Glass Scintillators

V. Berdnikov, H. Gan, T. Horn, I.L. Pegg
## Selection of Inorganic Scintillators

<table>
<thead>
<tr>
<th>Material/ Parameter</th>
<th>Density (g/cm³)</th>
<th>Melt. Point (°C)</th>
<th>Rad. Length (cm)</th>
<th>Moliere Radius (cm)</th>
<th>Refr. Index</th>
<th>Emission peak</th>
<th>Decay time (ns)</th>
<th>Light Yield (γ/MeV)</th>
<th>Rad. Hard. (krad)</th>
<th>Radiation type</th>
<th>Radiation type</th>
<th>Z Eff</th>
</tr>
</thead>
<tbody>
<tr>
<td>BaF₂</td>
<td>4.89</td>
<td>1280</td>
<td>2.03 2.06</td>
<td>3.10 3.40</td>
<td>1.50</td>
<td>300 220</td>
<td>650 0.9</td>
<td>16000 2000</td>
<td>&gt;50</td>
<td>Scint.</td>
<td>52.7</td>
<td></td>
</tr>
<tr>
<td>CeF₃</td>
<td>6.16</td>
<td>1460</td>
<td>1.70 1.68</td>
<td>2.41 2.60</td>
<td>1.62 1.68</td>
<td>340 300</td>
<td>5 30</td>
<td>2800</td>
<td>&gt;100</td>
<td>Scint.</td>
<td>50.8</td>
<td></td>
</tr>
<tr>
<td>(BGO)Bi₄Ge₃O₁₂</td>
<td>7.13</td>
<td>1050</td>
<td>1.12</td>
<td>2.23 2.30</td>
<td>2.15</td>
<td>480</td>
<td>300</td>
<td>8000 4000</td>
<td>&gt;1000</td>
<td>.98 scint.</td>
<td>.02 Č</td>
<td>83</td>
</tr>
<tr>
<td>(PWO)PbWO₄</td>
<td>8.30</td>
<td>1123</td>
<td>0.89 0.92</td>
<td>2.00</td>
<td>2.20</td>
<td>560 420</td>
<td>50 10</td>
<td>40 240</td>
<td>&gt;1000</td>
<td>.90 scint.</td>
<td>.10 Č</td>
<td>75.6</td>
</tr>
<tr>
<td>PbF₂</td>
<td>7.77</td>
<td>824</td>
<td>0.93</td>
<td>2.21</td>
<td>1.82</td>
<td>280 310</td>
<td>&lt;30</td>
<td>2-6</td>
<td>50</td>
<td>Pure Č</td>
<td>77</td>
<td></td>
</tr>
<tr>
<td>(BSO):CeBi₄Si₃O₁₂</td>
<td>6.80</td>
<td>1030</td>
<td>1.85</td>
<td>≈5</td>
<td>2.06</td>
<td>470 505</td>
<td>≈100</td>
<td>1000 4000</td>
<td>&gt;10</td>
<td>Scint.</td>
<td>75</td>
<td></td>
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<tr>
<td>(LSO):CeLu₂SiO₅</td>
<td>7.40</td>
<td>2050</td>
<td>1.14</td>
<td>2.07</td>
<td>1.82</td>
<td>420</td>
<td>40</td>
<td>30000</td>
<td>&gt;1000</td>
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<td>64.8</td>
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<td>(LYSO):Ce[LuY]₂SiO₅</td>
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<td>2050</td>
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<td>2.07</td>
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Identical Volume: X₀³
Crystals in EMCal: PbWO$_4$

- PbWO$_4$ material of choice for many EMCals – high density, fast response, large and granular solid angle, etc., but also limitations, e.g. hadron radiation damage

**temperature dependence of different scintillators**

![Graph showing temperature dependence of different scintillators]

PbWO$_4$ light yield temperature dependence: 2%/°C

**1.8 x 10$^{13}$p/cm$^2$ @ KVI**

PbWO$_4$ radiation resistance
Crystals in EMCal: PbWO$_4$

- Another consideration: expensive ($15-25$/cm$^3$) and manufacturing uncertainty
  - Despite progress (work with SICCAS and now also CRYTUR) still a struggle to work with vendors to get reliable PbWO$_4$ crystals that would be compatible with experiment requirements, e.g. NPS, EIC EMCal.
Glass-based Scintillators for Detector Applications

An alternative active calorimeter material that is more cost effective and easier to manufacture than, e.g. crystals

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<td>(BaO*2SiO₂):Ce glass</td>
<td>3.7</td>
<td>3.6</td>
<td>2-3</td>
<td>~20</td>
<td>22</td>
<td>72</td>
<td>450</td>
<td>&gt;100</td>
<td></td>
<td>10 (no tests &gt;10krad yet)</td>
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<td>(BaO*2SiO₂):Ce glass loaded with Gd</td>
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Also: (BaO*2SiO₂):Ce shows no temperature dependence
Premier materials science facility with unique capabilities and expertise in glass R&D.

