018); https://doi.org/10.1063/1.5025819
Submitted: 13 February 2018 . Accepted: 05 April 2018 . Published Online: 25 April 2018

G. Pilania, K. J. McClellan, C. R. Stanek, and B. P. Uberuaga

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- Objectives of this work
  - Provide experimental data to feed back into model
  - Identify promising scintillator compounds to pursue further

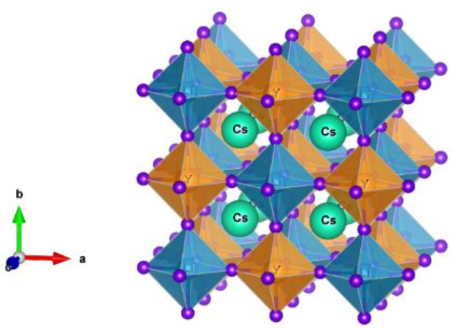
- Motivation:
  - Ideal scintillator does not exist
  - Discovery of new scintillator compounds can be lengthy and tedious
  - Complimentary use of modeling and experimental work can provide a more efficient approach than conventional “trial and error”

□ **Rapid screening, vast chemical spaces**

Project Overview

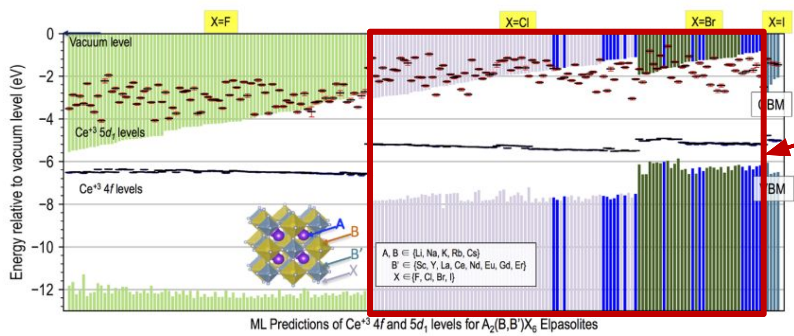
- Compositional space – **double perovskite halides** → **$A_2BB'X_6$:Ce** (A = Rb^+, Cs^+ ; B = Na^+, K^+ ; B' = RE^{3+} ; X = Cl^-, Br^-)
- Selected a set of *new compounds* to pursue experimentally, based on calculated electronic band structures
- Fabrication method – single crystal growth via Bridgman technique
- Characterization – physical, optical, and scintillation properties

Double perovskite structural model (Cs_2LiYCl_6)



Cubic space group *Fm*-3*m*

Positions of Ce^{3+} 5*d* and 4*f* energy levels relative to CB and VB for 200 Double Perovskite Halides



ML model predicts that some of these will be efficient scintillators

G. Pilania *et al.* "Physics-informed machine learning for inorganic scintillator discovery" J. Chem. Phys. **148**, 241729 (2018)

Initial Screening Done w/ RbCl-based Compounds

3 of the predicted scintillators, based on calculated positions of Ce^{3+} energy levels

Composition	Calc. Density (g/cm3)	Z_{eff}
$Rb_2NaLaCl_6$:Ce 2%	2.67	39.1
$Rb_2NaGdCl_6$:Ce 2%	2.93	43.5
Rb_2NaYCl_6:Ce 2%	2.61	30.7

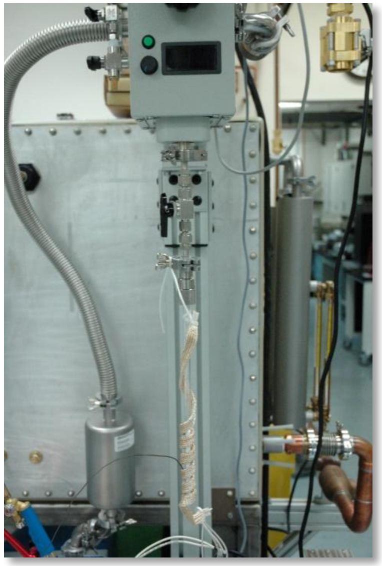
Screening process

Set up equipment and capabilities → **Synthesize and evaluate polycrystalline samples** → **Grow single crystals**

