Gamma-Ray Induced Radiation Damage Up to 340 Mrad in Various Scintillation Crystals

Fan Yang, *Member, IEEE*, Liyuan Zhang, *Member, IEEE*, and Ren-Yuan Zhu, *Senior Member, IEEE*

***Abstract*—Because of their superb energy resolution and detection efficiency scintillation crystals are widely used in high energy and nuclear physics experiments. A crucial issue is radiation damage in crystals. We report an investigation on γ-ray induced radiation damage in various crystal scintillators of large size, including BaF_2, BGO, CeF_3, pure CsI, LSO/LYSO/LFS and PWO, with an integrated dose up to 340 Mrad and a dose rate up to 1 Mrad/h. Optical and scintillation properties of these crystal samples were measured before and after irradiations. The results show that pure CsI has good radiation hardness below 100 krad. BaF_2, BGO and LYSO have good radiation hardness beyond 1 Mrad. In terms of light output degradation LYSO is clearly the best among all scintillation crystals.**

***Index Terms*—Emission, light output, longitudinal transmittance, radiation damage, scintillation crystal.**

I. INTRODUCTION

BECAUSE of their superb energy resolution and detection efficiency, crystal scintillators are widely used in HEP experiments. The CMS lead tungstate ($PbWO_4$ or PWO) crystal calorimeter, for example, has played an important role in the discovery of the Higgs boson [1]. One crucial issue, however, is their radiation damage in a severe radiation environment, which requires precision monitoring to correct variations of crystal's transparency [2]. During the two years of the 1st run, up to 70% loss of light output in CMS PWO crystals at large rapidity was observed *in situ* at the LHC when the experiment was running at a luminosity of 5×10^{33} cm^{-2} s^{-1} and a half of its designed energy [3]. The proposed HL-LHC with 5×10^{34} cm^{-2} s^{-1} luminosity and 3,000 fb^{-1} integrated luminosity presents an extreme severe radiation environment, where up to 130 Mrad ionization dose, 3×10^{14} charged hadrons/cm^2 and 5×10^{15} neutrons/cm^2 are expected [4]. To face these challenges, bright, fast and radiation hard cerium doped lutetium yttrium oxyorthosilicate ($Lu_{2(1-x)}Y_{2x}SiO_5Ce$ or LYSO) crystals was proposed to construct a sampling Shashlik calorimeter for the CMS upgrade for the HL-LHC [5]. Future high energy physics experiments at both the energy and intensity frontiers require fast crystal scintillators with good radiation hardness for precision electromagnetic calorimetry.

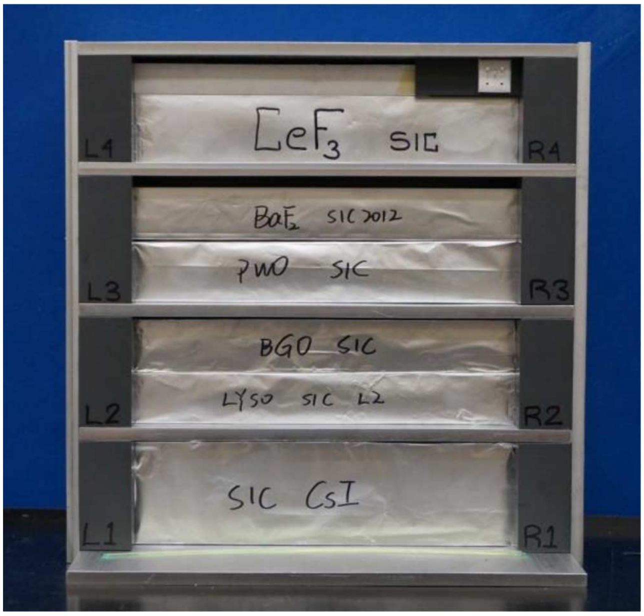

Fig. 1. Seven samples in an aluminum box.

We report an investigation on γ-ray induced radiation damage in various fast crystal scintillators of large size, such as BaF_2 ($20 \times 20 \times 250$ mm^3), BGO ($25 \times 25 \times 200$ mm^3), CeF_3 ($33 \times 32 \times 191$ mm^3), pure CsI ($50 \times 50 \times 300$ mm^3), LYSO ($25 \times 25 \times 200$ mm^3) and PWO ($28.5^2 \times 30^2 \times 220$ mm^3). The 30 cm long pure CsI sample was irradiated to 1 Mrad, and was cut to 20 cm for irradiations beyond 1 Mrad.

Gamma-ray irradiations up to 1 Mrad were carried out at two irradiation facilities at Caltech: an open ^{60}Co source and a sealed ^{137}Cs source. The former provides dose rates between 2 and 100 rad/h by placing samples at appropriate distances. The later provides a dose rate of about 7 krad/h in 2015 with 5% uniformity along the sample's longitudinal axis when the samples are placed at the center of the irradiation chamber [6].

Gamma-ray irradiations beyond 1 Mrad or 7 krad/h were carried out at the Total Ionization Dose (TID) facility of Jet Propulsion Laboratory (JPL), where a group of high intensity ^{60}Co sources provides an adjustable dose rate up to 1 Mrad/h in an opening throat of $10 \times 10 \times 13.5$ inch. Fig. 1 shows a photo of an aluminum box of ten inch square containing six large size crystal samples. The box was inserted in the throat with the 10×10 inch side facing the source so that samples are uniformly irradiated. All samples were pre-irradiated to 1 Mrad at Caltech. The irradiations at the TID facility were carried out in steps: 9, 90 and several steps of 100 Mrad each to reach a total

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The authors are with the California Institute of Technology, Pasadena, CA 91125 USA (e-mail: zhu@hep.caltech.edu).

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of 340 Mrad. The dose rate was 180 krad/h for the 9 Mrad irradiation and 1 Mrad/h for the rest.

