Progress in Polysiloxane Organic Scintillators

Mackenzie Duce

LANNS Symposium

May 12, 2023

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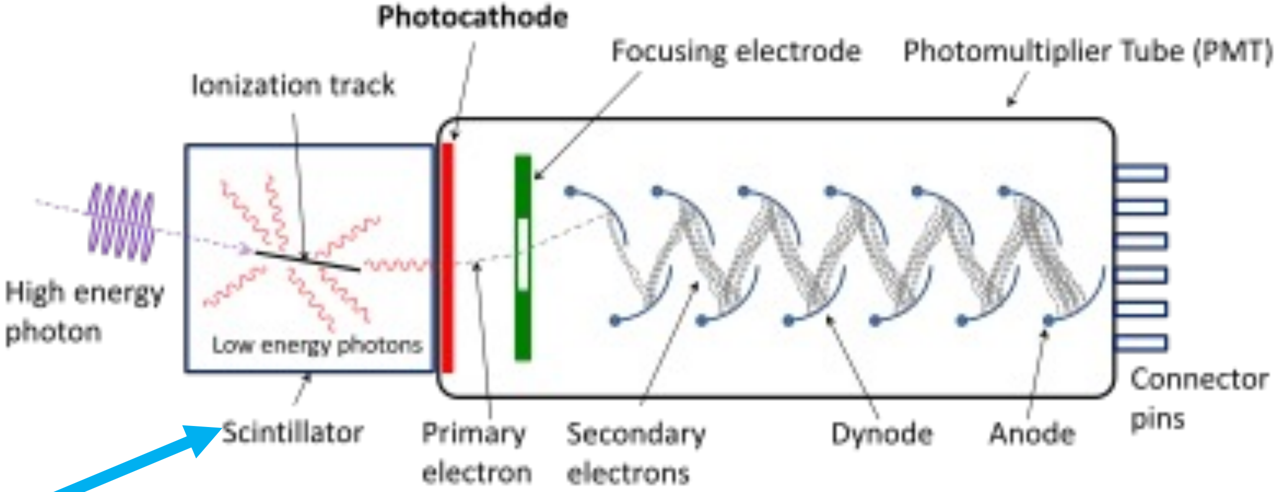
Scintillators – What are they again?

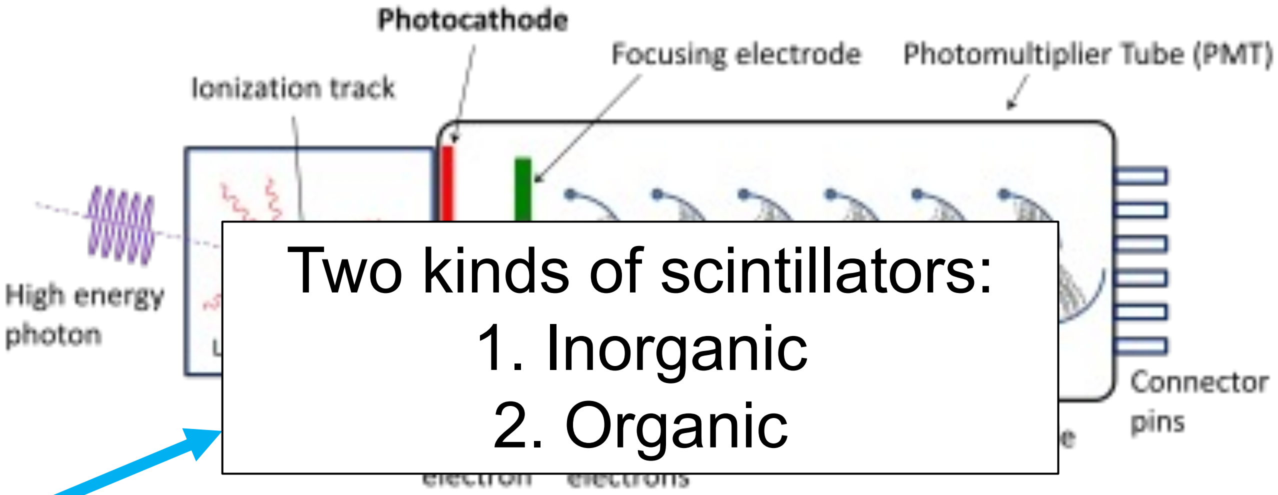


Scintillator

PMT

NaI(Tl) detector

Scintillators – What are they again?

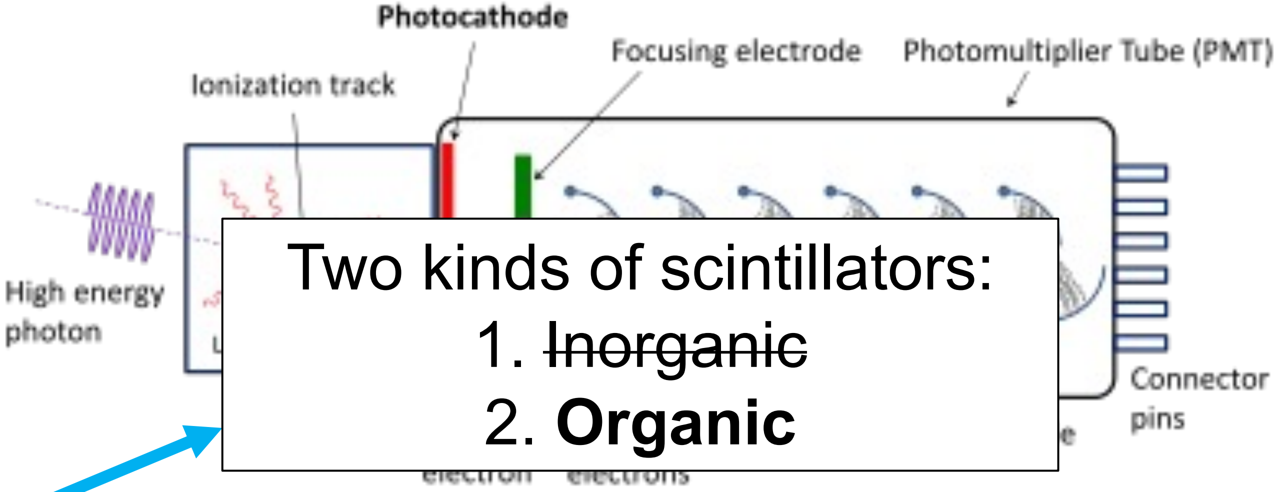


Two kinds of scintillators:

1. Inorganic
2. Organic

NaI(Tl) detector

Scintillators – What are they again?



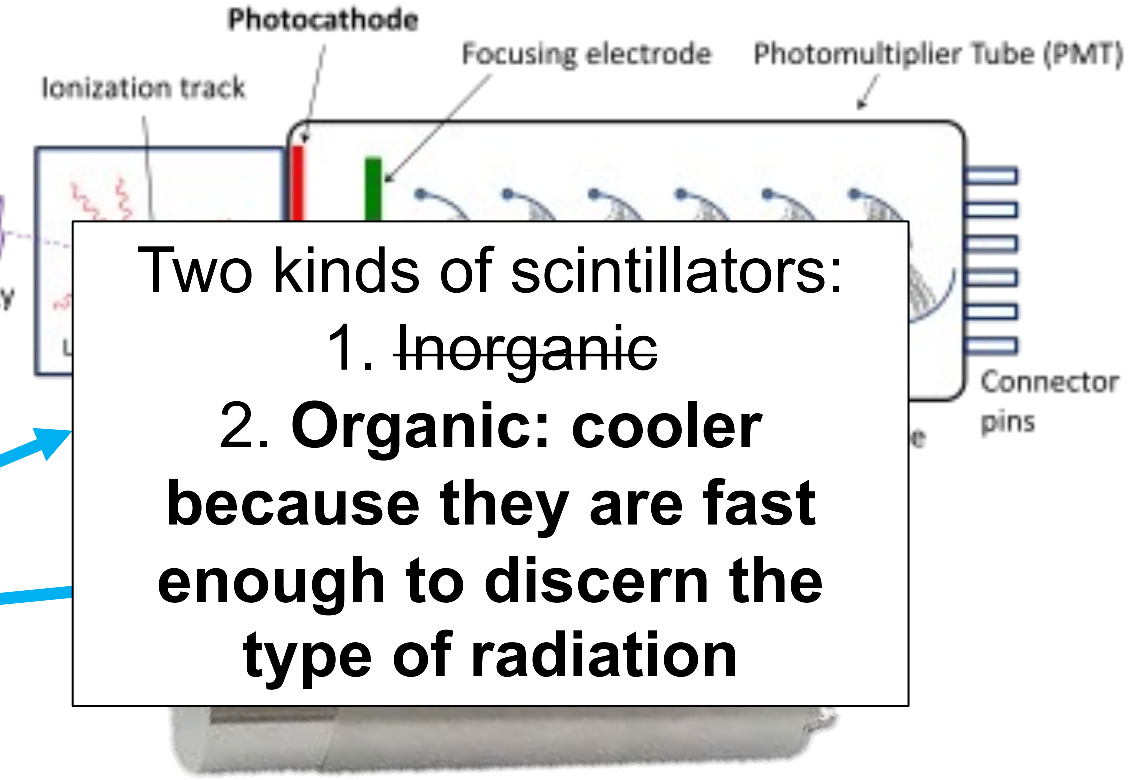
Two kinds of scintillators:

1. ~~Inorganic~~
2. **Organic**

Scintillator PMT

NaI(Tl) detector

Scintillators – What are they again?

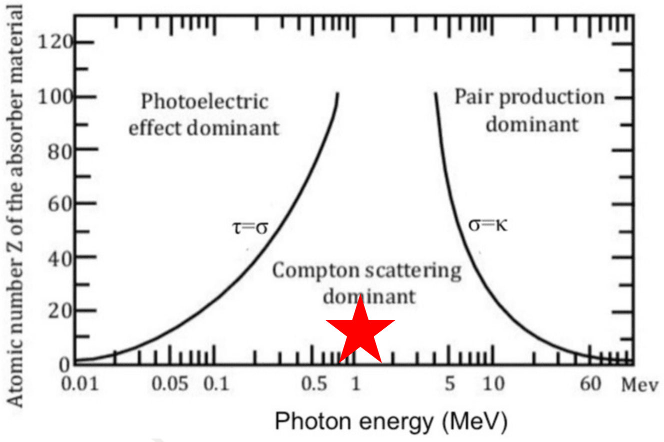


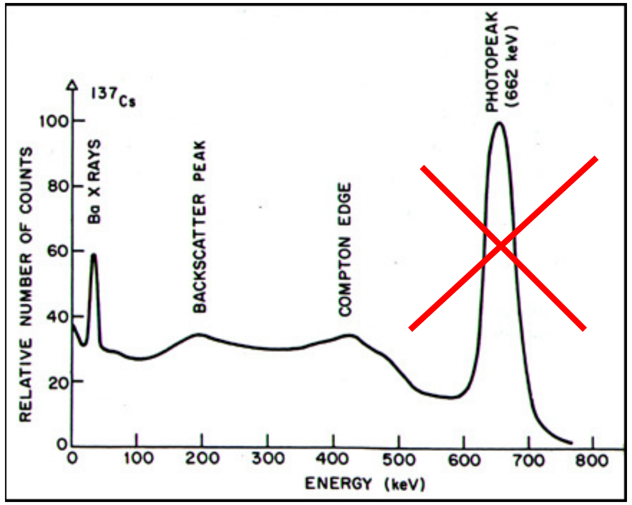
Two kinds of scintillators:

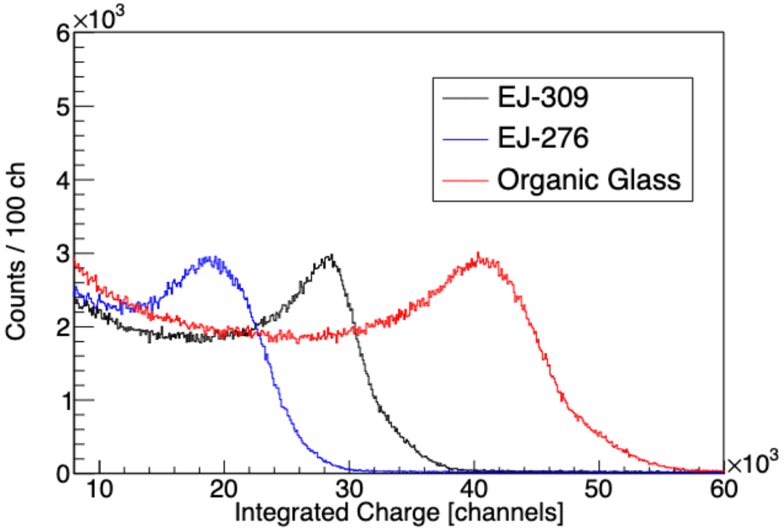
1. ~~Inorganic~~
2. **Organic: cooler because they are fast enough to discern the type of radiation**

NaI(Tl) detector

Metrics: Light Yield







Zaitseva, NIMA, Recent Developments of Plastic Scintillators

- Light yield: Photons emitted by scintillator per absorbed energy
  - Convention: 50% of Compton peak
  - Alternative: Location of most derivative

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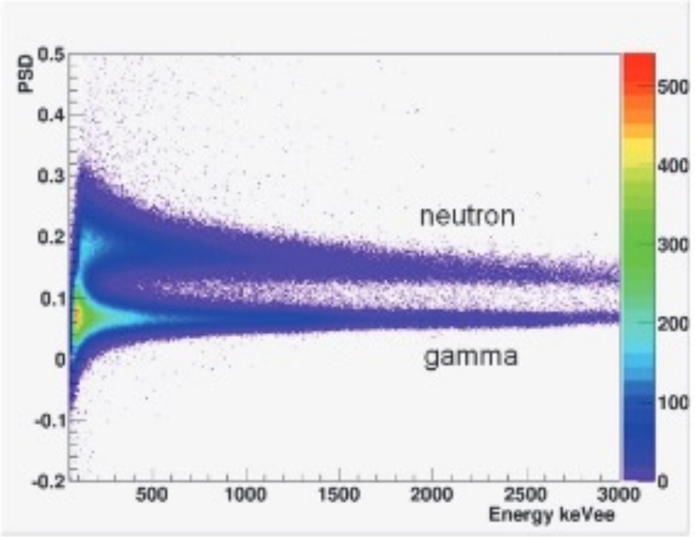
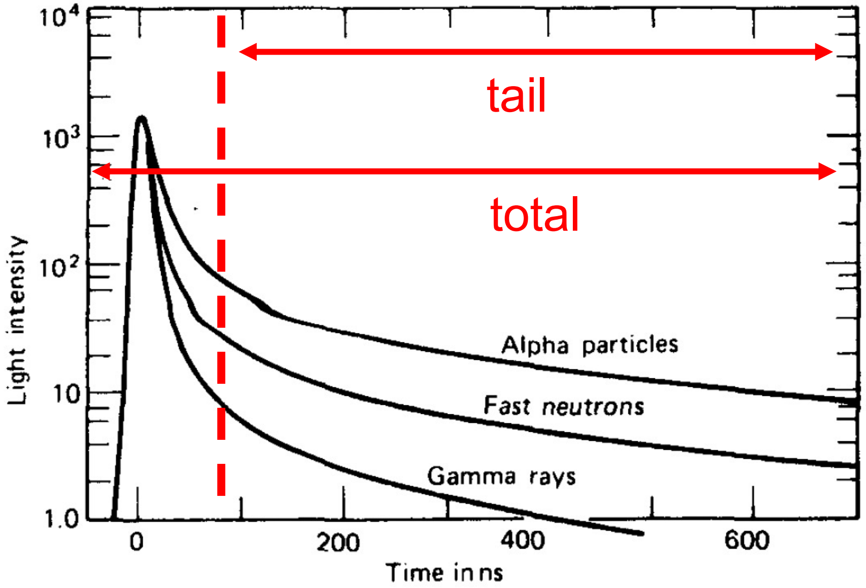
Metrics: Pulse Shape Discrimination

- Pulse shape discrimination (PSD)

$$PSD = \frac{Q_{tail}}{Q_{total}}$$

- PSD ϵ [0,1]





Ellis et al, Pulse Shape Discrimination for Homeland Security Applications, 2016

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When do we use plastic (organic) scintillators?