1. Confirm structure/phase (X-ray diffraction)
2. Measure scintillation properties
3. Provide experimental data to feed back into prediction models

Setting Up Equipment and Capabilities

(1) Vacuum Drying Station



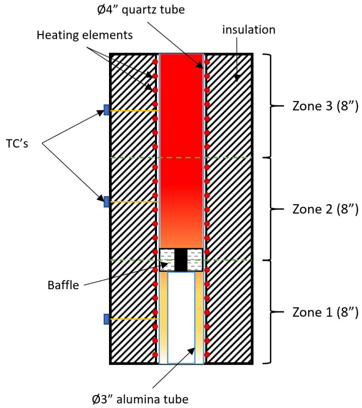
Raw material is dried under vacuum (~10^{-5} mbar) to remove moisture / hydrates

Seal with torch

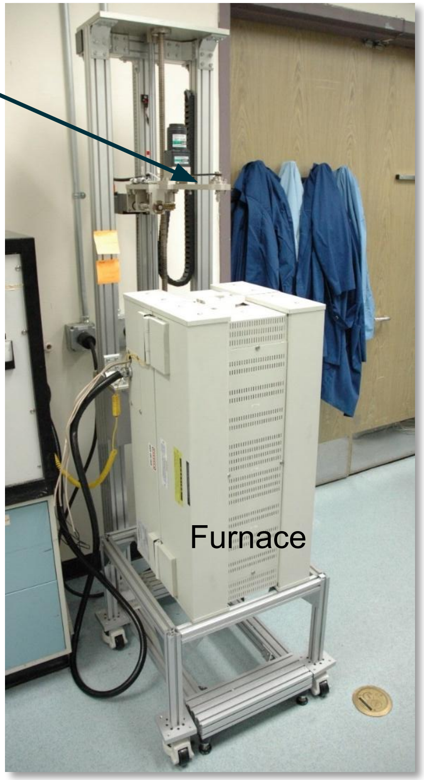
Encapsulated and dried powder, ready for synthesis

(2) Growth Furnace

Furnace schematic



Translation Mechanism

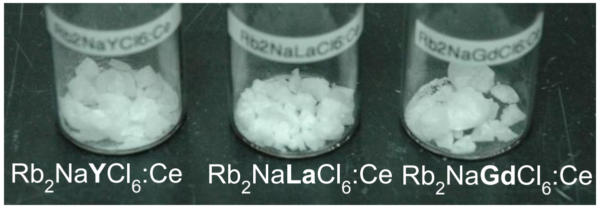


Lab was previously equipped for growth of *oxides*, not *halides*; require different handling and synthesis/growth methods.

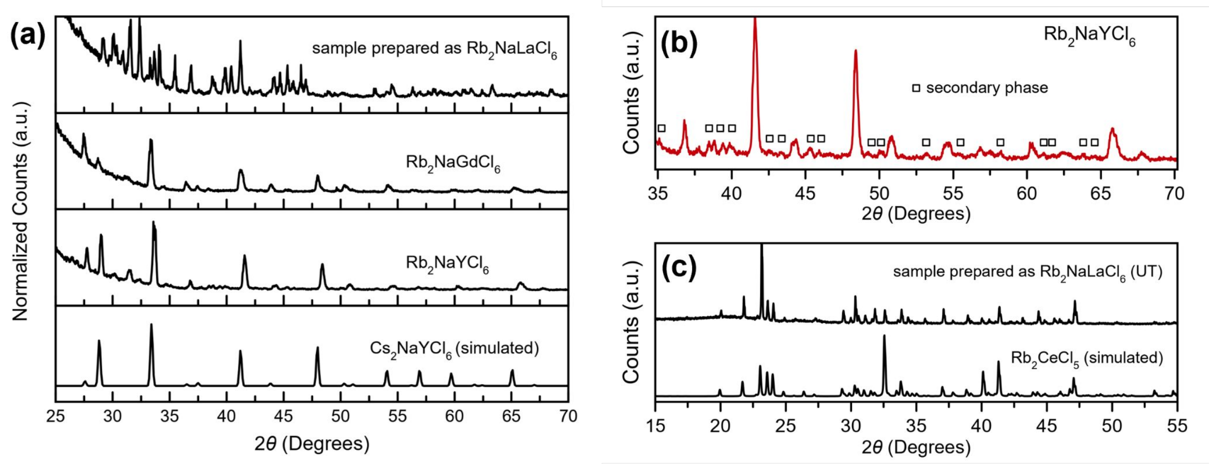
Successfully Synthesized Rb_2NaYCl_6:Ce and $Rb_2NaGdCl_6$:Ce

