Polymer-blend Organic Glass Scintillators for next generation neutron detectors at FRIB

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Radiation Monitoring Devices, Inc.

Mission

- Perform world class research
- Develop exceptional commercial products

Overview

- Founded 1974
- R&D and commercial products
- Acquired by Dynasil in 2008
- 80 employees

Research Expertise

- Semiconductors
- Scintillators
- Advanced Sensors
  - Nuclear
  - Optical
  - Magnetic
- Instruments & Systems
- Imaging
- Coatings

Organic Glass Scintillators for Nuclear Physics Experiments

Urmila Shirwadkar[1], Edgar V. van Loef[1], Tawan Jamdee[1], Lakshmi S. Pandian[1], Chuncheng Ji[1], Jarek Glodo[1], Patrick L. Feng[2], Annabelle Benin[2] Ryan Witzke[2], Remco G. T. Zegers[3], Jorge Pereira[3], and Cavan Maher[3]

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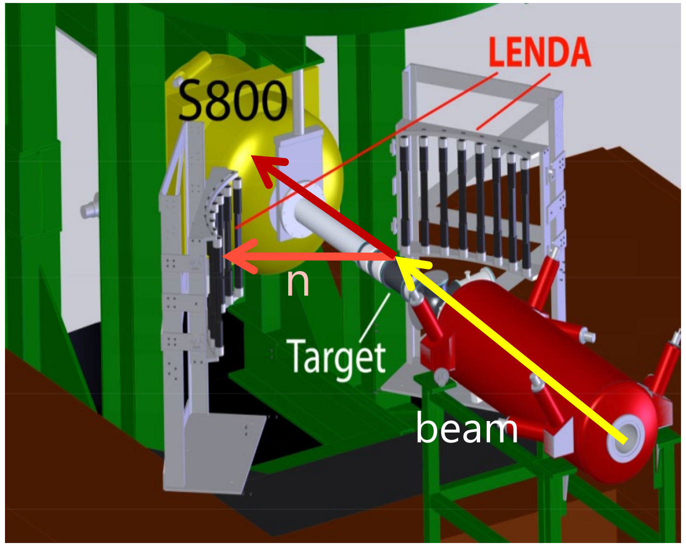
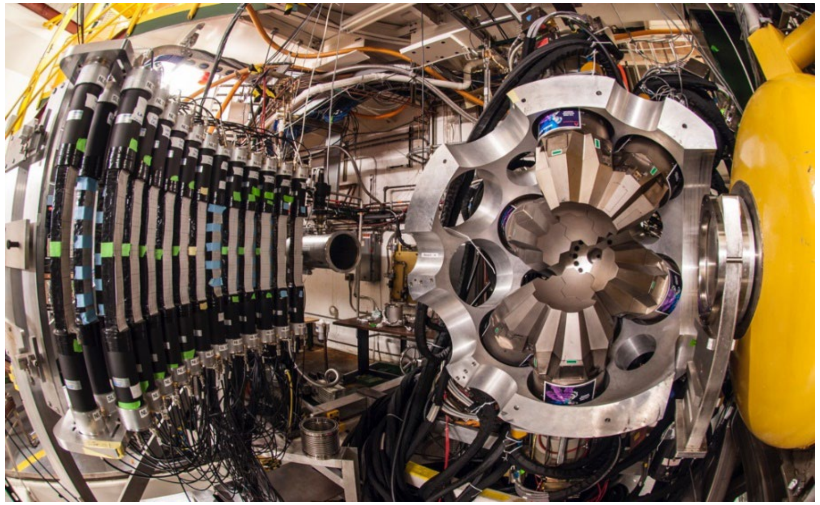
POP: 4/4/2022 - 4/3/2024

Neutron detectors at FRIB



- Low Energy Neutron Detector Array (LENDA)* is used as a neutron time-of-flight (ToF) spectrometer developed for inverse kinematics (p,n) charge-exchange experiments in which low energy neutrons (~150 keV to 10 MeV) are detected.
- Versatile Array of Neutron Detectors at Low Energy (VANDLE) array is a hybrid neutron time-of-flight (TOF) detector consisting of plastic bars optimized for measurements of beta-delayed neutrons between 100 keV and 5 MeV
- LENDA Consists of 24 BC-408 plastic scintillator bars with dimensions 30 x 4.5 x 2.5 cm, each coupled to Hamamatsu H6410 PMT to each end of the bar
- These detector arrays are unable to differentiate between neutrons and gammas, complicating the background subtraction
- Separation between neutrons and photons is possible by time-of-flight, but it doesn't work for indirectly produced photons (e.g. produced by neutron-induced photons of surrounding materials)



LENDA+GRETINA@S800 picture: S. Noji

Ref: G. Perdikakis et. al. LENDA: A low energy neutron detector array for experiments with radioactive beams in inverse kinematics, NIMA, Volume 686, 2012, Pages 117-124

Ideal detector requirements

- Separation between neutrons and photons by pulse-shape discrimination (PSD)
- Need to cost-effectively cover large area
- Good timing resolution (< 1 ns)
- Position resolution (~5 cm)
- Low neutron-detection threshold (<200 keV)

