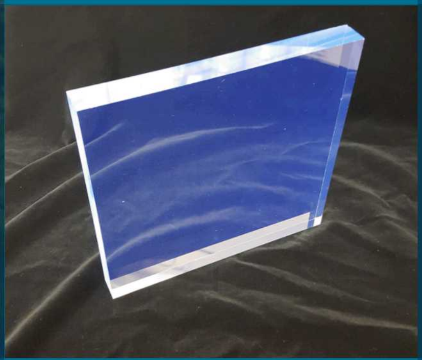
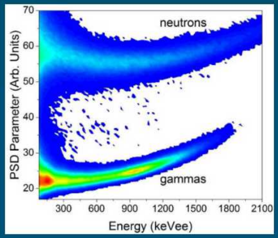
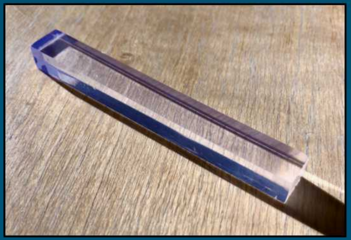
SAND2019-2877PE

Organic Glass Scintillators (OGS) for Improved Fast Neutron Detection







As part of the "Advanced Materials for Detectors" multi-lab collaboration:

LBNL, **SNL**, ANL, PNNL, UC-Berkeley, Wake Forest Univ., Northwestern, Univ. of Minnesota

With contributions from:
University of Michigan

Joseph Carlson, **Patrick Feng**,
Huu Tran, Nicholas Myllenbeck, Bethany Goldblum, Thibault Laplace

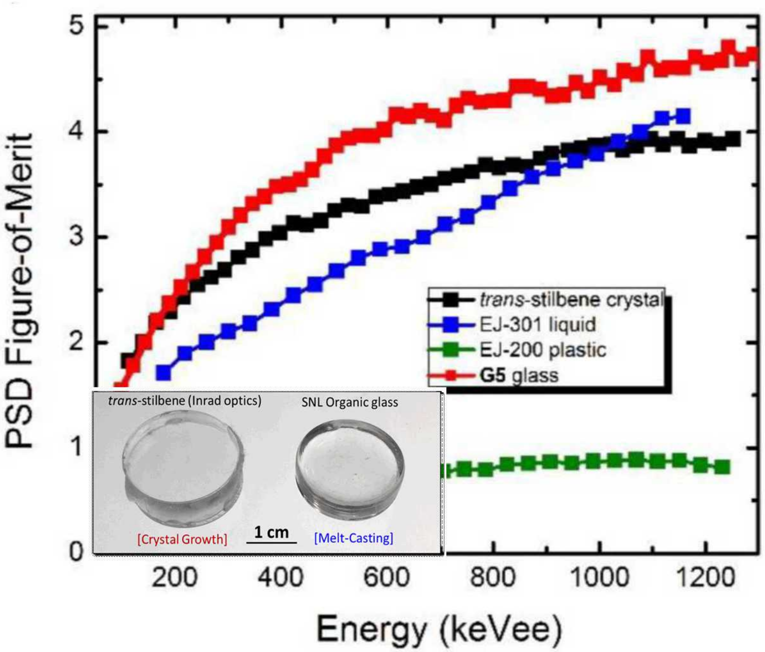
Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Project Overview: Background and Prior Work

What is an organic glass?
Amorphous organic solid that resides below it's glass transition temperature.

How are small-molecule organic glasses similar to plastic scintillators?
Both are highly adaptable platforms with isotropic optical, mechanical, and transport properties

How do small-molecule organic glasses differ from plastic scintillators?
Polymers: Distribution of chain lengths/properties (PSD), ↓ Q.Y.'s.
OGS: Identical molecules arranged in random orientations (PSD), ↑ Q.Y.'s. (high light yields)



Highlights of Prior Work:

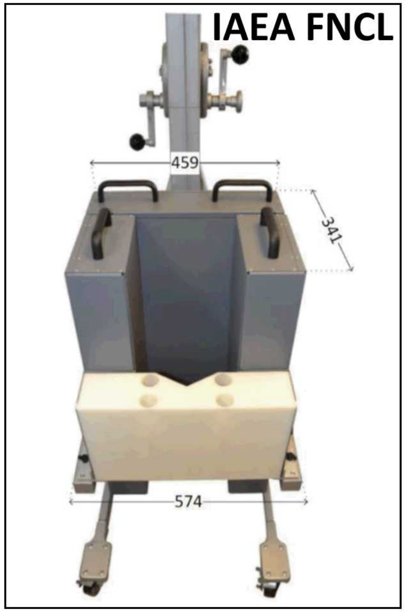
- Pathway to low-cost / large-volume production
- Indefinitely stable, even under accelerated aging
- T_g comparable to plastic scintillators
- Transparent monoliths can be directly melt-cast
- 0.75" diameter specimens provided light yields and PSD that exceeded *trans*-stilbene

Are these attributes sufficiently interesting?

Project Overview: Why pursue OGS materials?

