

PSD Scintillator Development at LLNL

Reactor Antineutrino Spectrum and Anomaly Measurements:
U.S. Planning Meeting

December 11, 2012

Andrew Glenn



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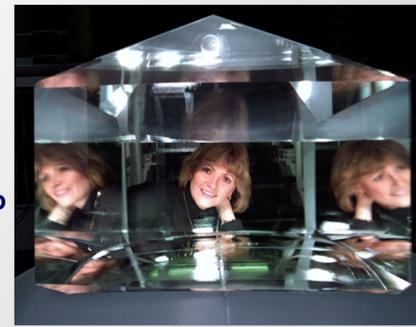
LLNL-PRES-607694

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Team

Natalia Zaitseva (PI): Ph.D. Physics/Mathematics, Moscow State University, Russia. Development of the Rapid Growth Techniques for production of super-large KDP-DKDP crystals; Crystal growth; Crystal characterization; Work planning and management.



Leslie Carman: Chemist, UC Santa Barbara. Played active part in NIF KDP development. Crystal growth; Chemical analysis; Material purification; Solubility measurements; Crystallizer design modification.

Andrew Glenn: Ph.D. in High Energy Nuclear Physics, University of Tennessee. Nuclear scientist, Neutron detection characterization, PSD data analysis, Design of electronic instrumentation.

Iwona Pawełczak: Ph.D. in (Nuclear) Chemistry, University of Rochester. Neutron detection characterization, PSD data analysis, Spectroscopy, Crystal studies for incorporation in detection systems.

Paul Martinez: Ph.D. in Chemistry, UC San Diego. Organic/inorganic synthetic techniques, Material purification, Chemical analysis, and Optical spectroscopy.

Michelle Faust: Lawrence Scholar Program graduate student, University of California – Davis. Crystal characterization, Solubility measurements, Spectroscopic analyses, Crystallography.

Keith Lewis: Mechanical technician – Crystallizer modifications and manufacturing, Crystal cutting procedures.

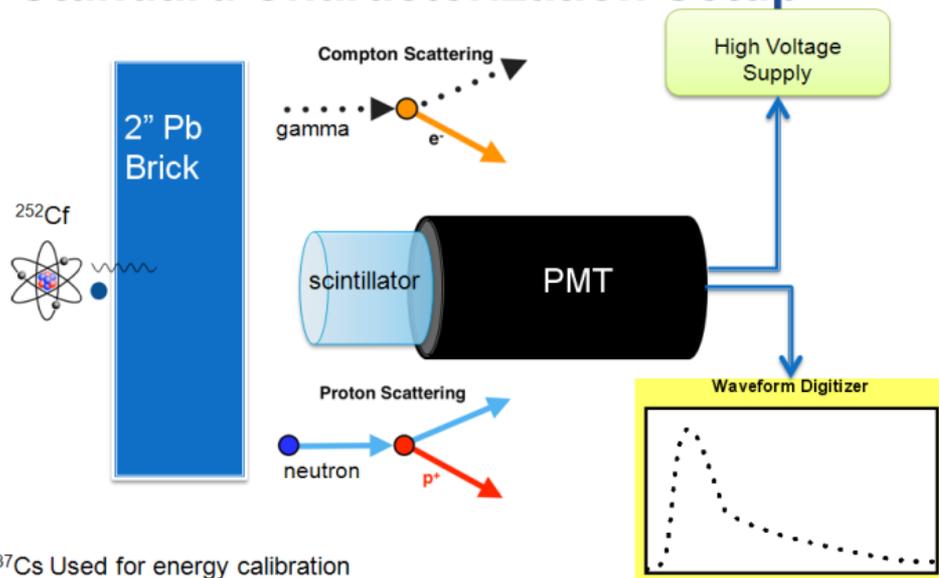
Stephen Payne (PM): Ph.D. Physical Chemistry, Princeton University. Work coordination; Modeling; Data analysis and discussions.

Goals and projects

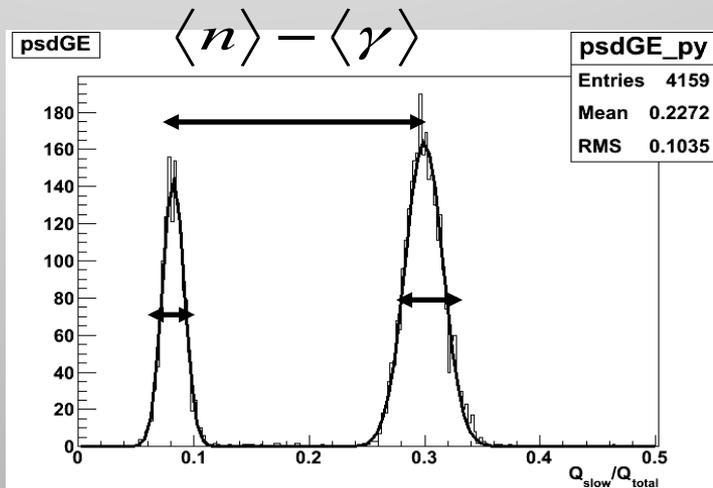
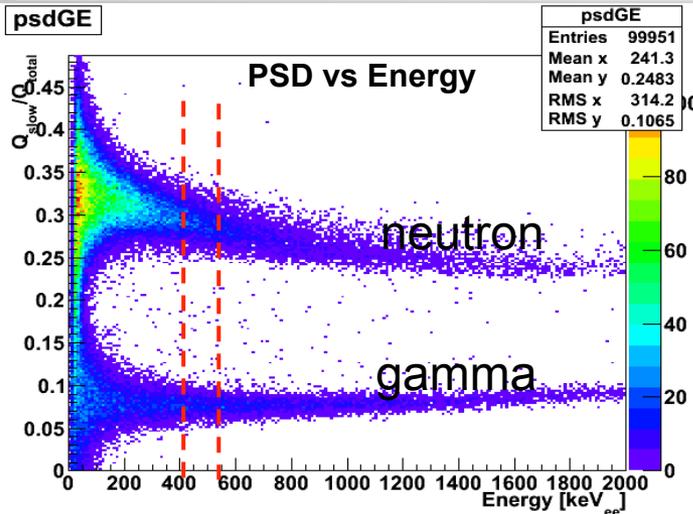
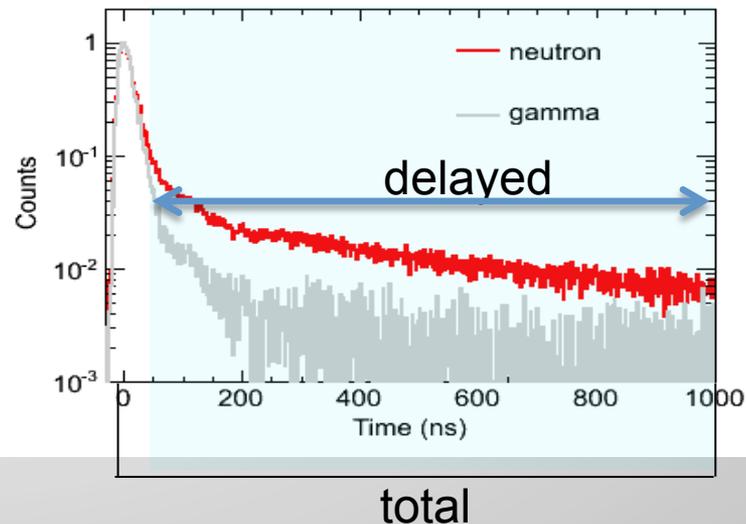
- **Develop advanced organic scintillators for radiation detection**
 - Emphasis on solid state neutron detectors
 - Enhance homeland security, safeguards and related capabilities
- Started with LDRD for survey and down selection of crystal scintillators with neutron/gamma pulse shape discrimination
- Ongoing DNDO funded stilbene growth project
- Ongoing NA22 funded advanced crystals/plastics
- Starting boron loaded plastics development for DTRA

