Reactor Antineutrino Detector Development at LLNL using PSD capable Plastic Scintillators with Li-6 Doping

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Applied Antineutrino Physics 2019

Lawrence Livermore National Laboratory

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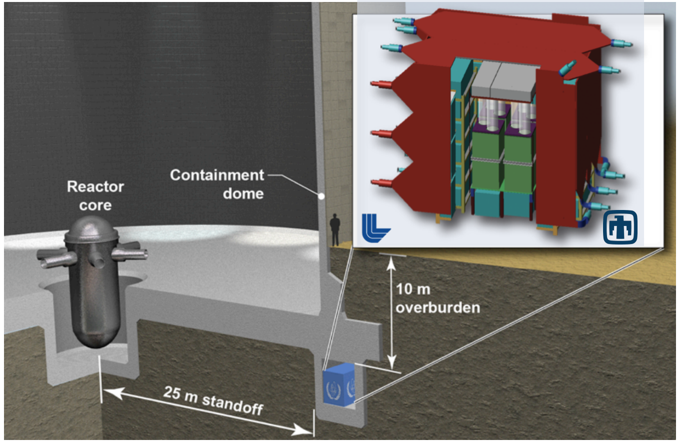
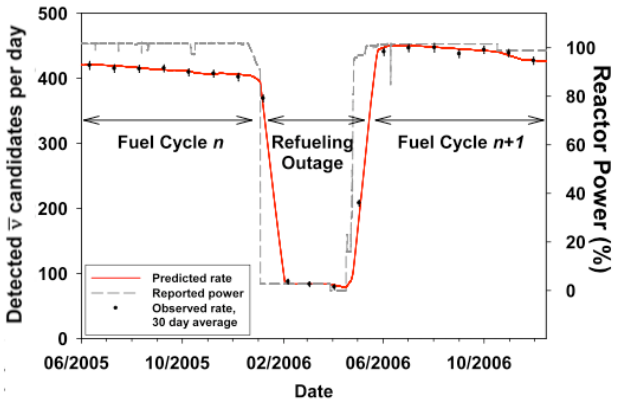
Reactor Monitoring using Antineutrino Detection

- Tools and techniques from neutrino physics have enabled remote unattended reactor monitoring demonstrations, *using Inverse Beta Decay*
- Provides means to verify reactor operational history & fuel loading
- Aboveground operation and packaging/mobility are among the utility considerations raised by potential end-users

IAEA Report STR-361



Schematic diagram of SONGS1 monitoring detector deployed at the San Onofre Nuclear Generating Station



SONGS1 data (points) compared to expectation (red) showing sensitivity to power history and fuel burnup

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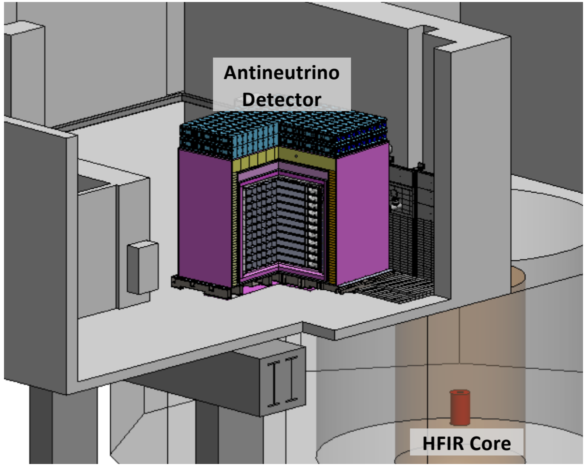
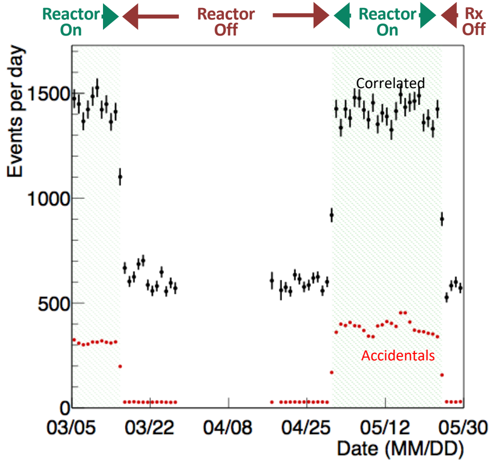


*Schematic diagram of **PROSPECT** experiment at High Flux Isotope Reactor (HFIR), **with almost no overburden***



***PROSPECT** measurement of the HFIR antineutrino flux with >1:1 S:B on the earth's surface*

Reactor Monitoring using Antineutrino Detection

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- Provides means to verify reactor operational history & fuel loading
- Aboveground operation and **packaging/mobility** are among the utility considerations raised by potential end-users


IAEA
International Atomic Energy Agency
Atoms for Peace

Final Report:
Focused Workshop on
Antineutrino Detection
for Safeguards Applications

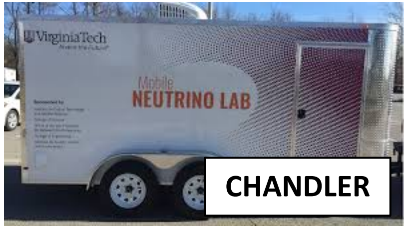
IAEA Report STR-361


Angra


PANDA

VIDAAR


LLNL/SNL

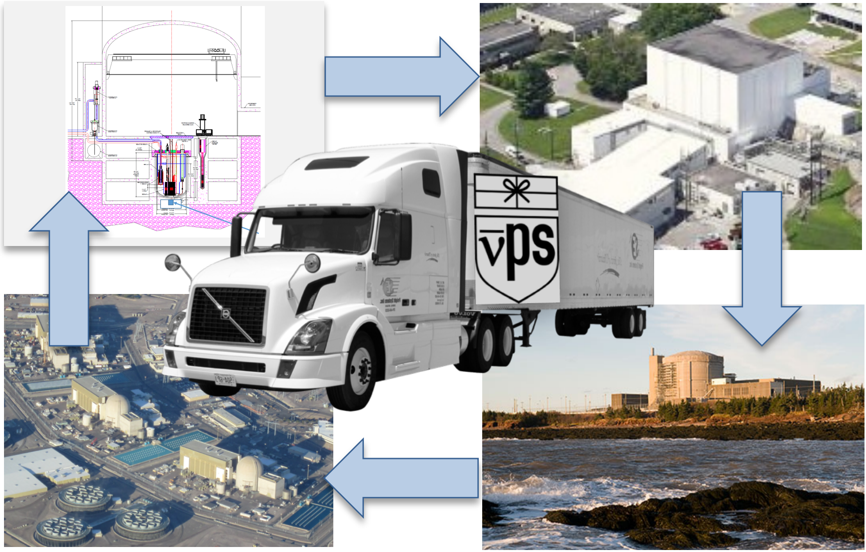


CHANDLER

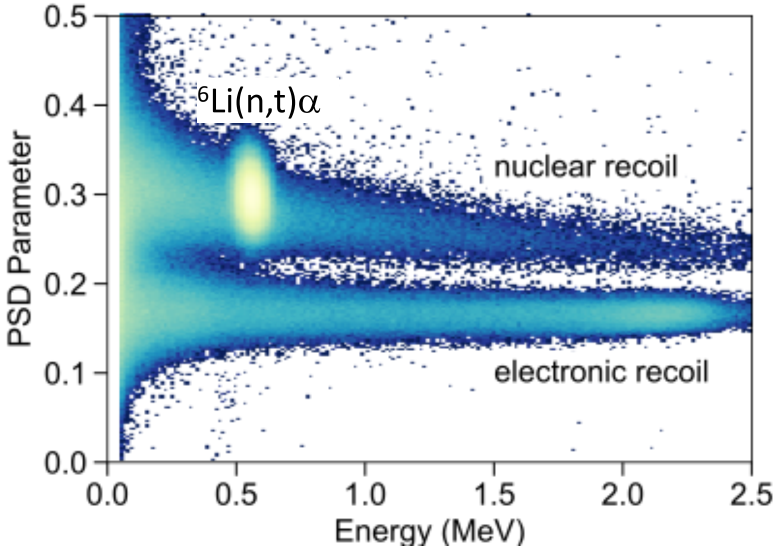
Additional motivation for high sensitivity mobile aboveground systems

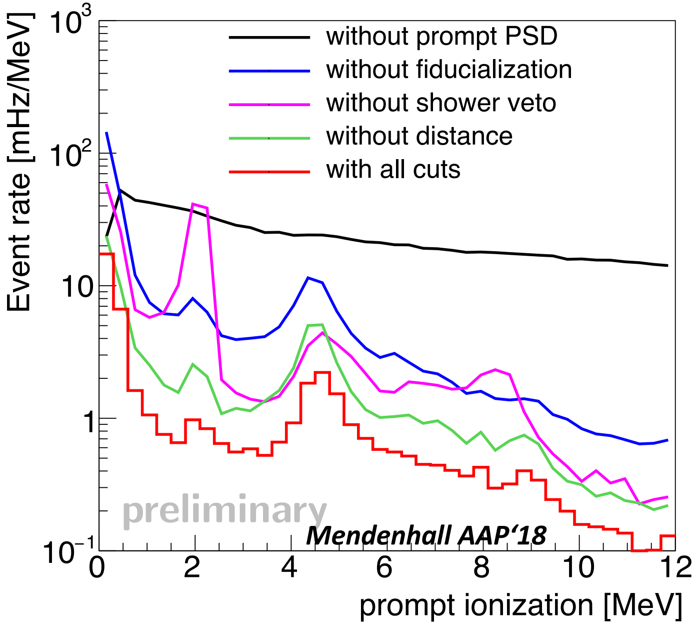
Multi-reactor type spectrum benchmarking campaign with same mobile detector design

- Same detector has never measured spectrum from more than one reactor
- For example, in North America could measure spectrum from ^{235}U research reactor, PWR, CANDU, VTR, …?
- More work needed to define resolution, systematic, and statistical/sensitivity requirements for such a campaign…



Approach to aboveground IBD detection: Particle Identification via ^{6}Li(n,t)α, PSD, segmentation

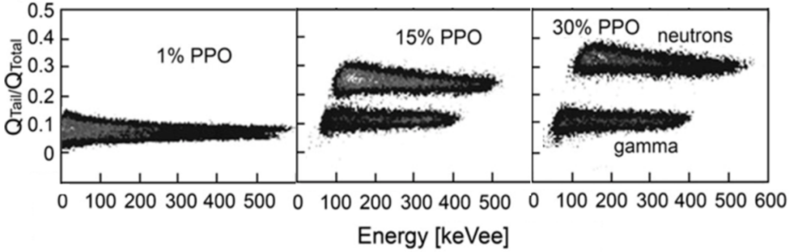


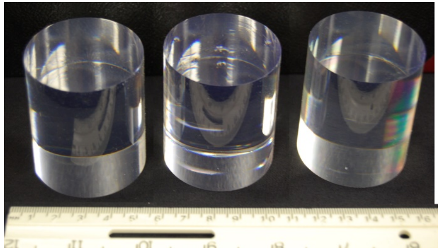


To pursue high sensitivity mobile antineutrino detection systems, our focus is on materials that provide both fast neutron recoil and 6Li(n,t)α identification

