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

P.R. Menge, K. Yang, and V. Ouspenski
Saint-Gobain Crystals
Hiram, OH, USA
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:TI)
A BRIEF INTRODUCTION

- Invented by Dr. Robert Hofstadter in 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
Neutron detectors are important components in systems for illicit nuclear material interdiction:
- only fissile isotopes emit neutrons
- efficient, inexpensive neutron detectors are wanted (3He crisis)
- simultaneous detection of gamma rays and neutrons is also desired

Li can be incorporated into NaI:Tl to introduce thermal neutron detection:
- \( n + {}^6\text{Li} \rightarrow t \, (2.75 \text{ MeV}) + \alpha \, (2.05 \text{ MeV}) \)
- 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

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)
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

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

i.e. “tail-to-total”

\( \varnothing 2.5 \times 2.5 \text{ cm crystal, } [^{6}\text{Li}] = 0.6\% \)

moderated \(^{252}\text{Cf source} \)

Gammas

Neutrons

\( \varnothing 2.5 \times 2.5 \text{ cm crystal, } [^{6}\text{Li}] = 0.6\% \)
### APPLICATIONS FOR NAIL

<table>
<thead>
<tr>
<th>Target Application</th>
<th>Competing Technology</th>
<th>Advantage of NaI/L</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Radiation Portal Monitor</strong></td>
<td>Neutrons: $^3$He tubes, $^{10}$B tubes, $^6$LiF/ZnS films</td>
<td>Cost, Spectroscopic gamma, Dual mode</td>
</tr>
<tr>
<td></td>
<td>Gammas: Plastic scintillator</td>
<td></td>
</tr>
<tr>
<td><strong>Mobile</strong></td>
<td>Neutrons: as above</td>
<td>Cost, Size, Weight, Dual mode</td>
</tr>
<tr>
<td></td>
<td>Gammas: NaI:Tl</td>
<td></td>
</tr>
<tr>
<td><strong>Backpack</strong></td>
<td>Neutrons: as above</td>
<td>Cost, Size, Weight</td>
</tr>
<tr>
<td></td>
<td>Gammas: NaI:Tl</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Dual: $Cs_2^6LiLaBr_6$ (CLLB), $Cs_2^6LiYCl_6$ (CLYC)</td>
<td></td>
</tr>
<tr>
<td><strong>Handheld</strong></td>
<td>Neutrons: $^3$He $Cs_2^6LiYCl_6$ $^6$LiF/ZnS films</td>
<td>Cost, Size, Weight</td>
</tr>
<tr>
<td></td>
<td>Gammas: NaI, CsI, LaBr$_3$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Dual: $Cs_2^6LiLaBr_6$</td>
<td></td>
</tr>
</tbody>
</table>
A FEW % $^6\text{Li}$ DOPING IS SUFFICIENT FOR MOST SECURITY APPLICATIONS

Handheld size

$\Phi5\times5\text{cm}$

Portal monitor size

thickness$x10\times40\text{cm}$

Na & I have small $\sigma_n$ abs,

$\therefore n_{th}$ detection efficiency grows strongly with $[^6\text{Li}]$
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

<table>
<thead>
<tr>
<th>Size</th>
<th>(^{6}\text{Li} ) in crystal</th>
<th>thermal neutron ( \varepsilon )</th>
<th>MCNPX ( \varepsilon ) prediction</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \phi 2.5 \times 2.5 ) cm</td>
<td>1.37%</td>
<td>34.5±0.2%</td>
<td>32.8%</td>
</tr>
<tr>
<td>( \phi 5.1 \times 5.1 ) cm</td>
<td>0.24%</td>
<td>10.6±0.3%</td>
<td>11.1%</td>
</tr>
</tbody>
</table>

Detected neutron count rate (counts/s) vs. neutron flux (n/s)

- \( \phi 25 \times 25 \) mm, \([^{6}\text{Li}] = 1.4\%\)
- \( \phi 50 \times 50 \) mm, \([^{6}\text{Li}] = 0.24\%\)

Slope = 0.345 ct/n for \( \phi 25 \times 25 \) mm

Slope = 0.106 ct/n for \( \phi 50 \times 50 \) mm

MCNPX simulation results compared with experimental data.
ACHIEVING LARGE NAIL INGOTS

2015
Φ6.4 cm
250 cm³

2016
Φ20cm NaI/L
5800 cm³

2017
Φ80cm NaI/L
120000 cm³
A 5X10X40 cm$^3$ was chosen as a large crystal for testing

This is a popular large format

a cut 5x10x40 cm$^3$ NaI$\text{L}$ crystal

$[^6\text{Li}] = 0.37\%$ (measured by ICP-OES)
GAMMA TEST RESULTS: 5X10X40 CM$^3$ DETECTOR

$^{137}$Cs source
662 keV gammas
$E_{\text{res}} = 9.8\%$

Typical NaI(Tl) of this size is 7.0 – 8.0%
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.
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 =
$0.40 \text{ n/s/ng of } ^{252}\text{Cf at 2 m}$

$^3$He tube in Radiation Portal Monitors
$\Phi 5 \times 173 \text{ cm, 3 atm } = \sim 3 \text{ n/s/ng of } ^{252}\text{Cf at 2 m}$

∴ Three of these NaI(L) detectors with $[^{6}\text{Li}]=1-2\%$ 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 \(\nabla[Li]\)
    - fix with overall higher [Li]

- **Upcoming work**
  - Finalize optimized growth process by 2018Q1
  - >70 crystals \(\exists\) of various sizes to package for demos and sales
  - Start offering NaIL at all sizes in 2018Q2