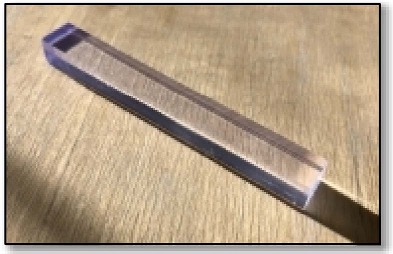
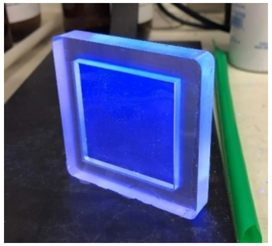
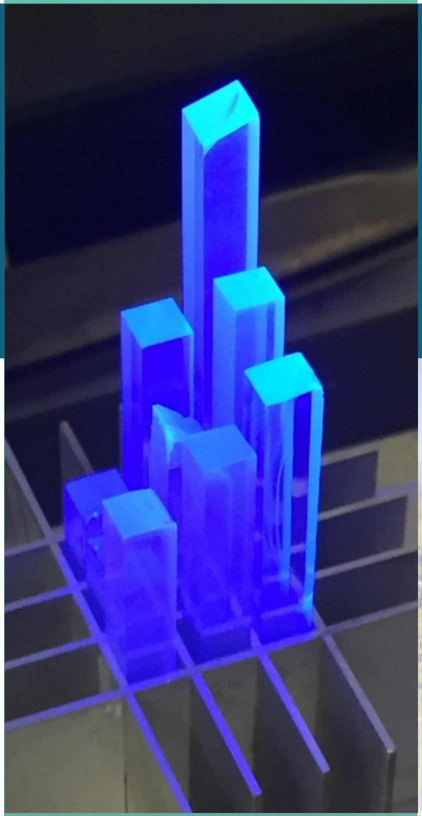
SAND2021-3192PE

Organic Glass Scintillators for Neutron Detection and Radiography







NSARD 2021 Program Review

Patrick Feng, Joseph Carlson, Huu Tran, Nicholas Myllenbeck, Lucas Nguyen, Annabelle Benin – SNL

Sven Vogel, Alex Long, Cort Gautier, Danielle Schaper – LANL
David Welker, Neil McIntyre – Incom, Inc. (subcontract)

Project Overview and Technical Approach

Problem / Need:

Existing organic scintillators have limitations that hinder their use and/or effectiveness in several application areas:

- Fast neutron transmission radiography (e.g. Non-destructive evaluation):
- Time-of-flight, high-rate environments (e.g. Single-volume scatter camera, pixelated neutron detector):
- High-efficiency and performance n detection/discrimination (e.g. Nuclear Physics)
- Challenging environments: (e.g. Radiation portal monitors, IC, space applications)

Light Yield	**PSD**	**Cost**	**Size**
Mechanical Prop.	**Thermal Prop.**	**Stability**	**Timing**

Differentiating Approach:

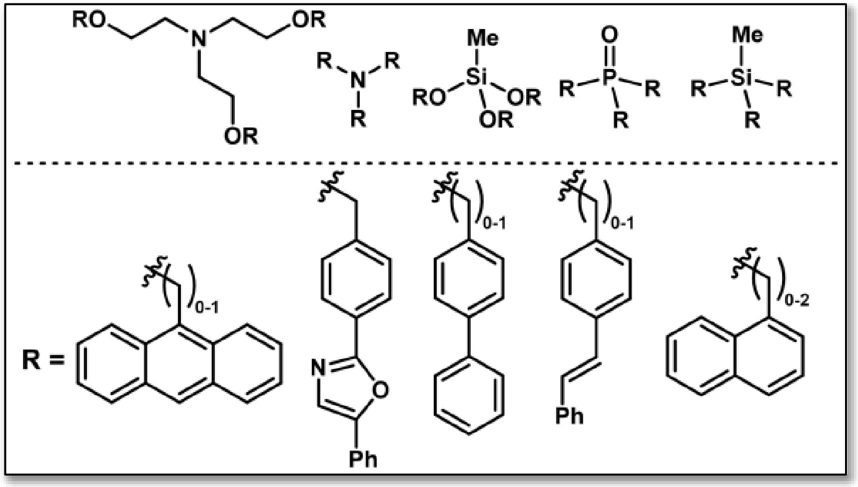
Apply and further develop Organic Glass Scintilators (OGS) to meet stated needs

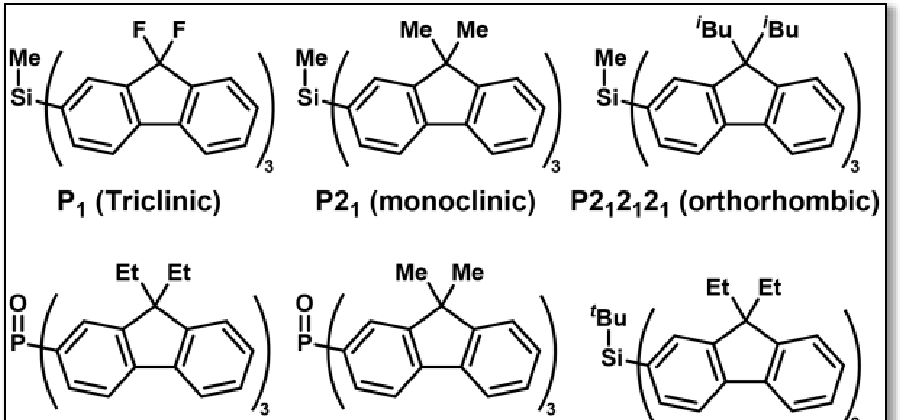
- Integrate OGS with Nanoguide optical waveguide technology developed at Incom

Organic Glass Scintillator (OGS): Definition

Material Class	Identical Molecules?	Thermoplastic?	Compatible with Additives?
Molecular crystals	✓		
Plastics		✓	✓
Liquids	✓		✓
(Molecular) Organic Glass	✓	✓	✓

- OGSs are based on random packing of identical molecules into stable amorphous phase
- Properties are a combination of crystals, plastics, and liquids
- OGS are fabricated by melt-casting or other thermal processing methods





P1 (Triclinic) P21 (monoclinic) P212121 (orthorhombic)

Pc (monoclinic) R-3 (trigonal) P-3 (trigonal)