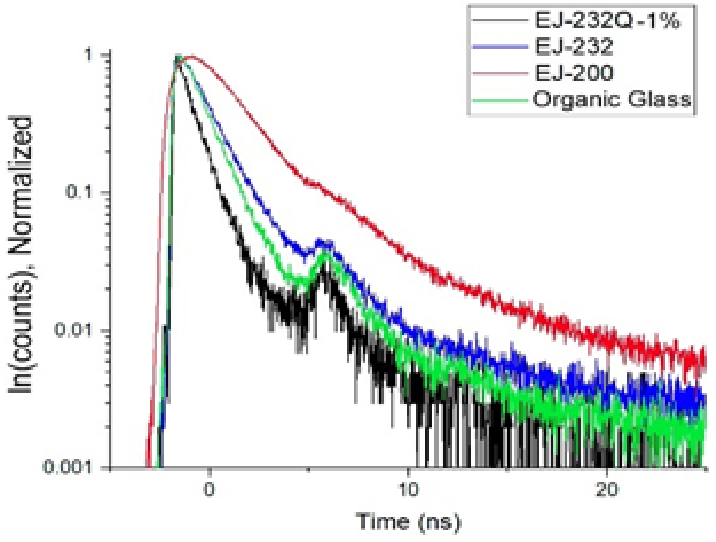
Issues with current detectors

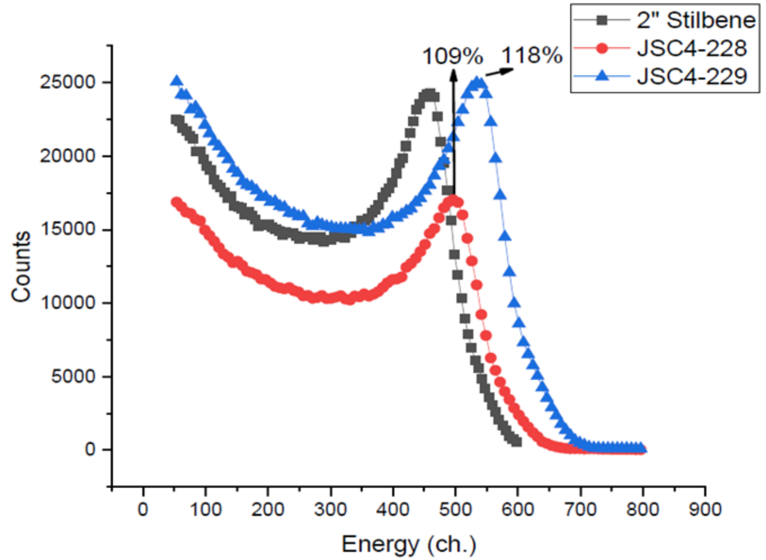
- PSD slows down speed – affects timing
- Difficult to achieve with liquid scintillators that have PSD capability – toxicity, size, and mobility

Limited number of alternative options available and further development of capabilities is required

Organic glass scintillators as a solution

- Ability to provide PSD between gamma rays and neutrons
- Ability to scale up to large sizes
- High light yields of OGS allows for lower energy thresholds in PSD measurements and therefore a wider range of neutron energies to be detected
- Fast response
- Low cost



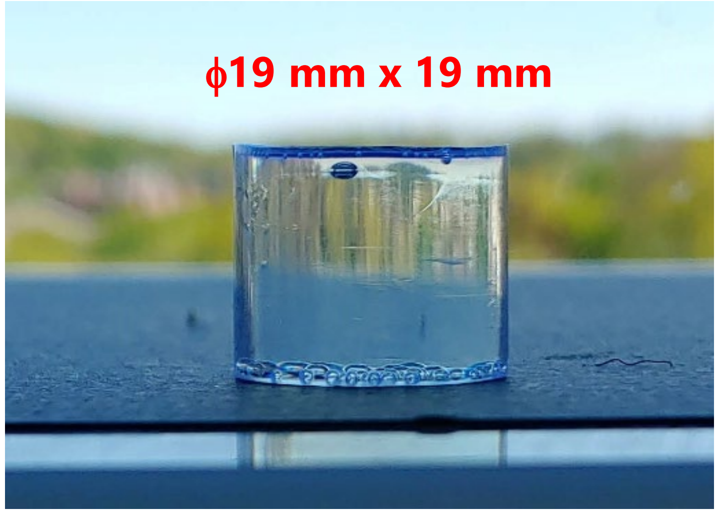


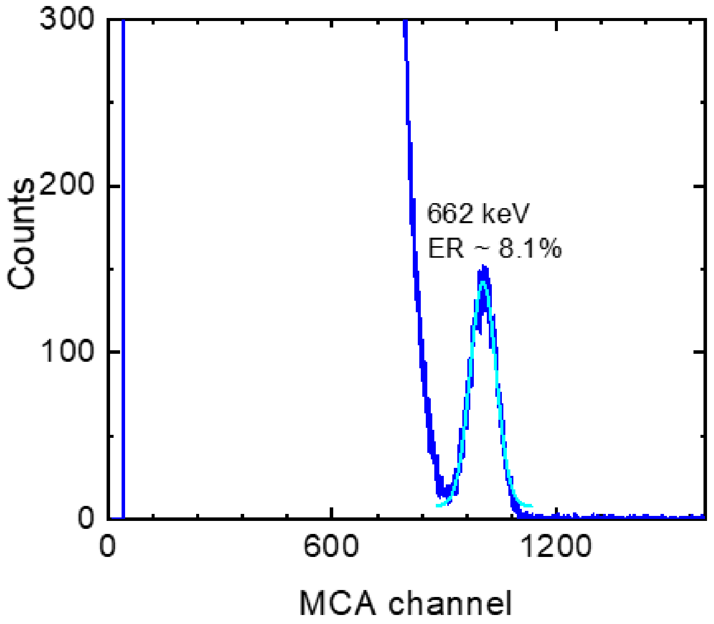
Scintillator	Emission (nm)	Light Yield (Ph/MeV)	Decay (ns)	PSD (FOM)
Stilbene	390	16,000	2.4-4.5	> 3
EJ-200	425	10,000	2.1	NO
EJ-232	370	8400	1.6	NO
EJ-232Q-1%	370	1700	0.7	NO
EJ-276	425	8,600	13	~ 2
Organic Glass Scintillators*	430	18,000	1.46	> 3

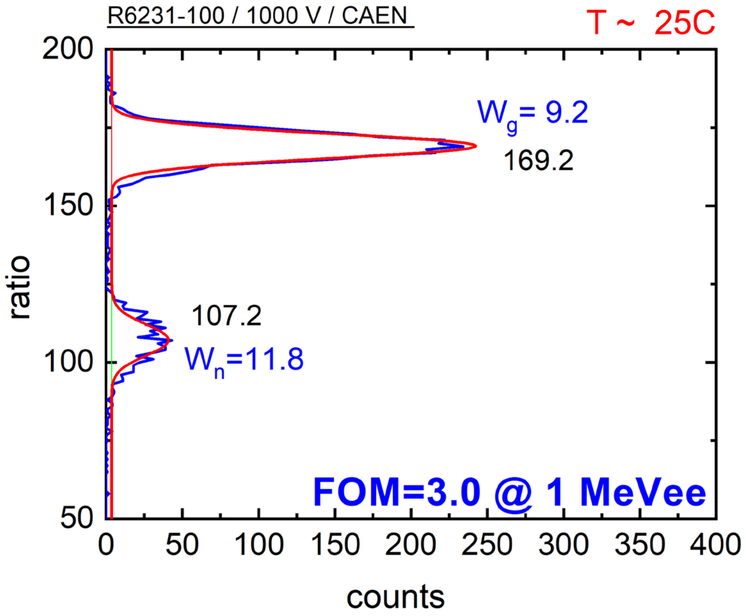
****Ref: Joseph S. Carlson, Patrick L . Feng, "Melt-cast organic glasses as high-efficiency fast neutron scintillators", Nucl. Instr. Meth. Phys. Res. A 832 (2016) 152-156***