- XRD shows primary phase is cubic elpasolite (space group F*m*-3*m*)
- Low intensity peaks from secondary phase(s) also observed in XRD
  - Match with NaCl and $Rb_3(RE)Cl_6$ phases
  - Expected due to incongruent melting
- $Rb_2Na\mathbf{La}Cl_6$:Ce appears to be unstable compound
  - Instead, decomposes into Rb_2LaCl_5 and NaCl
  - Results confirmed w/ second sample prepared at UT

Synthesized Polycrystalline Samples



Measured XRD of Polycrystalline Samples vs Reference Patterns



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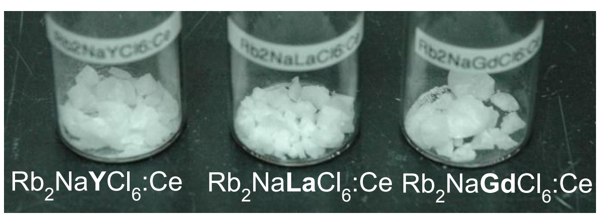
2/3 double perovskite compounds correctly predicted

Synthesized Polycrystalline Samples



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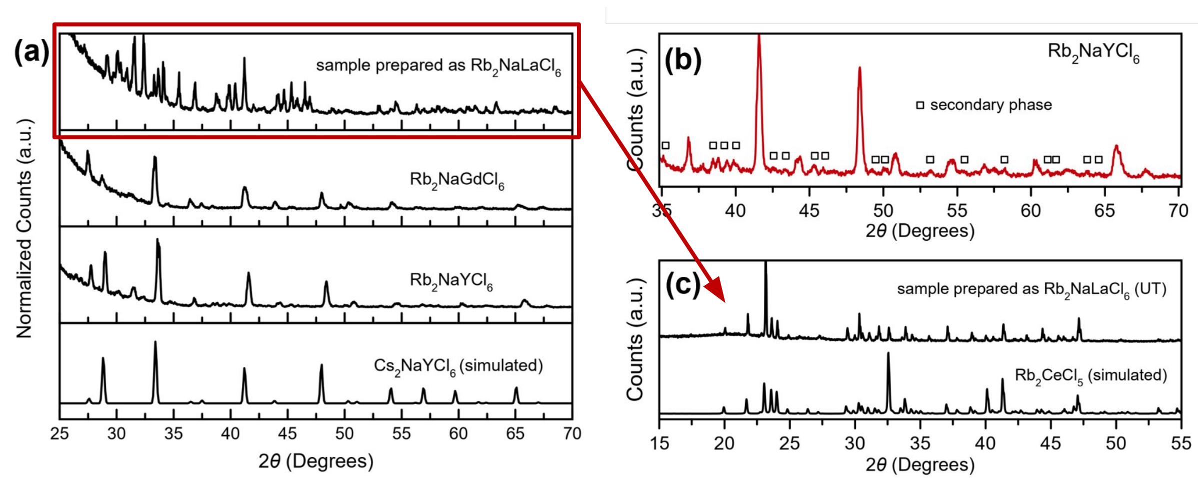
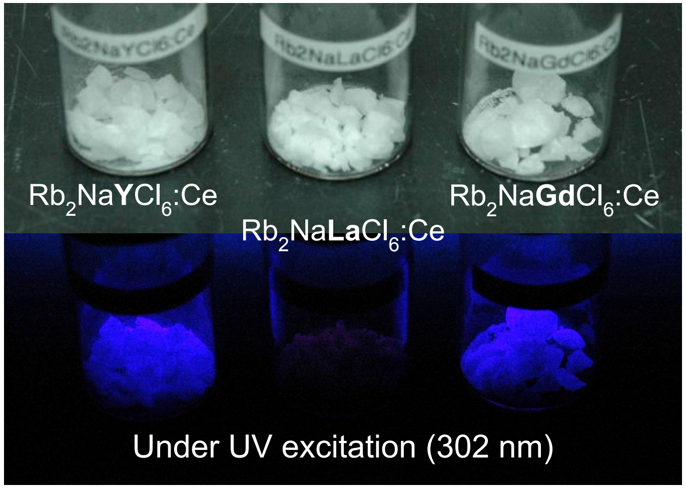
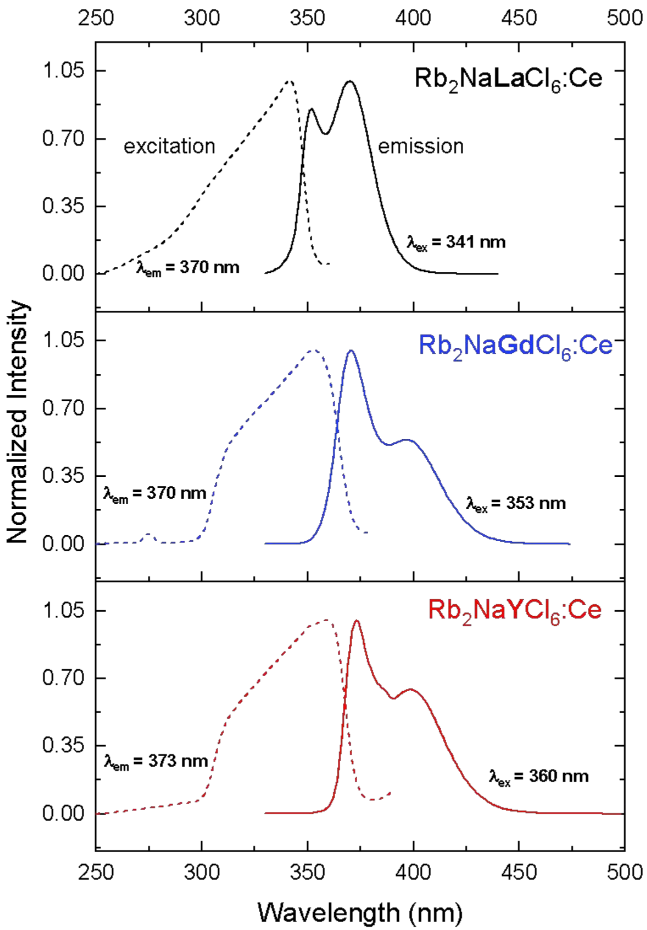


Photoluminescence Measurements Indicate Successful Incorporation of Ce^{3+} Activator

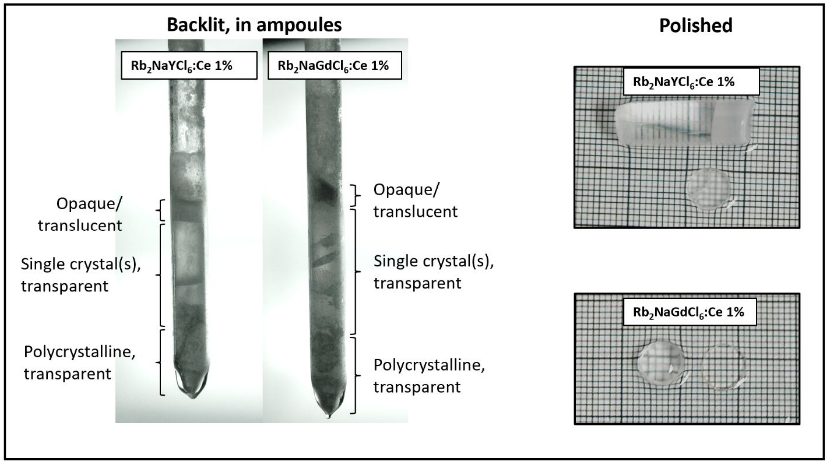
- Measured photoluminescence (PL) of the polycrystalline samples to assess which are promising
  - Uses *light* as excitation source
  - If no emission, could suggest unfavorable Ce^{3+} energy levels
- Strong doublet emission is observed from Ce^{3+} 5*d* to 4*f* electronic transition
- Based on result, proceeded with growth