Key Milestones and Progress to Date

Key Milestones	Task #	Results Highlights
Nanoguide production methods	2.1.1	Performed several full-scale draws using commercial plastic scintillator feedstock. First 'mini-draw' nearly complete using OGS-based scintillator material
Evaluate thermomechanical properties of OGS feedstock	2.1.2	Discovered additives and methods to modify melt-flow, T_g, fracture toughness of OGS
Light yield, PSD, and timing characterization/optimization	2.1.3	Characterized high light yield (up to ~2.5x EJ-200), fast timing (τ_1= 1.5 ns), excellent PSD
Photophysical and multi-functional additives	2.1.4	Wavelength shifters, thermal n capture compounds, viscosity modifiers synthesized and evaluated
OGS detector fabrication methods	2.1.5.1 2.1.5.3	Removable molds for optical-quality surfaces and 'in-situ' castings into permanent molds.
Scale-up of OGS-based feedstock for Nanoguide evaluation	2.1.5.2	Developed OGS-polymer blends compatible with Nanoguide processing requirements
Environmental aging-resistant formulations	2.1.5.5	Long-term stability via chemical composition, formulation, vacuum processing, and coatings.
X-ray and fast neutron radiography imaging and analysis	2.1.6	Constructed neutron radiography system at LANSCE FN-60R beamline. Completed two measurement campaigns to characterize Nanoguide and OGS formulations.

Project Timeline: Mid-way through a 3-year project (FY20-FY22)

Differentiating Photophysical Characteristics of OGS

Crystals	Quantum Yield (Φ_D)	Decay Time (ns)	(Donor) Peak Emission (nm)	Rel. ^{137}Cs Scint. Light Yield
trans-Stilbene	0.62	2.4–8.1*	385	1.00
Anthracene	0.64	12–26*	440	-
p-Terphenyl	0.67	4.2–11.5*	367	-
Plastics				
Poly(9-vinylcarbazole)	0.076-0.14	2.5 (monomer) 14 (excimer)	368 (monomer) 425 (excimer)	-
Polystyrene / PVT	0.12-0.18	12 (monomer) 20 (excimer)	285 (monomer) 332 (excimer)	0.54 (EJ-200)
Liquids				
Toluene	0.14	34	302	-
1-Methylnaphthalene	0.25	27.6	337 (monomer) 428 (excimer)	0.30
4-Isopropylbiphenyl	0.25	9.7	328	0.10
N-(2-ethylhexyl)carbazole	0.30	20.6	368 (monomer) 420 (excimer)	0.50
2,6-Diisopropylnaphthalene	0.36	30.6	346	-
p-Xylene	0.40	8.2	290	-
1,2,4-Trimethylbenzene (pseudocumene)	0.41	2.15	362	0.63 (EJ-301)
Organic Glass				
bis-(9,9'-dimethylfluoren-2-yl)diphenylsilane (**P2**)	0.66 (glass)	1.7	387	1.14
tris-(9,9'-dipropylfluoren-2-yl)methylsilane	0.78 (crystal) 0.48 (glass)	-	395	1.23
4,4'-*bis*-((*E*)-4-(9H-carbazol-9-yl)styryl)biphenyl	0.69	1.6	453	-
bis-(9-phenylcarbazolylbenzofuran), (**BPBFCz2**)	0.87	1.8	427	-

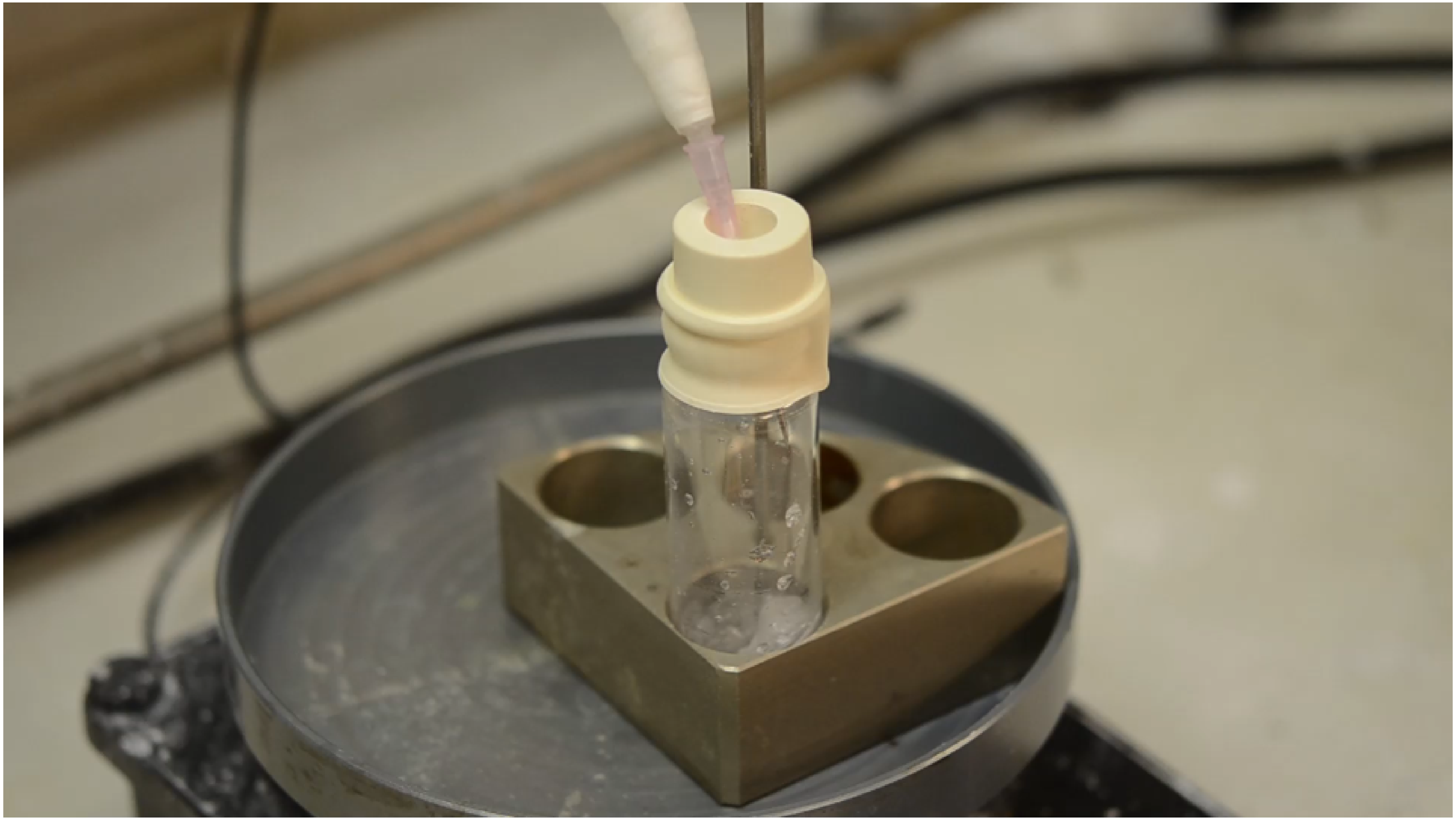
Förster Equations: $E = \dfrac{1}{1 + \left(r/R_0\right)^6}$ *(FRET efficiency)*