Pulse Shape Discrimination

Standard Characterization Setup



Scintillator Time Response



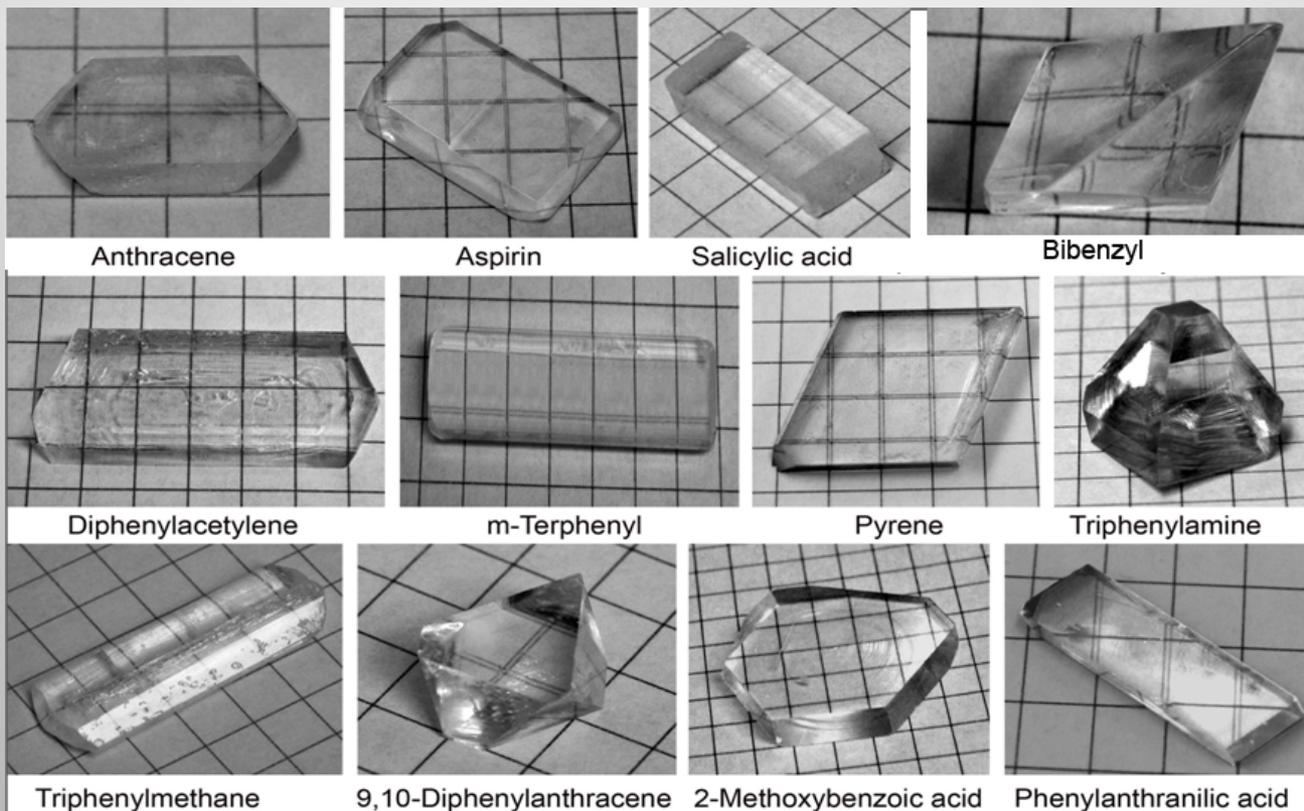
$$PSD = \frac{\int_{delayed} Q dt}{\int_{total} Q dt}$$

$$FoM = \frac{\langle n \rangle - \langle \gamma \rangle}{\Gamma_{FWHMn} + \Gamma_{FWHM\gamma}}$$

We conducted an extensive survey of aromatic compounds and found many new materials with efficient PSD

Solution technique applied to grow organic crystals traditionally grown from melt

Broad variety of organic compounds explored for neutron detection properties and PSD

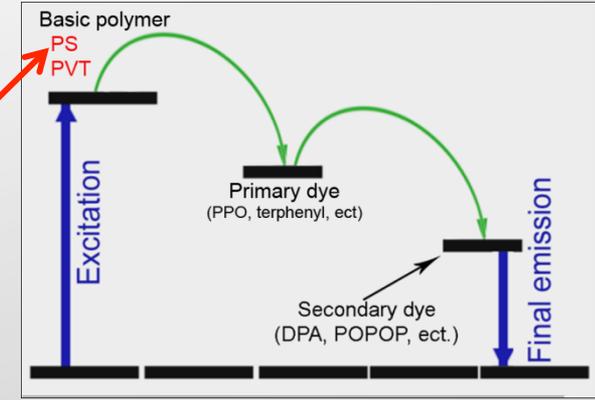
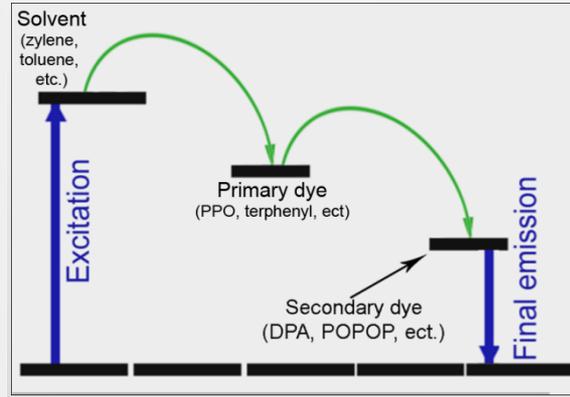
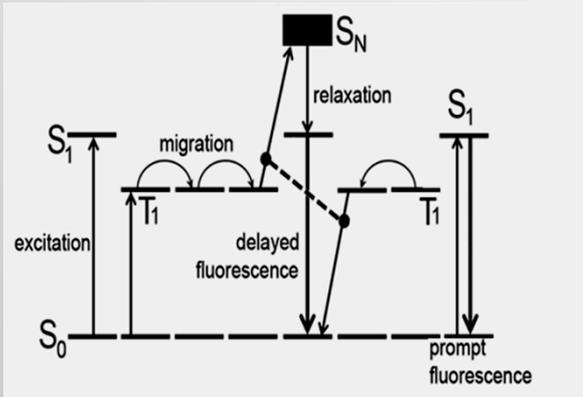


