

Triple PSD with Polysiloxane-Based Scintillators

Mackenzie Duce

2023 LANNS Symposium-2



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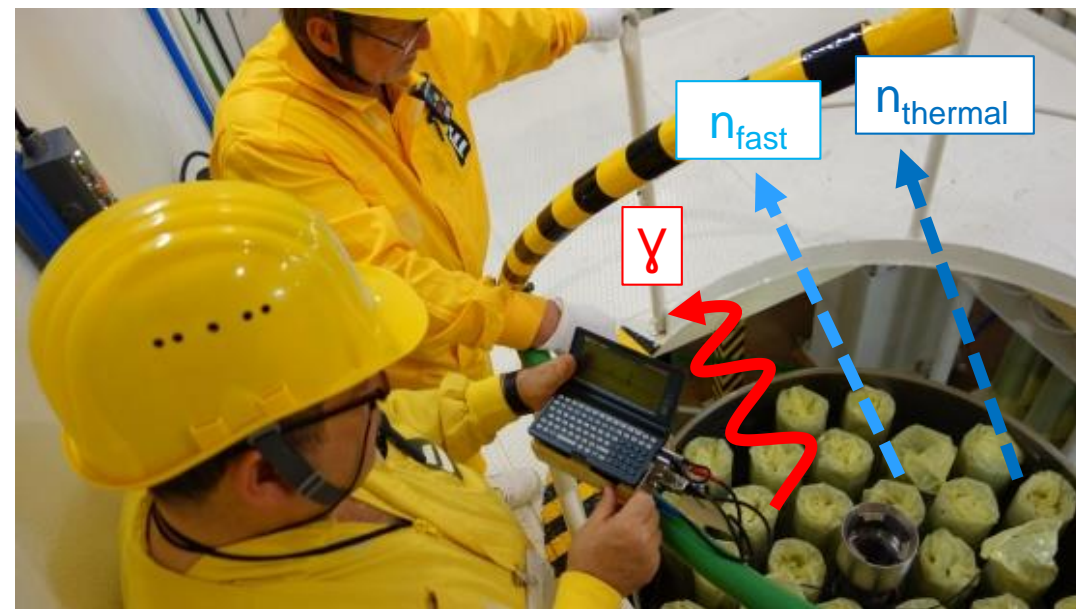
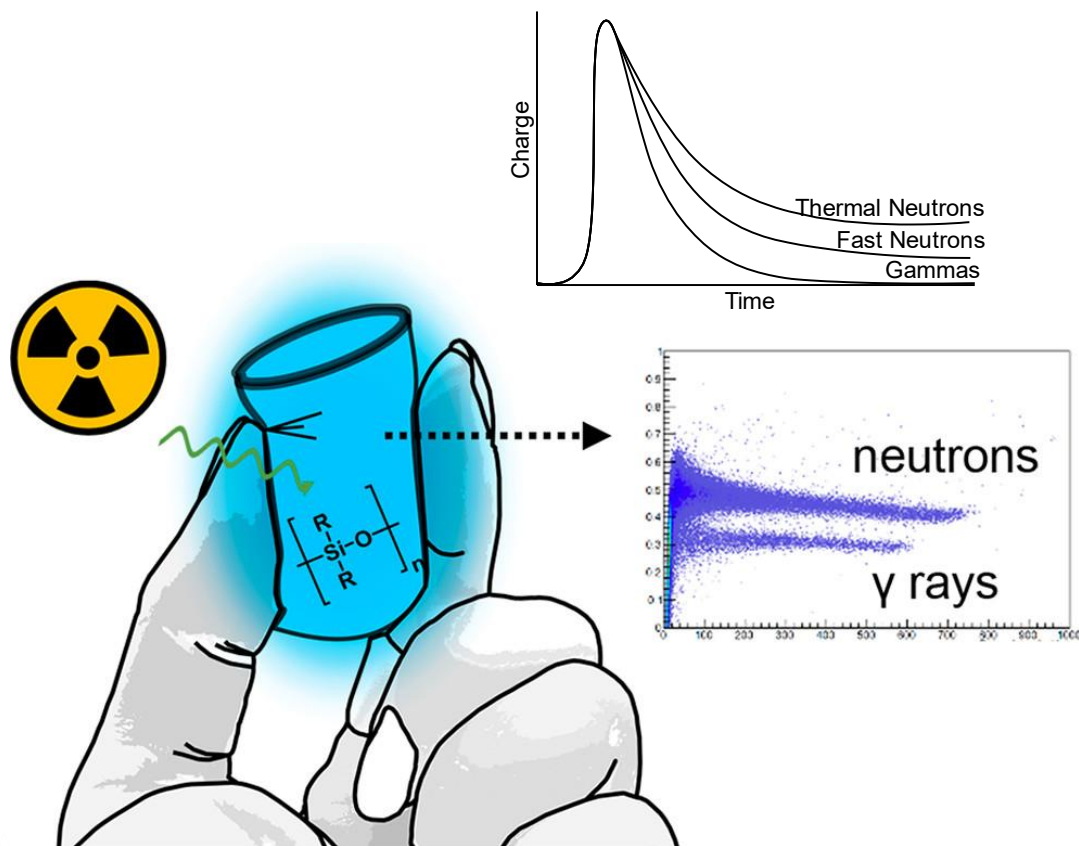
LABORATORY FOR ADVANCED NUCLEAR
NONPROLIFERATION AND SAFETY

November 03, 2023



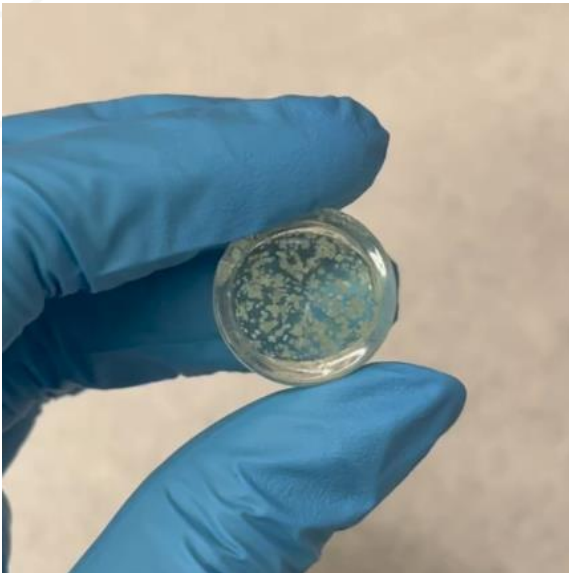
Organic scintillators for neutron detection

- Triple particle detection can support many detection applications

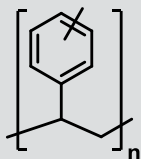
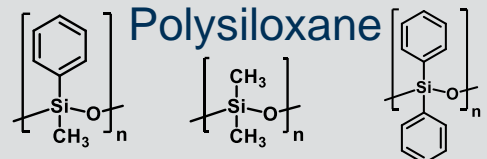


[IAEA inspectors conducting fresh fuel verification](#)

Polysiloxanes as alternative to PVT matrices



Polysiloxane-based scintillator at Georgia Tech, fabricated November 2022 at Colorado School of Mines by Caleb Chandler. (KER6000/PHF/Phenyl)

	 PVT	 Polysiloxane
Transparency	Yes	Yes
Physical Properties	Hard, rigid	Variable
Radiation Hardness	No	Yes ^{2,6}
Thermal Stability	No ²	Yes ^{3,7}
Fabrication	5 days, air sensitive	3 hrs, in air ¹
Cost	\$0.08/g ⁵	\$0.75 - \$16/g ⁵
PSD	@ 20%wt dopant	@ 5%wt dopant ^{4,1}

¹A. Lim, J. Arrue, P. Rose, A. Sellinger, A. Erickson, *ACS Appl. Polym. Mater.*, 2020, 2, 8, 3657-3662

²Bertrand, G. H. V.; Hamel, M.; Sguerra, F. *Chem. Eur. J.* 2014, 20 (48), 15660–15685.

³Arrue, J., et al., *NIM A*, 2023, 1056, 168650

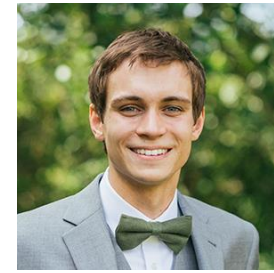
⁴Marchi, T. et al. *Sci. Rep.* 2019, 9 (1), 9154.

⁵Prices from Sigma, Gelest, TCI America, and Nusil

⁶Quaranta, et al. *Mats. Chem. And Phys.* 2013, 137 (3) 951-958

⁷Deshpande, G, Rezac, M., *Polymer Deg. And Stability.* 2002, 76 (1) 17-24

Fabrication at Colorado School of Mines



Caleb Chandler
[Sellinger Research Group](#)



Prepare
silanized vial
(30-60 mins)



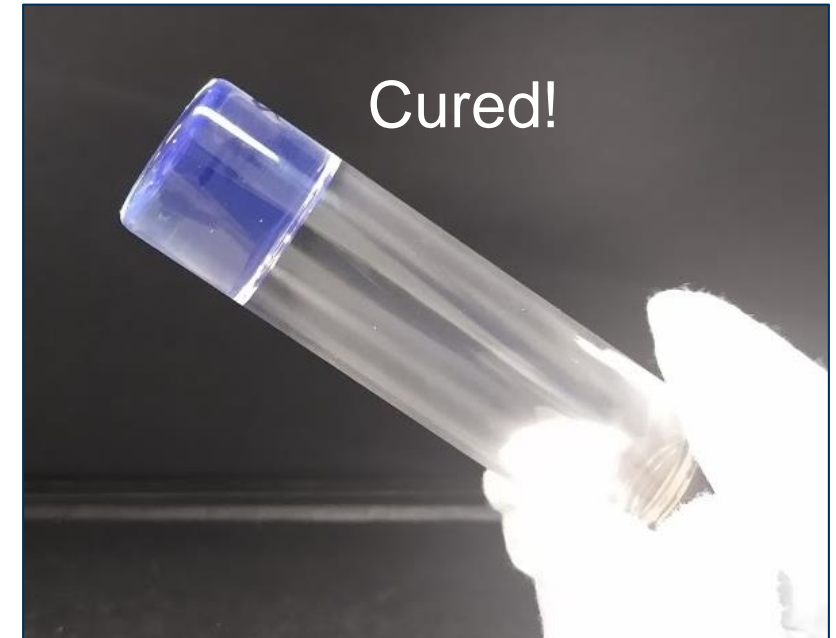
Add SFS, PPO



Add xylenes

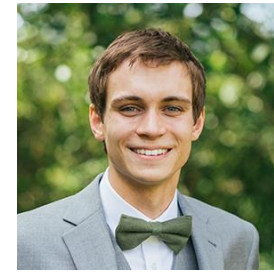


Solubilize and
add Part A,
mix, add Part B



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

Common Fabrication Challenges

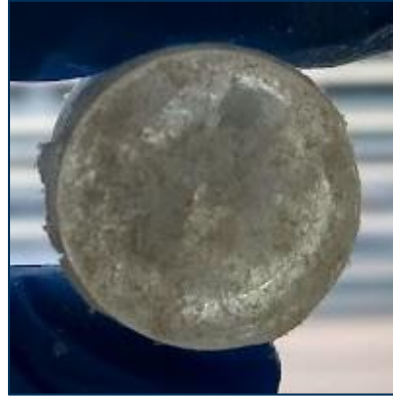


Caleb Chandler
[Sellinger Research Group](#)