Photoluminescence Spectra

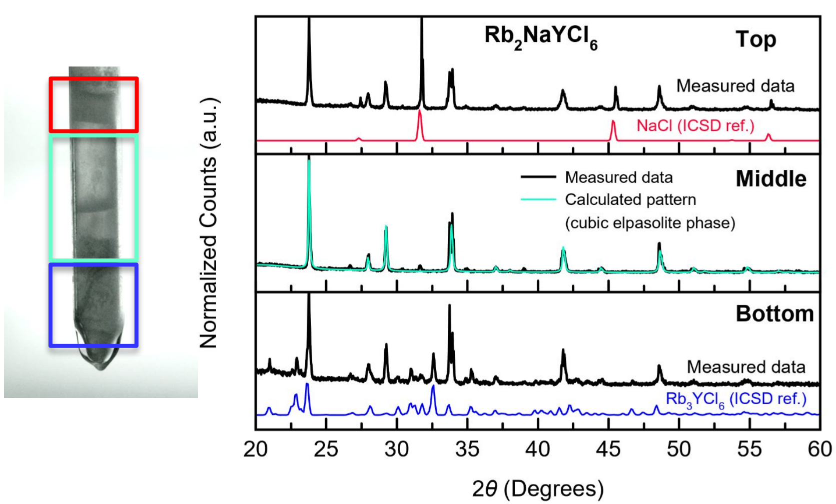


Growth Yielded Transparent Single Crystals of Rb_2NaYCl_6:Ce and $Rb_2NaGdCl_6$:Ce

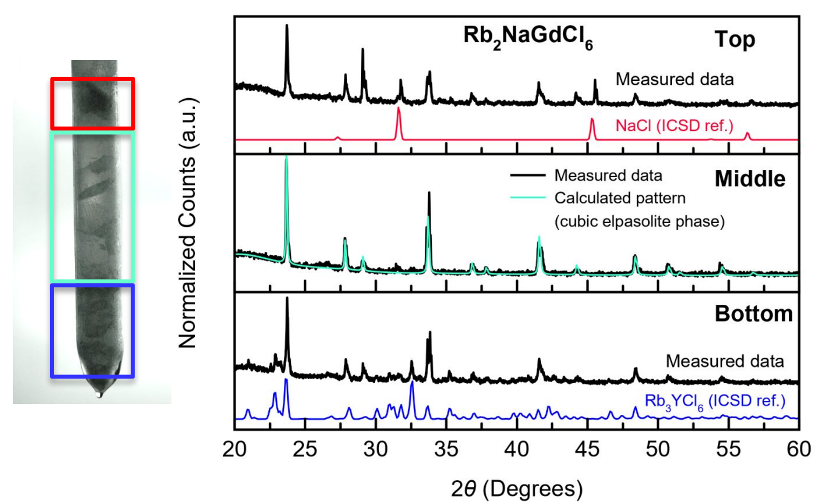


- Middle section of each crystal is transparent
- Phase purity analyzed w/ XRD
  - Shows middle is nearly phase pure for both
  - 3 distinct sections observed, common for elpasolites grown by directional solidification

