Recent developments in neutron detection with scintillators: some new data on liquid scintillators for neutron / gamma discrimination

P. Schotanus
SCIONIX Holland B.V.
Dedicated Scintillation Detectors
www.scionix.nl

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Detection of Neutrons:

- Physics (e.g. particle Physics, HEP)
- Security (SNM e.g. Pu, U)
- Health Physics (dosimetry, often non spectrometric)

Neutron energy: Thermalised 0.025 eV – MeVs
(fast neutrons > 50 keV)

Interaction with Nucleus of absorber:

A. Scattering or
B. Nuclear reactions

- Elastic scattering (protons)
- Inelastic scattering prompt gammas

Nuclear Reactions e.g.: $^{10}\text{B}(n \alpha)^7\text{Li}$, $^3\text{He}(n p)^3\text{H}$, $^6\text{Li}(n \gamma)^3\text{H}$, $^{157}\text{Gd}(n \alpha)^{158}\text{Gd}$
Detection of neutrons with scintillators:

A. Thermal neutrons via nuclear reactions on Li, B or Gd in the material.

B. Fast neutrons via elastic (recoil) scattering in proton containing materials (organic scintillators)

Detection of neutrons with Gas Tubes:

Thermal(ised) neutrons with He-3 Tubes: Rather slow and low eff. (gas)

- Pressurized Gas tubes (e.g. 4-8 Bar to increase sensitivity)
- Used mainly in Health Physics and security
- Large lengths (meters) possible

Usually, neutrons associated with gammas, both will interact with most scintillators but:

Neutron + 6-Li $\rightarrow$ alpha + Triton (4.78 MeV total)

(particles !) $\rightarrow$ Peak
6-Li containing scintillators

1. (96 % enriched) 6-Li glass
2. (96 % enriched) 6-LiI(Eu)
   - 3 mm material sufficient to stop 90 % of thermal neutrons
   - Low density → Low gamma efficiency

Alpha/ gamma ratio determines location in gamma spectrum (GEE)

1. Li-glass : 1.8 MeV
2. LiI(Eu) : 3 – 4 MeV

Some more complex alternatives known such as Cs$_2$LiYCl$_6$ ; LiI has highest alpha gamma ratio known ( > 0.90).

Energy resolutions for the thermal neutron peak approx 3 –3.5 % FWHM
Advantages

96 % enriched 6-LiI(Eu)

High GEE (> 3 MeV)
High Eff.

96 % enriched 6-Li glass (Ce doped)

Fast decay time (60 ns)
non hygroscopic

Disadvantages

Size < 76 mm
Cost
Decay time 1.4 μs

Lower GEE (1.8 MeV)
Cost
No gamma ray spectr. (low Z)
low light output

Boron Loaded scintillators:
No inorganics known except for Li Borate glasses with low light output
FAST NEUTRON DETECTION (Physics)

Interaction with Protons (plastics, Liquids,): elastic recoil

ORGANIC SCINTILLATORS:
Manufactured by ELJEN Technology (USA)
(represented by SCIONIX in Europe)