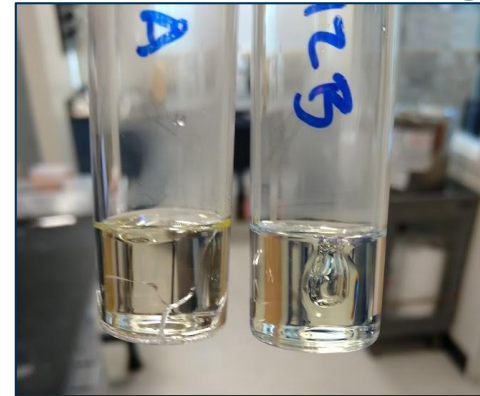
**Dopant Precipitation
on Surface**



**Internal
Precipitation**

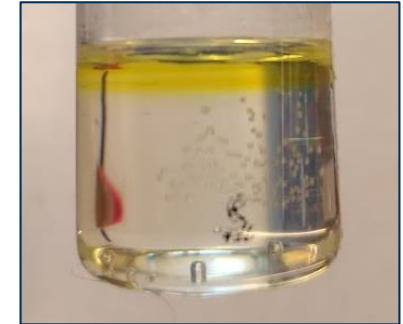


**Glass
Cracked**

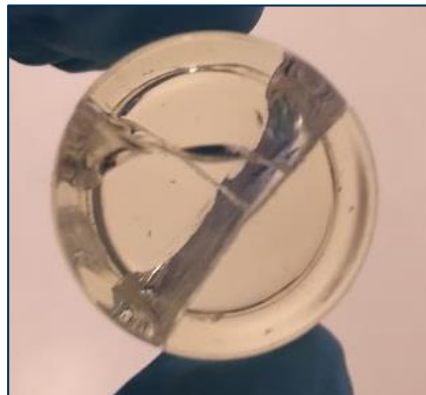


**Internal
Bubbling**

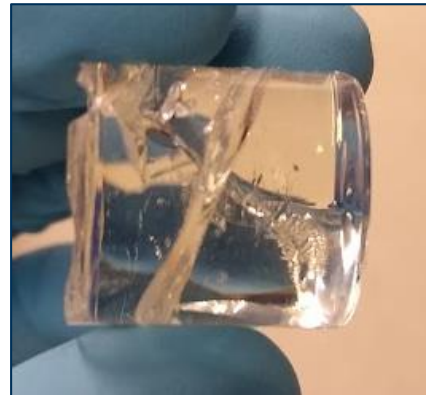
Surface Coloration



Cracked



Cracked



Air Bubbles Trapped



Toward 3 Particle Imaging

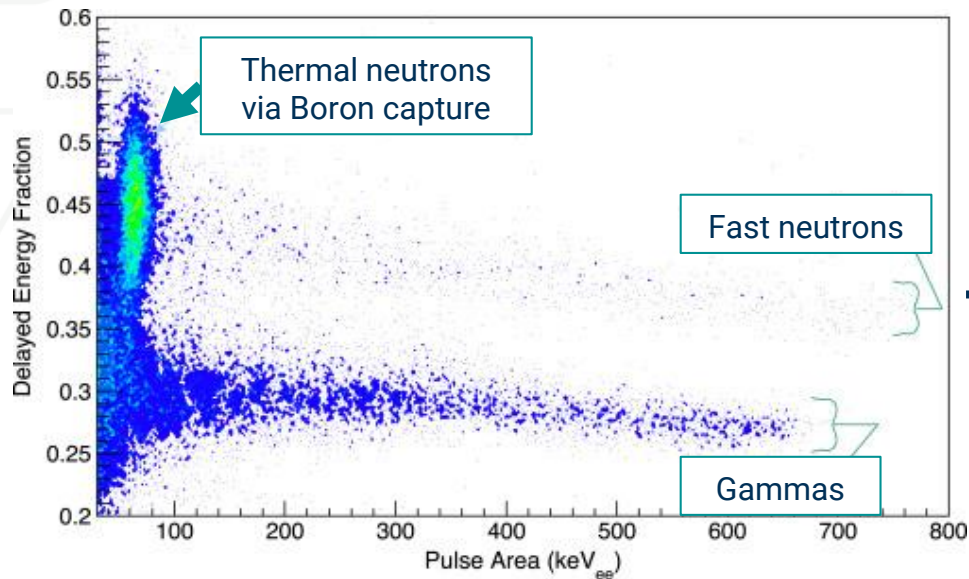
- Add thermal-neutron sensitive material to polysiloxane scintillators
- Iterate recipe for:
 - Stability
 - High light output
 - Pulse shape discrimination-capable
 - Minimal quenching
- Fabricate array
- Characterize
- Add image reconstruction



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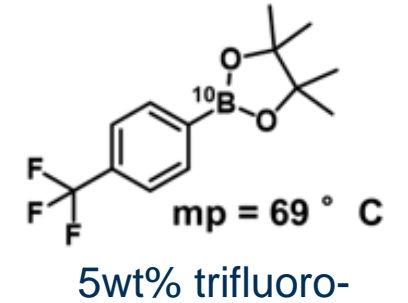
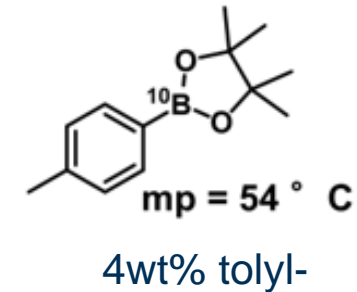
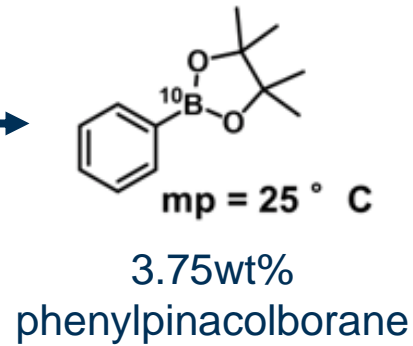
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(Enriched) Boron-10 Doped PVT Scintillators

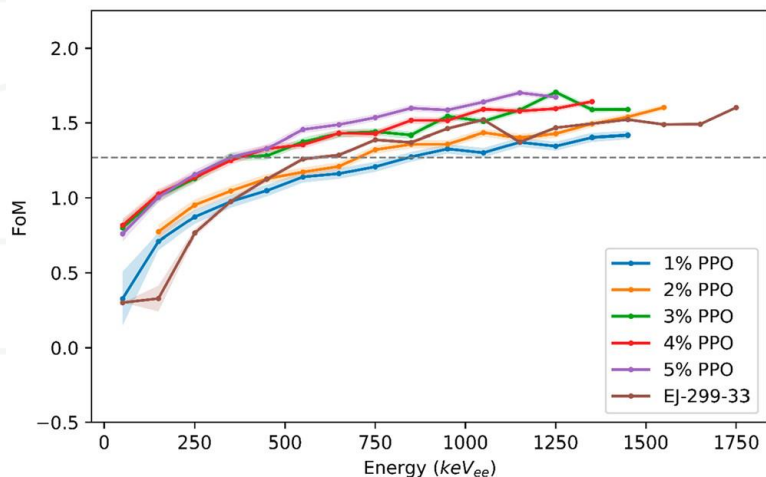


Previous work synthesizing and incorporating MBB in PVT scintillators

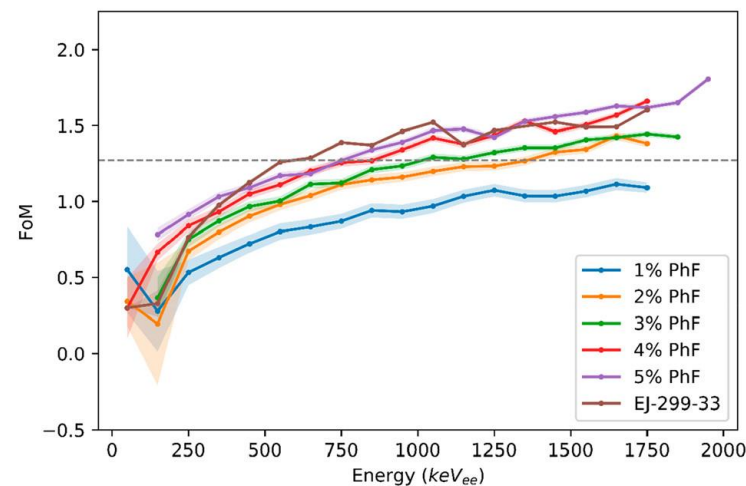
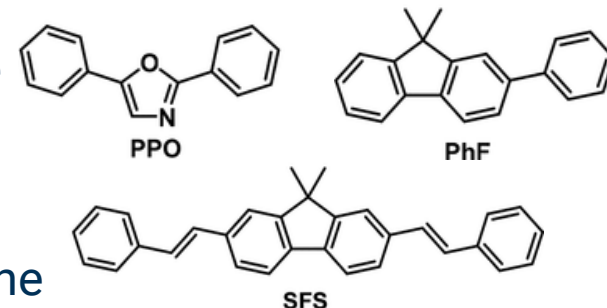
- Derivatives of MBB used in this study
 - Synthesized from enriched boric acid (>96% ^{10}B)



Polysiloxane Matrices & Fluorophores Tested



- Primary dopants:
 - PHF: 9,9-dimethyl-2-phenyl-9H-fluorene
 - PPO: 2,5-diphenyloxazole
- Secondary dopant (wavelength shifter):
 - SFS: 9,9-dimethyl-2,7-distyryl-9H-fluorene
- Polymer resins modified:



PSD FOM results for Wacker LUMISIL-based polysiloxane scintillators of varying dopant concentrations

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

- [Shin-Etsu KER-6000](#)
- LED encapsulant
- [Wacker SILRES H62C](#)
- electronics encapsulant





	<u>R.I.</u>	<u>Hardness</u>	<u>Mix</u>	<u>Cure</u>
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