There is considerable user 'pull' and engagement with industry:

- Evaluation as potential replacement for fast neutron collar (NA-241/IAEA)
- FRIB 'NEXT' neutron detector (DOE NP SBIR with XIA, Inc.)
- Memory-resistant radioxenon detection system (XIA collaboration)
- Structural scintillator materials for high strength and/or impact-resistant applications (DTRA, other agencies)
- n-TOF applications (i.e. Single-Volume Scatter Camera, EDUG/AWE)
- Fast scintillators for active interrogation (CWMD)
- High-efficiency and spatial resolution scintillators for fast neutron radiography
- Radiation portal monitors (CWMD)
- Ongoing collaboration with industrial partner for trial-scale production evaluation



Types of Samples Produced to Date:

- 2" and 3" diameter cylinders, 4-8" long rods, 2" x 2" windows, 8" x 8" slabs

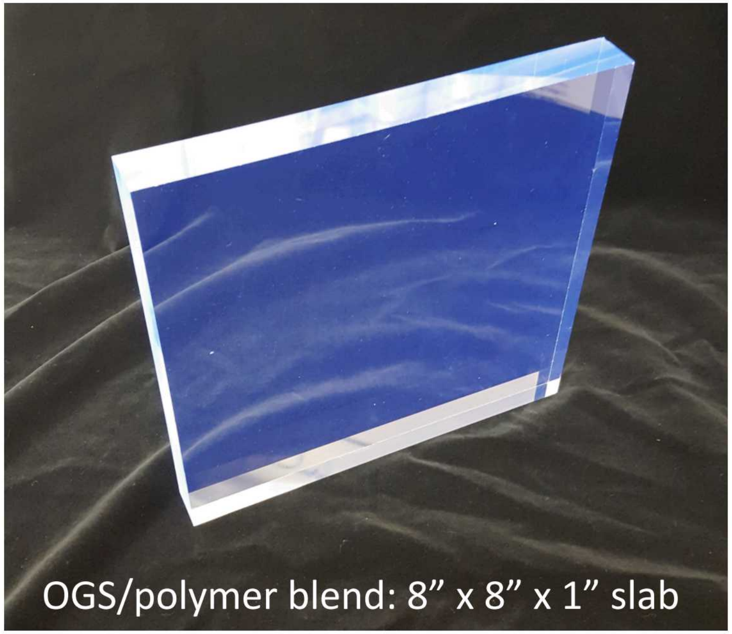
Current Phase Results: Key OGS Highlights

<u>Demonstrated that OGS provides a flexible scintillator matrix (‘platform’)</u>

- Fabricated diverse sizes/shapes using rapid casting process
- Excellent neutron proportionality, efficiency, and light yields (UCB, UM collaboration)
- High performance is maintained in larger sizes (currently 2-3”)
- OGS/polymer blends are especially promising
  - High-strength blends
  - Very fast timing + very high light yield scintillators
- Compatibility with a wide range of additives
  - Wavelength shifters
  - Polymers
  - Thermal n capture isotopes
  - Organometallic compounds
  - Environmental aging-resistant compounds
- Range of OGS-based materials span TRLs 2-6

Scale-up Efforts

OGS: 2" x 2" cylinder



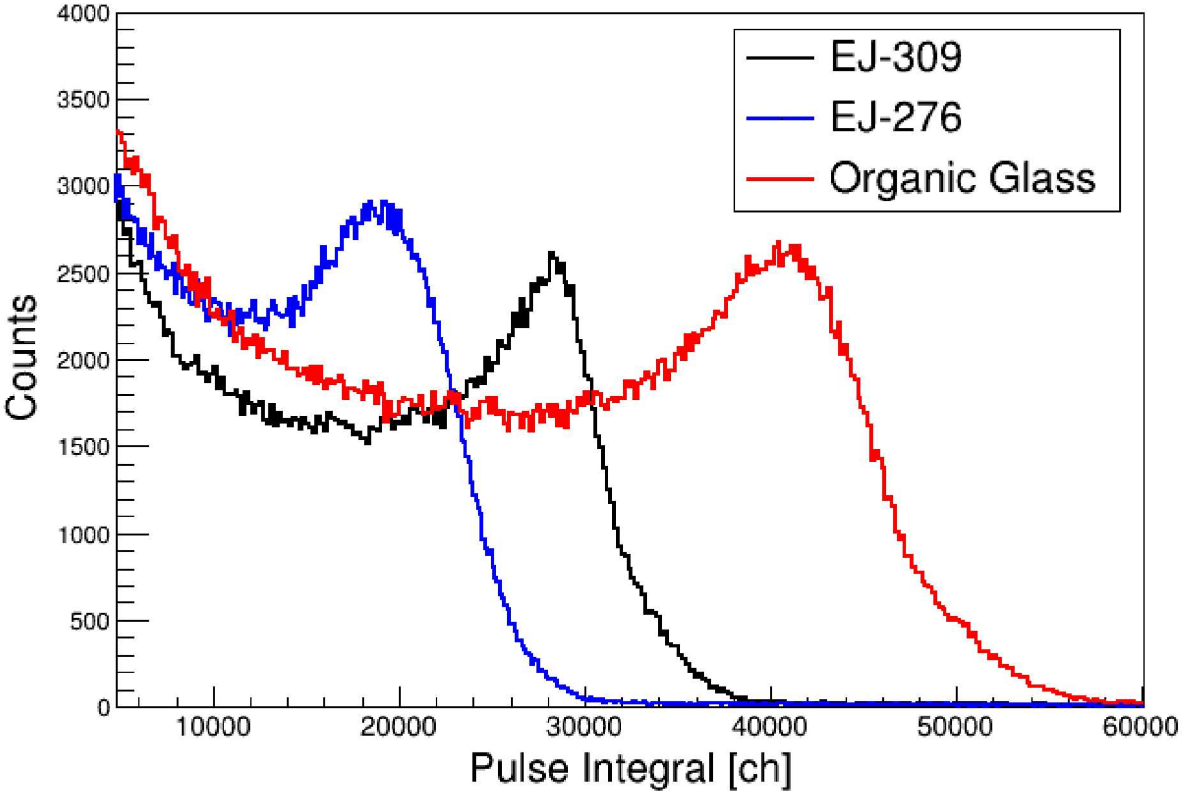
OGS/polymer blend: 8" x 8" x 1" slab

Highlights:

