Lithium-Doped PSD-Capable Liquid Scintillator for the PROSPECT Experiment

Pieter Mumm, Denis Bergeron, Svetlana Nour, Jerry LaRosa, and Mark Tyra
National Institute of Standards and Technology

For the PROSPECT Collaboration

PROSPECT Motivation: multiple experimental anomalies

Overall reactor neutrino flux deficit

Possibility of sterile neutrino oscillation as an explanation of observed electron antineutrino deficits

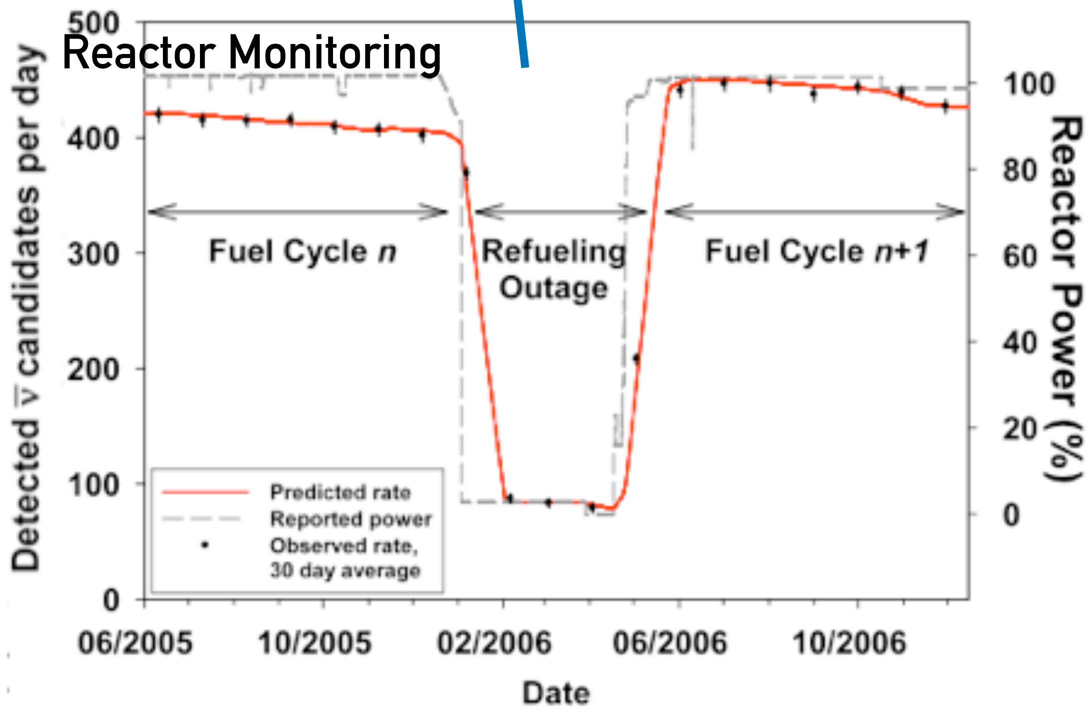
Reactor spectrum spectral features and Reactor fuel evolution not as predicted

Anomalies in spectral shape at ~ 5-6 MeV
Provide complementary measurement of pure ^{235}U

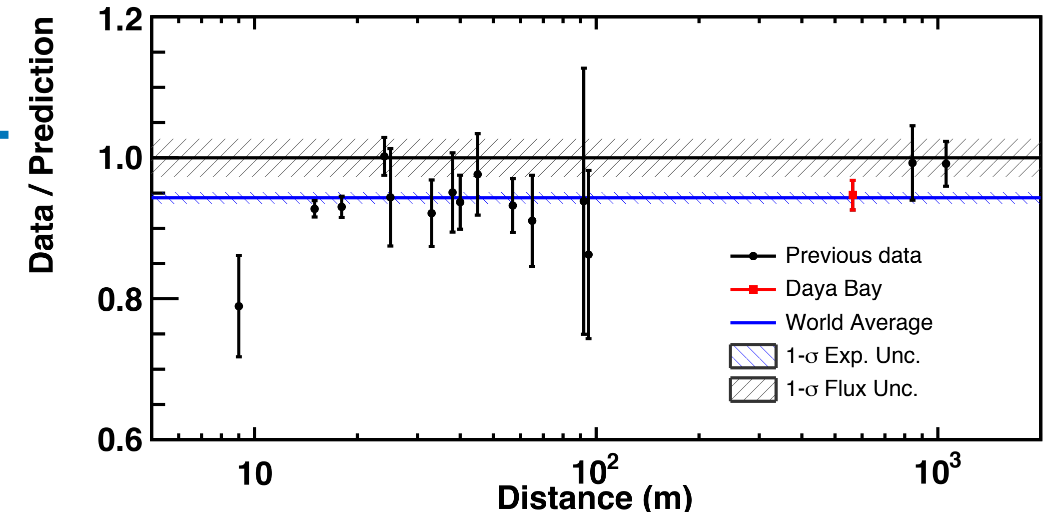
Safeguards - Passive Standoff Capability

Provides a remote, non-intrusive reactor power and Pu production monitoring

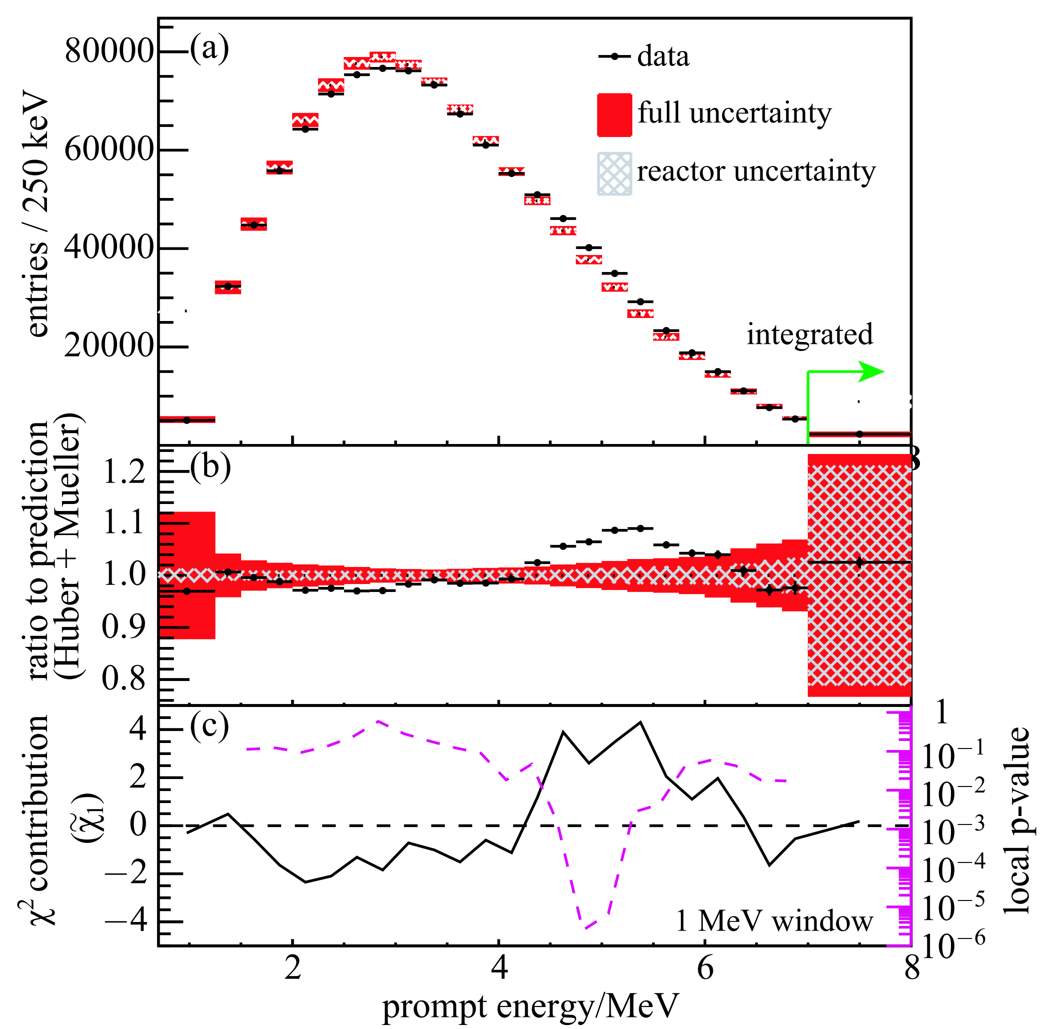
Reactor Monitoring



Flux Deficit



Spectral Anomaly

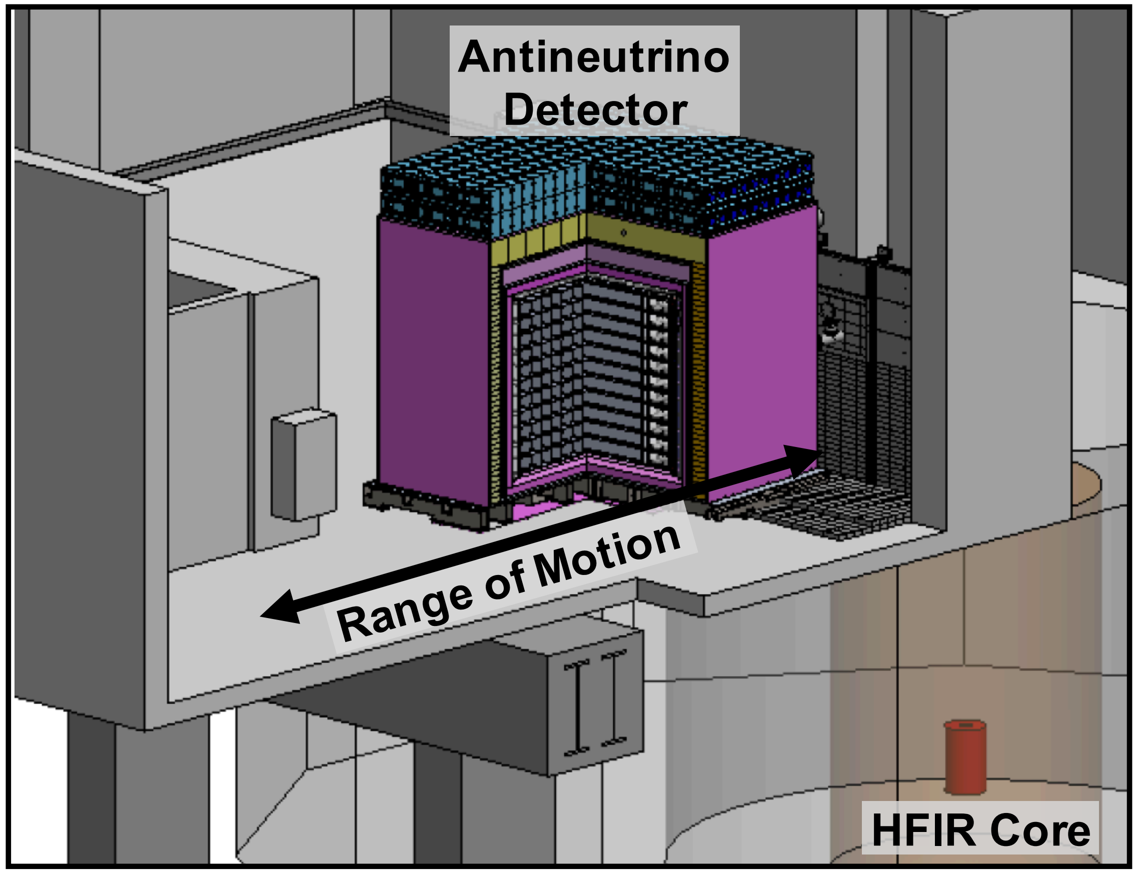


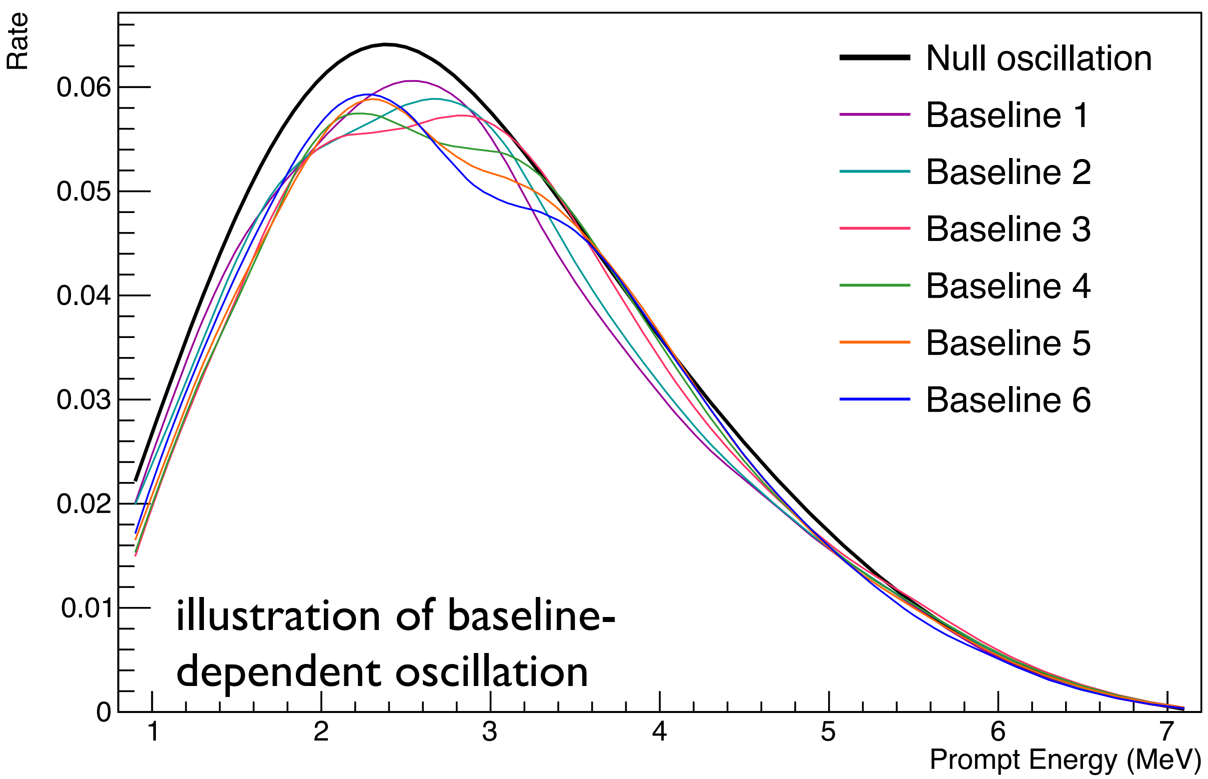
Experimental Goals and Strategy

1. Search for short-baseline sterile-neutrino oscillations independent of reactor models
2. Measure antineutrino spectrum due to ^{235}U
3. Demonstrate near-field surface operation