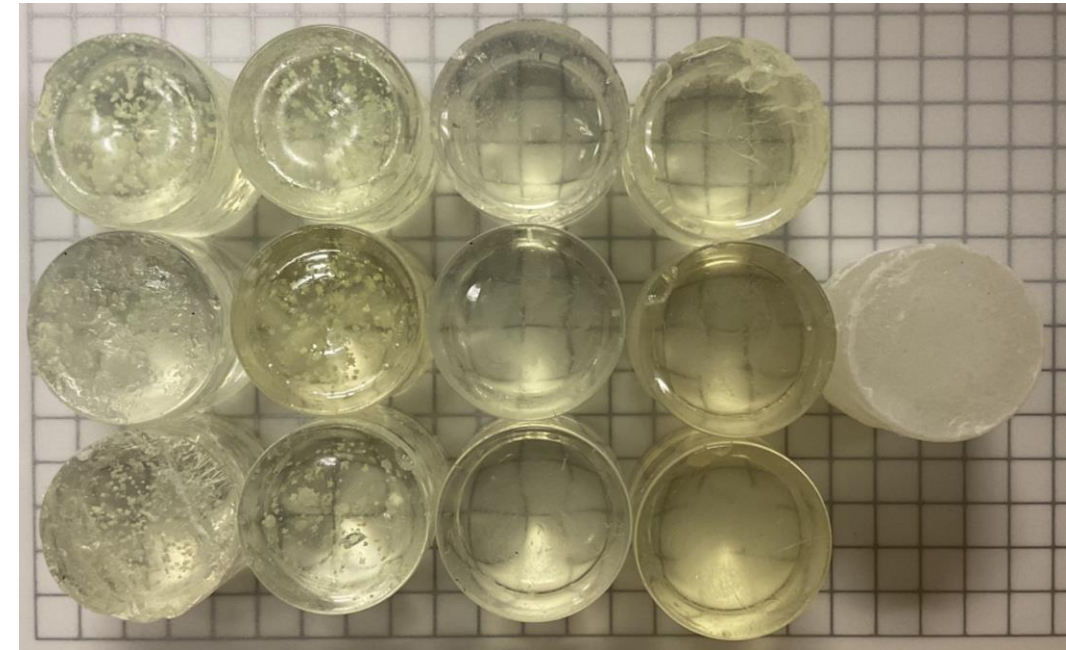
Testing Scintillator Compositions



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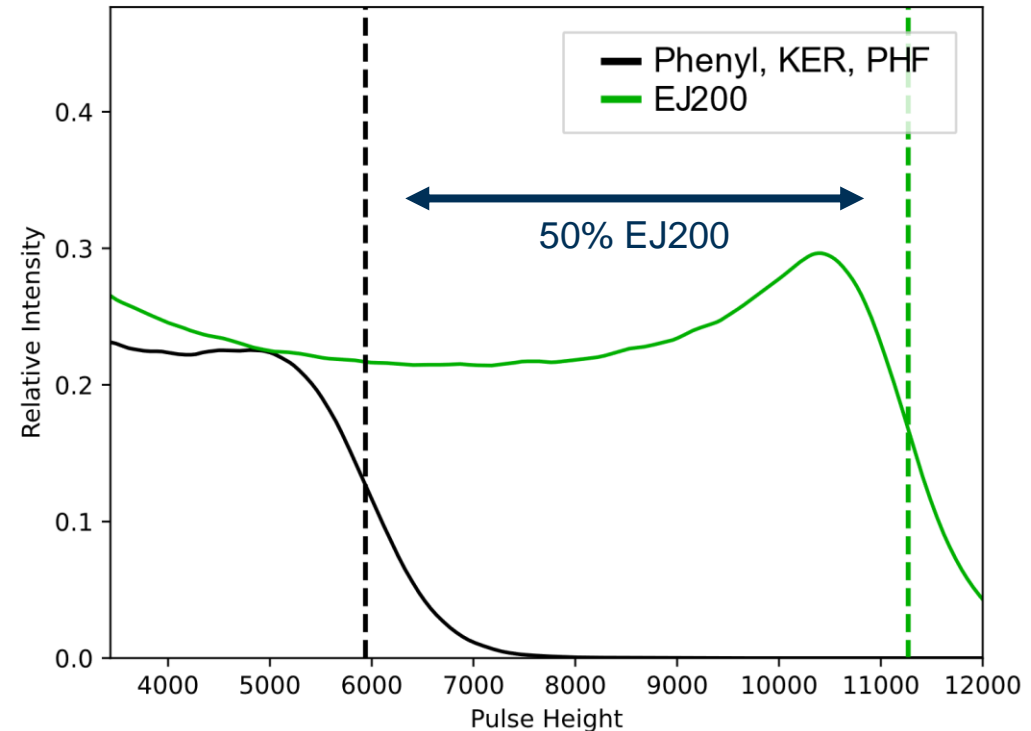
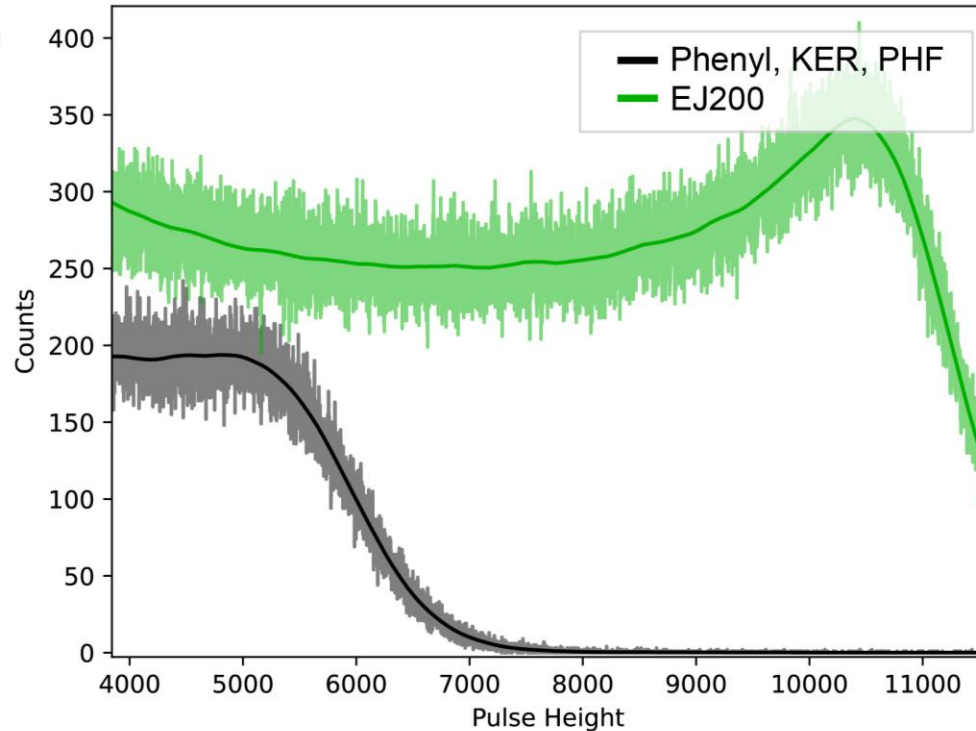
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	KER6000		SILRES		SILRES
	PHF	PPO	PHF	PPO	20% PHF
3.75% Phenyl-					
4% Toly-					
5% Trifluoro-					



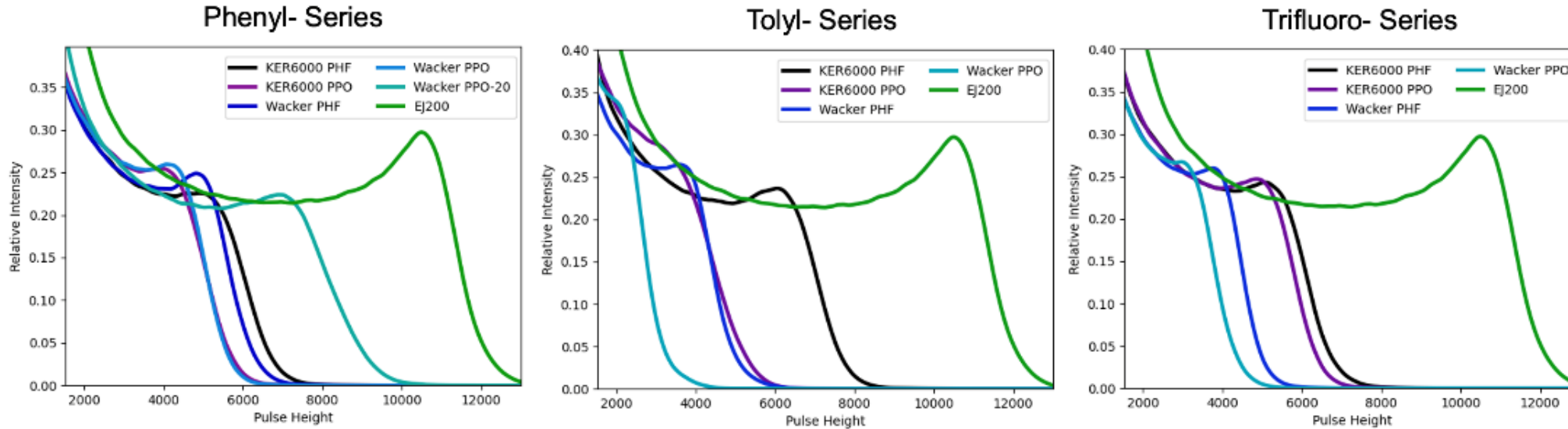
One year later!

Measuring Light Yield (electron equivalent)



- 477 keV Cs-137 Compton edge
- Differentiation method
- Compare to EJ200: 10,000 ph/MeVe

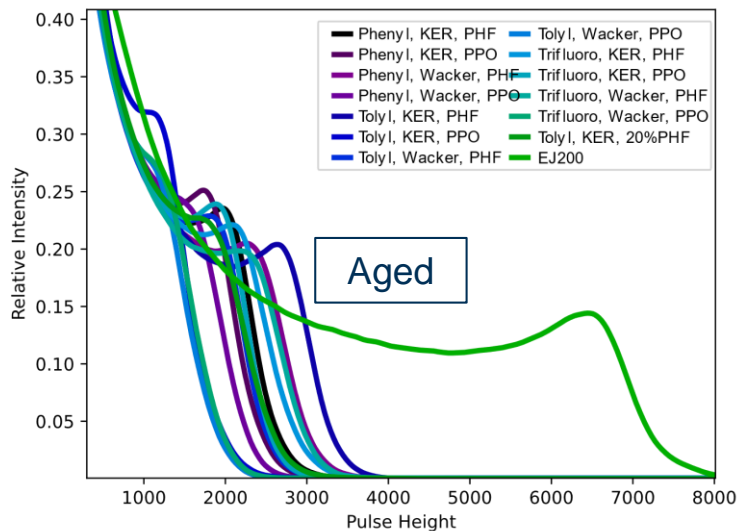
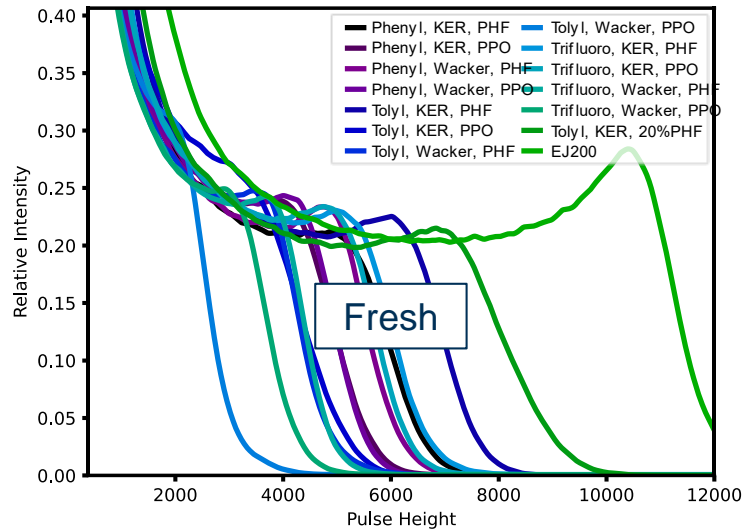
Measuring Light Yield (electron equivalent)



		5% PHF	5% PPO	20% PHF
B10	Matrix	<i>Fresh</i>	<i>Fresh</i>	<i>Fresh</i>
Phenyl	KER6000	0.53	0.45	
	SILRES	0.49	0.45	
Toly	KER6000	0.62	0.39	
	SILRES	0.39	0.23	0.71
Trifluoro	KER6000	0.55	0.51	
	SILRES	0.39	0.33	

Toly, KER6000, 5%PHF
~6,200 photons/MeV
 Toly, SILRES, 20%PHF
~7,100 photons/MeV

Aging Effects on Light Yield



B10	Matrix	5% PHF			5% PPO			20% PHF		
		<i>Fresh</i>	<i>Aged</i>	<i>Change</i>	<i>Fresh</i>	<i>Aged</i>	<i>Change</i>	<i>Fresh</i>	<i>Aged</i>	<i>Change</i>
Phenyl	KER6000	0.53	0.36	-32%	0.45	0.33	-27%			
	SILRES	0.49	0.42	-14%	0.45	0.31	-69%			
TolyI	KER6000	0.62	0.46	-26%	0.39	0.35	-10%			
	SILRES	0.39	0.35	-10%	0.23	0.24	4%	0.71	0.35	-51%
Trifluoro	KER6000	0.55	0.38	-31%	0.51	0.35	-31%			
	SILRES	0.39	0.41	5%	0.33	0.26	-21%			

