A low-background structural scintillator for rare event physics experiments

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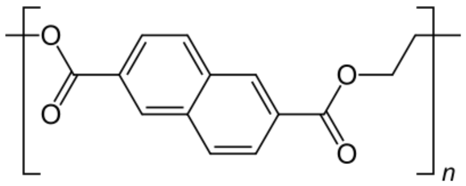
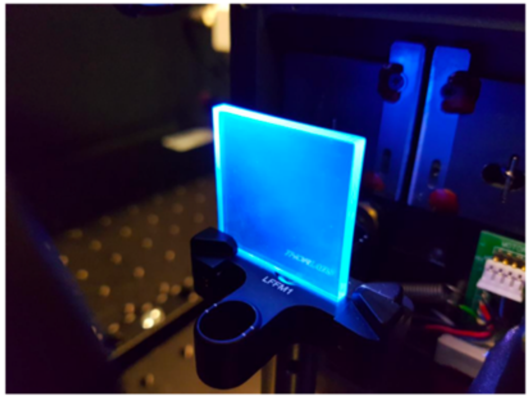
On behalf of the PEN working group

Motivation

- Active vetos are a crucial component of detection systems designed for rare event physics
  - Needed to reduce backgrounds to ultra-low levels required for dark matter, 0v2β, neutrino physics,…
  - Ideally, we'd like to limit the amount of *inactive* components near the sensitive detection volume
- Inactive materials
  - Structural components, cables and connectors, electronics, …
  - Typically electroformed copper, PTFE, …
- Can we replace some of these *inactive* components with an *active* component such as a scintillator?
  - Once possibly is recently discovered scintillator: poly(ethylene 2,6-naphthalate) (PEN)



Poly(ethylene 2,6-naphthalate) (PEN)