Optical and scintillation properties, such as emission and transmittance spectra, light output and light response uniformity were measured before and after each irradiation step. Photo-luminescence was measured by using a HITACHI F4500 fluorescence spectrophotometer. The angle between the excitation UV light and the sample was set to be 10° so that the photoluminescence spectra collected are not affected by internal bulk absorption. Longitudinal transmittance (LT) was measured by using a PerkinElmer LAMBDA 950 UV/Vis spectrophotometer with double beam, double monochromator and an integrating sphere in a large sample compartment. The systematic uncertainty in repeated measurements is 0.15%.

Scintillation light output (LO) was measured by using a Hamamatsu R2059 PMT for $\mathrm{BaF_2}$, $\mathrm{CeF_3}$, pure CsI and PWO, and a Hamamatsu R1306 PMT for BGO and LYSO/LSO/LFS. For the LO measurement the large end of tapered samples or one end of rectangular samples was coupled to the PMT, while all other faces of the sample were wrapped with Tyvek paper. Dow Corning 200 fluid was used between crystals and PMT for most samples except pure CsI, for which air gap was used so that its soft surface was not damaged by the cleaning procedure after measurements and thus reduced systematic uncertainties for its light output measurement. Collimated $^{137}\mathrm{Cs}$ or $^{22}\mathrm{Na}$ sources were used to excite the samples. The γ-ray peak positions were determined by a simple Gaussian fit. The LO of a crystal is defined as an average of the light output measured at seven positions evenly distributed along the crystal axis with a systematic uncertainty of 1%.

II. LYSO/LSO/LFS CRYSTALS

LYSO, LSO and LFS crystals by six different vendors are investigated. The top plot of Fig. 2 shows the photo-luminescence spectra measured before and after 90 Mrad irradiation for a Saint-Gobain LYSO crystal sample. Their difference normalized to the area within FWHM between 380 and 450 nm is shown in the bottom plot. The average absolute value of the difference is 0.45% much less than the systematic uncertainties, indicating that γ-ray irradiation up to 90 Mrad does not change the scintillation mechanism in LYSO crystals. This result is consistent with our previous observation [6].

Fig. 3 shows the longitudinal transmittance spectra measured before and after irradiation of 1, 10 and 100 Mrad for six LYSO/LSO/LFS samples grown by different vendors. Also listed in the figure are the numerical values of the emission weighted longitudinal transmittance (EWLT) defined as [7]:

$$EWLT = \frac{\int LT(\lambda)\, Em(\lambda)\, d\lambda}{\int Em(\lambda)\, d\lambda}. \tag{1}$$

EWLT provides a presentation of the LT across crystal's emission spectrum. It is a direct measure of transparency for crystal's scintillation light. No significant transparency degradation was observed up to 10 Mrad, indicating excellent radiation hardness of crystal scintillators of this type.

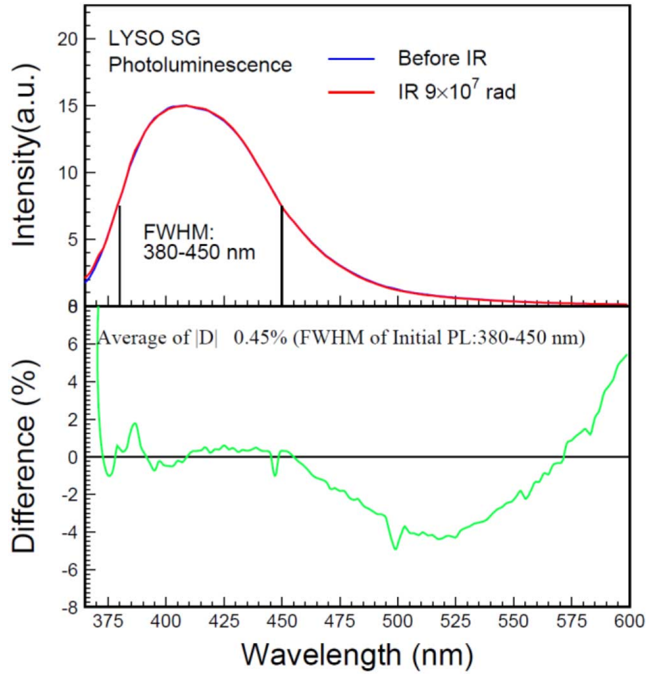

Fig. 2. Top: Photo-luminescence spectra measured before (blue) and after (red) γ-ray irradiation of 90 Mrad are shown for a SG LYSO sample. Bottom: corresponding normalized difference (green).

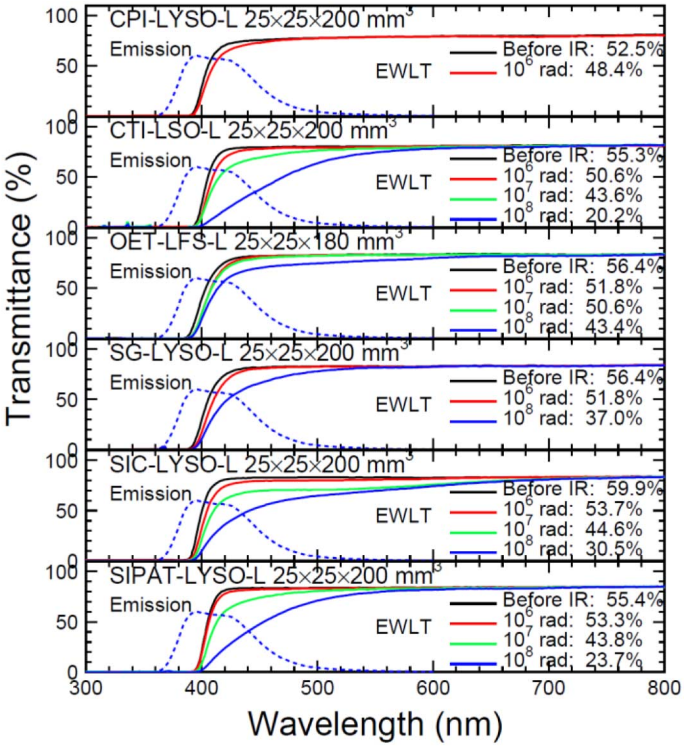

Fig. 3. Longitudinal transmittance spectra are shown for six LSO/LYSO/LFS samples from different vendors before and after γ-ray irradiations.