- Determined cost-effective synthetic pathway to industrial production of OGS powder
  - For production quantities of 100's of kilograms
- Optimized melt-casting procedures to enable net shape production
  - No polishing required
- Collaboration with industrial partner: successful pre-production OGS/polymer blends

UC-Berkeley – Thibault LaPlace, Bethany Goldblum

Electron Light Yield Comparison (2”x2”)



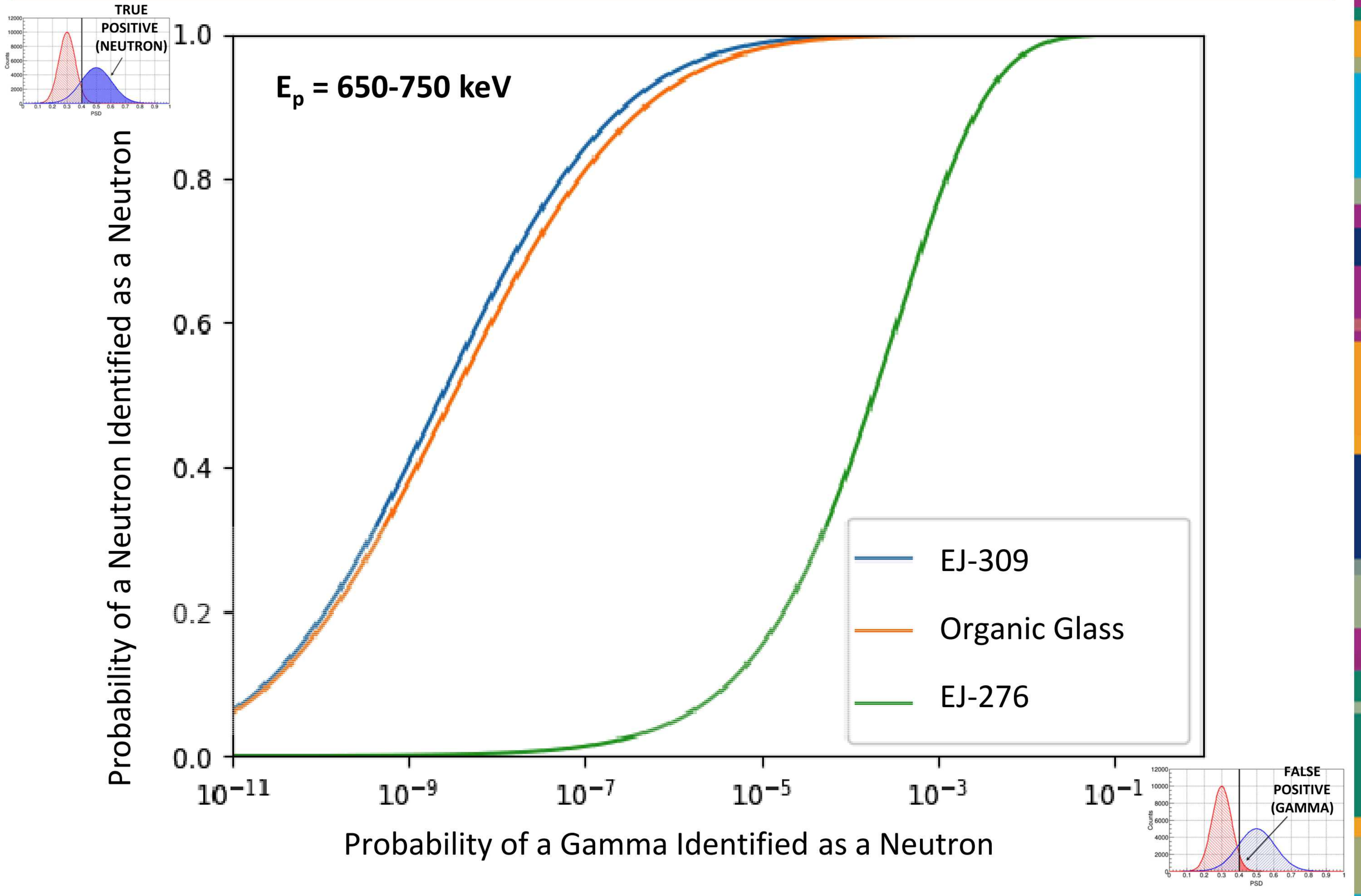
Scintillator	Light Output
EJ-309	1.00
EJ-276	0.72(3)
OGS	**1.46(5)**