Materials development at LLNL:
Plastic Scintillator with Enhanced Delayed Light: *PSD plastics*

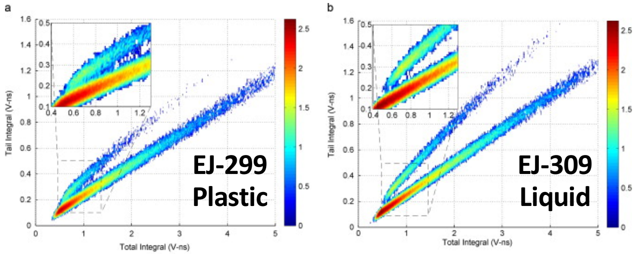
- High concentrations (>20%) of fluorescent dyes increases fraction of delayed light

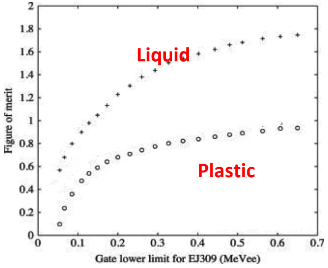




N. Zaitseva et. al., *Nucl. Instrum. Meth. A.*, 668 (2012) 88.

- Technology was licensed for commercial production by *Eljen (EJ-299)* in 2012





S. Pozzi et. al., *Nucl. Instrum. Meth. A*, 723 (2013) 19.

C. Lawrence et. al., *Nucl. Instrum. Meth. A*, 759 (2014) 16.

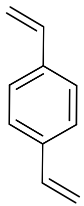
- Tests of the first EJ-299 plastics showed PSD performance inferior to liquid scintillators (*EJ-309*)

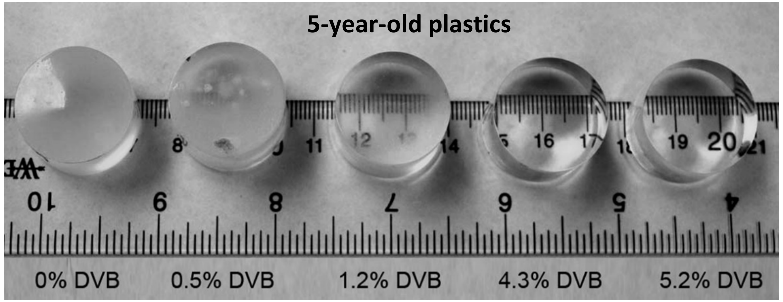
Improve PSD in Plastics to the Level of Liquid Scintillators

Critical modifications made in plastic composition

Matrix		Primary Dye		Secondary Dye (Waveshifter)
PS	+	20-36%	+	~0.2%

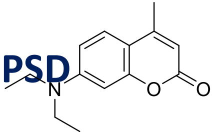
DVB (divinylbenzene) crosslinker added to the matrix: Thermoset Scintillator





- DVB improves mechanical & thermal properties
- Enhances resistance to dye precipitation
- No physical degradation with DVB > ~2%

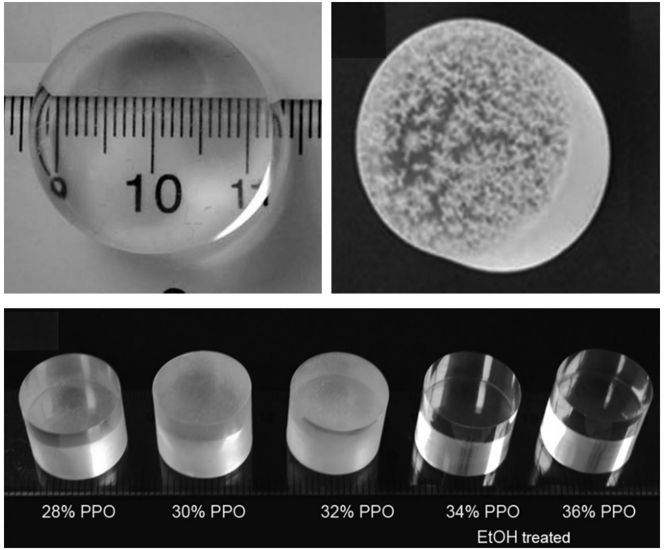
New secondary dye *(MDAC)* found to enhance LO and PSD

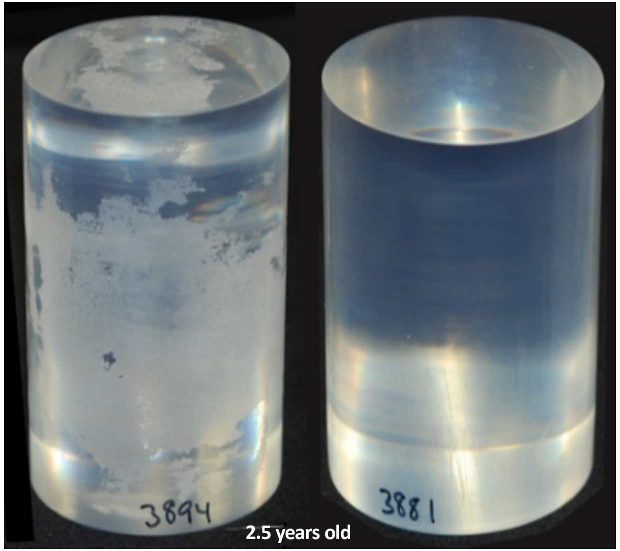


N. Zaitseva et. al., *Nucl. Instrum. Meth. A*, 889 (2018) 97.

Post-Treatment Found to Prevent Dye Precipitation & Leaching

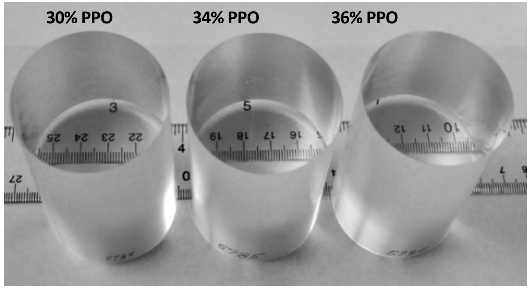
- **Simple Ethanol treatments prevents PPO leaching and surface degradation**

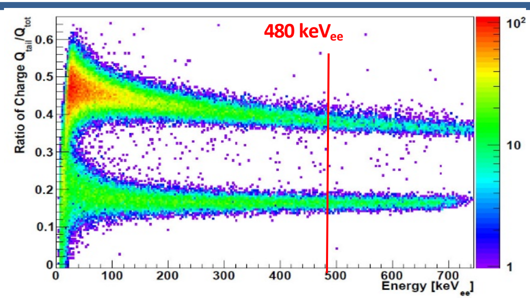


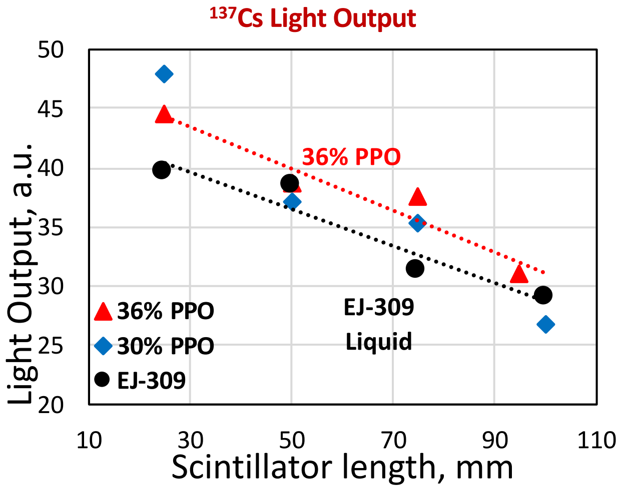


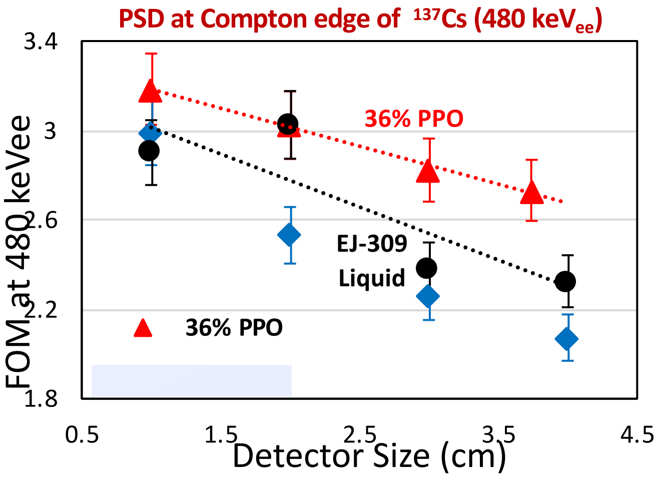
- **Modifications enabled production of stable plastics with PPO load up to 40%**
  - **Compared to previously used maximum 30%**

Modified Plastics Show LO & PSD Better than Commercial Liquid Scintillators



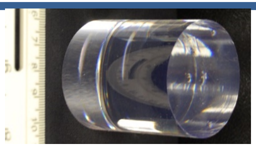


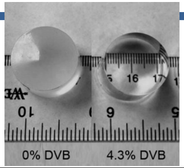




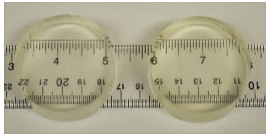
N. Zaitseva et. al., *Nucl. Instrum. Meth. A*, 889 (2018) 97.

