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Abstract. Future HEP experiments at the energy and intensity frontiers require fast and ultrafast inorganic scintillators with excellent radiation hardness to face the challenges of unprecedented event rate and severe radiation environment. This paper reports recent progresses in fast and ultrafast inorganic scintillators, such as LYSO:Ce crystals and LuAG:Ce ceramics for an inorganic scintillator based shashlik sampling calorimeter and yttrium doped BaF$_2$ crystals for the proposed Mu2e-II experiment. Applications of ultrafast inorganic scintillators in Gigahertz hard X-ray imaging will also be discussed.

1. Introduction
Inorganic scintillators have been used widely in high energy and nuclear physics experiments, medical instruments and homeland security applications. In high energy physics (HEP) and nuclear physics experiments, total absorption electromagnetic calorimeters made of inorganic crystals are known for their superb energy resolution and detection efficiency for photon and electron measurements [1]. An inorganic crystal calorimeter is thus the choice for those experiments where precision measurements of photons and electrons are crucial for their physics missions.

Among all existing crystal calorimeters, the CMS lead tungstate (PbWO$_4$ or PWO) crystal calorimeter, consisting of 75,848 crystals of 11 m$^3$, is the largest. Because of its superb energy resolution and detection efficiency, the CMS PWO calorimeter has played an important role for the discovery of the Higgs boson by the CMS experiment [2]. Crystal calorimeters currently under construction are: an undoped CsI calorimeter for the Mu2e experiment at Fermilab, a PWO calorimeter for PANDA at FAIR, a LYSO calorimeter for COMET at JPARC and a PbF$_2$ calorimeter the g-2 experiment at Fermilab.

Future HEP calorimeters will be operated under unprecedented luminosity. An important issue is thus the decay time of scintillation light. Table 1 lists the optical and scintillation properties for fast inorganic crystal scintillators with a scintillation decay time ranged from sub-nanosecond to a few tens nanosecond, and compared to plastic scintillator [1]. Among the fast crystals listed in Table 1 the mass-production cost of barium fluoride (BaF$_2$) and undoped CsI crystals is significantly lower than others because of their low raw material cost and low melting point.

Crystal calorimeters for future HEP experiments at the energy frontier face a challenge of severe radiation environment. Significant losses of light output have been observed in the CMS PWO crystals at large rapidity in situ at the LHC caused by both ionization dose and hadrons [3]. Controlling oxygen contamination in halide crystals, e.g. CsI:TI, or oxygen vacancies in oxide crystals, e.g. PWO, was found effective [4]. Co-doping with yttrium and lanthanum was also found effective for CMS PWO crystals [5]. For experiments to be operated at the HL-LHC with 3,000 fb$^{-1}$, crystals should survive an environment with an absorbed dose of 100 Mrad, charged hadron fluence of $6\times10^{14}$ p cm$^{-2}$ and fast
A neutron fluence of $3 \times 10^{15}$ n cm$^{-2}$. To mitigate this challenge, efforts have been made to reduce the light path length in inorganic scintillators by designing an inorganic scintillator-based shashlik sampling calorimeter [6]. On the other hand, radiation damage in various inorganic scintillators has been investigated for an ionization dose up to 340 Mrad [7] and a fluence of protons up to $3 \times 10^{15}$ p cm$^{-2}$ [8]. Progress on neutron induced radiation damage up to $3.6 \times 10^{15}$ n cm$^{-2}$ was also investigated [9]. BaF$_2$, GAGG, LuAG and LYSO based inorganic scintillators are found to be radiation hard for the HL-LHC. Following these investigations, a LYSO MIP timing detector has been proposed for the CMS upgrade for the HL-LHC [10].