Organic scintillator for real-time neutron **dosimetry**
KA Beyer, A Di Fulvio, L Stolarczyk… - … protection **dosimetry**, 2018 - academic.oup.com
… an **organic scintillator** for spectrometry and **dosimetry** of out-of-field secondary neutrons from clinical proton beams. The detector consists of an EJ-299-34 crystalline **organic scintillator**, …
☆ Save 99 Cite Cited by 8 Related articles All 8 versions

Warhead verification as inverse problem: **Applications** of neutron spectrum unfolding from **organic-scintillator** measurements
CC Lawrence, M Febbraro, M Flaska… - Journal of Applied …, 2016 - aip.scitation.org
… -spectrum unfolding with **organic** scintillation detectors, with an aim at future **applications** in the … The unfolding capability of three different **organic scintillators** will be compared—the …
☆ Save 99 Cite Cited by 22 Related articles All 5 versions

[HTML] Digital pulse shape discrimination in **organic scintillators** for fusion **applications**
B Esposito, Y Kaschuck, A Rizzo, L Bertalot… - Nuclear Instruments and …, 2004 - Elsevier
… Stilbene and NE213 **organic scintillators** are commonly used for neutron and γ-ray detection … The importance of DPSD for fusion **applications** and its advantages with respect to analog …
☆ Save 99 Cite Cited by 77 Related articles All 9 versions

[HTML] Real-time source localisation by passive, fast-neutron time-of-flight with **organic scintillators** for facility-installed **applications**
V Astromskas, SC Bradnam, LW Packer… - Nuclear Instruments and …, 2021 - Elsevier
Fast neutron time-of-flight (ToF) has been used to characterise the location of a source of a mixed radiation field. Two EJ-309 **organic scintillators** and a fast, digital, data acquisition …
☆ Save 99 Cite Cited by 3 Related articles All 3 versions

[HTML] **Organic scintillators** with long luminescent lifetimes for radiotherapy dosimetry
AR Beierholm, LR Lindvold, CE Andersen - Radiation measurements, 2011 - Elsevier
… the **application** of temporal stem signal removal for fibre-coupled **organic scintillators** seems … dose measurements is insufficient if the **scintillator** luminescent lifetime is not significantly …
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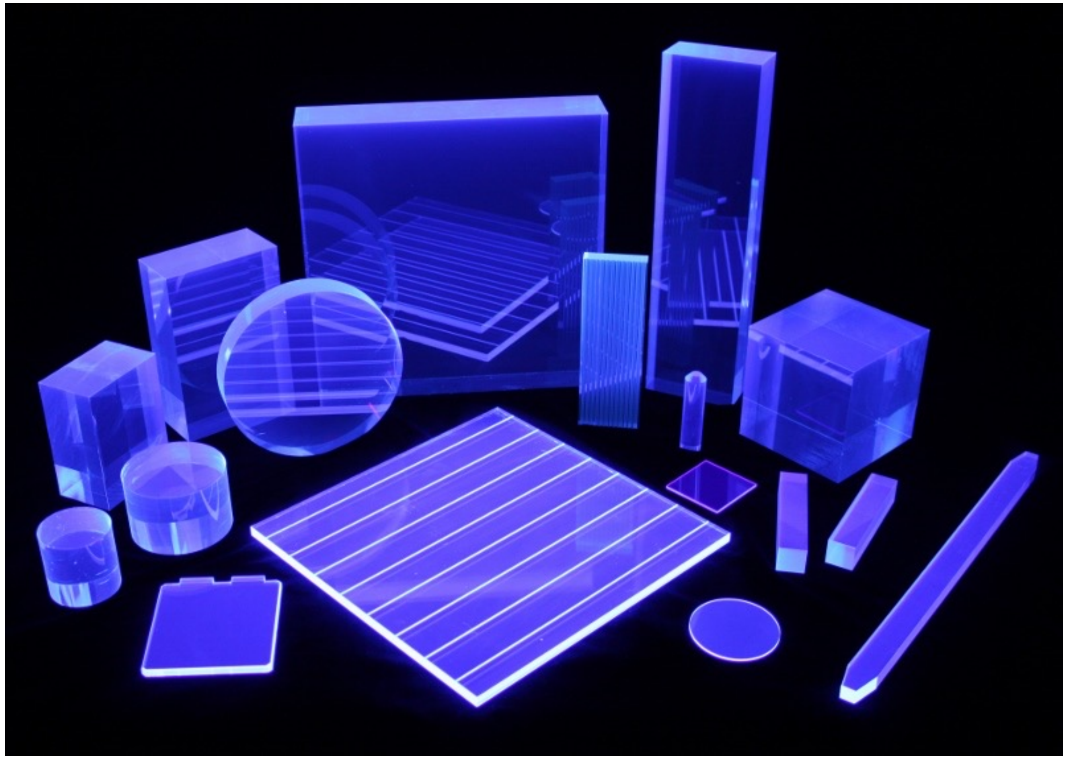
Aren’t we done with scintillator science?



Eljen

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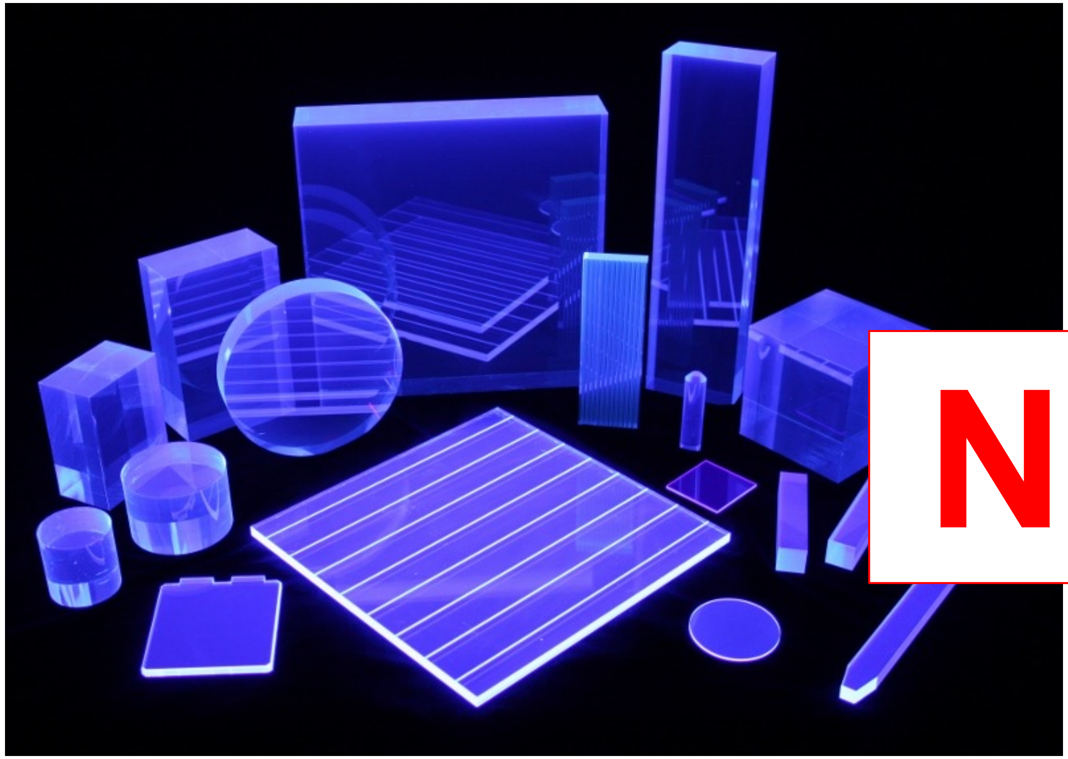
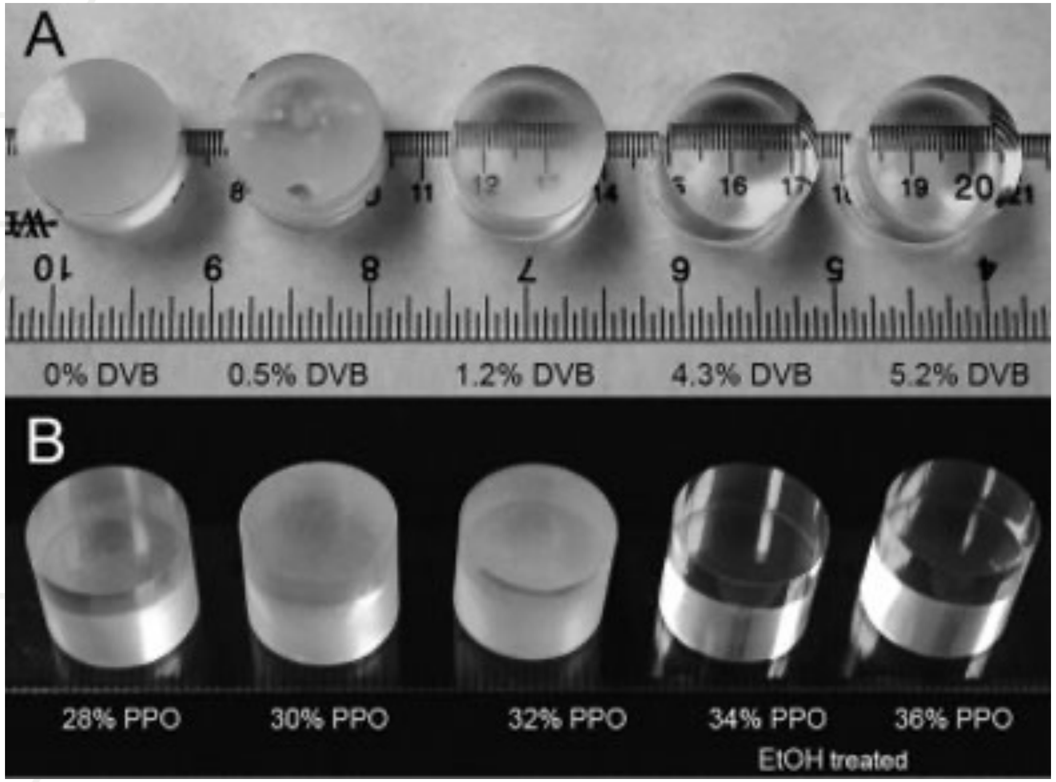
Aren't we done with scintillator science?



No

Eljen

Stability Issues in Plastic (PVT) Scintillators



Zaitseva, NIMA

- Eljen plastic scintillators made with PVT plastic matrix
- PVT requires overloading of dopant -> poor stability
- **30%** decrease in light yield in 6 months

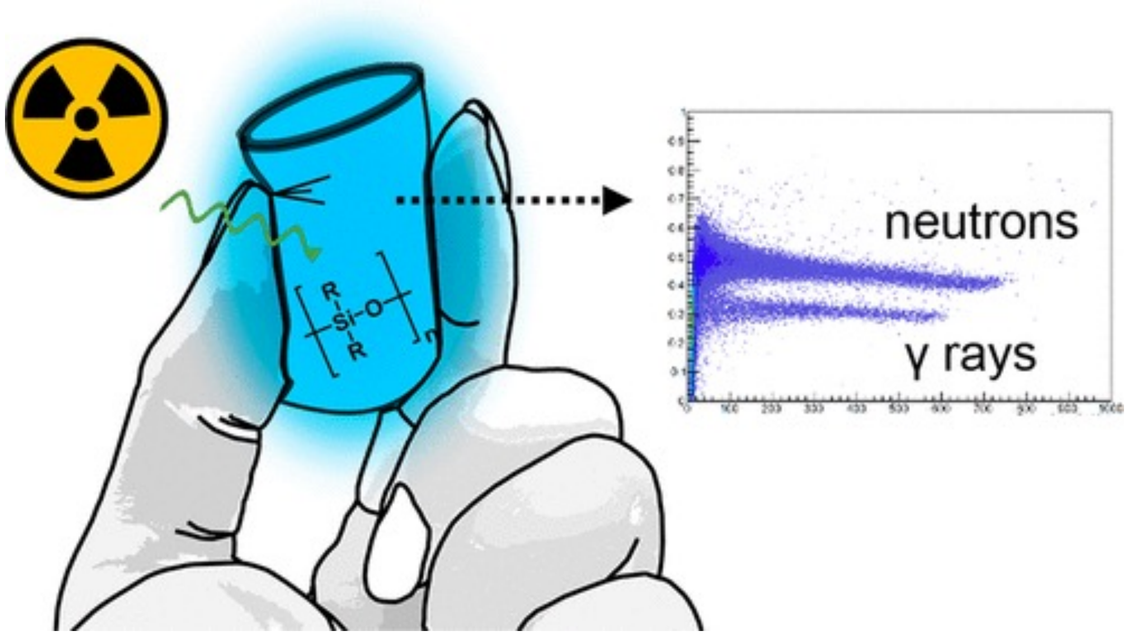
Polysiloxane Scintillators as an Alternative

	PVT	Polysiloxane
Transparency	Yes	Yes
Physical Properties	Hard, rigid	Variable
Radiation Hardness	No	Yes
Thermal Stability	No	Yes
Fabrication	5 days, air sensitive	3hrs, in air
PSD	@ 20wt% dopant	@ 5wt% dopant
TRL	9, deployed	4-5, lab work

https://www.sciencedirect.com/science/article/pii/S0254058412009376?via%3Dihub

Improving Polysiloxane Scintillators

- Goals:
  - Short processing time (3h in air)
  - Long term stability
  - Good pulse shape discrimination (PSD)
  - High light yield (LY)



ACS APPLIED POLYMER MATERIALS

Polysiloxane Scintillators for Efficient Neutron and Gamma-Ray Pulse Shape Discrimination

Allison Lim, Jonathan Arrue, Paul B. Rose, Alan Sellinger*, and Anna S. Erickson*

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A. Lim, J. Arrue, P. Rose, A. Sellinger,, A. Erickson, ACS Appl. Polym. Mater., 2020, 2, 8, 3657-3662