XRD of Different Sections of Crystal

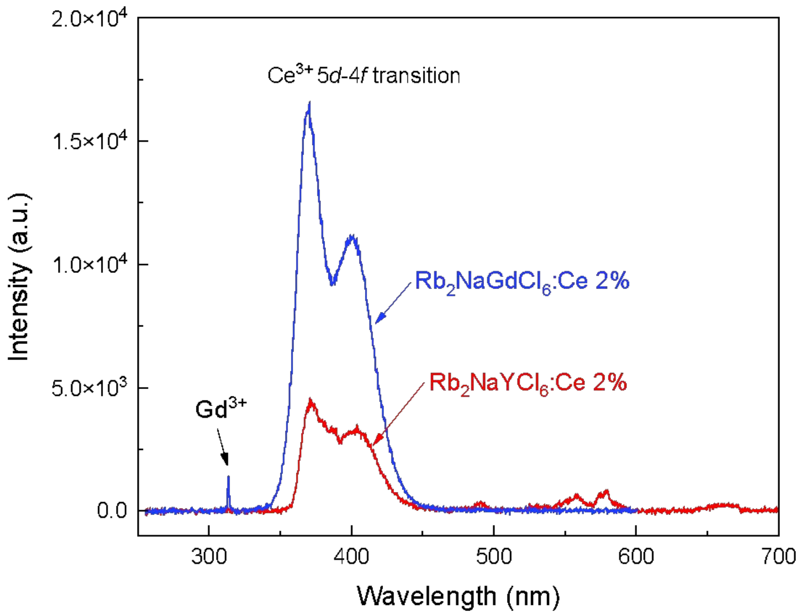


XRD of Different Sections of Crystal

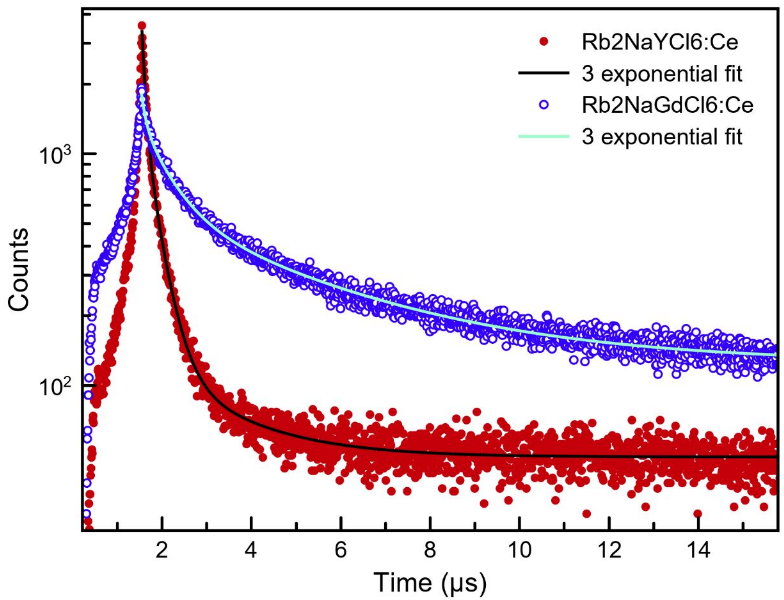


Both Crystals Scintillate Under X-ray and Gamma Excitation

Radioluminescence Spectra (excitation by X-rays)



Scintillation Decay Profiles (^{137}Cs)



Decay Constants

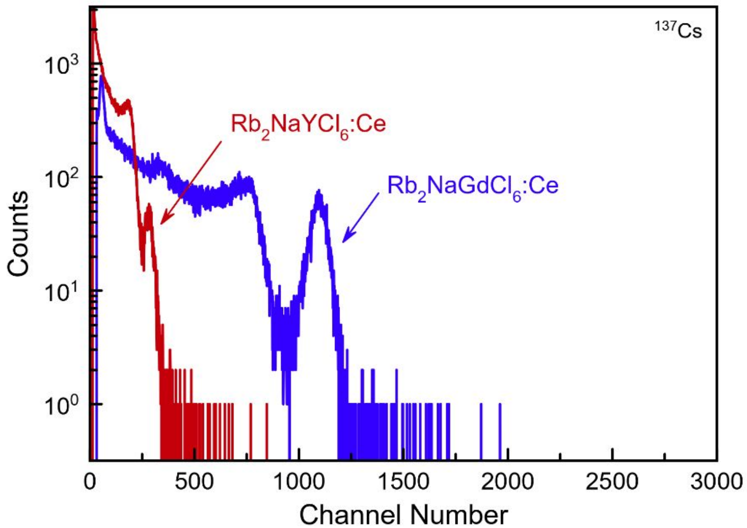
	T_1 (ns)	T_2 (ns)	T_3 (μs)
$Rb_2NaGdCl_6$:Ce	94 (2%)	632 (22%)	3.72 (76%)
Rb_2NaYCl_6:Ce	85 (25%)	340 (56%)	1.78 (19%)