Tin-Loaded OGS

- Increased gamma-ray sensitivity, High Z_{eff} - spectroscopic capabilities
- Several reaction channels in one experiment, optimization of scientific output of complex and high-cost FRIB runs
- No compromise on the neutron detector solid angle to measure simultaneous gamma rays







Polymer-blended OGS

- Standard OGS are too brittle for high aspect ratio detectors that need to be self-supporting (30 x 4.5 x 2.5 cm3)
- Polymer blending with OGS can significantly enhance the mechanical properties of OGS*
- The core matrix material is an OGS-polymer mixture that comprises a small amount of polycarbonate or PVT blended into the OGS matrix.
- This polymer-blended OGS composition can be scaled up while maintaining high-performance similar to the standard OGS

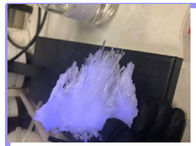
Ref: Nicholas R. Myllenbeck et. al. "Nano-segmented optical fibers containing molecular organic glass scintillator for fast neutron imaging", Proc. SPIE 11838, Hard X-Ray, Gamma-Ray, and Neutron Detector Physics XXIII, 118380R (1 September 2021); https://doi.org/10.1117/12.2596532

Starting Materials, Synthesis and Purification

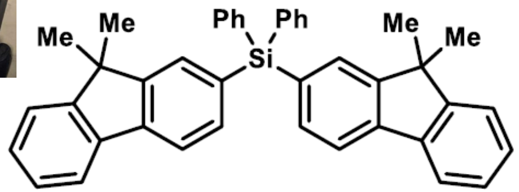
- **Starting Materials**
  - Vinyltoluene/Polycarbonate - Alfa-Aesar/Fisher Scientific/Sigma-Aldrich
  - OGS base material, bis(9,9′-dimethylfluoren-2-yl)diphenylsilane (P2) - Jubilant Biosys
  - Organotin compound, trimethyl(4-vinylphenyl)stannane - Adesis, Inc. (New Castle, DE)
  - bis-MSB - Sigma-Aldrich
- **Synthesis**
  - Sandia prepared polycarbonate, Bis-MSB, and OGS compound
  - RMD synthesized tin compound (TMS) as additives for incorporation into the OGS-Polymer blend
- **Purification**
  - Done at RMD and Sandia

Polymer Organic Glass Scintillators

- Controlled polymerization of mixtures by dissolve OGS base material and a small amount of 1,4-bis[2-methylstyryl]-benzene (bis-MSB) into the styrene monomer or polycarbonate solution, and for tin version also add the organotin compound.
- Vary the concentrations of OGS and polymer to optimize polymer OGS composition
- Vary the concentrations of organotin compound to optimize light yield and PSD

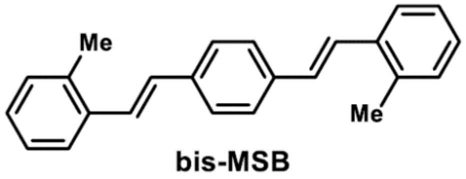


Primary fluorophores



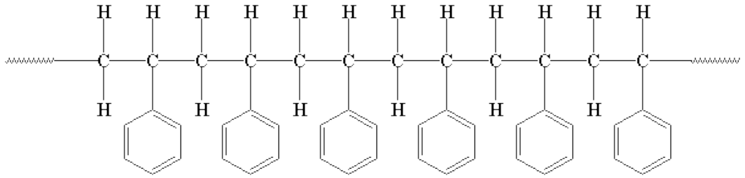
Diphenyl-bis(n,n-dimethylfluorene)silane