Fig. 4 shows the EWLT values as a function of time measured after 200 Mrad irradiation for two LYSO samples and the corresponding fits. The recovery time constants extracted are thousands days for both samples. This negligible recovery indicates that radiation damage in crystals of this type has no dose rate

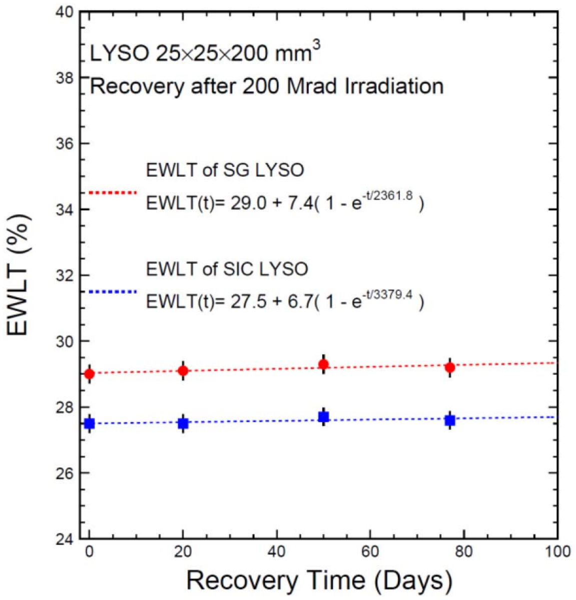

Fig. 4. The EWLT values are shown as a function of time for two LYSO samples after 200 Mrad irradiation.

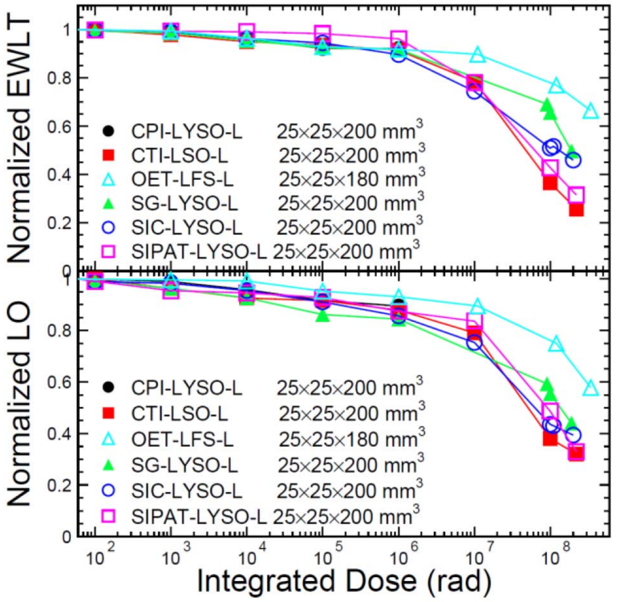

Fig. 5. Normalized EWLT and LO are shown as a function of integrated dose up to 340 Mrad for six LYSO/LSO/LFS crystals from different vendors.

dependence, so that high dose irradiation can be carried out in high dose rate [8]. In other words, the radiation damage effect in crystals of this type depends only on the integrated dose, not the dose rate used. This result is also consistent with our previous observation [6].

Fig. 5 shows normalized EWLT (top) and LO (bottom) as a function of the integrated dose for six LYSO/LSO/LFS samples. The loss of EWLT and LO of the OET LFS is about 35% and 42% respectively after 340 Mrad irradiation. The average LO loss after 10 Mrad is about 18% with a divergence of 7.5%, indicating good consistency of radiation hardness for commercially available crystals from six vendors.

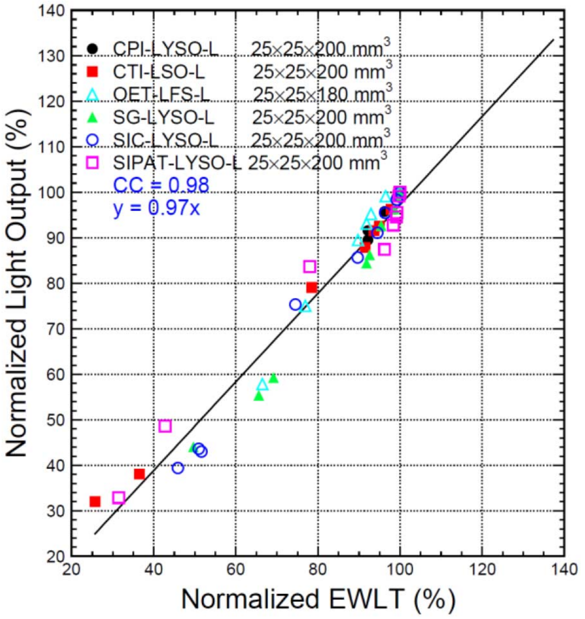

Fig. 6. Correlation between normalized EWLT and LO is shown for LYSO/LSO/LFS crystals after irradiation of various doses.