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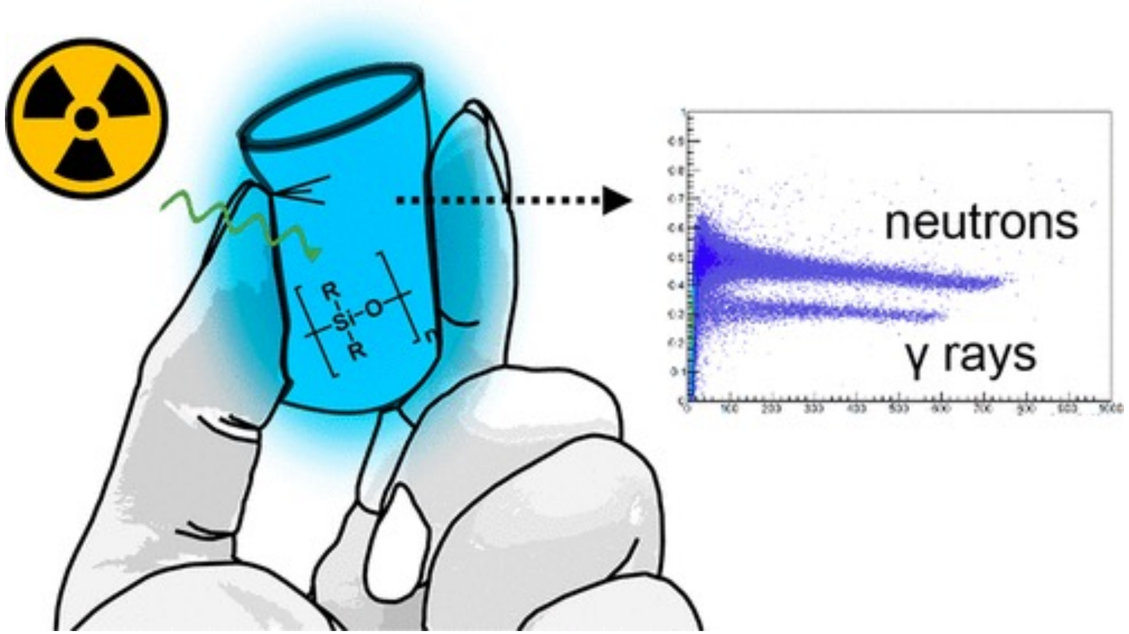
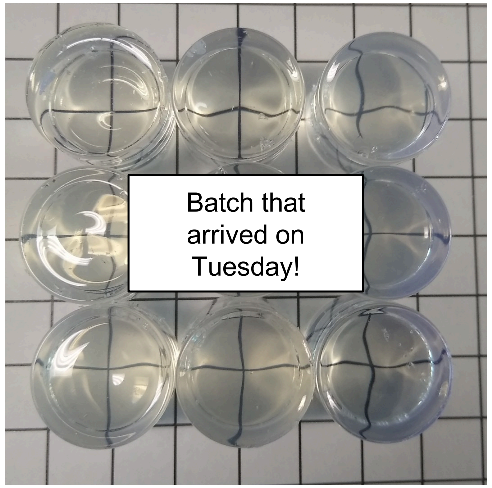
Improving Polysiloxane Scintillators: Boron Loading

- Goals:
  - Short processing time (3h in air)
  - Long term stability
  - Good pulse shape discrimination (PSD)
  - High light yield (LY)
  - Construct a three particle imager



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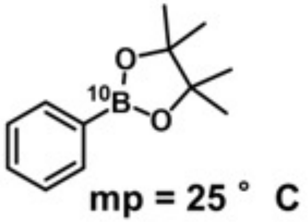
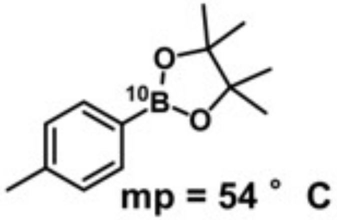
Boron Loading

- Boron-10 enriched molecules

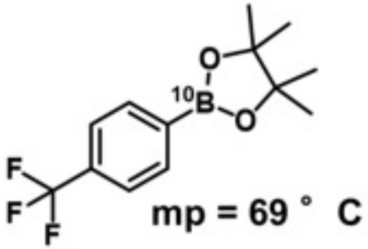
3.75wt% phenylpinacolborane[1]



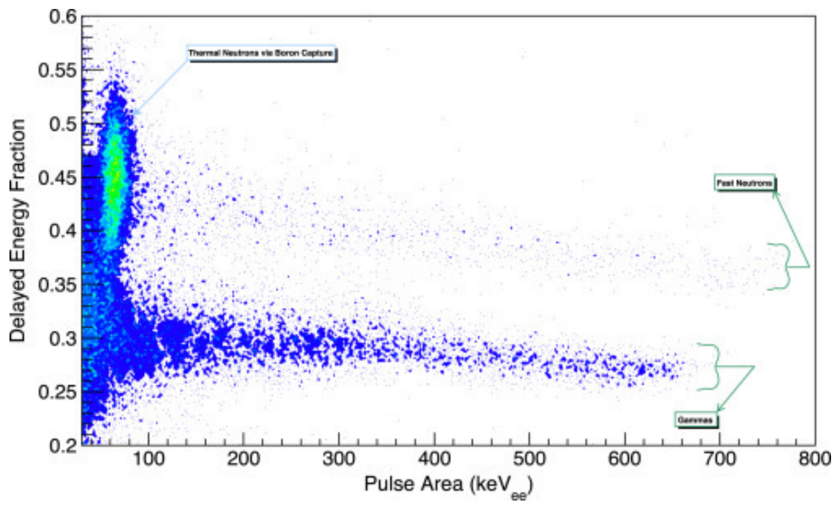
3wt% tolyl-



5wt% trifluoro-



$$
{}^{10}B + n_{th} \rightarrow \begin{cases} {}^{7}Li_{(1.01\ \mathrm{MeV})} + \alpha_{(1.78\ \mathrm{MeV})} \\ Q = 2.790\ \mathrm{MeV}, \quad 6.3\% \\ \\ {}^{7}Li^{*}_{(0.84\ \mathrm{MeV})} + \alpha_{(1.47\ \mathrm{MeV})} \\ \hookrightarrow {}^{7}Li^{*} \rightarrow {}^{7}Li + \gamma_{(478\ \mathrm{keV})} \\ Q = 2.310\ \mathrm{MeV}, \quad 93.7\% \end{cases}
$$



Previous work with phenyl-derivative for thermal neutron detection with PVT matrix, PPO, POPOP

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[1]A. Mahl, H.A. Yemam, R. Fernando, J.T. Koubek, A. Sellinger, U. Greife, NIM A, 2018, 880, 1-5

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Metrics: PSD/FOM

- Pulse shape discrimination (PSD)

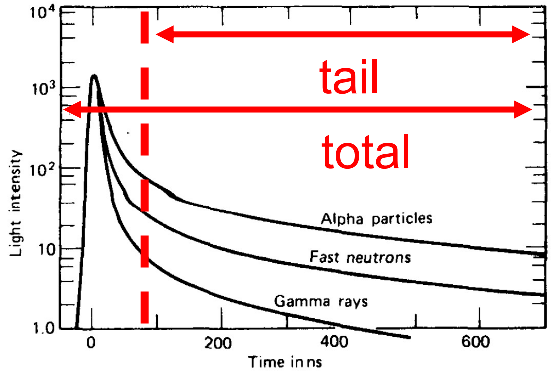
$$PSD = \frac{Q_{tail}}{Q_{total}}$$

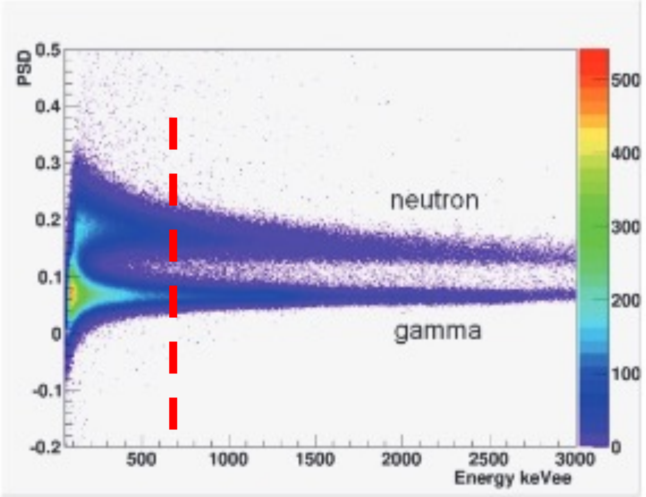
- PSD ϵ [0,1]

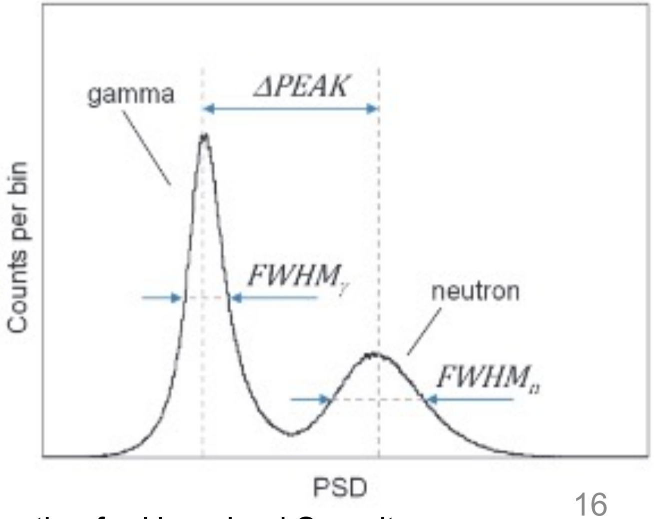
- S = separation of n and gamma lobe
- Figure of merit (FoM)

$$FOM = \frac{S}{FWHM_{\gamma} + FWHM_{n}}$$

  - “Efficient” FOM at 1.27



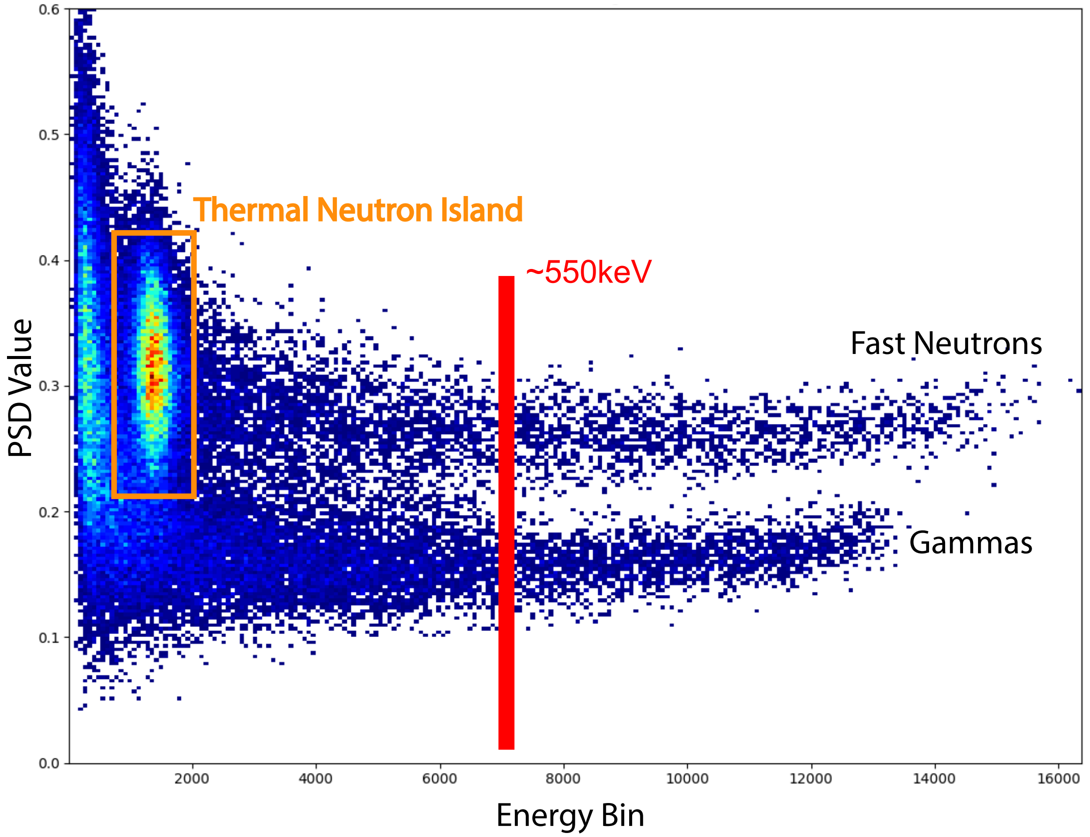




Ellis et al, Pulse Shape Discrimination for Homeland Security Applications, 2016

Successful Thermal Neutron Sensitivity

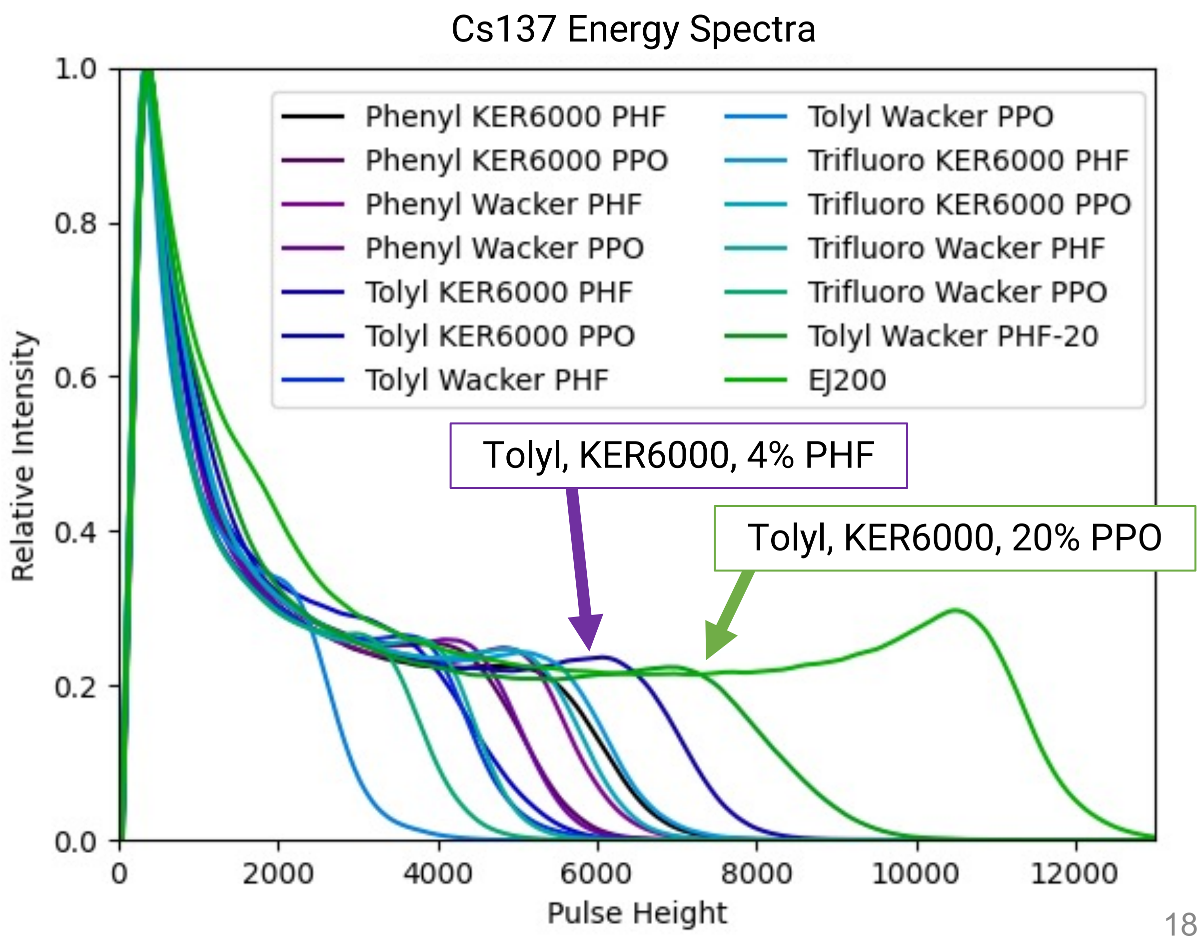
Polysiloxane PSD Heatmap (Tolyl-b10, PHF, KER6000)