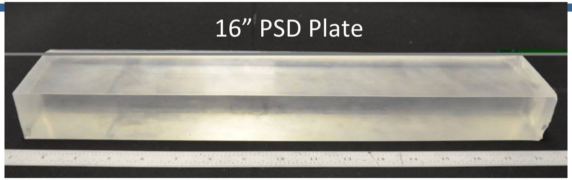
Highlights in the Development of PSD Plastics Formulations





Development of Purification Protocols





Commercial Production of PSD Plastics

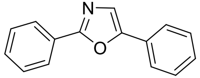
Development of Mechanical Stabilization

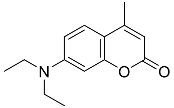
Observation and Diagnosis of Aging Behavior

Development of Scaling Techniques for > 1 kg

2012 — 2013 — 2014 — 2015 — 2016 — 2017 — 2018 — 2019

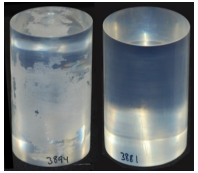
“Exhaustive” Evaluation of Primary and Secondary Dyes



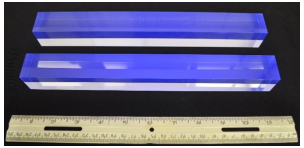


Evaluation of Boron-Loaded PSD Plastics (DTRA)

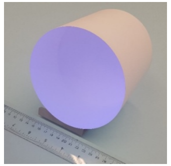
Development of Post-Treatment

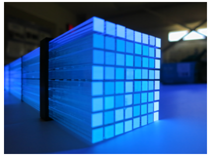


Analytical Development of Polymerization Profiles



Commercial Production of EJ-276

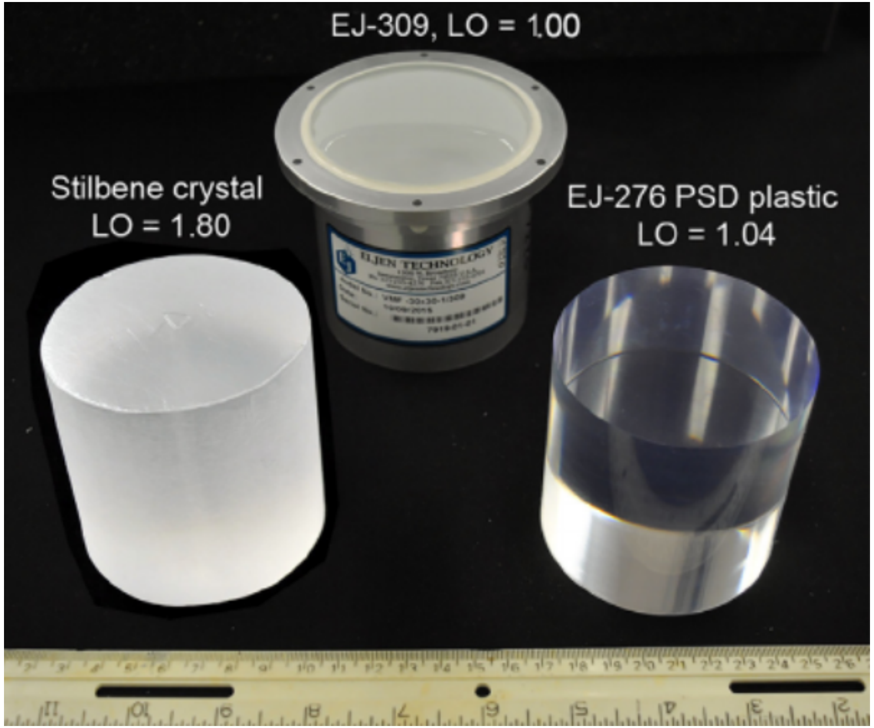


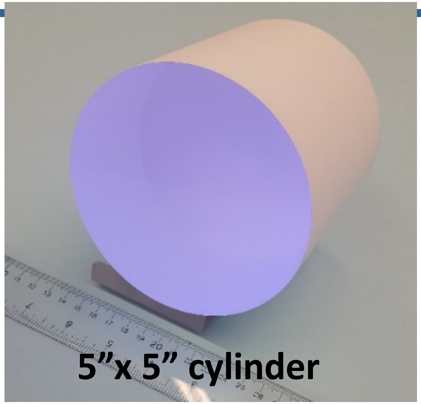


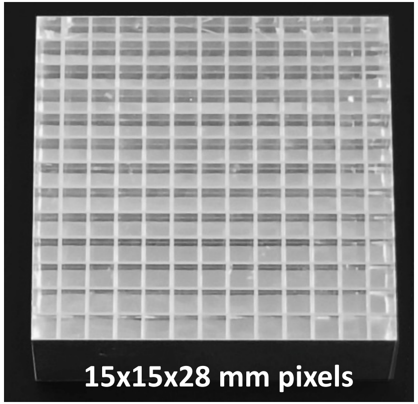
Development of Lithium Complexation Chemistry

Development of SANDD Directional Detector based on Li Plastics

Commercialization of New Formulation: EJ-276



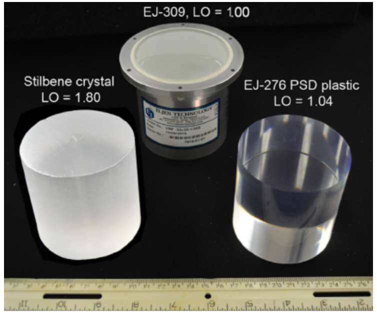


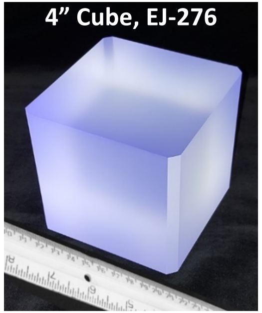


Commercial Production of EJ-276 Courtesy of Eljen Technology
(http://www.eljentechnology.com/)

Lithium-Loaded PSD Plastics

- Work on lithium is an extension from developing new PSD plastics based on high load of primary dye PPO (EJ-276)*

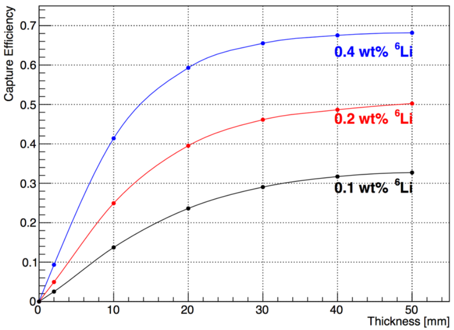




Thermal Neutron Capture Reactions

Nuclide	*Reaction*	*σ*	*Q (MeV)*
^{3}He	^{3}He(n,p^{+})α^{2+}	5333 b	0.764 MeV
^{6}Li	^{6}Li(n,α^{2+})^{3}H^{+}	940 b	4.78 MeV
^{10}B	^{10}B(n,^{7}Li*)α^{2+}	3835 b	2.792 MeV
	^{10}B(n,^{7}Li*)α^{2+}	3835 b	2.310 MeV
^{113}Cd	^{113}Cd(n,γ)^{114}Cd	20,600 b	Various
^{115}In	^{115}In(n,γ)^{116}In	202 b	Various
^{157}Gd	^{157}Gd(n,γ)^{158}Gd	259,000 b	7.94 MeV

GEANT4 Simulation for 49cm^2 plastic scintillator



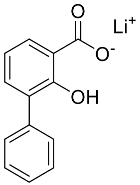
- Regularly produce 0.2% ^{6}Li loading
  - Users report that 0.2% ^{6}Li is adequate for neutron imaging
  - Antineutrino reactor monitoring project requires 0.1% ^{6}Li
  - PROSPECT uses liquid scintillator 0.07% ^{6}Li
- We use 0.1% ^{6}Li as our regular test composition

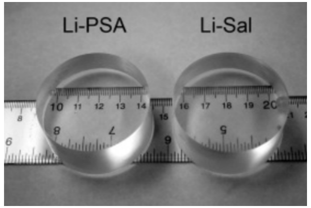
*N. Zaitseva et. al., *NIM. A*, 889 (2018) 97-104
Commercial production courtesy of Eljen Technology.

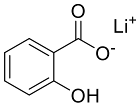
Organolithium Compounds and Plastic Scintillators

- Organolithium salts are characterized by organic group, carboxylate group, and lithium ion
- Insoluble in precursor monomer solutions (e.g., styrene)
- Use different strategies to incorporate into plastic scintillators, depending on the chemistry of the lithium salt
- To date, have tested >50 organolithium compounds and fabricated around 400 lithium-loaded plastics

Aromatic Lithium Salts

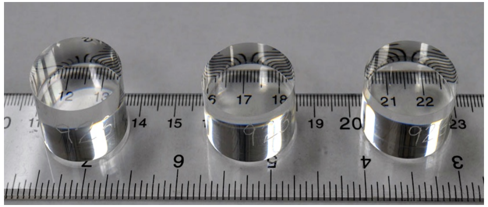




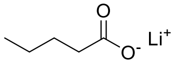


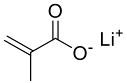
0.28% ^{6}Li **0.40% ^{6}Li**

Aliphatic Lithium Salts



0.5% ^{6}Li

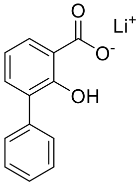


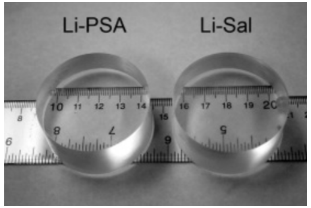


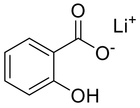
N. Zaitseva et. al., *NIM. A*, 729 (2013) 747-754
A. Mabe et. al., *NIM. A*, 806 (2016) 80-86

Strategies for incorporating Li Compounds

Aromatic Lithium Salts







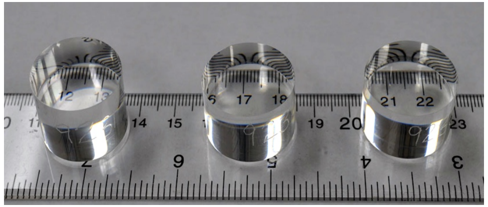
0.28% ^{6}Li **0.40% ^{6}Li**

To disperse in plastic scintillator:

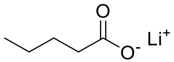
- Use polar complexing solvent containing electron donating atom (acetone, tetrahydrofuran, etc.)
- Add polar nonaromatic comonomer (e.g., poly(methyl methacrylate), PMMA)

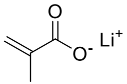
***But**,* lithium salts and PMMA matrix reduce performance → tradeoff between Li solubility, excitation production in matrix and quenching from lithium salt

Aliphatic Lithium Salts



0.5% ^{6}Li



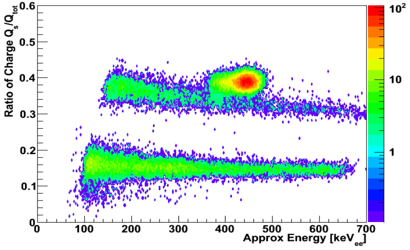


- Form a complex and copolymerize with PMMA to prevents lithium from phase-separating

***But**,* PMMA matrix reduces performance → experiment with different aliphatic compounds (chain length) to optimize

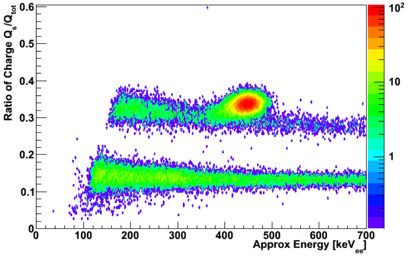
Comparison of Aromatic and Aliphatic Lithium in PSD plastics