Fig. 6 shows correlation between normalized EWLT and LO for six LYSO/LSO/LFS crystals after irradiation of various integrated dose. Also shown in the plot is a linear fit and the linear correlation coefficients (CC), defined as [9]:

$$\mathrm{CC} = \frac{\sum (x - \bar{x})(y - \bar{y})}{\sqrt{\sum (x - \bar{x})^2 \sum (y - \bar{y})^2}} \tag{2}$$

An excellent positive correlation of 98% is observed between normalized EWLT and LO for six LYSO/LSO/LFS crystals from different vendors, indicating that LO loss in crystals of this type is caused by transmittance loss, so may be corrected by using a light pulse based monitoring system.

Fig. 7 shows emission weighted radiation induced absorption coefficient (EWRIAC) as a function of integrated dose for six long LYSO/LSO/LFS crystal samples from different vendors. The radiation induced absorption coefficient (RIAC) and EWRIAC are measures of the radiation damage in crystal transparency, defined as [10], [11]:

$$RIAC = \frac{1}{l} ln \frac{T_0(\lambda)}{T(\lambda)} \tag{3}$$

$$EWRIAC = \frac{\int RIAC(\lambda)\, Em(\lambda)\, d\lambda}{\int Em(\lambda)\, d\lambda} \tag{4}$$

where T_0 is the transmittance along crystal length l measured before irradiation; T is the transmittance measured after irradiation; $Em(\lambda)$ is the emission spectrum. The EWRIAC values of the best sample is 0.62, 1.5 and 2.4 m^{-1} after 10, 120 and 340 Mrad respectively. These EWRIAC values extracted from long crystals have a small uncertainty of a few cm, so can be used as an input to estimate light output loss for crystals of different shape by using a ray-tracing simulation.

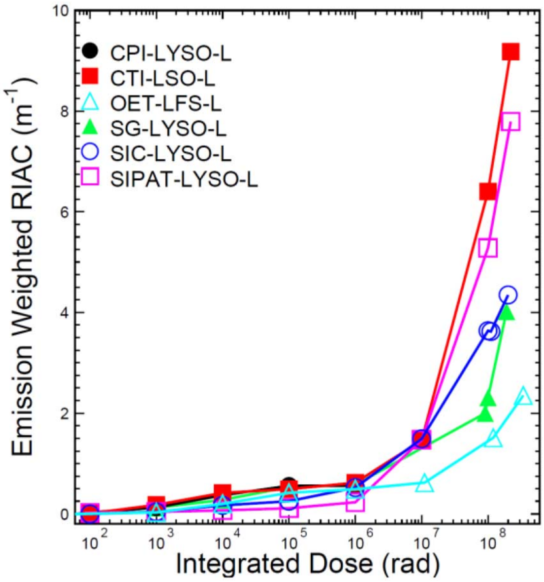

Fig. 7. Emission weighted radiation induced absorption coefficient (EWRIAC) is shown as a function of integrated dose for six LSO/LYSO/LFS crystals.

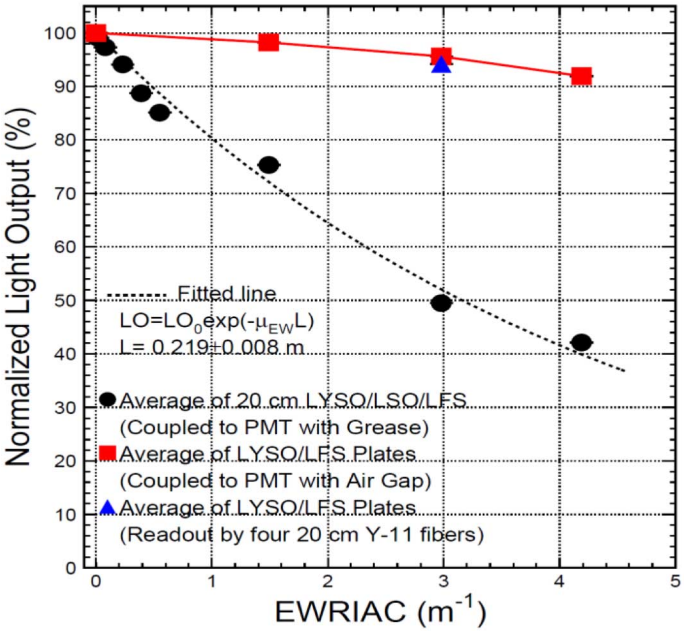

Fig. 8. Normalized LO as a function of EWRIAC is shown for 20 cm long LYSO/LSO/LFS crystals and LYSO/LFS plates of $14 \times 14 \times 1.5$ mm^3.

Fig. 8 shows normalized LO as a function of EWRIAC for 20 cm long LYSO/LSO/LFS crystal samples and $14 \times 14 \times 1.5$ mm^3 LYSO/LFS plates with five holes designed for the proposed LYSO/W Shashlik calorimeter [5]. While the LO of 20 cm long crystals (black dots) was measured by a PMT coupled to the samples directly, the LO of plates was measured by two methods. One is with the plates coupled to a PMT directly (red squares). The other is with the plate coupled a PMT through four Y-11 wavelength shift fibers (blue triangles) [5]. After 100 Mrad irradiation, the LYSO/LSO/LFS samples have a EWRIAC value of about 3 m^{-1}. The corresponding measured LO loss is about 50% for 20 cm long crystals, and 4% and 6% respectively for the $14 \times 14 \times 1.5$ mm^3 plates directly coupled to the PMT and through Y-11 WLS fibers.

