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Ultrafast and Radiation Hard Inorganic Scintillators for Future HEP Experiments

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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 (PbWO4 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:Tl, 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 6×10^{14} p cm^{-2} and fast

neutron fluence of 3×10^{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 3×10^{15} p cm^{-2} [8]. Progress on neutron induced radiation damage up to 3.6×10^{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

|	LSO/LYSO	GSO	YSO[1]	CsI	BaF$_2$	CeF$_3$	CeBr$_3$[2]	LaCl$_3$	LaBr$_3$	Plastic scintillator (BC 404)[3]
Density (g/cm³)	7.40	6.71	4.44	4.51	4.89	6.16	5.23	3.86	5.29	1.03
Melting point (ºC)	2050	1950	1980	621	1280	1460	722	858	783	70#
Radiation Length (cm)	1.14	1.38	3.11	1.86	2.03	1.70	1.96	2.81	1.88	42.54
Molière Radius (cm)	2.07	2.23	2.93	3.57	3.10	2.41	2.97	3.71	2.85	9.59
Interaction Length (cm)	20.9	22.2	27.9	39.3	30.7	23.2	31.5	37.6	30.4	78.8
Z value	64.8	57.9	33.3	54.0	51.6	50.8	45.6	47.3	45.6	-
dE/dX (MeV/cm)	9.55	8.88	6.56	5.56	6.52	8.42	6.65	5.27	6.90	2.02
Emission Peak[a] (nm)	420	430	420	310	300 / 220	340 / 300	371	335	356	408
Refractive Index[b]	1.82	1.85	1.80	1.95	1.50	1.62	1.9	1.9	1.9	1.58
Relative Light Yield[a,c]	100	45	76	4.2 / 1.3	42 / 4.8	8.6	141	15 / 49	153	35
Decay Time[a] (ns)	40	73	60	30 / 6	650 / 0.9	30	17	570 / 24	20	1.8
d(LY)/dT[d] (%/ºC)	-0.2	-0.4	-0.3	-1.4	-1.9 / 0.1	~0	-0.1	0.1	0.2	~0

a. Top line: slow component, bottom line: fast component.
b. At the wavelength of the emission maximum.

c. Relative light yield normalized to the light yield of LSO
d. At room temperature (20ºC)
#. Softening point

1. N. Tsuchida et al *Nucl. Instrum. Methods Phys. Res. A*, 385 (1997) 290-298
   http://www.hitachi-chem.co.jp/english/products/cc/017.html
2. W. Drozdowski et al. *IEEE TRANS. NUCL. SCI*, VOL.55, NO.3 (2008) 1391-1396
   Chenliang Li et al, *Solid State Commun*, Volume 144, Issues 5–6 (2007),220–224
   http://scintillator.lbl.gov/
3. http://www.detectors.saint-gobain.com/Plastic-Scintillator.aspx
   http://pdg.lbl.gov/2008/AtomicNuclearProperties/HTML_PAGES/216.html

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 (HHCAL) 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].
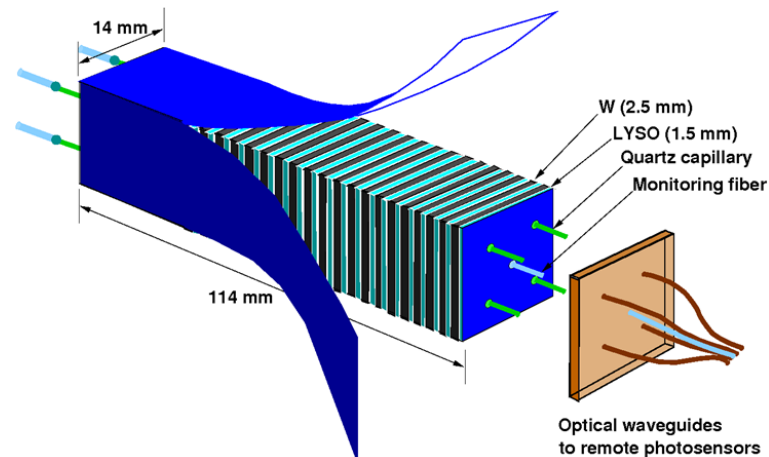
**Figure 1** An LYSO/W/Quartz Capillary-based shashlik sampling calorimeter concept.