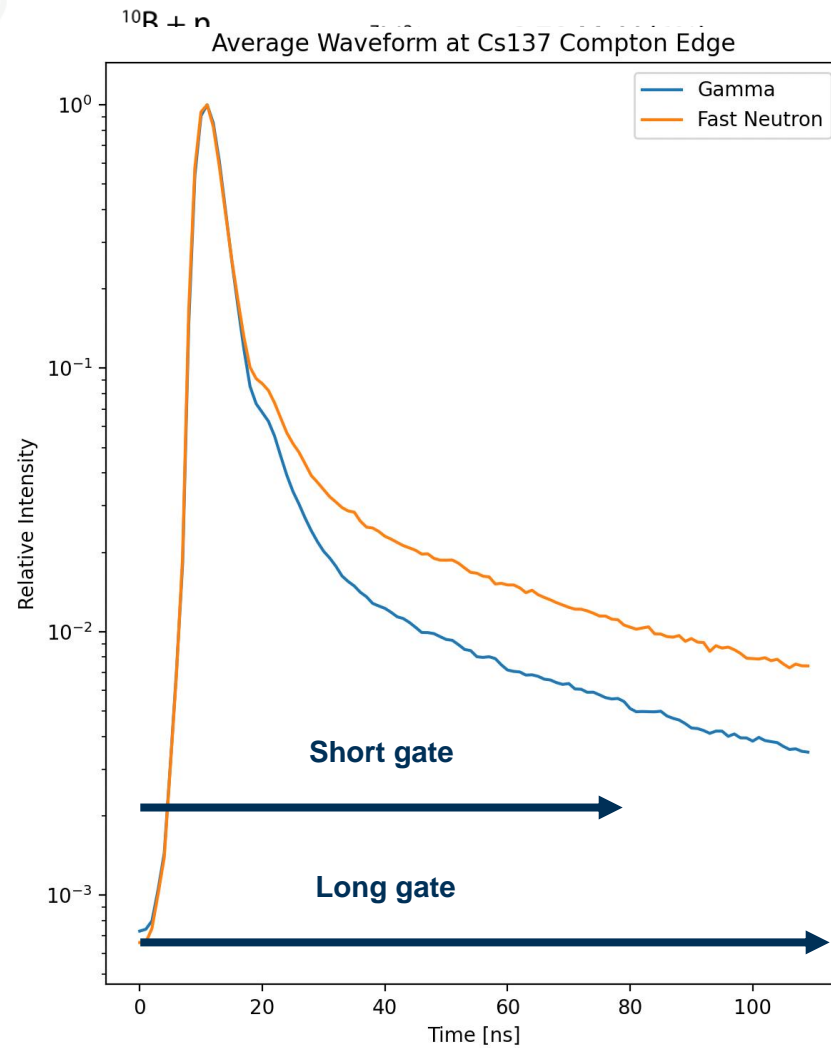
Values reported as fraction of similar size EJ200 under same experiment conditions.

KER6000 reduced ~-30%
SILRES reduced ~-15%

Pulse Shape Discrimination Measurements



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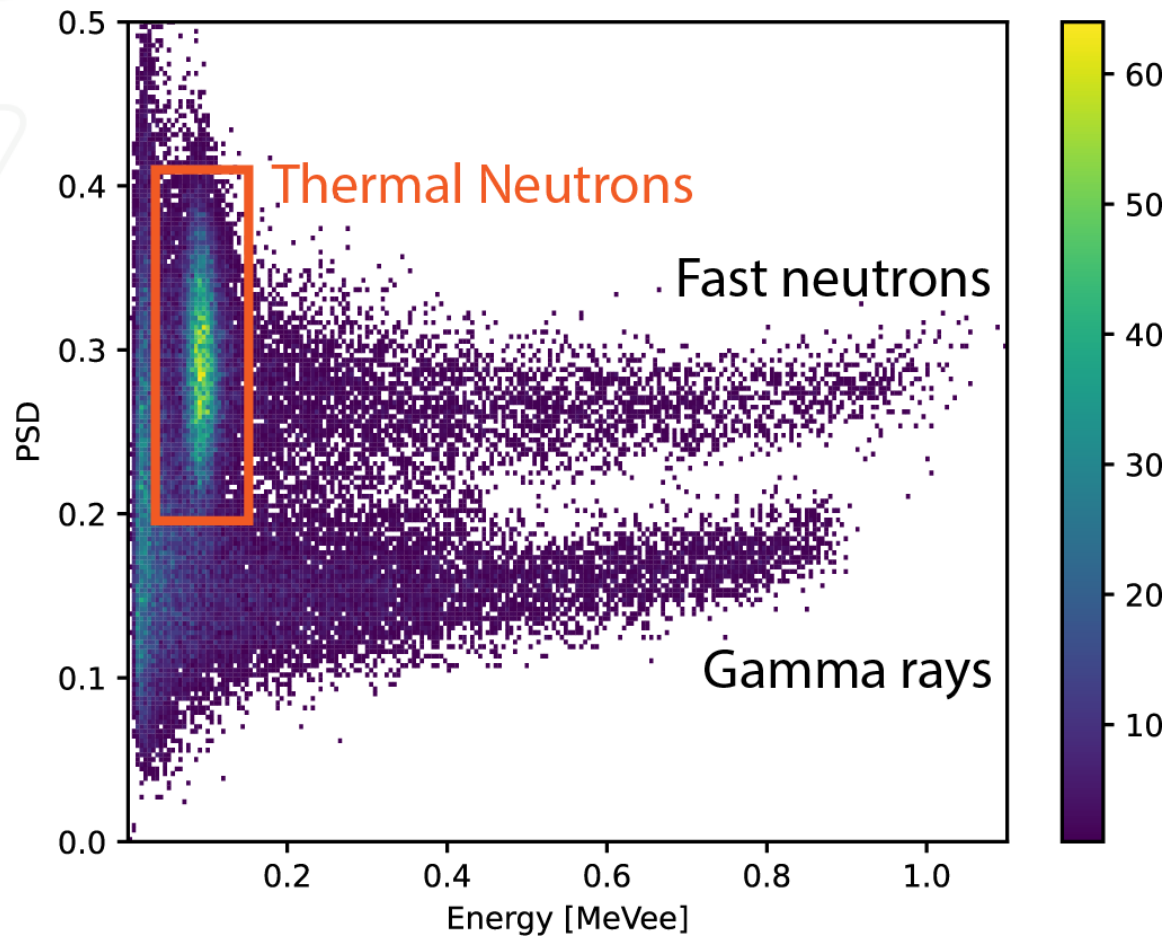
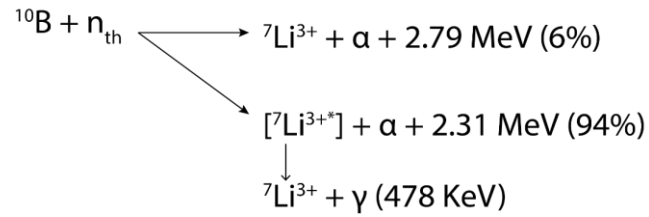
AmBe setup: Poly, concrete, water, 3" lead to knock down gammas

Pulse Shape Discrimination Measurements



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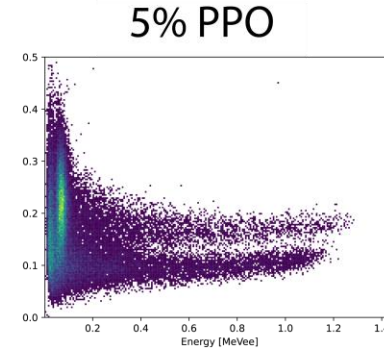
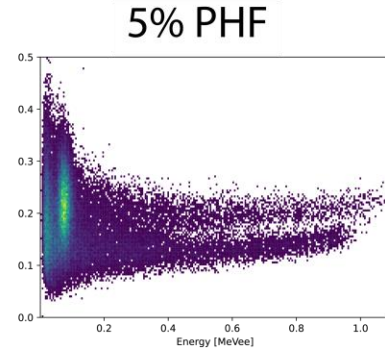
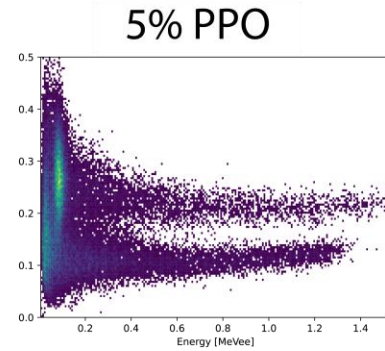
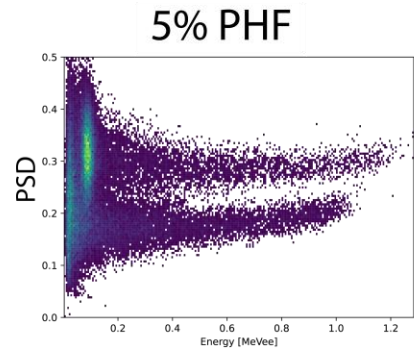
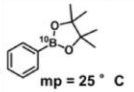
AmBe setup: Poly, concrete, water, 3" lead to knock down gammas

Pulse Shape Discrimination for all Samples

KER6000

SILRESH62C

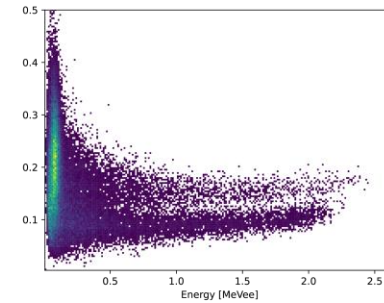
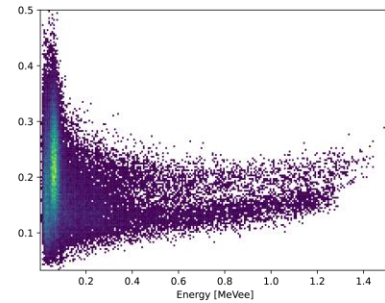
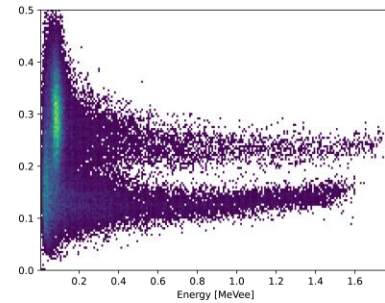
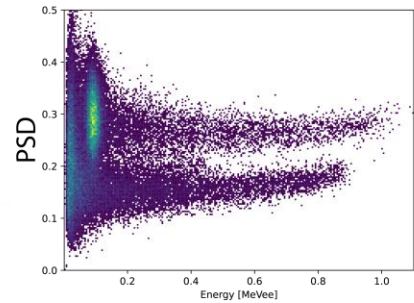
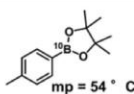
Phenyl



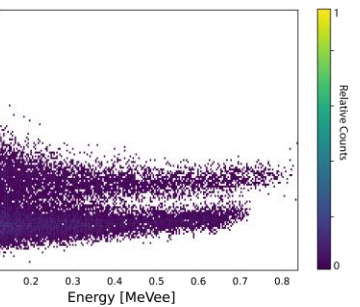
Relative Counts
0 1

20% PHF

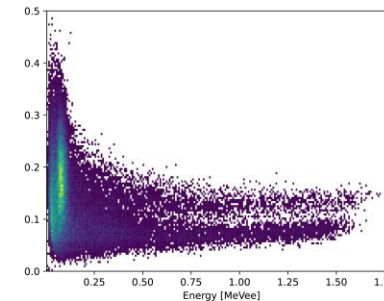
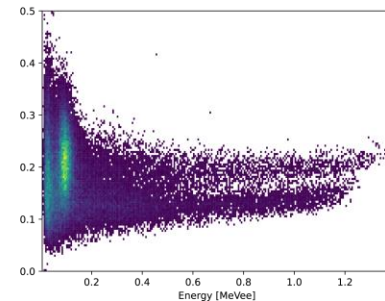
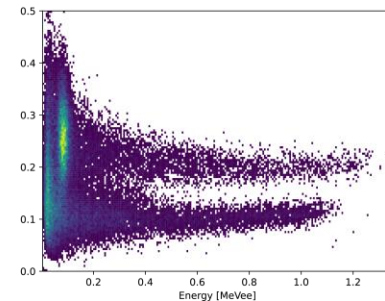
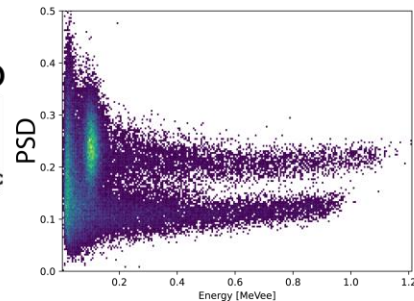
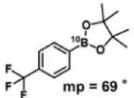
Tolyl