PEN excited with UV light

PEN working group

- 20+ active members
- 7 Institutions

Oak Ridge National Laboratory

Max-Planck-Institut für Physik

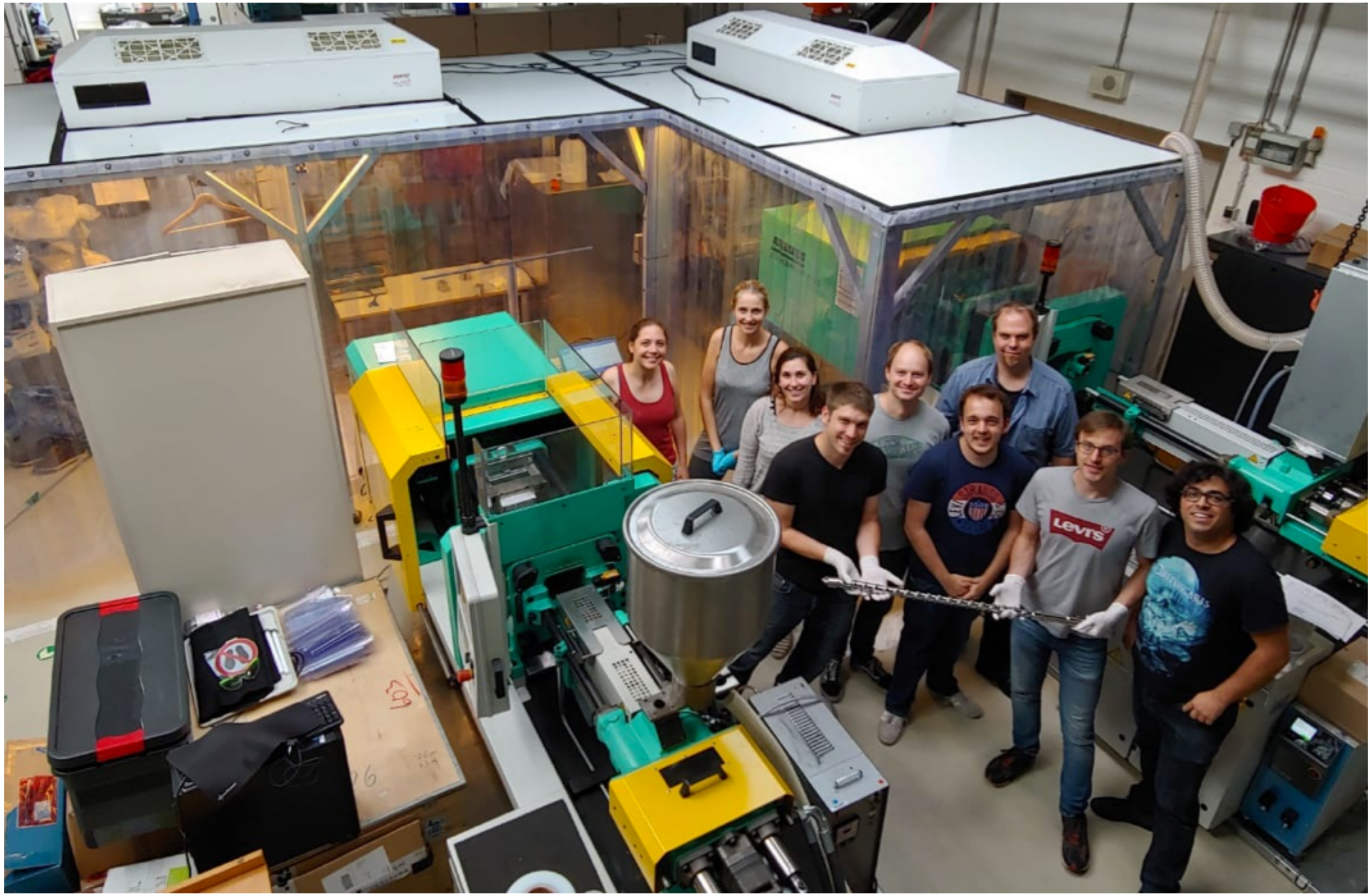
Lancaster University

Technische Universität München

CTU Czech Technical University in Prague

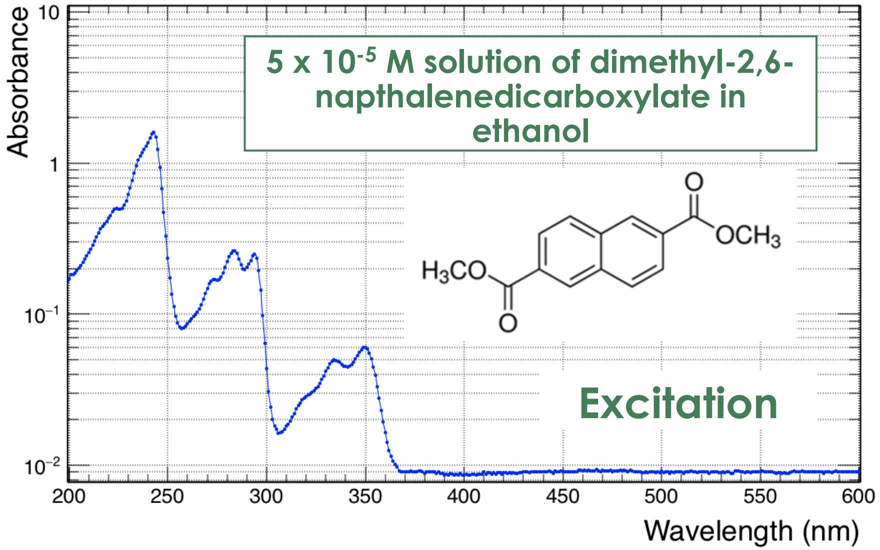
The University of Tennessee Knoxville

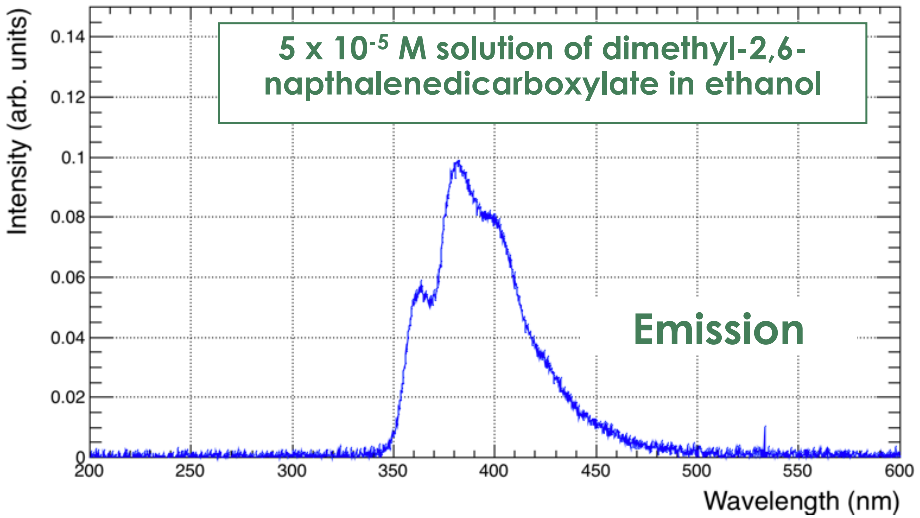
technische universität dortmund

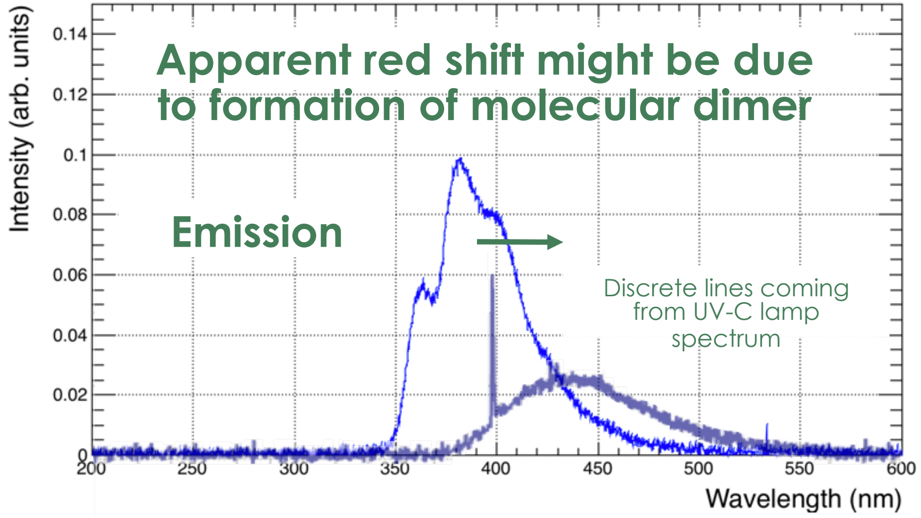


Luminescence of PEN

- PEN is a semi crystalline aromatic polyester composed of naphthalene repeat units
- PEN inherently scintillates with emission ~445 nm without addition of fluors
- Origin of scintillation likely due to a short-lived dimer state ?
  - Red-shift observed with increasing in concentration of naphthalene dicarboxylate molecules