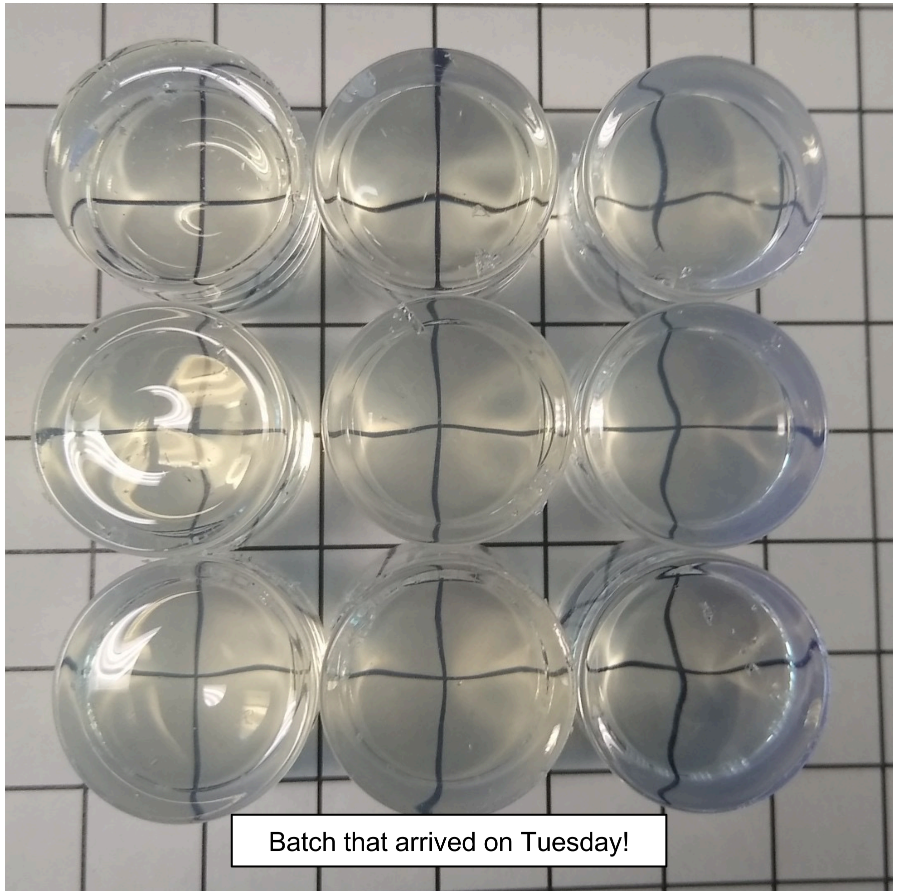
		Thermal Neutron Island FoM						~550keV FoM	
		Gamma-Thermal		Fast-Thermal		Gamma-Fast		Gamma-Fast	
B-10	Matrix	5% PHF	5% PPO	5% PHF	5% PPO	5% PHF	5% PPO	5% PHF	5% PPO
5% Phenyl	KER6000	0.56	0.57	0.34	0.36	0.85	0.85	1.09	0.81
	Wacker	x	0.47	x	0.36	x	0.76	0.69	x
4% Tolyl	**KER6000**	**0.58**	**x**	**0.38**	**x**	**0.89**	**x**	**1.17**	**0.78**
	Wacker	x	x	x	x	0.34	0.36	0.64	0.62
5% Trifluoro	**KER6000**	**0.58**	**0.61**	**0.35**	**0.35**	**0.85**	**0.89**	**1.10**	**1.21**
	Wacker	x	0.45	x	0.36	x	0.73	0.56	0.69

Boron-Doped Polysiloxanes: Light Yield

		Light Yield (%EJ200)	
B-10 enriched molecule	Matrix	5% PHF	5% PPO
5% Phenyl-	KER6000	53	45
	Wacker	49	45
4% Tolyl-	KER6000	**62**	39
	Wacker	39	23
5% Trifluoro-	KER6000	55	51
	Wacker	39	33



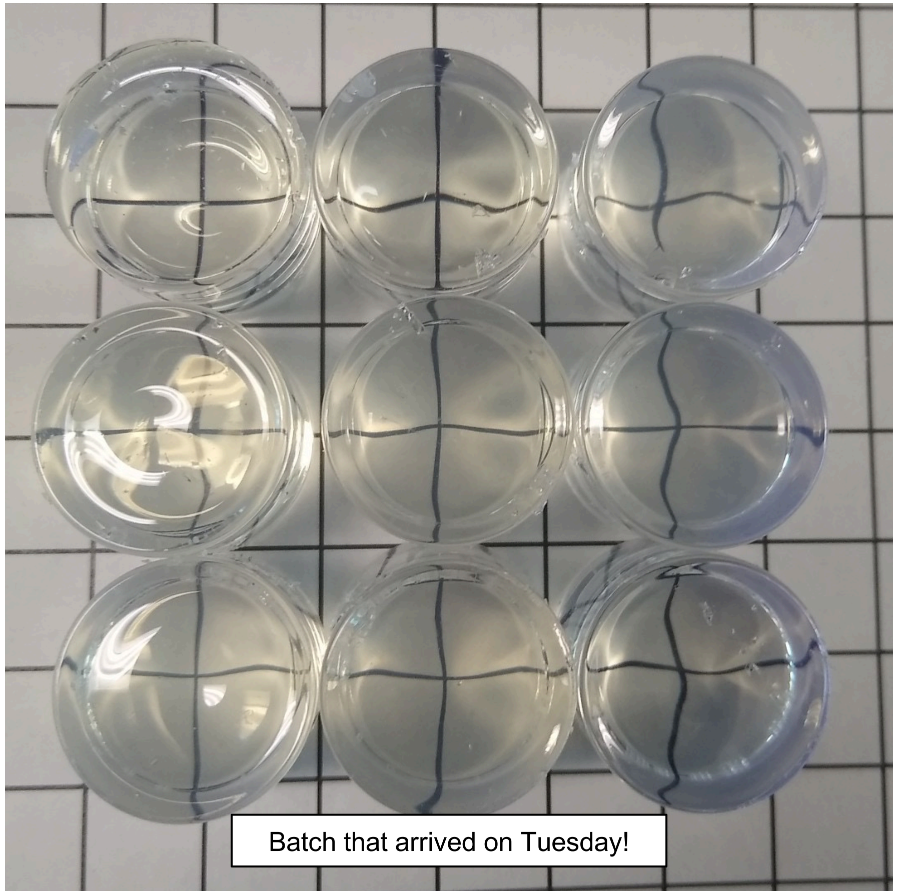
Best Array Candidate: Tolyl-, KER6000, PHF



Batch that arrived on Tuesday!

- Thermal Island FOM:
  - Gamma-Thermal: 0.58
  - Fast-Thermal: 0.38
  - Gamma-Fast: 0.89
- Cs-137 Compton Edge FOM:
  - 1.17
- LY: 62% EJ200

Best Array Candidate: Tolyl-, KER6000, PHF



- Thermal Island FOM:
  - Gamma-Thermal: 0.58
  - Fast-Thermal: 0.38
  - Gamma-Fast: 0.89
- Cs-137 Compton Edge FOM:
  - 1.17
- LY: 62% EJ200
- Challenge: Separating contributions from each particle

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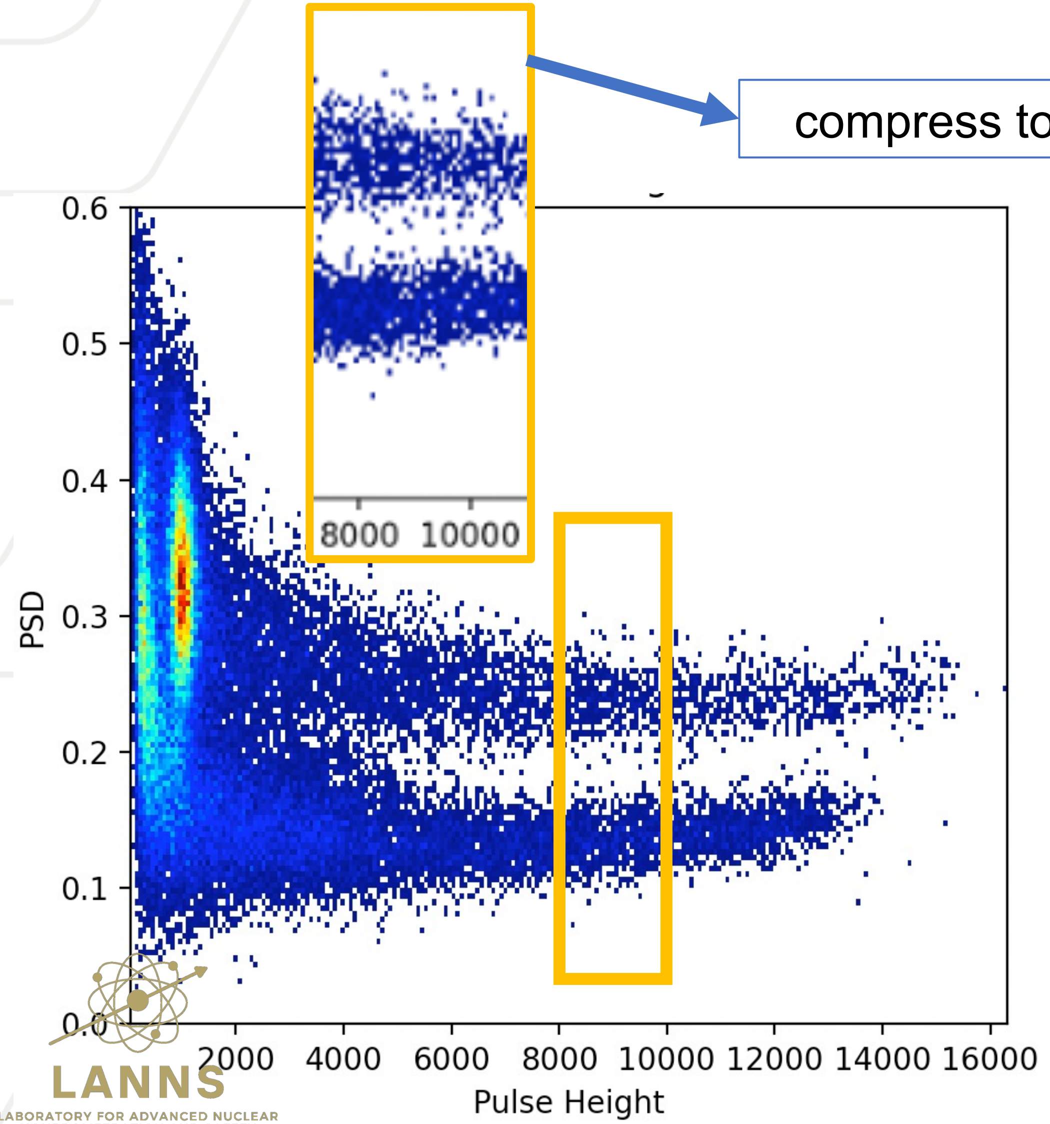
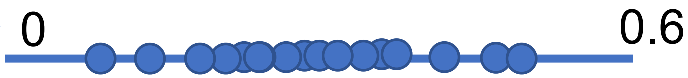
Gaussian Mixture Model: Fitting Routine

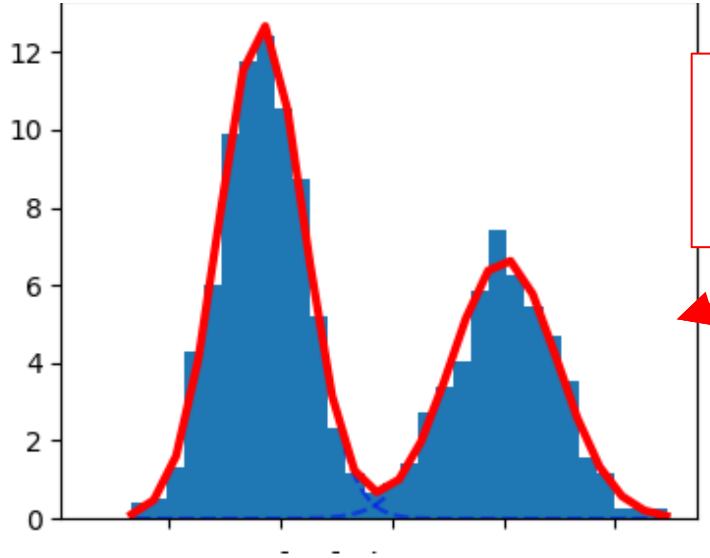


compress to 1d



PSD

as a histogram



apply gaussian mixture model to 1D data

plot GMM gaussian fits (red) on data (blue)