Secondary fluorophores



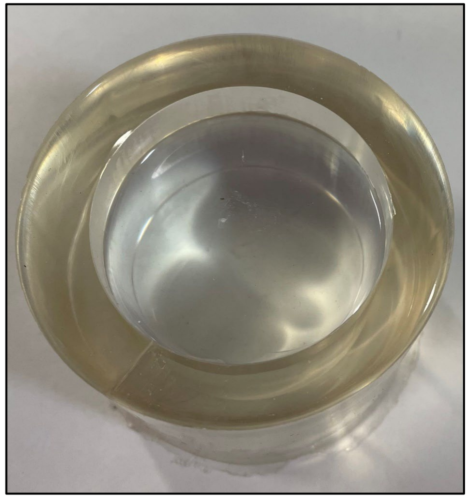
bis(methylstyryl)benzene

Polymer

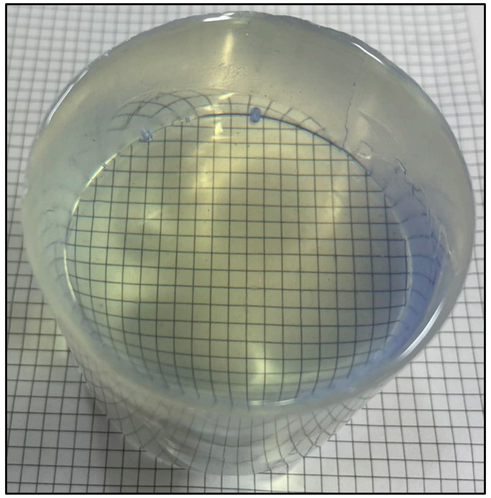


Polystyrene

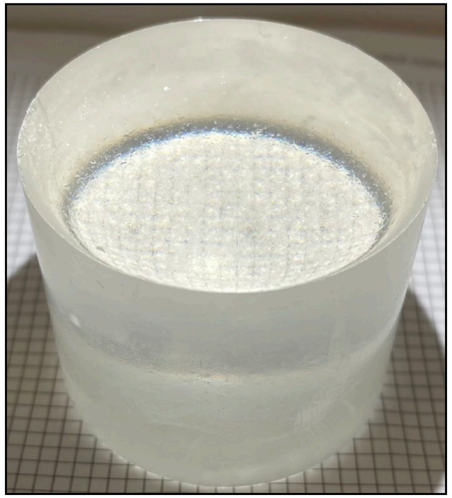
Polycarbonate doped OGS



AIB-6-101-B
1% PC, bis-MSB



AIB-6-116-2in
1% PC, BSBCz-Bu

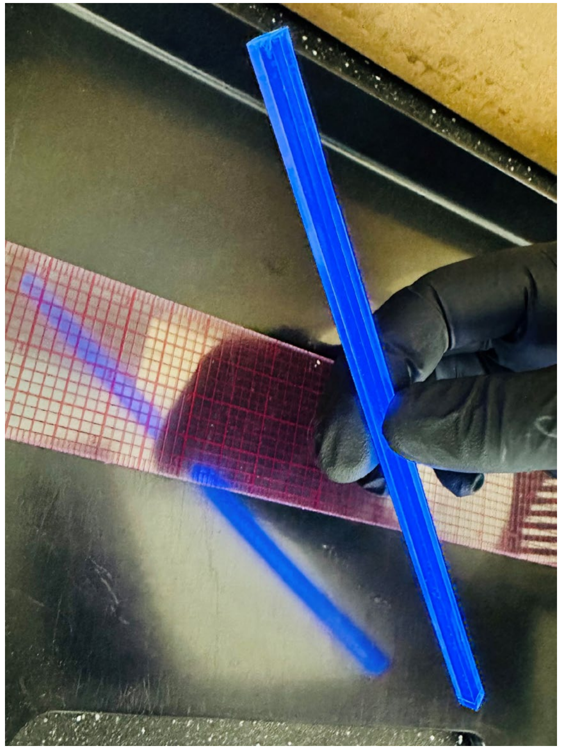


BSO-PC3-2in
3% PC, bis-MSB

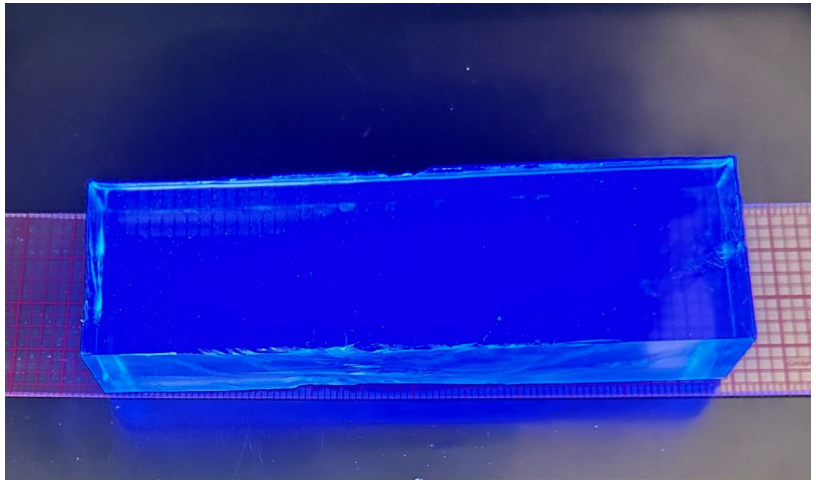
Sample	% PC	WLS	Rel LY EJ200	FOM
AIB-6-101-B	1	bis-MSB	1.40	2.31
AIB-6-116-2in	1	BSBCz-Bu	0.94	1.91
BSO-PC3-2in-PVA	3	bis-MSB	1.20	2.07

Large format Polymer OGS samples

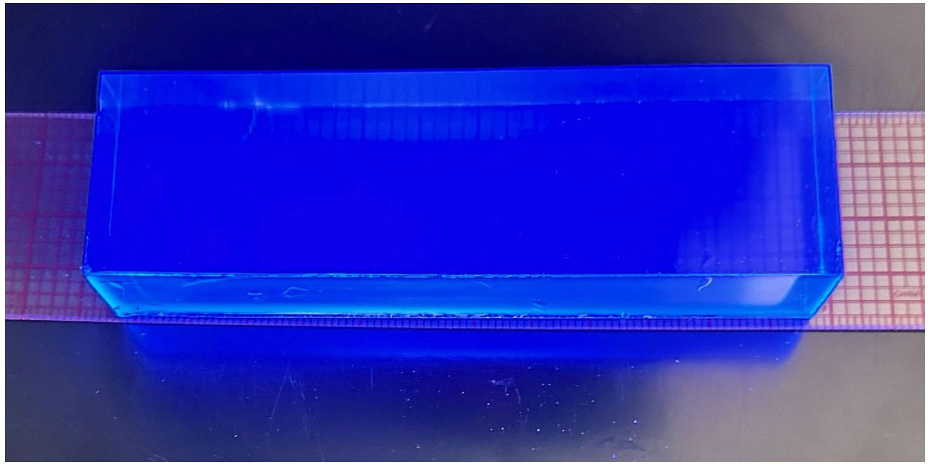
3 wt% PC in OGS,
20 cm x 0.5 cm x 0.5 cm



1 wt% PC in OGS,
15 cm x 4.5 cm x 2.5 cm



3 wt% PS in OGS,
15 cm x 4.5 cm x 2.5 cm



Measurements at MSU

Objectives

- Evaluate minimum measurable energy-loss signal (threshold)
- Evaluate pulse-shape discrimination capabilities
- Evaluate timing resolution
- Measure neutron efficiency
- Compare results with LENDA bar

Energy calibration and timing resolution

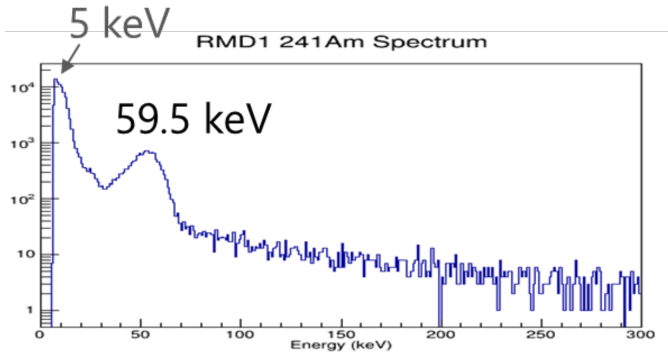
PSD and neutron efficiency measurements using two ELJEN liquid scintillators with the OGS samples