Experimental Strategy:

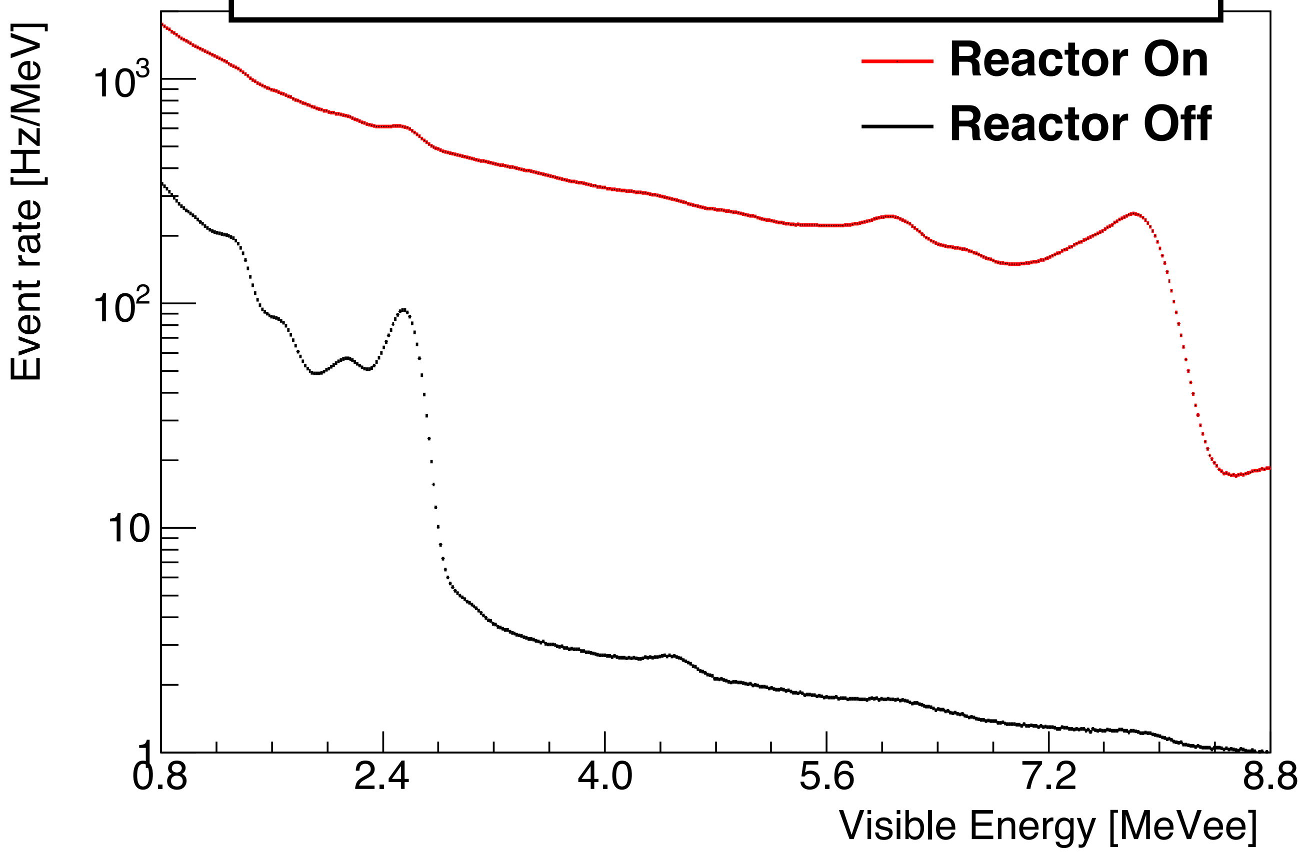
- Compact HEU reactor (HFIR at ORNL)
- ^{6}Li-doped liquid scintillator provides unique compact tag and light yield
- Segmented detector localizes events and supports background rejection
- Measure high-resolution spectra at range of baselines (currently 6.5-9m)
- Search for characteristic relative spectral distortions within detector volume



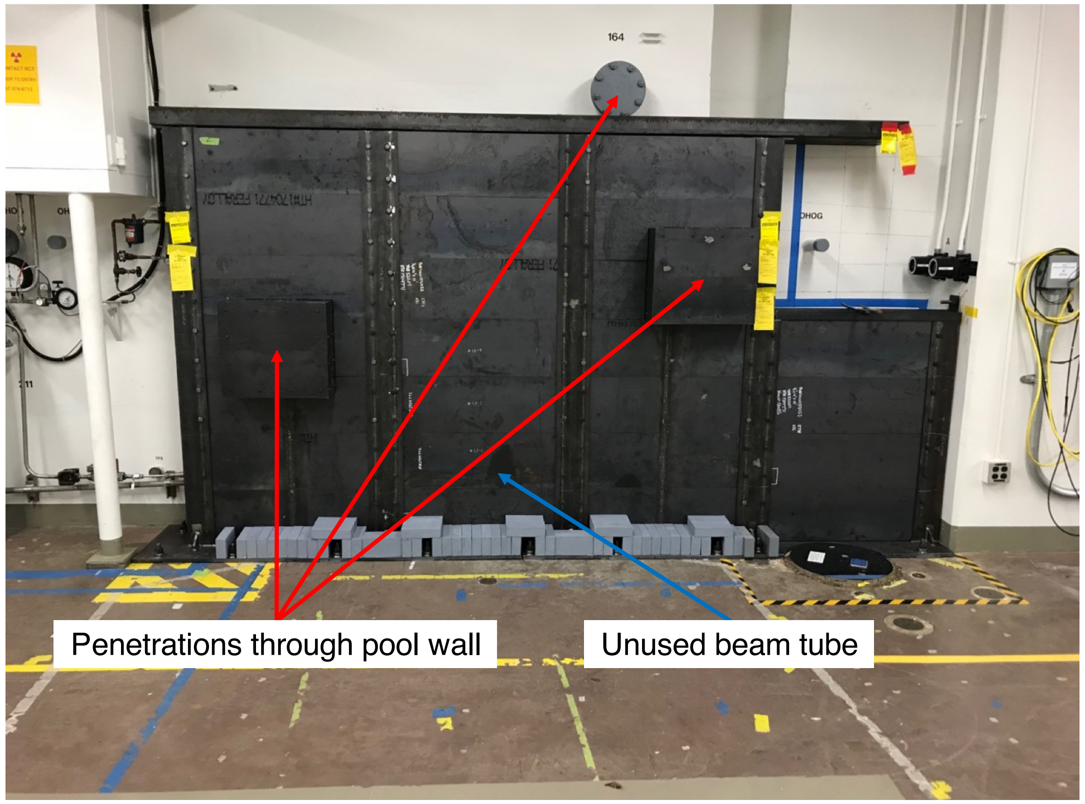


Ambient Backgrounds

Electron-like event rate in detector



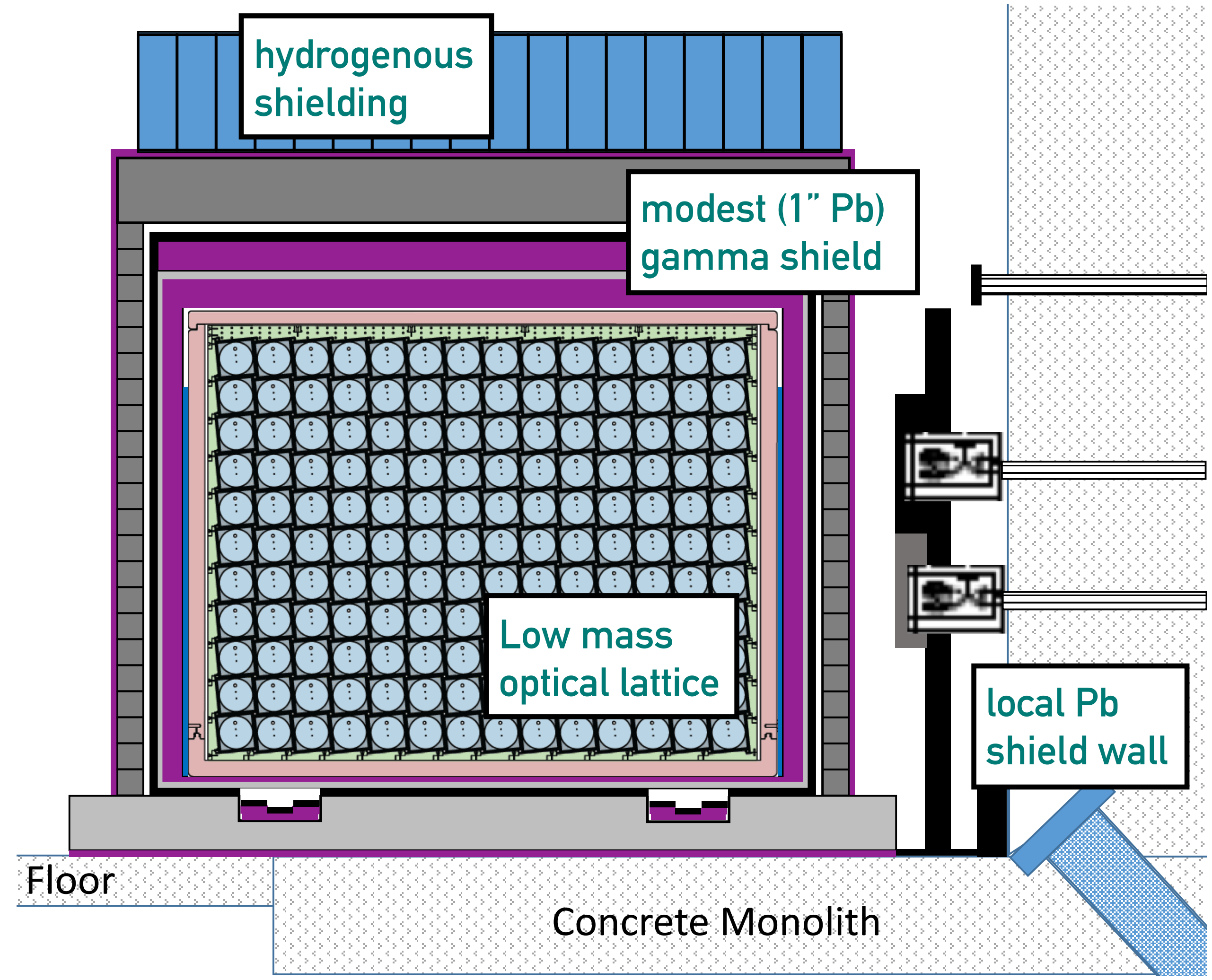
Local shield against pool wall

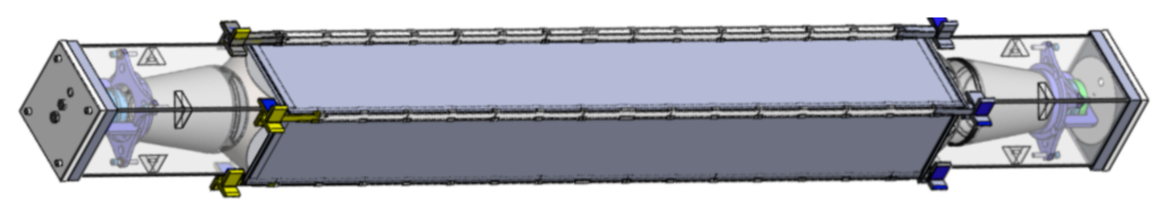


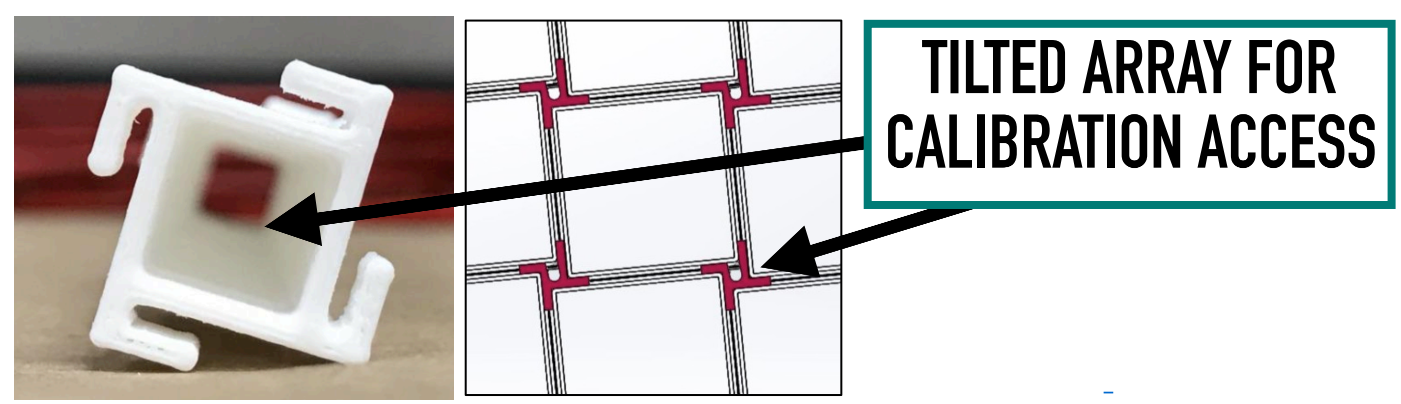
- Shortest baselines at HFIR imply in-building operation
  - high time-dependent gamma rates (in some locations approaching 5 mrem/hr)
  - time and spatially varying thermal neutron fields
  - only facility overburden concrete roof (<1 mwe), atmospheric neutron interactions highly significant
- Design for background rejection (Liquid Scintillator is key)