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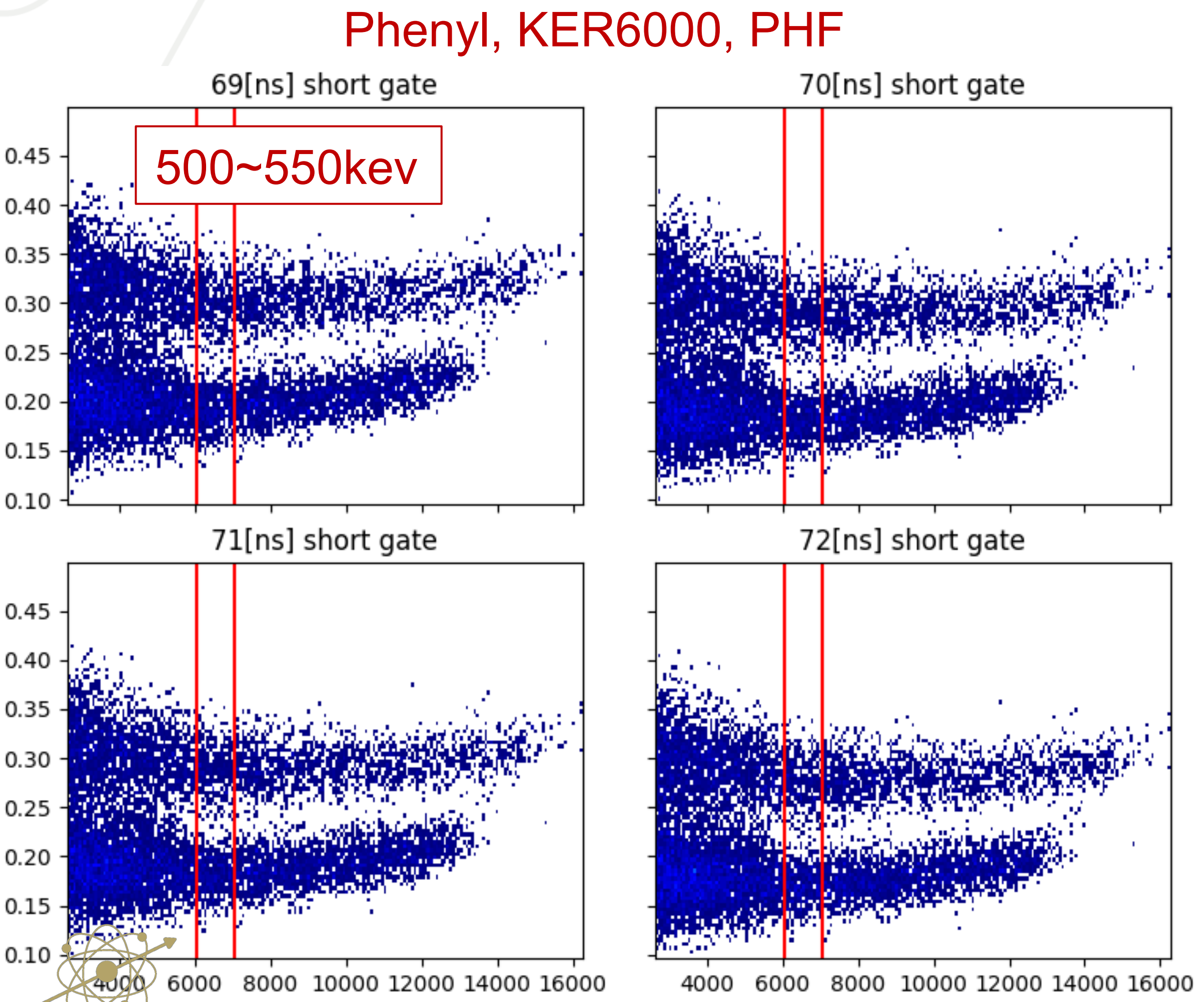
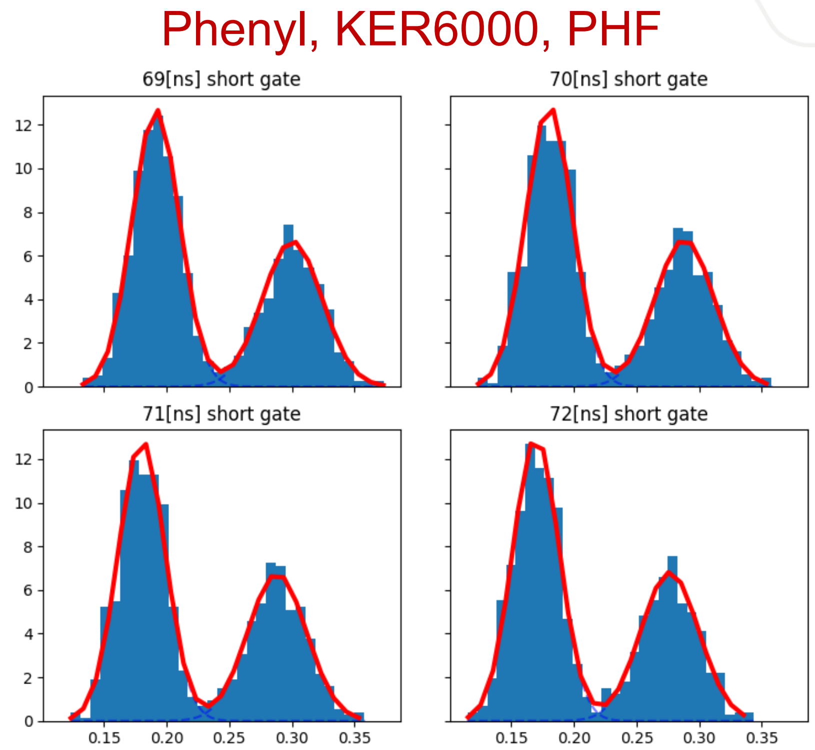
Cs137 Compton Edge: 2 Gaussians





Gate	Means	Stds	FWHMs	FoM	R^2
69	0.191, 0.300	0.019, 0.024	0.044, 0.064	1.08	**0.96**
70	0.180, 0.288	0.019, 0.023	0.044, 0.055	1.09	**0.95**
71	0.180, 0.288	0.019, 0.023	0.044, 0.055	1.09	**0.95**
72	0.170, 0.275	0.018, 0.023	0.043, 0.055	1.09	**0.93**

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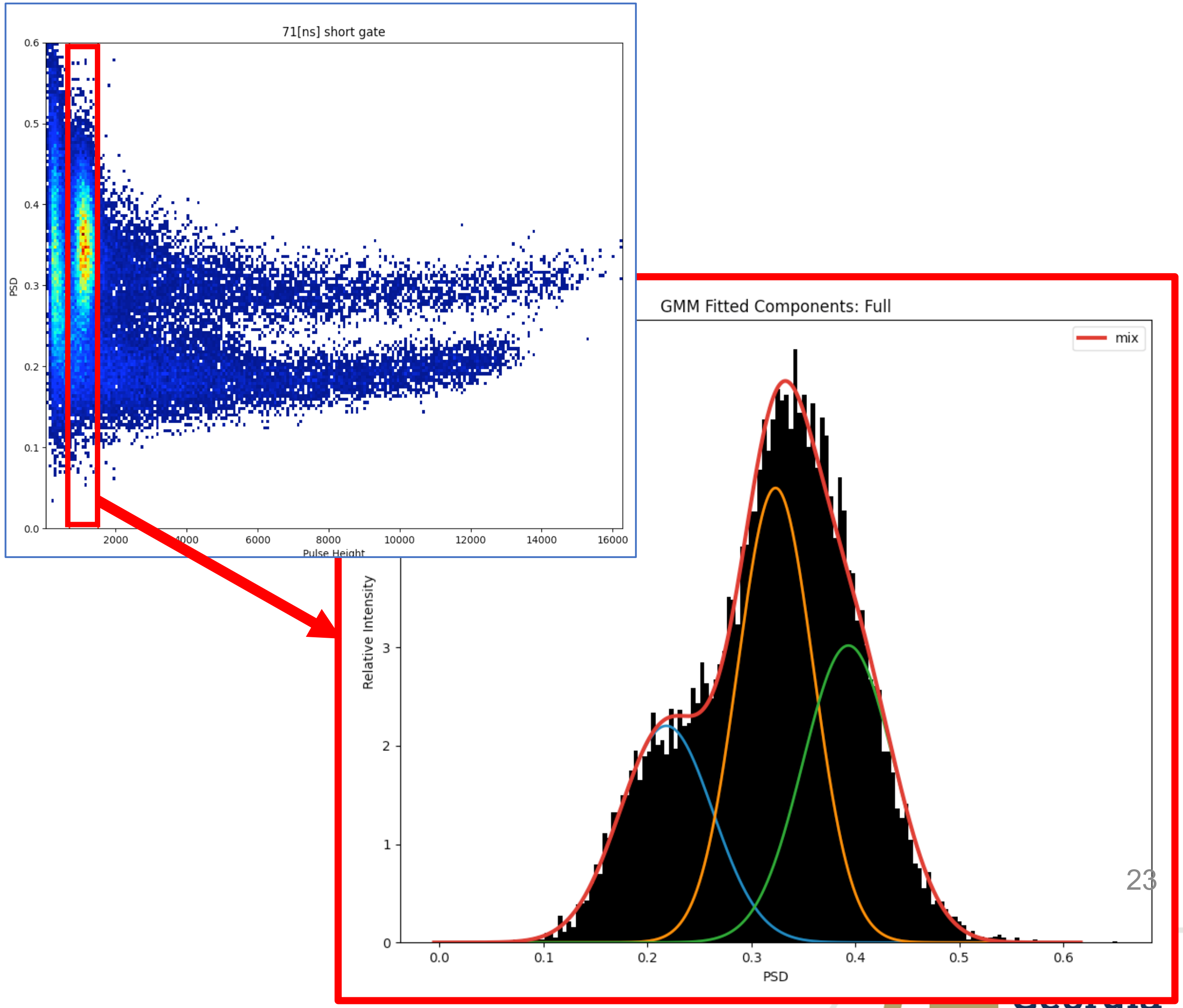
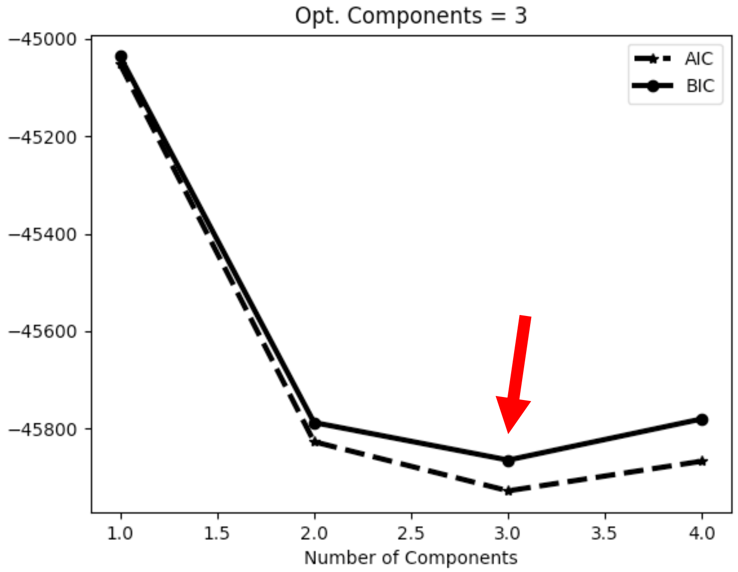
Thermal Neutron Island: 3 Gaussians

- Fit results for Phenyl, KER6000, PhF sample’s thermal neutron island.
- **FOMs**
  - **nfast, nthermal = 0.59 +/- 0.01**
  - **nfast, gamma = 0.84 +/- 0.01**
  - **nthermal, gamma = 0.37 +/- 0.01**





Conclusions & Future Work

Conclusions

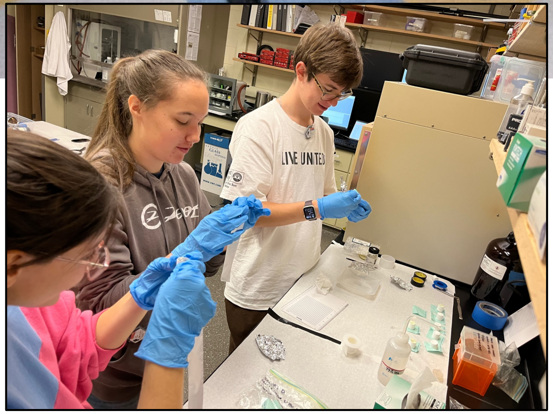
- Found a good scintillator candidate for three particle imaging
- Developed strategy for separating particle contributions
- New samples for array construction have arrived

Future Work

- Construct array, physically
- Particle separation strategy may need to account for gamma from boron-capture interaction
- Error of GMM must be more closely examined
- Develop training set and keep GMM through iterations
- Implement Luke Maloney's image reconstruction algorithm

Thank you

- LANNS group members supporting polysiloxane work
  - Alex England
  - Ian Schreiber
  - **Jana Shade**
  - **Pierre O'Driscoll**
  - **Anna Schafer**
  - Dr. Anna Erickson
  - Dr. Yuguo Tao
- Caleb Chandler, Dr. Allen Sellinger



