Large Format Li Co-doped **NaI:Tl (NaIL™)** Scintillation Detector for Gamma-ray and Neutron Detection

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CRYSTALS
SAINT-GOBAIN

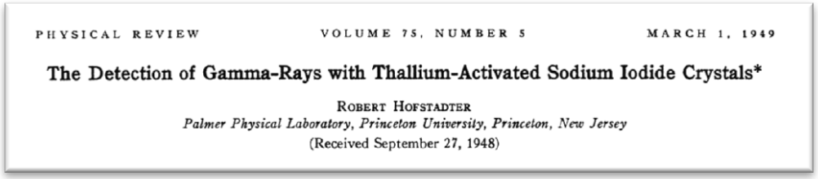
OUTLINE

- ❑ Key Properties of NaIL
- ❑ Applications of NaIL
- ❑ Development of large crystal detectors

- ❑ Main purpose: Present first results on very large dual detection NaIL scintillators

THALLIUM ACTIVATED SODIUM IODIDE (NaI:Tl)
A BRIEF INTRODUCTION

- **Invented by *Dr. Robert Hofstadter* in 1948**



PHYSICAL REVIEW — VOLUME 75, NUMBER 5 — MARCH 1, 1949

The Detection of Gamma-Rays with Thallium-Activated Sodium Iodide Crystals*

ROBERT HOFSTADTER
Palmer Physical Laboratory, Princeton University, Princeton, New Jersey
(Received September 27, 1948)

- **Commercial production first started by *Harshaw Chemical* in the 1950's**

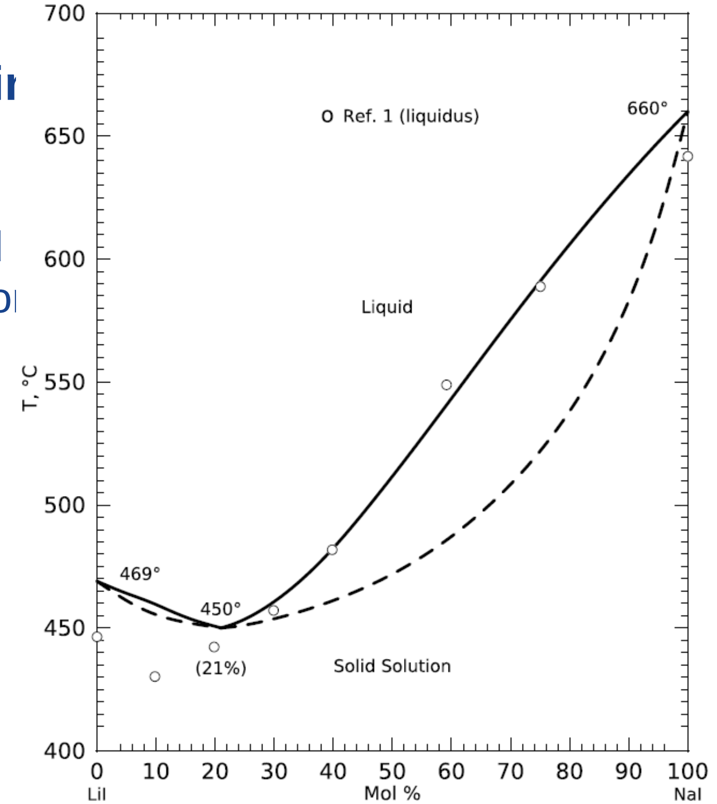
- **Still world's most widely used scintillator (by volume)**
- **Industrial scintillator with mature detector and system designs**

INCORPORATING Li INTO NaI:Tl

- **Neutron detectors are important components in for illicit nuclear material interdiction**
  - only fissile isotopes emit neutrons
  - efficient, inexpensive neutron detectors are wanted
  - **simultaneous** detection of gamma rays and neutron desired

- **Li can be incorporated into NaI:Tl to introduce thermal neutron detection**
  - n + ^{6}Li → t (2.75 MeV) + α (2.05 MeV)
  - 4.8 MeV events create a lot of scintillation light

- **NaI and LiI forms solid solution at any ratio.**
  - Tunable neutron detection efficiency
  - Crystal growth is tolerant of high Li gradients



Phase diagram of NaI - LiI

- **Seems easy, yes?**
  - Need to be able to not mistake gamma-rays for neutrons (no false positives)
  - Previous work in this area had been unable to tell the difference (pulse shape discrimination not workable)