Design Overview

- Single 4,000 L ^{6}Li-loaded liquid scintillator (3,000 L fiducial volume)
- 11 x 14 (154) array of optically separated segments
- Very low mass separators (1.5 mm thick)
  - Corner support rods allow for full in situ calibration access
- Double ended PMT readout
  - High light collection
  - Full X,Y,Z event reconstruction
- Optimized shielding to reduce cosmogenic and local backgrounds

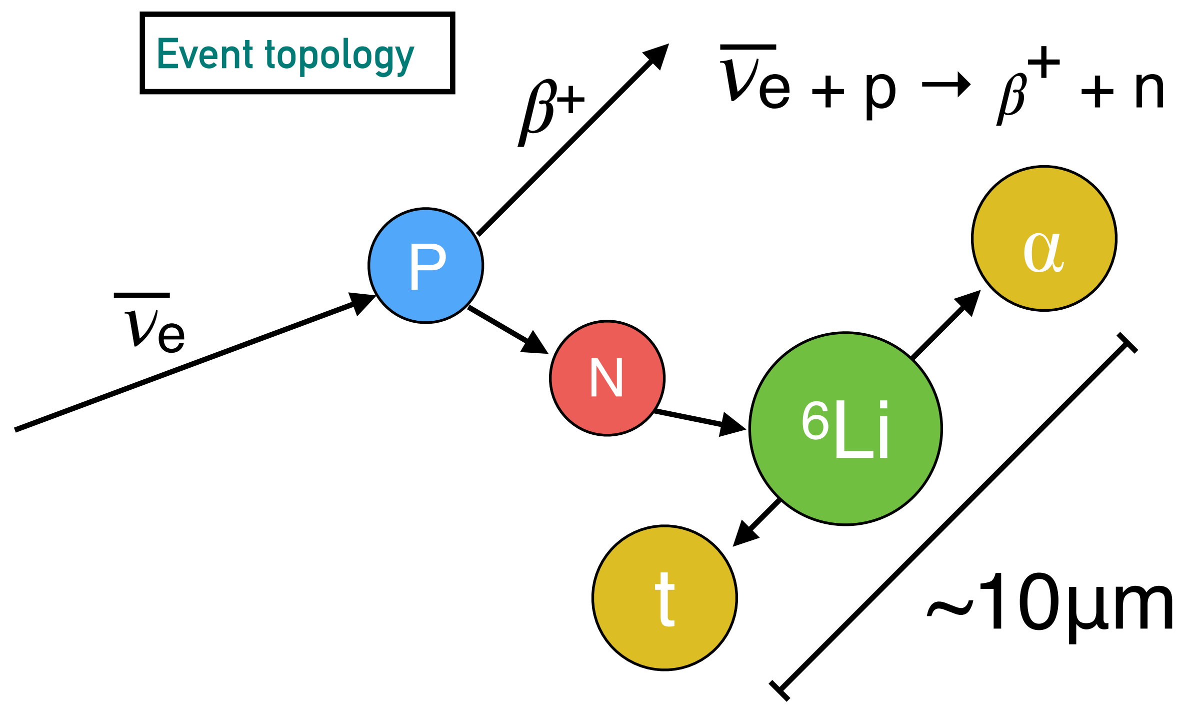




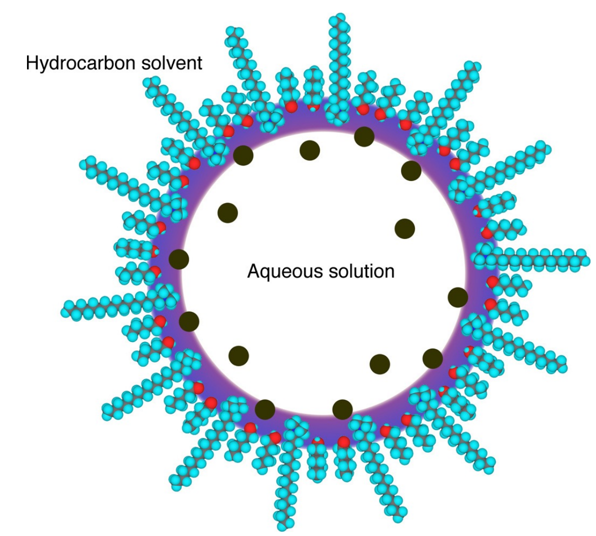


Unique ^{6}Li Loaded Liquid Scintillator

- Compact, segmented detector needs a capture signal that is highly localized.
  - Lithium-6 neutron capture daughter nuclei fit the bill!
  - Also gives even efficiency, and good prompt-delayed distance definition

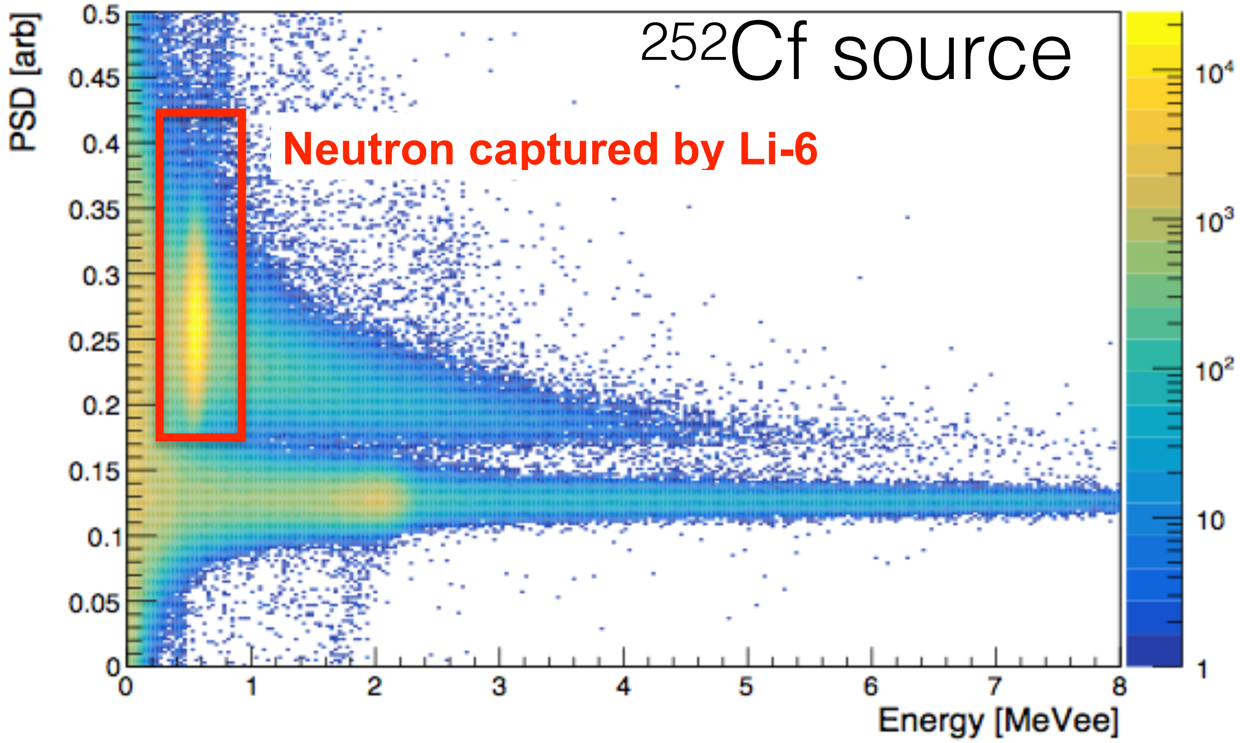


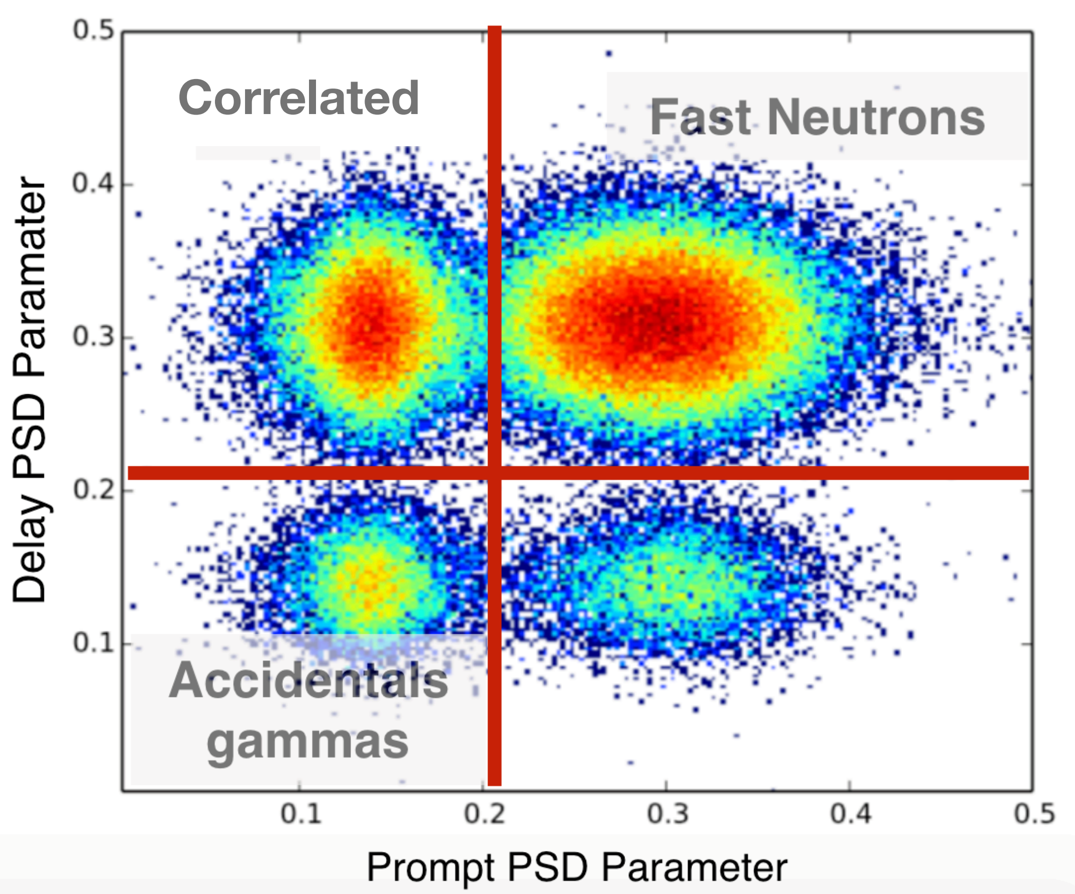
- R&D program led to 0.07% ^{6}LiLS loaded liquid scintillator, meets all requirements.
  - capture time long compared to scattering physics, short compared to accidental rate.
  - High light yield (~8200ph/MeV) for energy resolution
  - Particle ID through pulse-shape discrimination (PSD)
  - Long term stability, material compatibility, nonflammable



^{6}Li loading via Reverse micelles:
- Surfactants added to base liquid scintillator
- Dynamically stable
- relatively high loading possible > 0.1%
- minimal reduction in light yield
- minimal reduction of PSD performance

Unique IBD Signature





Coincidence + PSD to reject backgrounds

Event Coincidence Signature:
e-like prompt signal, followed by a ~40-50μs delayed neutron capture

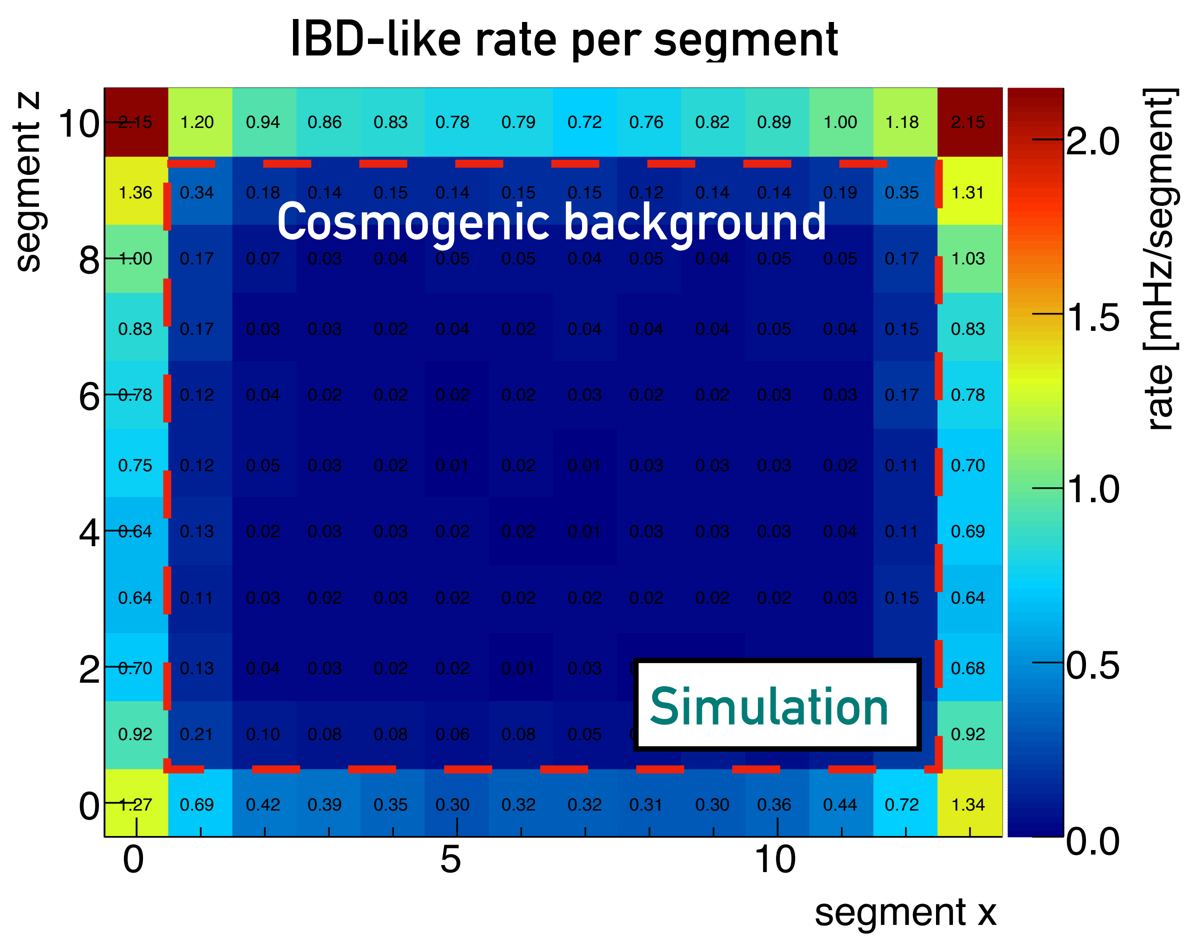
Pulse-shape Discrimination (PSD) Signatures