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.
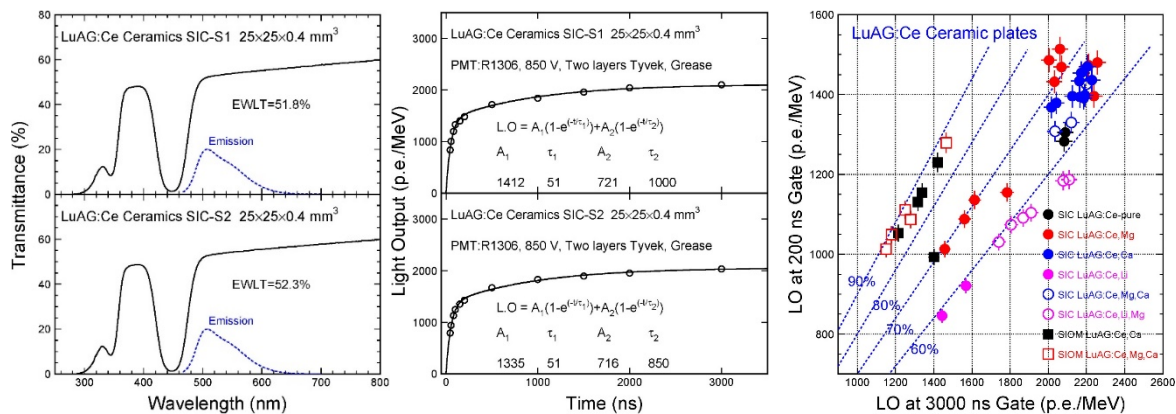
Figure 2. Optical and scintillation properties of LuAG:Ce ceramic samples

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].
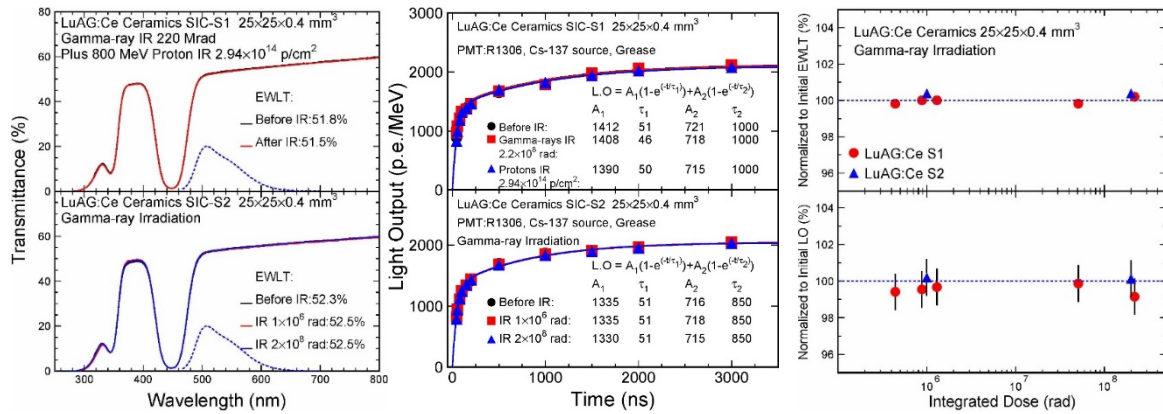

Figure 3. Ionization dose and proton induced radiation damage in LuAG:Ce ceramic samples.

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 2.9×10^{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 BaF$_2$: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.
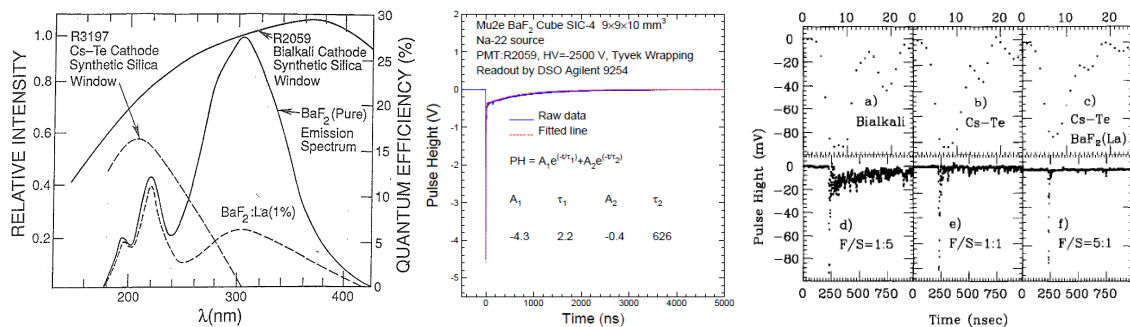
Figure 4. Fast and slow scintillation in BaF$_2$ and slow suppression.