Relative Counts
0 1



Trifluoro



Relative Counts
0 1

Pulse Shape Discrimination for all Samples (Aged)



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KER6000

SILRESH62C

5% PHF

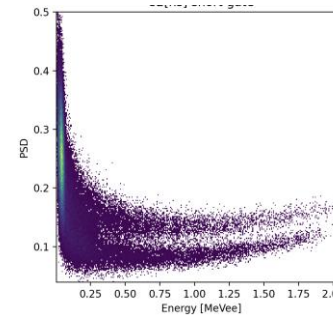
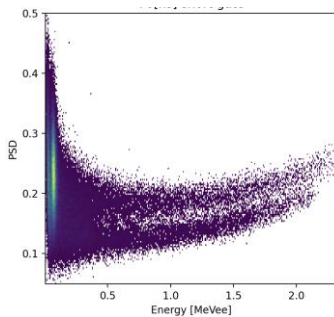
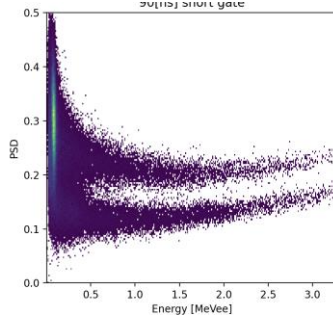
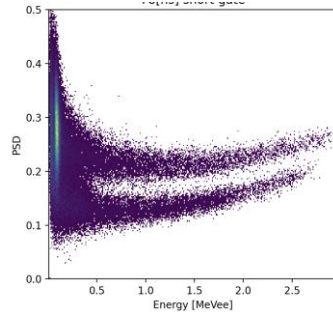
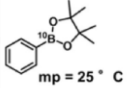
5% PPO

5% PHF

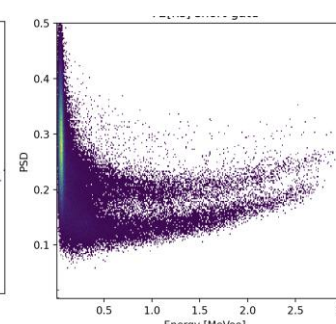
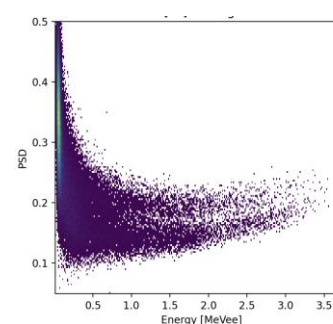
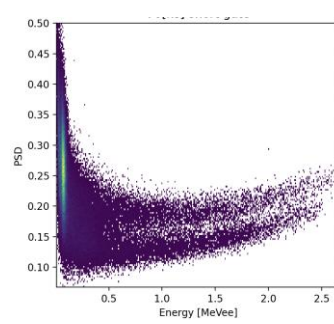
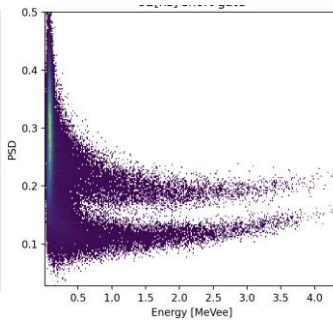
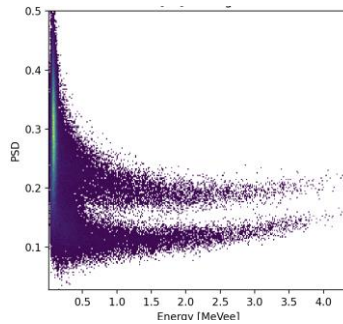
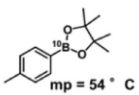
5% PPO

20% PHF

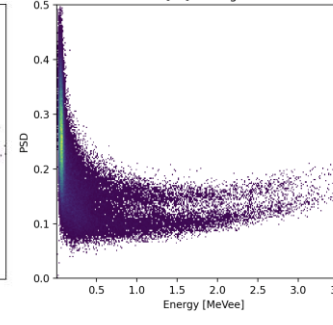
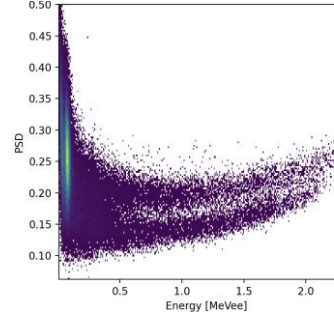
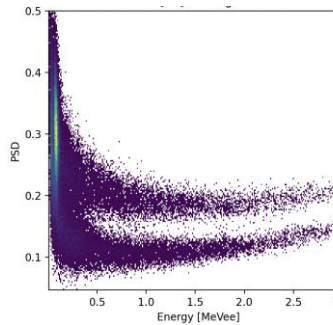
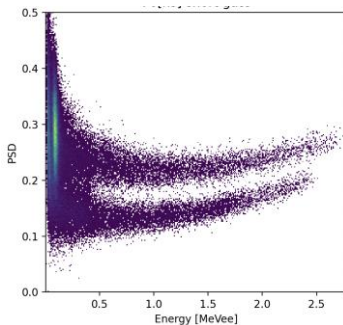
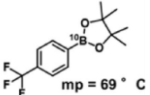
Phenyl



Tolyl



Trifluoro

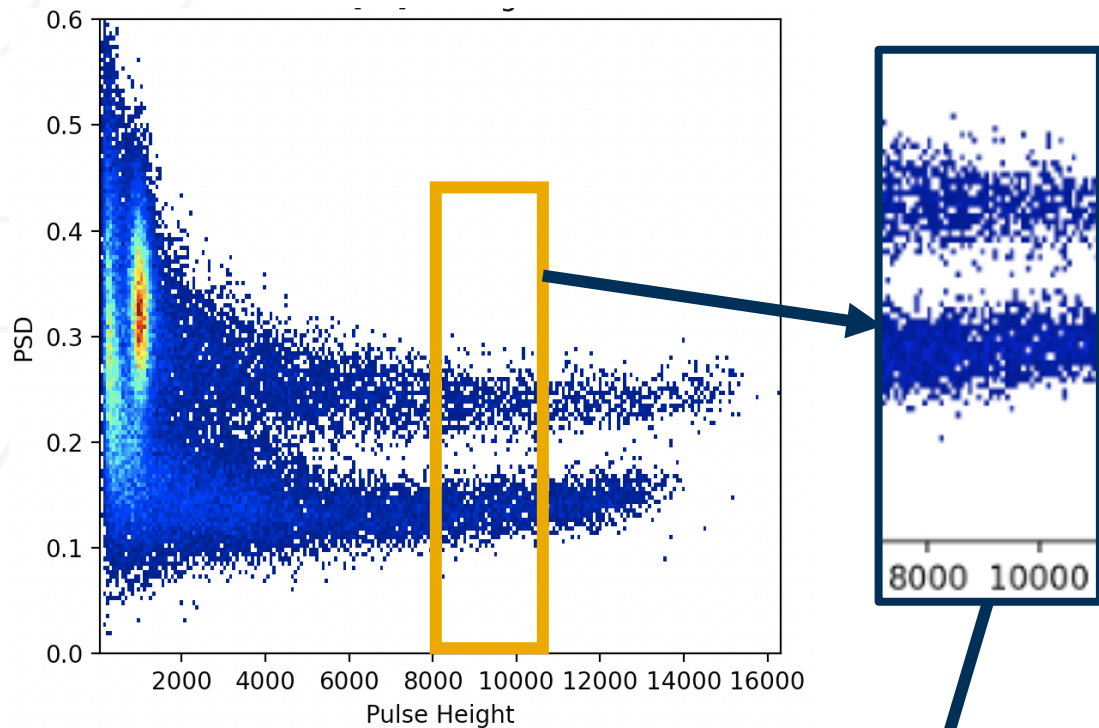




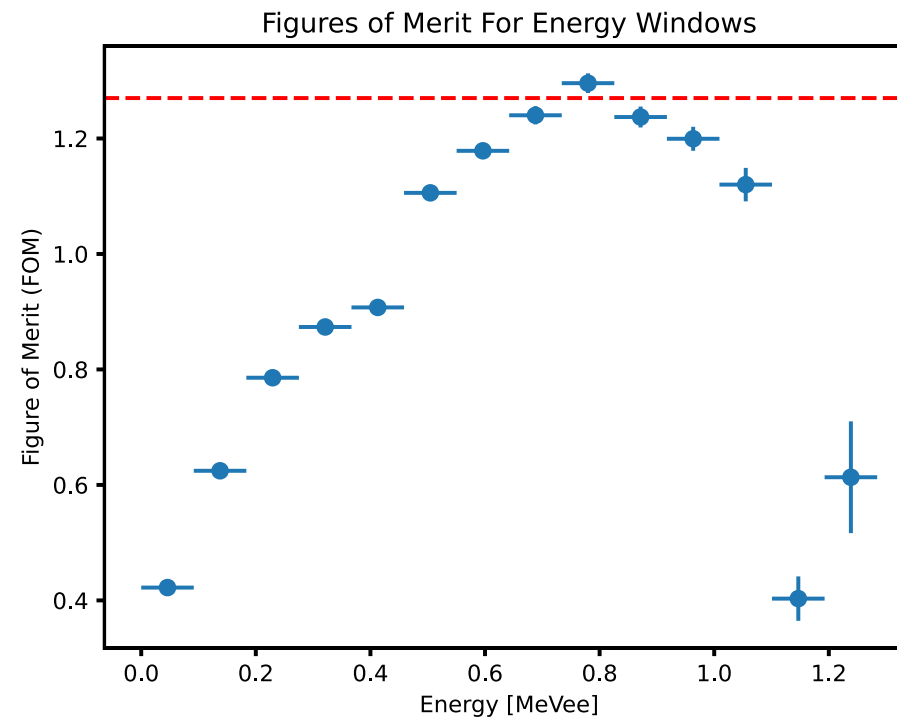
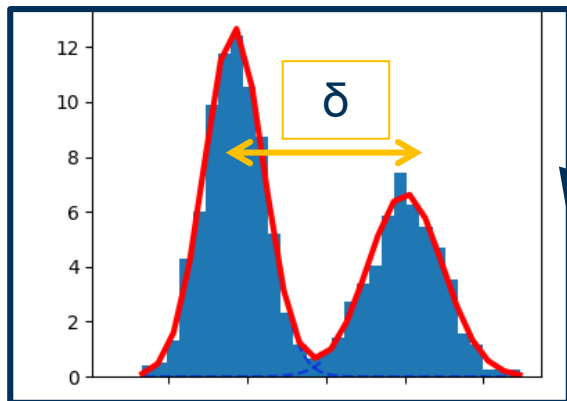
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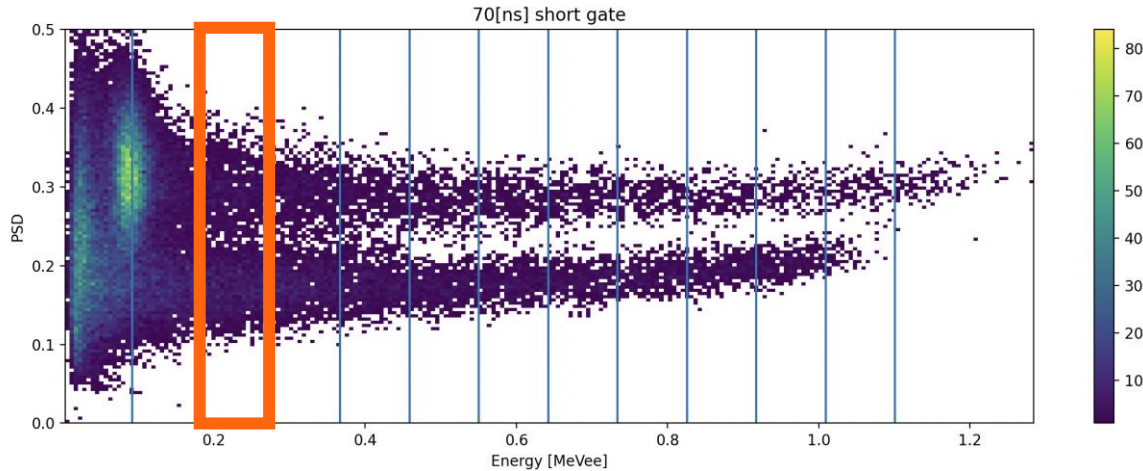
PSD Effectiveness: Figure of Merit