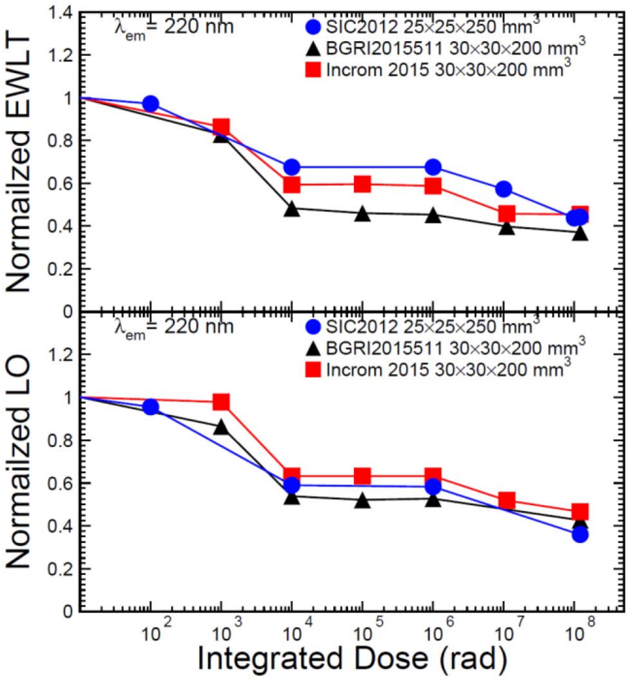

Fig. 9. Normalized EWLT and LO of the fast scintillation component are shown as a function of integrated dose for BaF_2 crystals from three vendors.

Compared to 20 cm long crystals, about a ten times better radiation hardness is observed in $14 \times 14 \times 1.5$ mm^3 plates, indicating that the expected radiation hardness of the proposed LYSO/W Shashlik calorimeter is greatly enhanced as compared to a LYSO calorimeter of total absorption because of the reduced light path length. An exponential fit reveals that the average scintillation light path length in 20 cm long crystals is about 22 cm. The data shown in Fig. 7 and 8 may be used to estimate performance of LYSO/LSO/LFS crystals in various radiation environment.

III. BaF_2 AND PURE CSI CRYSTALS

Similar to LSO/LYSO/LFS γ-ray induced radiation damage in BaF_2 and pure CsI does not recover at room temperature [12], [13], so is not dose rate dependent.

Fig. 9 and 10 show normalized EWLT (top) and LO (bottom) as a function of integrated dose for the fast and slow components respectively for three long BaF_2 crystals from different vendors. The fast and slow components refer to scintillation light peaked at 220 and 300 nm with scintillation decay time of less than 1 ns and 600 ns respectively [12]. While the sample SIC2012 was grown by SICCAS in 2012, the samples BGRI 2015 and Incrom 2015 were grown respectively by BGRI and Incrom in 2015.

The average EWLT and LO values after 120 Mrad are 40% and 45% respectively for the fast and slow scintillation component in these samples from thee different vendors, indicating that long BaF_2 crystals with excellent radiation hardness against ionization dose up to 100 Mrad are available from industry. It is also noticed that the radiation hardness of

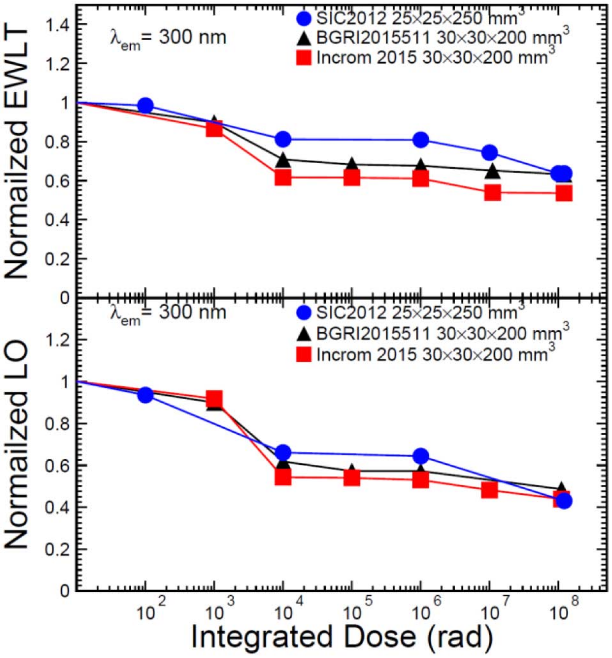

Fig. 10. Normalized EWLT and LO of the slow scintillation component are shown as a function of integrated dose for BaF_2 crystals from three vendors.

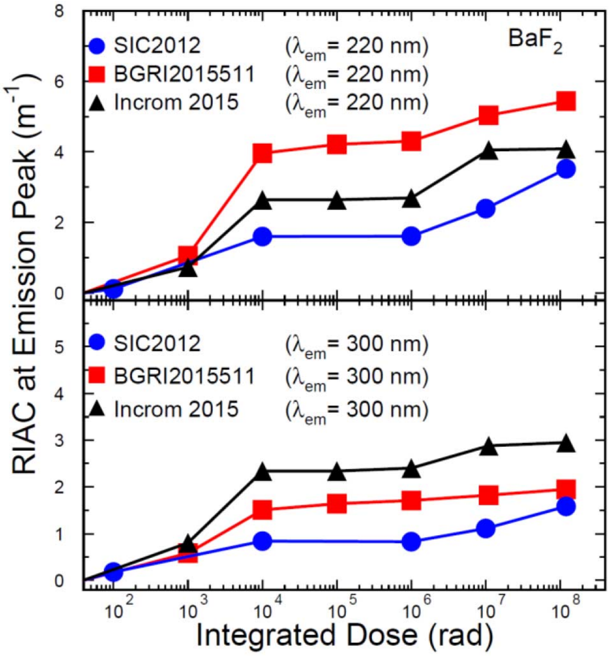

Fig. 11. RIAC values at the emission peak are shown as a function of the integrated dose for BaF_2 crystals from three vendors.

the sample SIC2012 is consistent with BaF_2 crystals batch produced twenty years ago [12].