Background square size is 6.5 mm

More than 150 organic crystals tested in survey of PSD phenomenon

Scintillation mixtures exhibit complex PSD behavior

Liquid and plastic scintillators always prepared as mixtures of two or more organic compounds

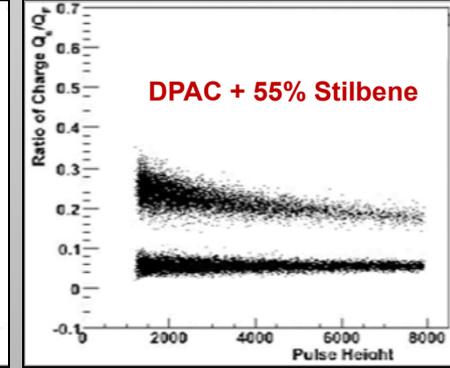
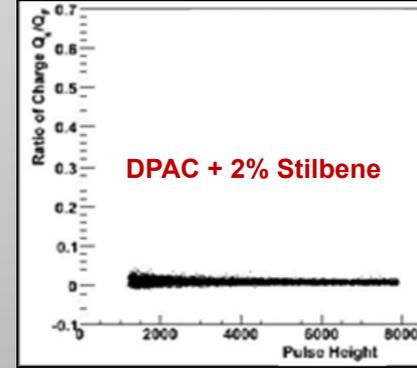
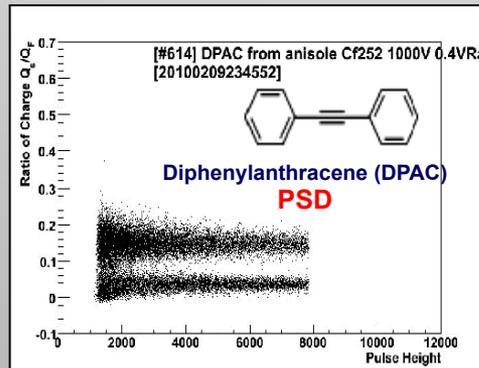
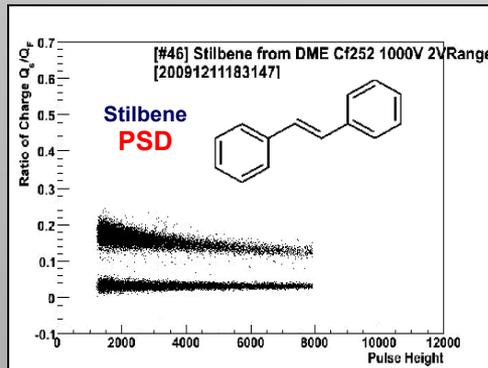


Single-component system

Liquid scintillator mixture

Plastic scintillator mixture

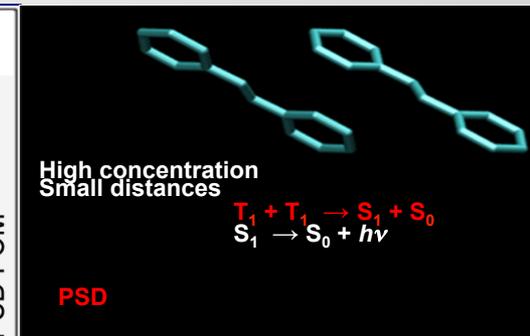
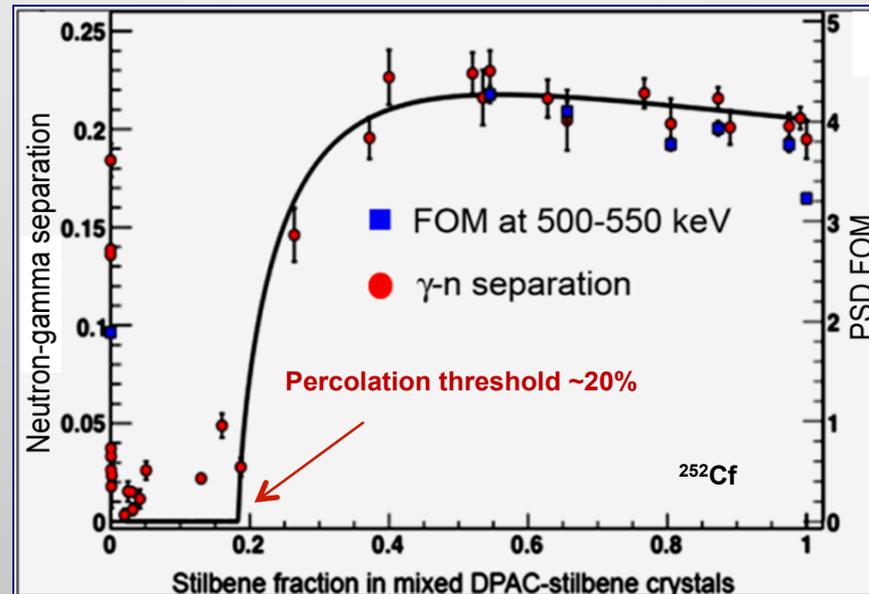
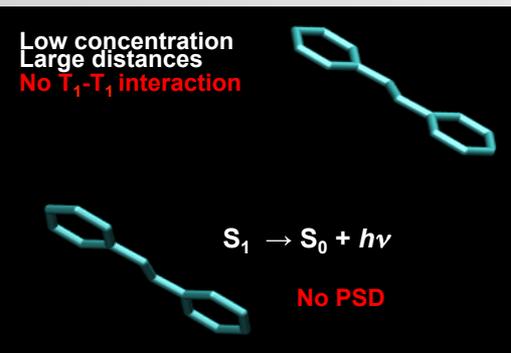
Studies of mixed crystals resulted in new understanding of PSD mechanisms



PVT = Polyvinyltoluene; PS = Polystyrene; PPO = 2,5-Diphenyloxazole

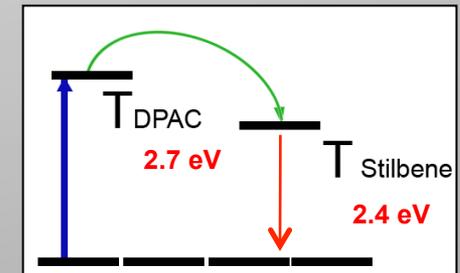
PSD in mixed crystals found to depend on the concentration of constituent dyes