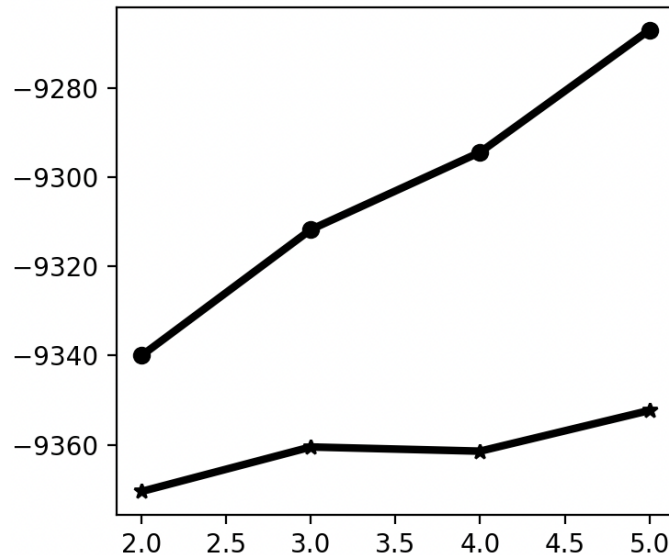
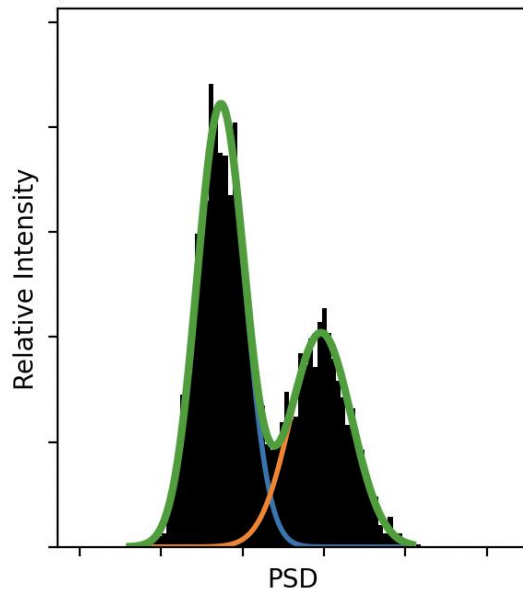
- $FOM = (\delta) / (FWHM_y + FWHM_n)$
- Efficiency separation at $FOM = 1.27$ ($\delta > 3\text{std}$)



Unsupervised Gaussian Mixture Model for FOM



- No pre-labeled data required
- Gaussian assumption aligns with data and physics
- Low energy discrimination
- BIC/AIC tests



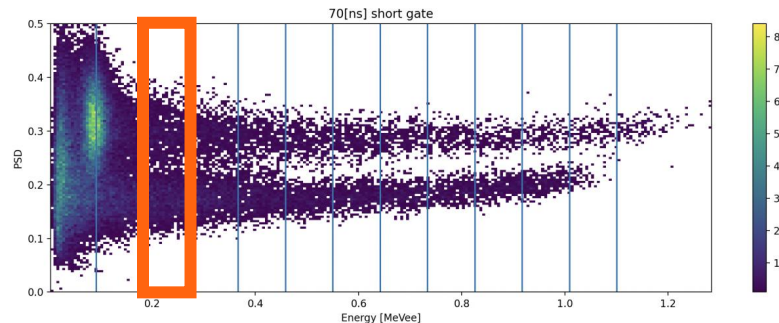
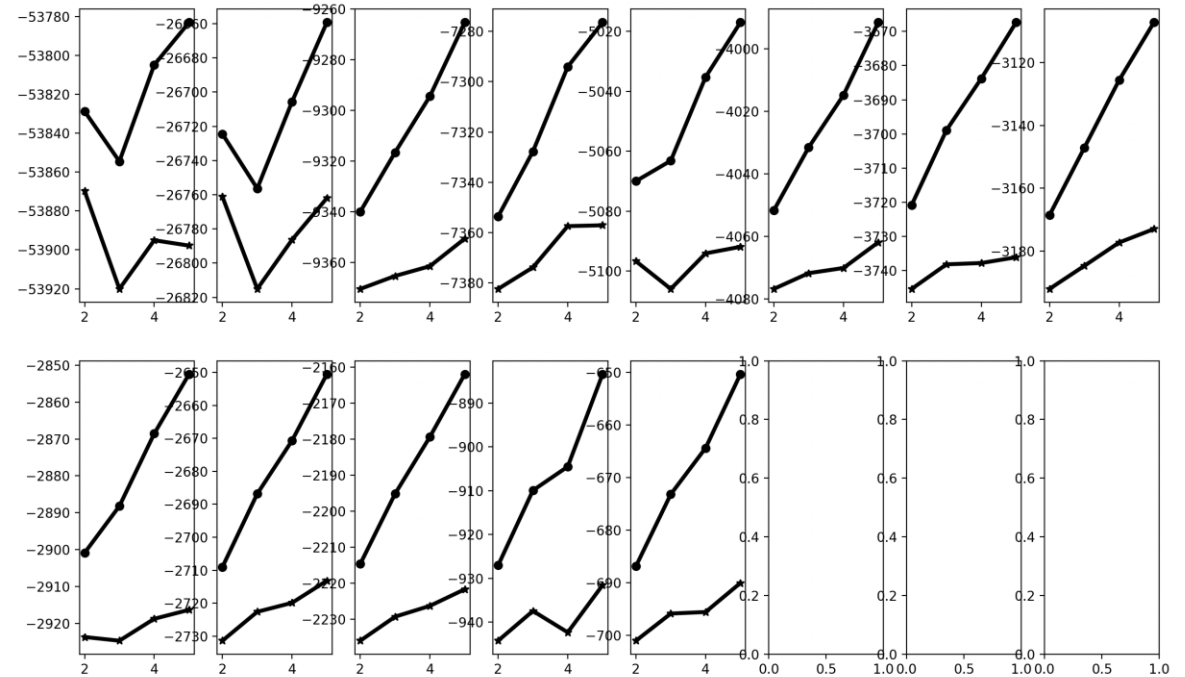
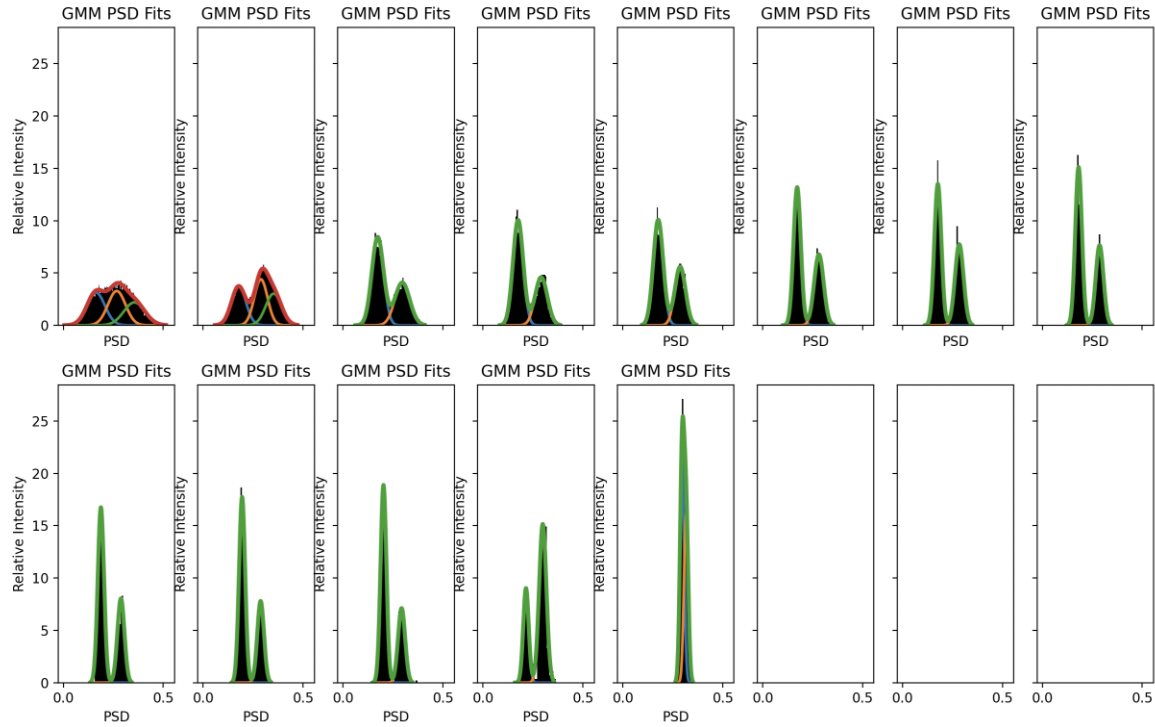
Blair, B., *Stat. Anal. Data Min.: The ASA Data Science Journal*. 2019, 12 (6) 479-488