Table 1. Basic Properties of Fast Inorganic Crystal Scintillators

| Scintillator | Density (g/cm$^3$) | Melting point (°C) | Radiation Length (cm) | Molière Radius (cm) | Interaction Length (cm) | Z value | $d(C)/dT$ (MeV/cm) | Emission Peak (nm) | Refractive Index | Relative Light Yield | Decay Time (ns) | $d(LY)/dT$ (%/°C) |
|-------------|-------------------|--------------------|----------------------|-------------------|-----------------------|--------|----------------|-----------------|----------------|------------------|---------------|----------------|
| LSO/LSO     | 7.40              | 6.71               | 4.44                 | 4.51              | 4.89                  | 6.16   | 5.23           | 3.86            | 5.29           | 1.03             | -             | -              |
| GSO         | 1950              | 1980               | 621                  | 1280              | 1460                  | 722    | 858            | 783             | 79             | -                | -             | -              |
| YSO         | 1.14              | 1.38               | 3.11                 | 1.86              | 2.03                  | 1.70   | 1.96           | 2.81            | 1.88           | 42.54            | -             | -              |
| BaF$_2$     | 2.07              | 2.23               | 2.93                 | 3.57              | 3.10                  | 2.41   | 2.97           | 3.71            | 2.85           | 9.59             | -             | -              |
| CaF$_2$     | 20.9              | 22.2               | 27.9                 | 39.3              | 30.7                  | 23.2   | 31.5           | 37.6            | 30.4           | 78.8             | -             | -              |
| CsI         | 64.8              | 57.9               | 33.3                 | 54.0              | 51.6                  | 50.8   | 45.6           | 47.3            | 45.6           | -                | -             | -              |
| BaCl$_2$    | 9.55              | 8.88               | 6.56                 | 5.56              | 6.52                  | 8.42   | 6.65           | 5.27            | 6.90           | 2.02             | -             | -              |
| CsI         | 420               | 430                | 420                  | 310               | 300                   | 220    | 340            | 335             | 356           | 408              | -             | -              |
| YSO         | 1.82              | 1.85               | 1.80                 | 1.95              | 1.50                  | 1.62   | 1.9            | 1.9             | 1.9            | 1.54             | -             | -              |
| CsI         | 100               | 45                 | 76                   | 4.2               | 4.8                   | 8.62   | 141            | 15              | 153           | 35              | -             | -              |
| BaF$_2$     | 40                | 73                 | 60                   | 30                | 650                   | 9.0    | 17             | 570             | 24             | 1.8              | -             | -              |
| CsI         | -0.2              | -0.4               | -0.3                 | -1.4             | -1.9                   | -0.1   | -0.1           | 0.1             | 0.2            | -0              | -             | -              |

Another challenge for future HEP experiments at the intensity frontier, such as Mu2e-II, where the event rate, and thus the radiation dose, will be increased by a factor of ten as compared to the Mu2e-I experiment [11]. Such a fast rate requires faster and radiation harder inorganic scintillators to mitigate the pile-up effect. R&D aimed at developing ultrafast inorganic scintillators has been pursued. Yttrium doping in BaF$_2$ crystals was found to be effective to suppress the slow scintillation component in BaF$_2$ while maintaining its sub-ns fast scintillation component [12]. ZnO nano-particle imbedded in polystyrene was also reported with a sub-ns decay time [13]. An interesting investigation along this direction is to combine confined excitons and bi-excitons into a form of nanocrystals in inorganic scintillators [14].

For HEP experiments at future lepton colliders, inorganic scintillators have also been proposed to build a Homogeneous Hadron Calorimeter (HHICAL) to achieve unprecedented jet mass resolution by dual readout of both Cherenkov and scintillation light [15,16]. For this application, development of cost-effective crystal detectors is a crucial issue because of the huge crystal volume required, while the requirements on radiation hardness is much relaxed because of the lepton collider environment [17]. Investigation along this line has been concentrated on developing cost-effective UV transparent inorganic scintillators, including crystals and glasses. Progress on UV transparent cerium-doped and co-doped fluorophosphate glasses was reported in several conferences [18, 19].
2. Fast and Radiation Hard LuAG:Ce Ceramics

To face the challenge of severe radiation environment at the HL-LHC, an LYSO/W/quartz capillary-based shashlik calorimeter concept was proposed. As shown in Fig. 1, it consists of 1.5 mm LYSO plates as active material interleaved with 2.5 mm tungsten plates as absorbers, four quartz capillaries as wavelength shifters, and one quartz leaky fiber for injection of monitoring light [6].

![Figure 1 An LYSO/W/Quartz Capillary-based shashlik sampling calorimeter concept.](image)

With a density of 6.73 g/cm$^3$, a fast scintillation with a decay time of about 50 ns and a high light yield of 26,000 photons/MeV cerium-doped Lu$_3$Al$_5$O$_{12}$ aluminium garnet (LuAG:Ce) is another attractive bright and fast scintillator [6]. Compared to crystals the fabrication process for scintillating ceramics features a lower temperature, a more uniform doping distribution and a more effective use of raw materials. LuAG:Ce ceramics thus is a cost-effective alternative for LYSO:Ce crystals for the proposed shashlik calorimeter concept.