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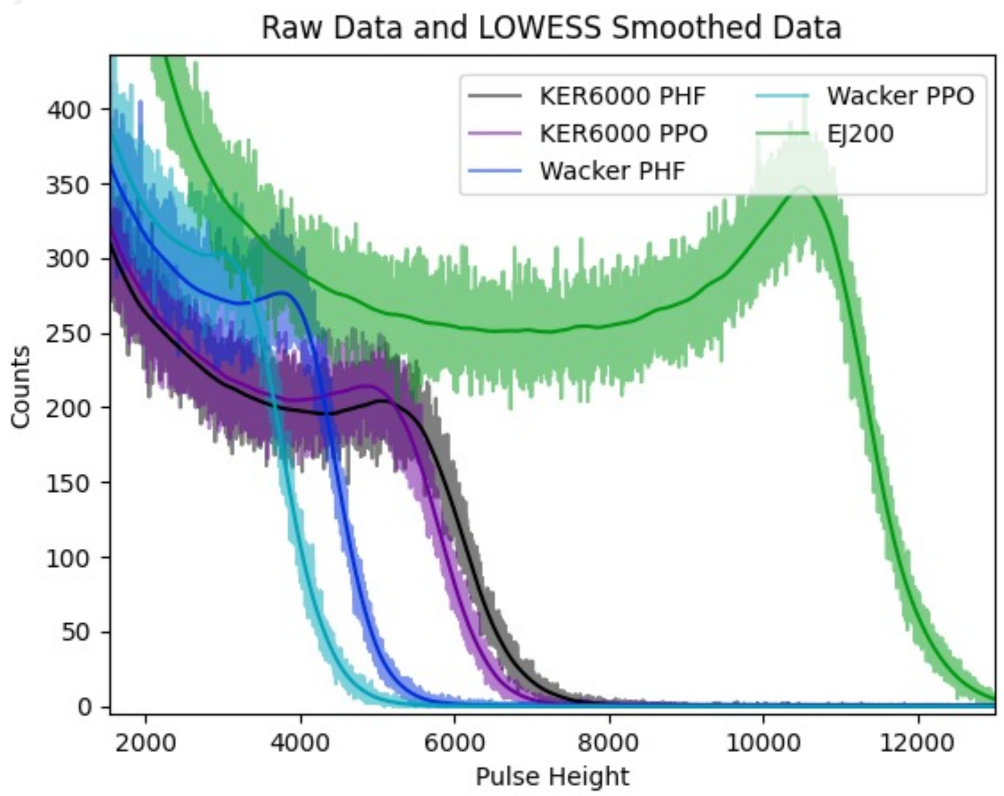
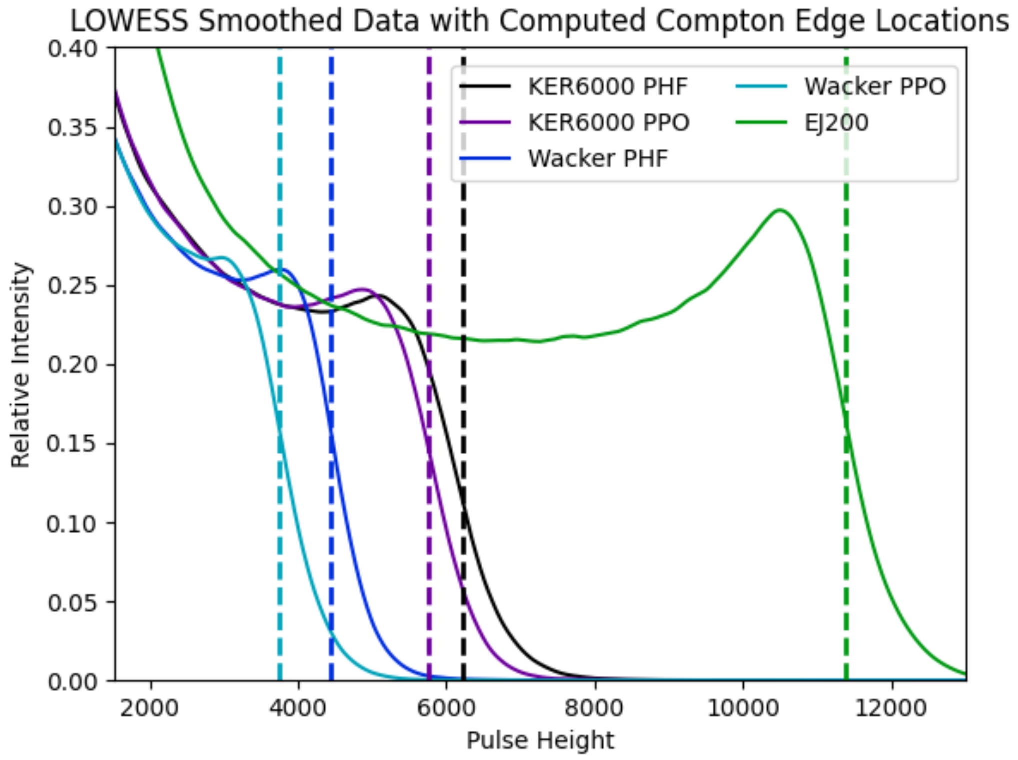
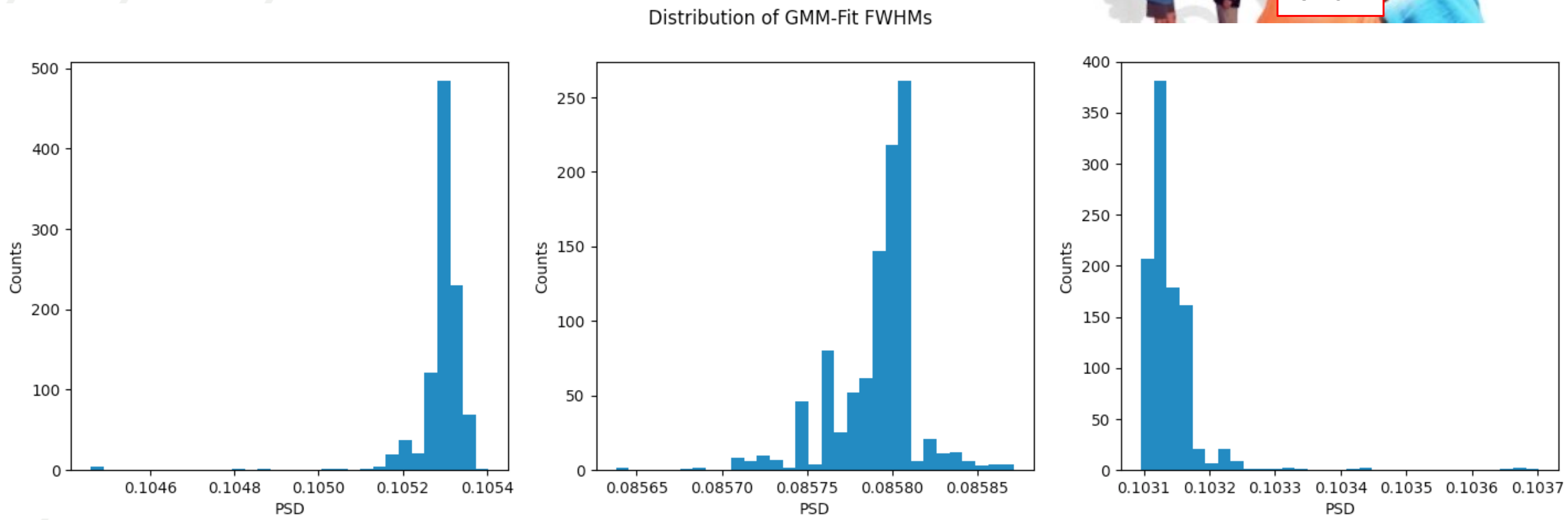
Back Up

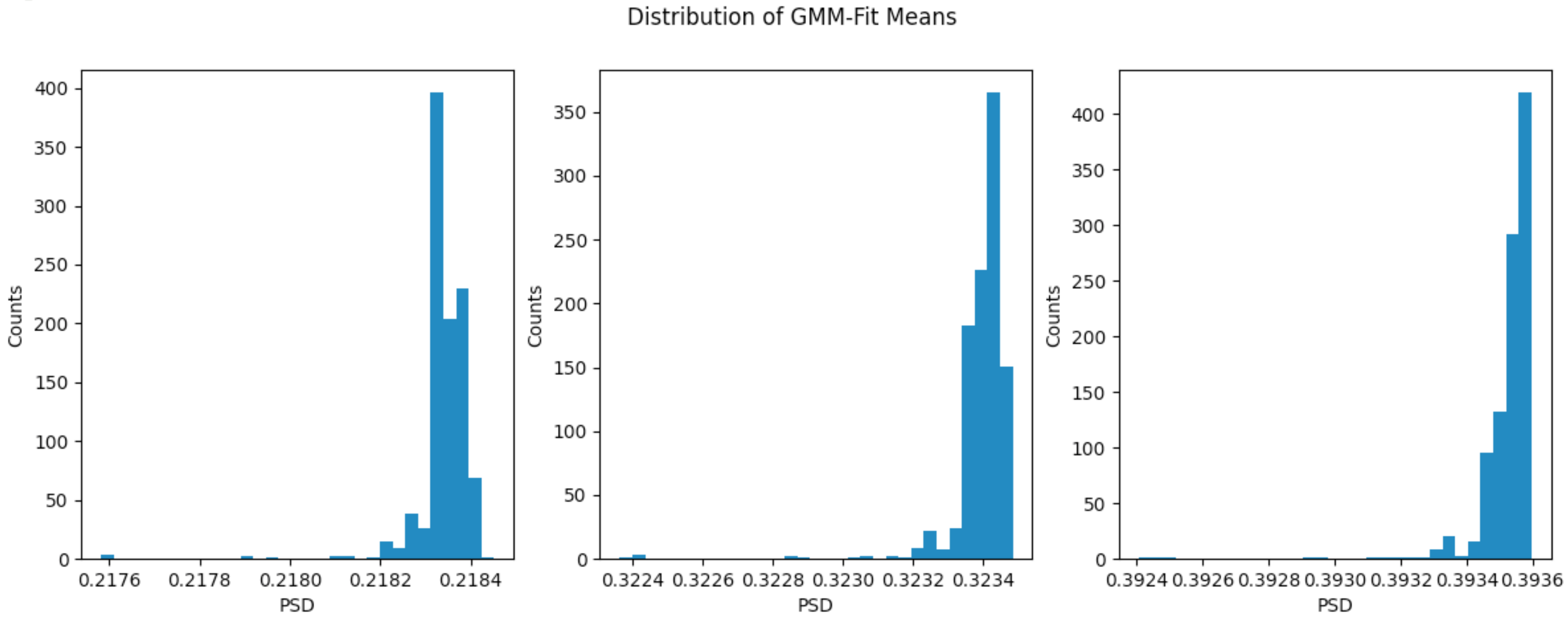
Light Yield – Compton Edge





Bootstrapping

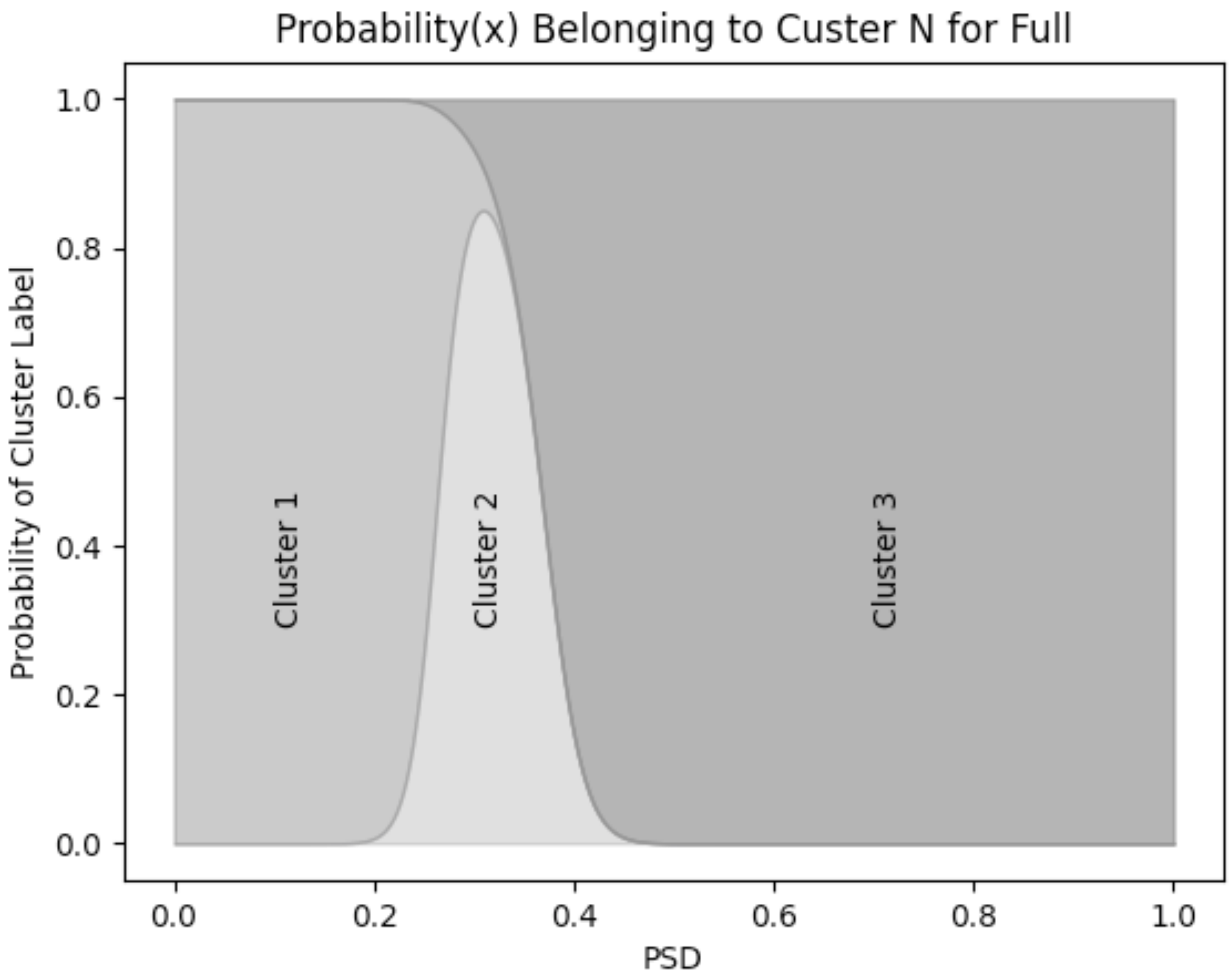




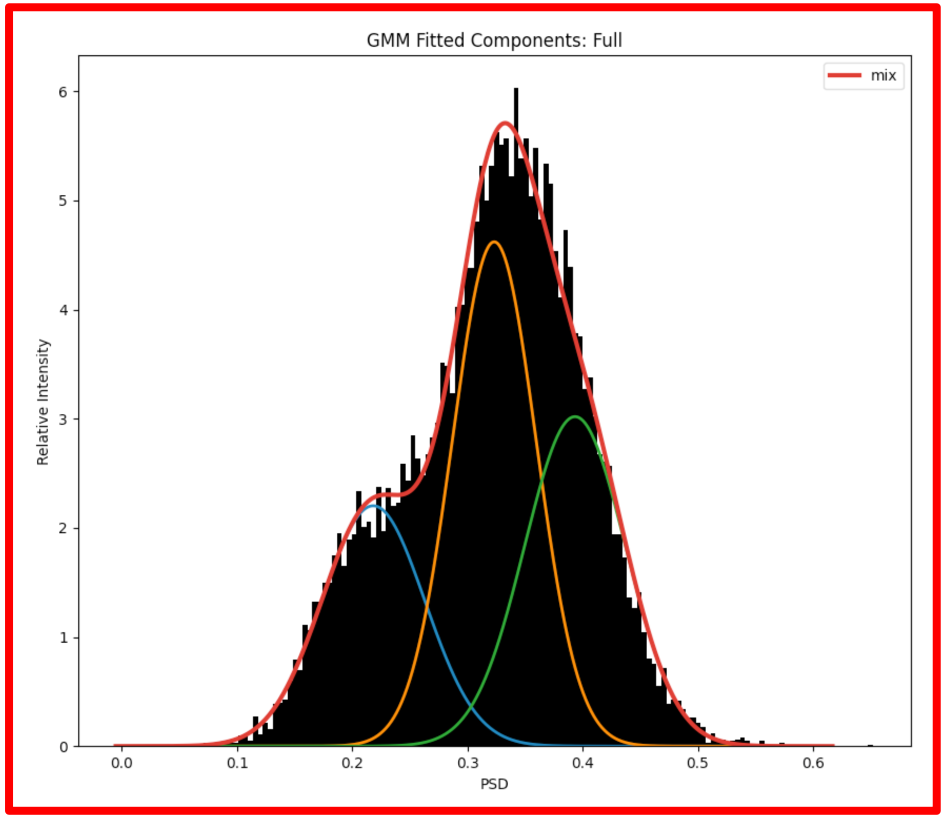
- Ran 1,000 GMM fits seeding with a random number each time on the full dataset and recorded the:
  - means of the gaussian fits
  - fwhms of the gaussian fits
- Results
  - The smallest and largest of the values and the difference between them. Take difference as the error on FOM
- **FOMs**
  - **nfast, nthermal = 0.598 +/- 0.008**
  - **nfast, gamma = 0.840 +/- 0.008**
  - **nthermal, gamma = 0.371 +/- 0.008**

```
Smallest Value, Largest Value, Difference For Component 1
Mean: 0.21845162257846895 0.21758129093607434 [0.00087033]
FWHM:  0.10540377188715863 0.10445907039499769 [0.0009447]
Smallest Value, Largest Value, Difference For Component 2
Mean:  0.3234839645169943 0.3223623305385313 [0.00112163]
FWHM: 0.08587132918822132 0.08563786531844203 [0.00023346]
Smallest Value, Largest Value, Difference For Component 3
Mean: 0.39359613409664856 0.39240555859781895 [0.00119058]
FWHM:  0.10370180192759926 0.10309565876408398 [0.00060614]
```