Aromatic Lithium (0.1% ^{6}Li)
LO = 72%
FOM (n_{th}/γ) = 3.91



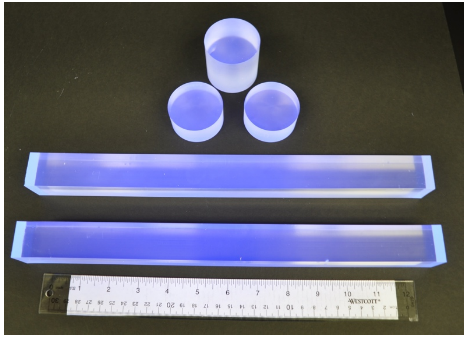
Lower LO
Better PSD
Difficult fabrication
Scalable

Aliphatic Lithium (0.1% ^{6}Li)
LO = 95%
FOM (n_{th}/γ) = 3.59



Higher LO
Lower PSD
Easier fabrication
Scalable

Aliphatic lithium formulation scalable to large size



Investigation of multiple chemistries and techniques is developing a fundamental understanding of scintillation quenching mechanisms and improved performance

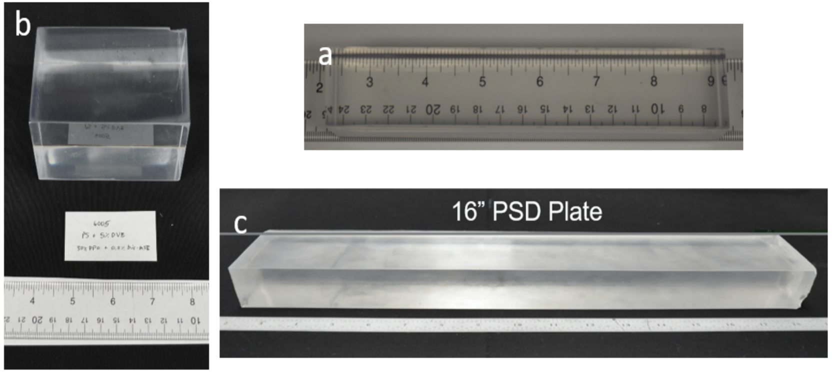
Scaling Lithium Plastics to Large Size

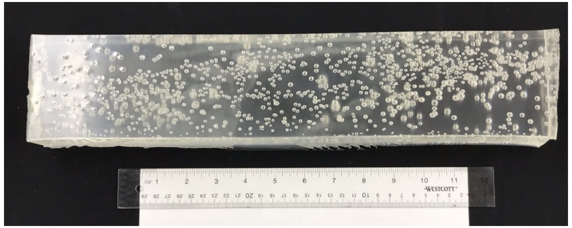
Scaling of Standard PSD plastics

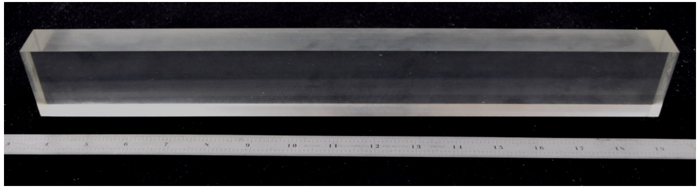
- Bubbling & yellowing are problems
- Global optimization of cook program uses kinetic models
- To hate: Have produced 2 kg scale element

Scaling of Lithium PSD plastics

- Similar problems as standard PSD plastic, but more reactive → runaway polymerization
- To date: using lower temperature profile, have produced bubble-free 500 g elements (2” x 1” x 16”)

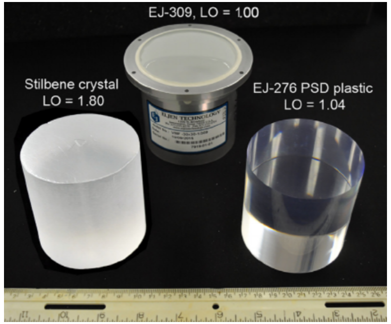


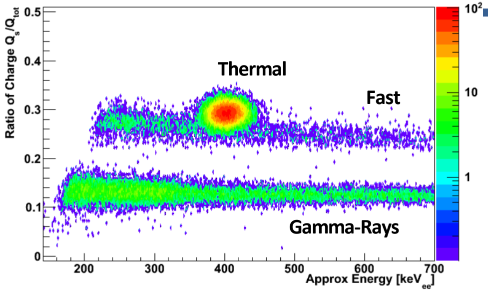


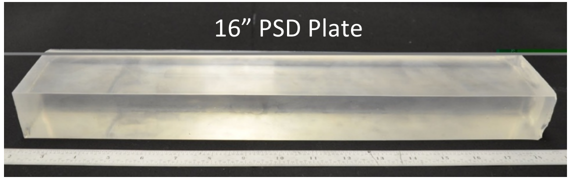


PSD Material summary and Future Work

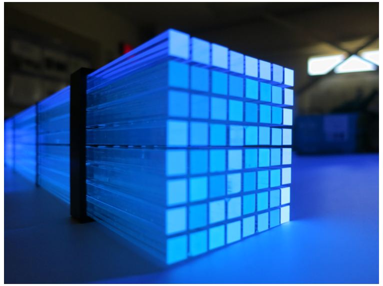
- PSD plastics markedly improved
  - Can increase loads up to 36% PPO
  - New secondary dye MDAC
  - Can produce at kg / half-meter scale
  - New commercial product: EJ-276







- Lithium doped materials
  - Strategies developed for both aromatic and nonaromatic compounds
  - Can produce plastics up to 0.5% ^{6}Li
  - Aromatic Li gives better PSD, but nonaromatic Li gives better LO
  - Current/Future materials work:
    - Understand and mitigate quenching
    - Improve manufacturing process and understand cost drivers in detail
    - Investigate these ^{6}Li chemistries in liquid scintillator



SANDD: A small directionally sensitive detector prototype

Aim for as ~cm-scale position resolution to provide IBD directionality

Full detector → 3 detector granularities

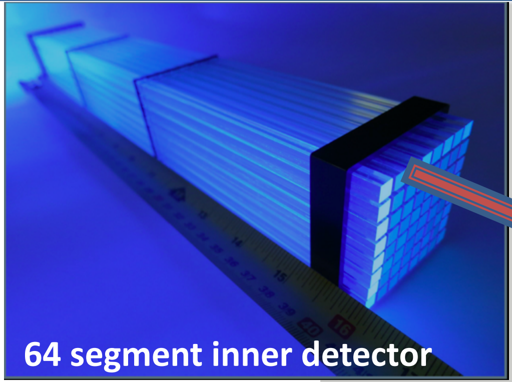
1. Central module – 5 mm x 5 mm x 40 cm segments
2. Middle section – 2.5 cm x 2.5 cm x 40 cm segments
3. Outer section – 5 cm x 2.5 cm x 40 cm segments

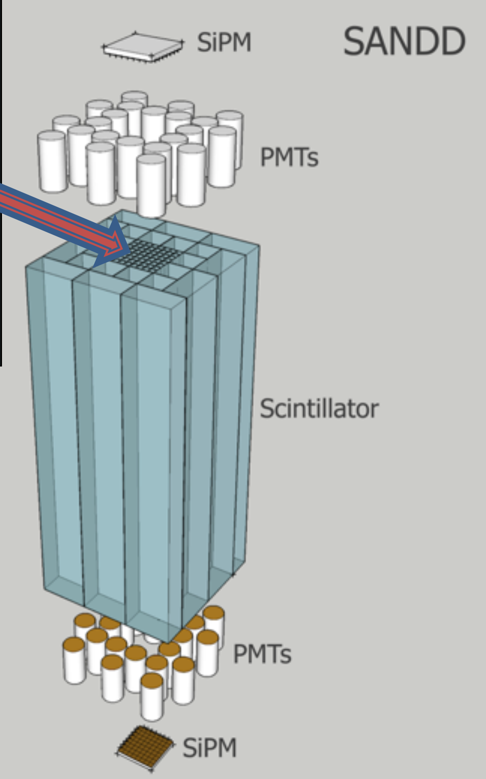
Total volume

- ~10 liters → ~4 ev/day at HIFR,

  ~1 ev/day at Hartlepool (50m standoff)

- IDB efficiency ~40% - includes neutron capture efficiency
- Detector placed inside ~30 → 40 cm boron doped poly shielding, surrounded by a muon veto.
- Whole detector (including shielding) – refrigerator size

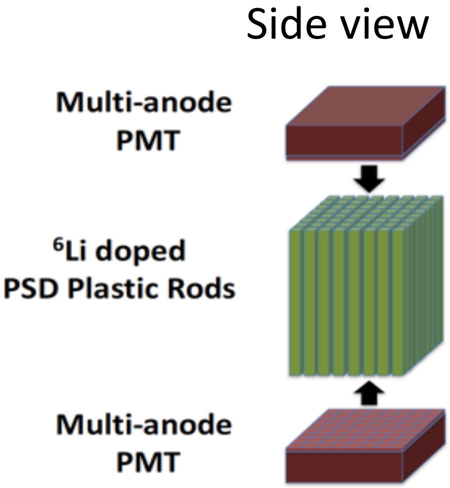

64 segment inner detector

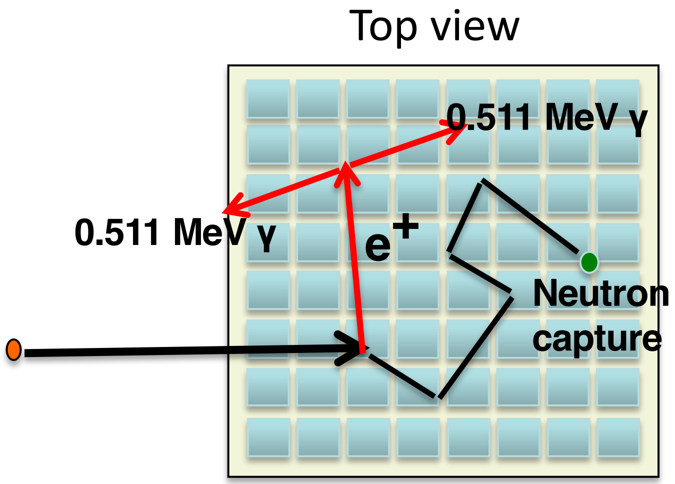


- S. Dazeley
- V. Li
- A. Mabe
- T. Classen
- F. Sutano

V. Li et al. NIMA 942, P162334 (2019)

IBD directionality in central module

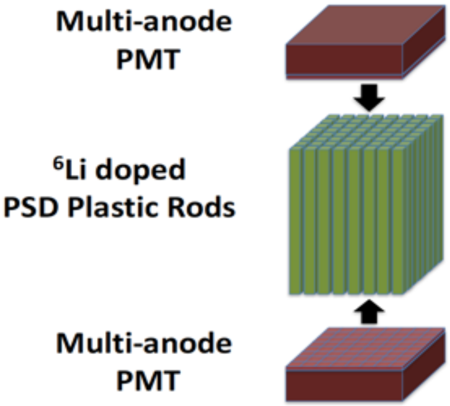




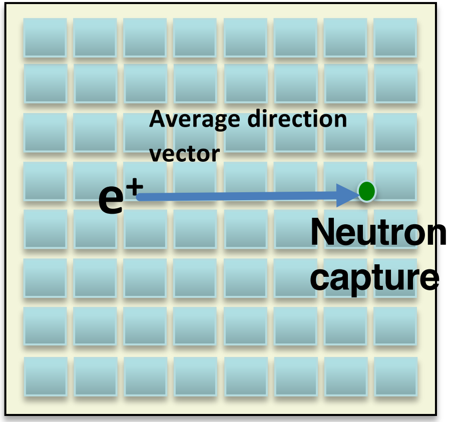
Plastic form of ^{6}Li PSD scintillator enables a highly segmented, position sensitive readout with no dead material.