$Rb_2NaGdCl_6$:Ce Has Moderate Light Yield and Energy Resolution

- Scintillation properties further evaluated by pulse height analysis
  - performed on 8 mm diameter slabs, ~3-4 mm thick
- $Rb_2NaGdCl_6$:Ce has LY of 14,000 ph/MeV
- Energy resolution is 5.5% at 662 keV
- Rb_2NaYCl_6:Ce has LY of 4,400 ph/MeV

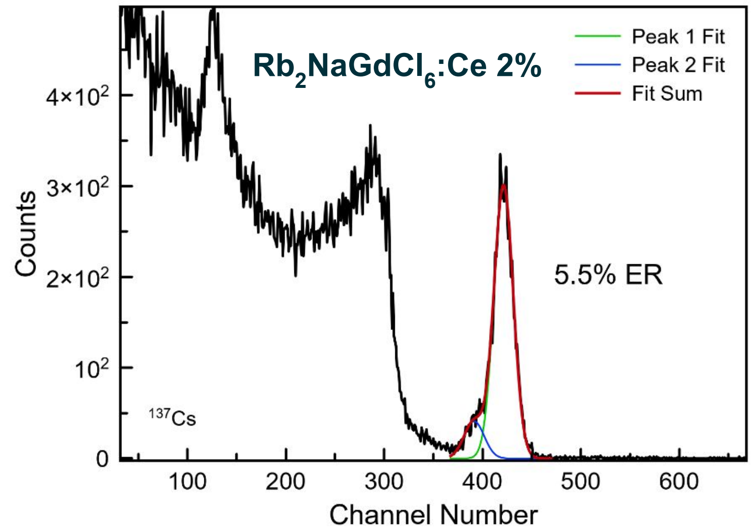
$$\mathrm{ER} = \frac{FWHM}{photopeak\ centroid}$$

Pulse height spectra used to calculate LY



Channel # is proportional to LY

Fitting for energy resolution calculation



*Gd escape peak at 43 keV lower than the full energy photopeak

Summary and Conclusions

- Single crystals of Rb_2NaYCl_6:Ce and $Rb_2NaGdCl_6$:Ce were grown
- Both were correctly predicted to scintillate
- Generated the first set of experimental data for a ML-based model for scintillator discovery
- Capabilities now set up for MST-8 group at LANL to continue work on synthesis and growth of novel halide scintillators

Although there is a wide range of compositions that will eventually need to be studied experimentally, this work is an important first step toward validating the ML model developed by Pilania *et al.*

The NSSC Experience

NSSC sponsored events

- Presented at UPR (2018, 2019)
- Presented at NSSC Fall Workshop (LLNL 2019)
- NSSC-LANL Keepin Nonproliferation Science Summer Program (2021)

Other conferences, workshops, etc. attended w/ funding from NSSC

- PNNL Radiation Detection for Nuclear Security Summer School (2018)
- International Summer School on Crystal Growth (ISSCG-17) in Granby, CO (2019)
- Presented at several conferences: SORMA (2018), ACCGE (2017, 2021), ICCGE (2019), IEEE (2020, 2021)

Connection with national labs

- Conducted summer research at LANL (2021)
- Spent 2 weeks at LLNL performing measurements (2019)
- Still collaborating w/ group at LANL

Plaza Blanca (Abiquiu, NM)

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Mentors and Collaborators

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Questions?