EJ-309: PSD-capable organic liquid scintillator

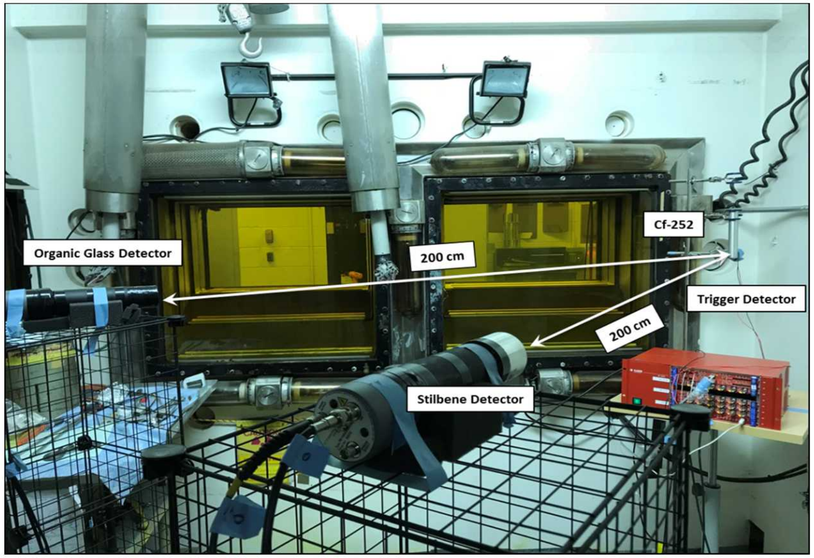
EJ-276: PSD-capable organic plastic scintillator

Organic Glass: PSD-capable organic glass scintillator

ROC Curve (2”x2”)

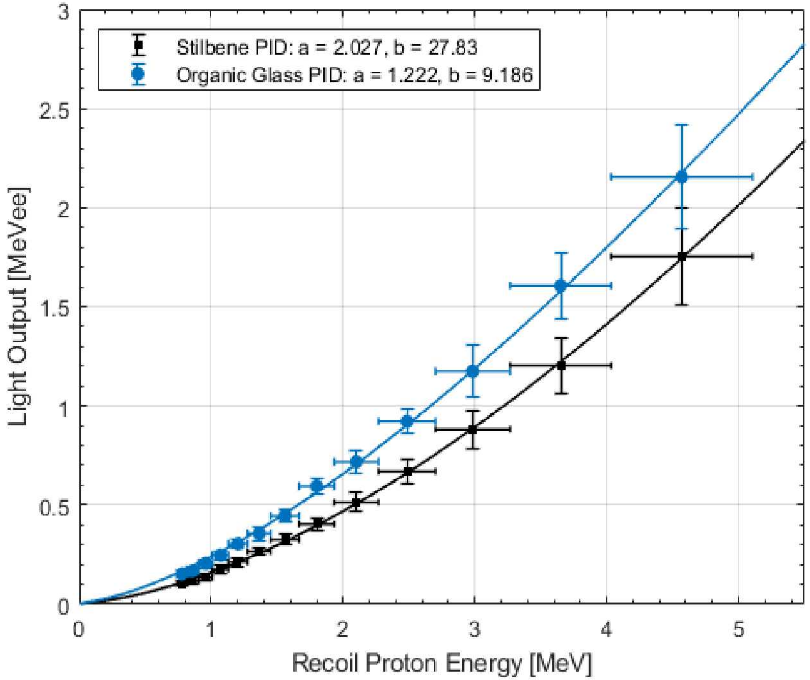


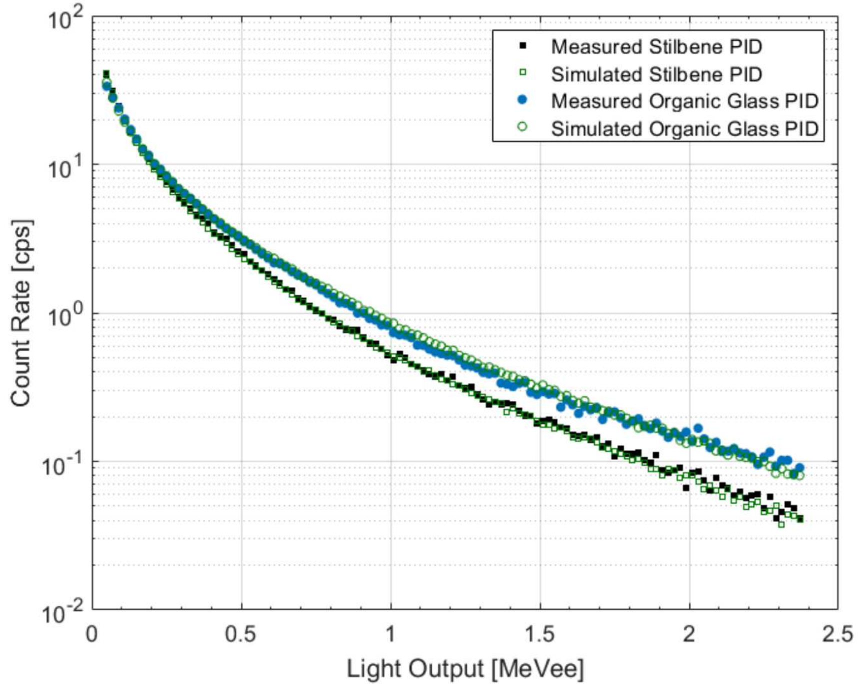
Cf-252 Time-of-Flight Experiments (2”x2”)