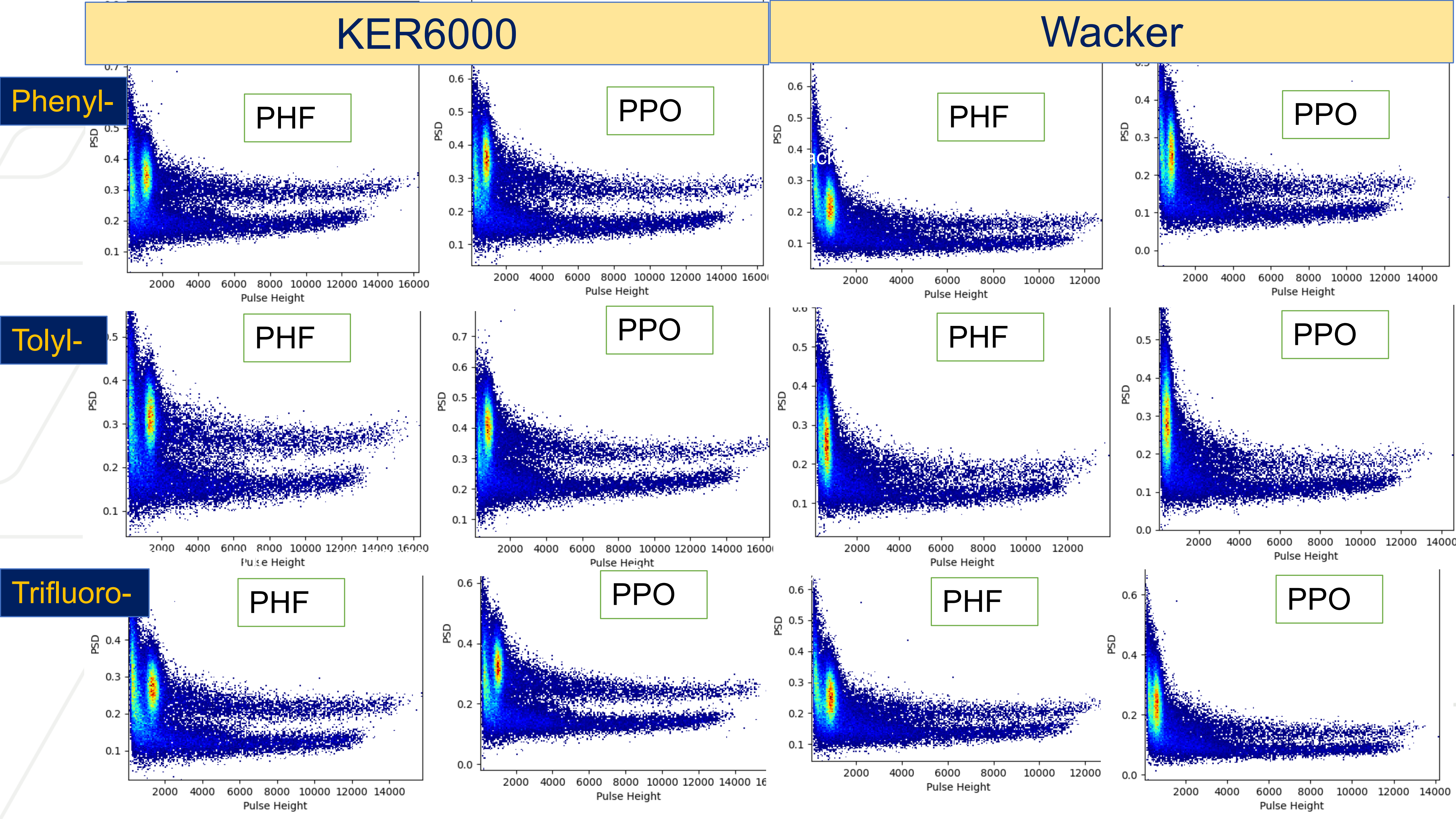
Clustering



Probability(x) Belonging to Custer N for Full
Probability of Cluster Label
1.0
0.8
0.6
0.4
0.2
0.0
Cluster 1
Cluster 2
Cluster 3
0.0
0.2
0.4
0.6
0.8
1.0
PSD



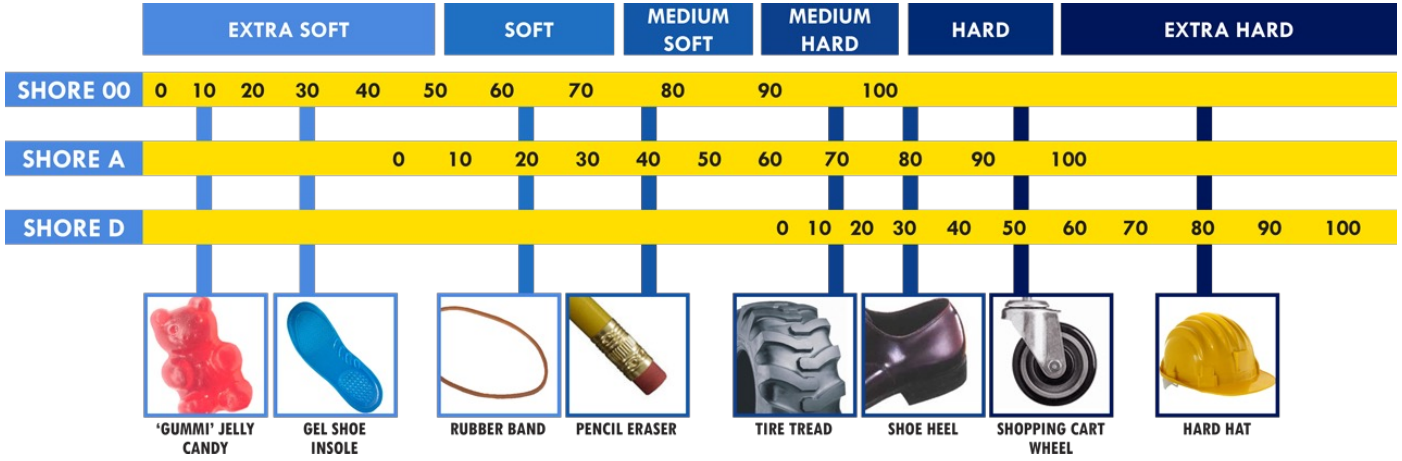
GMM Fitted Components: Full
mix
Relative Intensity
PSD

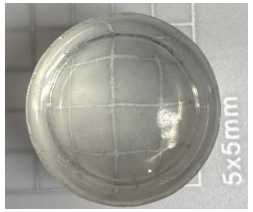

KER6000
Wacker
Phenyl-
Tolyl-
Trifluoro-
PHF
PPO
PHF
PPO
PHF
PPO
PHF
PPO
PHF
PPO
PHF
PPO
PSD
Pulse Height

CSM-GT Polysiloxanes (Matrices)

- Polymer resins used:
  - Wacker Lumisil 579
    - LED encapsulant
  - Shin-Etsu KER-6000
    - LED encapsulant
  - Wacker SILRES H62C
    - electronics encapsulant

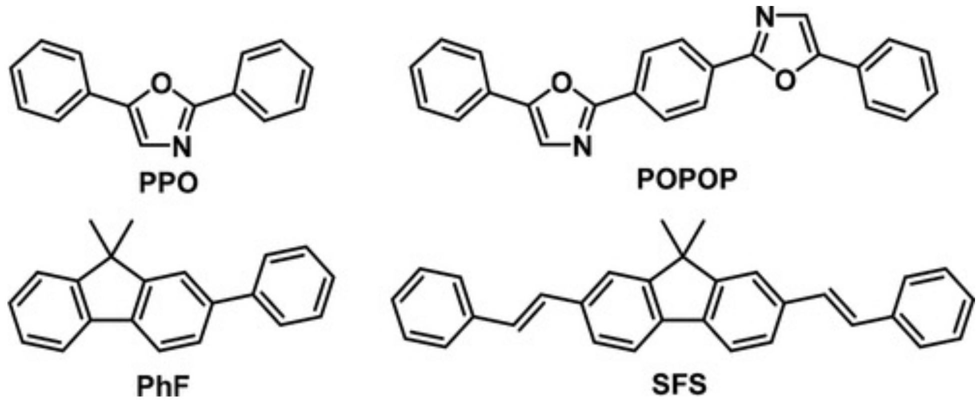
	R.I.	Hardness	Mix	Cure
Wacker 579	1.53	25 Shore A	2-component	150 °C / 1 hr
KER-6000	1.51	22 Shore A	2-component	100 °C / 1 hr, 150 °C / 2 hr
SilRes H62C	1.50	65 Shore D	1-component	150 °C / 10 hr

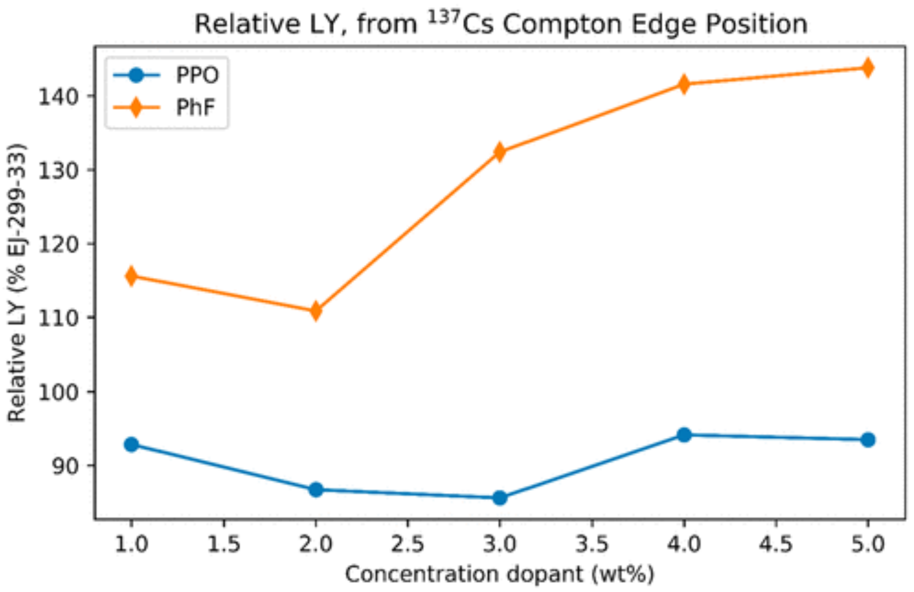




CSM-GT Polysiloxanes (Dopants)

- Primary dopants (fluorophore):
  - 9,9-dimethyl-2-phenyl-9H-fluorene (PHF) : Custom fluorophore developed by Colorado
  - 2,5-diphenyloxazole (PPO) : Industry-standard fluorophore
- Secondary dopant (wavelength shifter):
  - 9,9-dimethyl-2,7-distyryl-9H-fluorene (SFS)





A. Lim, J. Arrue, P. Rose, A. Sellinger,, A. Erickson, ACS Appl. Polym. Mater., 2020, 2, 8, 3657-3662

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Fabrication Process (Colorado School of Mines)

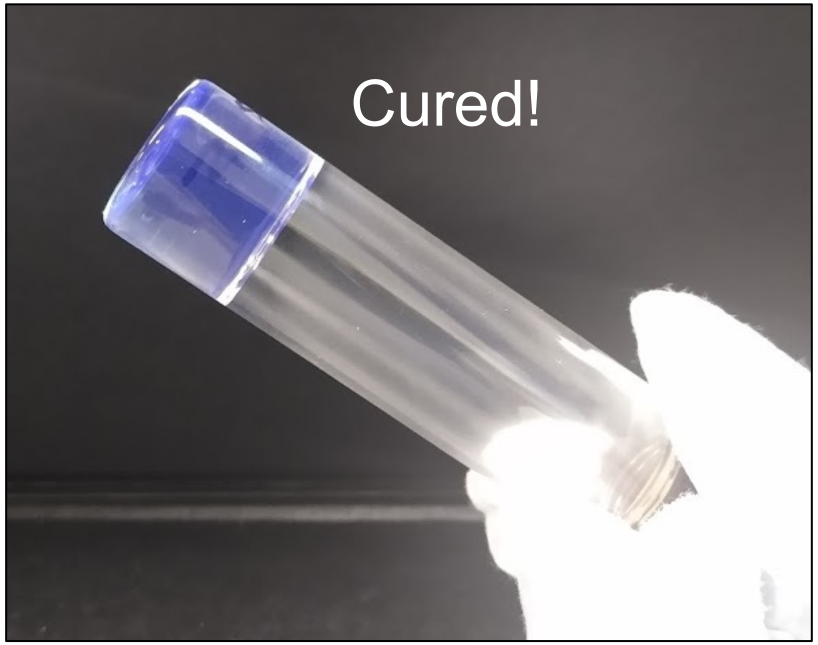
- Thanks: Caleb Chandler, Alan Sellinger

Prepare silanized vial (30-60 mins)

Add SFS, PPO

Add xylenes

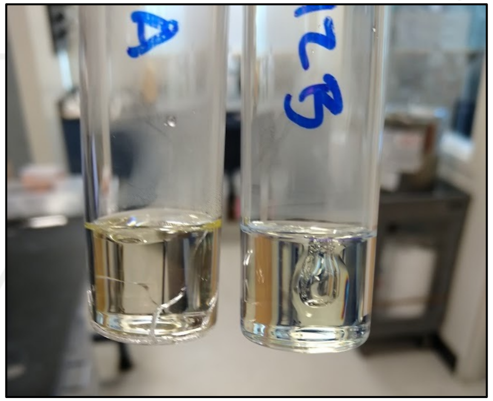
Solubilize and add Part A, mix, add Part B



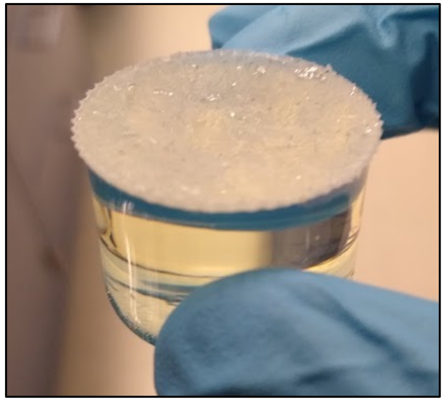
Vortex, then cure at 150 C for 3 hrs in air