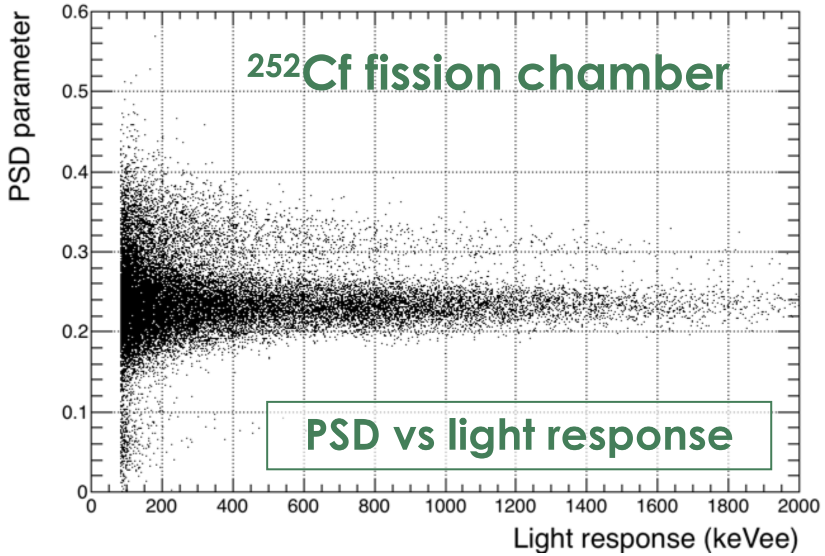


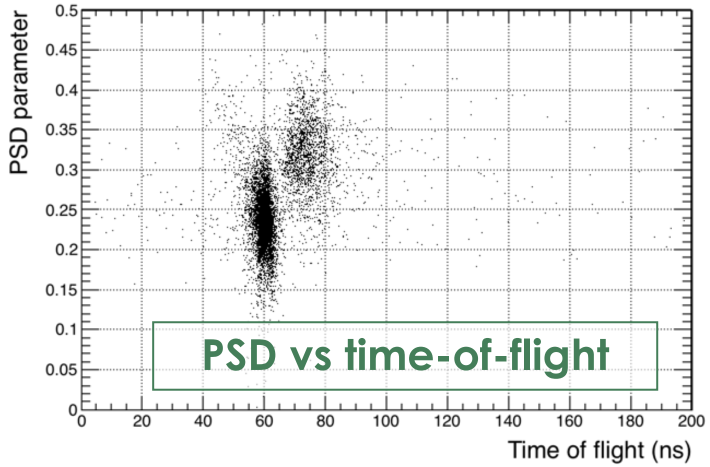


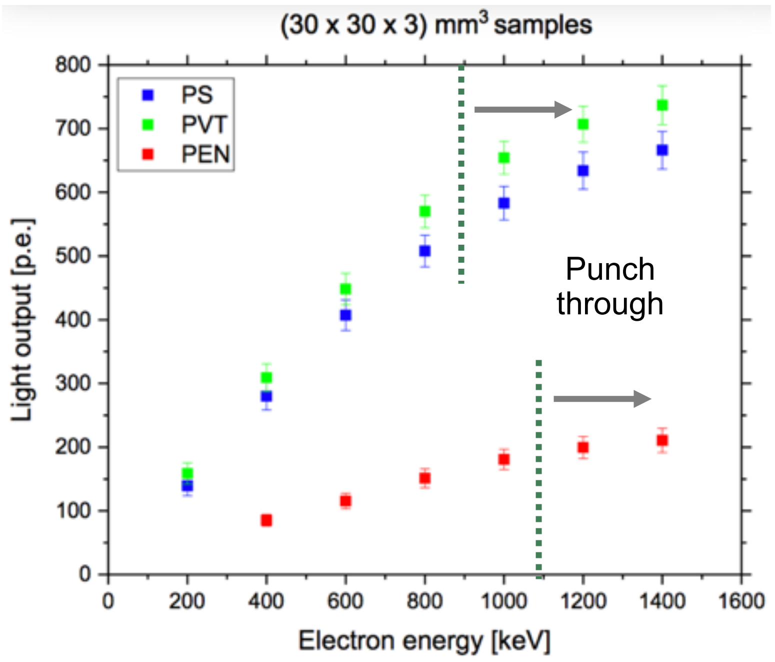


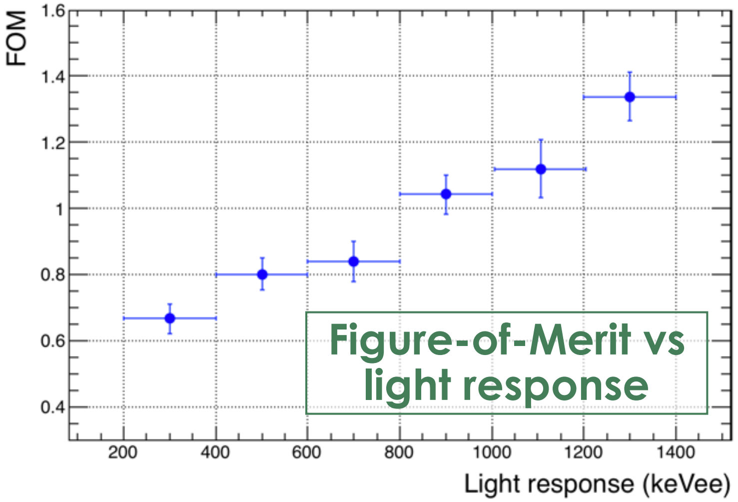
PEN scintillation & WLS properties

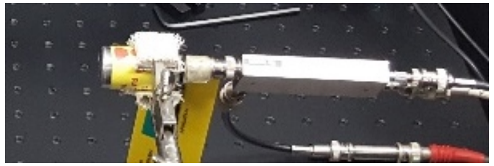
- Light yield ~1/3 of conventional plastic scintillators
  - Recall PEN has no fluors – limited by dimer decay?
- Particle identification using pulse shape discrimination (PSD) possible
- PEN is a wavelength shifter for LAr scintillation light (128 nm)







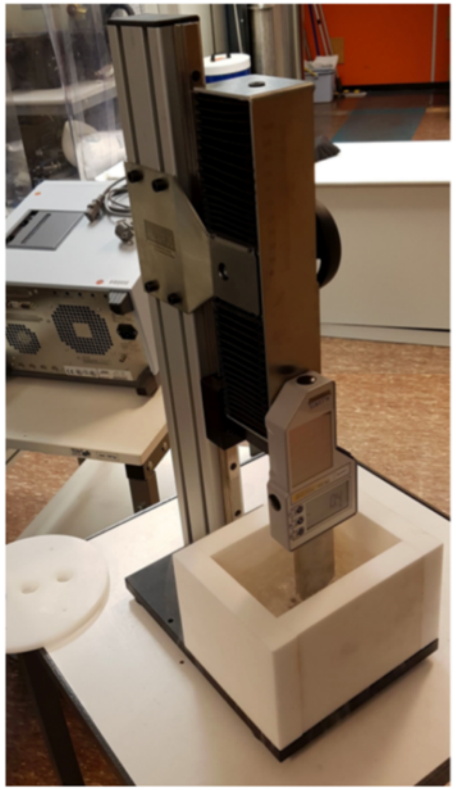


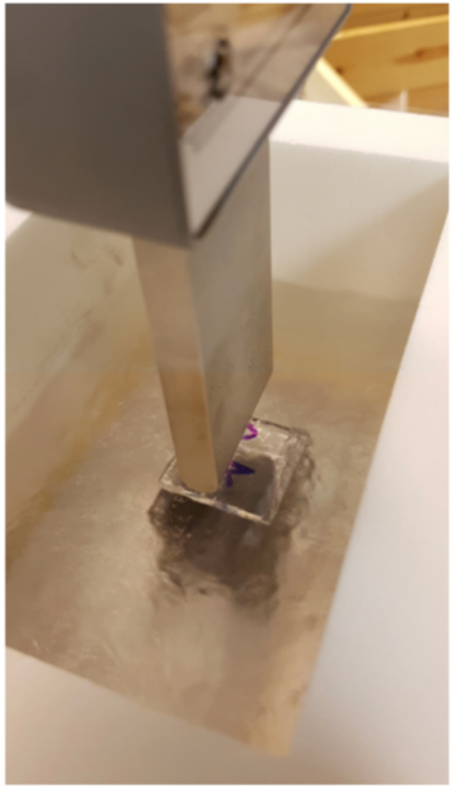


ORNL ^{252}Cf fission chamber

PEN mechanical properties

- Very chemically resistant to most acids and organic solvents
  - Can be aggressively cleaned
  - Requirement for low background experiments!
- 3-point bending test of material at room and LN_2 temperatures at MPI
  - High structural stability at room and cryogenic temperatures



