Lawrence Livermore National Laboratory

Plastic Scintillator Materials Development at LLNL

Andrew N. Mabe

M. Leslie Carman, Andrew M. Glenn, Steven A. Dazeley, Natalia P. Zaitseva, and Stephen A. Payne

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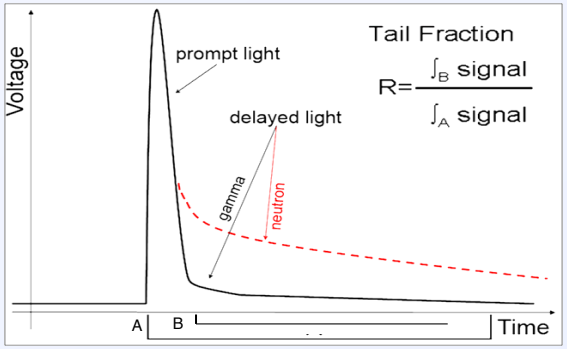
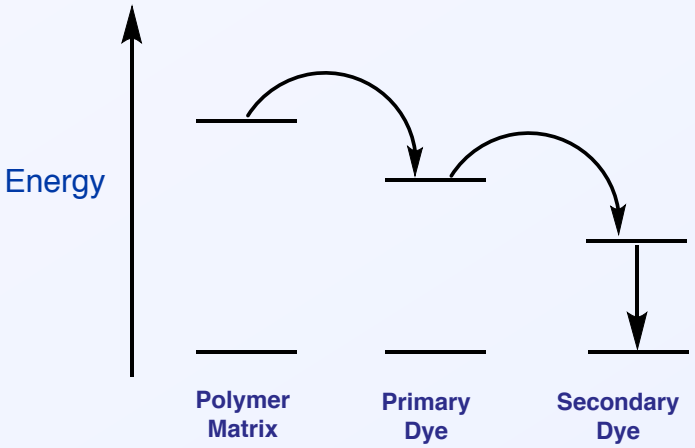
LLNL-PRES-759723

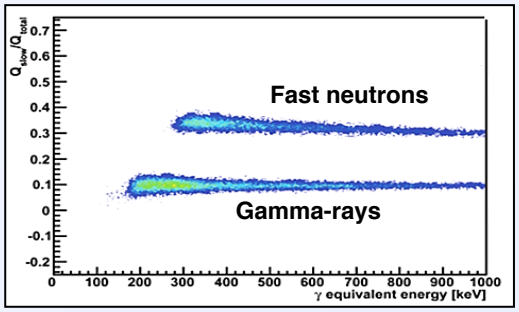
Scintillation Process in Organic Scintillators

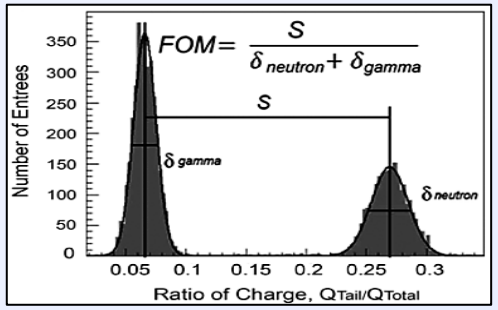
Compton e⁻ → Primarily prompt light: $S_1 \rightarrow S_0 + h\nu$

Heavy Ion → More delayed light: $T_1 + T_1 \rightarrow S_1 + S_0$, $S_1 \rightarrow S_0 + h\nu$





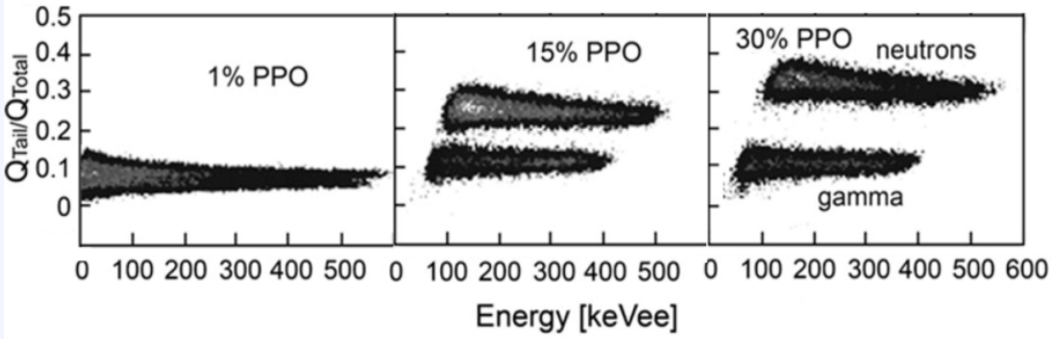


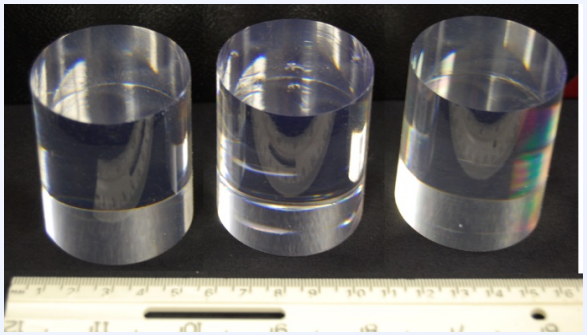


- **The PSD technique exploits difference in time characteristics in signals induced by heavy ions (recoil protons) and Compton electrons**

Plastic scintillator with enhanced delayed light: *PSD plastics*

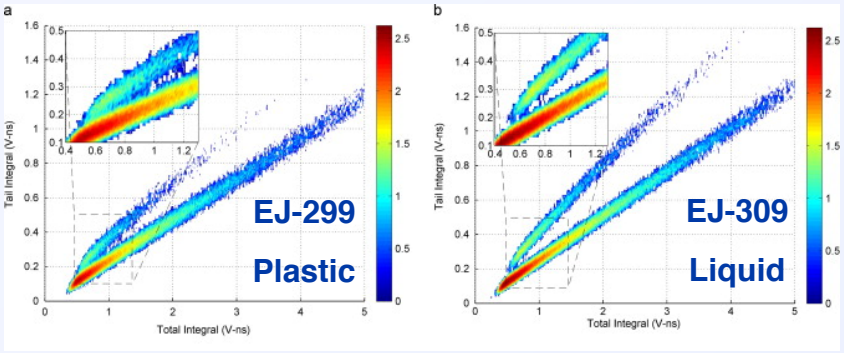
- **High concentrations (>20%) of fluorescent dyes increases fraction of delayed light**

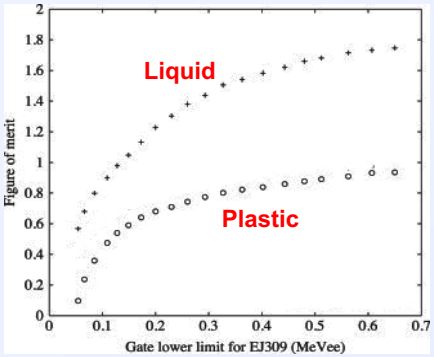




N. Zaitseva et. al., *Nucl. Instrum. Meth. A.*, 668 (2012) 88.

- **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.

