Glass Scintillators for Homogeneous Calorimetry

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Outline

• Glasses in Calorimetry
  – OPAL ECAL (PbO glass Barrel and Endcap)
• Heavy Metal Fluoride Cerium-doped glasses
  – Historic work in 1990s (LHC motivated)
  – 21st Century work
• New Oxide Glasses
• Summary and outlook
Large Glass Calorimeters

• They *have* been built, at least for EM calorimetry, e.g. the OPAL experiment at LEP

Lead glass blocks approx 94×94 mm² in cross-section and of length up to 520 mm (25 $X_0$).

This picture shows half of the barrel ECAL. The barrel contained 9440 individual blocks.

These used Cherenkov light to generate the signal.
Heavy Metal Fluoride (HMF) Glasses

- High(ish) Density (up to \( \sim 6 \text{ g.cm}^{-3} \))
- Low refractive index (\( n \sim 1.5 \)) compared to typical dense oxide glasses (\( n \sim 2.0 \)).
- Excellent transparency well into the UV (limited by Cerium if present as luminescent centre). Good for Cherenkov light.
- Some commercial manufacturing & some commercial applications.
- Considerable work in the 1990’s (SSC/LHC experiments). Emphasis here on fast scintillators.
Scintillating HMF Glasses

• “ZBLAN” composition well developed and understood from work on optical fibres. Substitute Hf for Zr and add approx 5% CeF$_3$

• Hafnium is key change and increases density to $\sim$ 6 g.cm$^{-3}$

• Cerium in the 3+ state added as a luminescent centre, fairly fast decay time (10’s of ns).

• Block size limited by crystallisation.

• Cost of raw materials, issue of tolerated vs critical impurities.
"Crystal Clear" Collaboration


Transmission (%)

T = 295 K

Intensity (a.u.)

wavelength (nm)

sample #
HFG796 (5%Ce)

radiation induced abs.coeff. ()

counts

\[ \lambda = 325 \text{ nm} \]

\[ \lambda = 500 \text{ nm} \]

\[ \lambda = 600 \text{ nm} \]

\[ \tau_1 = 1 \text{ ns} \]

\[ \tau_2 = 18 \text{ ns} \]

\[ \tau_3 = 40 \text{ ns} \]
Improving Scintillation Yield

- Cerium in silicate glasses (e.g. GS1) is a reasonable scintillator, somewhat better than pure CeF$_3$ crystal, but low density.
- Ce can only be added in small quantities to HBLAN composition thus low scintillation yield.

<table>
<thead>
<tr>
<th>Scintillator</th>
<th>Density g.cm$^{-3}$</th>
<th>$\lambda$ emission (nm)</th>
<th>Lifetime (ns)</th>
<th>Relative yield</th>
</tr>
</thead>
<tbody>
<tr>
<td>CeF$_3$</td>
<td>6.2</td>
<td>310, 340</td>
<td>5, 30</td>
<td>100%</td>
</tr>
<tr>
<td>GS1 glass</td>
<td>2.6</td>
<td>395</td>
<td>60</td>
<td>140%</td>
</tr>
<tr>
<td>Hf HMF glass</td>
<td>5.7</td>
<td>320</td>
<td>20</td>
<td>38%</td>
</tr>
<tr>
<td>Fluorophosphate</td>
<td>4.0</td>
<td>370</td>
<td>30</td>
<td>60%</td>
</tr>
</tbody>
</table>
UK Work (mid 1990’s)

- Shaukat S F *et al* Journal of Non-Crystalline Solids 244 (1999) 197-204
Indium doping provides significant enhancement in radiation tolerance

The effect of InF₃ doping on the light yield and radiation hardness of a glass with basic composition 'HBCeAN', containing 5% (molar) CeF₃. Curve I (triangles) shows the variation of light yield versus InF₃ concentration. The yield is normalised to the yield obtained from a crystal of CeF₃ of similar dimensions to the glass samples. Curve II (open circles) shows the variation of radiation-induced optical attenuation at 350 nm, measured immediately after a dose of 2.4 kGy. (Co⁶⁰-γ). Curve III (crosses) shows the optical attenuation induced by a dose of 7 kGy, followed by 4.5 months storage in the dark at room temperature.
Beam Test Data

- One of the very few tests in a particle beam reported in the literature. HMF block size is $20 \times 30 \times 140 \text{ mm}^3$
- Protons of 750 MeV/c momentum laterally exciting the block.