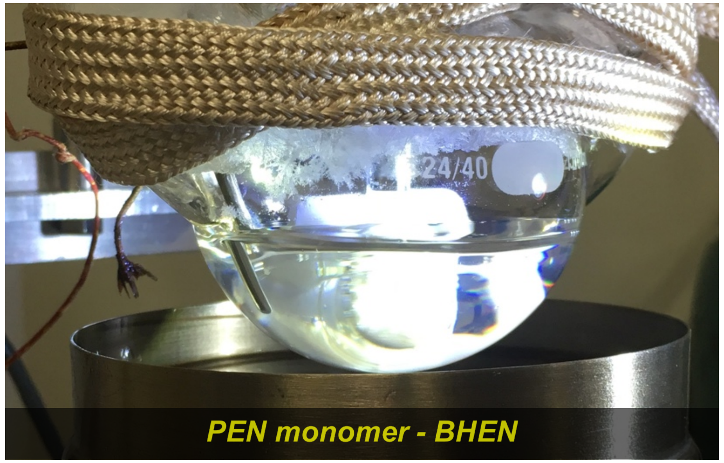


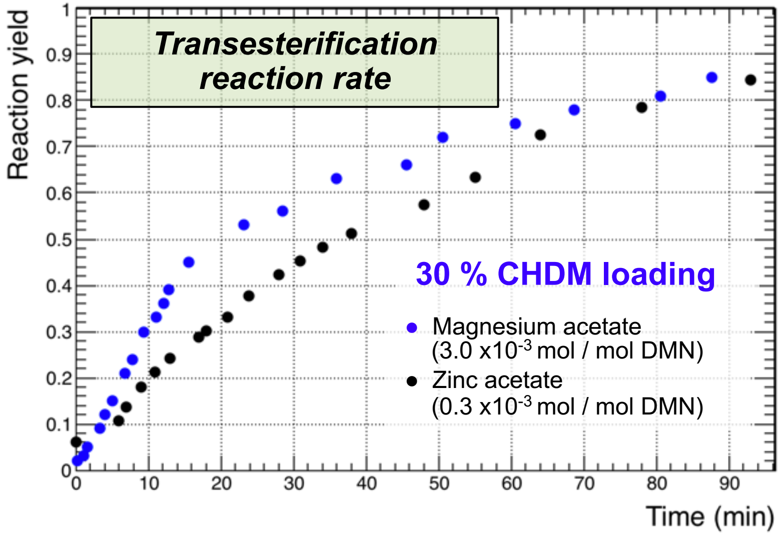
	PTFE[1]	Cu[2]	Electroformed Cu[5]	PEN	PEN at 77 K
Tensile Strength σ_{el} [MPa]	< 45.0	100	85.8 ± 7.8	108.6 ± 2.6	209 ± 2.8
Young's Modulus E [Gpa]	< 2.25	128	77.8 ± 15.6	1.86 ± 0.01	3.71 ± 0.08

1 https://www.treborintl.com/content/properties-molded-ptfe
2 http://www.memsnet.org/material/coppercubulk/
3 https://www.pnnl.gov/main/publications/external/technicalreports/PNNL − 21315.pdf

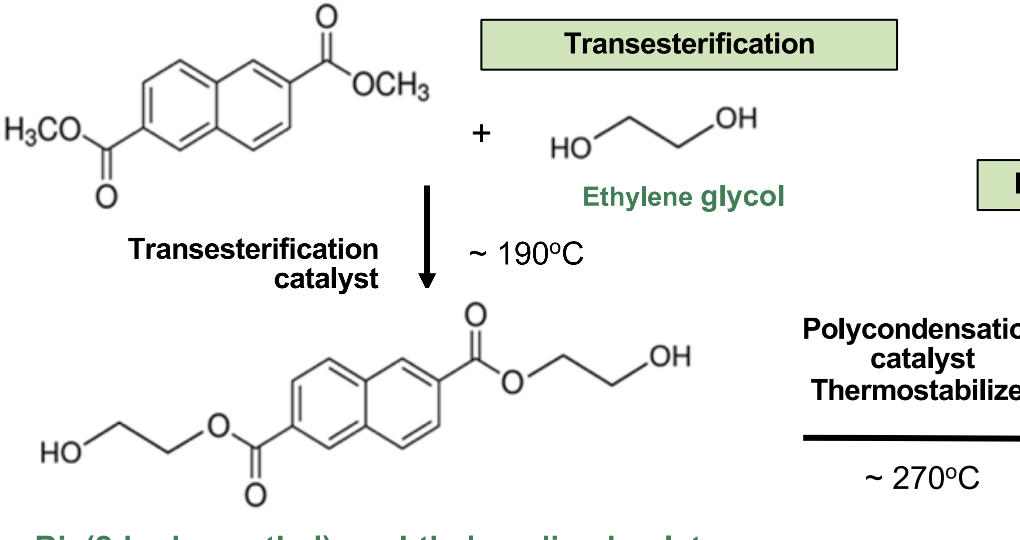
PEN synthesis at ORNL

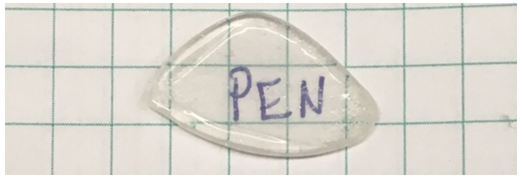
- *Can we make low-background scintillator grade PEN?*
- Synthesis efforts focused on low-background PEN derivatives
  - Higher light yield, reduction in radio impurities, improved optical properties
- Two-step synthesis method: *Transesterification → Polycondensation*

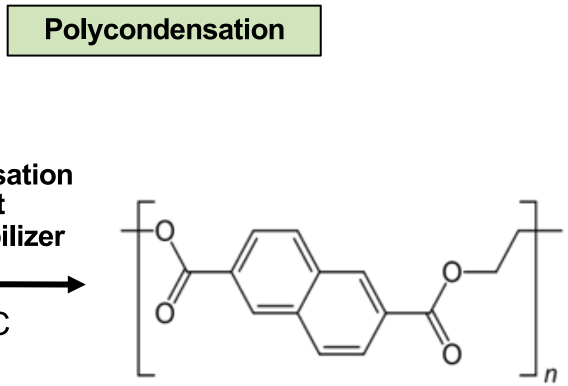




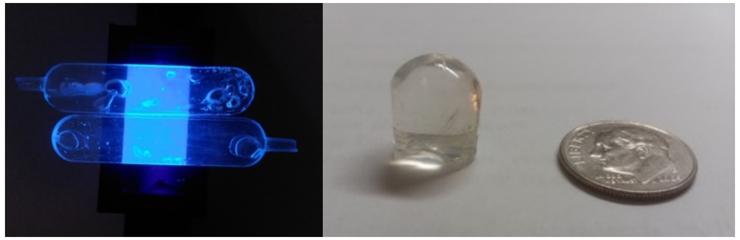
Dimethyl-2,6-naphthalenedicarboxlate







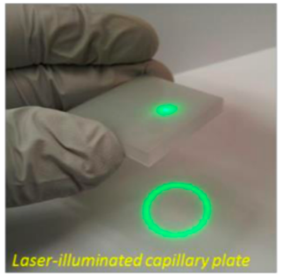
Bis(2-hydroxyethyl) naphthalenedicarboxlate



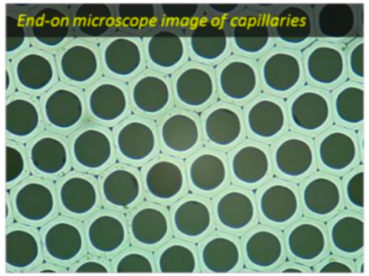
Scintillator Laboratory at ORNL

- Physics division's chemistry support laboratory is growing!
  - Synthesis, fabrication, and characterization of organic scintillator detectors
  - Experience with isotopically enriched scintillators
  - Organic synthesis setups, gloveboxes, Laminar flow hoods, chemical purification
  - Gas chromatography mass spectrometer (GCMS)

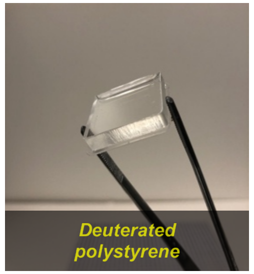
- Currently supports multiple projects
  - Low energy nuclear physics (FRIB)
  - Neutrinoless double beta decay (LEGEND)
  - Neutrino physics (COHERENT)
  - Applied nuclear science applications