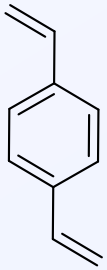
- **Tests of the first EJ-299 plastics showed PSD performance inferior to liquid scintillators (*EJ-309*)**
  - ***especially pronounced at low energies***

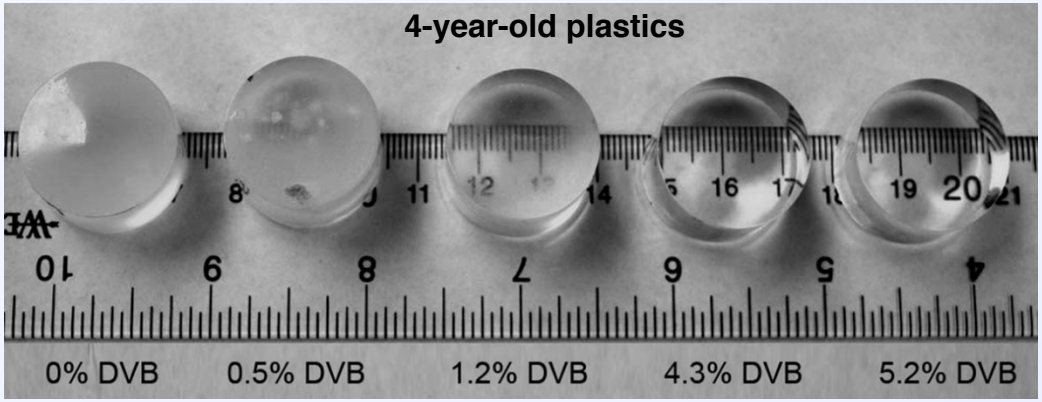
Focus of recent work: Improve PSD in Plastics to the Level of Liquid Scintillators

Critical modifications made in plastic composition

Matrix		Primary Dye		Secondary Dye (Waveshifter)
PS	+	20-36% PPO	+	~0.2% DPA, POPOP, Bis-MSB

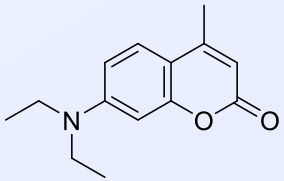
DVB (divinylbenzene) crosslinker added to the matrix: Thermoset Scintillator





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

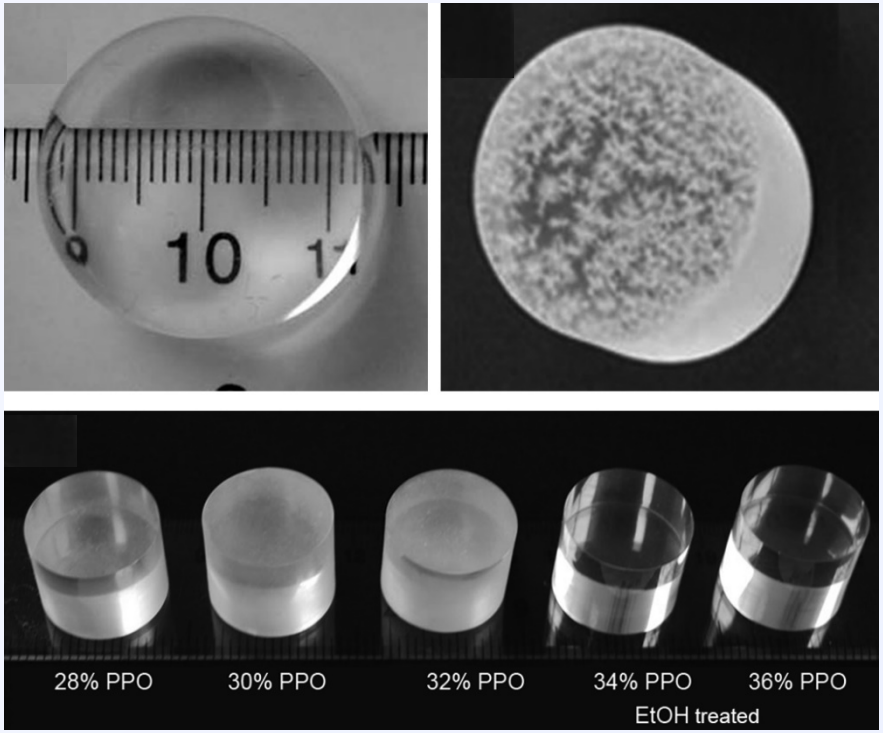
New secondary dye *(MDAC)* found to enhance LO and PSD