Fig. 11 shows the RIAC values at the emission peak of the fast (top) and slow (bottom) component as a function of the integrated dose for three BaF_2 crystals from different vendors. The RIAC values of the fast scintillation component are about 2.4 and 3.5 m^{-1} respectively after 10 and 120 Mrad for the 25 cm long BaF_2 sample SIC2012.

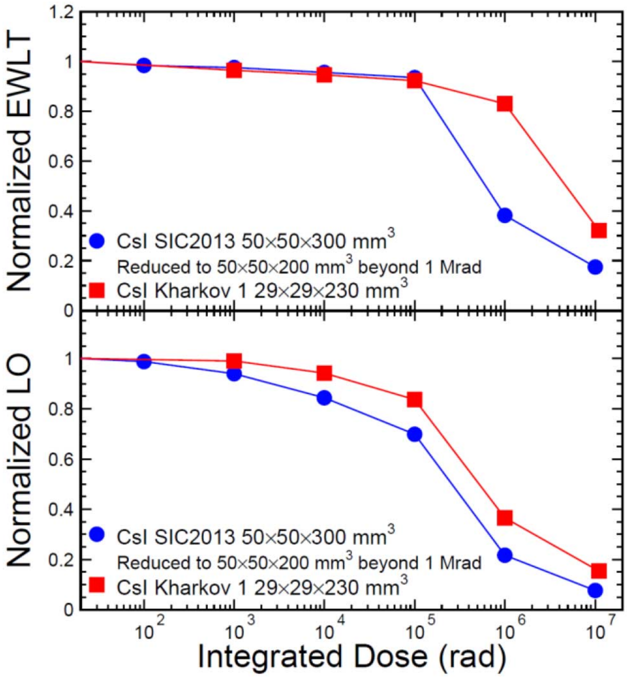

Fig. 12. Normalized EWLT and LO are shown as a function of integrated dose for two pure CsI crystals from different vendors.

Fig. 12 shows the normalized EWLT (top) and LO (bottom) as a function of the integrated dose for two pure CsI crystal samples from different vendors. The 30 cm long samples SIC2013 was grown by SICCAS in 2013. The 23 cm long sample Kharkov 1 was grown by Kharkov in 2015. The sample SIC2013 was 30 cm long when it was irradiated up to 1 Mrad, and was cut to 20 cm for irradiations beyond 1 Mrad at the TID facility of JPL.

The samples from SICCAS and Kharkov show consistent radiation hardness. The LO is about 80% after 100 krad, and was reduced to about 30% after 1 Mrad, indicating that pure CsI is radiation hard at low doses up to 100 krad but not beyond. It is also noticed that the radiation hardness of sample SIC 2013 is consistent with the sample Kharkov 2015 and two Kharkov samples grown twenty years ago [13].

IV. BGO, CeF_3 AND PWO CRYSTALS

Radiation damage in BGO, CeF_3 and PWO crystals recovers under room temperature, leading to a dose rate dependent damage [8]. Fig. 13 shows the normalized EWLT (top) and LO (bottom) as a function of time for the BGO sample NIIC-2013 during γ-ray irradiations at dose rates of 2, 8, 30 and 5,444 rad/h. Both EWLT and LO reached an equilibrium under a definite dose rate [8], [14]–[17]. Consistent time constants were found for both LO and EWLT.

Fig. 14 shows the normalized EWLT (top) and LO (bottom) in equilibrium as a function of the dose rates for two BGO crystal samples from different vendors. The radiation hardness of these two samples is more or less consistent. Early BGO crystals produced for the L3 experiment, however, are not as radiation hard as these samples [17], indicating that the quality of BGO crystals is improved during mass production for the medical industry.

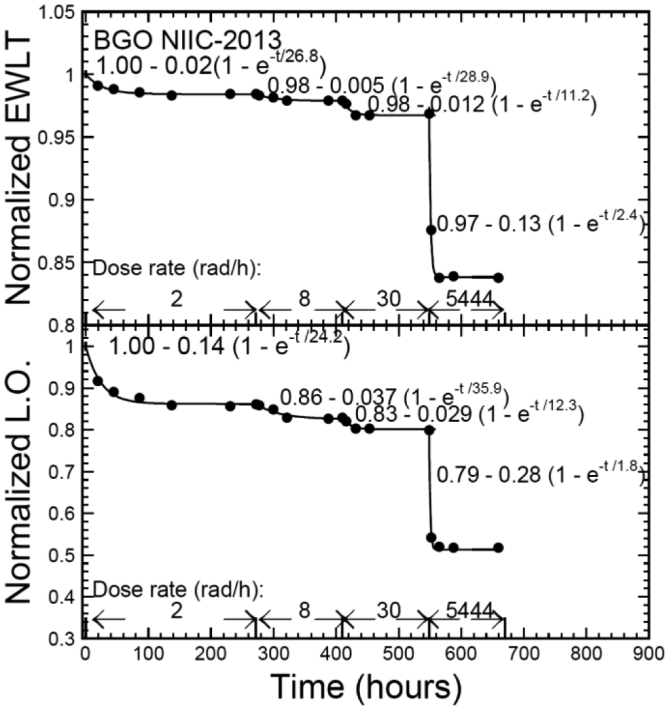

Fig. 13. Normalized EWLT (top) and LO (bottom) are shown as a function of time for the BGO crystal sample NIIC-2013.