Laser-illuminated capillary plate



End-on microscope image of capillaries



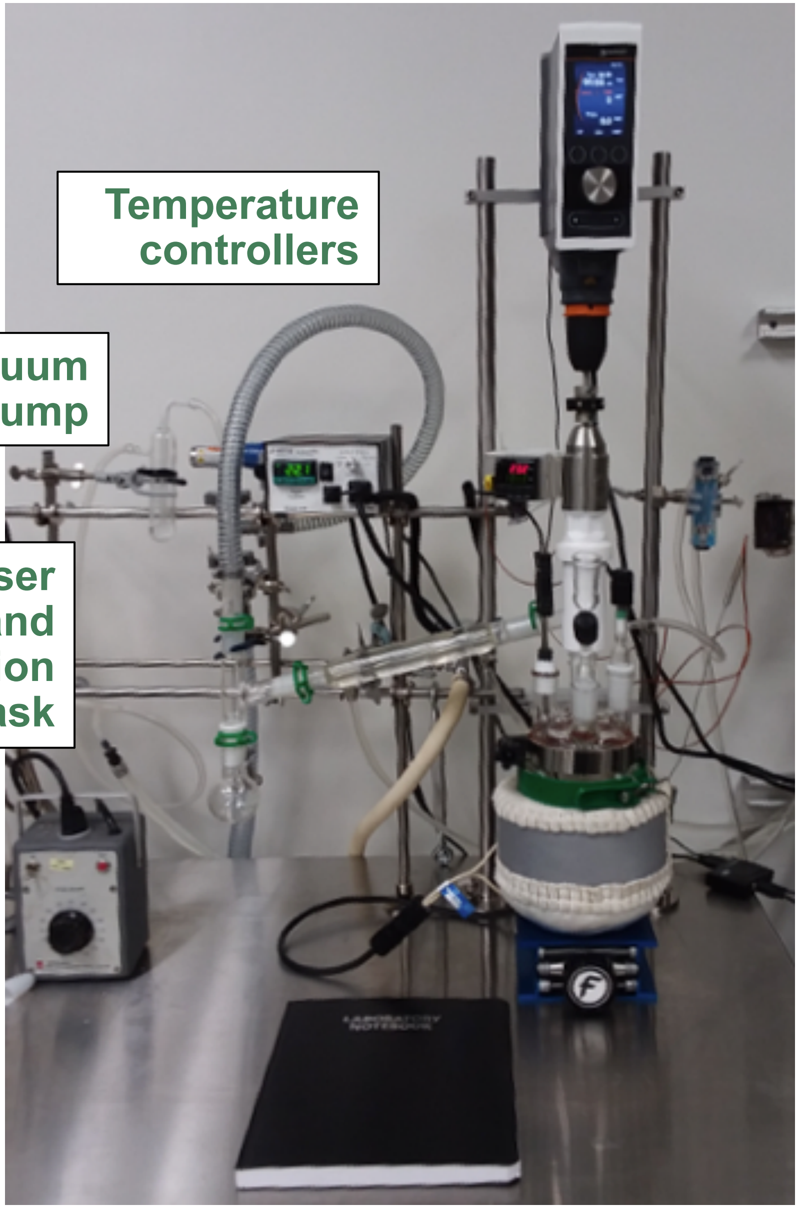
Deuterated polystyrene

Deuterated liquid scintillator

Reactor setup at ORNL

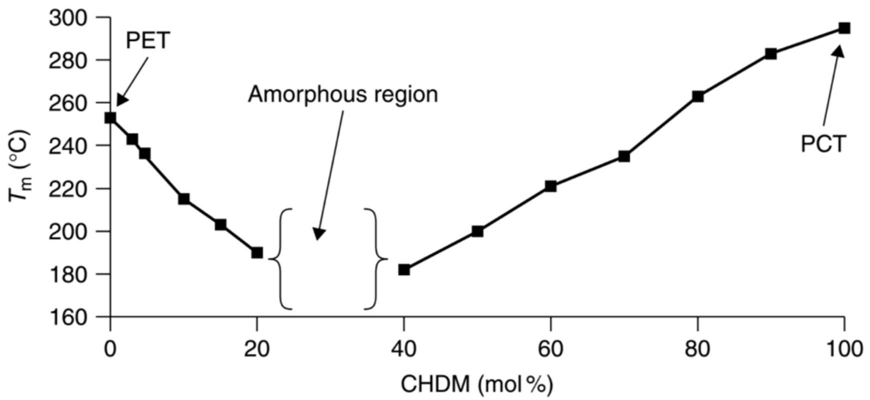
- 500 g batch reactor setup
  - Magnetically coupled-stirrer bearing → reduced oxygen contamination
  - Torque sensor for molecular weight monitoring
- Transesterification step is straight forward
- Challenge is the melt polycondensation, obtaining high MW, and reducing discoloration
  - GeO_2 used instead of Sb_2O_3 → radioclean catalyst!
  - Viscosity of the material increases with increasing molecular weight and hinders extraction of ethylene glycol needed for chain growth
  - Careful balance of catalysts, thermostabilizers, mixing, vacuum, and temperature