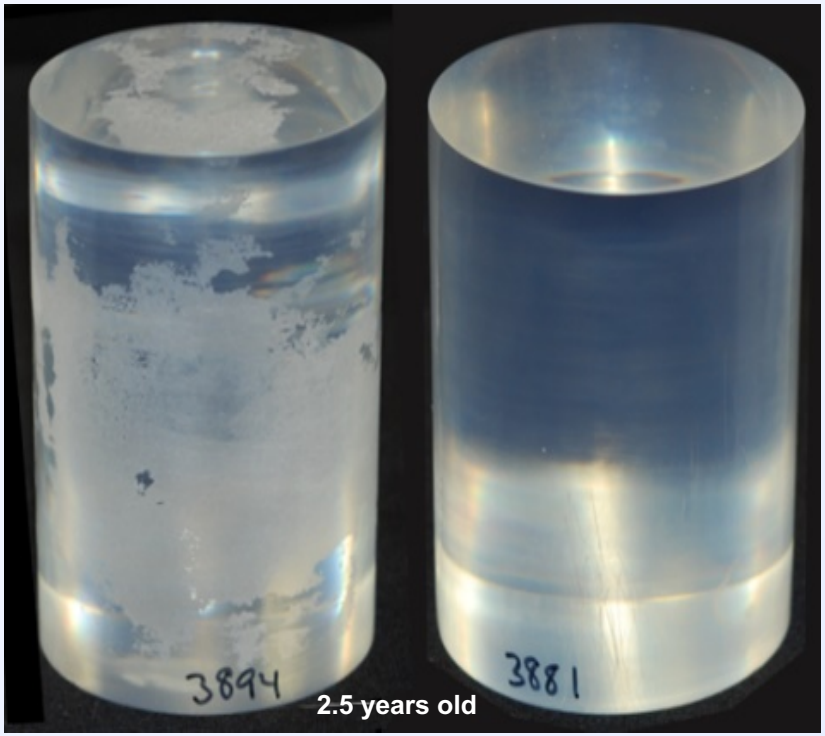


N. Zaitseva et. al., *Nucl. Instrum. Meth. A*, 889 (2018) 97.

Post-Treatment Technique Found to Prevent Dye Precipitation and Leaching

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

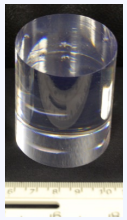


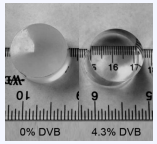


- **Modifications enabled production of stable plastics with PPO load up to 40%**
  - **Compared to previously used maximum 30%**

Highlights in the Development of PSD Plastics Formulation

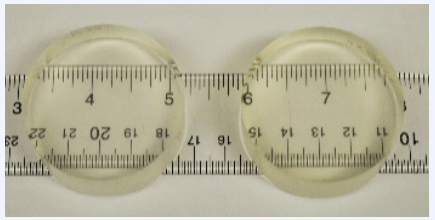
Commercial Production of PSD Plastics





Development of Mechanical Stabilization

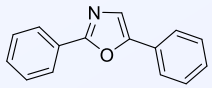
Development of Purification Protocols

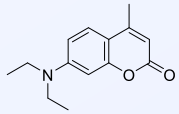


Observation and Diagnosis of Aging Behavior

2012 2013 2014 2015 2016 2017 2018

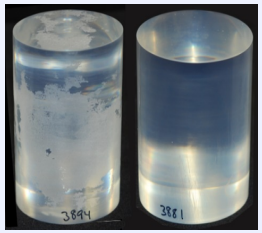
Exhaustive Evaluation of Primary and Secondary Dyes



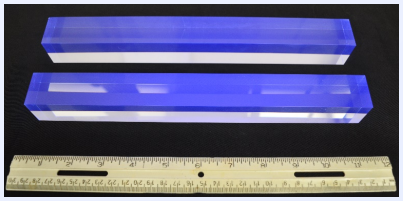


Evaluation of Boron-Loaded PSD Plastics (DTRA)

Development of Post-Treatment

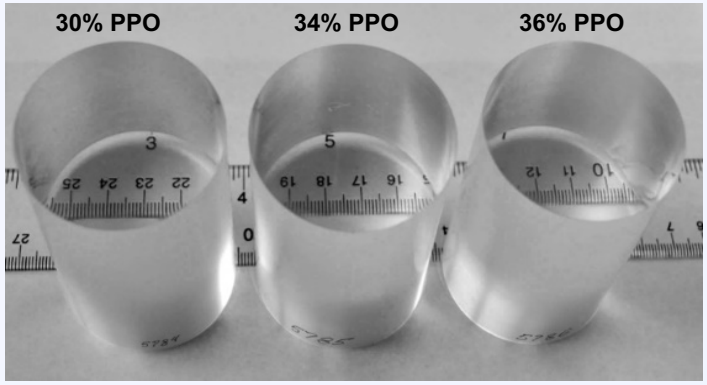


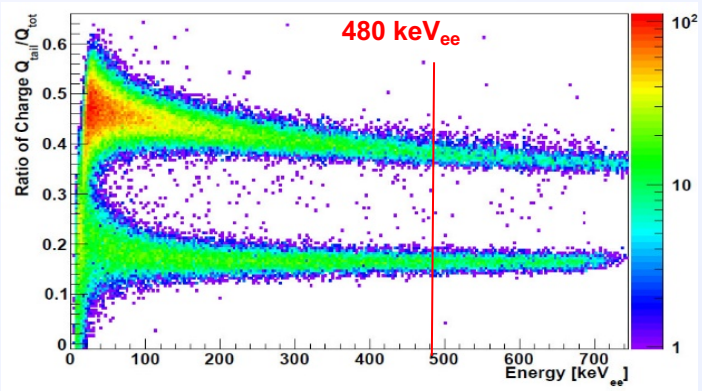
Analytical Development of Polymerization Profiles