GMM Applied to Full Dataset

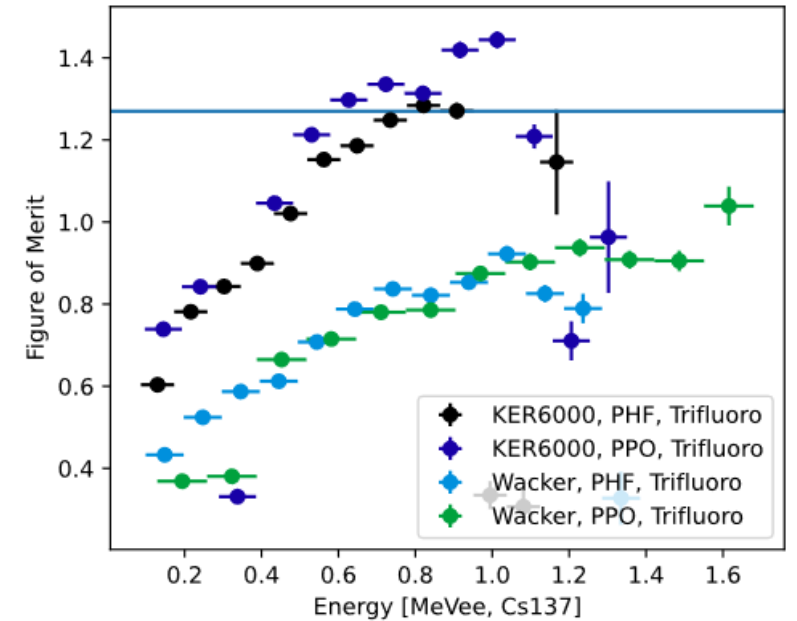
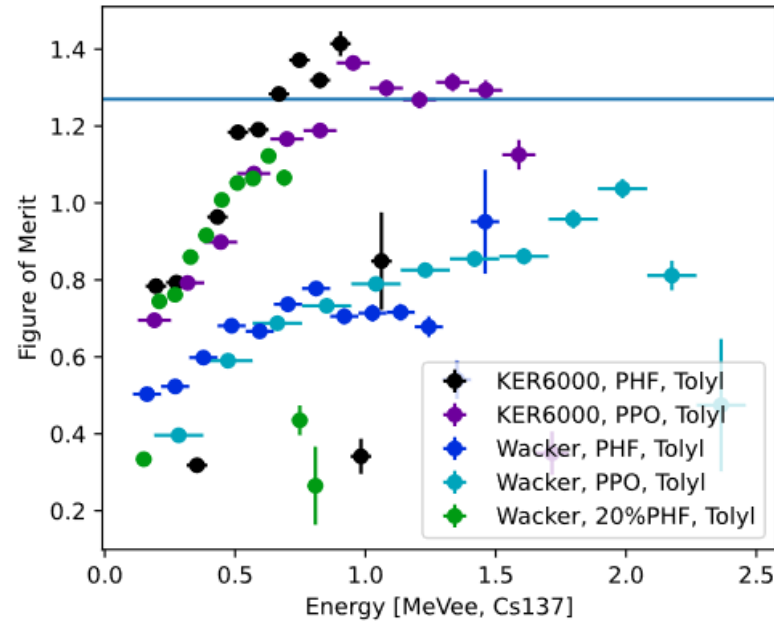
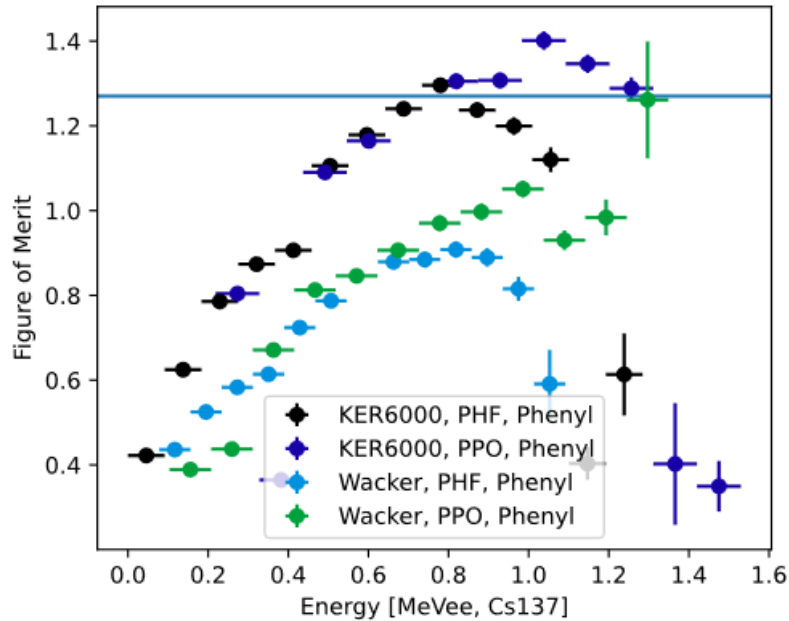


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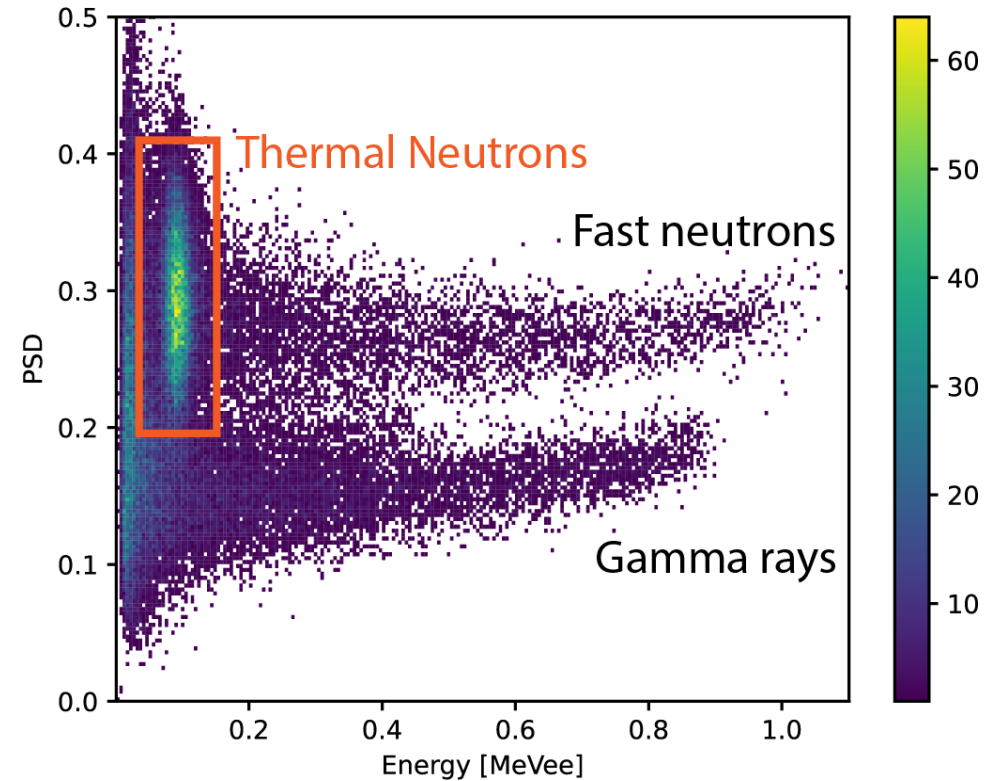
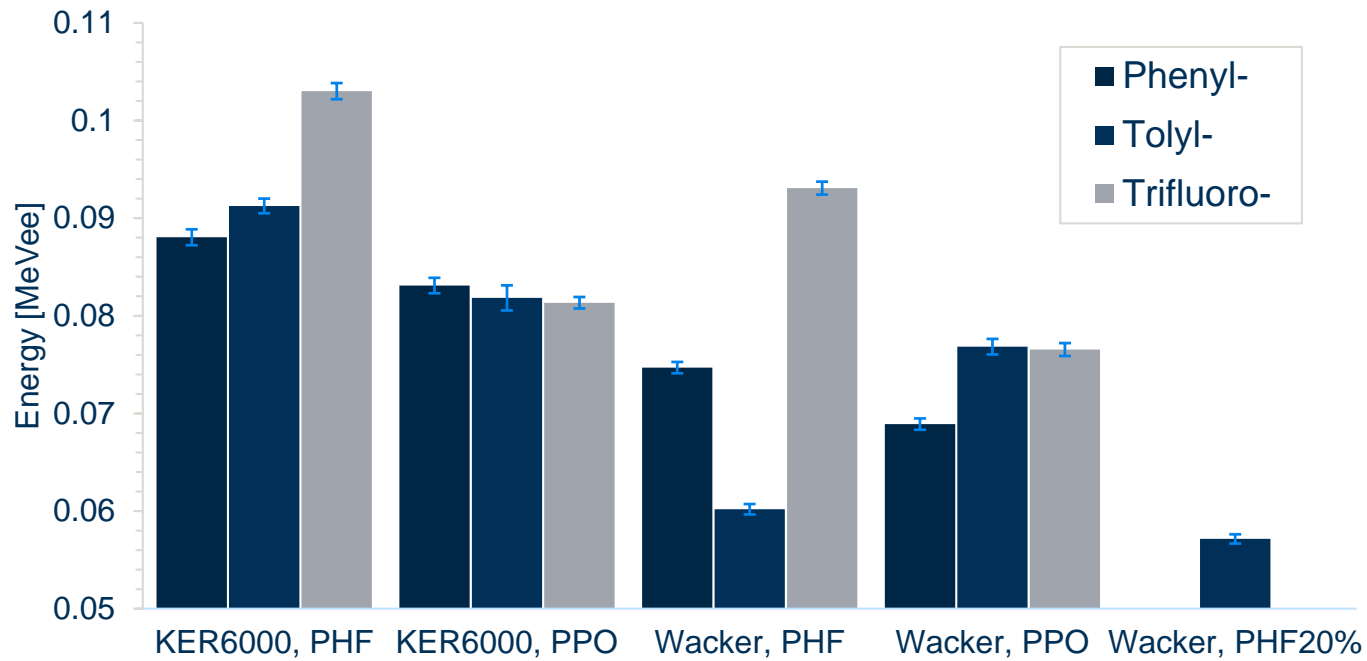


Unsupervised Gaussian Mixture Model for FOM



B-10	Matrix	5% PHF	5% PPO
5% Phenyl	KER6000	1.09	0.81
	Wacker	0.69	
4% Toly	KER6000	1.17	0.78
	Wacker	0.64	0.62
5% Trifluoro	KER6000	1.10	1.21
	Wacker	0.56	0.69

Thermal Neutron Island Evaluation



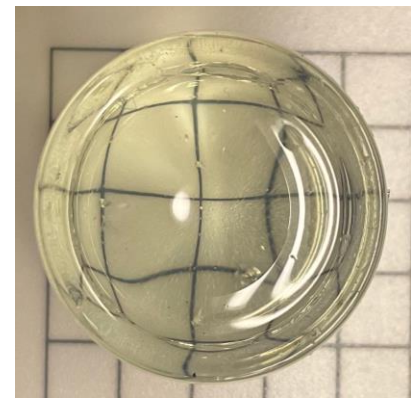
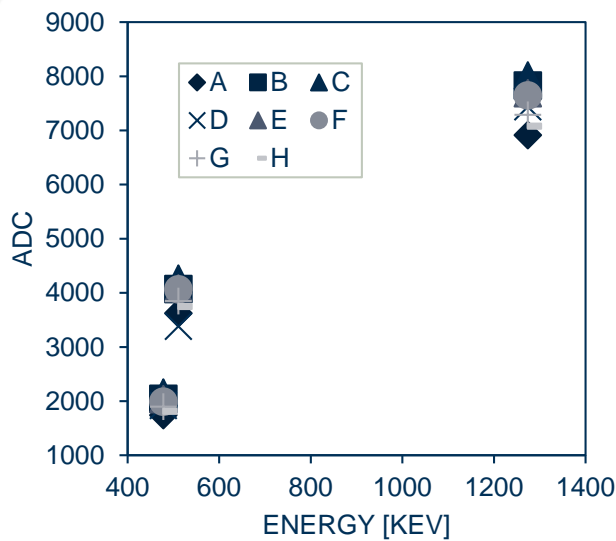


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Array Fabrication & Characterization

- Boron-enriched molecule doped did not diminish $FOM(n, \gamma)$ from previous polysiloxane trials
- Best $FOM(n_{th}, \gamma) = 0.54$ 83% of EJ309-B
- Best light yield 62% of EJ200
- -26% LY reduction over 1 year
- Successful triple particle detection!