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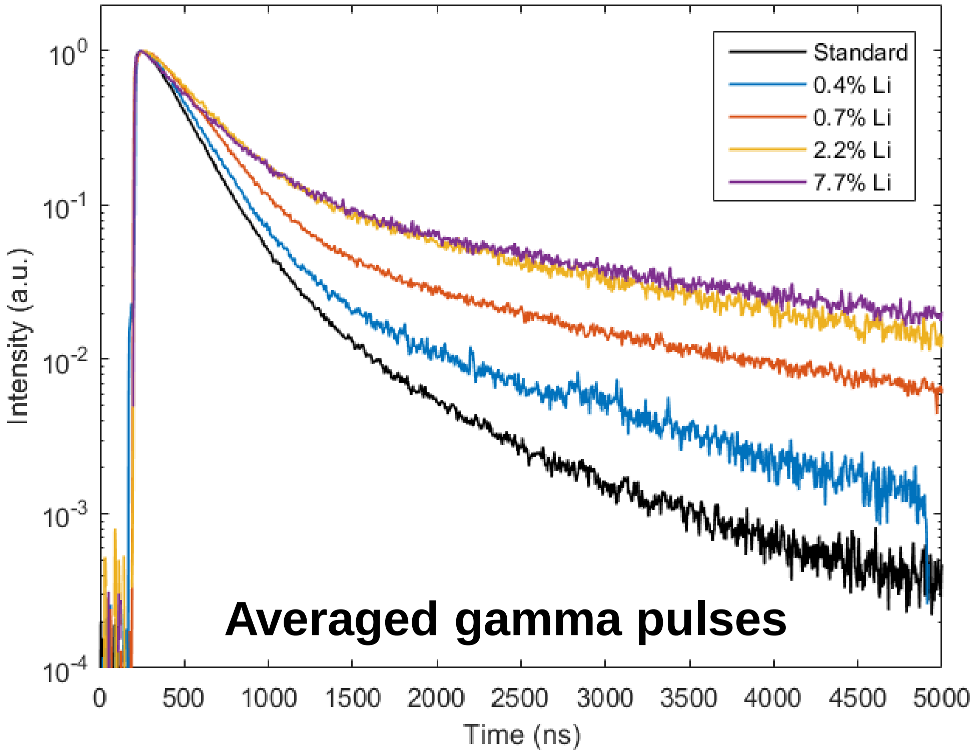
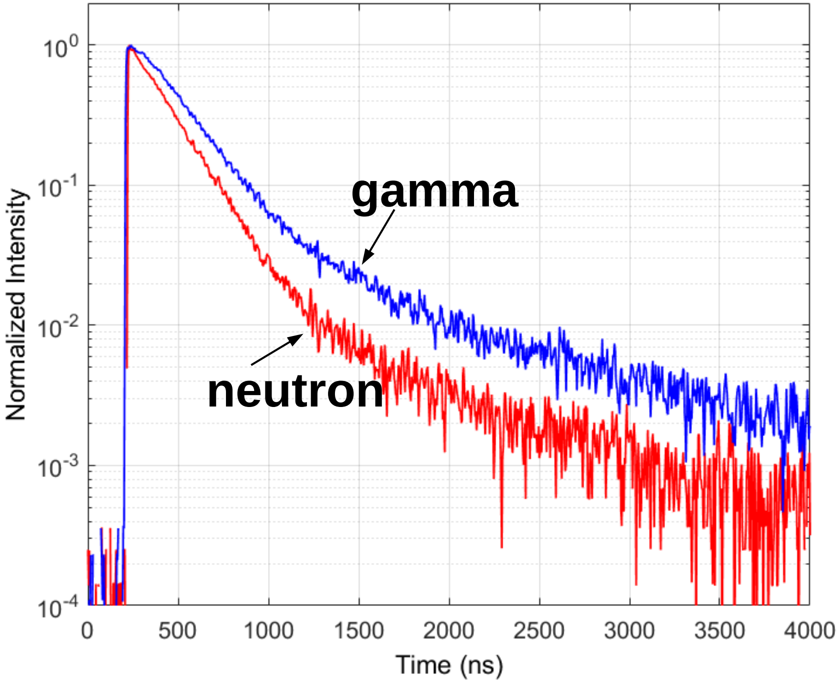
ADDING LITHIUM AFFECTS THE SCINTILLATION PULSE SHAPE