Detector	Lower light output threshold [MeVee]	Lower threshold in neutron-equivalent energy [MeV]	Upper light output threshold [MeVee]	Upper threshold in neutron-equivalent energy [MeV]	Calculated intrinsic neutron detection efficiency [%]
Stilbene	**0.06**	*0.60*	**2.40**	*6.40*	**28.66 ± 1.43%**
Organic Glass	**0.06**	*0.45*	**2.40**	*5.21*	**32.54 ± 1.63%**

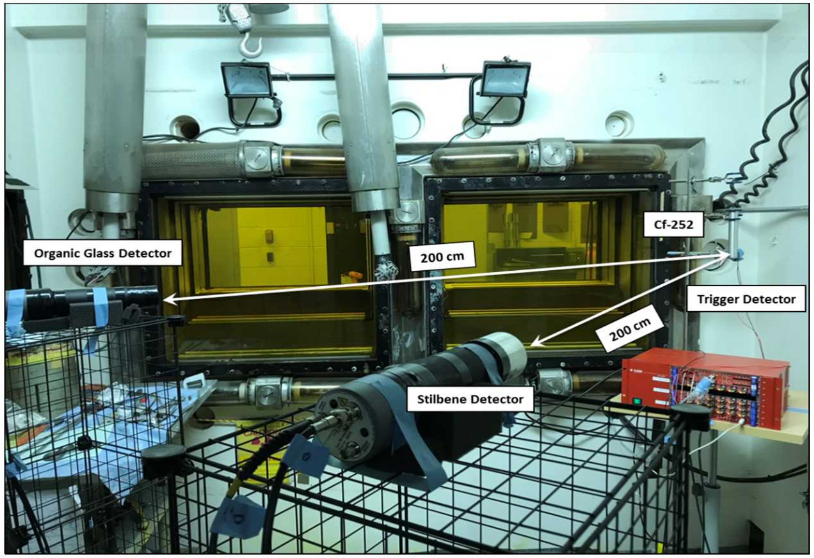
Higher n light yield for OGS provides improved low-energy detection





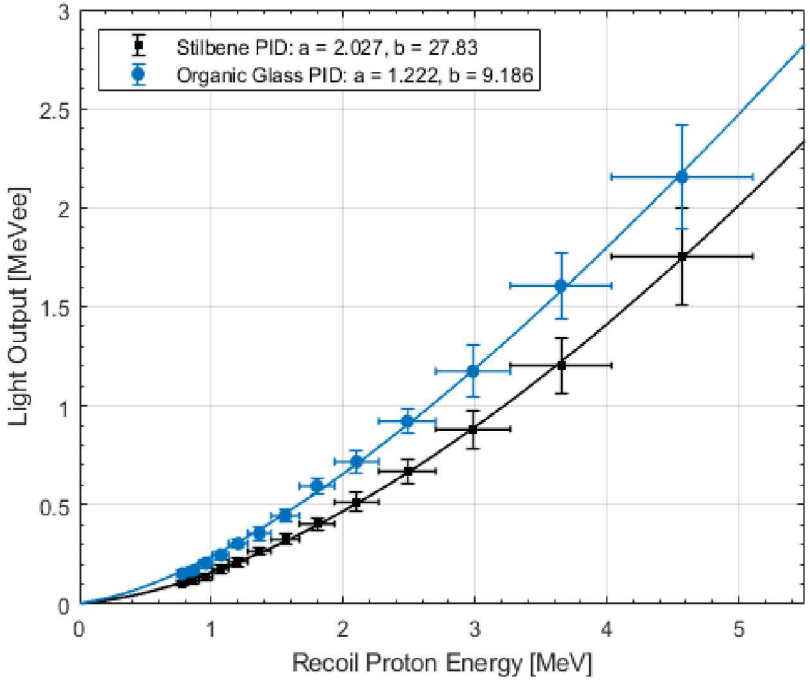
University of Michigan – Prof. Sara Pozzi, Tony Shin

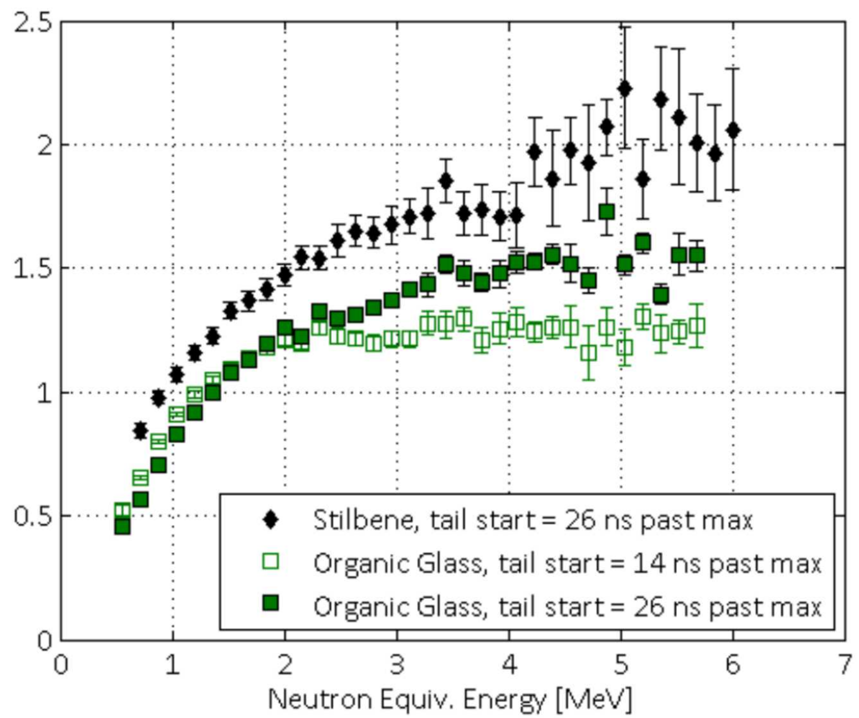
Cf-252 Time-of-Flight Experiments (2”x2”)