Delay light and PSD formation require T_1 - T_1 interaction of pairs of **closely located** molecules



Nuclear Science, IEEE Transactions on,
vol.58, no.6, pp.3411-3420, Dec. 2011

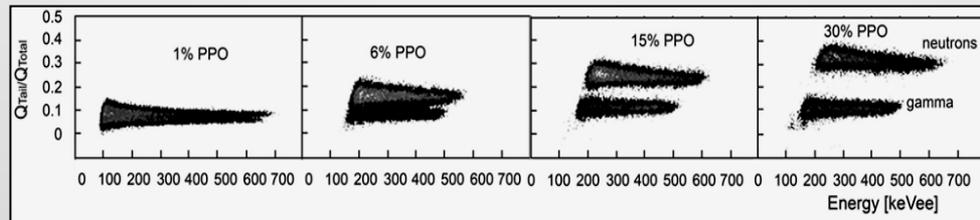
Excitation energy
transferred to lower-band-
gap dye (stilbene)



Studies conducted with crystals led to development of first plastic scintillators with efficient PSD

- Plastic scintillators traditionally prepared with low dye concentrations by analogy with liquids

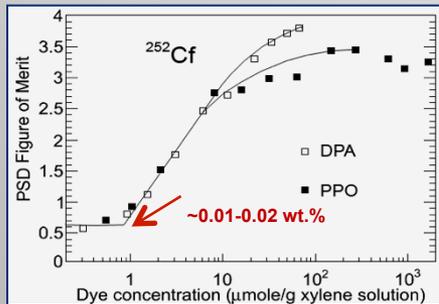
Plastics: PPO in PVT



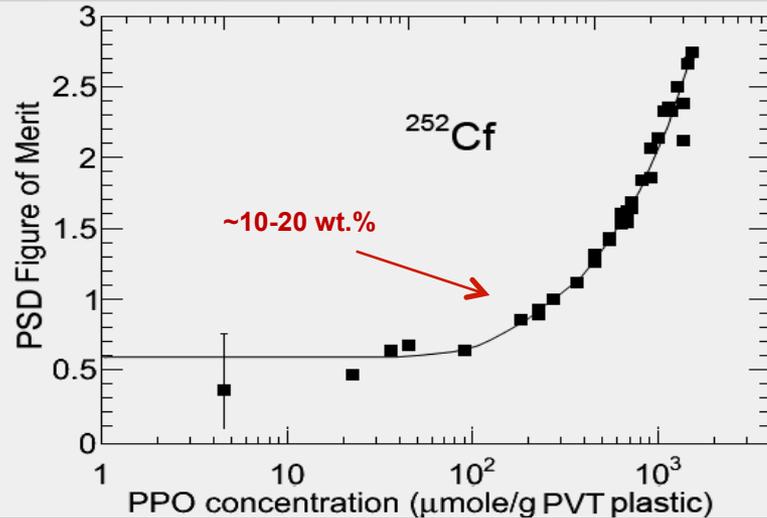
Low dye concentration is sufficient for PSD production in liquid scintillators

PSD in crystals requires 2-3 orders of magnitude higher dye concentration

Liquids

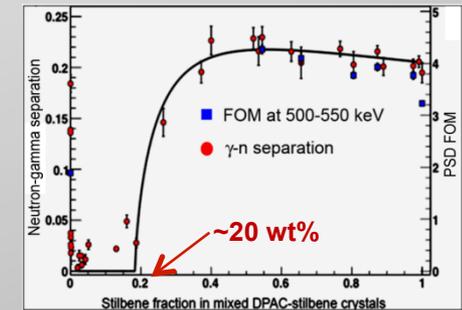


High diffusion mobility of molecular interaction



Efficient PSD at 10-20% (solid state)

Crystals



Fixed positions of molecules in crystal lattice

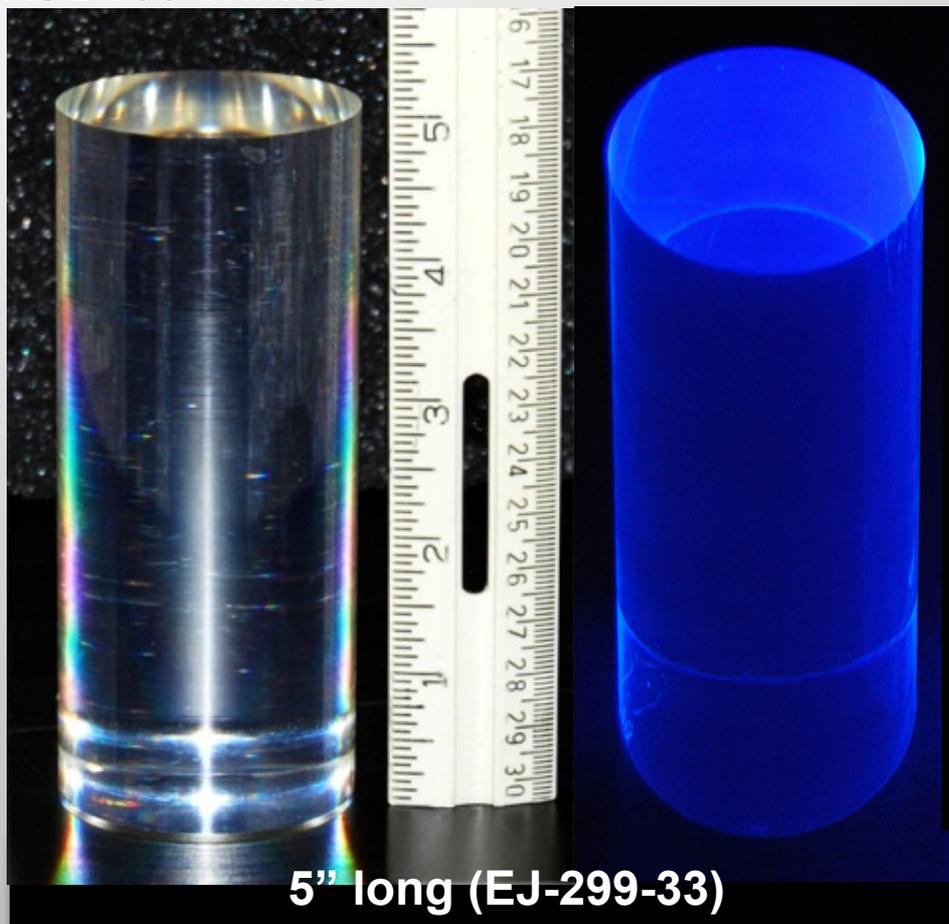
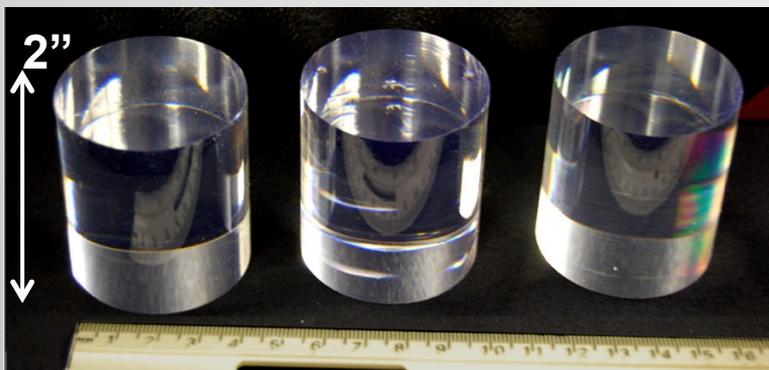
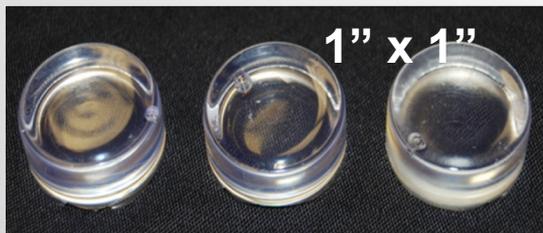
PSD in plastics requires concentration of a dye similar to that in crystals!