$$R_0^6 = \frac{2.07}{128\pi^5 N_A}\frac{\kappa^2 Q_D}{\eta^4}\int f_D(\lambda)\varepsilon_A(\lambda)\lambda^4 d\lambda$$

$$k_{ET} = \left(\frac{R_0}{r}\right)^6 \frac{1}{\tau_D}$$

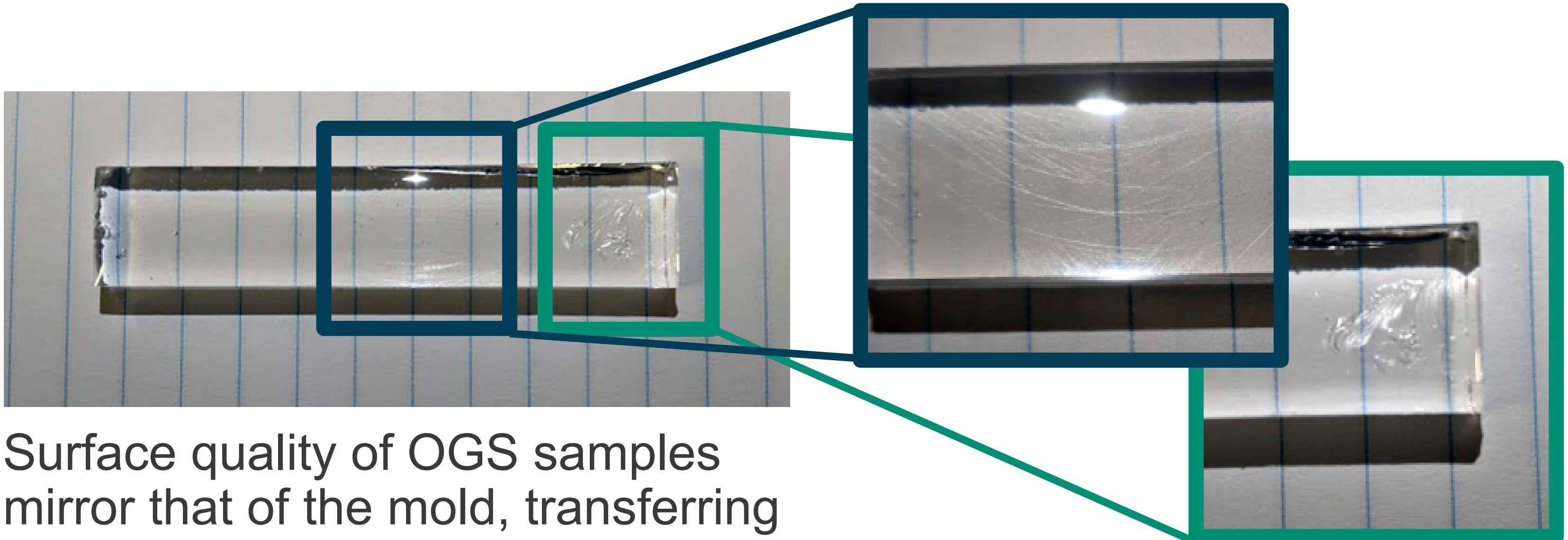
(FRET rate)

OGS: Melting and Casting Samples

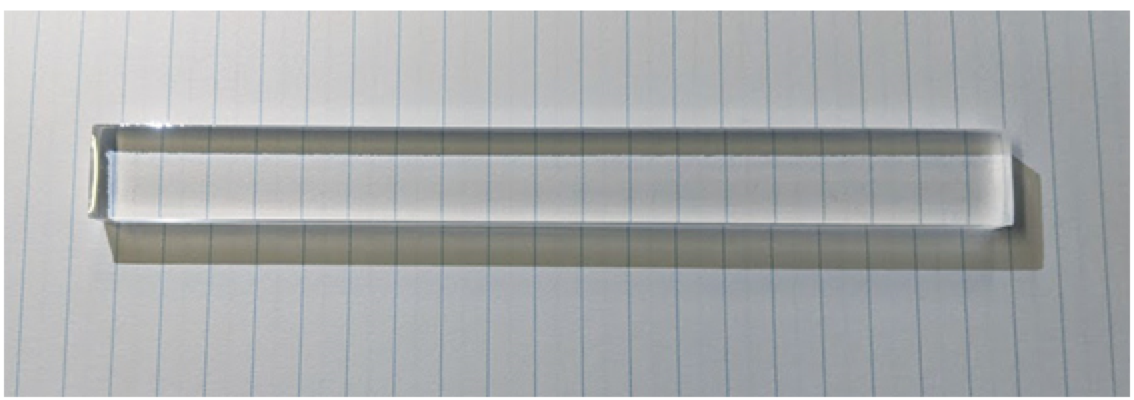


Video showing representative OGS melt-casting procedure

OGS Surface Quality depends on Mold Finish Quality



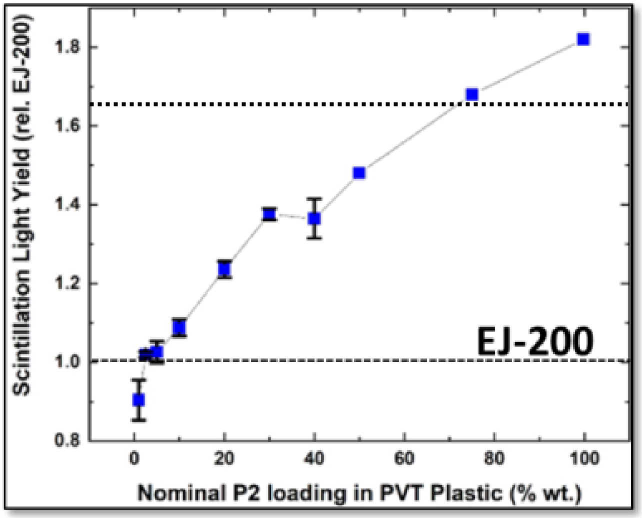
Surface quality of OGS samples mirror that of the mold, transferring any defects.