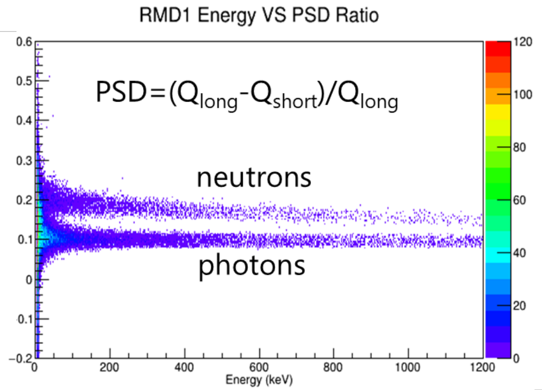
- DAQ used: NSCL/FRIB lab DAQ using two Pixie-16, 250 MHz digitizers
- ^{252}Cf fission source used for PSD and neutron efficiency measurements
- Various gamma sources used for detector characterization
- Well-developed ROOT-based analysis framework for LENDA detectors used*

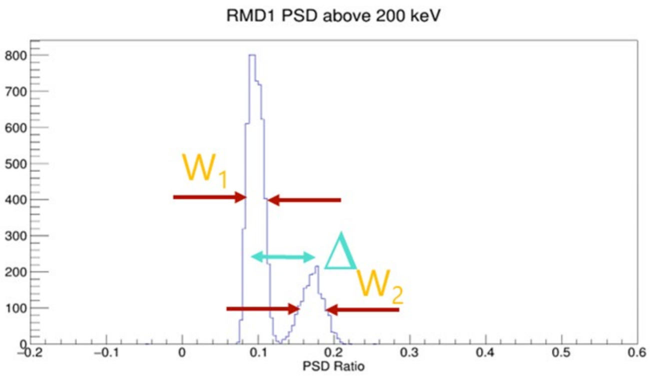
Ref: S. Lipschutz et al., NIMA Volume 815, 1 (2016)

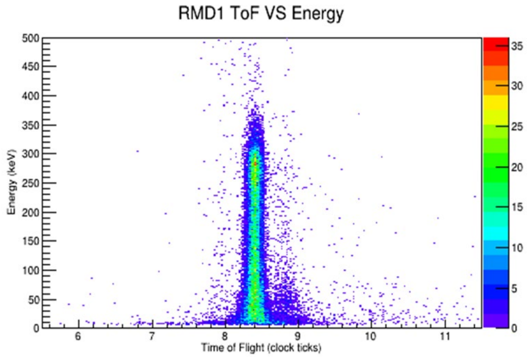
Composition selection in Phase-I

Detector	Comments	Width of ^{241}Am 59.6 keV line (FWHM; keVee)	Low Energy Threshold Limit (in keVee)	> 200 keVee	> 500 keVee	> threshold
RMD1	PC doped, 430 nm	17.59	~5 keV	1.22	1.51	.75
RMD2	PC doped, 530 nm	21.29	~20keV	.82	.88	.48
RMD3	Pure	14.22	~8 keV	1.08	1.31	.78
RMD4	Pure	13.69	~15 keV	1.23	1.27	N/A
RMD5	50% OGS in PVT	27.59	~28 keV	.62	.77	N/A
RMD6	Tin-loaded	19.59	~21 keV	1.14	1.185	N/A





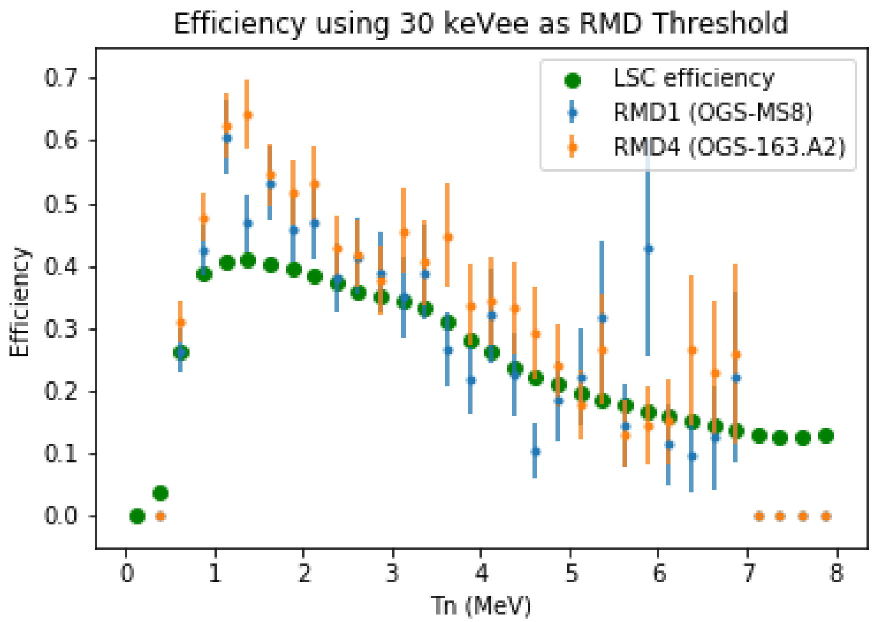


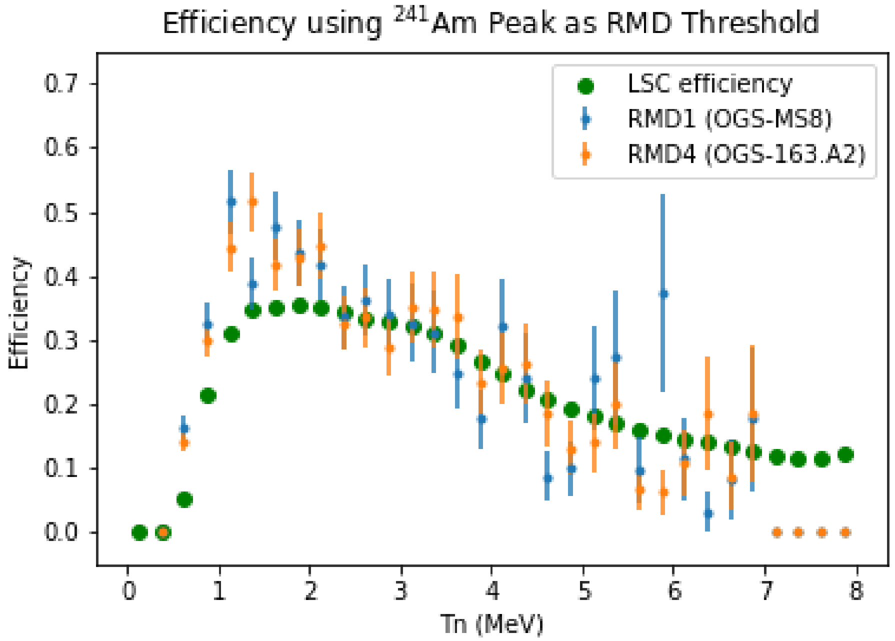


Neutron efficiency measurement

- Efficiency determined relative to that of an ELJEN liquid scintillator (LSC) with known efficiency and normalized for detector volume
- Efficiency of RMD samples calculated by taking