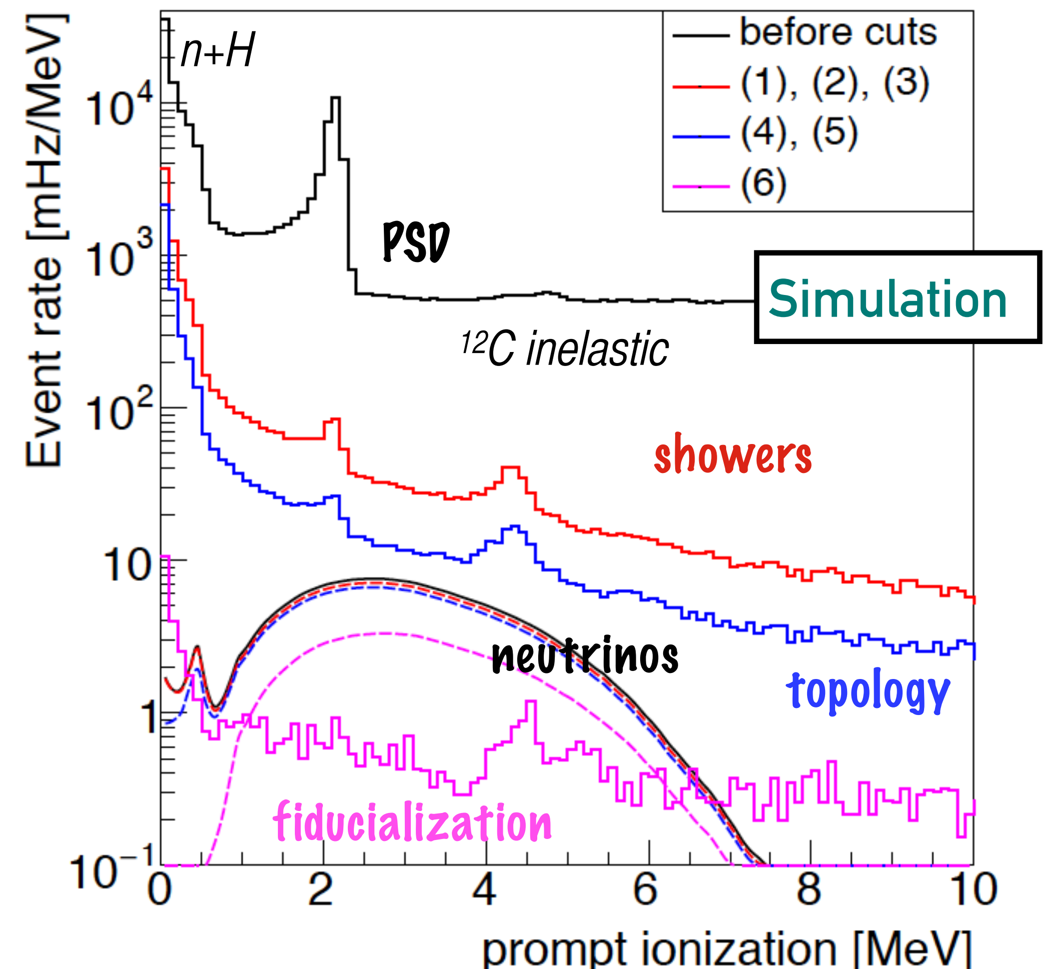
Inverse Beta Decay
γ-like prompt, n-like delay

Fast Neutron
n-like ~~prompt, n-like~~ delay

Accidental Gammas
γ-like ~~prompt, γ-like~~ delay

Background Rejection





- Detector design further optimized for background rejection
- A sequence of cuts leveraging spatial and timing characteristics of an IBD yields > 10^4 background suppression and signal to background of > 1:1.
- Rate and shape of residual IBD-like background can be measured during multiple interlaced reactor-off periods.

Combine:
- PSD
- Shower veto
- Event topology
- Fiducialization

Liquid Scintillator Production

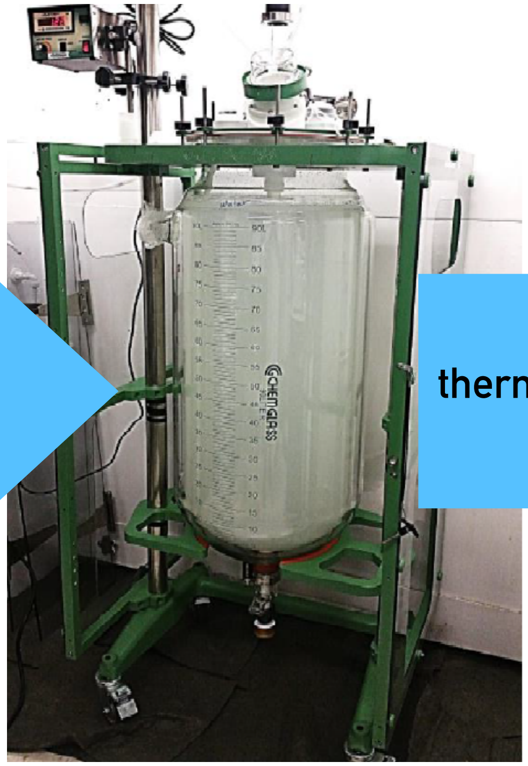
- Liquid scintillator developed for PROSPECT is composed of EJ-309 liquid scintillator, a nonionic surfactant and ^{6}LiCl. Total loading 0.07% Li by mass
  - 2,5-diphenyloxazole (PPO)
  - 1,4-bis(2- methylstyryl) benzene (bis-MSB)
  - di-isopropylnaphthalene (DIN)-based scintillator (EJ-309)
  - Surfactant: ether-based glycol
  - 10 M 6LiCl
- QA/QC performed on all raw EJ309 stock (absorbance between 200 and 1,000 nm)
- Roughly 5,000 L produced in 90 L batches over 9 months

Purification of raw materials (thin film vacuum distillation of by manufacturer)

90 L double wall reaction vessel

Mixing of raw materials

Addition of LiCl



Formation of thermodynamic micellular mico-emulsion

Final product

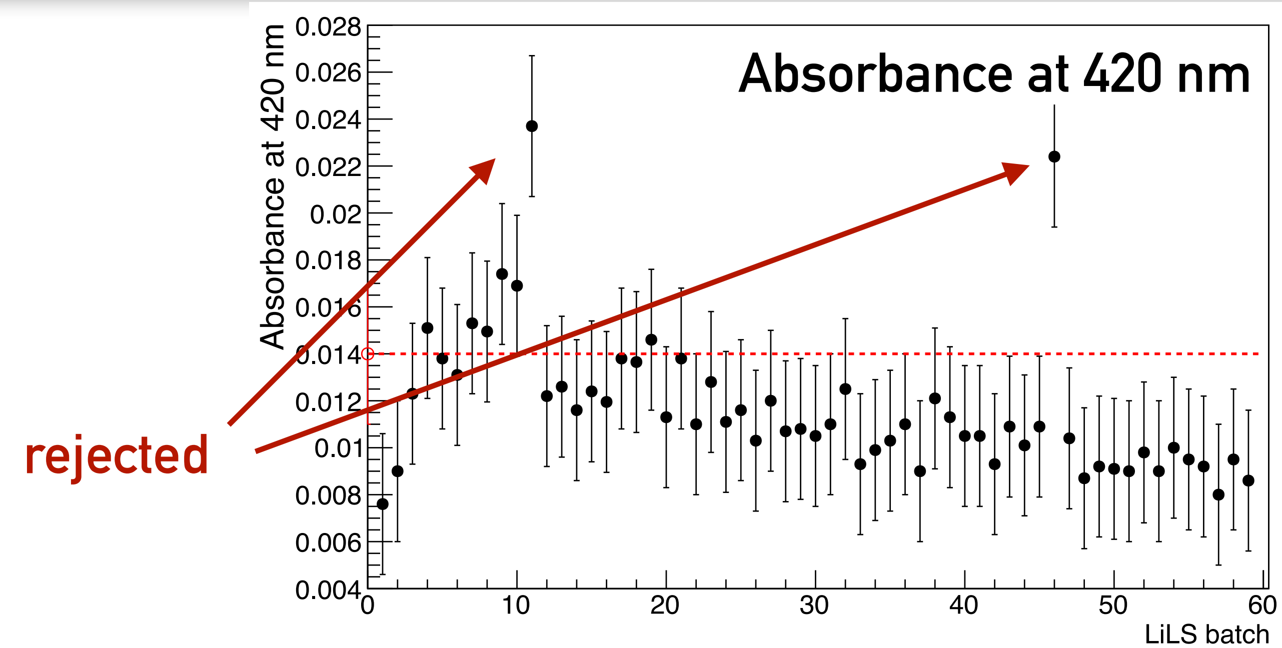
PROSPECT LS Production at BNL

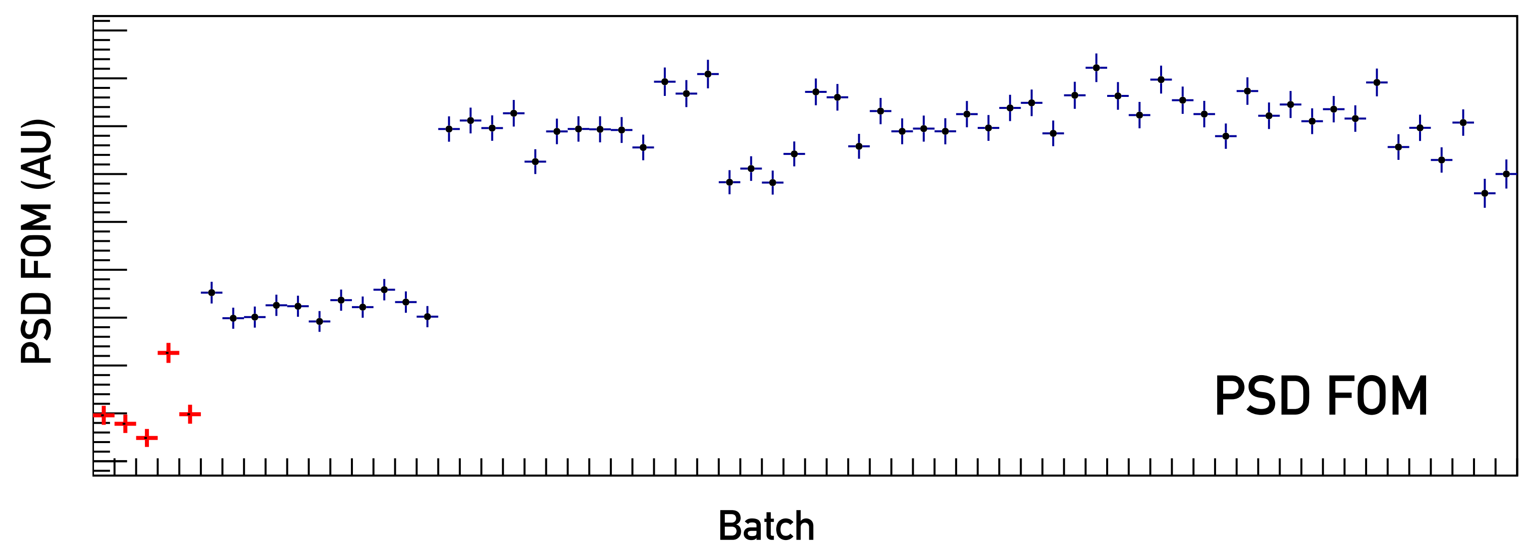
59 batches were produced. The QA/QC program compared several metrics to the performance of a well characterized prototype (P50)

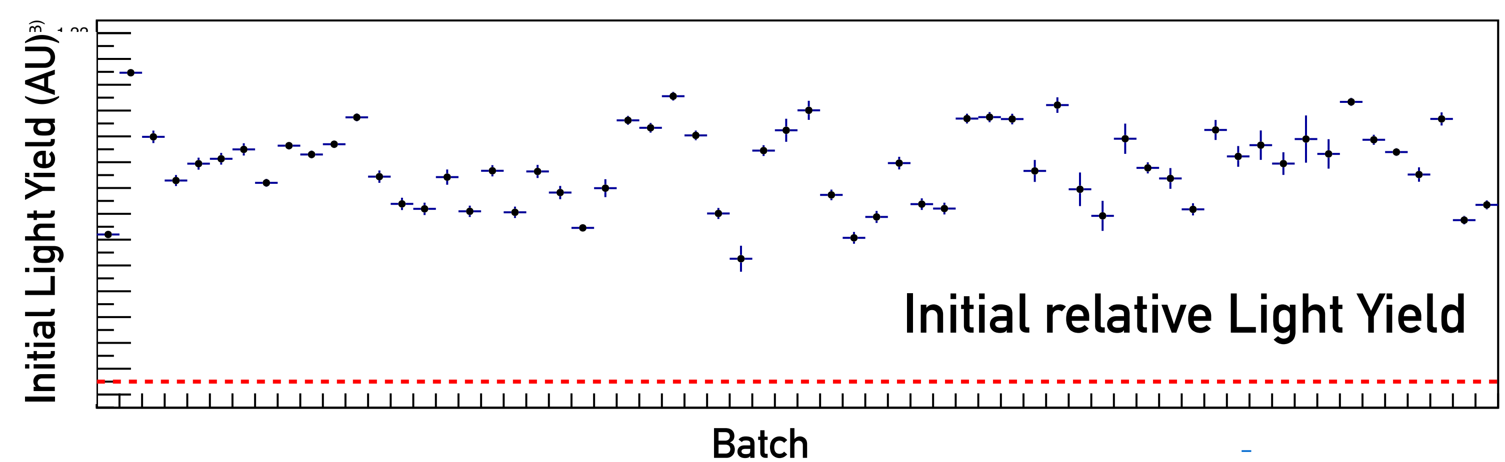
- Optical transparency (UV),
- Light Yield (LY)
- Pulse-Shape-Discrimination (PSD)

Variation in initial batches attributed to oxygen contamination of EJ309 base, manufacturing process improved

Overall, excellent consistency demonstrated, with observable improvements due to raw materials later in production







LiCl Preparation

Doped 0.07% by mass with ^6Li

Accomplished using surfactant and 10 M solution of lithium chloride.