HBLAN with Ce has physical properties very similar to CeF$_3$ crystal, but with lower scintillation yield. 5%Ce doped HBLAN produced about 1/16 the yield of pure CeF$_3$.
High energy electron beam-test data

$30 \times 30 \times 130 \text{ mm}^3 \ (8.2 \ X_0)$ long block. Top is data, bottom is MC simulation.
Industrial Scale HMF Glass Production

- Two key challenges: Bulk glass dimensions & affordability

- In the UK we engaged with a major European producer of hafnium, which is a by-product of the production of nuclear grade zirconium. **Tonne** quantities of Hf per annum are produced.

- Our research showed that the Ce$^{3+}$ light output dropped approximately linearly to zero as Zr substituted for Hf. Thus a few % Zr in HfF$_4$ is tolerable. The dominant cost in making very pure HfF$_4$ is the removal of Zr and Ti.

- Working with chemists at Johnson Matthey UK we showed that a fairly inexpensive purification stage would further reduce critical impurities (e.g. transition metals) to an acceptable level.

- We could see that a HMF glass raw material cost of ~ 1$/cm^3$ was possible

Both we and Crystal Clear Collaboration (working with the company Le Verre Fluoré) demonstrated that “moderate” blocks of Ce-doped HMF glass can be made but although we got close to what was needed for an ECAL cell size it is a long way off what is needed for HCAL.
One of our largest Ce-doped HBLAN glass blocks!
More recent activity

Work on cerium doped glasses does continue but at a lower level that 10 years ago.

Work on redox conditions, other additives, mixed fluoride/chloride systems, RE co-dopants, etc.


HM Oxide glasses

• Commercial oxide glasses with Cerium for radiation tolerance do exist (e.g. for “hot cell” windows).
• Some major commercial producers of oxide glasses (including moderately dense ones) e.g. Hoya, Schott, Corning.
• Some recent interesting work on dense oxide scintillating glasses has been published.
• A wider range of compositions is under study compared to the HBLAN (Ce) of HMF e.g.
  – ZnO in zinc barium silicate glasses (possible quantum-dot effects)
  – SnO$_2$ in phosphate glasses
  – Ce doped germanate glasses in the GeO$_2$-Gd$_2$O$_3$-BaO system
  – Tb doped silico-germanate glasses
Oxide glasses

SnO$_2$ in phosphate glasses

Shen CE et al, ASTROPARTICLE, PARTICLE AND SPACE PHYSICS, DETECTORS AND MEDICAL PHYSICS APPLICATIONS
Proceedings of the 11th Conference, 2009

Oxide Glasses

Tb doped oxide glasses containing Ln$_2$O$_3$ where Ln = Y, Gd, Lu luminesce under X-ray excitation and have densities ~6 g.cm$^{-3}$.

Conclusion and Prospects

Quite extensive work on HMF glasses has led to an optimum composition using HfF$_4$ and Ce$^{3+}$ as the scintillating ion, adding In$^{3+}$ significantly improves radiation tolerance (other ions have been evaluated).

It is expensive to remove some elements (Zr, Ti) but these can be tolerated at the ~1% level. We believe that a raw material cost of order 1$/cm^3$ is probably attainable commercially (EU economic costing).

Making blocks with smallest dimension > 30 mm was not demonstrated in the 1990’s and may not be achieved.

Some work on new oxide glass compositions has continued, but issues of radiation tolerance persist.

Dense oxide glasses have high refractive indices which is a disadvantage compared to fluorides.

Terbium doped gadolinium oxide glasses show some promise.

Possibly nano-crystals (e.g. ZnO) in radiation tolerant oxide glasses may be a way forward.