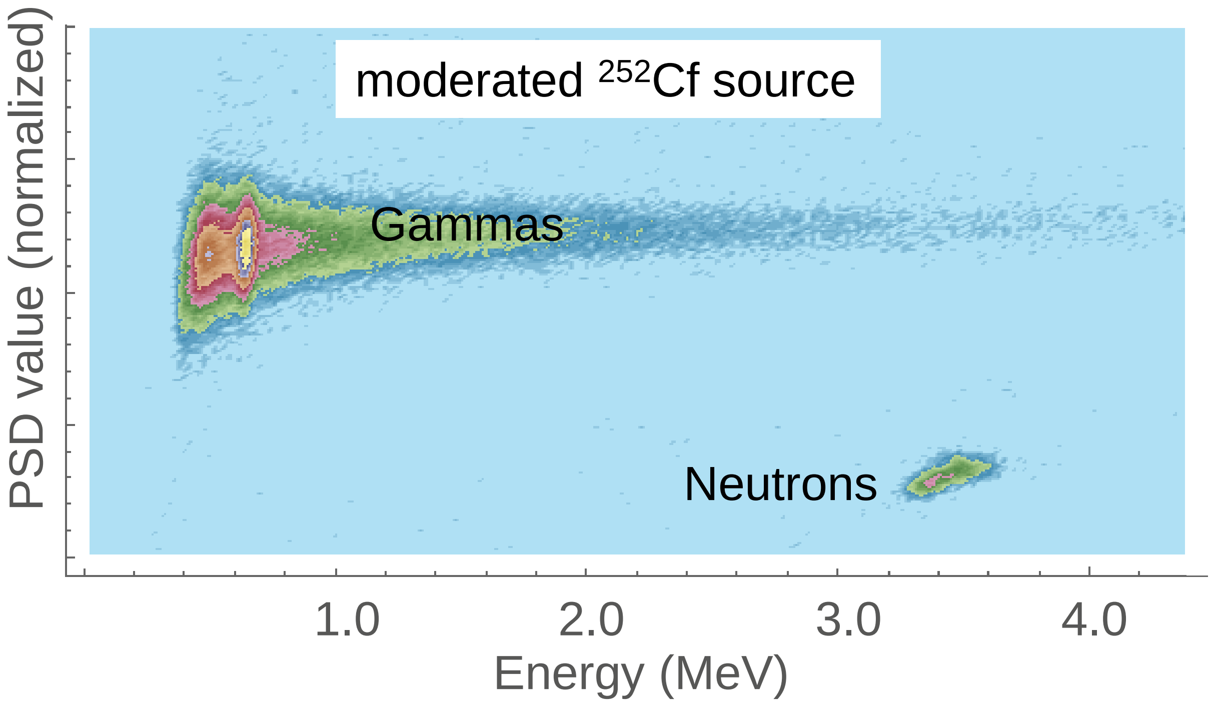
The higher the [Li], the longer the pulse





But, interactions with gammas make more of an increase than with neutrons

PULSE SHAPE DIFFERENCE GIVES EXCELLENT GAMMA/NEUTRON DISCRIMINATION



$$\text{PSD value} = \frac{\int_{400ns}^{1600ns} S(t)dt}{\int_{0}^{1600ns} S(t)dt}$$

i.e. “tail-to-total”