LiCl solution produced from ~95% enriched lithium carbonate

Process involves drying, dissolution, filtration, and purification

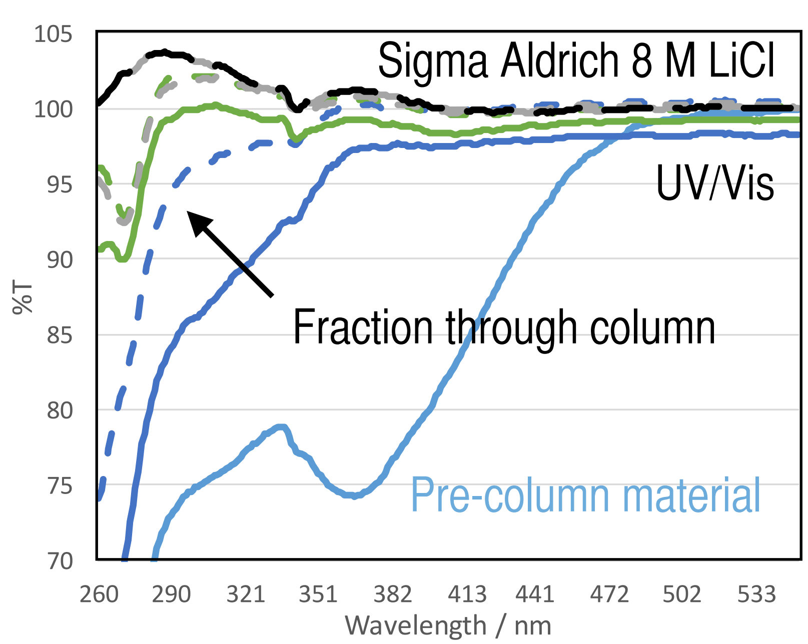
$2HCl + Li_2CO_3 = 2LiCl + H_2O + CO_2\uparrow$ (gas evolution)

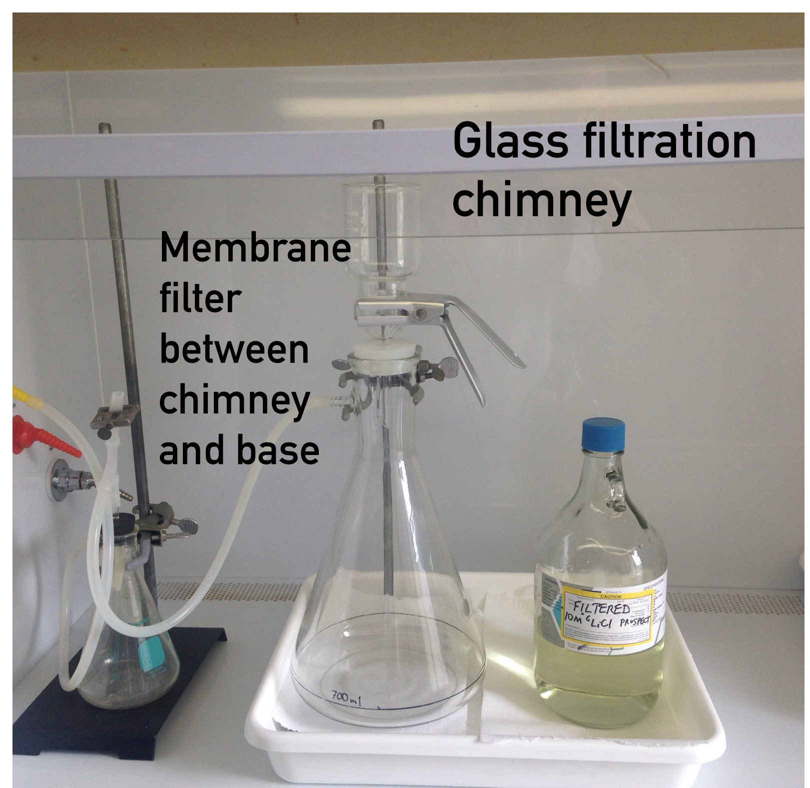
Purification removes anionic complex $FeCl_4^-$ that cause yellowing, Uranium and daughters characterized.

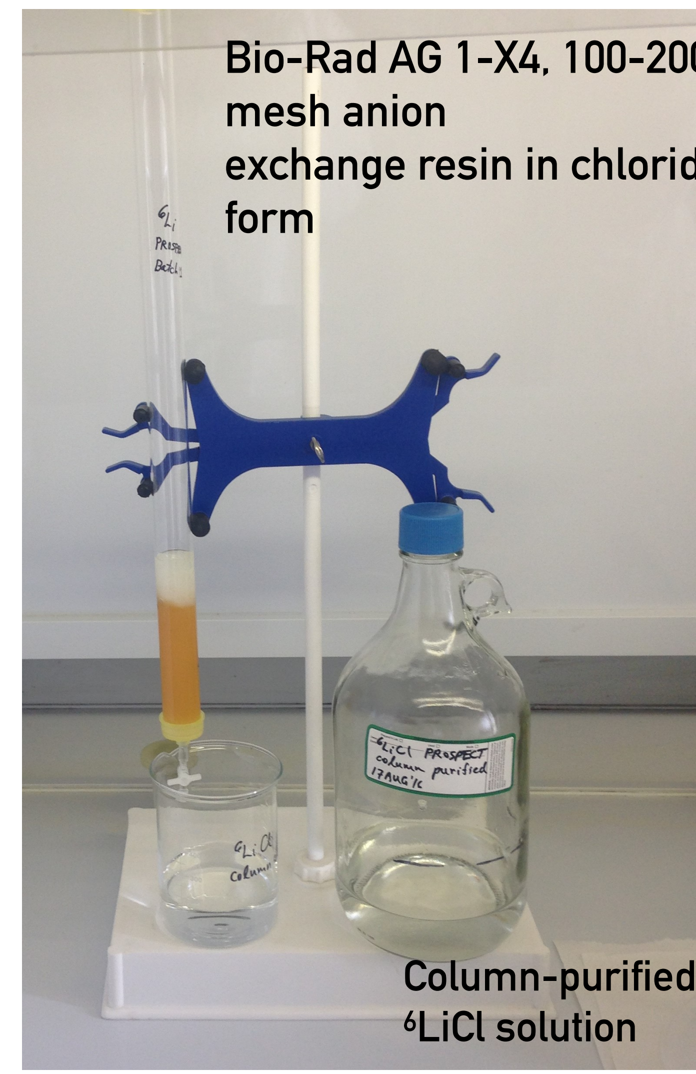
Mass spectrometry measurements give the precise $^6Li/^7Li$ isotopic ratio and ^6Li concentration in the final solution.

UV-Vis used for additional QA/QC.

Material shipped to BNL for addition to LS





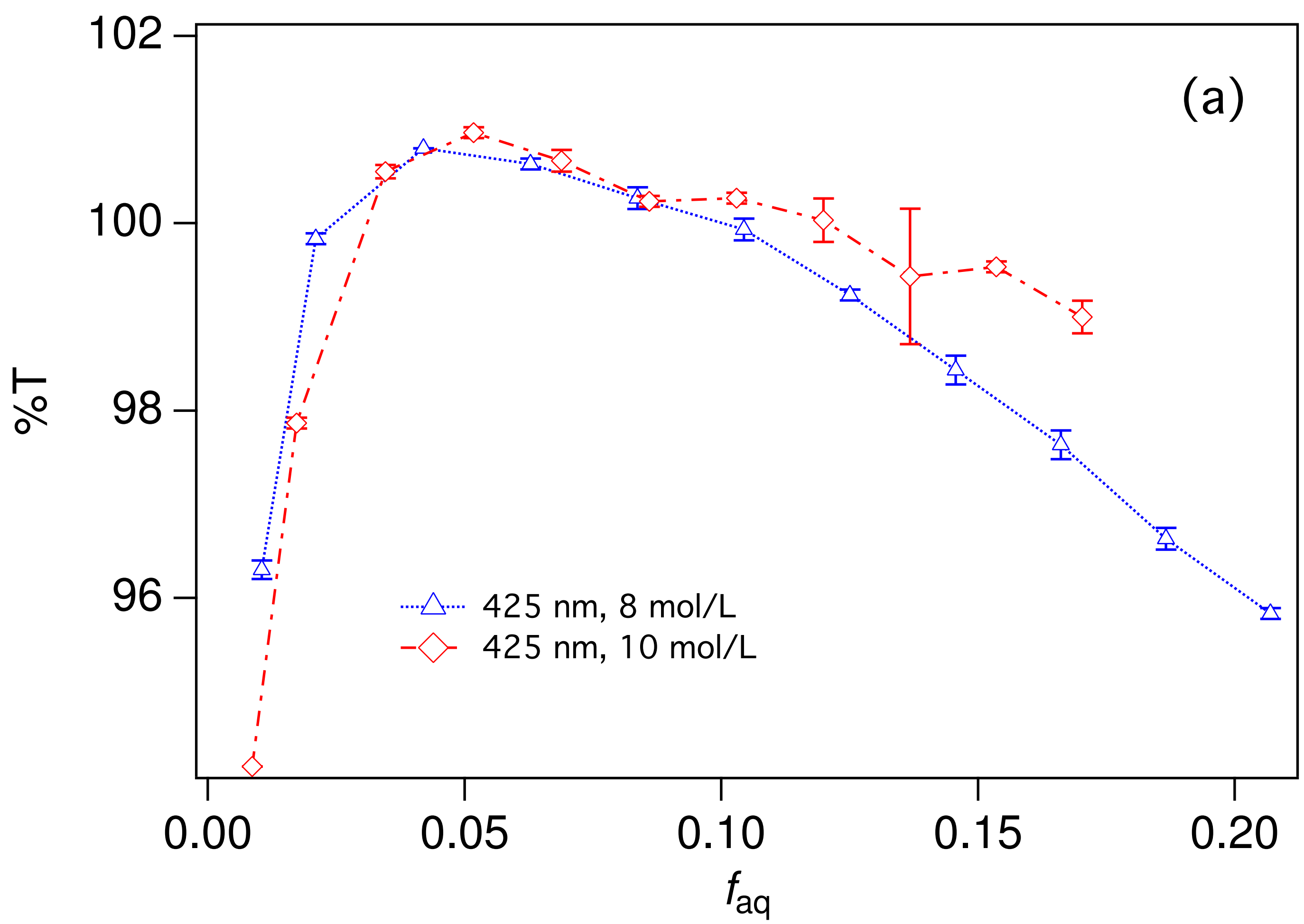


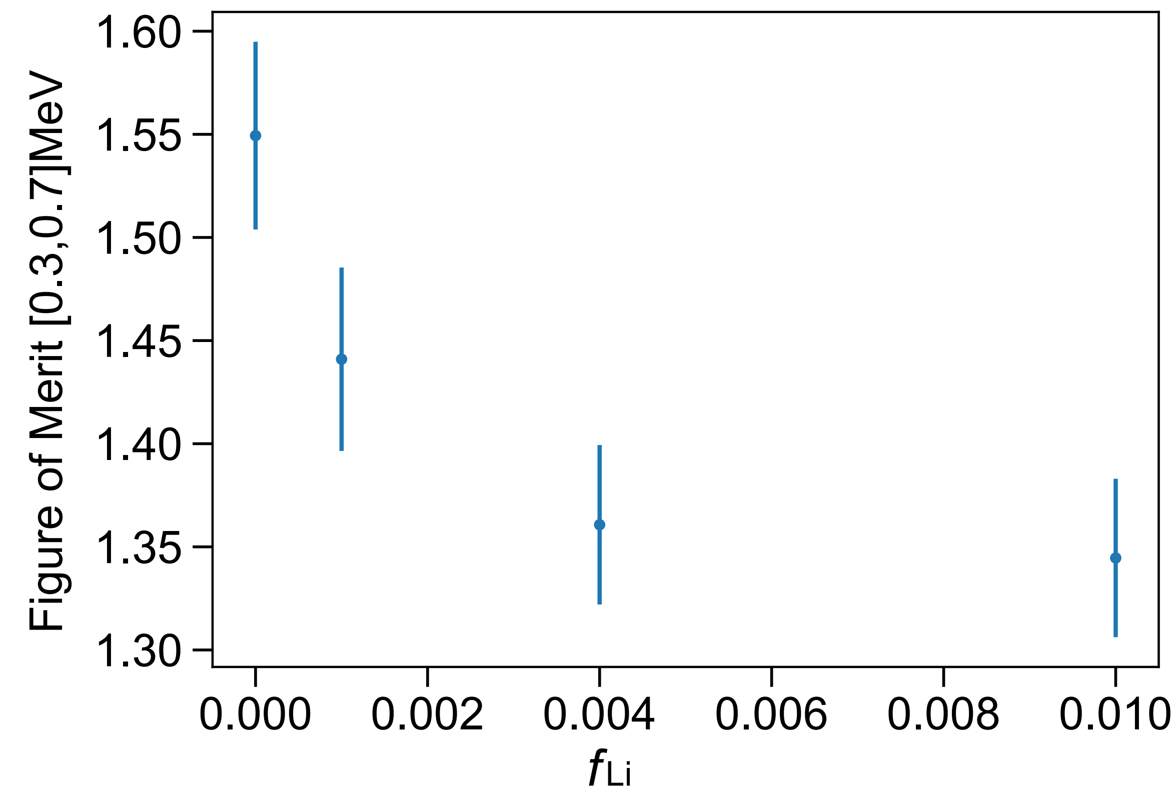
General trends observed in analogous system (LiCl doped UltimaGold AB)