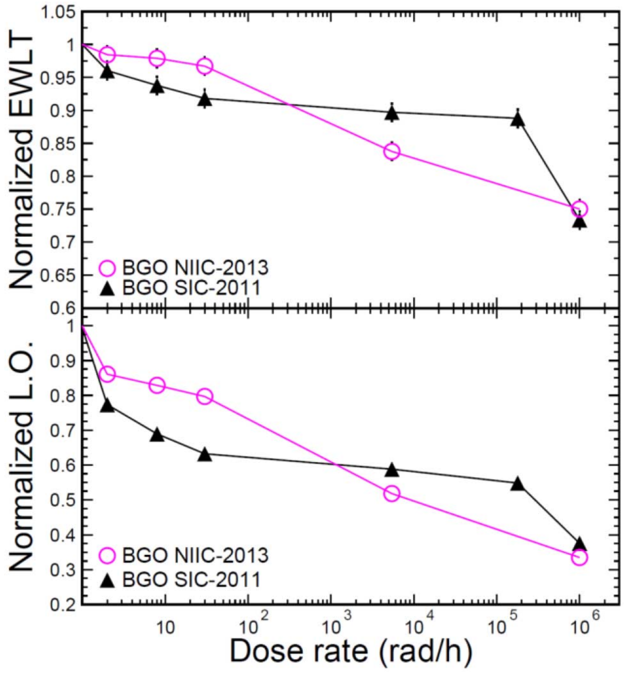

Fig. 14. Normalized EWLT (top) and LO (bottom) are shown as a function of dose rate for two BGO crystal samples.

Fig. 15 shows the normalized EWLT and LO as a function of time for a $\mathrm{CeF_3}$ sample SIC2014 during γ-ray irradiations in steps at dose rates of 2, 8, 30 and 7,062 rad/h. This sample was grown about twenty years ago, but was procured in 2014. Both EWLT and LO reached an equilibrium under a definite dose rate, which is similar to BGO and PWO [8], [14]–[17]. Consistent time constants were found for both LO and EWLT.

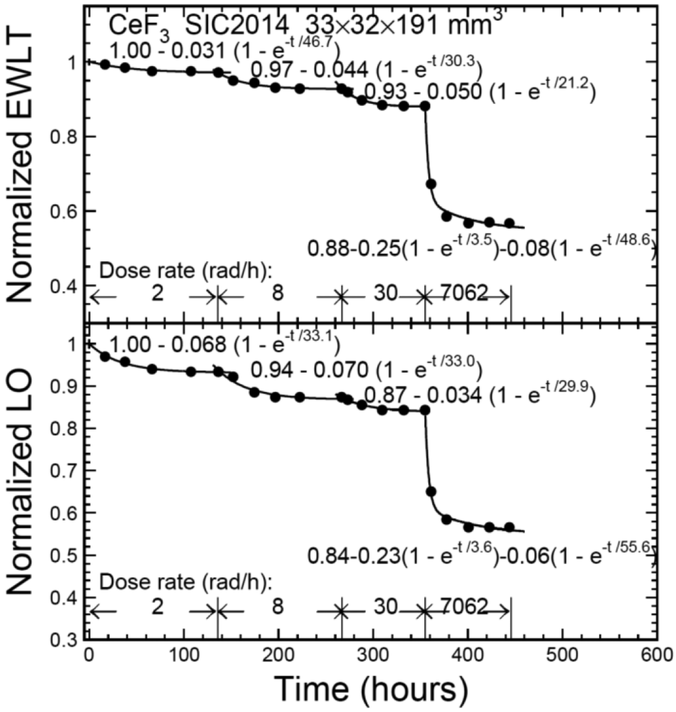

Fig. 15. Normalized EWLT (top) and LO (bottom) are shown as a function of time for the $\mathrm{CeF_3}$ crystal sample SIC2014.

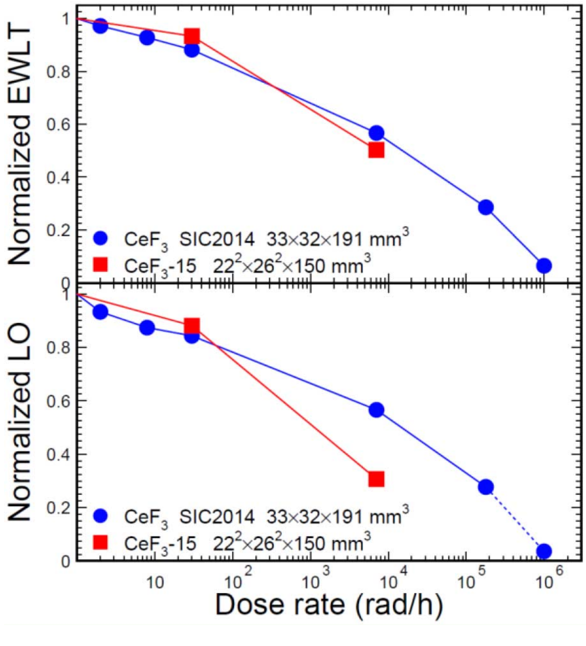

Fig. 16. Normalized EWLT (top) and LO (bottom) are shown as a function of dose rate for two $\mathrm{CeF_3}$ crystal samples.

Fig. 16 shows the normalized EWLT (top) and LO (bottom) in equilibrium as a function of the dose rates for two $\mathrm{CeF_3}$ crystal samples. Consistent radiation hardness was observed in these two samples. The light output of $\mathrm{CeF_3}$ crystal SIC2014 was too low to be measured under the 1 Mrad/h irradiation.