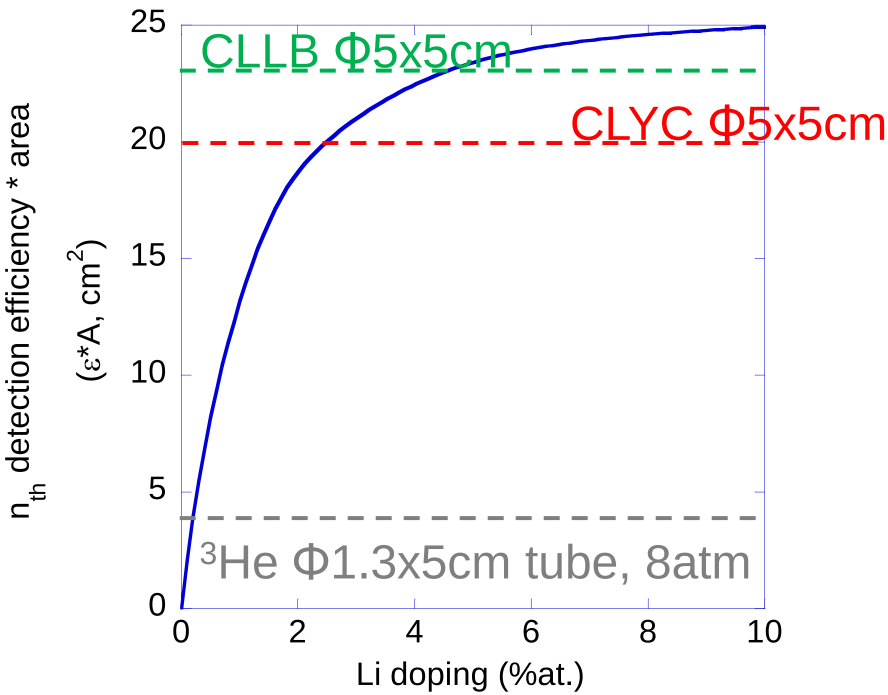
⌀2.5x2.5 cm crystal,
[^{6}Li]= 0.6%

APPLICATIONS FOR NAIL

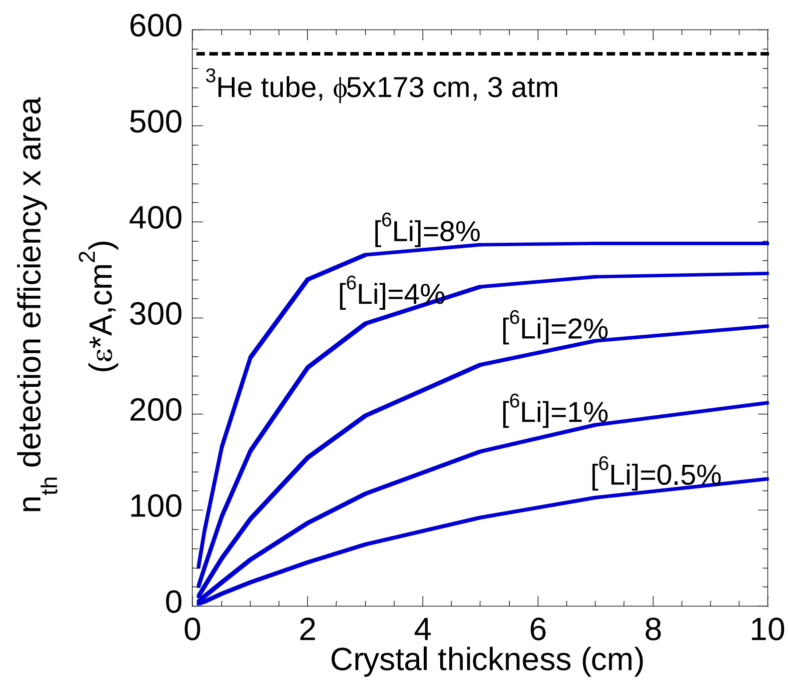
	Target Application	Competing Technology	Advantage of NaIL
	**Radiation Portal Monitor**	Neutrons: ^{3}He tubes ^{10}B tubes $^{6}LiF/ZnS$ films Gammas: Plastic scintillator	Cost, Spectroscopic gamma, Dual mode
	**Mobile**	Neutrons: as above Gammas: NaI:Tl	Cost, Size, Weight, Dual mode
	**Backpack**	Neutrons: as above Gammas: NaI:Tl Dual: $Cs_2\,^{6}LiLaBr_6$ (CLLB) $Cs_2\,^{6}LiYCl_6$ (CLYC)	Cost, Size, Weight
	**Handheld**	Neutrons: ^{3}He $Cs_2\,^{6}LiYCl_6$ $^{6}LiF/ZnS$ films Gammas: NaI, CsI, $LaBr_3$ Dual: $Cs_2\,^{6}LiLaBr_6$	Cost, Size, Weight

A FEW % ^{6}LI DOPING IS SUFFICIENT FOR MOST SECURITY APPLICATIONS

Handheld size
Φ5x5cm



Portal monitor size
thicknessx10x40cm



Na & I have small $\sigma_{n\ abs}$,
∴ n_{th} detection efficiency grows strongly with [^{6}Li]

& n_{th} detection efficiency grows with thickness

THE TRUE ADVANTAGE OF NAIL DETECTORS

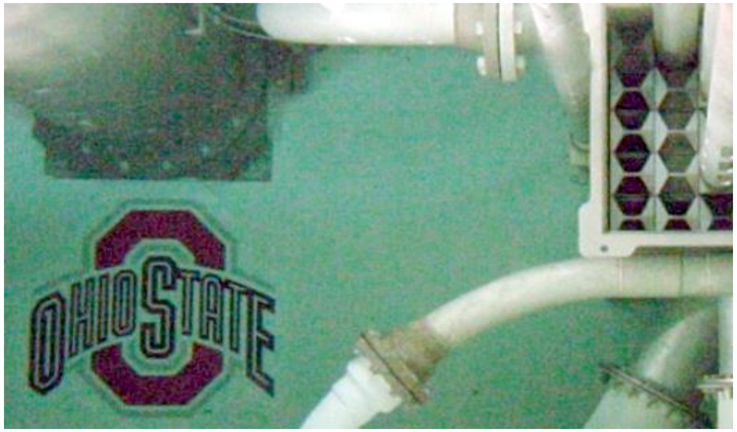
Fact 1: It is easy and inexpensive to grow large NaI crystals.

Fact 2: Na and I barely compete with ^{6}Li for neutron attenuation (0.53 and 6.25 vs. 940 barn).

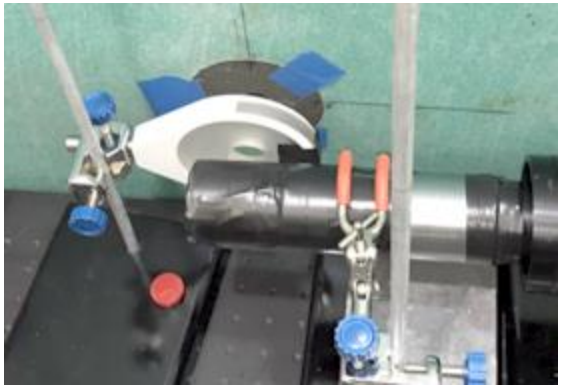
Advantage 1: The use of low ^{6}Li concentrations and large thicknesses can achieve the same neutron detection capabilities as ^{3}He or CLYC or CLLB detectors at a lower cost.