Detector	Lower light output threshold [MeVee]	Lower threshold in neutron-equivalent energy [MeV]	Upper light output threshold [MeVee]	Upper threshold in neutron-equivalent energy [MeV]	Calculated intrinsic neutron detection efficiency [%]
Stilbene	**0.06**	*0.60*	**2.40**	*6.40*	**28.66 ± 1.43%**
Organic Glass	**0.06**	*0.45*	**2.40**	*5.21*	**32.54 ± 1.63%**

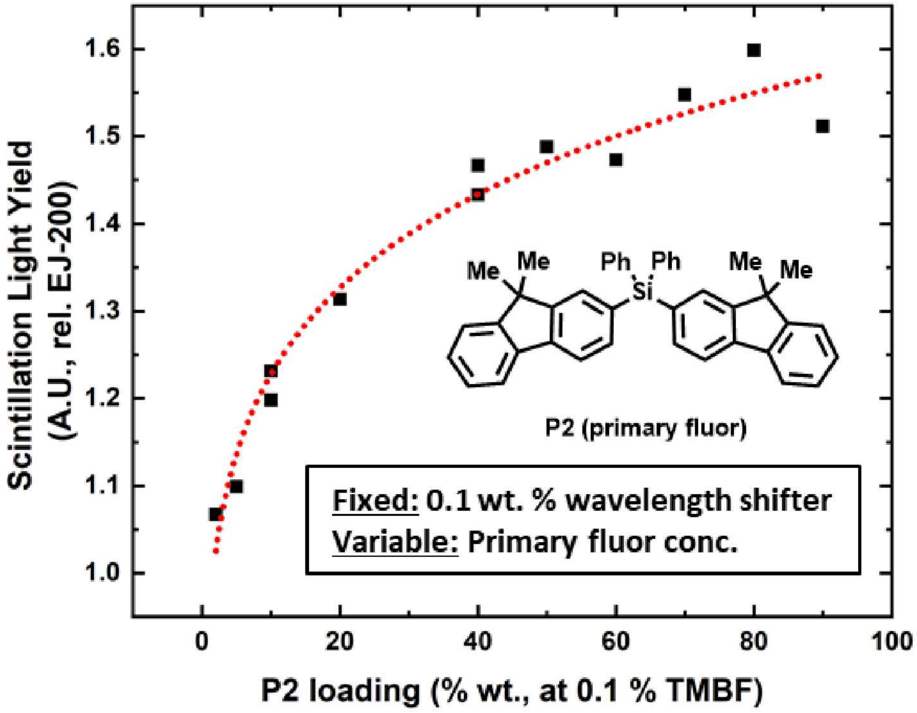
Reduction in OGS PSD for 2” size: Implies process and/or composition effects

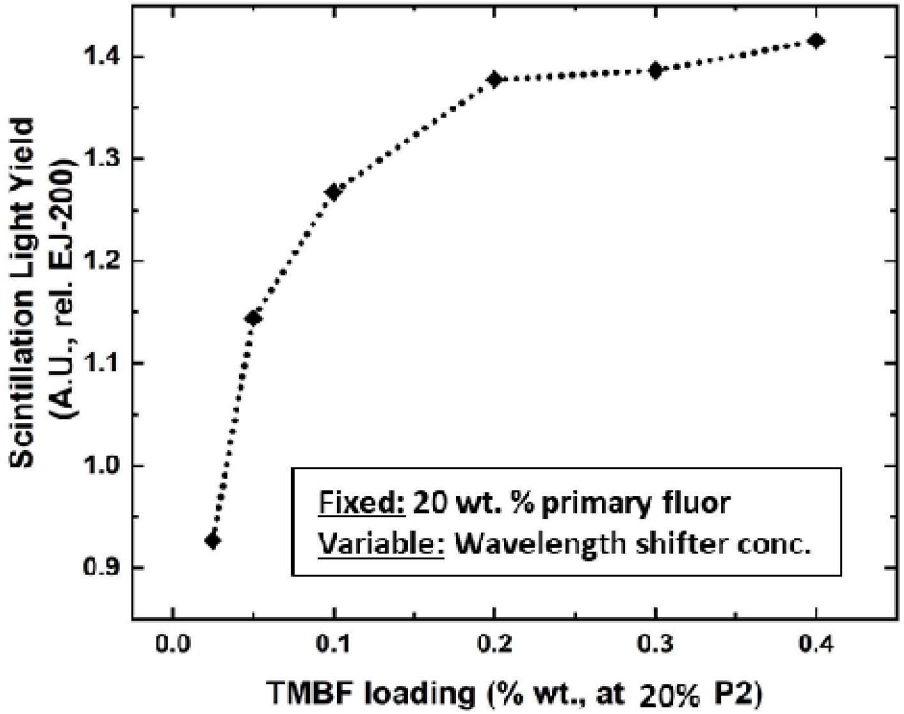




OGS/Polymer Blends

Light Output Optimization: P2/ TMBF wavelength shifter in PVT matrix





Conclusions:

1. 10-20 % P2, 0.2 % TMBF in PVT selected as economical high light yield formulation
2. Higher light yield can be selected per application by P2 loading
3. P2 loading at >30% gives rise to samples with γ-n PSD

Fast Timing Compositions

- Several applications require very fast lifetimes for excellent timing resolution (i.e. double-scatter n-TOF)
- A corresponding requirement comprises high light yield for improved detection efficiency
- **OGS and OGS/polymer blends exceeds state-of-the-art for both requirements**

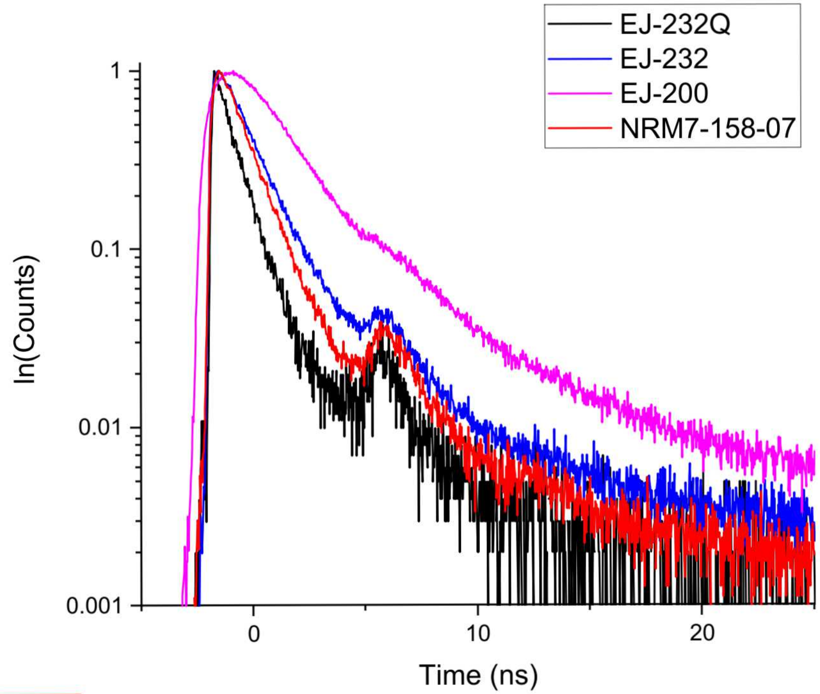


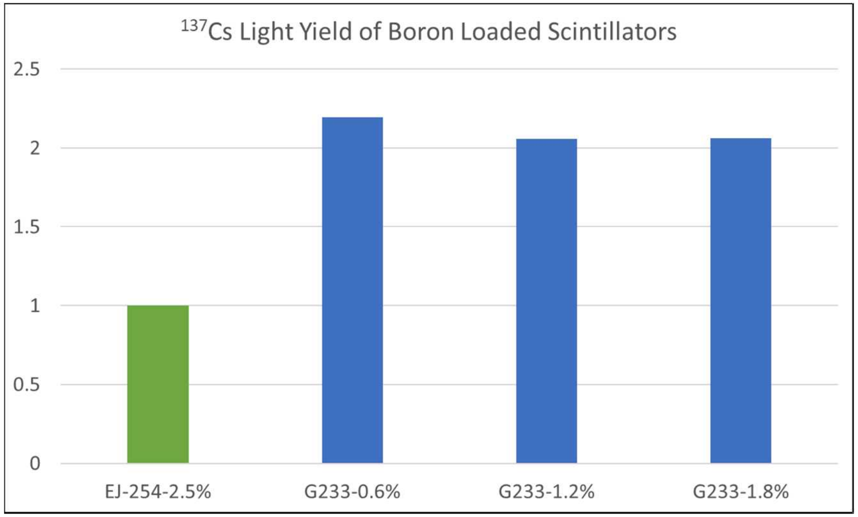
Table 1. Scintillation data for new formulations

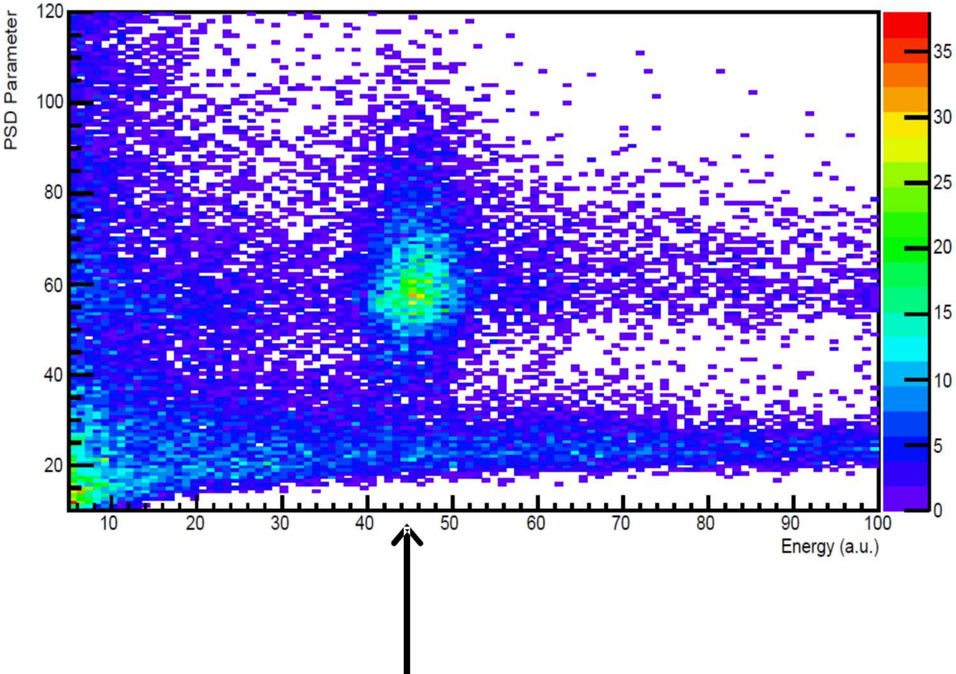
	Light Yield	Timing
Stilbene	100%	ca. 6 ns
EJ-200	61%	2.10 ns†
EJ-232	51%	1.60 ns†
Glass "Standard"	116%	1.71 ns
JSC6-88-A	**126%**	**1.46 ns**
JSC6-88-B	**99%**	**1.44 ns**
JSC6-88-C	**114%**	**1.24 ns**