IBD directionality in central module

Side view



Top view

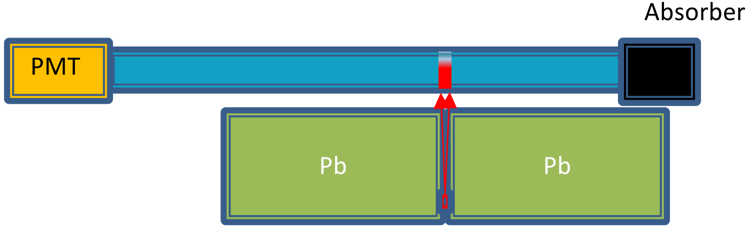


Plastic form of ^{6}Li PSD scintillator enables a highly segmented, position sensitive readout with no dead material.

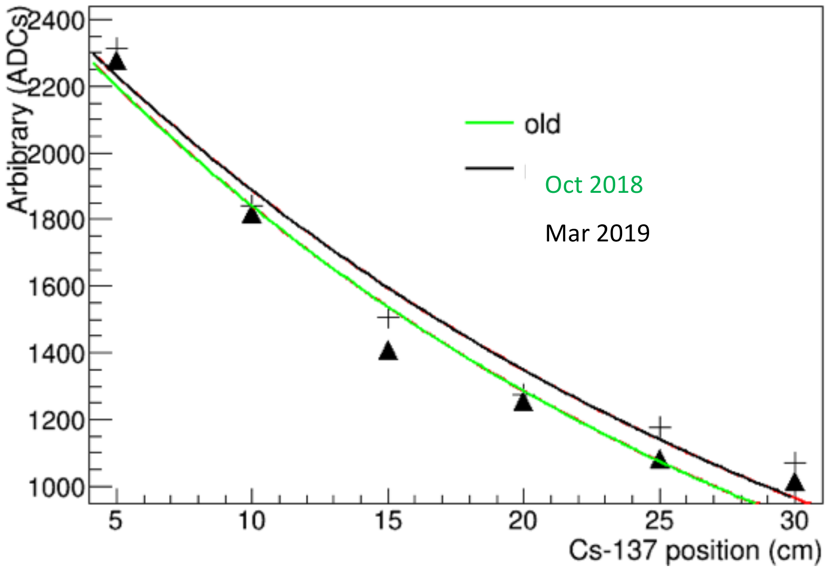
^{6}Li-doped plastic:
Apparent attenuation length measurements and stability

Attenuation setup

Scintillator: 30cm x 3cm x 3cm



- The apparent attenuation length includes effect of plastic transparency and reflectivity off walls.
- Apparent attenuation length → 25-27 cm
- Two samples stable performance for 6 months

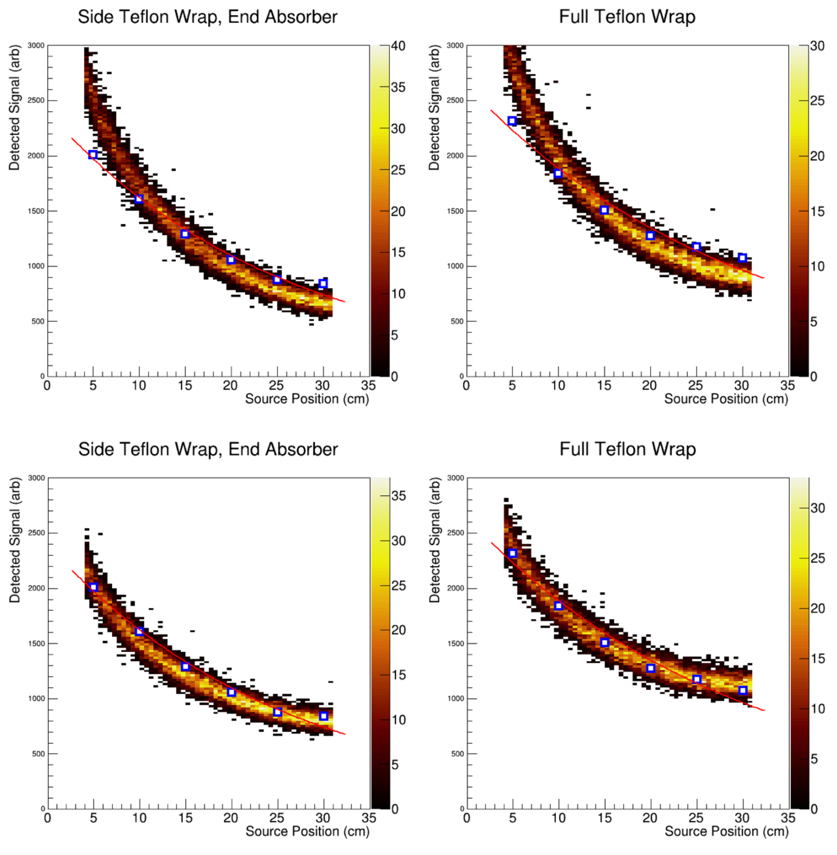


Note: single exponential is not a great fit here
Better is a double exponential – probably some wavelength dependence, or reflections?

Intrinsic attenuation length

GEANT4
30 cm
intrinsic

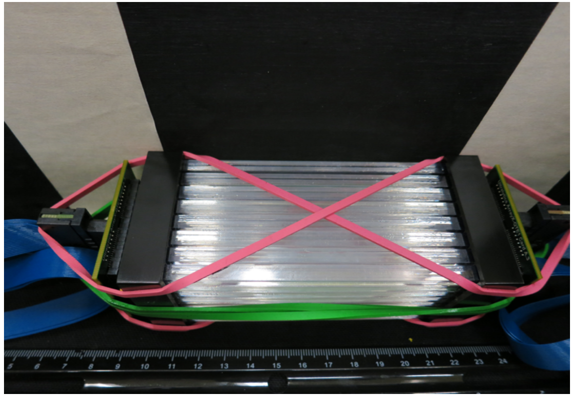
GEANT4
50 cm
intrinsic



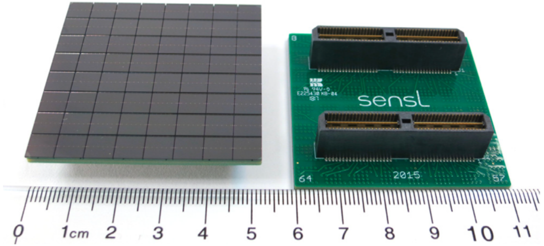
- Performance metric in small cross section bars is apparent attenuation length – includes intrinsic + reflectivity
- Attenuation results compared with GEANT4 simulation to get rough idea of the intrinsic attenuation length
- Best fit ~50 cm, and continues to improve with new development iterations

First prototype:

Pulse Shape Sensitive Plastic plus 64 Channel SiPM Slow Signal Readout……

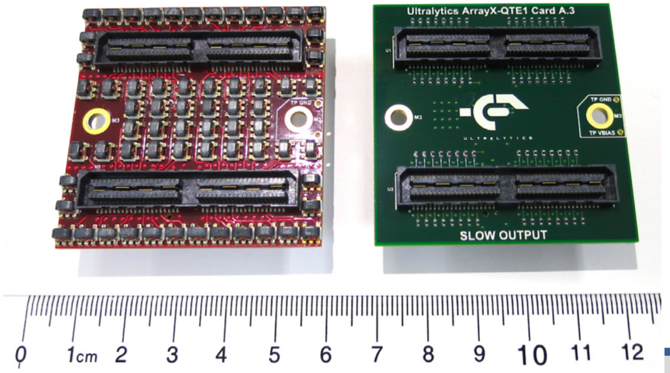


Photon readout
(SENSL J-series SiPM and new 64 channel readout)



Lack of commercial PSD-sensitive 64-channel readout for SiPM was a problem

Working with Ultralytics to solve this problem



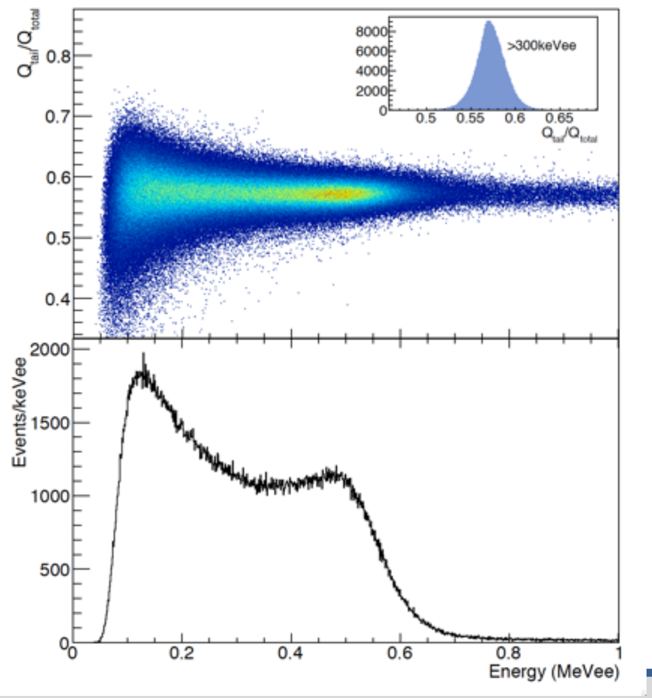
Advantages/Disadvantages

	SiPMs	Traditional PMTs
Quantum efficiency	40% → 50%	25% → 35%
Speed	~few ns	~few ns
Consistent Ch to Ch response	YES	NO
Cross talk	NO	YES

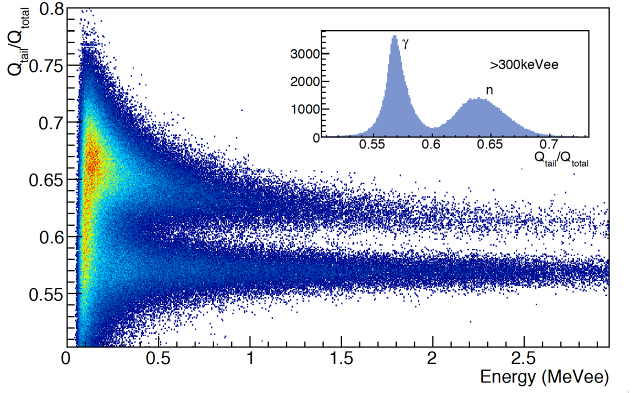
Prototype characterization published in V. Li et al. NIMA V942, P162334 (2019)