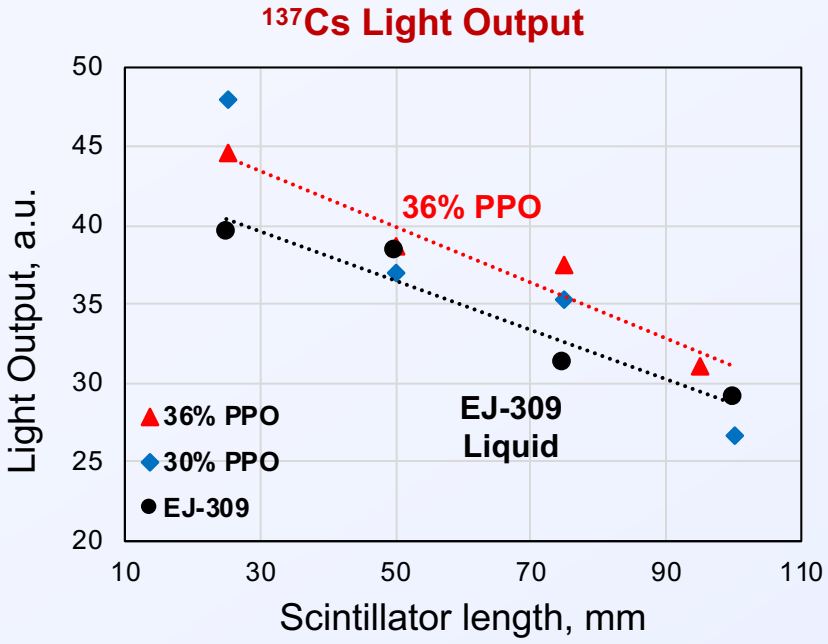


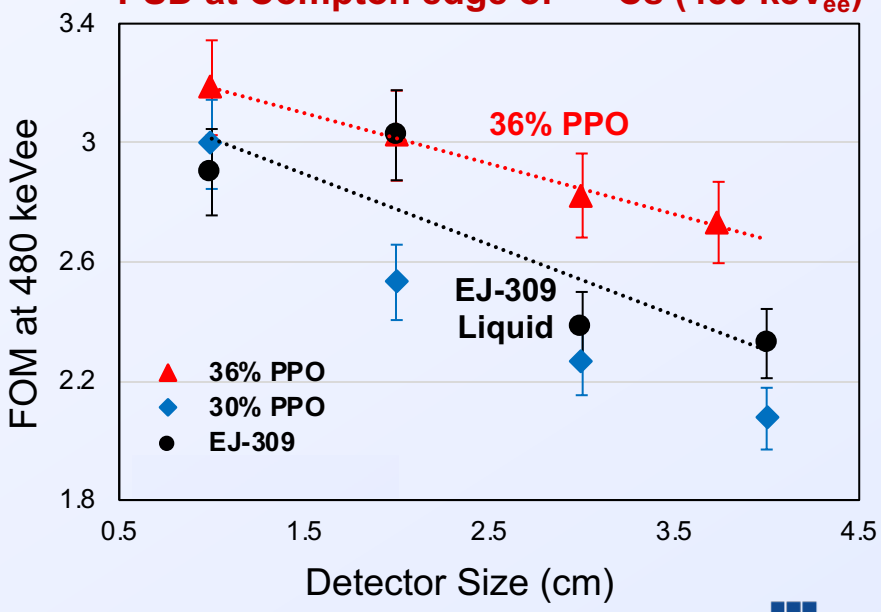
Commercial Production of New Formulation

Modified Plastics Show LO and PSD Competitive with Commercial Liquid Scintillators (EJ-309)



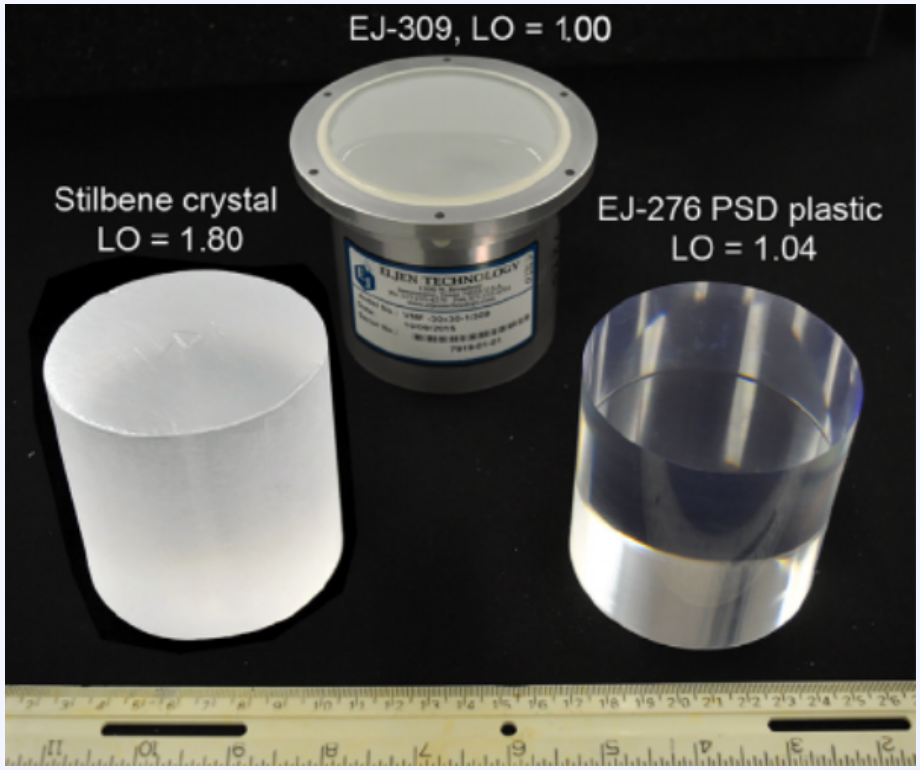


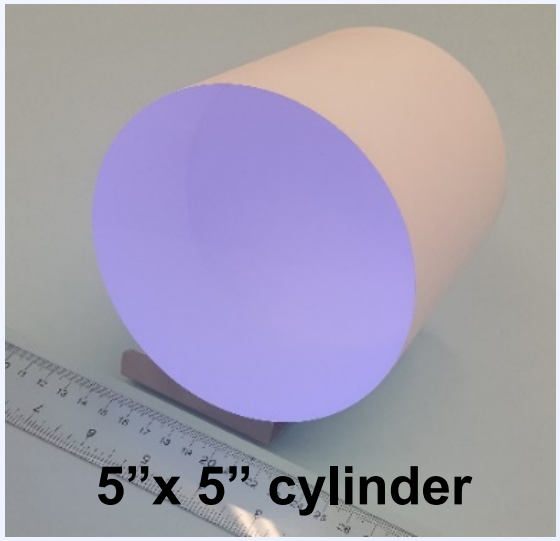


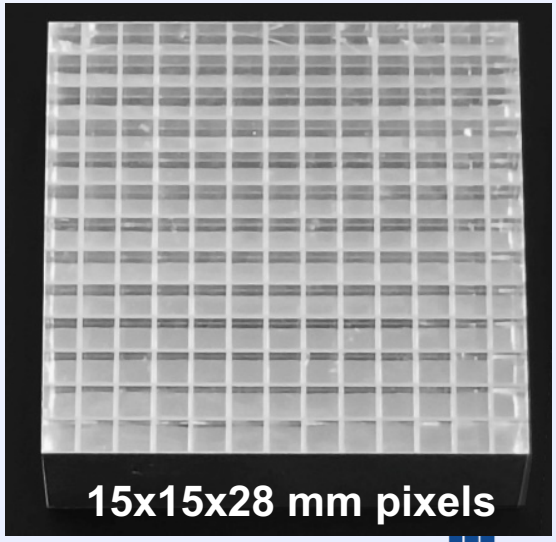


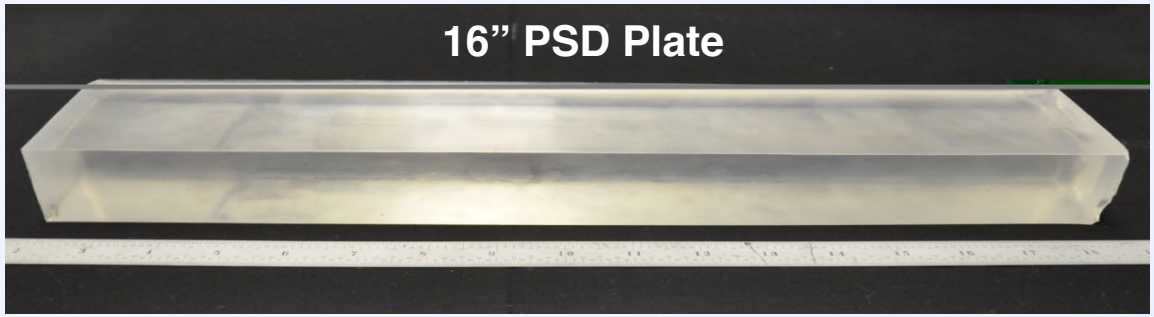
N. Zaitseva et. al., *Nucl. Instrum. Meth. A*, 889 (2018) 97.