Yttrium doping in BaF_2 was found to be effective in supressing 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 $\Phi19\times21$mm 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.
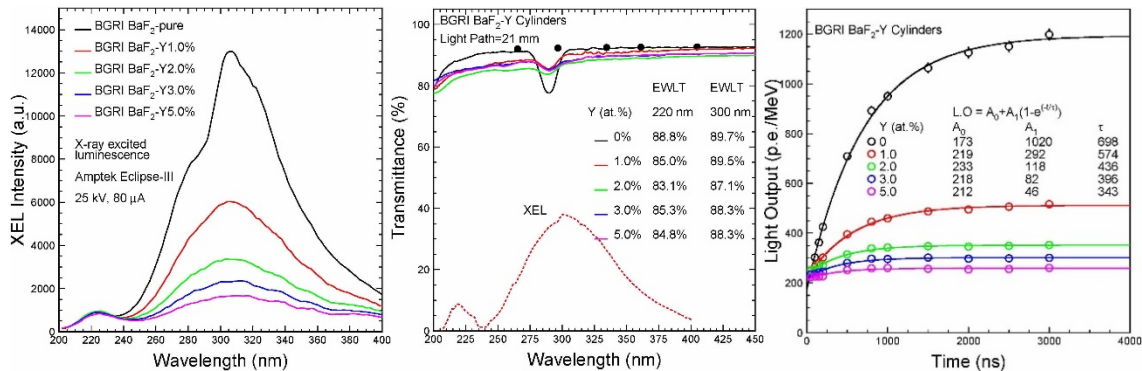

Figure 5. Optical and scintillation properties of BaF_2:Y samples.

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 $\Phi10 \times10$ 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.
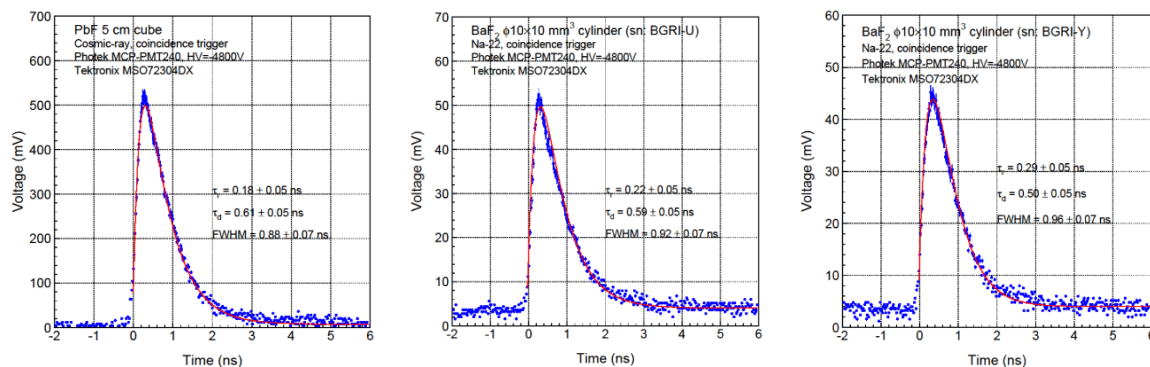

Figure 6. Ultrafast pulse shape in PbF_2 (left), BaF_2 (middle) and BaF_2:Y (right).