Current R&D program includes:

- Nuclear and hazardous waste stabilization
- Glass and ceramic materials development
  - Formulation optimization
  - Characterization
  - Property-composition models
- Materials corrosion and characterization
- Off-gas treatment
- Water treatment, ion exchange
- Cements, flyash
- Geopolymers
- Biophysics
- Nano-materials
- Thermoelectrics
- Spintronics
- Scintillation detectors
The Vitreous State Laboratory – unique facility

- Designing, constructing and testing large glass production systems
  - VSL Joule Heated Ceramic Melter (JHCM) Systems:
    - The largest array of JHCM test systems in the US
    - The largest JHCM test platform in the US

PILOT SYSTEM SCALE-UP

DM10 and DM100 JHCM Systems at VSL

VSL DM1200 HLW Pilot Melter System

About 400,000 kg glass made from about 1 million kg feed
XAS Studies on Silicate Glasses

- Na: Na\(^{+}\)O\(_{3-7}\) : Na-O = 2.30 - 2.60 Å
- Mn: Mn\(^{2+}\)O\(_{4-5}\) : Mn-O = 2.07 Å, Mn-Mn = 3.48 Å
- Cu: Cu\(^{2+}\)O\(_{4}\) : Cu-O = 1.96 Å, Cu-Cu = 2.98 Å
- Sr: Sr\(^{2+}\)O\(_{4-5}\) : Sr-O = 2.53 Å
- Zr: Zr\(^{4+}\)O\(_{6-7}\) : Zr-O = 2.08 Å
- Mo: Mo\(^{6+}\)O\(_{4}\) : Mo-O = 1.75 Å
- Ag: Ag\(^{+}\)O\(_{2}\) : Ag-O = 2.10 – 2.20 Å
- I: I\((\text{Na},\text{I})\)_\(_{4}\) : I-Li = 2.80 Å, I-Na = 3.04 Å
- Re: Re\(^{7+}\)O\(_{4}\) : Re-O = 1.74 Å
- Bi: Bi\(^{3+}\)O\(_{3}\) : Bi-O = 2.13 Å
- S: S\(^{6+}\)O\(_{4}\) surrounded by networkmodifiers; S\(^{2-}\); S-S
- Cl: Cl-O = 2.70 Å; Cl-Cl = 2.44 Å; Cl-Na; Cl-Ca
- V: V\(^{5+}\)O\(_{4}\); minor V\(^{4+}\)O\(_{5}\) under reducing conditions
- Cr: redox sensitive: Cr\(^{6+}\)O\(_{4}\) Cr-O = 1.64 Å; Cr\(^{3+}\)O\(_{6}\) Cr-O = 2.00 Å; Cr\(^{2+}\)O\(_{4}\) Cr-O ~ 2.02 Å
- Tc: redox sensitive, Tc\(^{4+}\)O\(_{6}\) Tc-O = 2.00Å; Tc\(^{7+}\)O\(_{4}\) Tc-O = 1.75 Å; evidence of Tc-Tc = 2.56 Å in hydrated, altered glass
- Sn: Sn\(^{4+}\)O\(_{6}\) (minor Sn\(^{2+}\)O\(_{4}\)) Sn-O = 2.03 Å; Sn-Sn = 3.50 Å
- Al: Al\(^{3+}\)O\(_{4}\) : Al-O: 1.77 Å
- Si: Si\(^{4+}\)O\(_{4}\) : various polymerizations
- Zn: Zn\(^{2+}\)O\(_{4}\) : Zr-O: 1.96 Å, Zn-Si 2\(^{\text{nd}}\) nearest-neighbor evidence
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Also: (BaO*2SiO₂):Ce shows no temperature dependence

### Shortcomings of earlier work:

- Macro defects, which can become increasingly acute on scale-up
- Sensitivity to electromagnetic probes
Uniformity remains a concern – manufacturing process requires optimization – progress with new method at CUA/VSL/Scintilex

Sample made at CUA/VSL based on previous DSB:Ce work

Samples made at CUA/VSL/Scintilex with our new method
Glass scintillators being developed at VSL/CUA/Scintilex

- Optical properties comparable or better than PbWO₄
- Preliminary tests on radiation damage look promising
- Ongoing optimization work

### Light Yield

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<tr>
<th>Material/Parameter</th>
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<th>Sample 2</th>
<th>Sample 3</th>
<th>Sample 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Luminescence (nm)</td>
<td>420</td>
<td>440</td>
<td>440</td>
<td>440</td>
<td>440</td>
</tr>
<tr>
<td>Relative light output</td>
<td>1</td>
<td>35</td>
<td>16</td>
<td>23</td>
<td>11</td>
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Red: after irradiation with 1000 Gy at IPNO
Status of New Glass/Ceramic Scintillator Material

- Color optimization – progress with new method at CUA/VSL/Scintilex

New glass has improved properties

New Scintilex formulations

- More transparent
- Higher light output

Optimized Transmittance

Excitation with 160 keV photons
Status of New Glass/Ceramic Scintillator Material

- High dose radiation tests – progress with new method at CUA/VSL/Scintilex

VSL-Scintilex-G1

VSL-Scintilex-G2

VSL-Scintilex-G3

Before irradiation

After 2 min 160KeV Xray at >3k Gy/min

After additional 2-5 min 160 KeV Xray at >3k Gy/min
Outlook

- Finalize optimization of glass composition - patent
- Larger samples for full characterization tests
  - Transmittance
  - Light Yield
  - Radiation hardness
  - Decay kinetics
  - Other
- Monte Carlo simulations to determine compactness of detectors
- Possible prototype tests in Hall D this fall
Summary

- PbWO$_4$ crystals are ideal for precision EMCal, but also have limitations – and are expensive

- Glass-based scintillators are cost-effective alternative to crystals, in particular EMCal regions with relaxed resolution requirements
  - Initial small samples produced at CUA/VSL have 35x light yield of PbWO$_4$
  - New method also eliminates bubbles, a major problem in earlier work
  - Ongoing optimizations