*) PVT = Polyvinyltoluene; PS = Polystyrene; PPO = 2,5-Diphenyloxazole

New PSD plastics: Easy to produce with traditional inexpensive ingredients, no major changes in existing technology

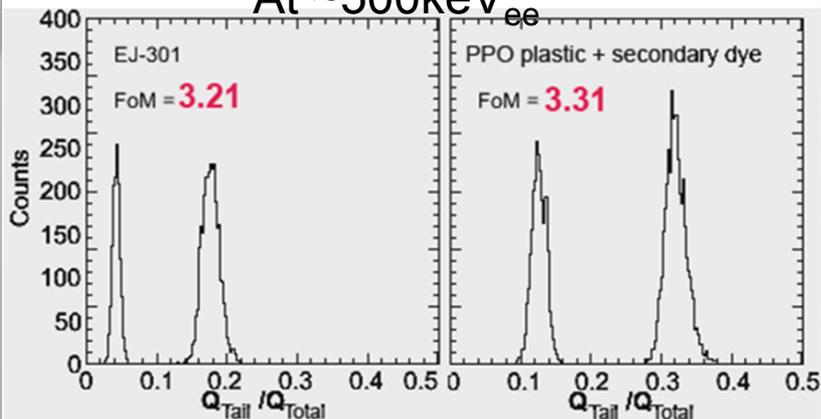
Quick transfer to industry!

Eljen technologies is producing an implementation
A 2012 R&D100 Winner

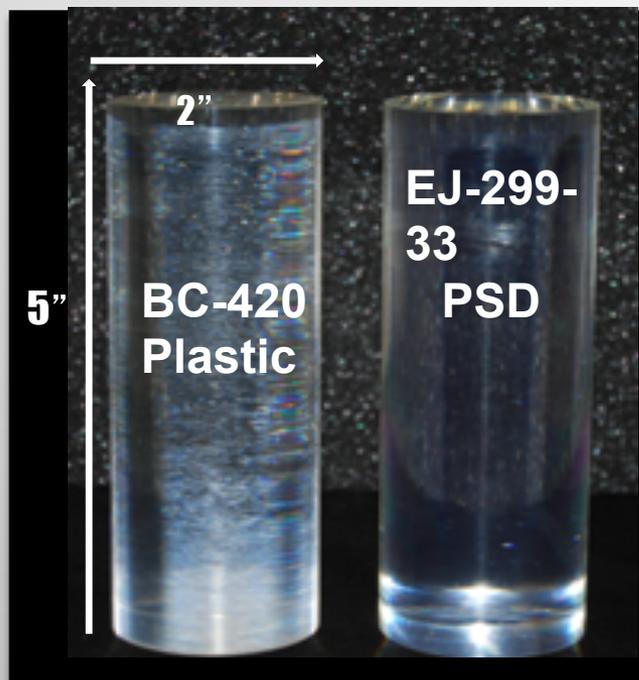


135mm x 175mm long cylinders
have been produced

At $\sim 500\text{keV}_{ee}$

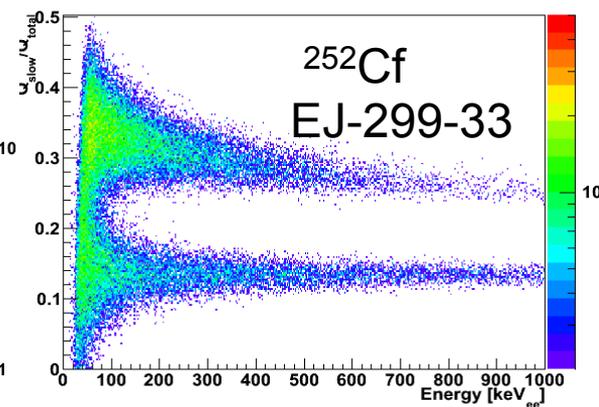
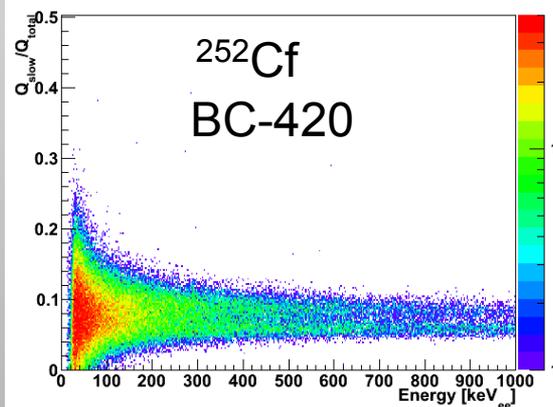
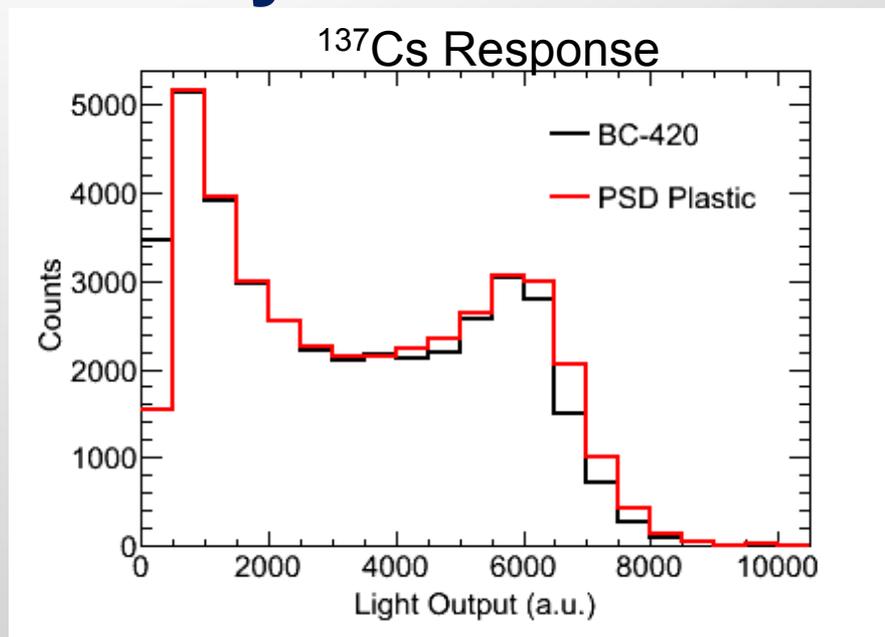