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.
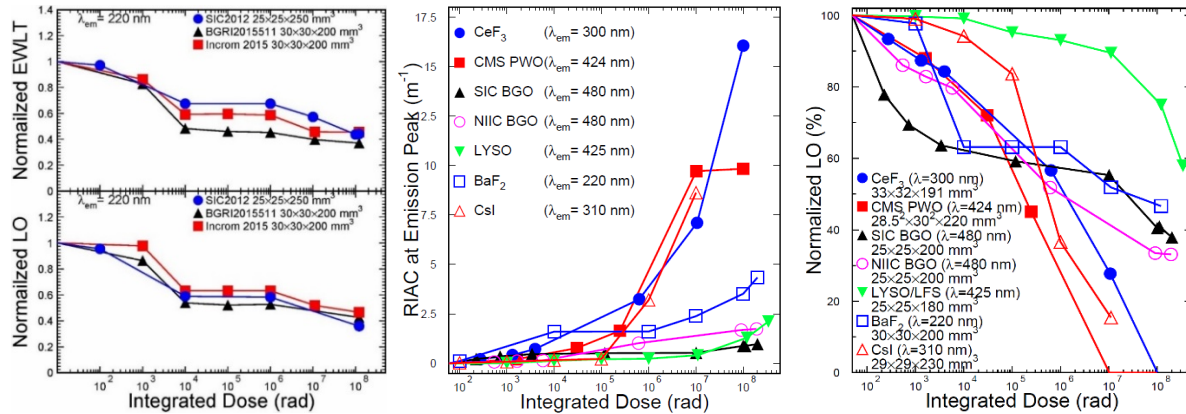

Figure 7. Ionization dose induced radiation damage in large size inorganic scintillators

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.
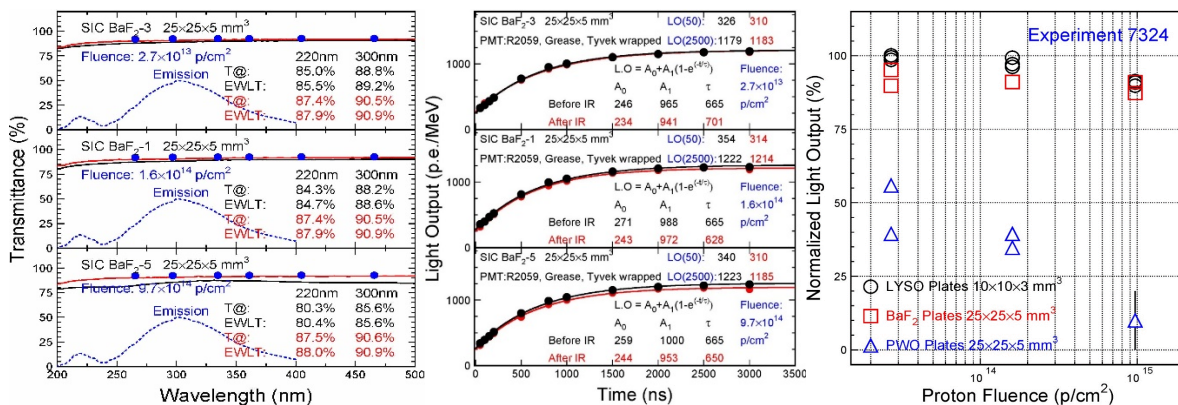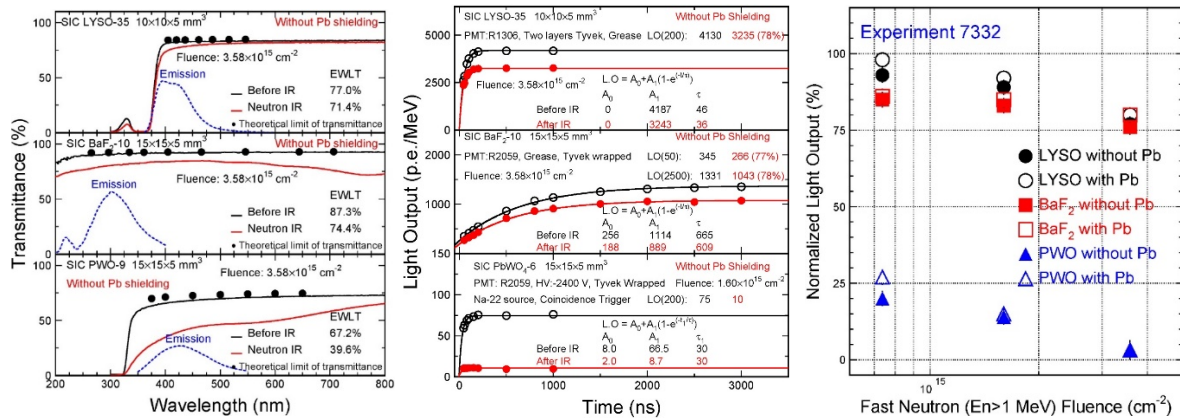

Figure 8. Proton induced radiation damage in LYSO, BaF_2 and PWO crystals.