4% Toly-, 5% PHF, KER6000



Separating Thermal Neutron Contributions

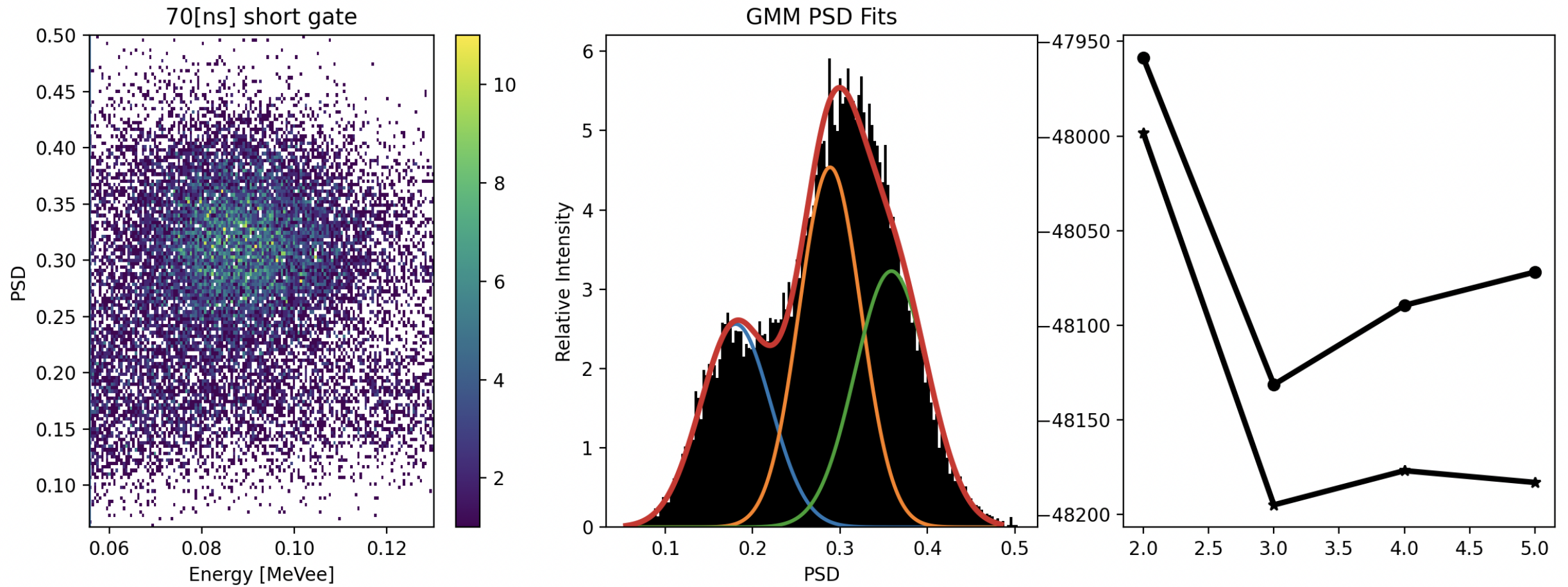
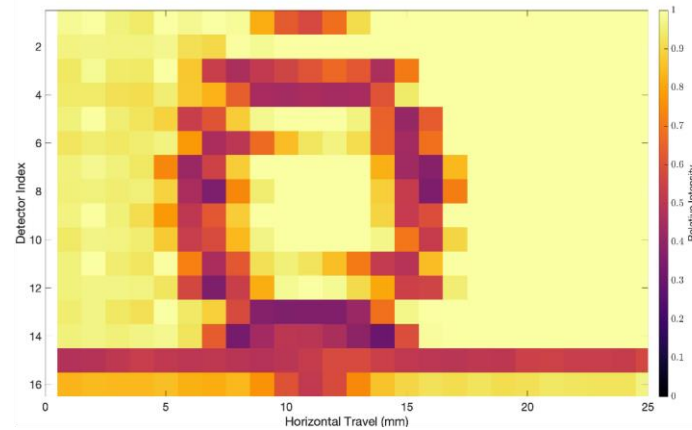
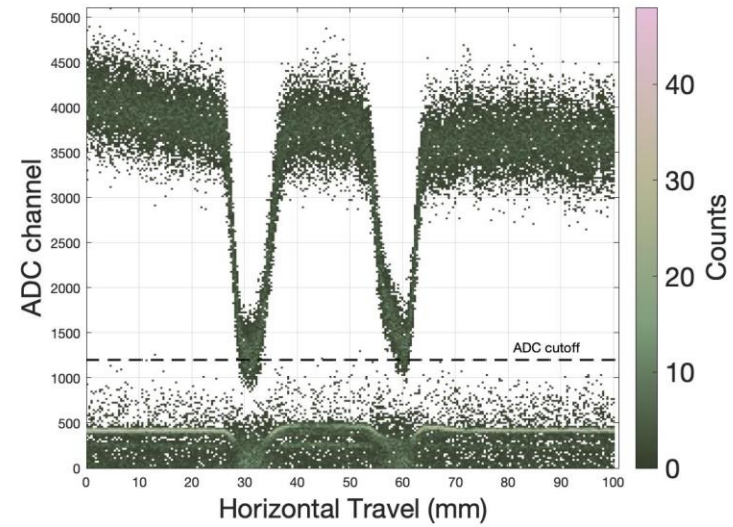
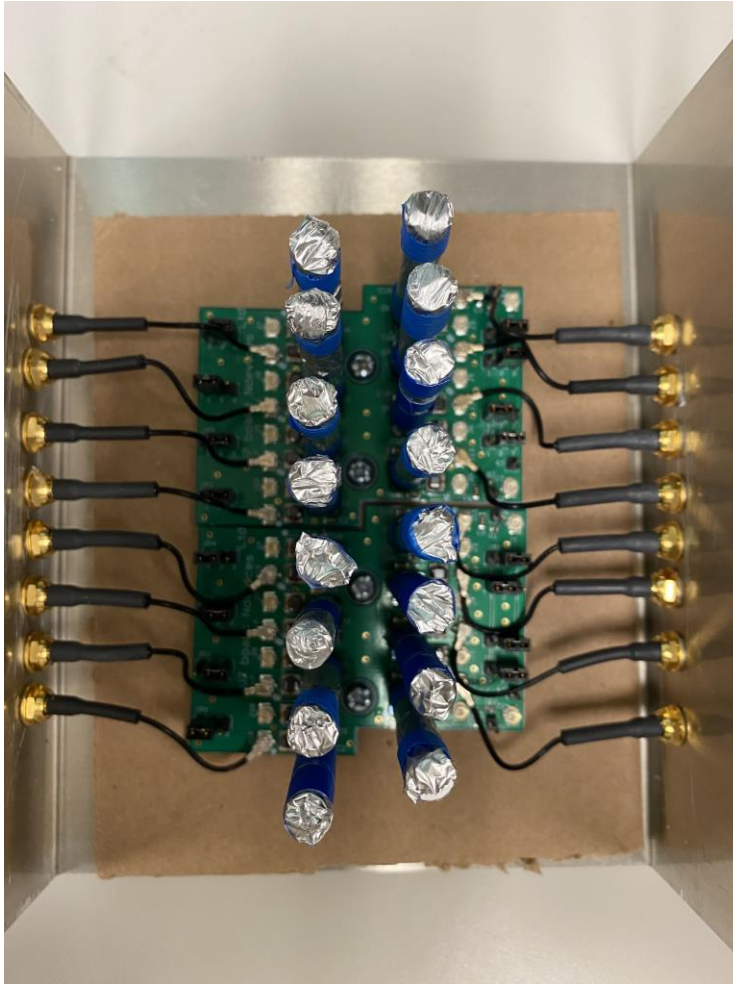


Image Reconstruction Algorithm Development



Maloney, Luke et al. "Assessment of image reconstruction algorithm coupled with fine-resolution array of Cherenkov detectors." *Scientific Reports* 12 (2022)

Acknowledgement

This work is supported by the Department of Energy / National Nuclear Security Administration under Award Number(s) DE-NA0003921.

Thank you



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Polysiloxanes as alternative to PVT matrices

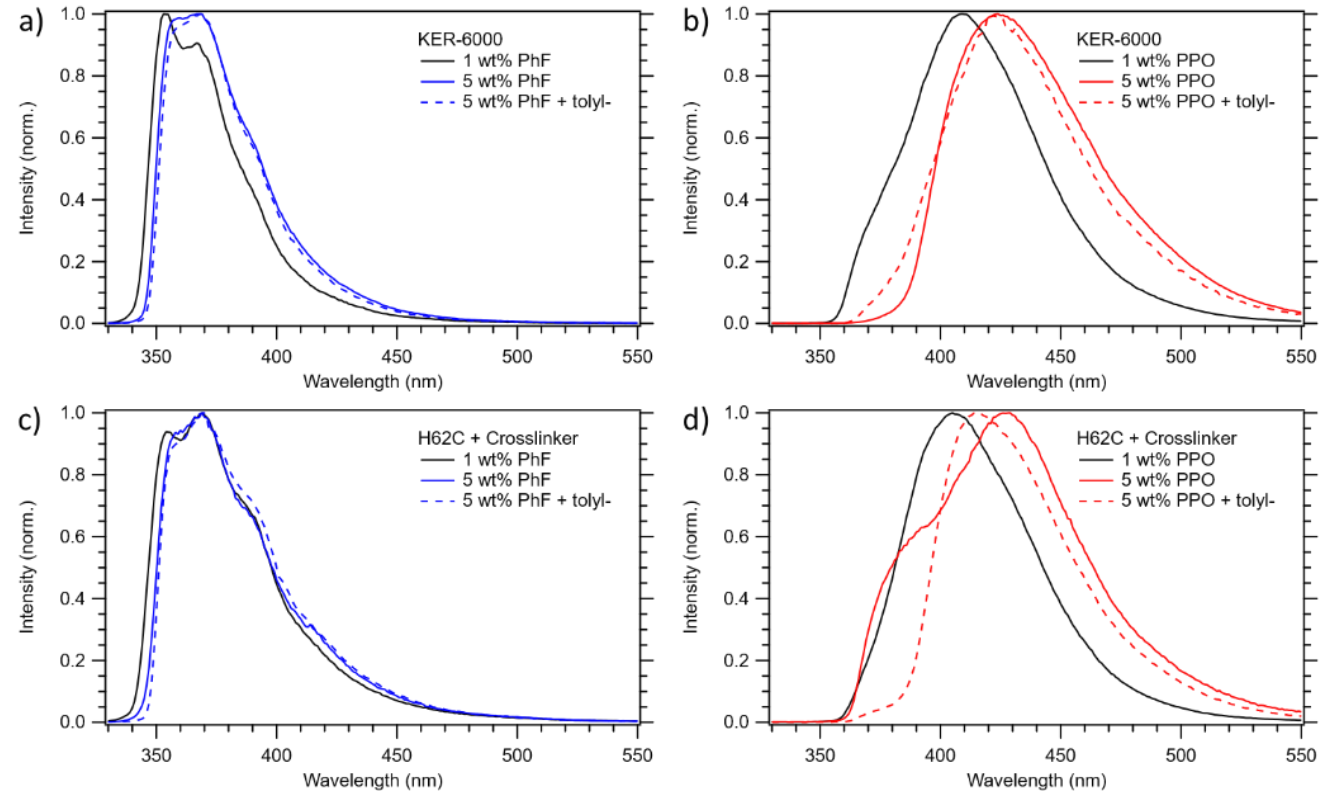


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- Internship at LBL supporting the EOS demonstrator
- Presentation at IEEE MIC/NSS 2022
 - Arrue, J., Chandler, C., Duce, M., Sellinger, A., Erickson, A., “Boron Doped Polysiloxane Organic Scintillators for Thermal and Fast Neutron Detection Via Pulse Shape Discrimination”
- Collaboration with Colorado School of Mines
- Ex-Core measurements with Texas A&M



PSD Capable Polysiloxane-based Scintillators



Fluorescence emission, samples excited at 290 nm