Light yield PSD plastic comparable to light yield in plastics with low-dye-concentration



Higher excitation density of secondary dye molecules compensates light absorption of high concentration primary dye

No noticeable change in response to collimated ^{137}Cs along length



Challenges and solutions of High Dye Loading In PSD Plastic Scintillators

Challenges

- Precipitation of dyes
- Softening\Clarity
- Enhanced Absorption (Yellowing)
- Inhomogeneity at Scaling



Solutions

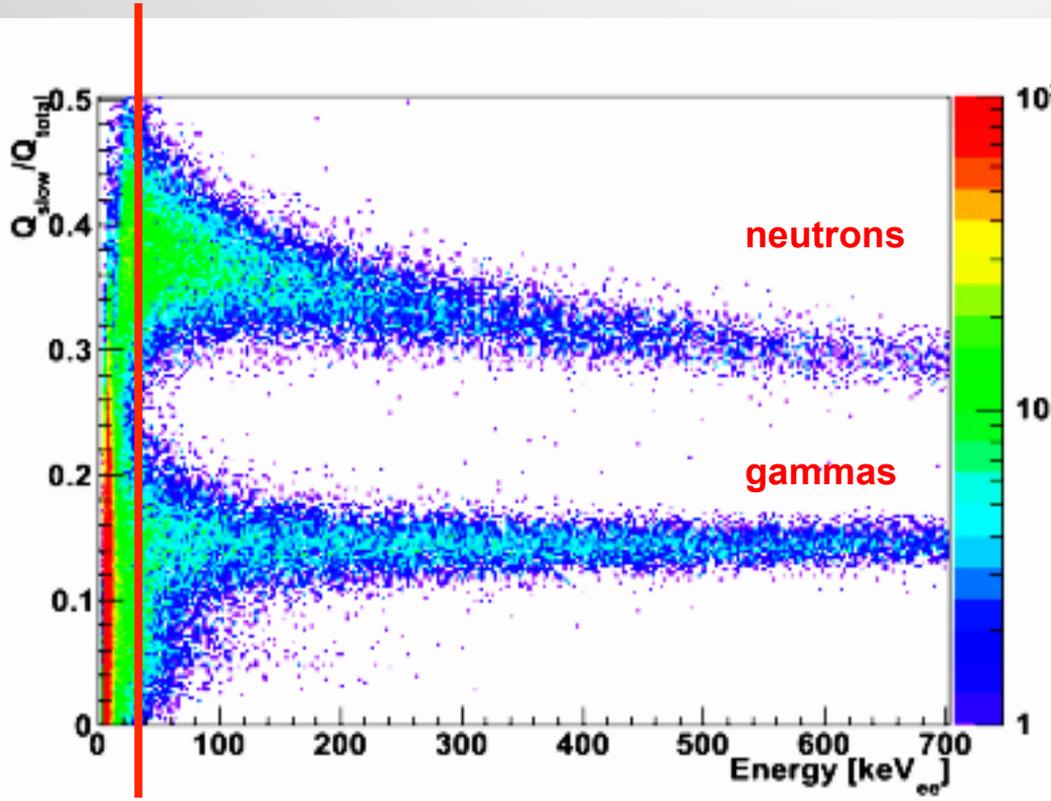
The addition of cross-linkers greatly limits any dye precipitation

The use of acrylate co-polymerization hardens and increases clarity

Slower curing time along with a temperature gradient aid both yellowing and scaling issues

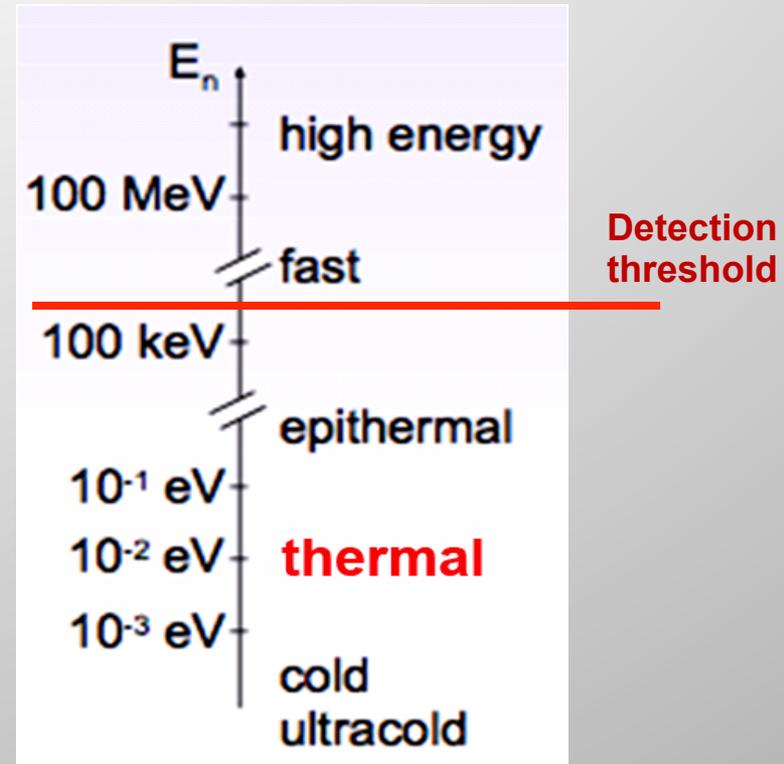
These PSD plastics can be used only for **fast** neutron detection

Detection threshold



~40-50 KeV gamma equivalent
~ 500 KeV neutron energy

Neutron energy



PSD in existing plastics
unsuitable for thermal
neutron detection

Neutron detection in wide dynamic range: Incorporation of nuclei with large capture cross sections