**Physical Constants of Plastic Scintillators**

<table>
<thead>
<tr>
<th>Scintillator</th>
<th>Light Output, % Anthracene</th>
<th>Wavelength of Maximum Emission nm</th>
<th>Decay Constant Main Components</th>
<th>Typical Light Attenuation Length cm</th>
<th>H : C Atomic Ratio</th>
<th>Refractive Index</th>
<th>Softening Point °C</th>
<th>Density</th>
<th>Principal Applications</th>
</tr>
</thead>
<tbody>
<tr>
<td>EJ-200</td>
<td>64</td>
<td>425</td>
<td>2.1</td>
<td>380</td>
<td>1.104</td>
<td>1.58</td>
<td>75</td>
<td>1.02</td>
<td>Best overall properties, TOF Counters, large area, p+, e−, etc.</td>
</tr>
<tr>
<td>EJ-204</td>
<td>66</td>
<td>408</td>
<td>1.8</td>
<td>160</td>
<td>1.107</td>
<td>1.58</td>
<td>75</td>
<td>1.02</td>
<td>Good general properties</td>
</tr>
<tr>
<td>EJ-208</td>
<td>60</td>
<td>434</td>
<td>3.3</td>
<td>400</td>
<td>1.104</td>
<td>1.58</td>
<td>75</td>
<td>1.02</td>
<td>Large area, protons, electrons, etc.</td>
</tr>
<tr>
<td>EJ-212</td>
<td>65</td>
<td>423</td>
<td>2.4</td>
<td>250</td>
<td>1.103</td>
<td>1.58</td>
<td>75</td>
<td>1.02</td>
<td>General Purpose, Thin Films</td>
</tr>
<tr>
<td>EJ-228</td>
<td>67</td>
<td>391</td>
<td>1.4</td>
<td>NA</td>
<td>1.102</td>
<td>1.58</td>
<td>75</td>
<td>1.02</td>
<td>Very fast timing, high pulse pair resolution</td>
</tr>
<tr>
<td>EJ-230</td>
<td>64</td>
<td>391</td>
<td>1.5</td>
<td>NA</td>
<td>1.102</td>
<td>1.58</td>
<td>75</td>
<td>1.02</td>
<td>Variant EJ-228 optimized for detector dimensions over 10 cm</td>
</tr>
<tr>
<td>EJ-232</td>
<td>55</td>
<td>370</td>
<td>1.4</td>
<td>NA</td>
<td>1.102</td>
<td>1.58</td>
<td>75</td>
<td>1.02</td>
<td>Ultra fast timing</td>
</tr>
<tr>
<td>EJ-240</td>
<td>41</td>
<td>435</td>
<td>−230</td>
<td>240</td>
<td>1.109</td>
<td>1.58</td>
<td>75</td>
<td>1.02</td>
<td>Long decay time, phoswich detectors</td>
</tr>
<tr>
<td>EJ-248</td>
<td>54</td>
<td>425</td>
<td>2.1</td>
<td>380</td>
<td>0.995</td>
<td>1.59</td>
<td>100</td>
<td>1.049</td>
<td>Elevated temperatures, general purpose</td>
</tr>
<tr>
<td>EJ-252</td>
<td>46</td>
<td>423</td>
<td>2.4</td>
<td>NA</td>
<td>1.098</td>
<td>1.58</td>
<td>75</td>
<td>1.037</td>
<td>Dosimetry, air-equivalent</td>
</tr>
<tr>
<td>EJ-256</td>
<td>32</td>
<td>425</td>
<td>2.1</td>
<td>NA</td>
<td>1.134</td>
<td>1.58</td>
<td>75</td>
<td>1.08</td>
<td>Lead loaded [5% standard] x-rays, dosimetry</td>
</tr>
</tbody>
</table>

1 MeV of energy deposited in EJ-200 from an energetic electron produces approximately 10,000 blue photons.
We see : Data on organic scintillators rather old (1950-1960s)

Often reference is made to very old numbers (e.g. NE 213)

Light output refers to barely existing material (antracene) !!

emissions in the 400 nm region, decay times rather fast (ns)
It is time for some NEW data using modern PMTs

Neutron spectroscopy with organics materials using “Time of Flight methods” implies TIME RESOLUTION of ORGANICS VERY IMPORTANT

**Time resolution:**
Accuracy with which the interaction time of an event can be determined

Dependent of:
- decaytime (speed) scintillator
- photoelectron statistics (first few photoelectrons important)
- PMT characteristics (rise time, time jitter, light travel time differences)

\[
\Delta \tau = 2.36 \cdot \frac{1}{\sqrt{N}} , H(\tau_1,\ldots,\tau_n)
\]

So:
- We need large photoelectron yield
- Short decay time, rel. fast PMT
- Good geometry (lots of “religion” around)
With some organics (esp. Liquids), neutrons and gamma produce differently shaped pulses due to a different scintillation processes for electrons and (recoil) protons (based on ionization density).