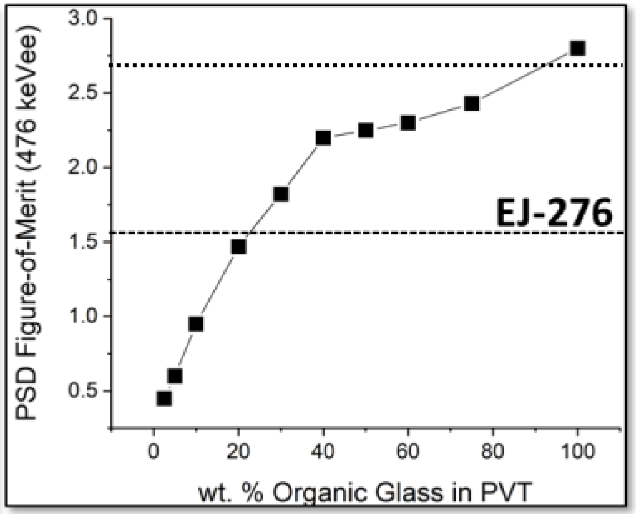
A mold with good finish enables high quality samples shown above. OGS slab sample was taken directly out of mold without polishing or coating.

Hybrid Scintillators: OGS Blended with Polymers

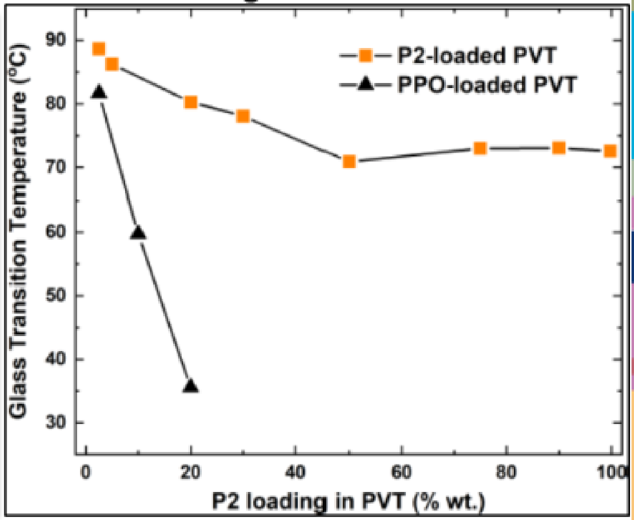
Light Yield vs. % OGS

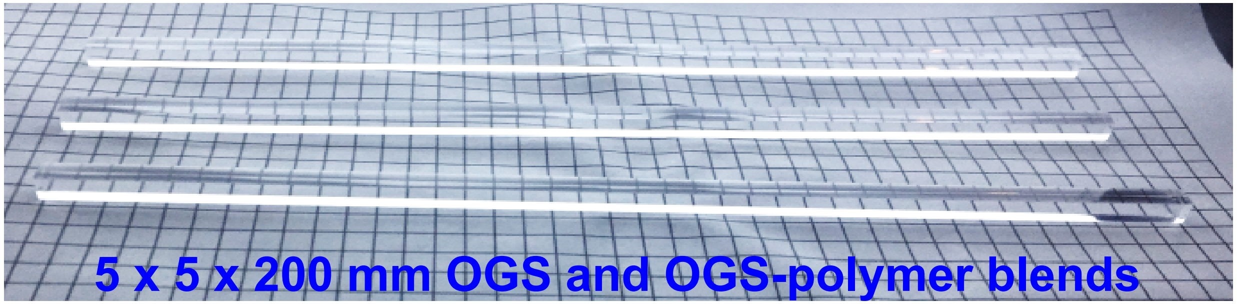


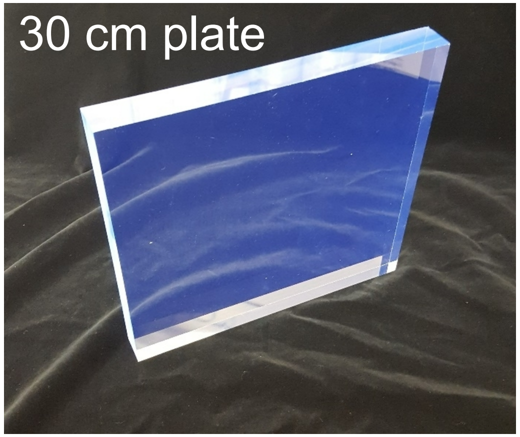
PSD vs. % OGS



T_g vs. % OGS



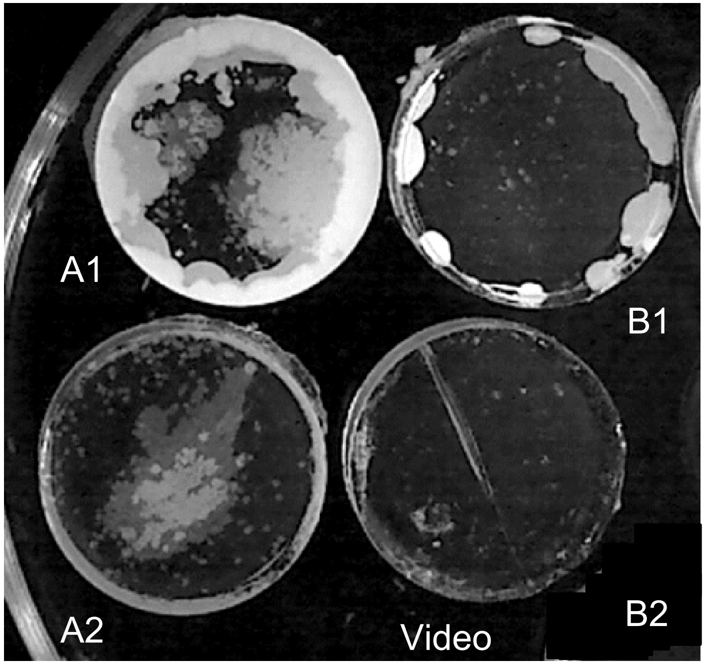


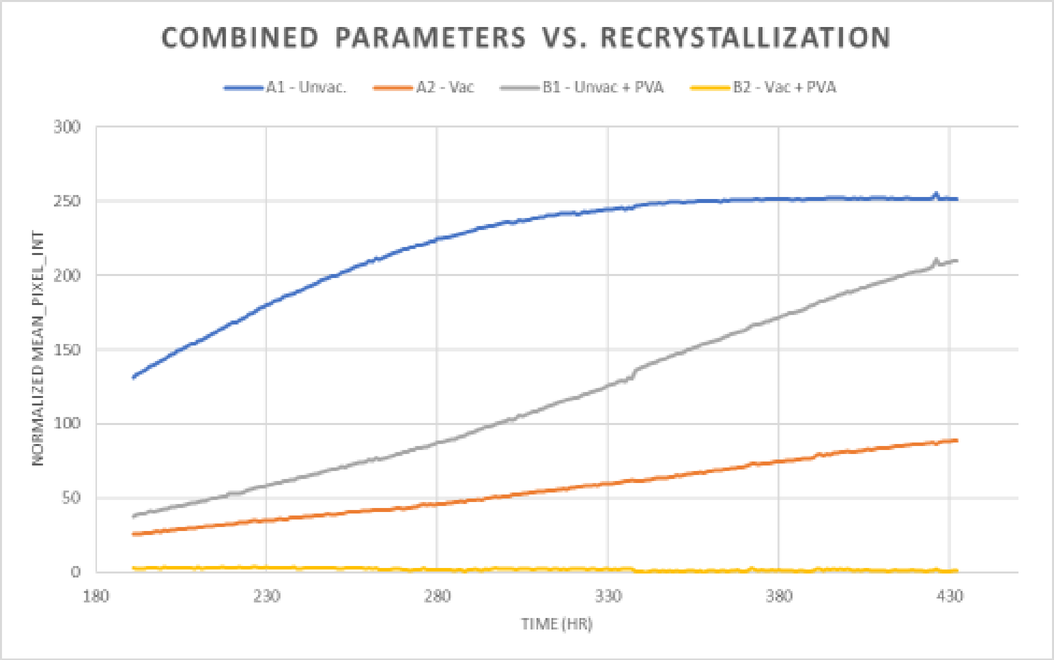


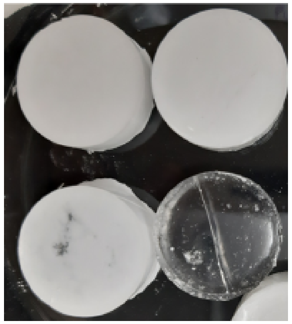
OGS fluorophores can be blended with polymers in all ratios (0-100%), leading to excellent scintillation properties and little to no change in T_g

Long-Term Stability of the Glassy State

Sample	10% PB3 Additive	Vacuum / Degas Applied to Molten OGS	PVA Coating
A1	+		
A2	+	+	
B1	+		+
B2	+	+	+