First prototype:
In early 2019 we began testing the new Ultralytics 64-channel readout

Cs-137 PSD and spectrum

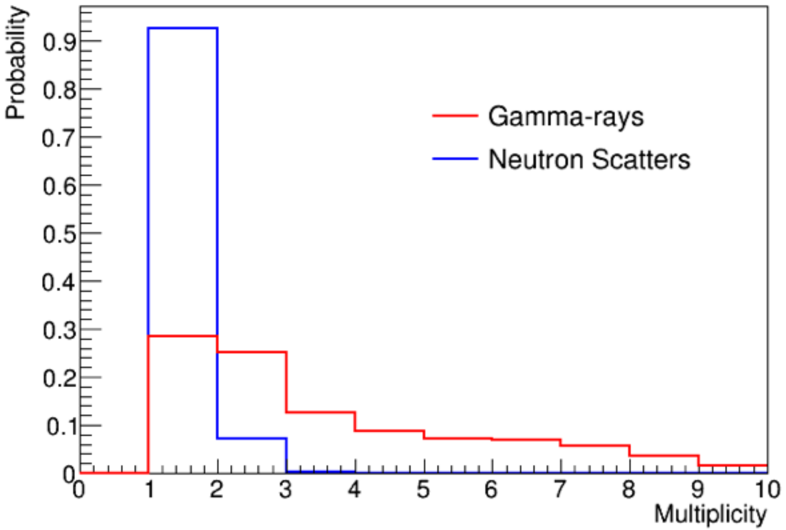


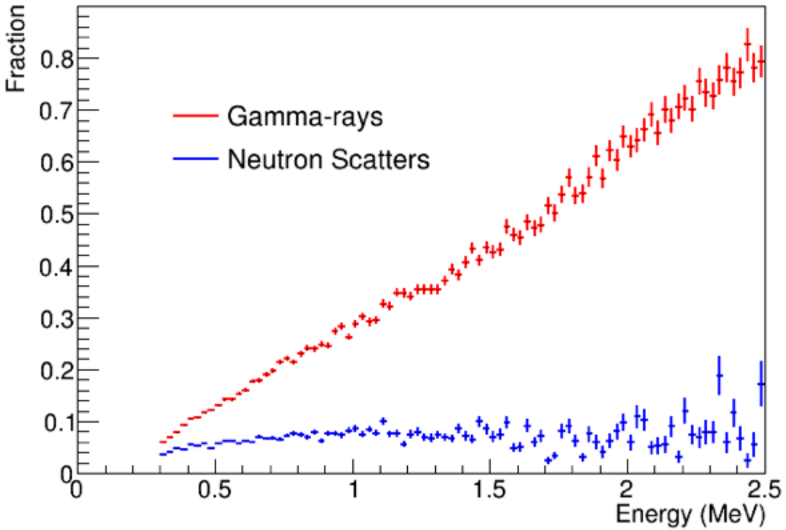
Cf-252
Pulse shape sensitivity with prototype



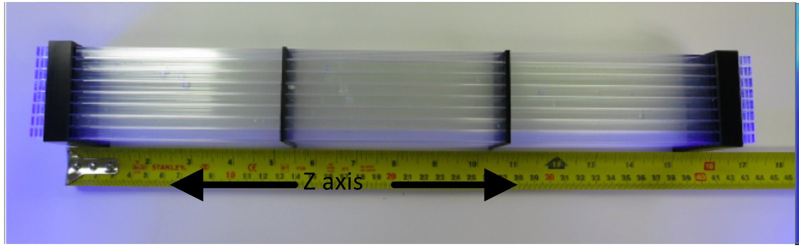
Particle ID via scintillator segment multiplicity

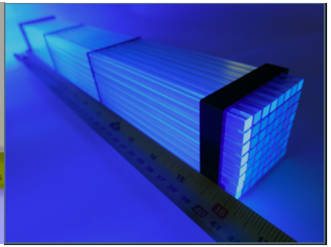
- Gain additional discrimination for/against neutron scattering via an independent measurement of particle range using segment multiplicity
- In addition to pulse shape sensitivity

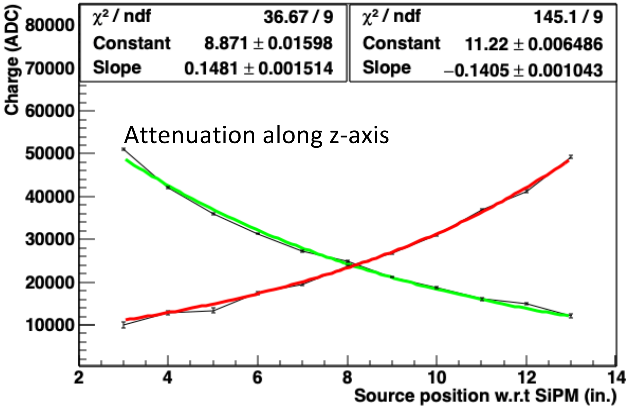




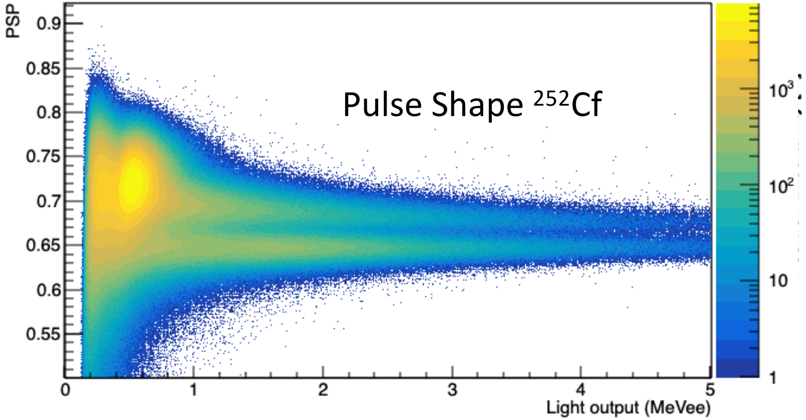
Completed central module (Preliminary results)

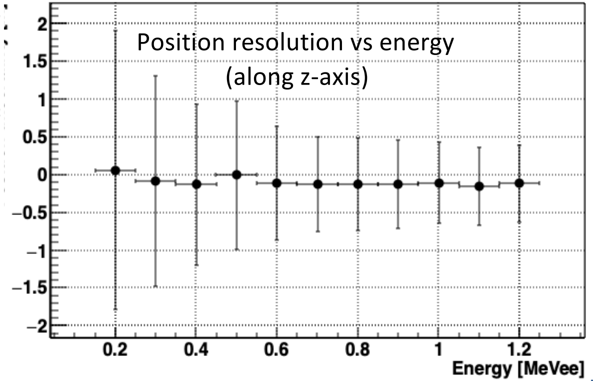






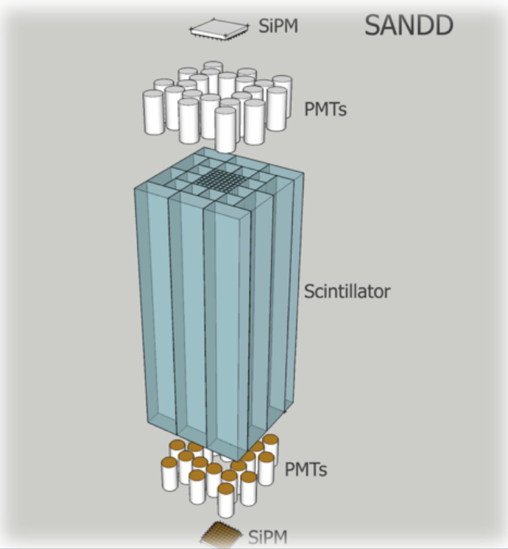
- Apparent attenuation length – 18 cm
- Position resolution @ 1 MeV (x,y) – ±0.5 cm, z – ±1.25 cm
- Energy resolution @ ^{137}Cs Compton edge – 22%

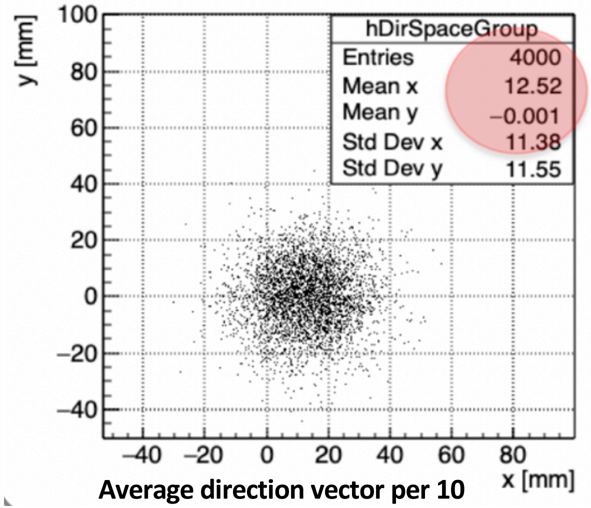




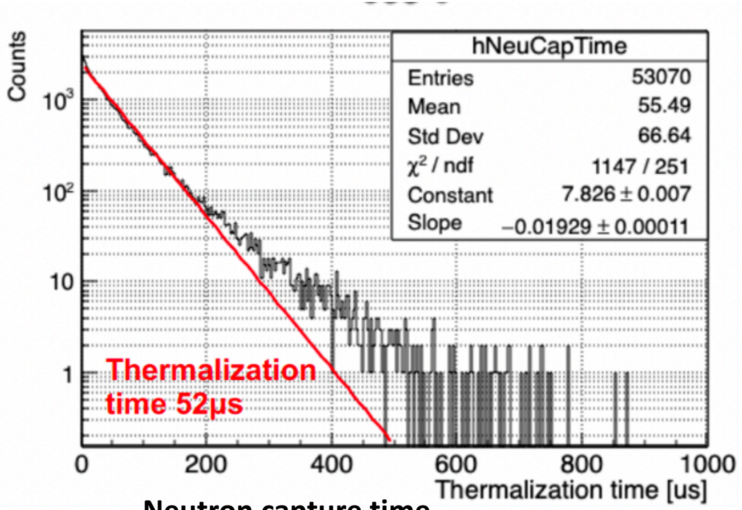
Characterization of full 10-liter SANDD

- In hand: central module and ~half outer bars; production ongoing
- Tuned simulation used for characterization of efficiency, directionality, background rejection
- Including PSD/energy/timing/multiplicity cuts Efficiency estimate ~14%





Average direction vector per 10 detected IBD events



Neutron capture time

SANDD Status:

- SANDD central module has ~1cm position resolution needed for sensitivity to antineutrino IBD direction
- ^{6}Li-doped scintillator (@0.1%) approaching ½ meter attenuation length. 50 microsecond neutron capture time
- Good light output. PSD could be better – but perhaps it is good enough for IBD
- SiPM 64-channel readout preserves PSD (a first)
- Producing remaining scintillator (about 7 liters more needed)
- Detector can be deployed in 3-6 months, possibly at HFIR (Oak Ridge NL, US), or Hartlepool (UK)

Ongoing Mobile Detector R&D

- We have just started a new effort to design and demonstrate a mobile system using ^{6}Li PSD scintillators, with a goal of resolution & light collection at least as good as PROSPECT
- Extending past work to study geometries using validated background and response simulation:
  - 2D vs 3D segmentation
  - Segmentation pitch
  - Internal dead material
- Continuing development of ^{6}Li doped PSD materials: Plastic, Liquid, and WLS PSD plastic
- Finalize design based on realized material performance & geometry studies in late-2020
- Aim for reactor deployment of ~500kg system in late-2021

Conclusion

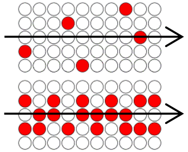
- Demonstrating ton-scale mobile systems with high resolution and sensitivity would provide important monitoring capabilities and enable benchmark spectrum measurements
- Detector designs and high performance materials that enable particle ID are key → segmentation, ^{6}Li loading, and PSD
- LLNL continues to make good progress developing ^{6}Li chemistries and production techniques for PSD capable scintillators, especially in plastic
- The first detector to use the newest generation of materials, SANDD, will provide data on the impact of very fine segmentation
- Work has started on a new project emphasizing mobility, background rejection, and energy response at the ton-scale

Backup

PSD Materials Development
Scintillation Process in Organic Scintillators

Compton e⁻

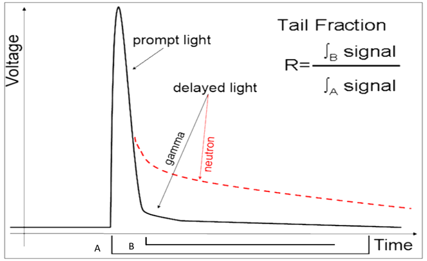
Heavy Ion

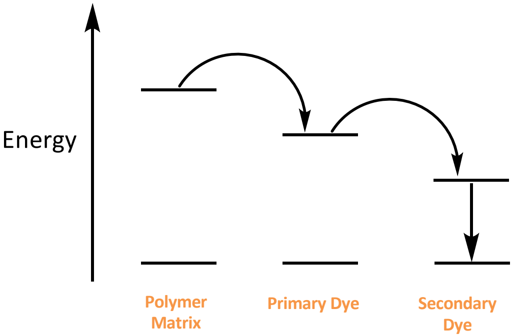


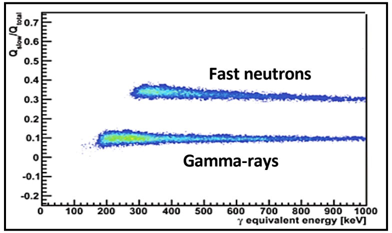
Primarily prompt light: $S_1 \rightarrow S_0 + h\nu$

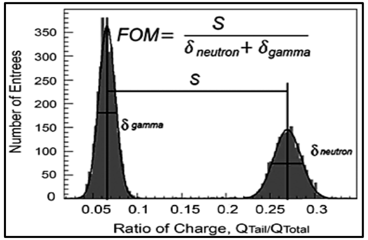
More delayed light: $T_1 + T_1 \rightarrow S_1 + S_0$

$S_1 \rightarrow S_0 + h\nu$







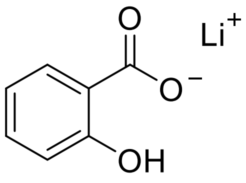


- **The PSD technique exploits difference in time characteristics in signals induced by different incident particles**

Incorporation of Aromatic Lithium Salts into Plastic Scintillators

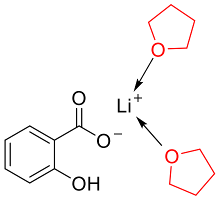
Must contain vicinal hydroxyl group

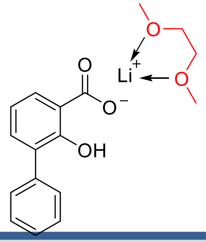
Salicylates are used in pharmaceuticals - nontoxic

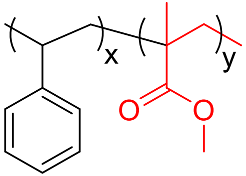


To disperse in plastic scintillator:

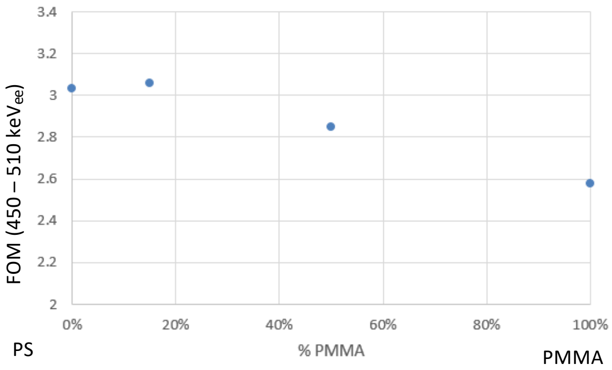
- Use **polar complexing solvent** containing electron donating atom (acetone, tetrahydrofuran, dimethoxyethane, etc.)
- Add **polar nonaromatic comonomer** (e.g., **poly(methyl methacrylate), PMMA**)







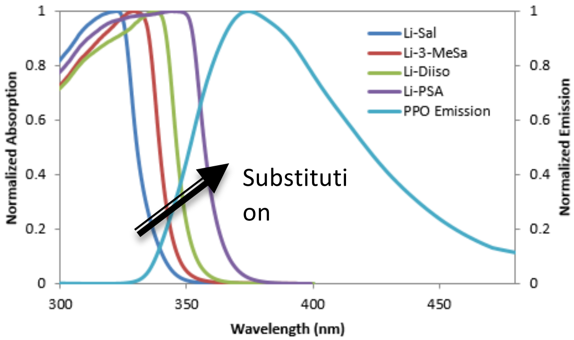
PS **PMMA**

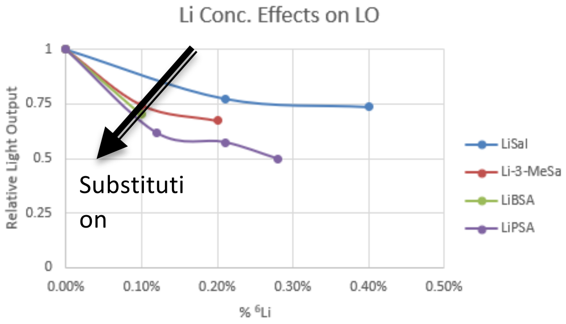


PMMA Matrix Reduces LO and PSD*

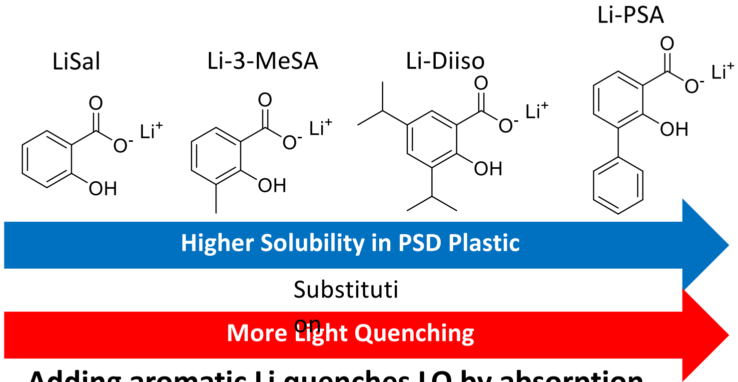
*H. P. Martinez et. al., *NIM. A*, 771 (2015) 28-31.

Light Output of Plastics with Aromatic Lithium Salts





- Aromatic lithium salts absorb PPO emission and are weak emitters
- Adding groups around ring red-shifts absorption edge



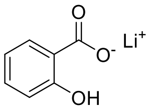
- Adding aromatic Li quenches LO by absorption
- Fraction of LO lost follows trend in absorption edge
- More overlap with PPO emission results in more light quenching

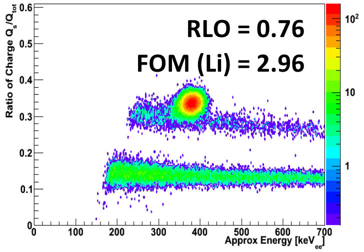
“Triple PSD” of Plastics with Aromatic Lithium

Substitution reduces amount of PMMA needed: Improves PSD

(RLO is relative to standard unloaded PSD plastic)

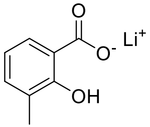
LiSal

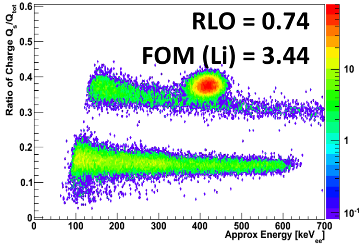




30% PS
70% PMMA

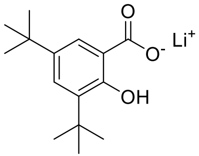
Li-3-MeSA

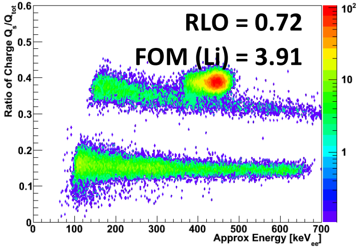




50% PS
50% PMMA

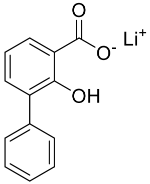
Li-BSA

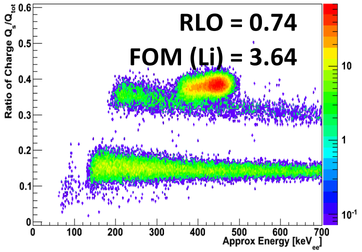




85% PS
15% PMMA

Li-PSA





85% PS
15% PMMA

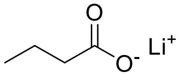
(minimum amount of PMMA needed for dissolution)

Trade-off between excitation production in matrix and quenching from lithium salt

- **Problem: These salts can be quite difficult to purify and keep clean**

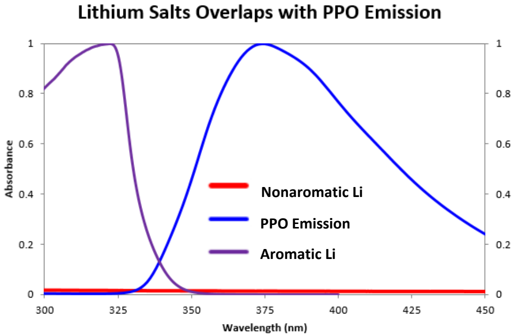
N. Zaitseva et. al., *Nucl. Instrum. Meth. A*, 729 (2013) 747-754
A. Mabe et. al., *Nucl. Instrum. Meth. A*, 806 (2016) 80-86