**Differences between organics** mainly:
- Optical clarity
- Decay time(s)
- Chemistry
- Loading yes or no
- PSD yes or no

Some liquids are considered **dangerous goods** due to low “flash point” carriers Like Benzene or Xylene.
(Problem in transport etc.)
Different ways to do neutron/gamma separation with PSD

1. QDC with 20 ns and 1 microseconds gate

2. Converts signals to time spectrum using a double delay line amplifier and CFDs (time spectrum shows two peaks)

3. Digitize the signal using waveform analyzer (500 Mhz to 1 GHz flash ADC

Data on most popular scintillators for n-gamma discr: EJ301 and new EJ 309
EJ301 is a scintillating liquid **equivalent to NE213**

**BC501A** specially designed for neutron / gamma discrimination.

**Properties**:

- Light output (rel. to antracene): 78% 75%
- Photon yield / MeV electrons: 12,000 11,500
- Maximum of emission wavelength: 425 nm 424 nm
- Density: 0.874 g/ cc 0.964 g/ cc
- H:C ratio: 1.21 1.25
- No. C atoms per cc: 4.0 $10^{22}$ 5.46 $10^{22}$
- No. H atoms per cc: 4.810 $22$ 4.37 $10^{22}$
- No. electrons per cc: 2.3 $10^{23}$ 3.17 $10^{22}$
- Flash point: 26 °C 144 °C
- Refractive index: 1.50 1.57
- Decay time short component: 3.2 ns approx. 3.5 ns
- Decay time long components: 32.3, 270 ns ----
EXPERIMENTAL

Measurements of pulse shapes for gammas and neutron/gammas

Sources:
- Cs-137 (gammas)
- 10 cm Pb shielded Cf252 source (Mixed neutrons/gammas)

Cells:
- 127 x 76 mm EJ 301
- 51 x 51 mm EJ 309

Technique:
- Delayed coincidence Techn. (Böllinger+Thomas)
EJ301 Neutron/Gamma response

**EJ301 Neutron/Gamma response serie 2**

Data: A301neutrons_C
Model: ExpDec2
Equation: \( y = A1 \cdot e^{-x/t1} + A2 \cdot e^{-x/t2} + y0 \)
Weighting: y No weighting
\( \chi^2/\text{DoF} = 832.54554 \)
\( R^2 = 0.99741 \)
\( y0 \quad 1.68 \pm 0 \)
\( A1 \quad 115031.30342 \pm 1518.17064 \)
\( t1 \quad 4.19862 \pm 0.01604 \)
\( A2 \quad 95.73834 \pm 0 \)
\( t2 \quad 16.84528 \pm 0 \)

**Time (ns)**

**Intensity (Arb. units)**

---

**EJ301 Neutron/Gamma response serie 1**

Data: A301neutrons_B
Model: ExpDec2
Equation: \( y = A1 \cdot e^{-x/t1} + A2 \cdot e^{-x/t2} + y0 \)
Weighting: y No weighting
\( \chi^2/\text{DoF} = 1003.76584 \)
\( R^2 = 0.99918 \)
\( y0 \quad 2 \pm 0 \)
\( A1 \quad 85271.99203 \pm 11671.0856 \)
\( t1 \quad 4.07022 \pm 0.34266 \)
\( A2 \quad 87.85202 \pm 70.36332 \)
\( t2 \quad 37.53046 \pm 15.14327 \)
\( A3 \quad 13610.79067 \pm 15572.40382 \)
\( t3 \quad 6.64412 \pm 1.48075 \)

**Data: A301neutrons_B**
Model: ExpDec3
Equation: \( y = A1 \cdot e^{-x/t1} + A2 \cdot e^{-x/t2} + A3 \cdot e^{-x/t3} + y0 \)
Weighting: y No weighting
\( \chi^2/\text{DoF} = 1019.22163 \)
\( R^2 = 0.99917 \)
\( y0 \quad 2 \pm 0 \)
\( A1 \quad 1479.59108 \pm 408.46306 \)
\( t1 \quad 12.61838 \pm 1.19748 \)
\( A2 \quad 91112.482 \pm 1019.35949 \)
\( t2 \quad 4.46786 \pm 0.04515 \)