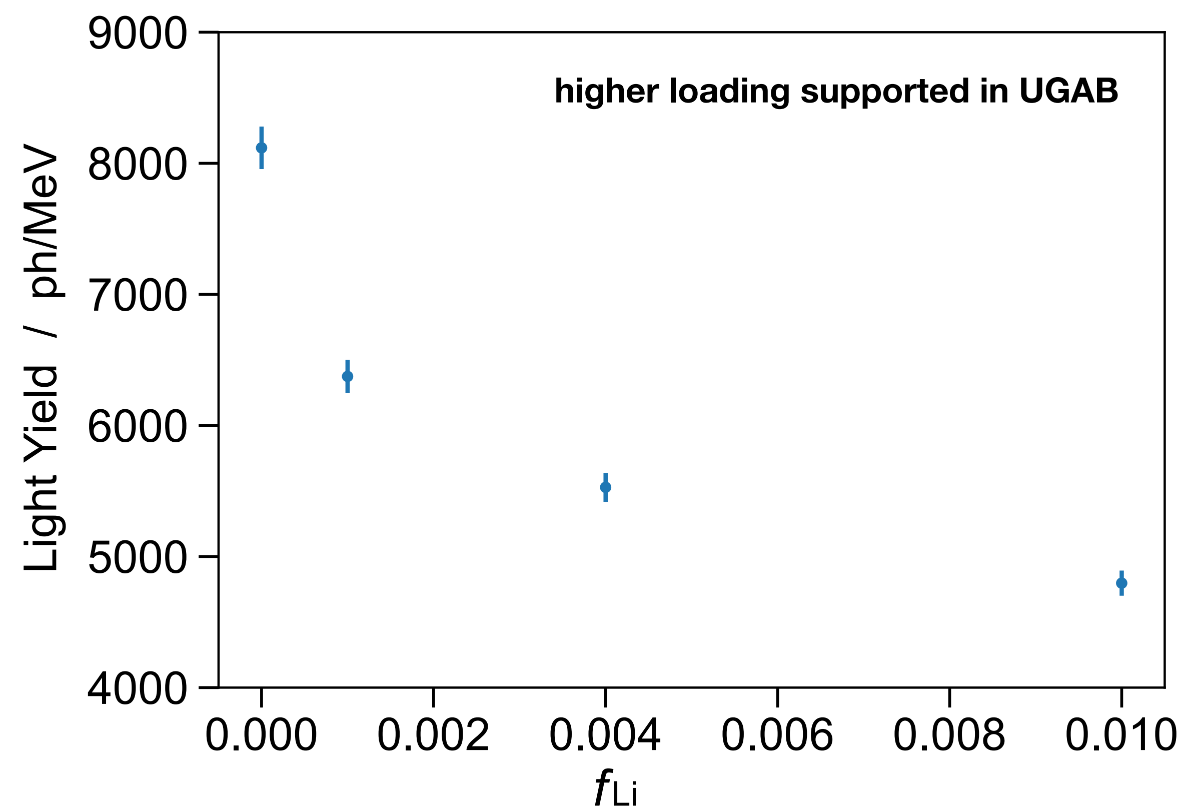
PSD FOM falls (~7 %) upon initial loading

37 % decrease in LY at maximum stable loading

Transmission dependent on aqueous fraction (micelle size)

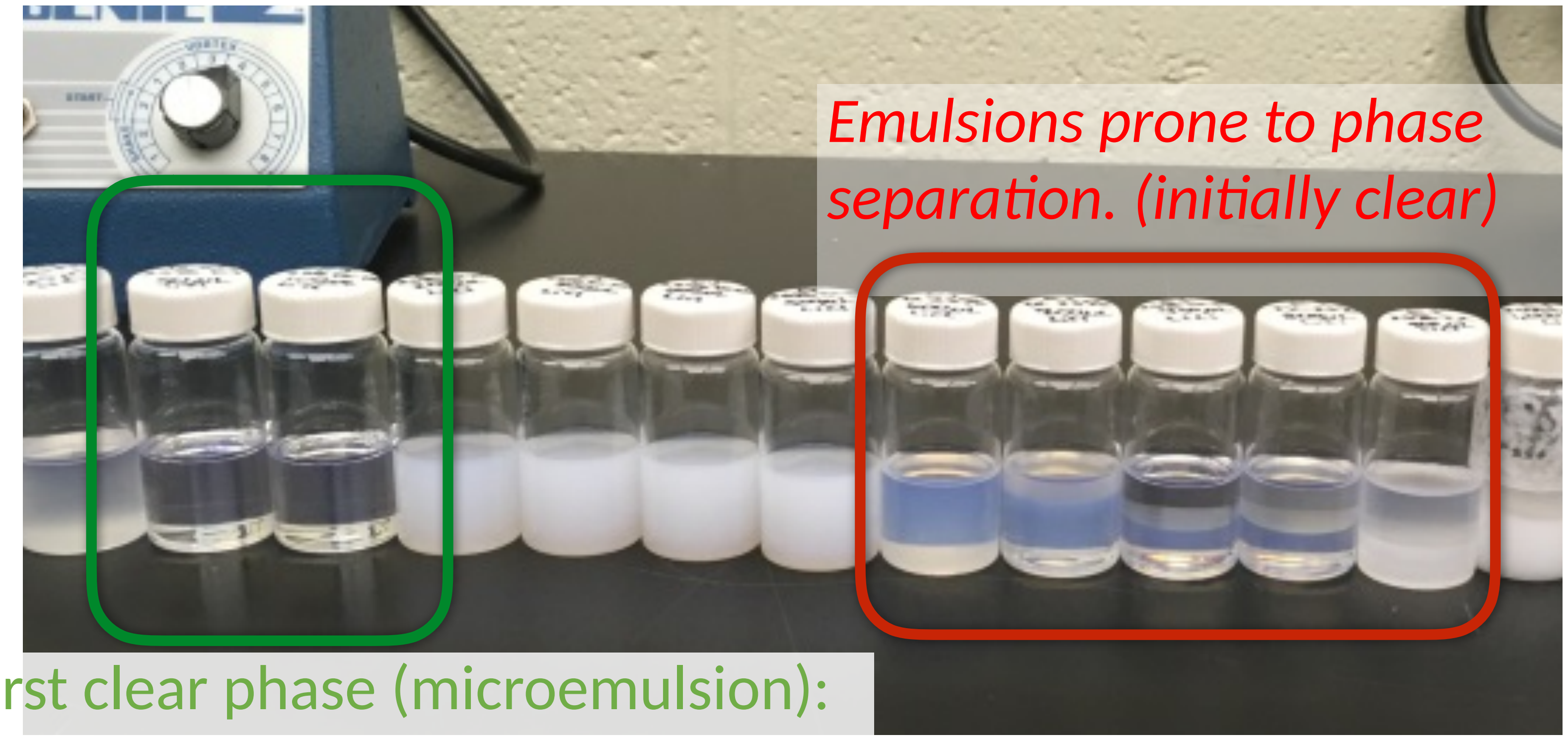






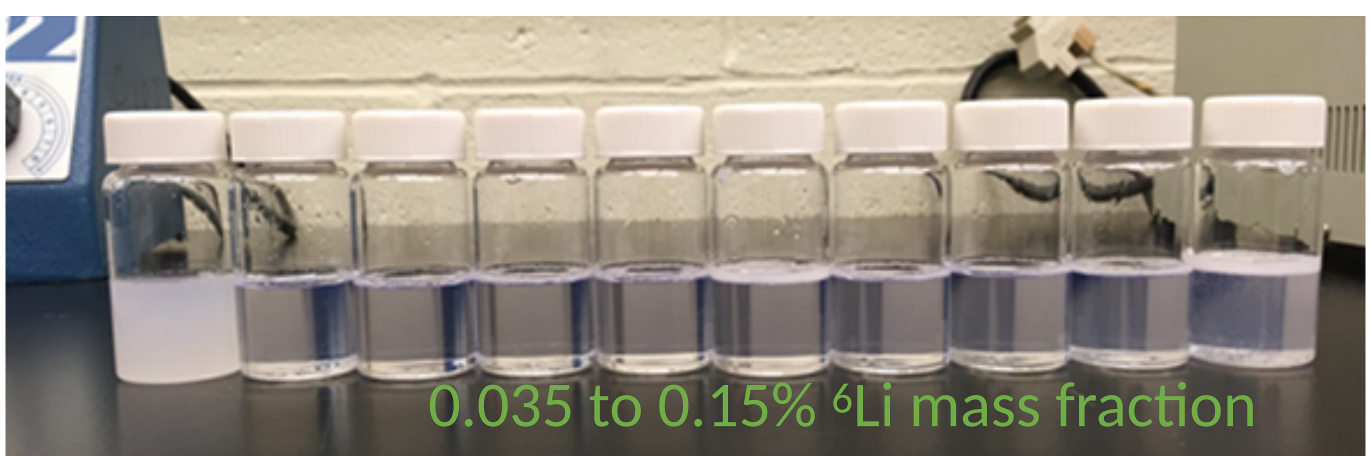
Micro-emulsion Stability

- True Micro-emulsions are thermodynamically stable.
- Uniformly distribute Li throughout the scintillator
- However, challenging to visually separate micro emulsions and emulsions (which separate over time)

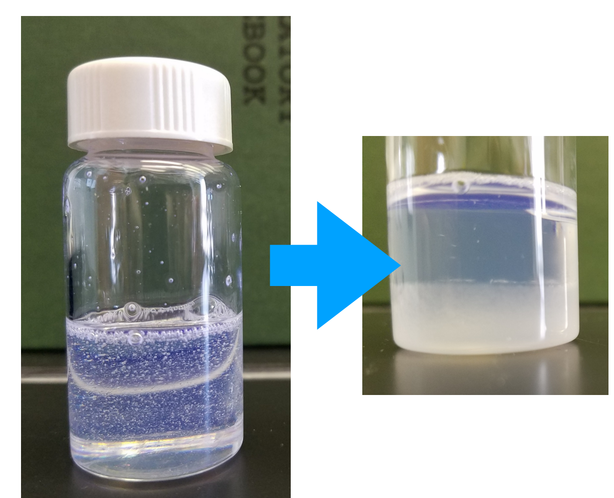


First clear phase (microemulsion):
0.035 to 0.15% ^{6}Li mass fraction

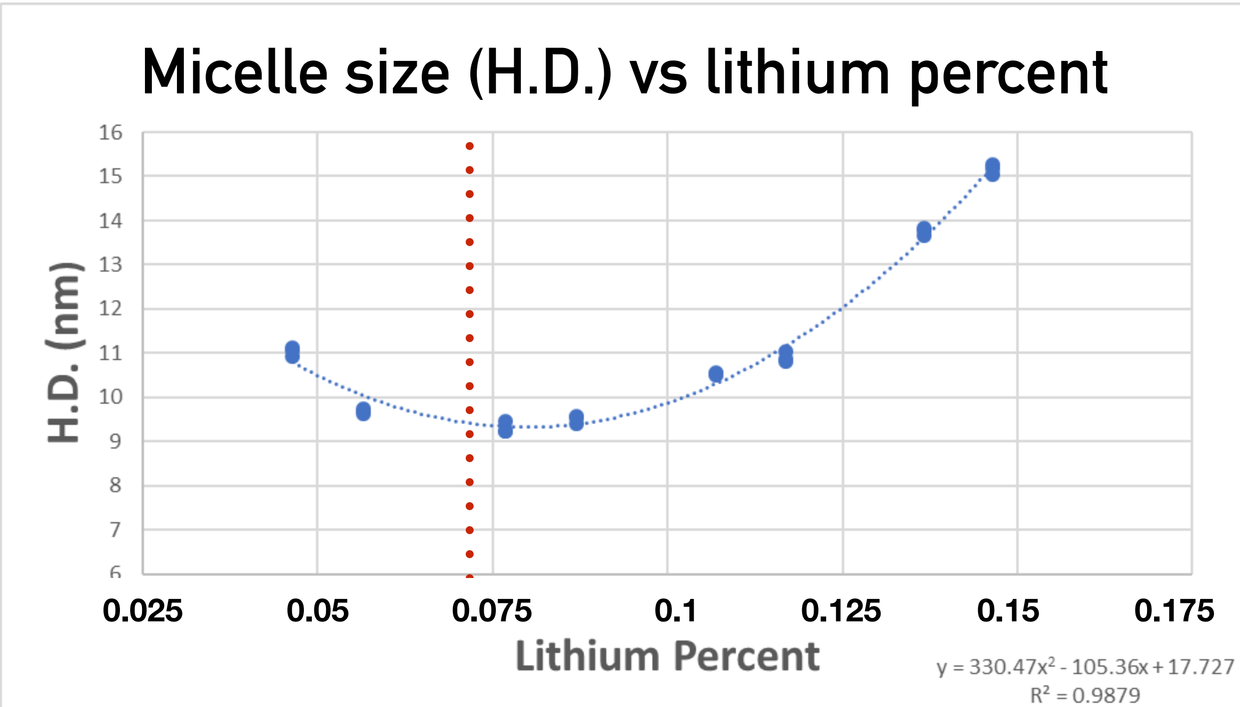
Gradually adding aqueous LiCl to the PROSPECT scintillant identifies turbid phase boundaries