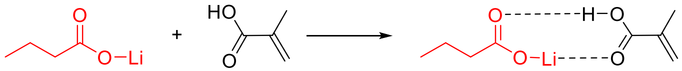
Incorporation of Nonaromatic (Aliphatic) Lithium



Aliphatic lithium salts have no chromophores

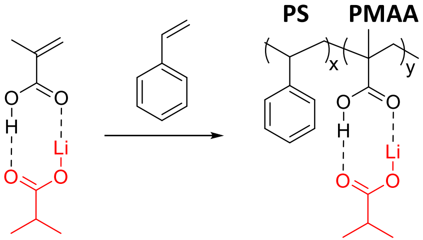
- ➢ No absorption in PPO emission region
- ➢ Hypothesis: Improved performance and attenuation properties may arise from using non-absorbing lithium compounds

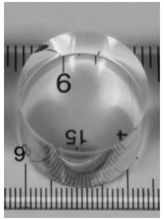




Strategy:

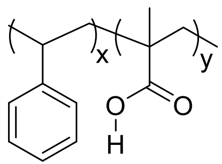
- ➢ Aliphatic lithium compounds are insoluble in organic solvents
- ➢ Form complex, copolymerize with styrene, prevents lithium from phase-separating



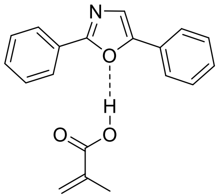


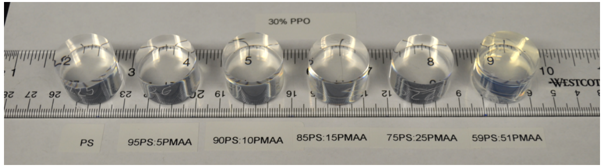
Effects of Poly(methacrylic acid) (PMAA) on Scintillation Properties

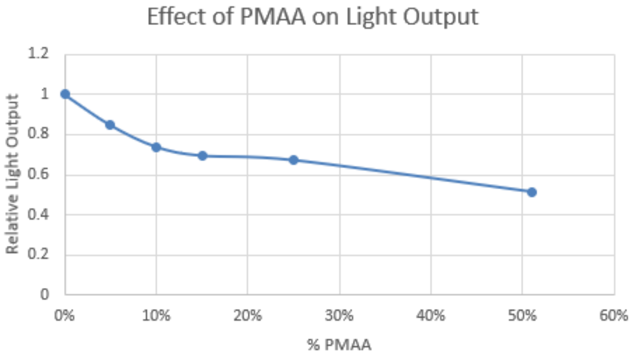
- PMAA even at 5% reduces light output by almost 20%.
  — More quenching than expected from simple dilution
  — No chromophores: suspect chemical quenching

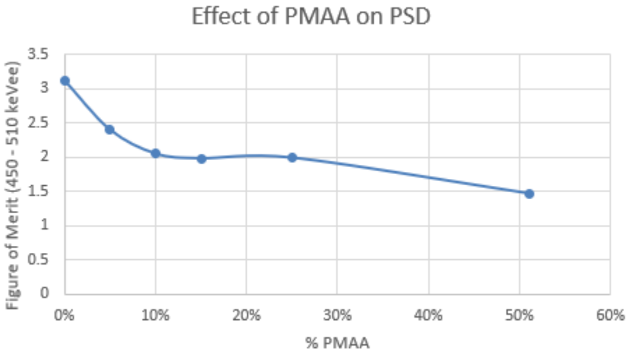


Suggested Mechanism: hydrogen bonding of MAA with PPO; produces a species with lower emission probability.
Plans to probe the existence of this species by diffusion time



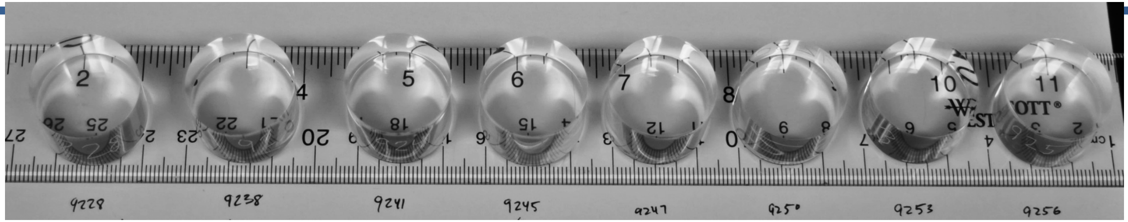


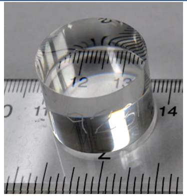




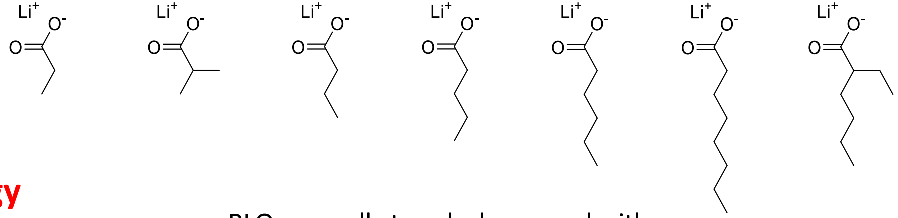
A Survey of Aliphatic Lithium Compounds in PSD Plastics

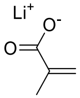
All at 0.1% ^{6}Li





No ^{6}Li

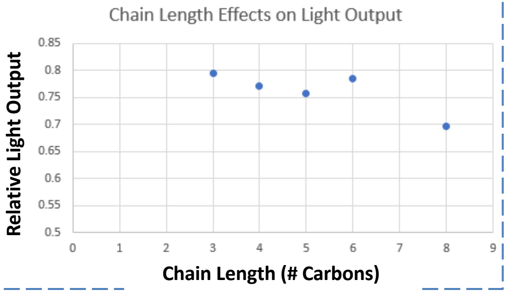




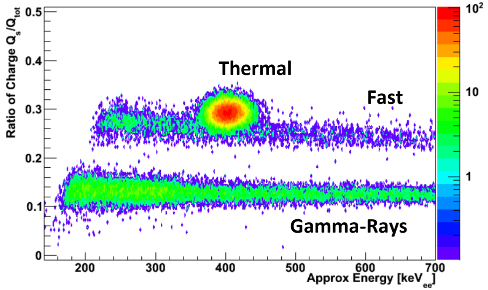
PMAA strategy works with all aliphatic lithium compounds tested.

- ➢ **Improved manufacturability relative to aromatic lithium**

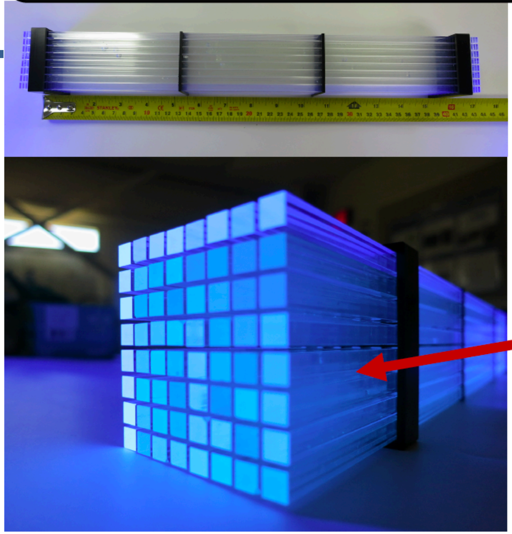
RLO generally trends downward with tail length, but the effect is small

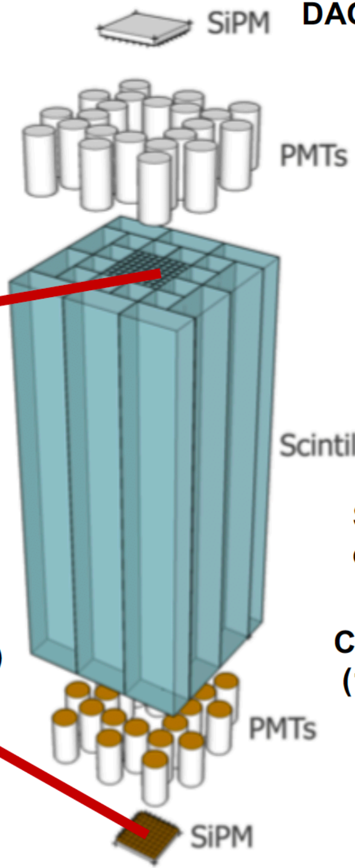


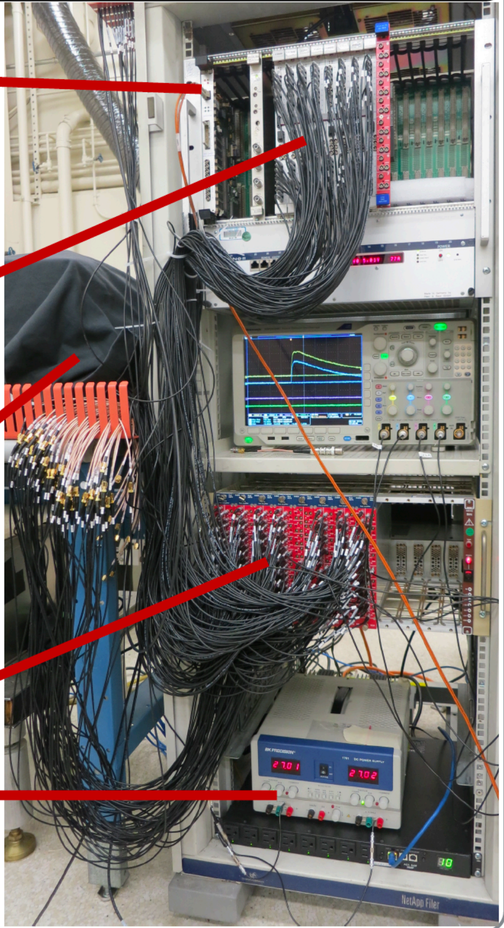
"Triple PSD" with aliphatic lithium



SANDD (Segmented AntiNeutrino Directional Detector)







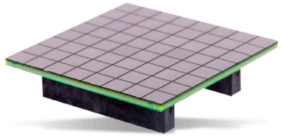
DAQ optical link

16-channel VME Struck SIS3316 digitizer (250 MS/s, 14-bit, 5 V dynamic range)

SANDD enclosure

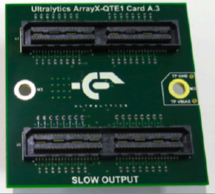
CAEN N979 (10x-gain)

SiPM Power supply



64-pixel SiPM array (SensL J-60035 series)

Slow readout board from Ultralytics



V. Li et al., NIMA 942 (2019) 162334