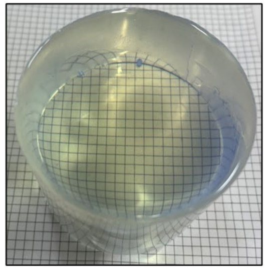
$$Eff_{RMD}(E) = Eff_{LSCI} * \frac{N_{RMD}(E)}{N_{LSCI}(E)}$$

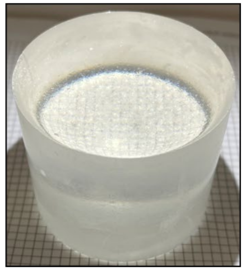




Measurements at MSU on 2” cylinder samples


AIB-6-101-B
1% PC, bis-MSB

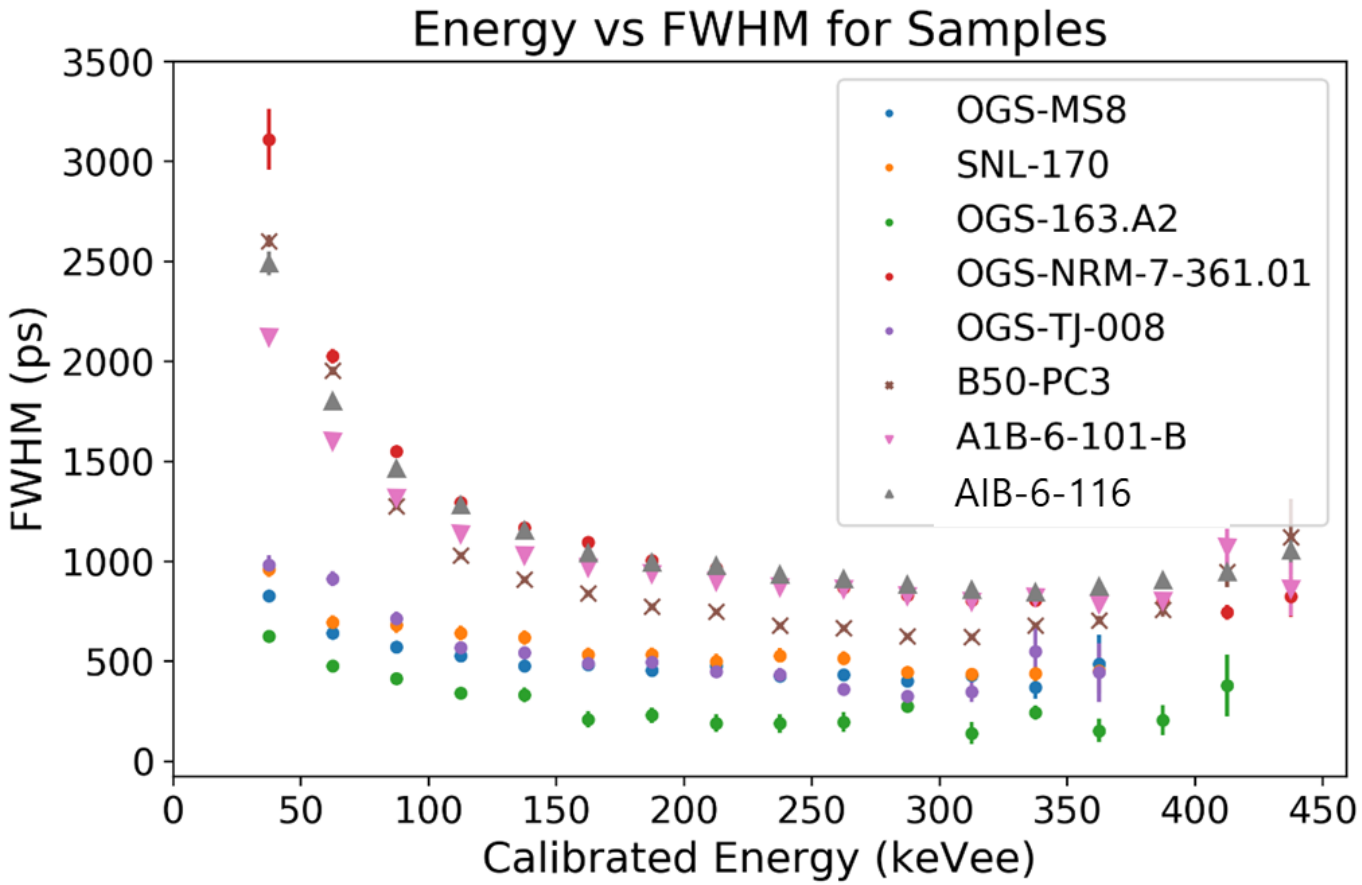

AIB-6-116-2in
1% PC, BSBCz-Bu


BSO-PC3-2in
3% PC, bis-MSB

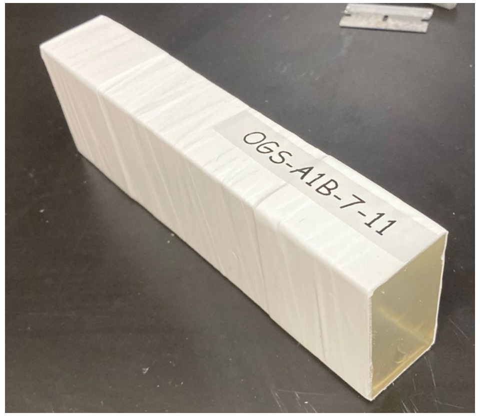
Detector	Sample number	Width of 59.6 keV line (FWHM; keV)	Low Threshold Limit (in keV)	22 Na First Compton Edge Channel #	PSD FOM > 200 keVee	PSD FOM: > 500 keVee
OGS-MS8	1	17.59	~5 keV	17924	1.22	1.51
OGS-3HF	2	21.29	~20keV	6837	.82	.88
SNL-170	3	14.22	~8 keV	16786	1.08	1.31
OGS-163.A2	4	13.69	~15 keV	17365	1.23	1.27
OGS-NRM-7-361.01	5	27.59	~28 keV	8872	.62	.77
OGS-TJ-008	6	19.59	~21 keV	9165	1.14	1.185
BS0-PC3	**7**	**18.4**	**~30 keV**	**8772**	**0.98**	**1.02**
AIB-6-101-B	**8**	**16.38**	**~25 keV**	**13712**	**1.12**	**1.11**
AIB-6-116	**9**	**16.693**	**~30 keV**	**9256**	**1.22**	**1.18**