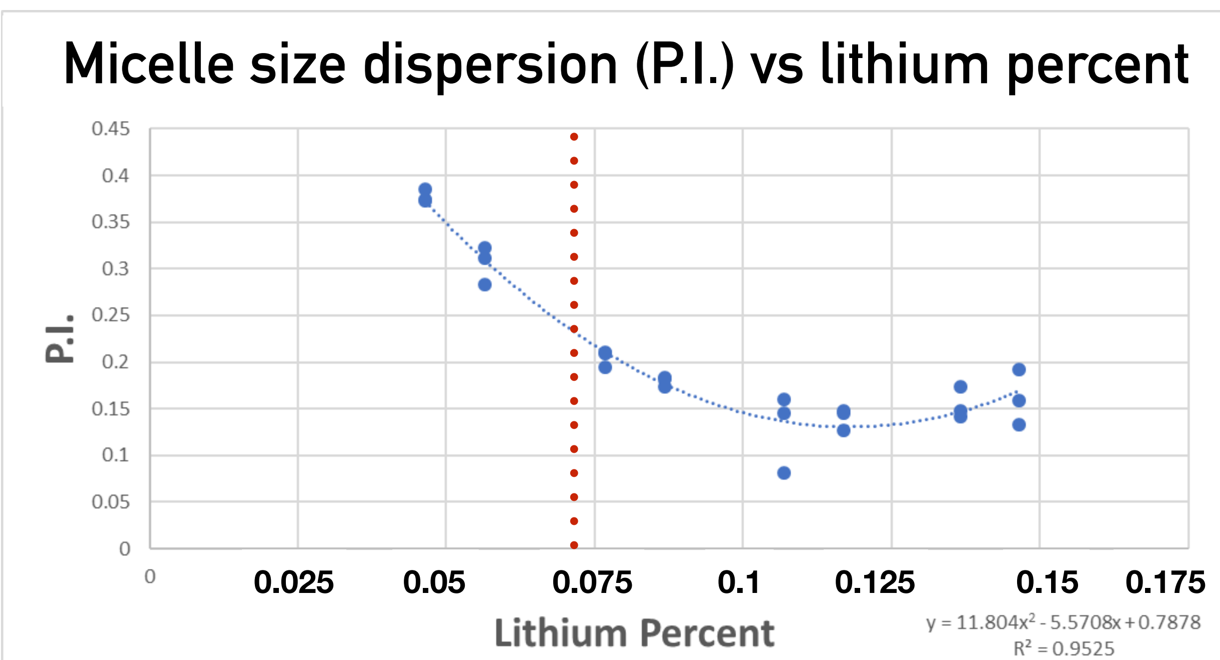


- Boundaries of stable phases confirmed using centrifugation



Micro-emulsion Stability





- nanometer-scale reverse micelles scatter light strongly
- Dynamic light scattering gives a measure of the size and polydispersity (size distribution) of reverse micelles in Li-loaded PROSPECT scintillant
- PROSPECT LS is near the middle of the range.
- Indicates well formed thermodynamically stable reverse-micelles: long term stability

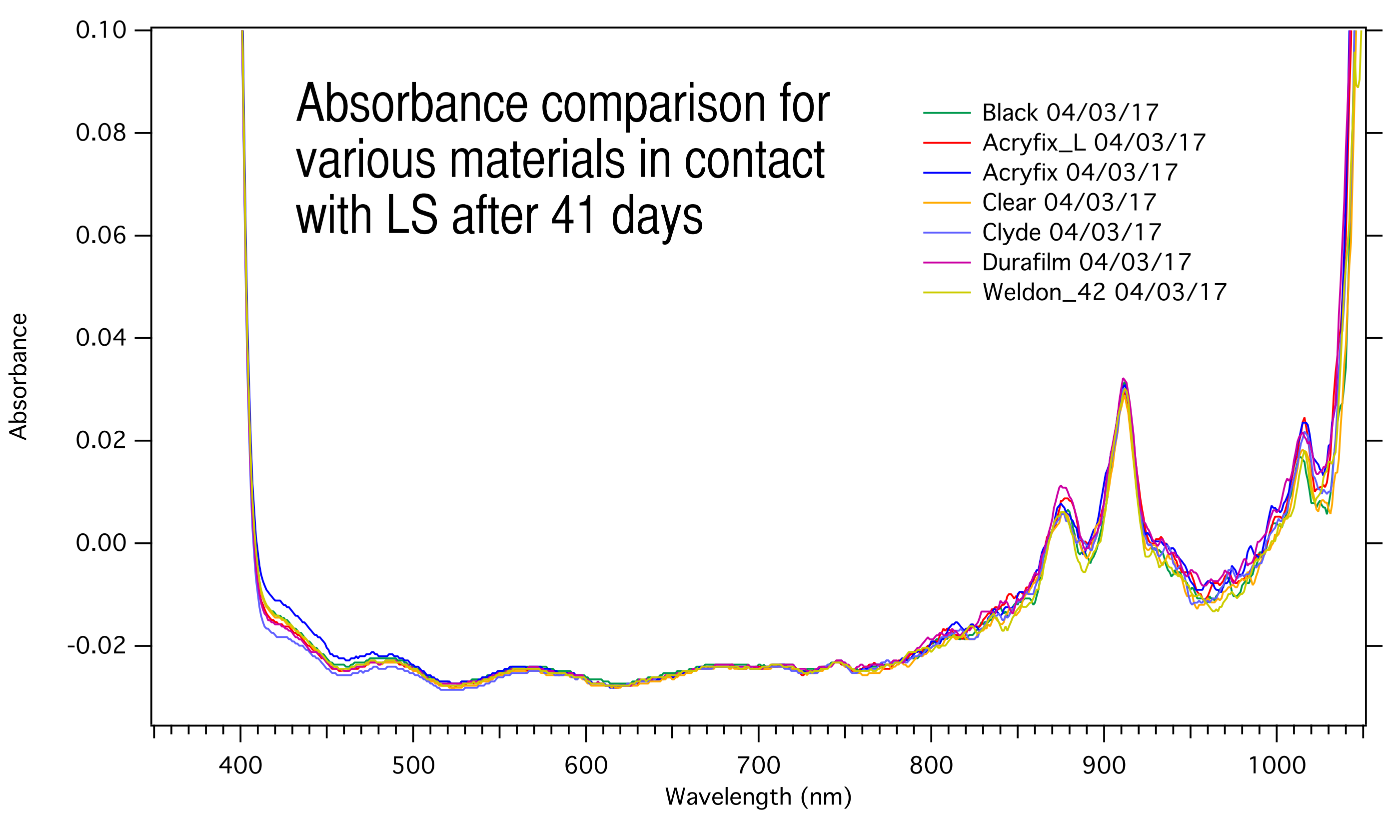
Extensive material compatibility campaign carried out

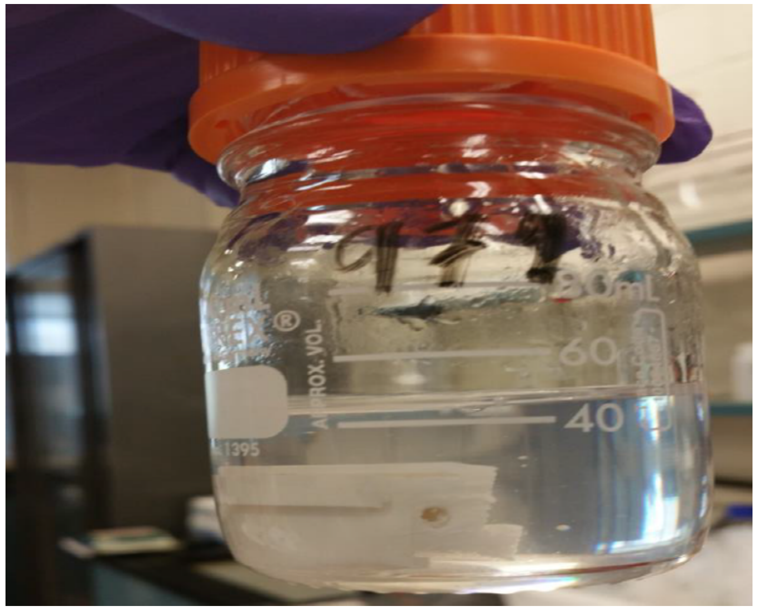
60+ materials tested for effect on loaded liquid scintillator.

PLA tested for material properties after scintillator exposure

- Samples carefully cleaned and sent to multiple insitutions
- Soaked for up to several months in LS
- Absorbance compared to reference samples via UV - Vis

All materials in direct contact with liquid scintillator qualified at multiple institutions.
Long term monitoring plan implemented at Le Moyne
Operation of various prototypes (e.g. P50X) validate final materials choice.