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} 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 $10 \times 10 \times 3$ mm^3 as well as BaF$_2$ and PWO plates of $25 \times 25 \times 5$ mm^3 after a proton fluence up to 1×10^{15} p cm^{-2}. While both LYSO and BaF$_2$ 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} p cm^{-2}. It is clear that LYSO and BaF$_2$ are much more radiation hard against protons than PWO.

Figure 9. Neutron induced radiation damage in LYSO, BaF2 and PWO crystals.

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, BaF$_2$ and PWO plates of $10 \times 10 \times 5$ mm^3 after a fast neutron (>1 MeV) fluence up to 3.6×10^{15} n cm^{-2}. While both LYSO and BaF$_2$ samples show a light output degradation of less than 25% after 3.6×10^{15} n cm^{-2}, about 86% loss in LO is observed in PWO plates after 1.6×10^{15} n cm^{-2}. It is clear that LYSO and BaF$_2$ are radiation hard against fast neutrons than PWO.

5. Summary

LYSO, BaF$_2$ crystals and LuAG ceramics show excellent radiation hardness up to 340 Mrad, 1×10^{15} p cm^{-2} and 3.6×10^{15} n cm^{-2}, promising a fast and robust detector in a severe radiation environment, such as HL-LHC. Commercially available undoped BaF$_2$ crystals provide ultrafast light with sub-ns decay time. Yttrium doping in BaF$_2$ 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} n cm^{-2} do not cause significant damage in LYSO and BaF$_2$ plates. We plan to continue investigating novel ultrafast crystals and radiation hardness of LuAG ceramics and BaF$_2$:Y crystals. Will also pay an attention to photodetector with DUV response for reading out the ultrafast scintillation light with sub-ns decay time.

Acknowledgments

References

[1]　R.-Y. Zhu, 'The next generation of crystal detectors,' Radiat Detect Technol Methods (2018) 2:2. DOI: 10.1007/s41605-017-0027-x.

[2]　S. Chatrchyan et al., 'Observation of a new boson at a mass of 125 GeV with the CMS experiment at the LHC,' Physics Letters B, 716, (2012) 30-61. DOI: 10.1016/j.physletb.2012.08.021.

[3]　T. T. de Fatis on behalf of the CMS Collaboration, 'Role of the CMS Electromagnetic Calorimeter in the hunt for the Higgs boson in the two-gamma channel,' J. Phys.: Conf. Ser. 404, 012002. DOI:10.1088/1742-6596/404/1/012002.

[4]   R.-Y. Zhu, 'Radiation damage in scintillating crystals,' Nucl. Instr. and Meth. In Phys. Res. A 413 (1998) 297—311. DOI: 10.1016/S0168-9002(98)00498-7.

[5]   A.N. Annennkov et al., 'Systematic study of the short-term instability of PbWO$_4$ scintillator parameters under irradiation,' Radiat Meas 29:27–38 (1998). DOI: 10.1016/S1350-4487(97)00230-8.

[6]   R.-Y. Zhu, 'A Very Compact Crystal Shashlik Electromagnetic Calorimeter for Future HEP Experiments,' J. Phys.: Conf. Ser. 928 (2017) 012015. DOI: 10.1088/1742-6596/928/1/012015.

[7]   F. Yang et al., 'Gamma-Ray Induced Radiation Damage Up to 340 Mrad in Various Scintillation Crystals', IEEE Trans. Nucl. Sci., 63, (2015) 612-619. DOI: 10.1109/TNS.2015.2505721.