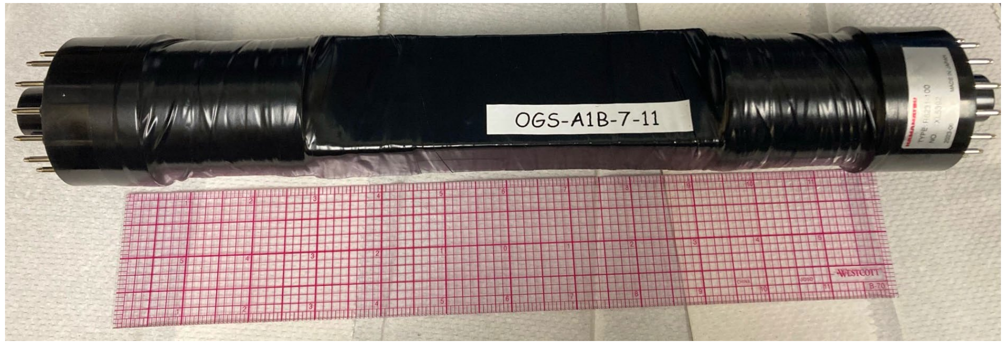
Timing resolution

- Timing measurement performed relative to timing signal from a LENDA bar
- Resolution determined as a function of energy - contribution of LENDA timing measurement subtracted