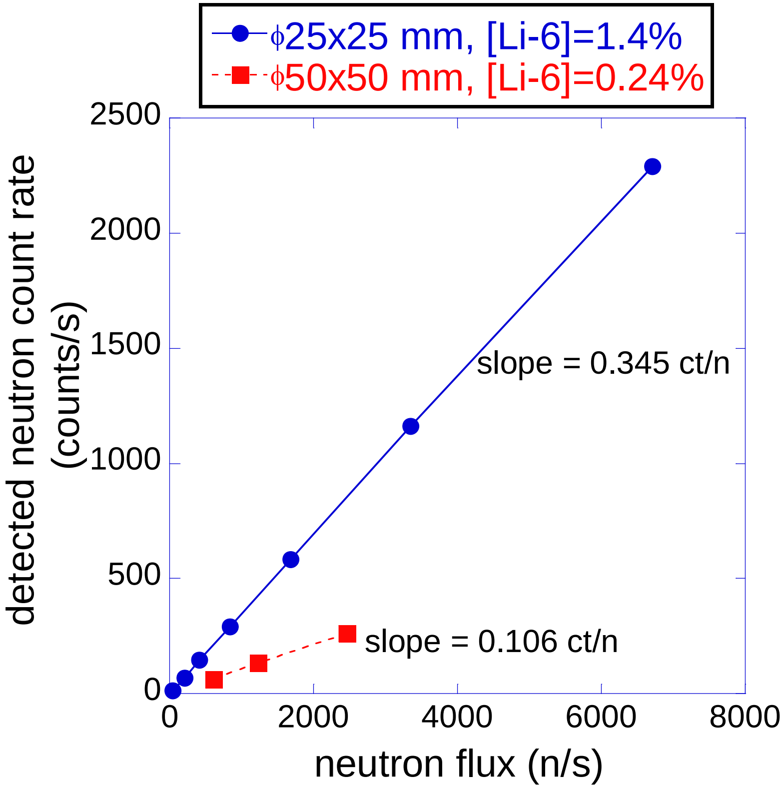
Advantage 2: Large volumes add efficient gamma ray detection capability as well.

NEUTRON EFFICIENCY IS AS EXPECTED



OSU reactor and neutron beam facility



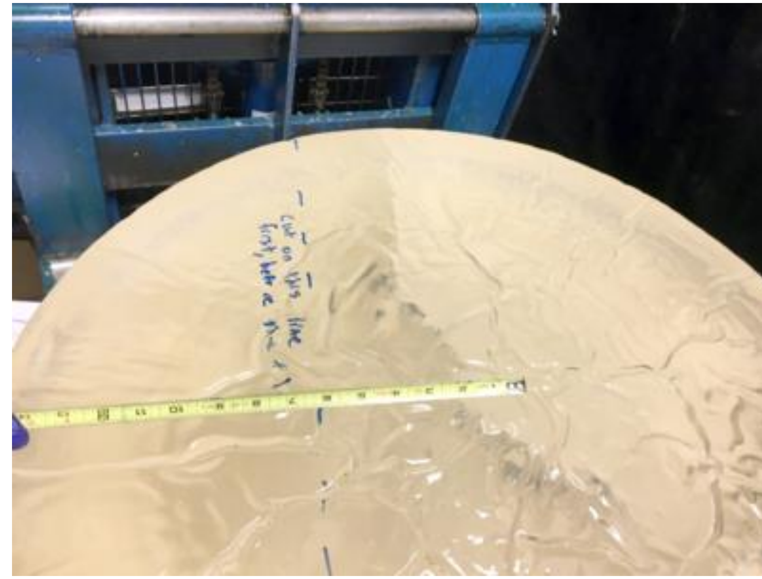


Size	$[^6Li]$ in crystal	thermal neutron ε	MCNPX ε prediction
ø2.5x2.5 cm	1.37%	34.5±0.2%	32.8%
ø5.1x5.1 cm	0.24%	10.6±0.3%	11.1%