Common Fabrication Issues

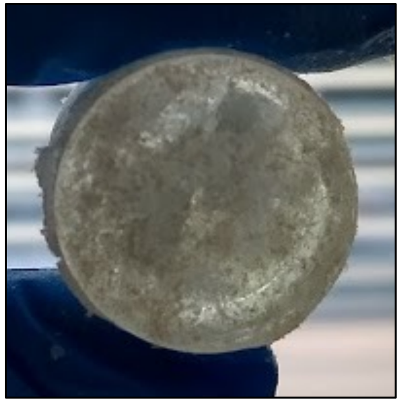
Glass Cracked
Internal Bubbling



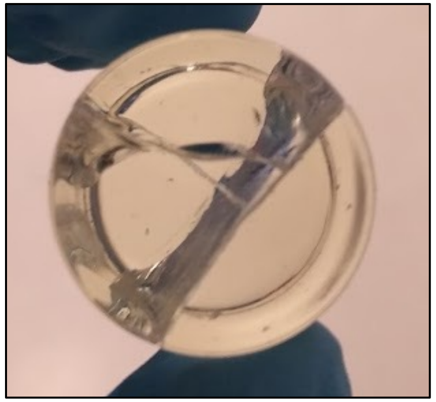
Dopant Precipitation on Surface



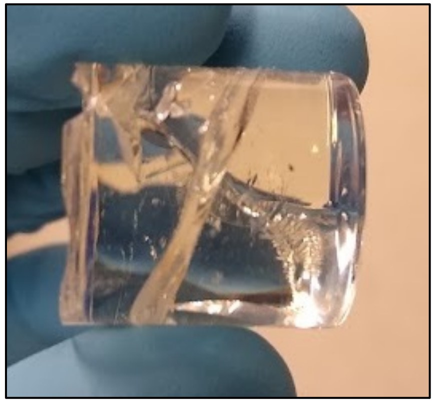
Internal Precipitation



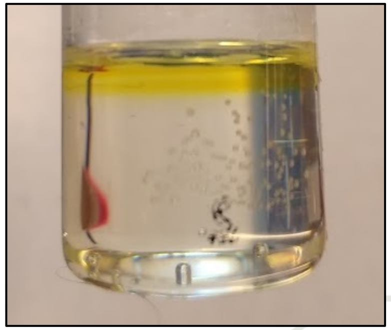
Cracked



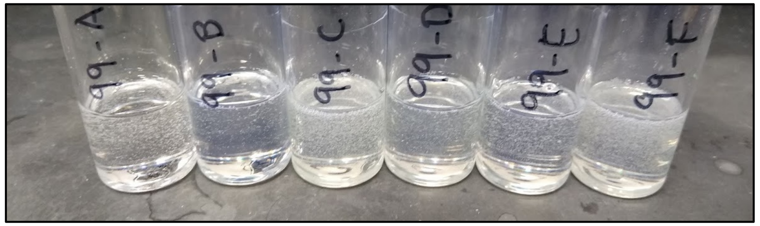
Cracked



Surface Coloration



Air Bubbles Trapped



Ion quenching in boron-doped organic scintillators

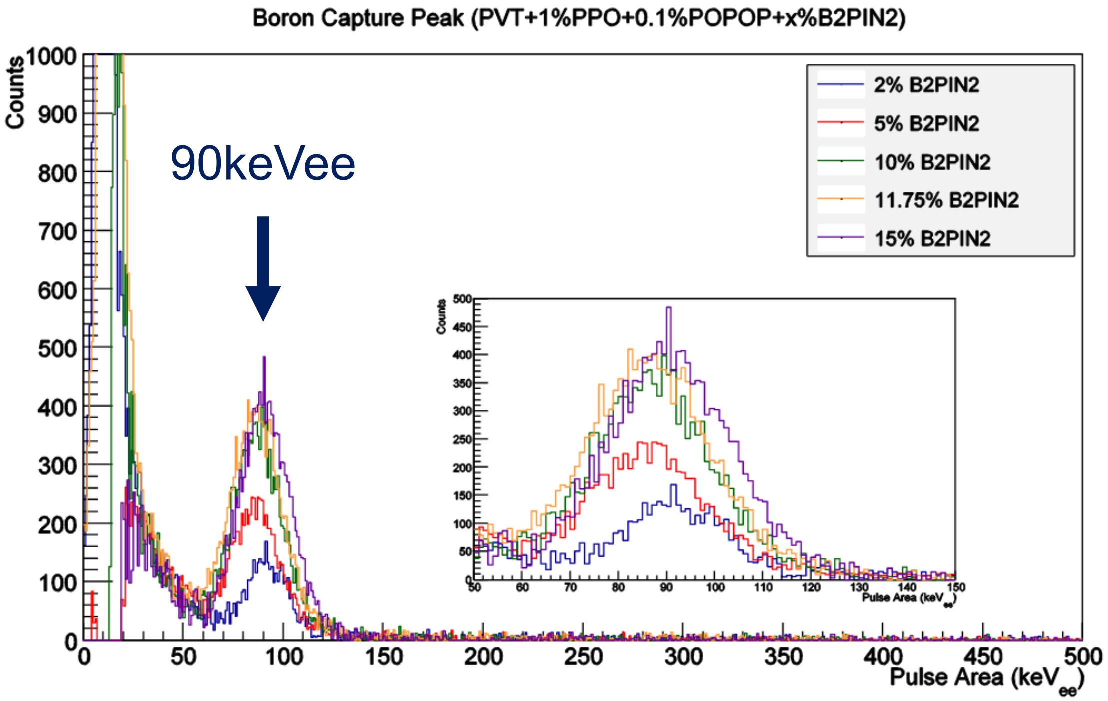


Figure 3: Number of counts versus calibrated pulse area (keV$_{ee}$) for ^{10}B thermal neutron reaction spectra with gamma response subtracted via cadmium shielded measurement. The keV$_{ee}$ sample scale adjusts all samples to the ^{137}Cs Compton edge position of each sample (light yield adjustment) to make changes in alpha quenching apparent.

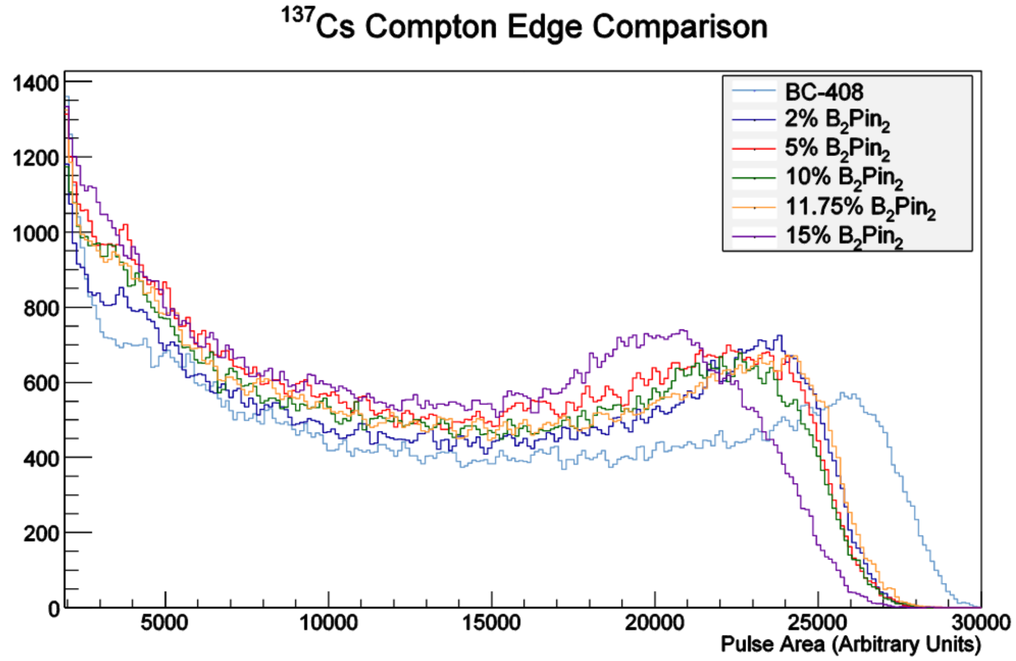


Figure 2: Number of counts vs uncalibrated pulse area in scintillating PVT based samples as measured through exposure to a ^{137}Cs γ-source using the same PMT bias value.

Adam Mahl*,2 Henok A. Yemam*,1 John Stuntz,1 Tyler Remedes,2 Alan Sellinger,1 Uwe Greife 2, et al, Bis(pinacolato)diboron as an additive for the detection of thermal neutrons in plastic scintillators, NIM A, Volume 816, 21 April 2016, Pages 96-100, https://doi.org/10.1016/j.nima.2016.01.073, 2016

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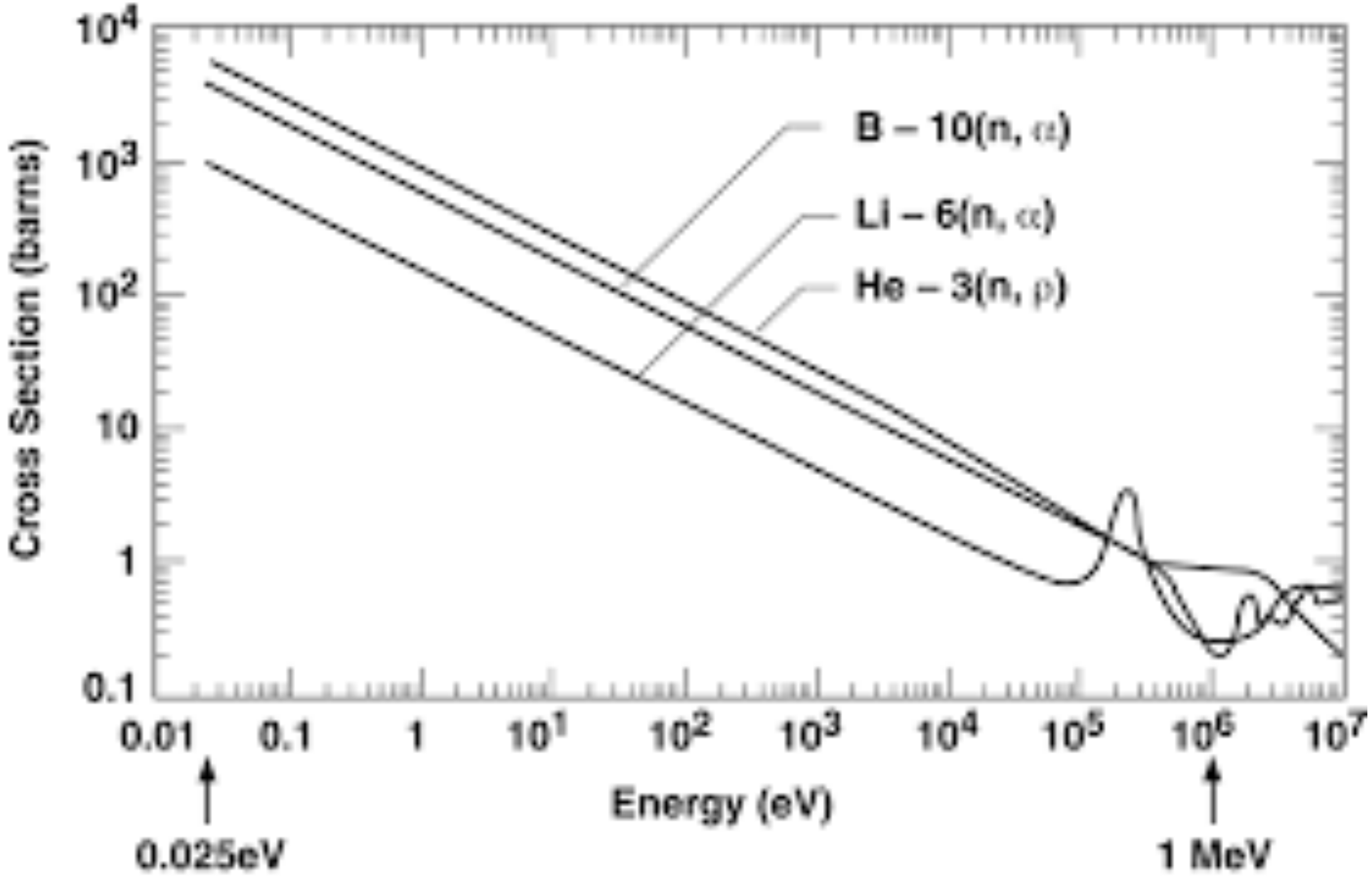
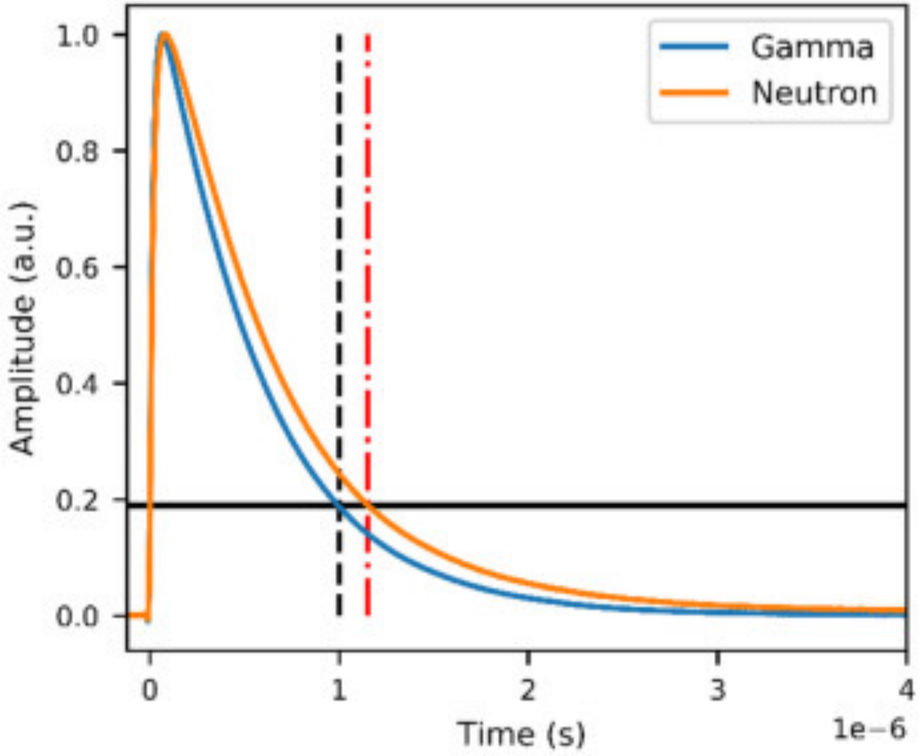
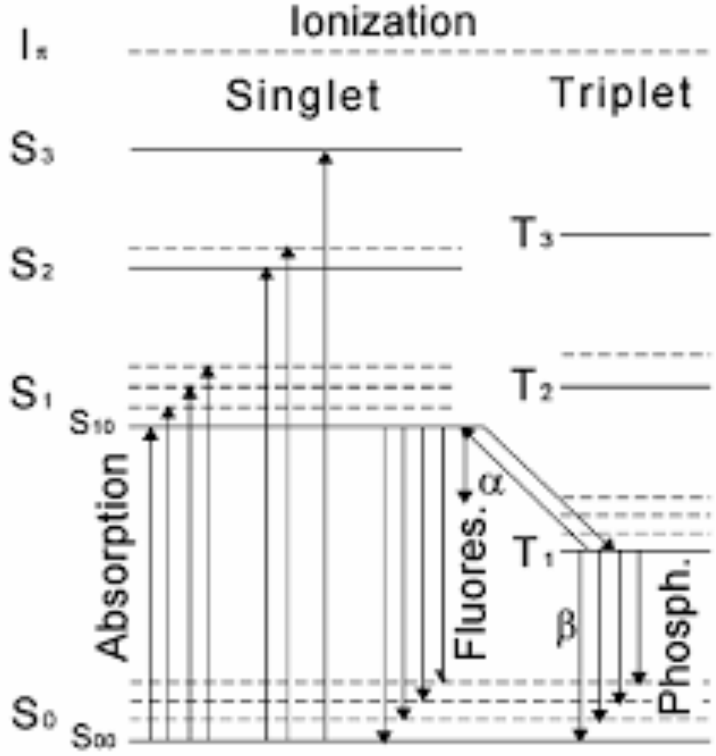
Neutron Capture Cross Section



Scintillators – What are they again?





Energy levels of an organic molecule
with pi-electron structure