Commercialization of New Formulation: EJ-276





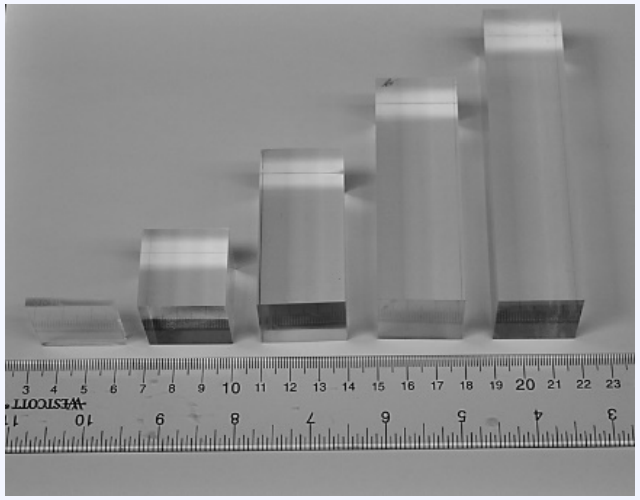


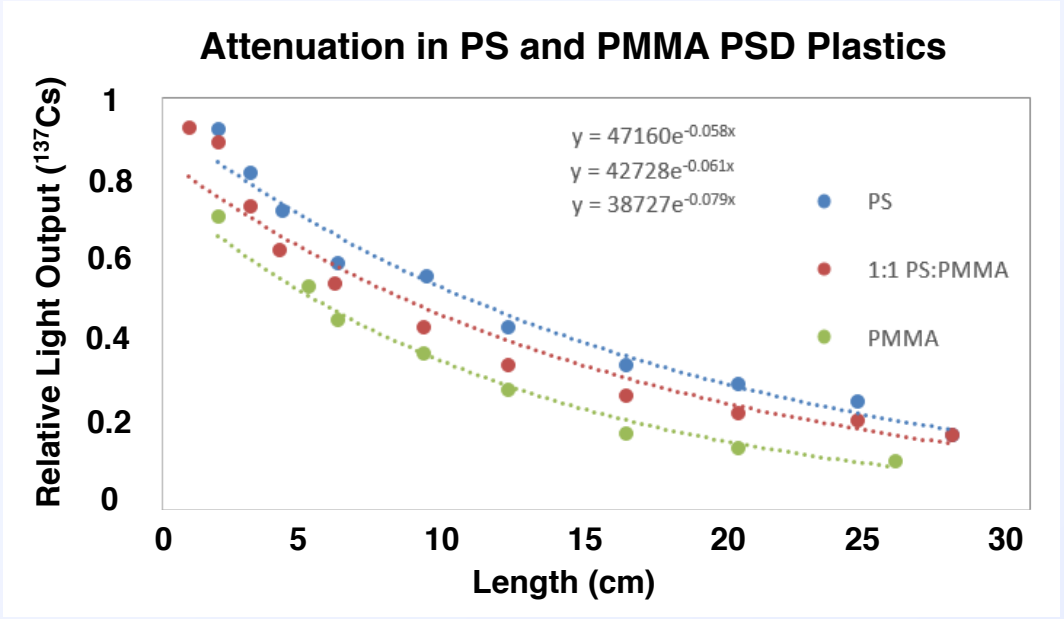


Commercial Production of EJ-276 Courtesy of Eljen Technology
(http://www.eljentechnology.com/)

Attenuation Length in PSD Plastics

- Sometimes it is necessary to add acrylic (PMMA) for specialty applications (E.g., Lithium)
  - It is known that **pure** PMMA has longer attenuation than PS



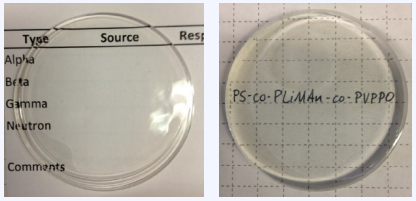


% PMMA	*μ (cm^{-1})*	***Attn. Length (cm)***	*ρ (g/cm^3)*	*μ/ρ (cm^2/g)*
0	0.058	**17.2**	1.090	18.8
50	0.061	**16.4**	1.125	18.4
100	0.079	**12.7**	1.176	14.9

Attenuation length not improved by using PMMA

Lithium-Loaded Polymer Scintillators Only a Recent Development: Oil doesn't dissolve salt

Lithium Copolymerized with Matrix



3.9% ^{6}Li 3.0% Li

Water-soluble, Low LO

A. Mabe et. al., *NIM. A*, 722 (2013) 29-33.
A. Mabe et. al., *Rad. Meas.*, 66 (2014) 5-11.

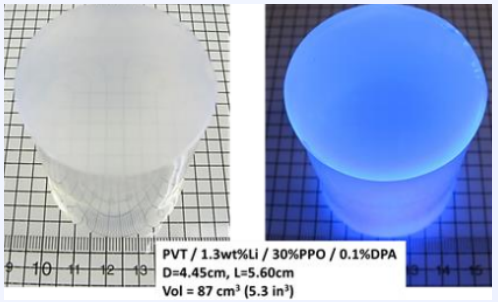
Lithium Methacrylate

0.08% ^{6}Li 0.67% ^{6}Li

Single Report, Limited Development

R. Breukers et. al. *NIM. A*, 701 (2013) 58-61

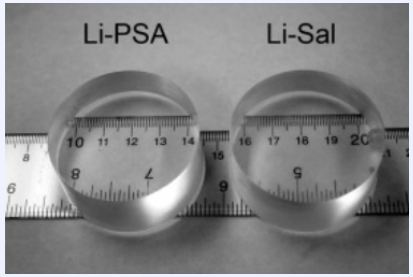
Recent LLNL Development: Lithium pivalate in PSD Plastic