[8]   C. Hu et al., 'Proton-Induced Radiation Damage in BaF$_2$, LYSO and PWO Crystal Scintillators,' in proceedings of the SCINT 2017 conference, and references therein. DOI: 10.1109/TNS.2018.2808841. F. Yang et al., 'Proton-Induced Radiation Damage in Fast Crystal Scintillators', IEEE Trans. Nucl. Sci., 64, (2016) 665-672. DOI: 10.1109/TNS.2016.2633427.

[9]   C. Hu et al., 'Neutron-Induced Radiation Damage in BaF$_2$, LYSO and PWO Crystals,' in these proceedings.

[10]   T. T. de. Fatis on behalf of the CMS Collaboration, 'Studies of precision time-tagging of charged tracks with scintillating crystals for the phase-II upgrade of CMS,' presented in the SCINT 2017 conference, Chamonix, France.

[11]   https://indico.hep.anl.gov/indico/conferenceDisplay.py?ovw=True&confId=1258, Mu2e-II Workshop, ANL, December 2017.

[12]   R.-Y. Zhu, "Applications of Very Fast Inorganic Crystal Scintillators in Future HEP Experiments," in Proceedings of the TIPP 2017 conference; J. Chen et al., 'Slow Scintillation Suppression in Yttrium Doped BaF$_2$ Crystals,' in Proceedings of the SCINT 2017 conference. DOI: 10.1109/TNS.2017.2786042

[13]   P. Lecoq, 'The 10 ps Timing-of-Flight PET Challenge,' presented in the SCINT 2017 conference, Chamonix, France.

[14]   R.M. Turtos et al., 'Ultrafast emission from colloidal nanocrystals under pulsed X-ray excitation,' JINST_068P_0616. DOI:10.1088/1748-0221/11/10/P10015.

[15]   A. Driutti et al., 'Towards jet reconstruction in a realistic dual readout total absorption calorimeter,' J. Phys. Conf. Ser. 293 (2011) 012034. DOI:10.1088/1742-6596/293/1/012034.

[16]   K. Pauwels et al., 'Single crystalline LuAG fibers for homogeneous dual-readout calorimeters,' Journal of Instrumentation (2013) 8 (09):09019. DOI: 10.1088/1748-0221/8/09/P09019

[17]   R. Mao et al., 'Crystals for the HHCAL Detector Concept,' IEEE Trans. Nucl. Sci., 59, (2012) 2229-2236. DOI: 10.1109/TNS.2012.2192290.

[18]   M. Lucchini et al., 'Scintillation properties and radiation tolerance of Alkali Free Fluorophosphate Glasses with different dopant concentrations,' presented in the SCINT 2017 conference, Chamonix, France.

[19]   C. Hu et al., 'Alkali-Free Ce-Doped and Co-Doped Fluorophosphate Glasses for Future HEP Experiments,' in proceedings of the SORMA 2018 conference.

[20]   C. Hu et al., 'LuAG Ceramic Scintillators for Future HEP Experiments for Future HEP Experiments,' in proceedings of the SORMA 2018 conference.

[21]   B. P. Sobolev et al., "Suppression of BaF2 Slow Component of X-ray Luminescence in Non-stoichiometric Ba$_{0.9}$R$_{0.1}$F$_{2.1}$ Crystals (R= Rare Earth Element)," Scintillator and Phosphor Materials, Materials Research Society Symposium Proceedings M. J. Weber, P. Lecoq, R. C. Ruchti, C. Woody, W. M. Yen and R. Y. Zhu, eds., pp. 277-283 (1994).

[22]   R.-Y. Zhu, "On quality requirements to the barium fluoride crystals," Nucl. Instrum. Meth. A, vol. 340, no. 3, pp. 442-457 (1994).

[23]   C. W. Barnes et al, 'Technology Risk Mitigation Research and Development for the Matter-Radiation Interactions in Extremes (MaRIE) Project,' Los Alamos National Laboratory Report LA-UR-17-26474 (2017); C. Hu et al., "Ultrafast Inorganic Scintillators for GHz Hard X-Ray Imaging", in proceedings of the SCINT 2017 conference. DOI: 10.1109/TNS.2018.2808103.

[24]   D. Ma, and R.-Y. Zhu, "Light attenuation length of barium fluoride crystals," Nucl. Instrum. Meth. A, vol. 333, no. 2-3, pp. 422-424 (1993).