ACHIEVING LARGE NAIL INGOTS

2015
Φ6.4 cm
250 cm³

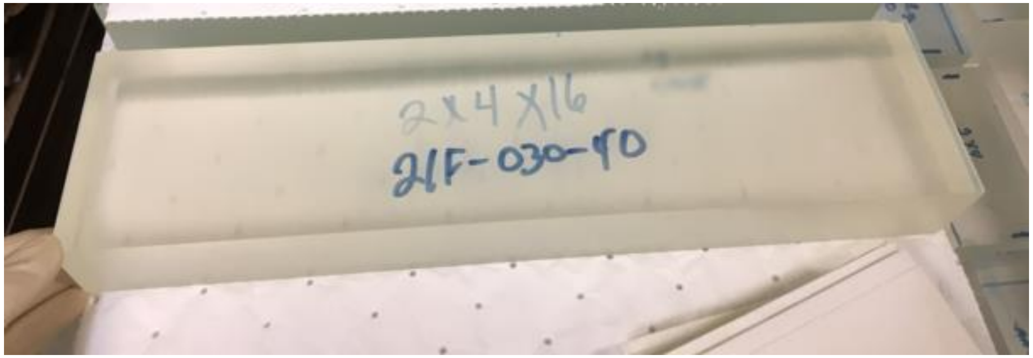
2016
Φ20cm NaIL
5800 cm³



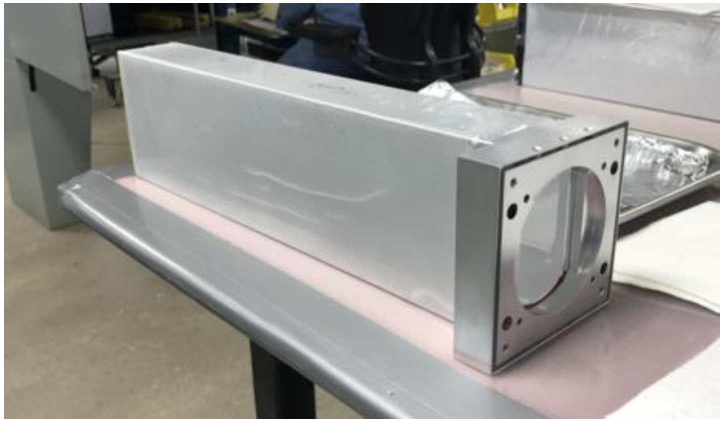
2017
Φ80cm NaIL
120000 cm³

A 5X10X40 CM³ WAS CHOSEN AS A LARGE CRYSTAL FOR TESTING

This is a popular large format

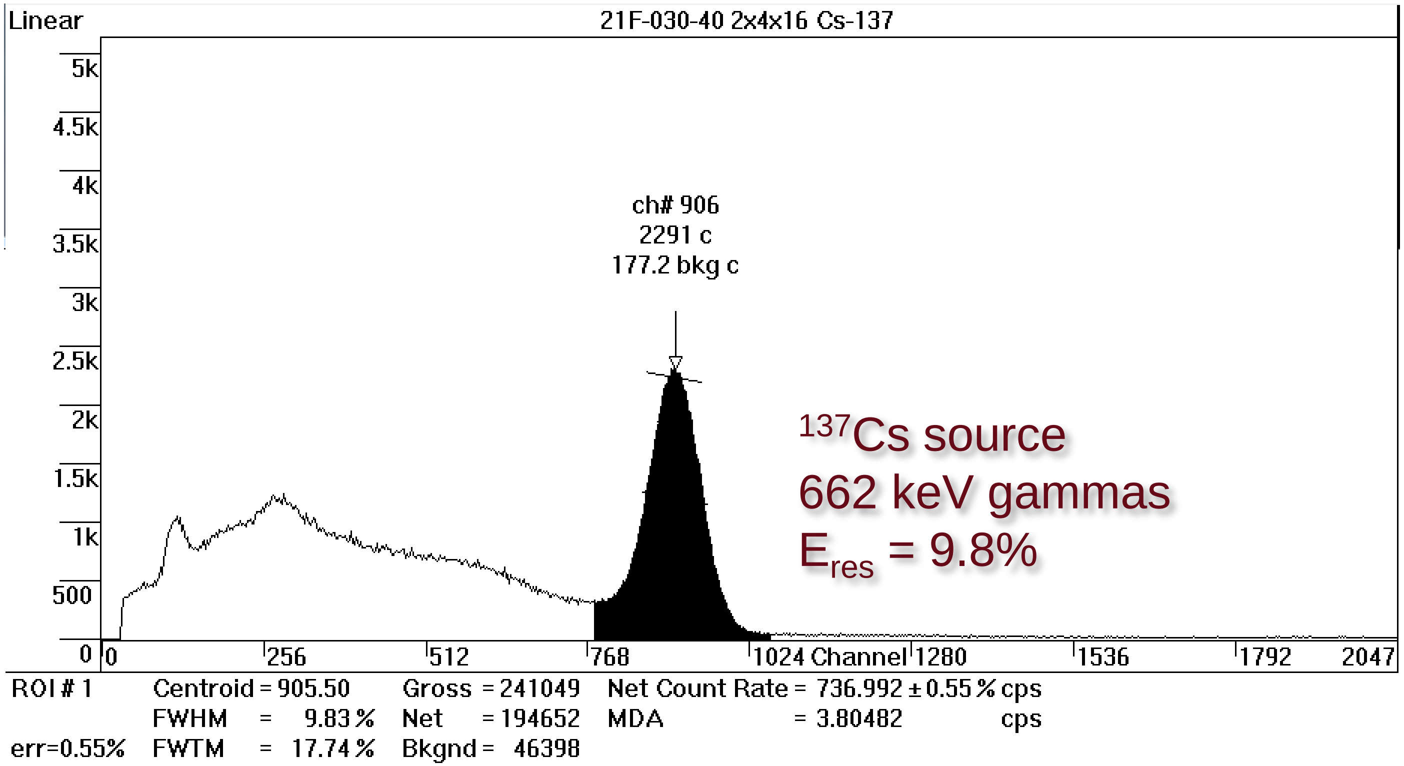


a cut 5x10x40 cm^3 NaIL crystal
[^{6}Li] = 0.37% (measured by ICP-OES)