High viscosity stirrer

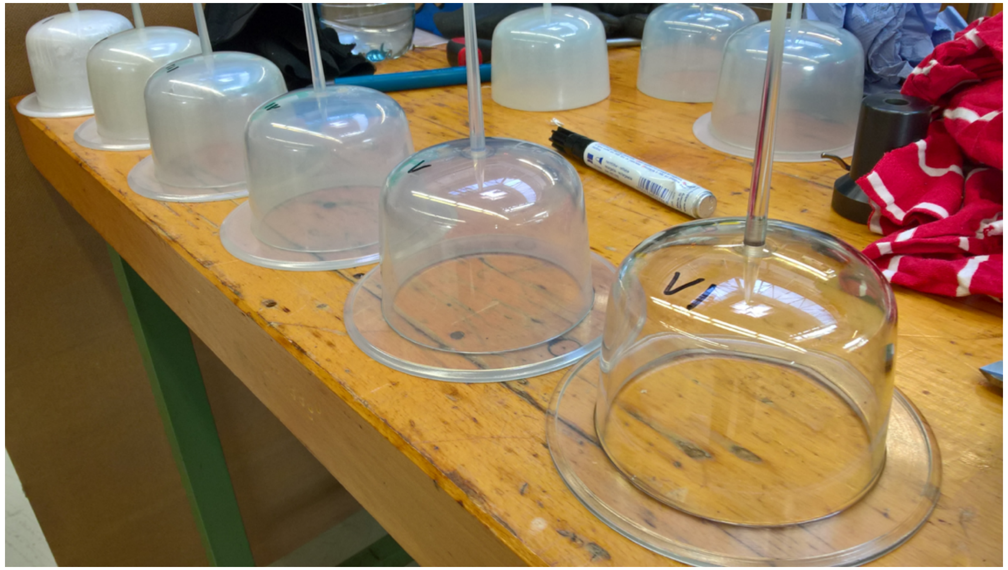


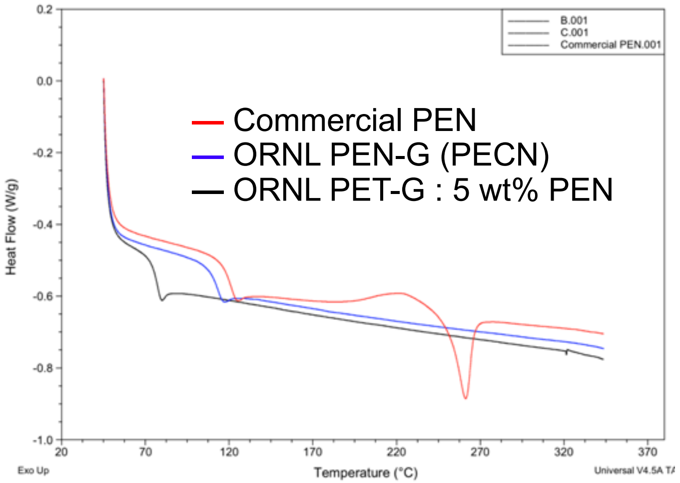
Transparency of PEN

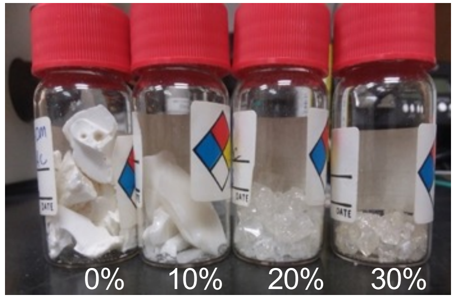
- Crystallization leads to scattering of light on boundaries
  - Polymer becomes opaque
  - Can be controlled using rapid cooling but not always possible for complex or large geometries
- Introduction of a copolymer can reduce crystallization
  - Demonstrated with PET
  - PETG or “glycol modified – PET”
  - Common copolymer is cyclohexanedimethanol or CHDM



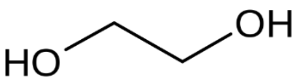
Modern Polyesters: Chemistry and Technology of Polyesters and Copolyesters. Edited by J. Scheirs and T. E. Long
 ISBN: 0-471-49856-4



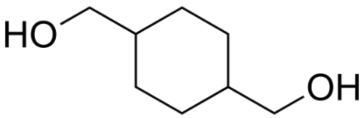




ORNL synthesized PEN - CHDM loading (mol %)



Ethylene glycol



1,4-Cyclohexane dimethanol (CHDM)

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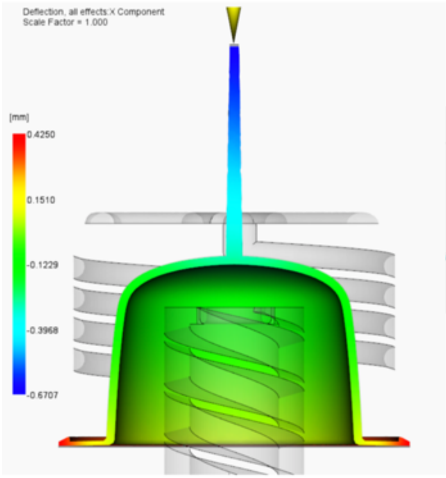
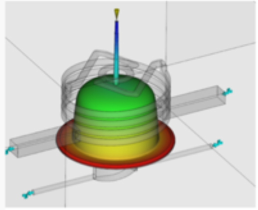
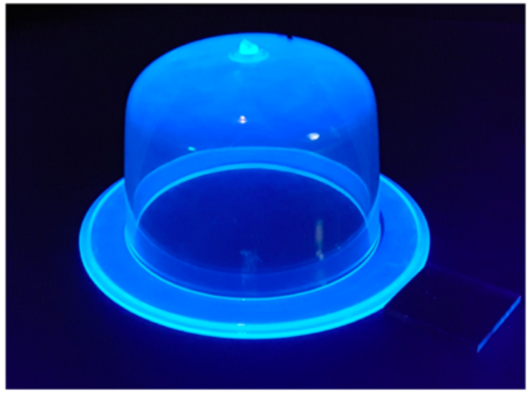
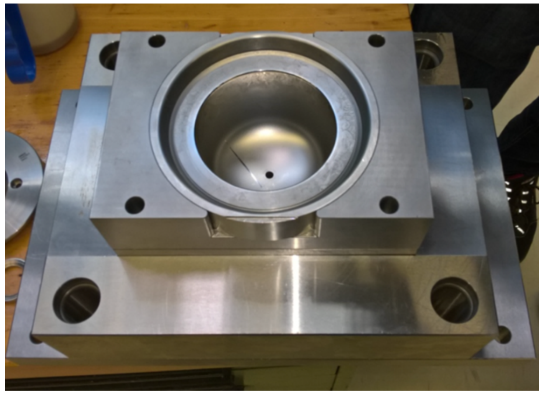
R&D on injection molding and bonding

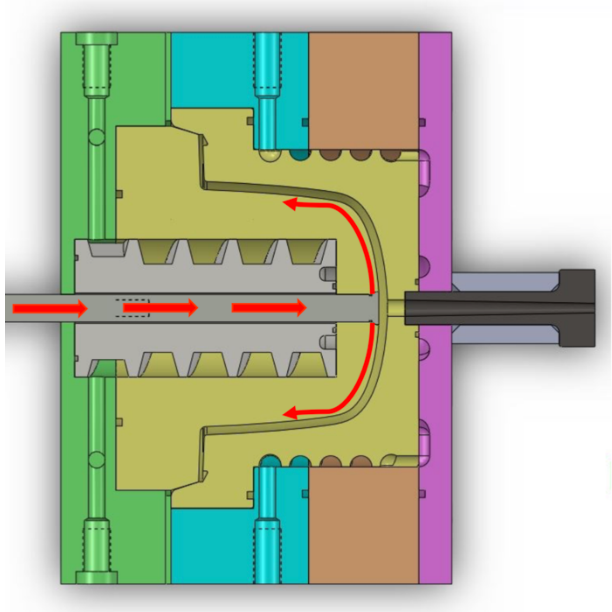
- Progress on producing arbitrary shapes
  - Plates / disks
  - Fibers
  - Capsules / containers
- Evaluation of radio-clean joining techniques
  - Ultrasonic welding
  - Low-background glues and adhesives











Can PEN components be made cleanly?