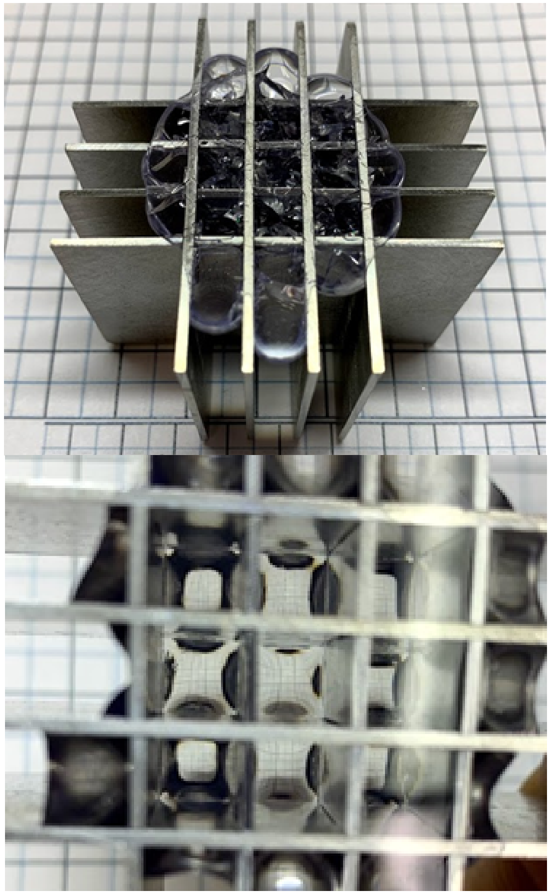




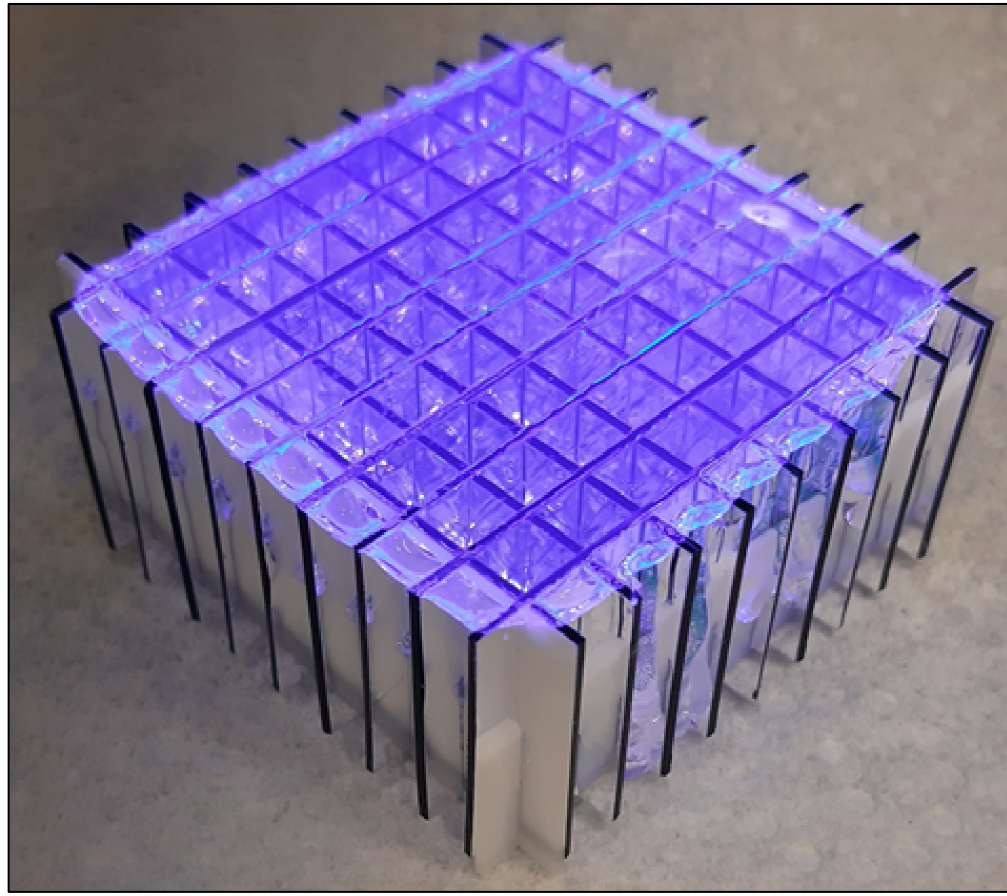


After 4 months at 60°C

‘Cast-in-Place’ Detector Arrays Based on OGS

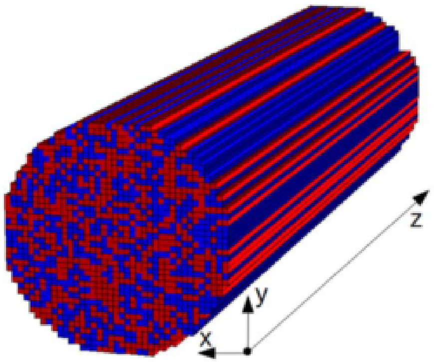


Higher loadings (15%) of mesitylene (top) or TDB (bottom) enable successful casting into metal or polymer arrays