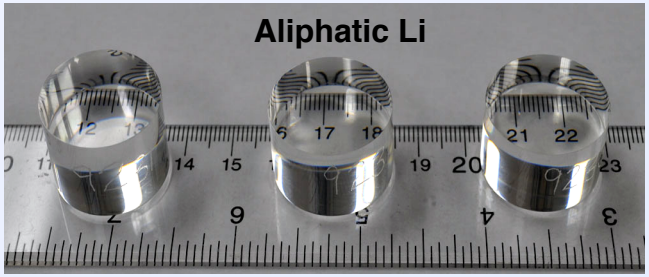


Initial Commercial Production

N. Cherepy et. al. *NIM. A*, 778 (2015) 126-132

Further LLNL Developments: Aromatic and Aliphatic Lithium in PSD Plastic





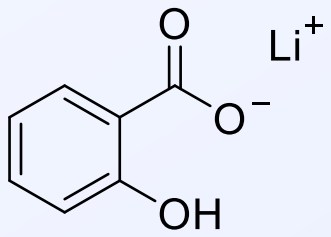
0.27% ^{6}Li 0.41% ^{6}Li > 0.2% ^{6}Li

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

Aromatic Lithium in PSD Plastics

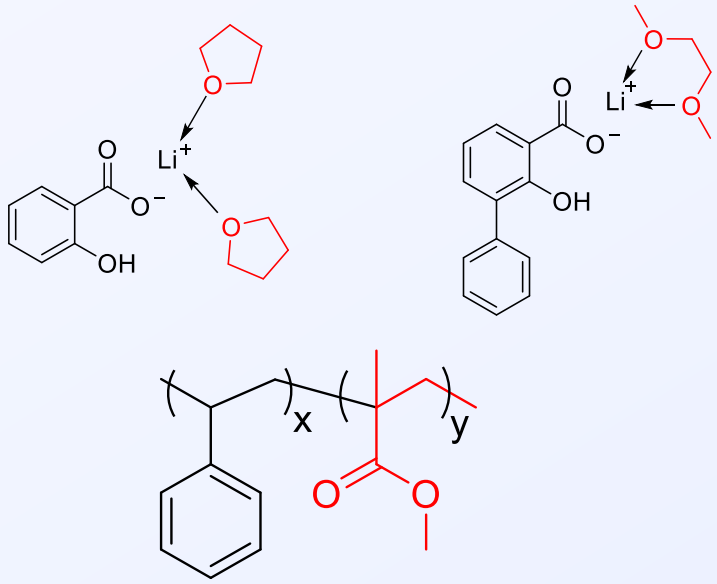
Organolithium compounds containing an aromatic ring

Salicylates are used in pharmaceuticals - nontoxic

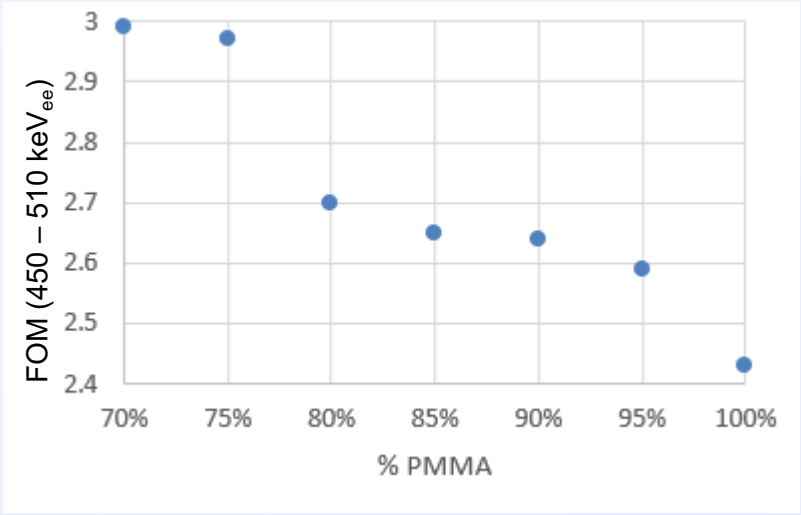


To disperse in plastic scintillator:

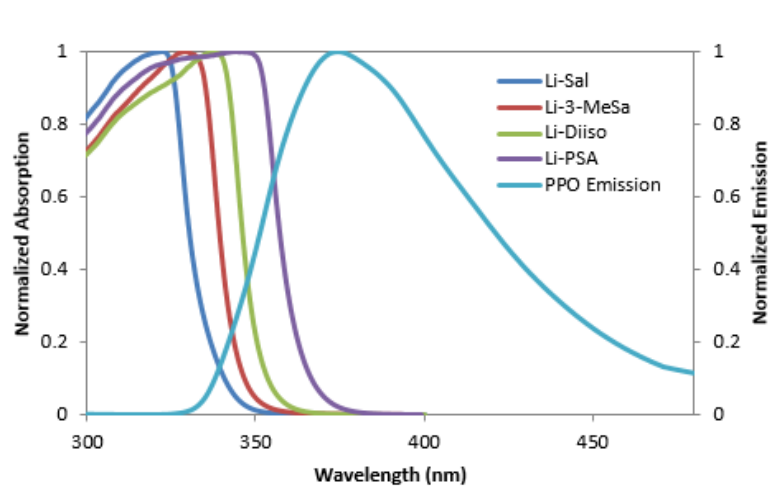
- Use **polar complexing solvent** containing electron donating atom (acetone, tetrahydrofuran, dimethoxyethane, etc.)
- Add **polar nonaromatic comonomer** (e.g., PMMA)



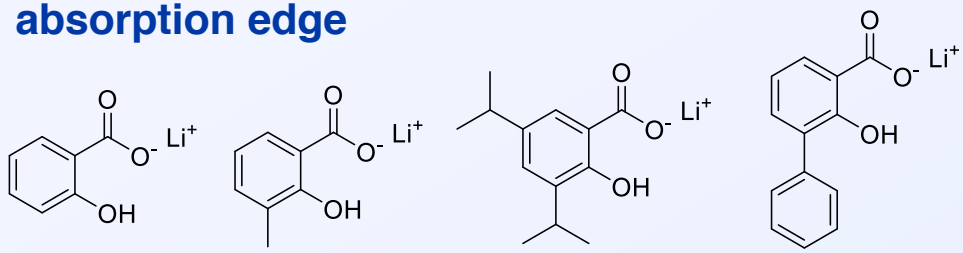
PMMA Matrix Reduces LO and PSD



Light Output of Plastics with Aromatic Lithium

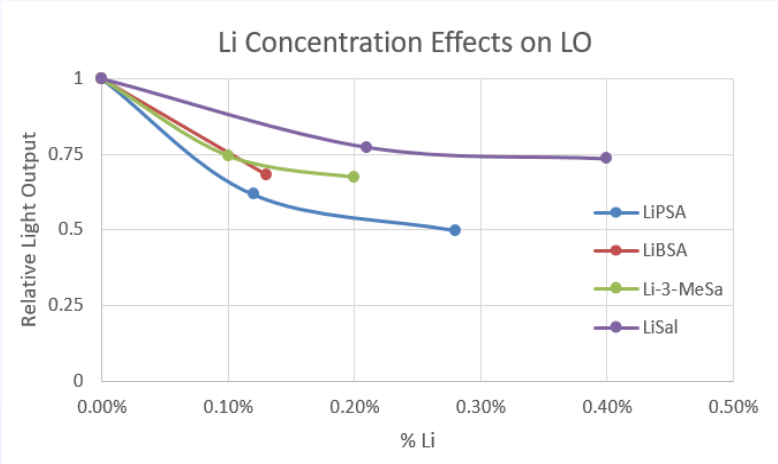


- Aromatic lithium salts absorb PPO emission
- Adding groups around ring red-shifts absorption edge