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**EJ 301 gammas only**

Components: 4.2 ns 99 %
16.84 ns < 1 %

(Literature 3.2, 32, 270 ns)

**EJ 301 gammas+neutrons**

Components: 4.1 ns 86 %
6.6 ns 13 %
37.5 ns < 1 %

(Literature 3.2, 32, 270 ns)
**EJ 309 Neutron/Gamma response serie 1**

Data: A309neutong_2006
Model: ExpDec2
Equation: \( y = A1 \times \exp(-x/t1) + A2 \times \exp(-x/t2) + y0 \)
Weighting: None

\[
\begin{align*}
\text{Chi}^2/\text{DoF} &= 128.88634 \\
R^2 &= 0.99981 \\
\end{align*}
\]

\[
\begin{align*}
y0 &= 1.000000 \\
A1 &= 637.20 \pm 145.94 \\
t1 &= 2.09 \pm 0.002 \\
A2 &= 32.12 \pm 3.38 \\
t2 &= 4.37 \pm 0.01 \\
\end{align*}
\]

**EJ 309 Neutron/Gamma response serie 2**

Data: A309neutong_2006C
Model: ExpDec3
Equation: \( y = A1 \times \exp(-x/t1) + A2 \times \exp(-x/t2) + A3 \times \exp(-x/t3) + y0 \)
Weighting: None

\[
\begin{align*}
\text{Chi}^2/\text{DoF} &= 107.33251 \\
R^2 &= 0.9997 \\
\end{align*}
\]

\[
\begin{align*}
y0 &= 1.200000 \\
A1 &= 1438.82 \pm 280.50 \\
t1 &= 0.26 \pm 0.03 \\
A2 &= 44.03 \pm 3.38 \\
t2 &= 2.74 \pm 0.01 \\
A3 &= 344.34 \pm 29.56 \\
t3 &= 13.64 \pm 0.64 \\
\end{align*}
\]

**Components:**

- EJ 309 gammas only
  - Components: 2.1 ns 95 % 4.4 ns 5 %

- EJ 309 gammas+neutrons
  - Components: 2.7 ns 99 % 12.5 ns 1 %

**Conclusions:**

- EJ 309 faster then EJ301
- Although self absorption for larger cells seems bigger (other data to be further investigated)
- Decay components different from old data (more study needed)
Design of Liquid cells for timing and PSD

General
1. Liquids expand (T) and expansion space has to be available (3-5% for temp. range –20 – +50 °C)

With properly designed windows and hardware, “bubble free” cells are possible (all orientations).

2. Optical windows of UV glass or quartz provide best optical transmission. (Boron Free requirement sometimes).

Readout
1. Classical readout with large PMTs is often NOT needed for good timing and/or PSD

2. PMT rise time not that crucial (few ns is O.K) but time jitter is.
Readout with smaller fast PMT provide good PSD

![Graph showing n-g spectrum EJ 301 127 x 76 mm readout 3" fast PMT]

Counts per Channel

Time 358 ps / channel

P/V = 14
fwhm gamma = 3.7 ns
Fwhm neutron = 8.3 ns
Delta = 24 ns

Larger volumes using small PMT also possible:

![SCIONIX logo]
25x25x8 cm  PSD liquid

P/V = 4.5
fwhm gamma = 5.7 ns
Fwhm neutron = 10.8 ns
Delta = 22 ns

Source in all cases 10mm Pb shielded Cf-252 source
These cells can be coupled to larger assemblies.

Larger cell with "NEW" EJ 309
Conclusions:

1. “Old” organics materials deserve renewed attention

2. Physical parameters should be remeasured (light output, decay times etc.)

3. Larger cells (even 50x50 cm) with few smaller PMTS are well suited to discriminate neutrons and gammas

4. 6-Li compounds for thermal neutrons are more and more employed and provide narrow (3 %) peaks at high energies for thermal neutrons

5. Very complex geometries are possible

Thanks to: Chuck Hurlbut (ELJEN)