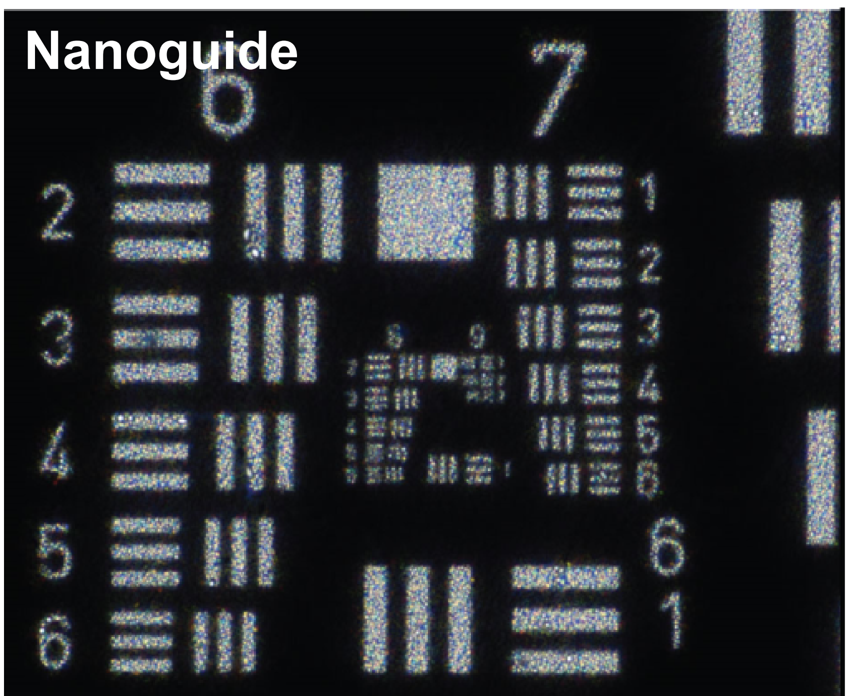
10% loading of TDB in OGS cast into an 8x8 array using black plastic acetal walls lined with ESR reflective material.

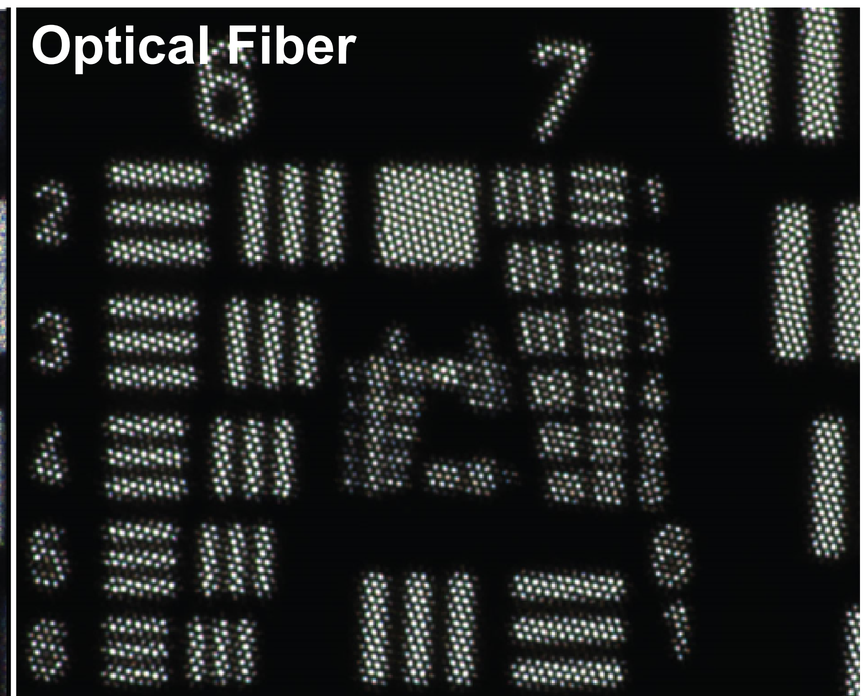
Comparison of Nanoguide vs. Optical Fiber Bundle



Nanoguide – Axial View
(~95% transmissive, ‘clear’)