Mould

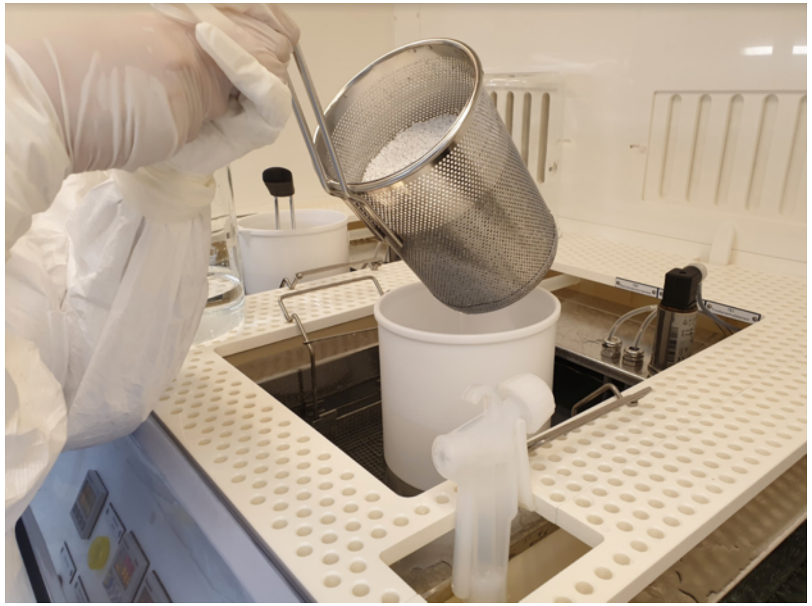
Material

Injection moulding machine

PEN tiles

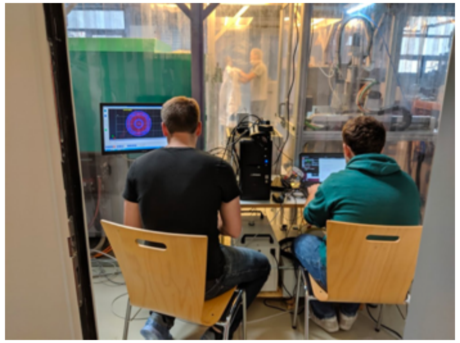
Radioclean production run

- First clean production run of PEN at TU-Dortmund
- Clean room ISO 6 (close to ISO 5)
- Use commercially available PEN granulate
  - First rinse with ultra-pure water
  - Ultrasonic bath with isopropanol
  - Final rinse with ultra-pure water and dried with boiloff nitrogen
- All parts which came in contact with PEN were new and etched in nitric acid or cleaned with micro-90
  - Injector assembly was completely rebuilt
  - New screw, injector nozzle, dosing hopper cleaned with micro-90 and ultraclean water
  - New mold plates which were acid etched
- Entire process from granulate to finish product performed in less than ***4 mins per part***
  - Injection compression molding
  - Optical characterization
  - CNC machining
  - Photography
  - Bagging and documentation

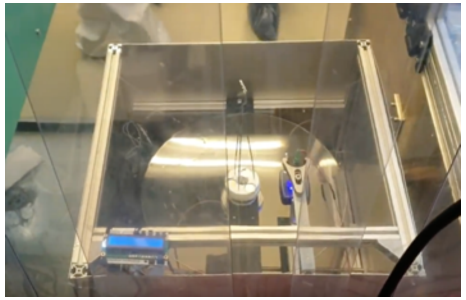


Overview of cleanroom layout

Control station / QA / Overseer

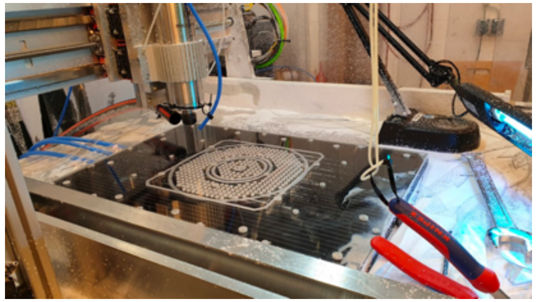


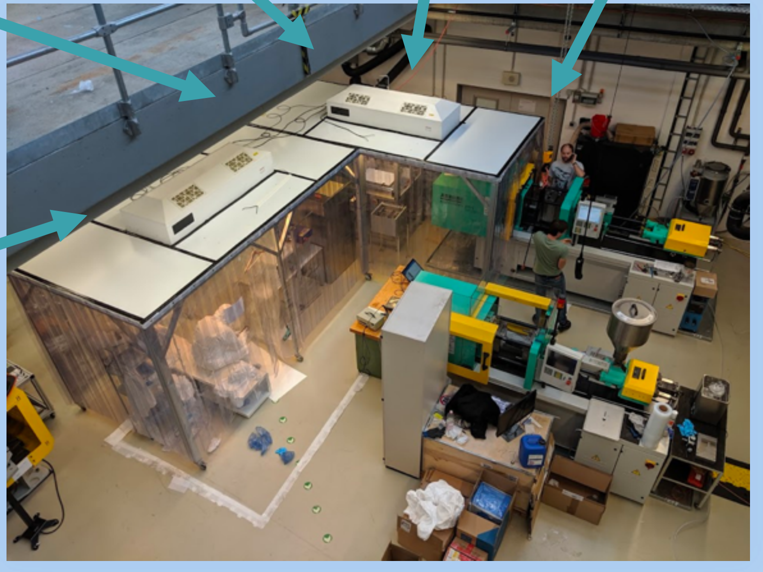
Optical scanner



Injection/compression molding

CNC machining

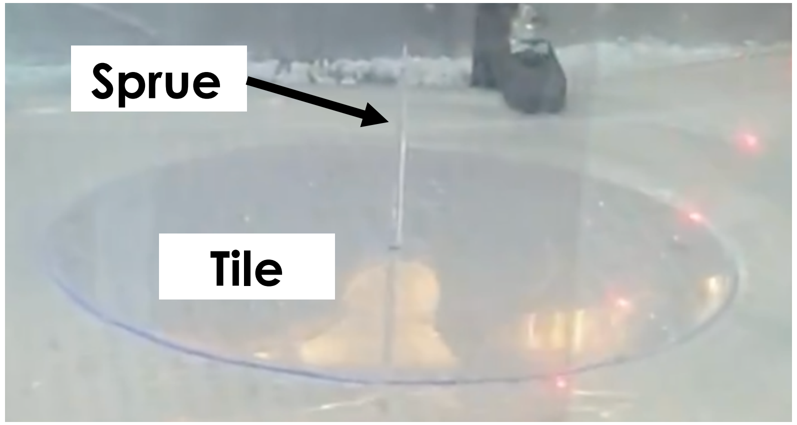


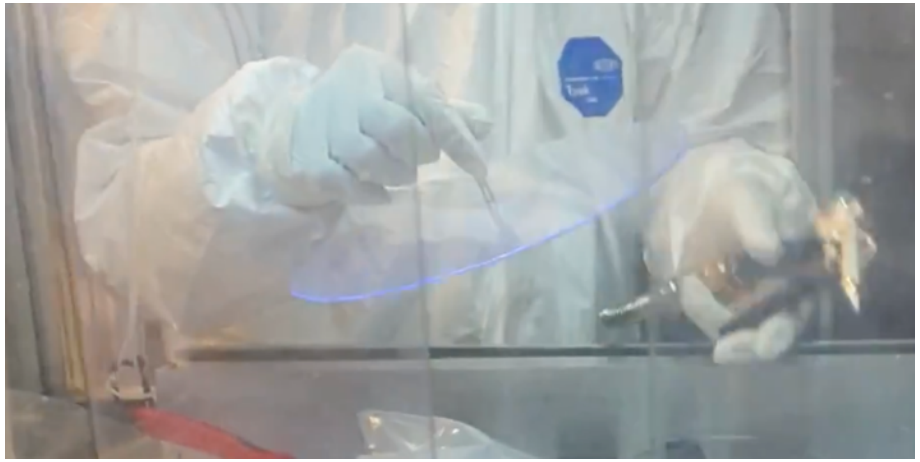


Photography, bagging, and labeling

Handling procedure

- Tile surface never handled directly with gloves or touched
- Operations performed using foot switches or from control station
- Pre-machining: tiles handled by sprue which is removed just before machining
- Post-machining: tiles handled using acid-etched stainless steel tongs using central hole
- Contact surfaces include: magnetic optical scanner stage, vacuum chuck for CNC, 3-point fixture for photography