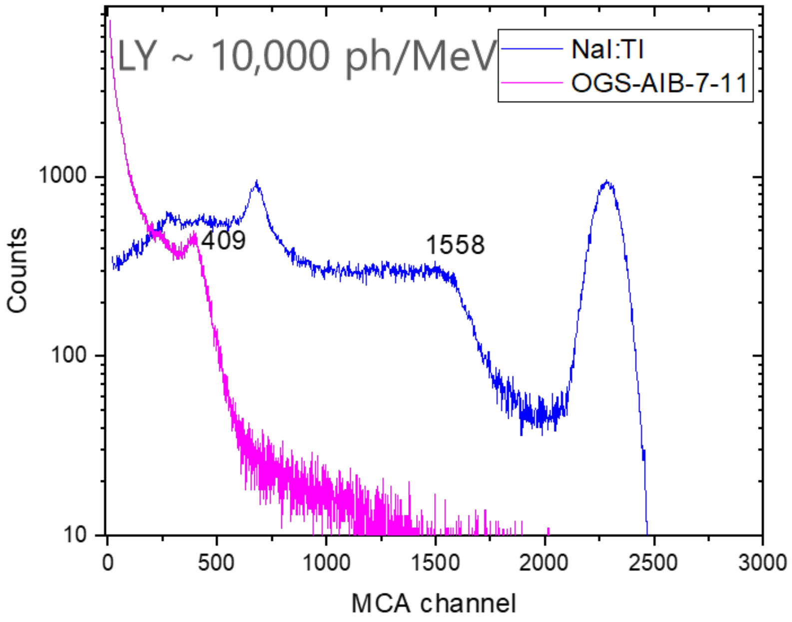


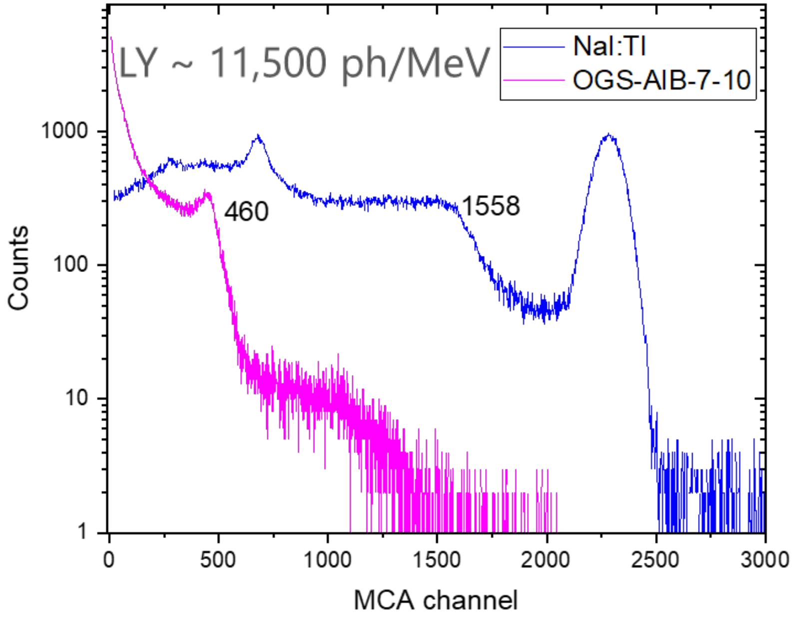
Measurement of half bars



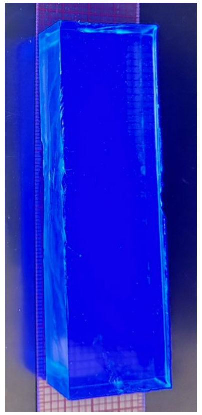


3% PS



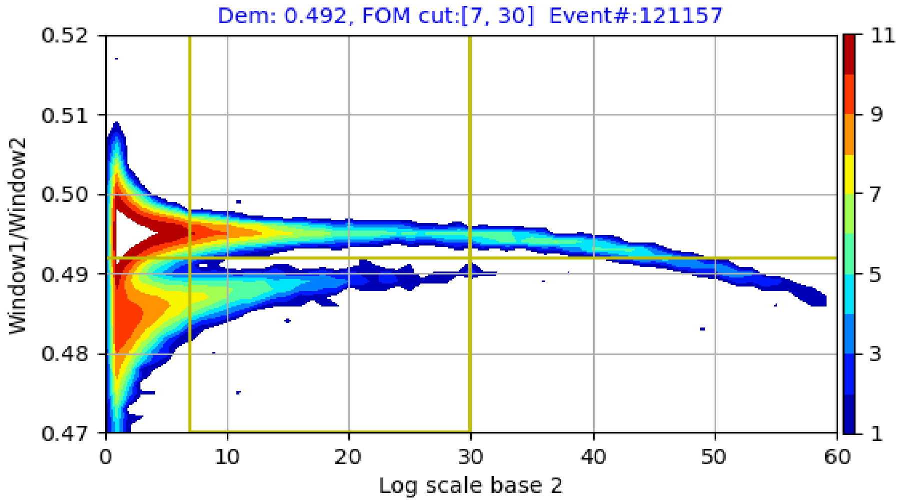


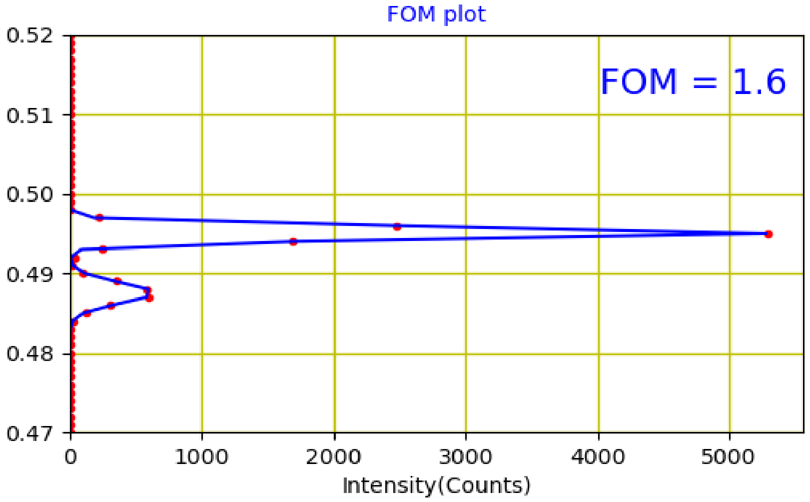
1% PC

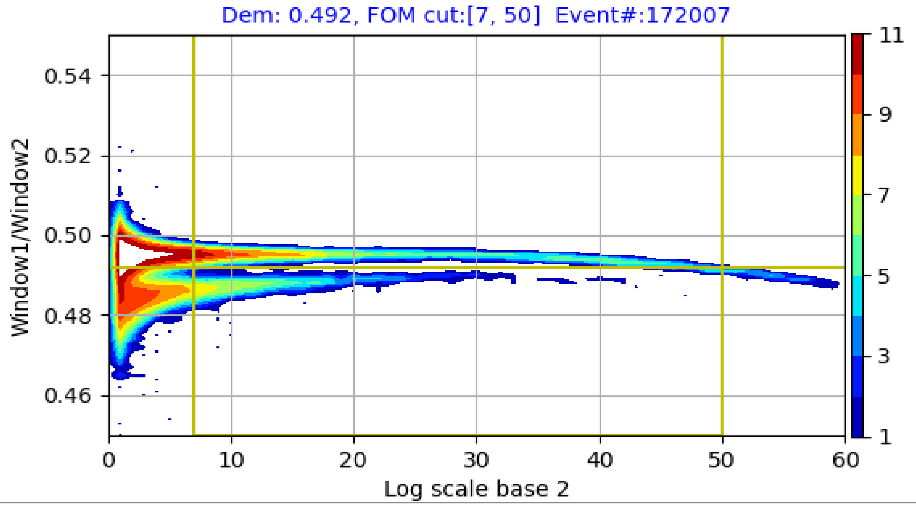


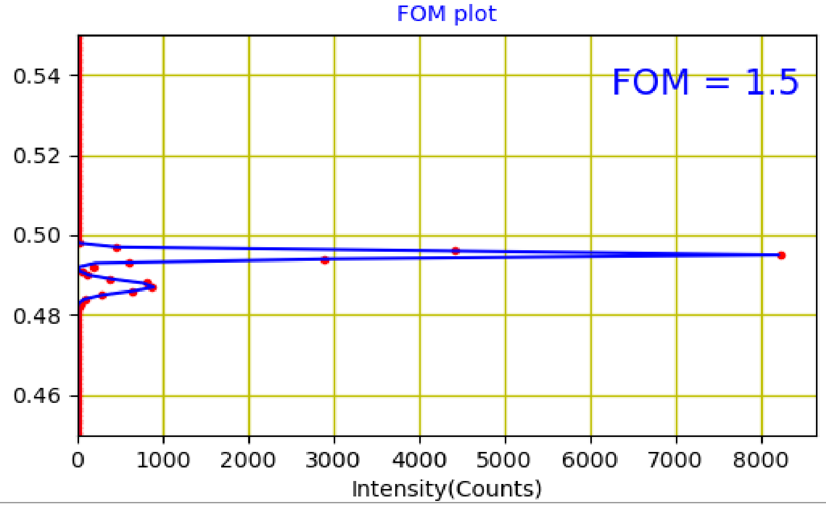
Measurement of half bars

3% PS

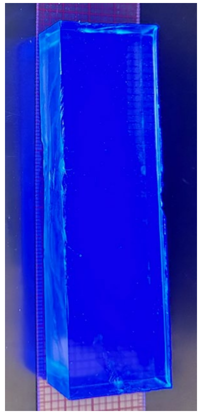








1% PC



Plans

- MSU to evaluate half bars
- Scale up of selected polymer OGS to 30 cm x 4.5 cm x 2.5 cm size
- Optimization of tin-loaded polymer OGS – use of vinyltoluene (aka methylstyrene)

Conclusions and Outlook

- LENDA is used for the detection of low-energy neutrons – options for upgrading the array with detectors that have pulse-shape discrimination for background reduction are being considered
- OGS composition has shown promise – scale up is possible due to polymer blending
- FRIB Charge-exchange group is characterizing half LENDA bar size PC-doped and PS-doped OGS samples are being evaluated at MSU
- Tin-loaded polymer OGS version is being developed

Acknowledgements

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