Nanoguide –Transverse View
(0% transmissive, ‘opaque')





Scale-Up for Custom Nanoguide Feedstock

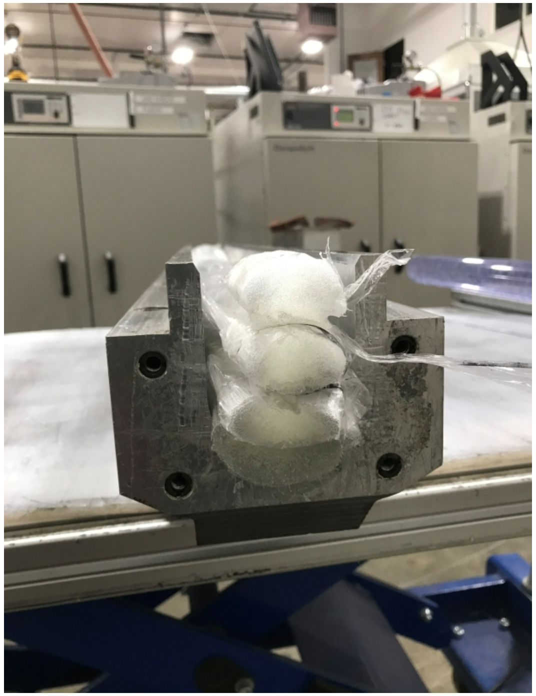
Polycarbonate end caps were used to take up additional space in fusing channel

Additionally, rod is showing visible scintillation in the presence of UV sun light

- Melt-flow measurements indicate up to ~35 wt. % OGS in PS is suitable for fiber drawing
- Multiple 3-4 kg batches synthesized and processed into 36” x 2.5” pre-forms, ~200 μm fibers, and eventually ~200 nm domain size Nanoguide

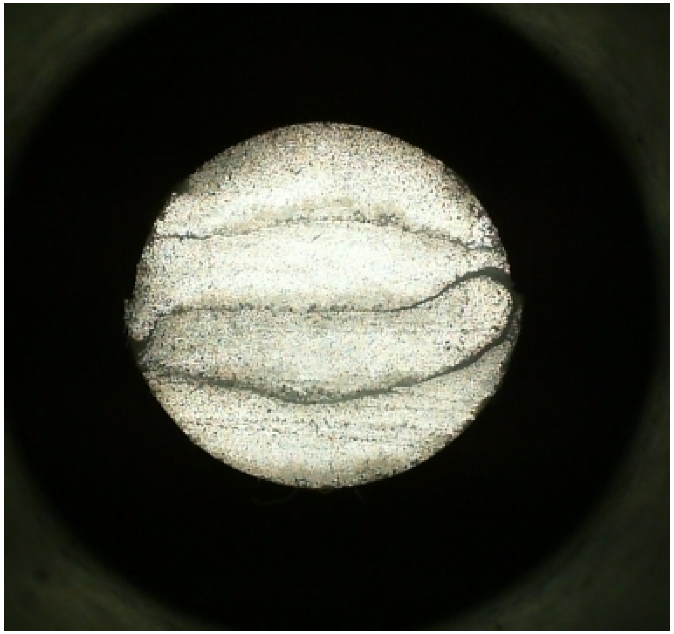
Quarter Nanoguide Draw with OGS-PS Material

Quarter Nano direct draw result (1st draw)