Multi-MeV neutron
detected by proton recoil

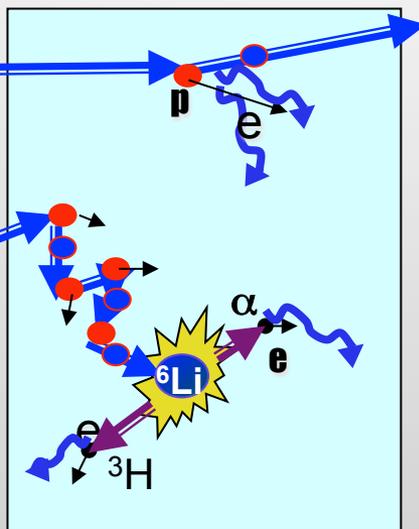
$n1$

Neutron below the threshold
for detection by proton recoil
measured by capture
reaction products

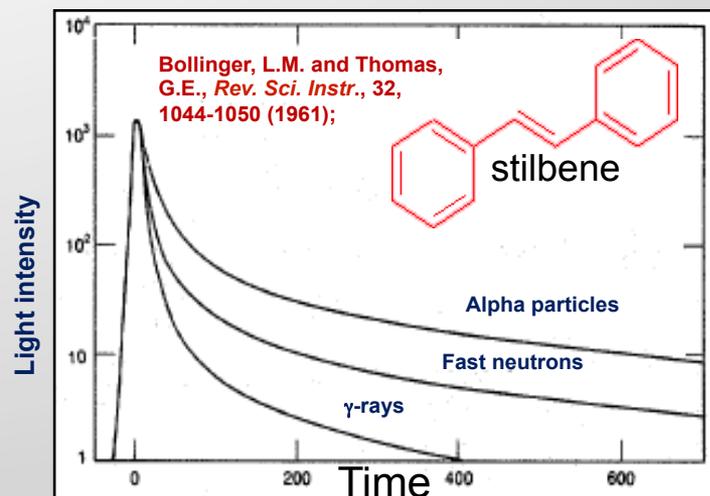
$n2$

Efficiency

PSD Scintillator Loaded with ^6Li

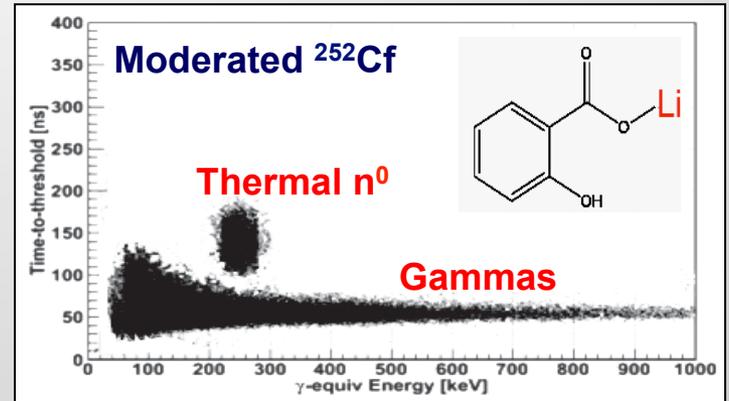
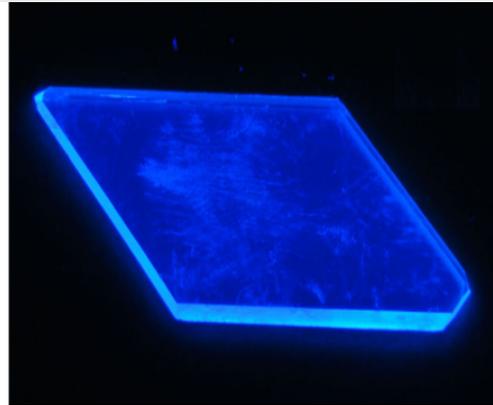
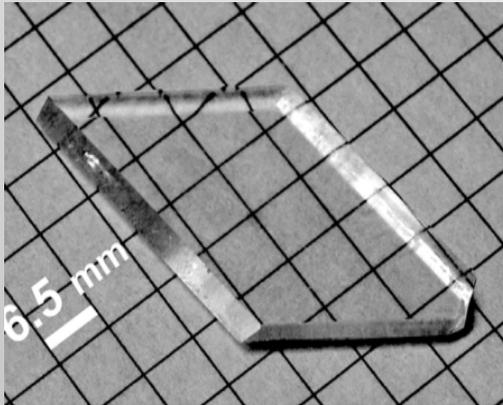


PSD – γ -ray background rejection

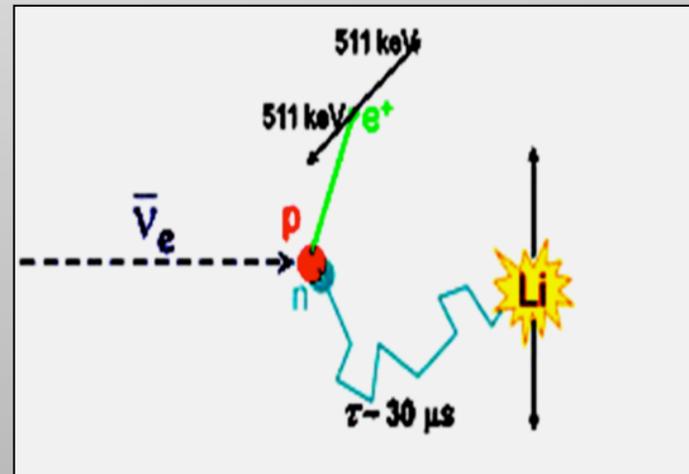
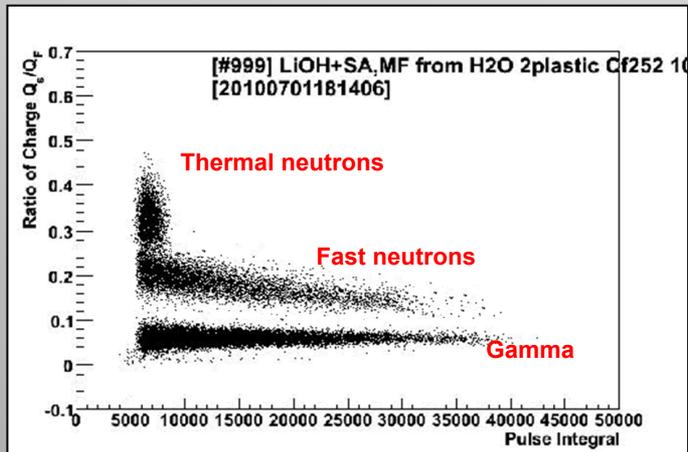


| Thermal Neutron Capture Reaction | | Cross Section (barn) | Q-value (MeV) | Natural Abundance (%) |
|--|-----|----------------------|---------------|-----------------------|
| $^{10}\text{B}(n,^4\text{He})^7\text{Li}$ | 6% | 3840 | 2.792 | 19.8 |
| $^{10}\text{B}(n,^4\text{He} \gamma(0.48\text{MeV}))^7\text{Li}$ | 94% | | 2.310 | |
| $^6\text{Li}(n,^3\text{H})^4\text{He}$ | | 940 | 4.78 | 7.40 |