Higher Solubility in PSD Plastic

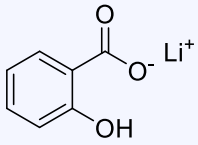
More Light Quenching

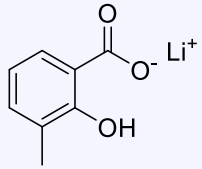


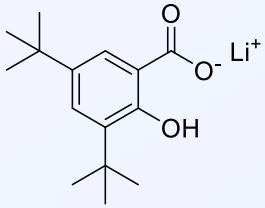
- Adding aromatic Li reduces LO by absorption
- Fraction of LO lost follows trend in absorption edge
- More overlap with PPO Emission results in greater reduction in light output

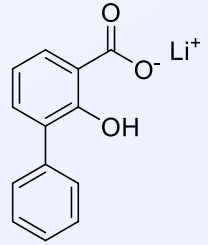
“Triple PSD” of Plastics with Aromatic Lithium

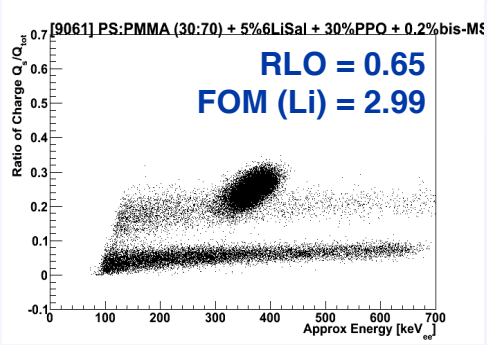
Substitution reduces amount of PMMA needed: Improves PSD



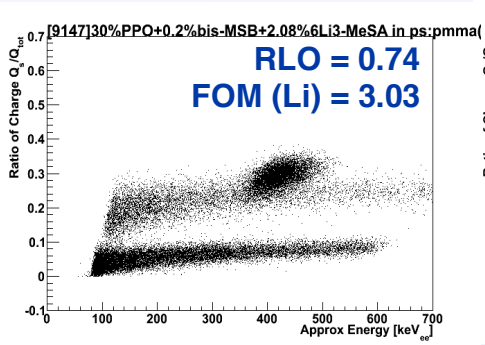




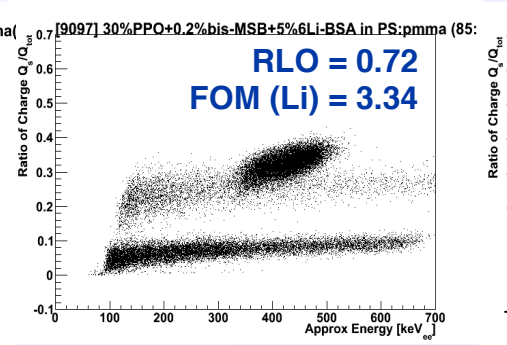




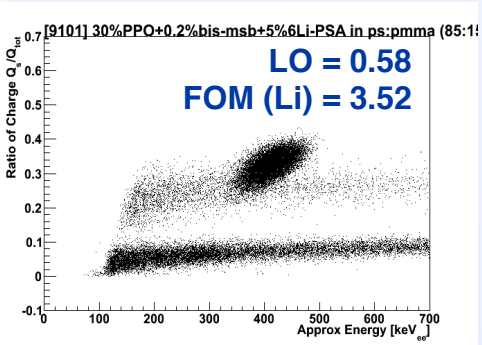
70% PMMA



50% PMMA



15% PMMA



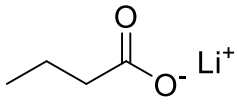
15% PMMA

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

- LO increases initially, due to increased fraction of aromatic matrix, decreases due to absorption
- PSD increases with increasing PS fraction

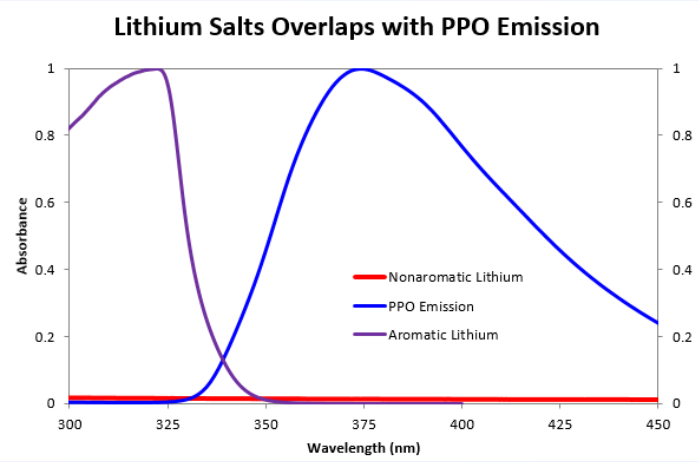
N. Zaitseva et. al., *Nucl. Instrum. Meth. A*, 729 (2013) 747-754
A. Mabe et. al., *Nucl. Instrum. Meth. A*, 806 (2016) 80-86

Incorporation of Nonaromatic (Aliphatic) Lithium



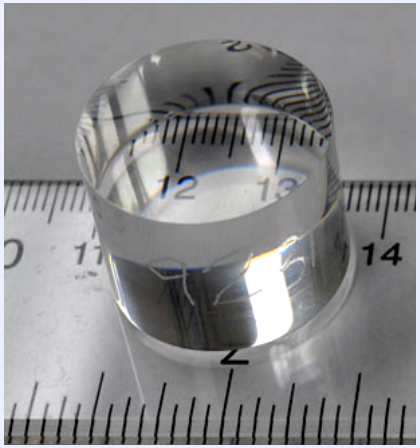
Aliphatic lithium salts have no chromophores