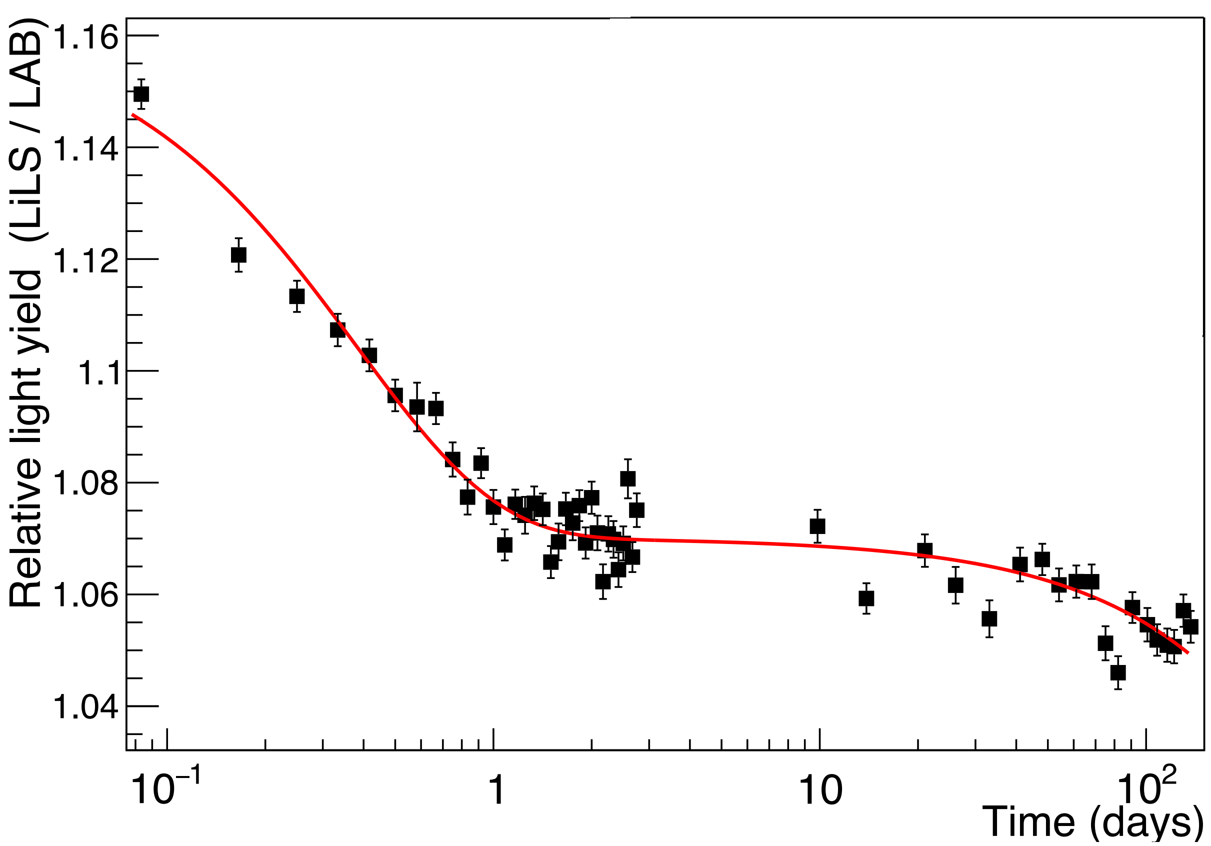


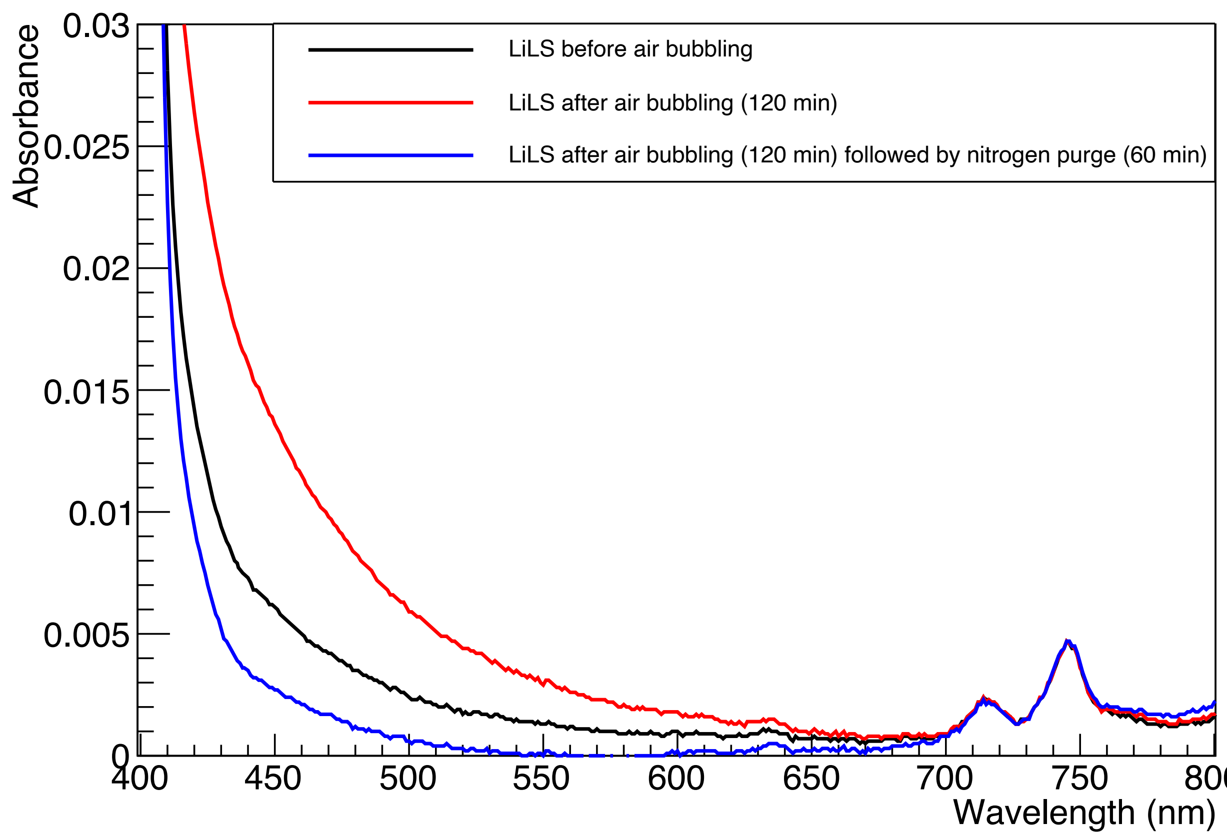
Printed PLA pinwheel in sample jar

Notable oxygen quenching effects observed

multiple time constants; short term effect partially recoverable

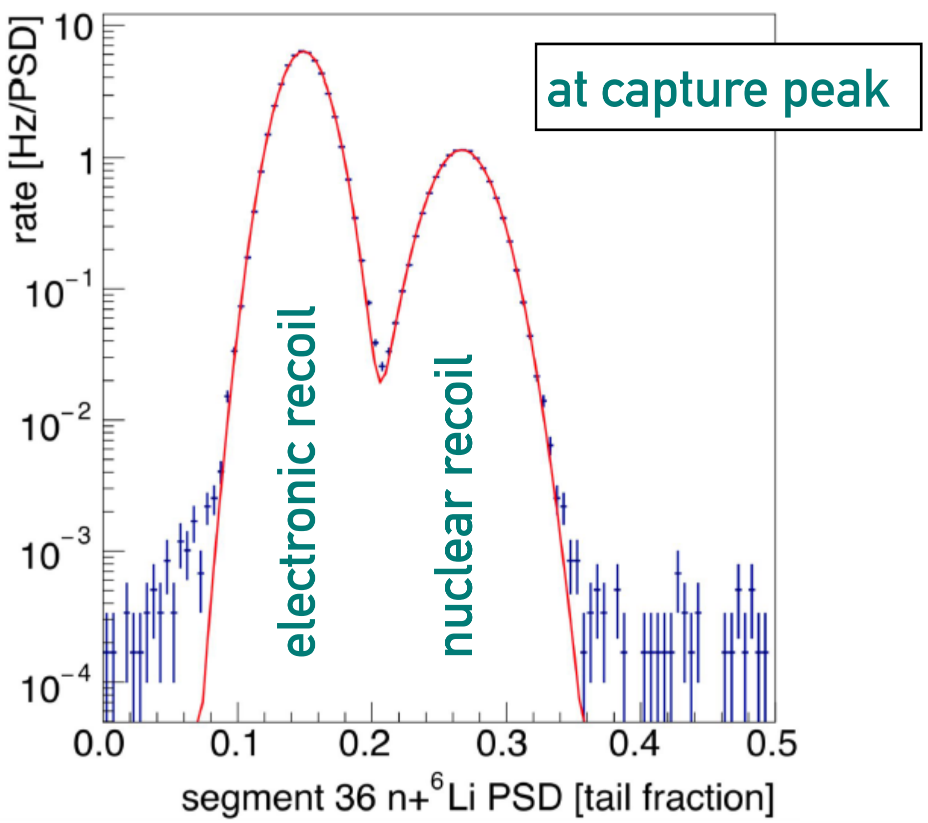
- Samples sparged at 30 ml/min dry N2 for 30 min prior to QA/QA measurements
- Final LS premixed in single volume and sparged with boil-off N2 prior to filling PROSPECT
- Cover gas of boil-off N2 maintained throughout



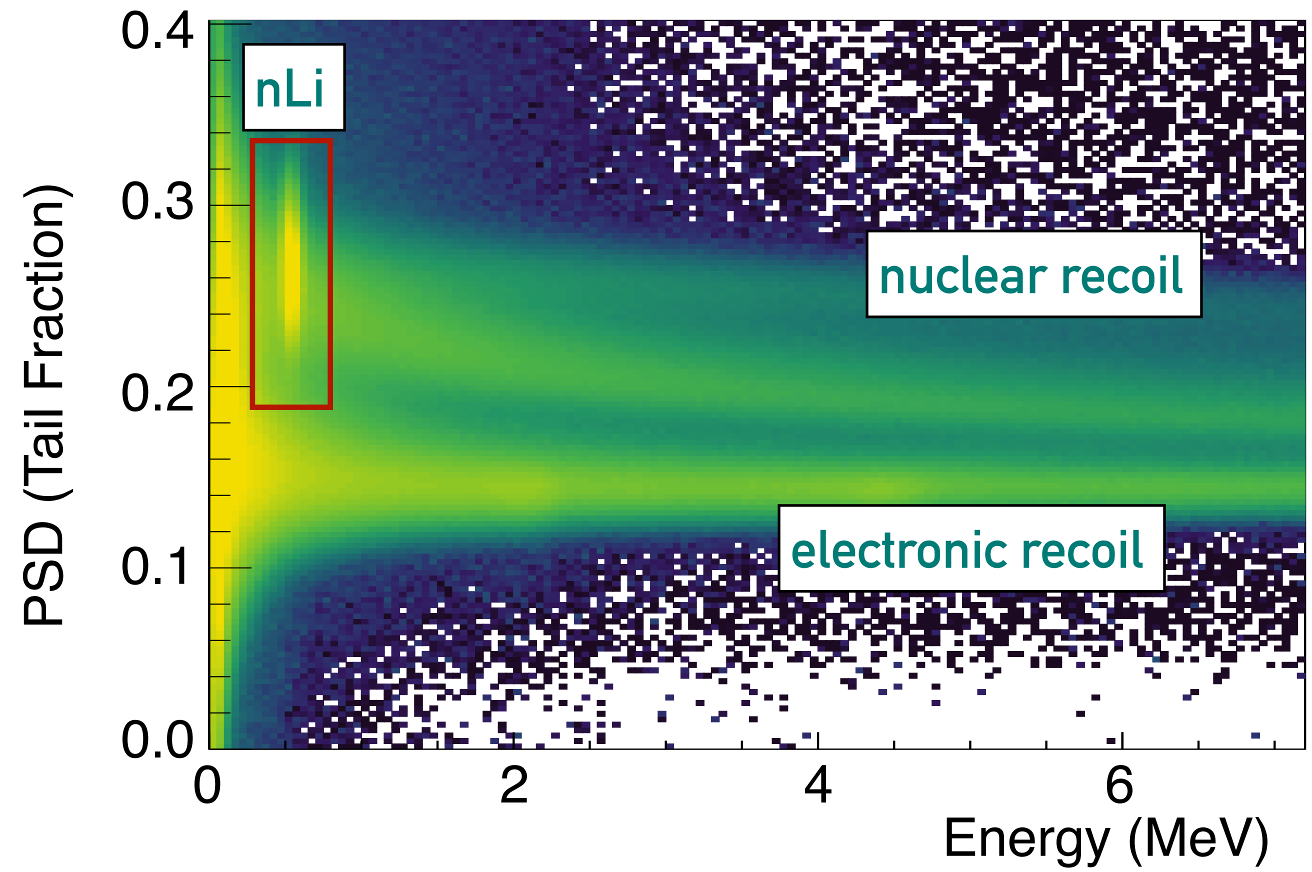


Pulse Shape Discrimination

Single segment PSD



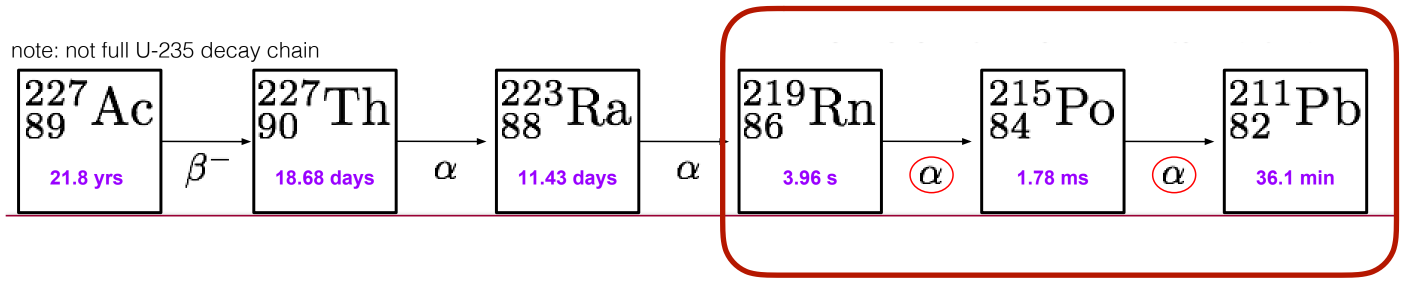
Full detector PSD



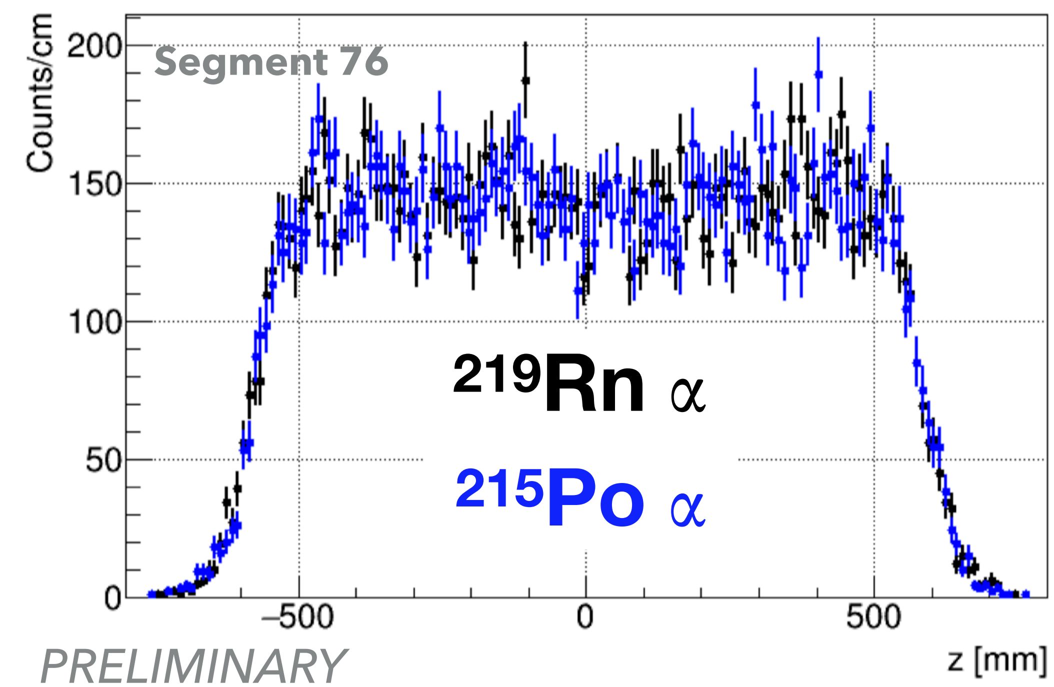
- Excellent discrimination of gamma interactions, and nuclear recoils
- Well separated ^{6}Li-n capture peak
- As dominant backgrounds are cosmogenic fast neutrons, reactor-related gamma rays, and reactor thermal neutrons:
  - Vast majority identified and rejected by Pulse Shape Discrimination for Prompt and Delayed signals

Relative target mass needed for oscillation search

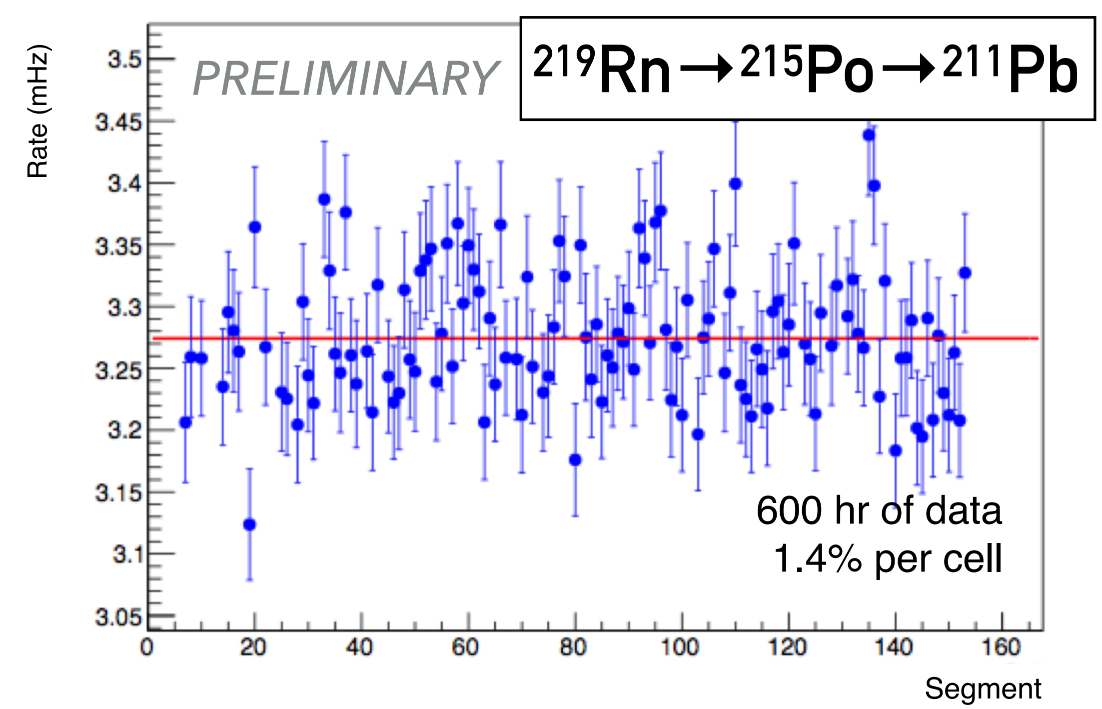
note: not full U-235 decay chain



- ^{227}Ac added to LS prior to filling
- Double alpha decay (^{219}Rn→^{215}Po→^{211}Pb), highly localized, easy to ID, 1.78ms lifetime
- Measured absolute z-position resolution of < 5cm
- Direct measurement of relative target mass in each segment



Uniformity in rates within segment



Uniformity in rates between segments

Conclusions

- PROSPECT has demonstrated a high-performance Li doped liquid scintillator critical to on-surface background rejection.
- Improvements in raw materials and purification methods have (and will continue to) led to higher performance.
- Better understanding of micelle behavior will allow tuning of micelle size to improve performance.
- Other applications for high light-yield PSD capable liquids

PROSPECT Collaboration

prospect.yale.edu

HEISING-SIMONS FOUNDATION

U.S. DEPARTMENT OF ENERGY

GRFP NSF GRADUATE RESEARCH FELLOWSHIP PROGRAM

BROOKHAVEN NATIONAL LABORATORY

NIST

Lawrence Livermore National Laboratory

OAK RIDGE National Laboratory

Drexel

Yale