Some crystals grown as lithium salts for thermal neutron detection

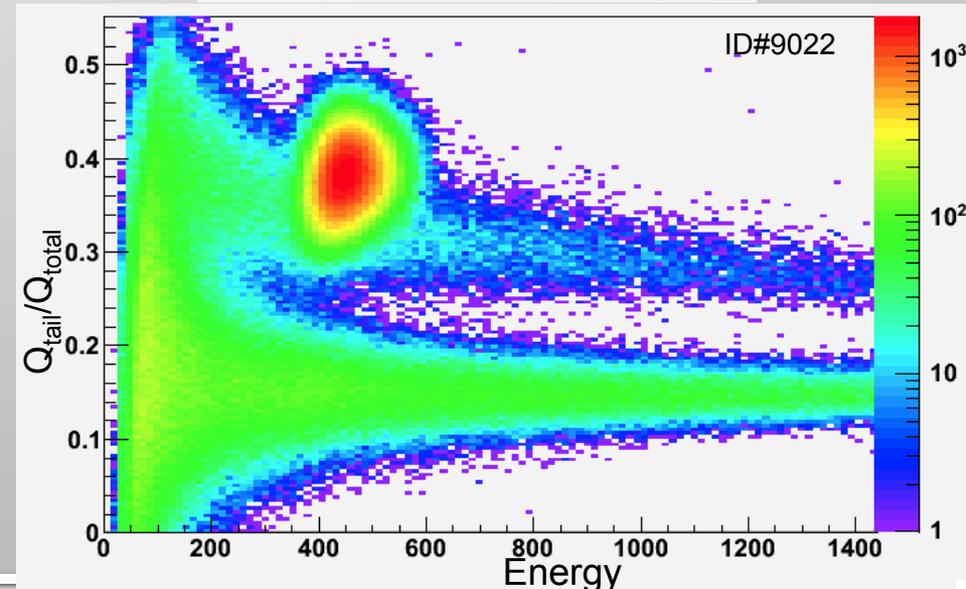
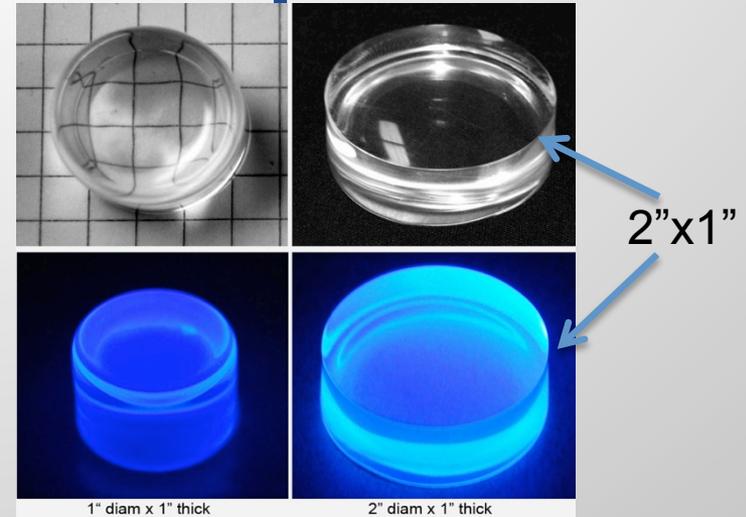


Simultaneous detection of fast and thermal neutrons serves a signature for anti-neutrinos

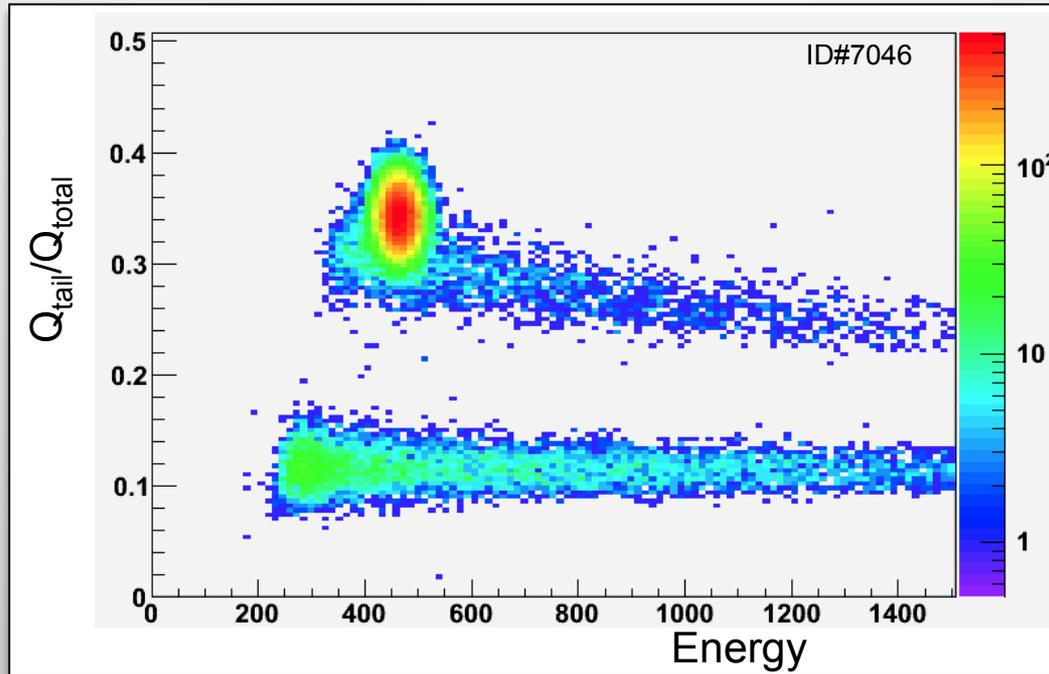


Organometallic lithium compounds can be incorporated in PSD plastic

- We have synthesized several organometallics which have allowed us to achieve homogeneous Li loadings of $\sim 0.15 - 0.3$ weight %
- We have ideas to increase loading
- No stability/shelf life issues have been observed
- Initial studies conducted with natural Li
- Now moving to enriched ${}^6\text{Li}$
 - **\$\$, Cost could be reduced by large quantity purchase**
- We plan to test performance with PMTs selected by N. Bowden et. al. in the near future



Organometallic Li compounds can be incorporated in liquid scintillators



- Similar loading as plastics
- So far only Xylene based
 - Not suitable for deployment due to handling/safety
- Less information on stability
- We will try safer alternatives SOON
 - Linear Alkyl Benzene (LAB)
 - EJ-309 liquid scintillator

Summary

- We are experienced in organic scintillator development
- We have developed the first plastic with efficient neutron/gamma PSD
 - And worked with industry to bring it to market
- We have developed the first ^6Li loaded plastic scintillators with PSD for thermal neutron, fast neutron and gamma detection
- The same method of Li incorporation appears applicable for liquid scintillators
- We look forward to working with the neutrino detection community