![Figure 2. Optical and scintillation properties of LuAG:Ce ceramic samples](image)

The left and middle plots of Fig. 2 show respectively the transmittance spectra and the light output as a function of integration time for LuAG:Ce ceramic samples produced at Shanghai Institute of Ceramics (SIC) [20]. While their light yield and decay time are compatible with LYSO:Ce crystals, these LuAG:Ce ceramic samples have also a slow scintillation component with a decay time of about a µs. Efforts have been made to suppress the slow component by varying cerium doping level and introducing various co-dopings [20]. The right plot of Fig. 2 shows a relationship between the light outputs in 200 ns (Fast) and 3,000 ns (Total) gate for LuAG:Ce ceramics with various mono- and...
divalent co-dopings. Li+ co-doping reduces both the F/T ratio and the LO of 200 ns gate; Mg2+ co-doping improves the F/T ratio and shows the highest LO in 200 ns gate; and Ca2+ co-doping is found effective with the highest F/T ratio ~90% [20].

Figure 3. Ionization dose and proton induced radiation damage in LuAG:Ce ceramic samples.

Fig. 3 shows excellent radiation hardness of LuAG:Ce ceramics. The left and middle plots of Fig. 3 show no visible damage in transmittance and light output respectively for two LuAG:Ce plate samples before and after an ionization dose of up to 220 Mrad plus an 800 MeV proton fluence of up to $2.9 \times 10^{15}$ p cm$^{-2}$. The right plot of Fig. 3 shows the numerical values of the normalized emission weighted longitudinal transmittance (EWLT) (top) and light output (bottom) as a function of ionization dose for two LuAG:Ce ceramic samples. It is clear that LuAG:Ce ceramics is radiation hard against the ionization dose and protons, so is a cost-effective alternative to replace LYSO for future HEP experiments in a severe radiation environment, such as the HL-LHC. R&D along this line will continue to investigate its radiation hardness against neutrons, and to develop radiation hard LuAG ceramic scintillators with suppressed slow scintillation component for future HEP experiments.

3. Ultrafast BaF2:Y Crystals

BaF$_2$ is a classical inorganic scintillator featured with an ultrafast cross-luminescence with sub-ns decay time. The left and middle plots of Fig. 4 show the emission and pulse shape of its ultrafast scintillation component peaked at 220 nm with sub-ns decay time, as well as a slow scintillation component peaked at 300 nm with 600 ns decay time and a five times brightness of its fast component. The slow component would cause pileup in an environment of fast event rate. Two approaches were used to reduce the slow component: selective doping with rare earth (La, Y, and Ce) [21] and selective readout with solar-blind photodetector [22] as shown in the right plot of Fig. 4.

Figure 4. Fast and slow scintillation in BaF$_2$ and slow suppression.
Yttrium doping in BaF$_2$ was found to be effective in suppressing the slow component. Fig. 5 shows x-ray excited emission (left) and transmittance (middle) spectra and light output as a function of integration time for BaF$_2$ cylinders of $\Phi$19×21mm grown at Beijing Glass Research Institute (BGRI) with yttrium doping levels from zero to 5 at% [12]. It is clear that yttrium doping is effective in suppressing the slow scintillation component while maintaining the fast component unchanged.

Figure 5. Optical and scintillation properties of BaF$_2$:Y samples.

Fig. 6 compares pulse shape measured by a Photek MCP-PMT 240 and an ultrafast 23 GHz Tektronix MSO 72304DX scope for Cherenkov light from a 5 cm PbF$_2$ cube sample (left) grown at SIC and ultrafast scintillation light from $\Phi$10×10 mm BaF$_2$ (middle) and yttrium doped BaF$_2$ (BaF$_2$:Y, right) samples grown at BGRI. It is interesting to note that all these light pulses show an ultrafast rise time of 0.2 ns and a decay time of about 0.6 ns, indicating that the ultrafast scintillation light from BaF$_2$ and BaF$_2$:Y is as fast as Cherenkov light from PbF$_2$. The data also confirm that yttrium doping in BaF$_2$ maintains the intensity of the ultrafast light. By using Photek MCP-PMT, BaF$_2$ crystals provides a sub-ns FWHM pulse width, which is unmatched by any other inorganic scintillators. Such ultrafast inorganic scintillator may also find applications in GHz hard X-ray imaging [23] and time of flight PET imaging, in addition to HEP and nuclear physics experiments.

Figure 6. Ultrafast pulse shape in PbF$_2$ (left), BaF$_2$ (middle) and BaF$_2$:Y (right).