in a hermetic housing

GAMMA TEST RESULTS: 5X10X40 CM³ DETECTOR



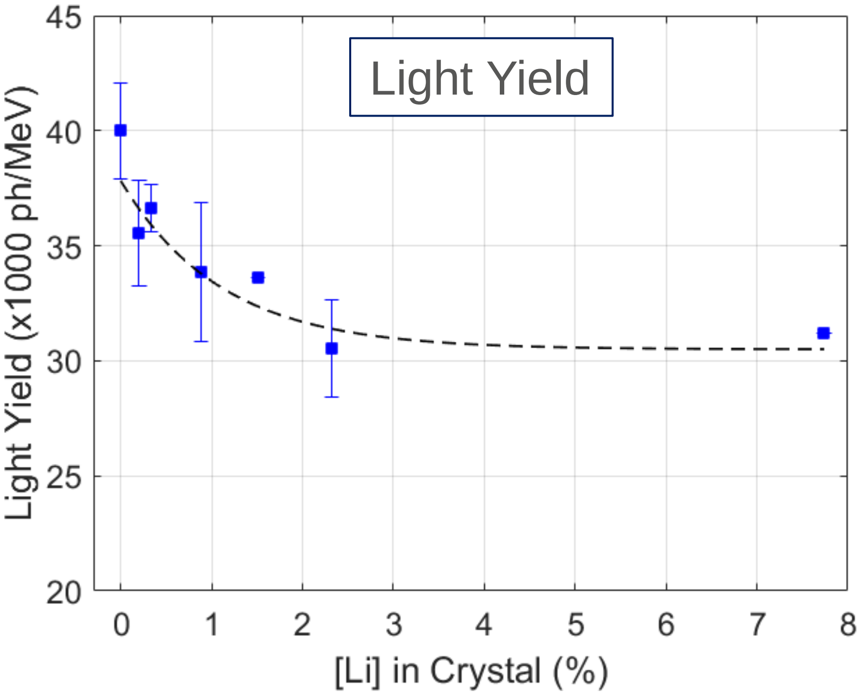
^{137}Cs source

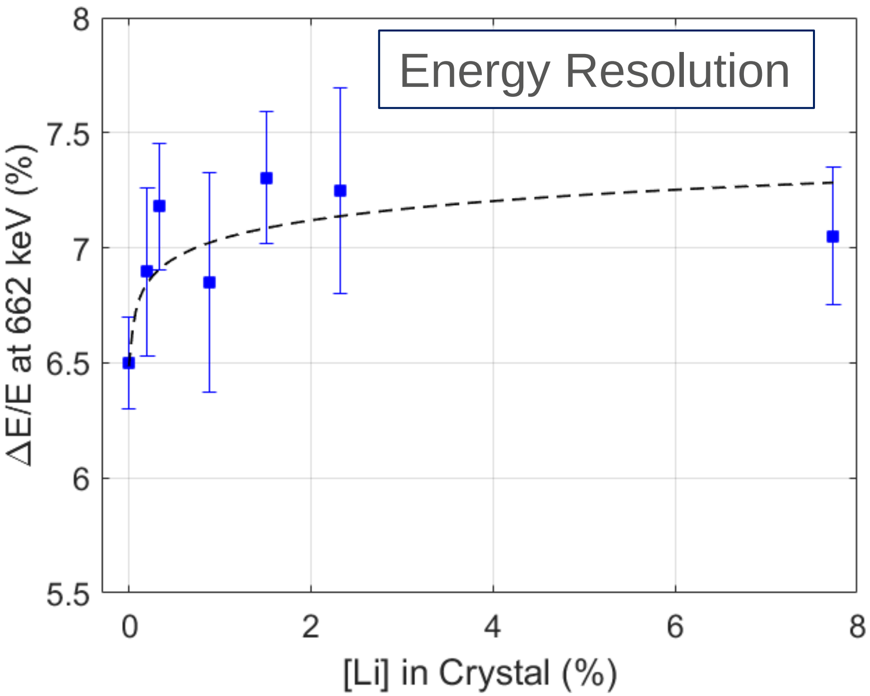
662 keV gammas

E_{res} = 9.8%

Typical NaI(Tl) of this size is 7.0 – 8.0%

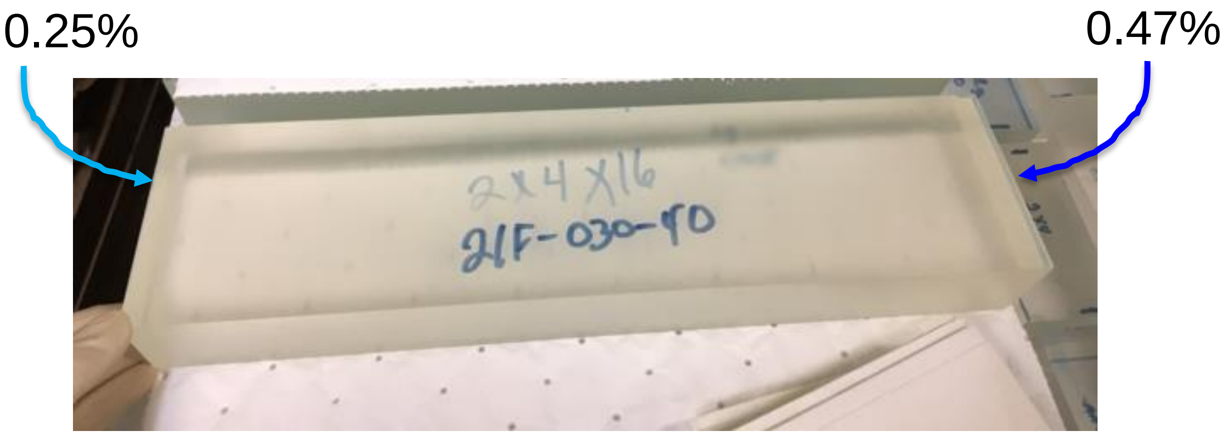
LIGHT YIELD AND ENERGY RESOLUTION DEPEND ON [LI]





- ❑ **Scintillation light yield decreases with [Li].**
  - **~ 34,000 ph/MeV @ 1% Li**
  - **~ 31,000 ph/MeV @ 2% Li and above**
  - **dependence is strongest at low [Li]**
- ❑ **Energy resolution is slightly increased.**

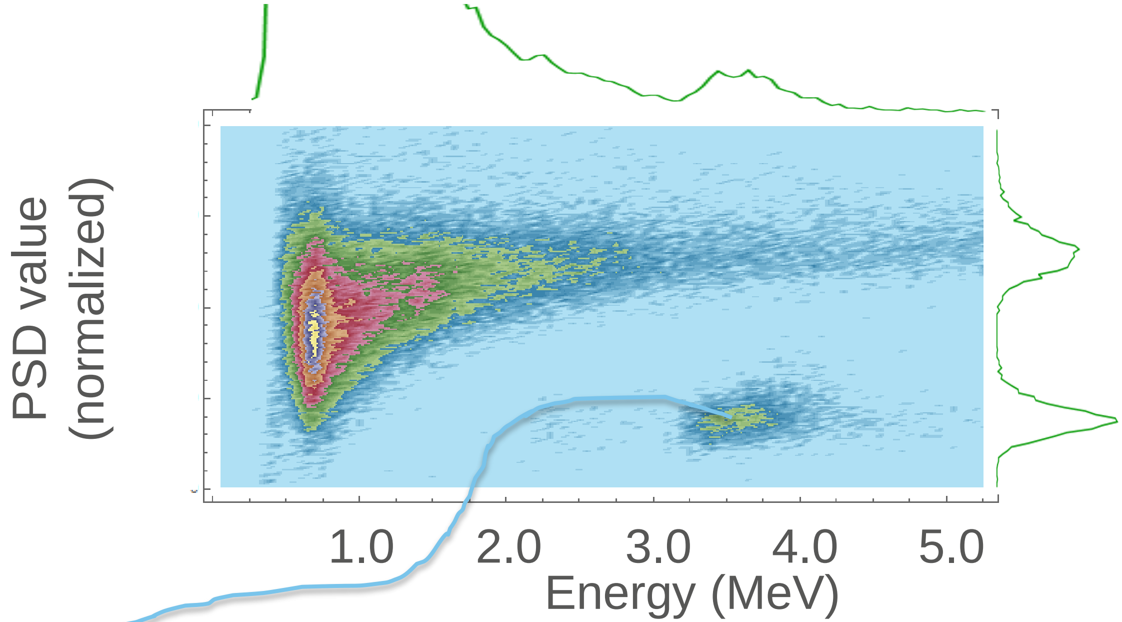
[Li] is not uniform - causing non-uniformity in light output



The difference in light yield is 4.5% from one end to the other.
This is the main source of degradation in energy resolution.

Future growths will utilize gradient reduction techniques
& higher lithium concentrations. Goal is <8% at 662 keV.

NEUTRON DETECTION CAPABILITY IS COMPLETELY ACCEPTABLE



moderated ^{252}Cf source
fission neutrons + gammas
PSD FoM = 2.0

net detection rate =
0.40 n/s/ng of ^{252}Cf at 2 m

^{3}He tube in Radiation Portal Monitors
Φ5x173 cm, 3 atm = ~3 n/s/ng of ^{252}Cf at 2 m

∴ Three of these NaIL detectors with [^{6}Li]=1-2% will have same n detection capability as one ^{3}He tube
at similar cost

SUMMARY AND OUTLOOK

❑ NaIL is the future for dual mode detection

- Spectroscopic, large volume & low cost

❑ First production scale NaI(Tl,^{6}Li) ingot grown (120 liter)

❑ First large detector (2 liter) fabricated

- neutron detection capability is excellent
- below par gamma energy resolution
  - fix with lower ∇[Li]
  - fix with overall higher [Li]

❑ Upcoming work

- Finalize optimized growth process by 2018Q1
- >70 crystals ∃ of various sizes to package for demos and sales
- Start offering NaIL at all sizes in 2018Q2