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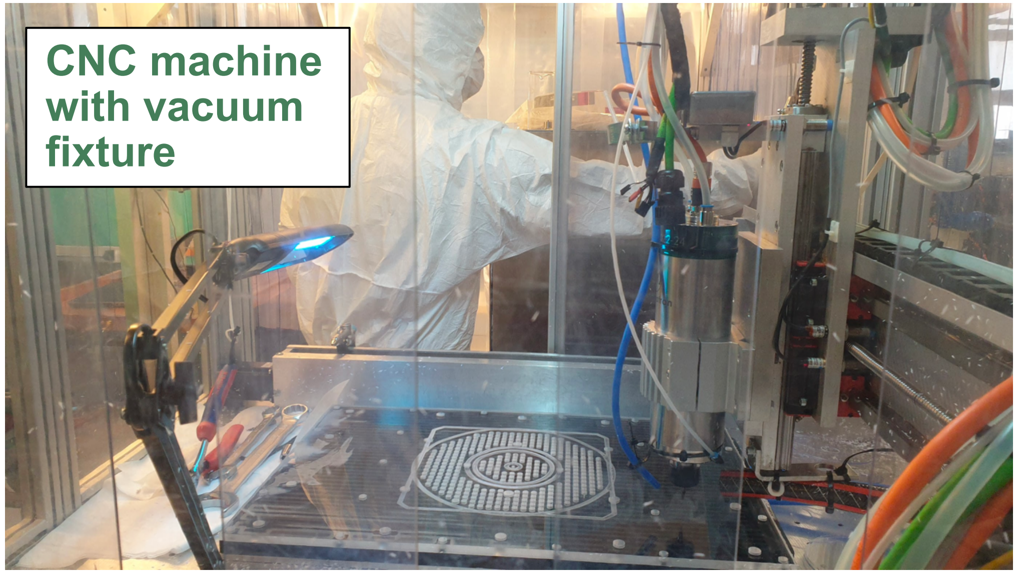
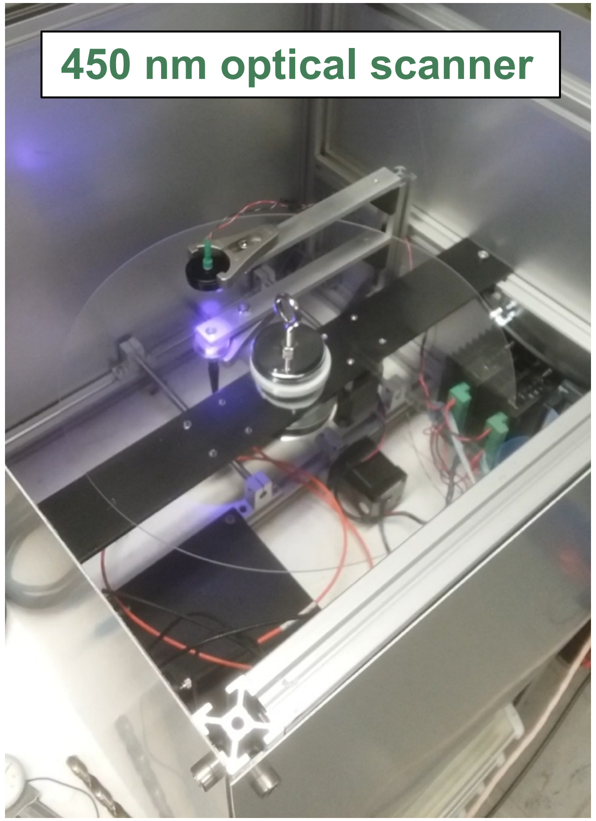
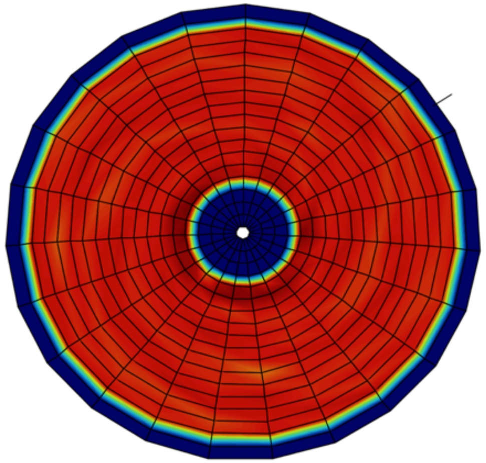
Low-background Production run

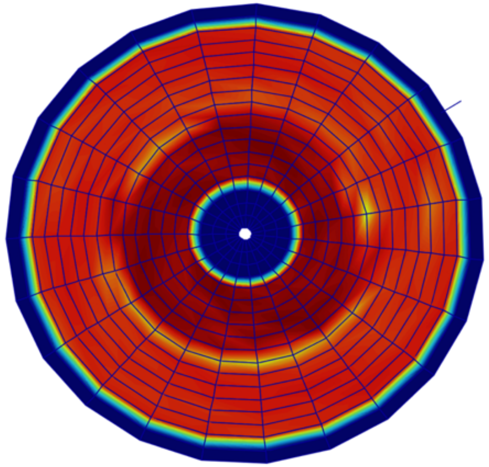
- Total of 291 tiles were produced
- Optical transmission scans at 450 nm
  - Well-match with PEN emission
- 242 tiles sent out for radioassay
  - 112 Obelix
  - 130 GeMPI







Optical scan – Accepted tile



Optical scan – Deflective tile

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Radiopurity – Intermediate results

- Intermediate results from LNGS
- Radioassay measurements from M. Laubenstein
- Upper limits with k=1.645, uncertainties are given with k=1 (approx. 68% CL)
- Tiles are still being counted

Radio assay of PEN tiles from production run			
Weight: 14.3 kg (131 tiles)			Preliminary
Live time: 43 days			
		μBq/kg	g/g
Th-232	Ra-228	80 ± 30	$19 \pm 7 \times 10^{-11}$
	Th-228	< 46	$< 1.1 \pm 7 \times 10^{-11}$
U-238	Ra-226	80 ± 20	$6 \pm 2 \times 10^{-12}$
	Th-234	< 2400	$< 1.9 \times 10^{-10}$
	Pa-234m	< 1500	$< 1.2 \times 10^{-10}$
	U-235	< 62	$< 1.1 \times 10^{-10}$
	K-40	< 230	$< 7.3 \times 10^{-9}$
	Cs-137	< 19	

Conclusion

- The polyester PEN has been demonstrated as a possible structural active veto scintillator
- Exhibits desirable mechanical properties at room and cryogenic temperatures
  - Good chemical resistance → Can be aggressively clean!
- Fluorescence observed at ~445 nm
  - Well-match with SiPM / PMT
  - Particle discrimination possible
  - Light output ~1/3 of conventional plastic scintillators
- New amorphous PECN (PEN-G) formulations produced at ORNL exhibit enhanced optical clarity during processing of complex or thick geometries
- Low-background PEN components can be prepared for rare-event physics experiments