4. Radiation Hardness of Fast Inorganic Scintillators

All known inorganic scintillators suffer from damage induced by ionization dose [1, 7] as well as charged [8] and neutral hadrons [9]. There are three possible radiation damage effects in crystal scintillators: (1) scintillation mechanism damage, (2) radiation induced phosphorescence (afterglow) and (3) radiation induced absorption (color centers). A damaged scintillation mechanism would reduce scintillation light yield and cause a degradation of light output. It may also change light response uniformity along a long crystal length since the radiation dose profile is usually not uniform. The radiation induced phosphorescence, commonly called afterglow, causes an increase of the dark current.
in the photo-detectors, and thus an increased readout noise. The radiation induced absorption reduces the light attenuation length [24], and thus the light output and possibly also the light response uniformity. There is so far no experimental evidence for a scintillation mechanism damage. All crystal scintillators, however, suffer from the radiation induced absorption and phosphorescence by ionization dose as well as charged and neutral hadrons.

**Figure 7.** Ionization dose induced radiation damage in large size inorganic scintillators

Ionization dose induced radiation damage was investigated for large size (about 200 mm long) inorganic scintillators may be used to construct total absorption crystal calorimeter [7]. The left plot of Fig. 7 shows normalized EWLT (top) and light output (bottom) as a function of ionization dose for the fast component in three BaF$_2$ crystals grown at different vendors: SIC, BGRI and Incrom. The average values of EWLT and light output are more than 40% after 120 Mrad. The middle and right plots of Fig. 7 compare the values of the radiation induced absorption coefficient (RIAC) at the emission peak and the normalized light output as a function of the integrated dose for various crystals. LYSO clearly shows the best radiation hardness among all crystal scintillators with its light output maintained at 75% and 60% respectively after 120 and 340 Mrad. On the other hand, the light output of BGO and BaF$_2$ is maintained at 35% and 45% respectively after 200 and 120 Mrad, so may be considered as cost-effective alternatives for LYSO.

**Figure 8.** Proton induced radiation damage in LYSO, BaF$_2$ and PWO crystals.

Protons were found to cause radiation damage in inorganic scintillators [8]. The left and middle plots of Fig. 8 show respectively the transmittance spectra and the light output as a function of integration time (middle) for BaF$_2$ plates of $25 \times 25 \times 5$ mm$^3$ after proton irradiation in three steps up to
1×10^{15} \text{ p cm}^{-2}. The right plot of Fig. 8 compares the normalized light output as a function of 800 MeV proton fluence for LYSO plates of 10 × 10 × 3 mm³ as well as BaF2 and PWO plates of 25 × 25 × 5 mm³ after a proton fluence up to 1×10^{15} \text{ p cm}^{-2}. While both LYSO and BaF2 samples showed a light output degradation of less than 20%, about 90% loss in LO is observed in PWO after a proton fluence of 1×10^{15} \text{ p cm}^{-2}. It is clear that LYSO and BaF2 are much more radiation hard against protons than PWO.

![Figure 8. Neutron induced radiation damage in LYSO, BaF2 and PWO crystals.](image)

Recently, neutron induced radiation damage in inorganic scintillators was also investigated [9]. Fig. 9 shows transmittance (left), light output as a function of integration time (middle) and the normalized light output as a function of fast neutron fluence (right) for LYSO, BaF2 and PWO plates of 10 × 10 × 5 mm³ after a fast neutron (>1 MeV) fluence up to 3.6×10^{15} \text{ n cm}^{-2}. While both LYSO and BaF2 samples show a light output degradation of less than 25% after 3.6×10^{15} \text{ n cm}^{-2}, about 86% loss in LO is observed in PWO plates after 1.6×10^{15} \text{ n cm}^{-2}. It is clear that LYSO and BaF2 are radiation hard against fast neutrons than PWO.

5. Summary

LYSO, BaF2 crystals and LuAG ceramics show excellent radiation hardness up to 340 Mrad, 1×10^{15} \text{ p cm}^{-2} and 3.6×10^{15} \text{ n cm}^{-2}, promising a fast and robust detector in a severe radiation environment, such as HL-LHC. Commercially available undoped BaF2 crystals provide ultrafast light with sub-ns decay time. Yttrium doping in BaF2 crystals increases its F/S ratio significantly while maintaining the ultrafast component. This material thus is promising for the Mu2e-II experiment and GHz hard X-ray imaging. Recent results of neutron irradiation at LANL show fast neutrons up to 3.6×10^{15} \text{ n cm}^{-2} do not cause significant damage in LYSO and BaF2 plates. We plan to continue investigating novel ultrafast crystals and radiation hardness of LuAG ceramics and BaF2:Y crystals. Will also pay an attention to photodetector with DUV response for reading out the ultrafast scintillation light with sub-ns decay time.

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