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



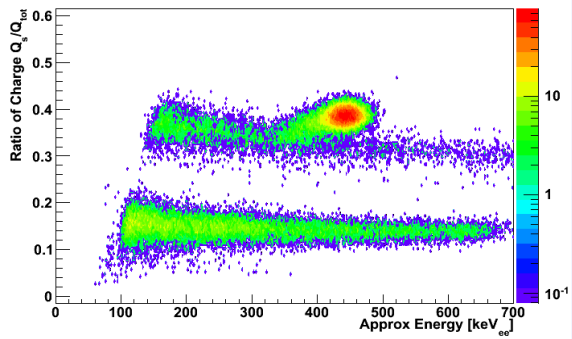
Strategy:

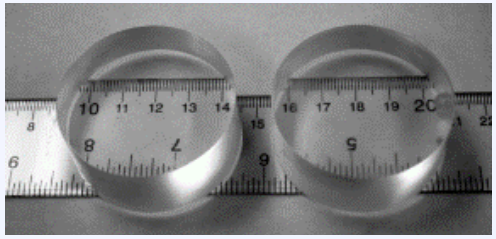
- Aliphatic lithium compounds have no solubility in organic solvents
- Complexation prevents lithium from phase-separating



Comparison of Aromatic and Aliphatic Lithium in PSD plastics

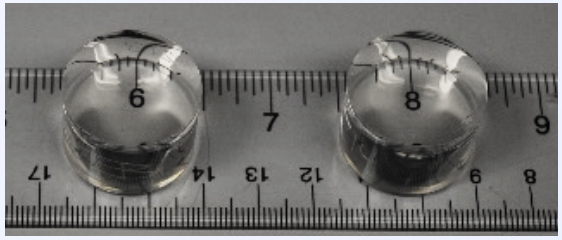
Aromatic Lithium
LO = 72%
FOM (n_{th}/y) = 3.34





Lower LO
Better PSD
Difficult fabrication
Scalable

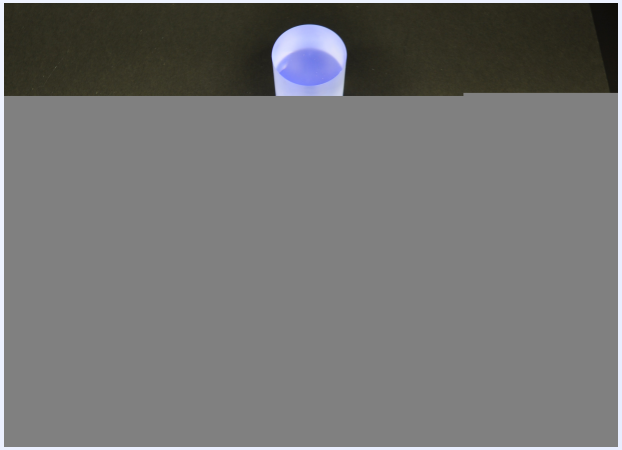
Aliphatic Lithium
LO = 79%
FOM (n_{th}/y) = 2.64



Higher LO
Lower PSD
Much easier fabrication
Scalable

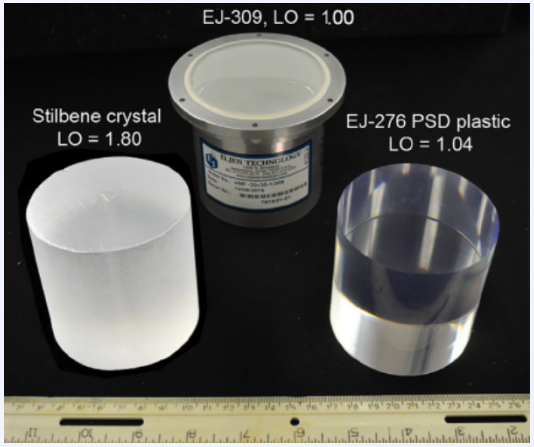
Aliphatic lithium formulation scalable to large size

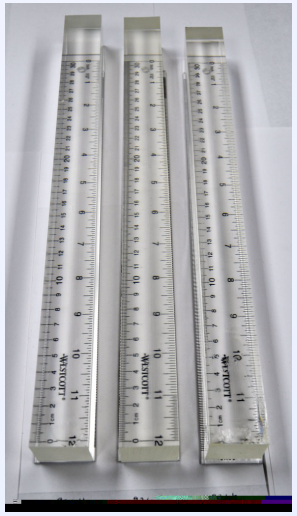
Attenuation studies are currently underway



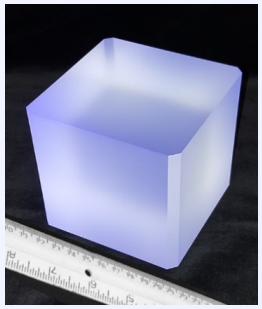
Conclusions and Future Work

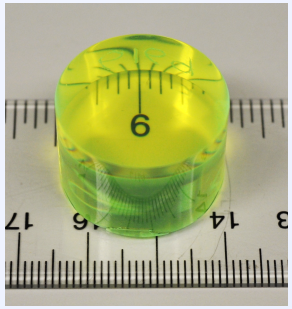
- PSD plastics performance and stability improved
  - Can increase loads up to (>) 36% PPO; stable.
  - New secondary dye MDAC
  - Can be produced on kg scale
  - New commercial product: EJ-276

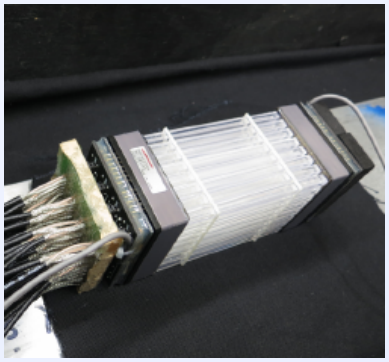


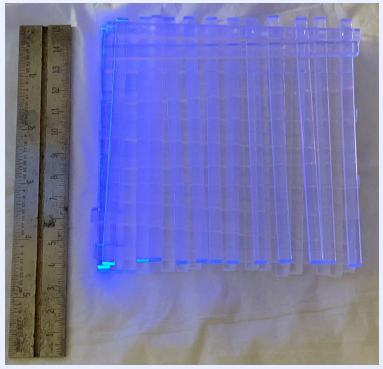


- Lithium Plastics
  - Can routinely produce plastics up to ~0.4% ^{6}Li
  - At 0.1% ^{6}Li, ~21% reduction in LO (Aliphatic)
    ~28% reduction in LO (Aromatic)
  - Mitigating problems associated with quenching
  - Segmented Li plastic
  - Evaluation of green-emitting PSD plastics









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