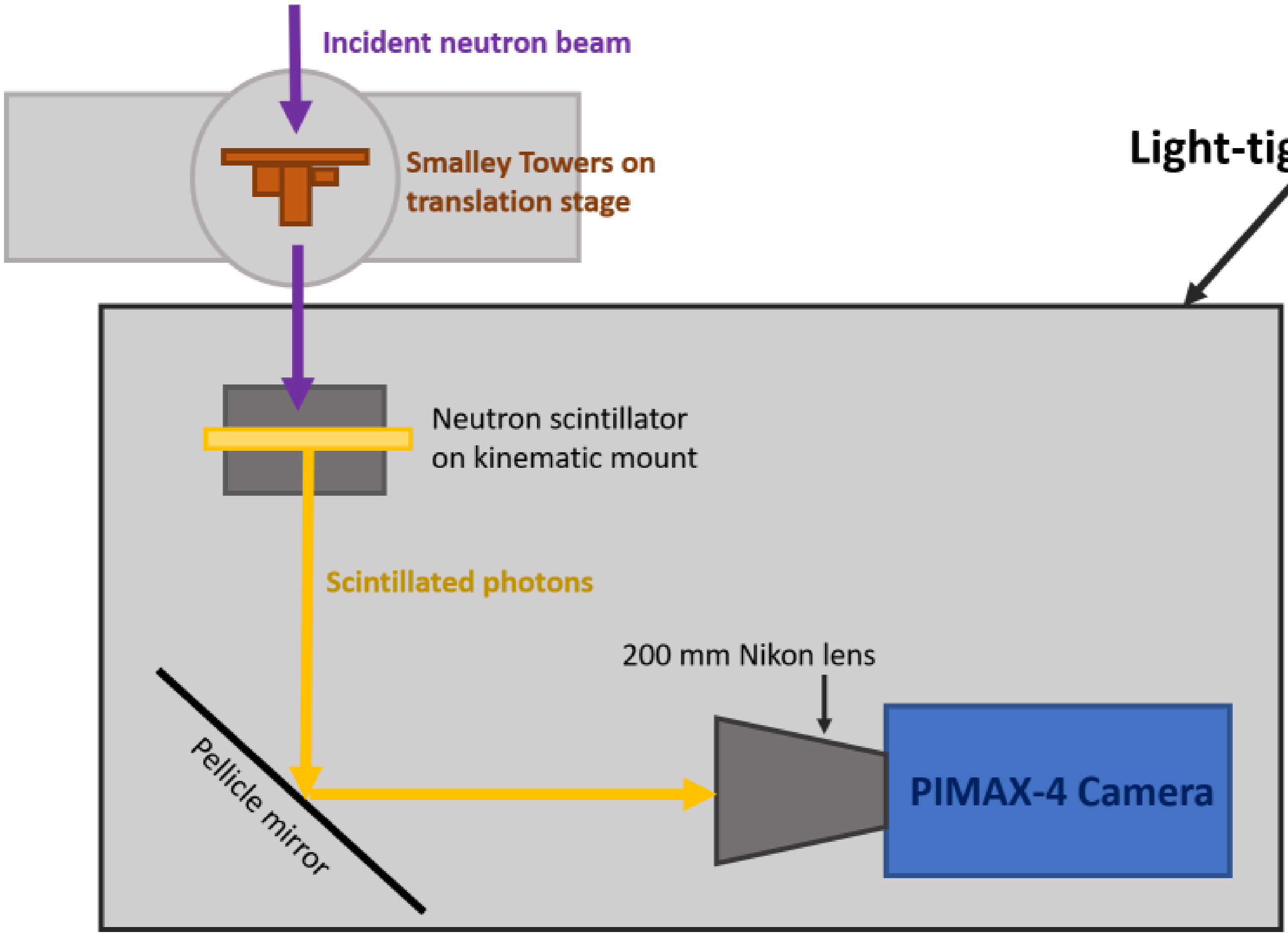


Quarter Nano direct draw (1st draw) – compressed into fusing die

Quarter Nano direct draw result (1st draw) – pre-fusing transmission photo



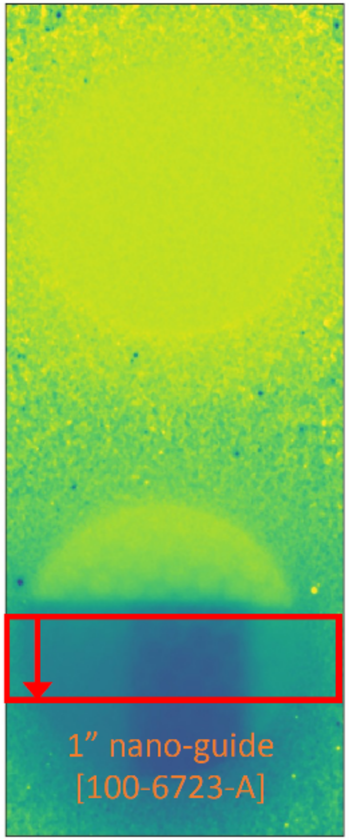
Neutron Radiography Setup at LANSCE (FP60R)



FP60R views an unmoderated tungsten source (Target 4)

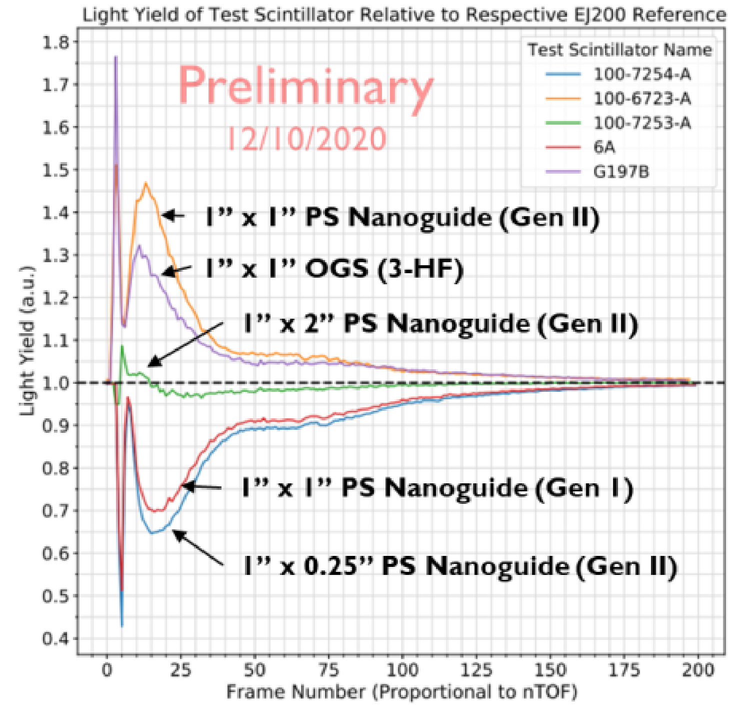
Flight path length used in calculations was L = 21.057 m

Initial Neutron Radiography Results (1-14 MeV)



Initial Observations:

- No apparent resolution reduction in 1” vs 2” thick Nanoguide
- Potential for higher *detected* light output in Nanoguide vs. bulk scintillator



Sample Description	Thickness (in.)	FWHM (microns)
OGS’ (Bulk)	1	247
50:50 Nanoguide-Gen1	1	218
50:50 Nanoguide-Gen1	2	219
80:20 Nanoguide-Gen2	1	274

Future Work

- Full analysis of neutron radiography data from 12/2020 beamtime, preparation for 2021 measurements
- Refinement of Nanoguide process conditions for custom OGS-polymer feedstock
- Multi-functional OGS compositions (e.g. gamma-ray spectroscopy, thermal neutron capture)
- Model-guided optimization of wavelength shifter concentration for each OGS composition
- In-situ mold design:
  - Dimensional tolerances for position-sensitive array coupling
  - Enhance light transport properties,
  - Alternative additives for ‘soft’ manufacturing approach
  - Strategies for ‘hard’ manufacturing approach
- Technology transfer to industry

Related Projects and Collaborations

Lab Collaborations:

- NA-22 project "Pixelated Neutron Detector", PI: Melinda Sweany (present-FY23)
- NA-22 project "Single Volume Scatter Camera", PI: Erik Brubaker (present-FY21)

SBIR/STTR Programs:

- Phase II DTRA STTR with RMD: *"Multimode Organic Scintillators for Neutron/Gamma Detection" Start date: 02/21/2021.*
- Phase I DOE-Nuclear Physics SBIR with RMD: *"Organic Scintillators for Nuclear Physics Experiments" Start date: 03/01/2021.*
- Phase I NSF SBIR with Blueshift Optics: *"Organic Glass Scintillator Integration into 64-channel Pixelated Array" Start date: 02/01/2021.*

Other Active Collaborations:

Bill Warburton (XIA, Inc.), Katherine Mesick (LANL), Sara Pozzi (Univ. of Michigan), Chris Allwork (AWE-Aldermaston), Ilker Meric (Western Norway Univ.), Marek Moszynski (Natl. Centre for Nucl. Res, Poland), Jose Valiente Dobon (Instituto Nazionale di Fisica Nucleare, Italy), Jason Legere (Univ. of New Hampshire), Tony Shin and Katherine Mesick (LANL)

Thank you!