[Faster than EJ-232 and >2x the light yield]

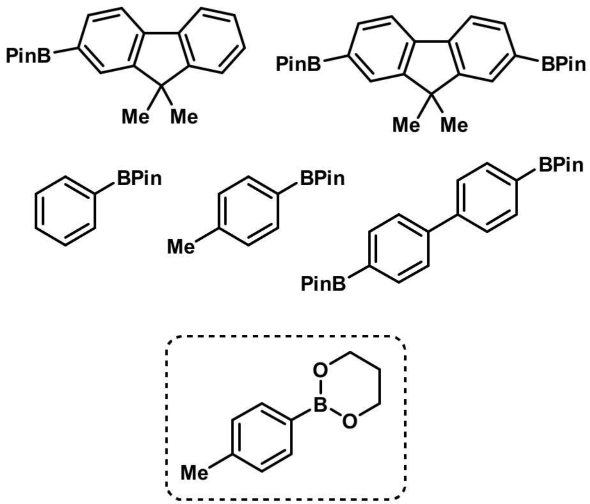
†From Eljen specification sheet

Boron-Loaded OGS





Boron Sources Used



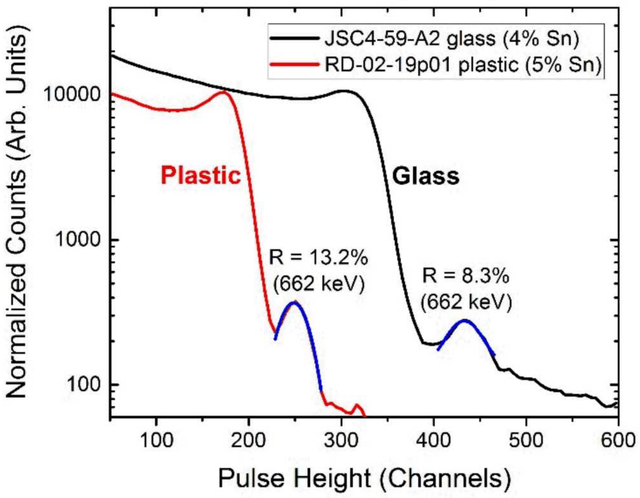
Sample: G205A-1.2%

1" diameter x 2" height sample
1.2% natural boron loading (0.24% ^{10}B)
30 min collection
Source: AmBe with HDPE moderation

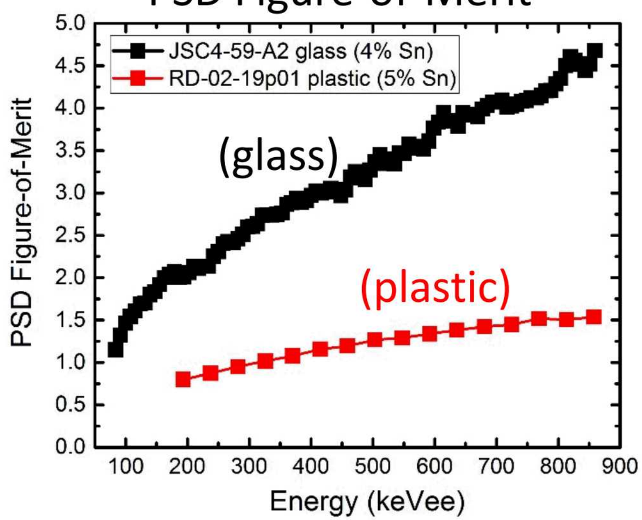
Combined PSD + Spectroscopy

[Proof-of-Concept in OGS Matrix]

^{137}Cs Pulse-Height Spectra



PSD Figure-of-Merit



Organic Glass as Host for Heavy-Metal Additives

Goal: Achieve gamma-ray spectroscopy and n/γ PSD in a single material

Prior Work: SNL and RMD have jointly demonstrated this capability in plastics (DNDO)

Limitations: Plastic scintillator host matrix provides modest light yield → limits spectroscopy and PSD performance

Significance of Present Results: Sn-loaded organic glass provides >1.5x the light yield and >2.5x better PSD-FOM than the highest-performing 'dual-mode' plastic scintillators.

Open Tasks: Scale-up, loading method, metal compound selection/synthesis