Fig. 17 shows the normalized EWLT (top) and LO (bottom) as a function of time for a PWO sample SIC 5 procured recently by JLAB during γ-ray irradiations in steps at dose rates of 2, 8,

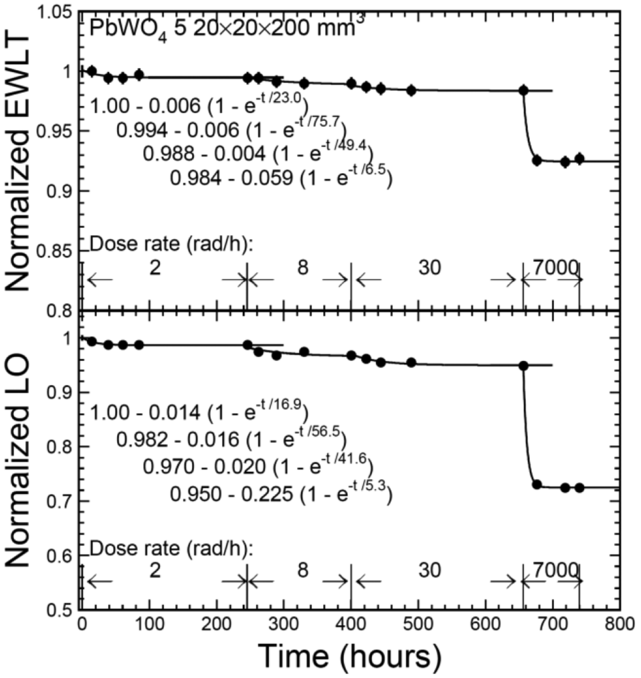

Fig. 17. Normalized EWLT (top) and LO (bottom) are shown as a function of time for the PWO crystal sample SIC 5.

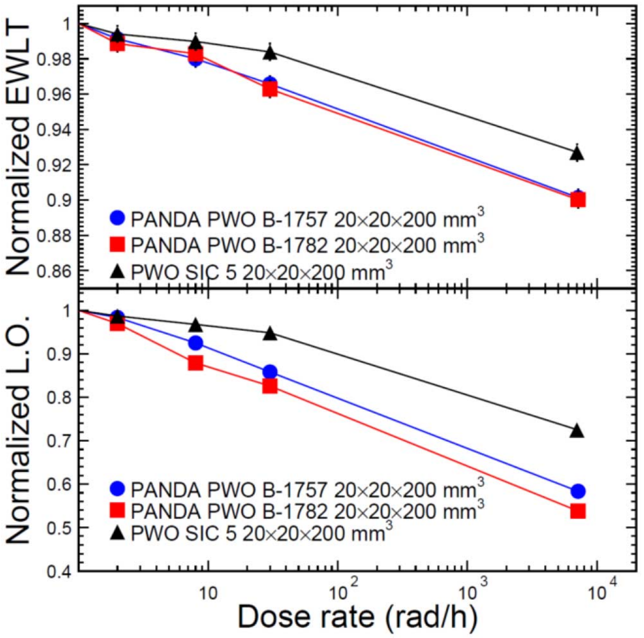

Fig. 18. Normalized EWLT (top) and LO (bottom) are shown as a function of dose rate for three PWO crystal samples.

30 and 7,000 rad/h. Both EWLT and LO reached an equilibrium under a definite dose rate with consistent time constants. This observation is consistent with previous publications [8], [14]–[17].

Fig. 18 shows the normalized EWLT (top) and LO (bottom) in equilibrium as a function of dose rates for three PWO crystals, including two recent grown PWO crystals by BTCP for Panda [17] and one recent grown PWO crystal by SIC for JLAB.

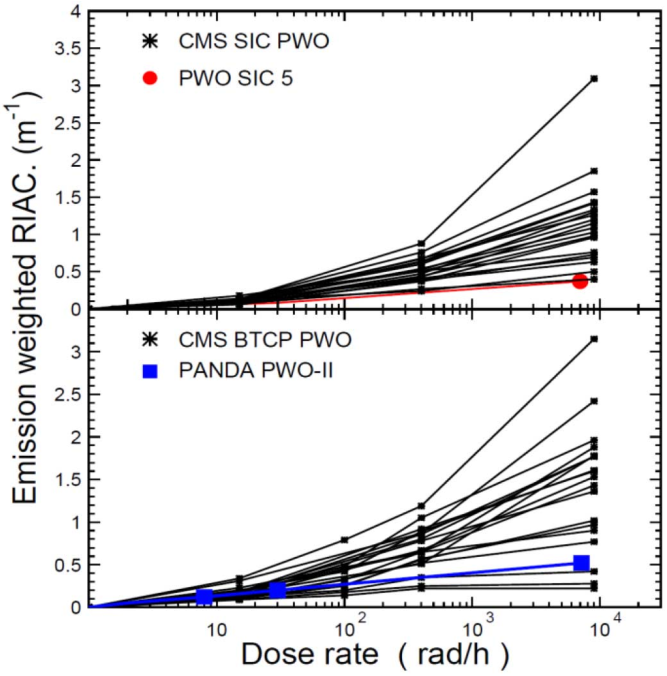

Fig. 19. EWRIAC is shown as a function of dose rate for PWO crystal samples.

Fig. 19 shows a comparison of EWRIAC of three PWO crystals grown recently to that of mass-produced PWO crystals for CMS. As shown in the figure that the radiation hardness of PWO crystals against ionization dose is diverse [11], and has no correlation to their initial optical quality [9]. Although selected samples grown recently are better than mass produced PWO crystals grown for CMS about 15 years ago, quality control is still required to ensure that PWO crystals used to construct future calorimeter meet a defined specification for crystal's radiation hardness.

V. SUMMARY

In this investigation no difference was observed between the photo-luminescence spectra of a LYSO crystal sample before and after 90 Mrad irradiation, indicating that its scintillation mechanism is not damaged by gamma-rays. No recovery was observed after 200 Mrad irradiation in two LYSO samples, indicating radiation damage in LYSO is not dose rate dependent. Consistent degradation in transmittance and light output was observed for 20 cm long LYSO/LSO/LFS crystals from six vendors, indicating that radiation damage in commercially produced crystals of this type is under control. Because of its high cost due to expensive raw materials and high melting point, however, alternative cost-effective crystals should also be considered for future crystal calorimeters.

Ignoring dose rate dependence, Fig. 20 shows the RIAC values at the emission peak as a function of the integrated dose for various crystal samples. Pure CsI crystal shows very small RIAC values below 10 krad. This is partly due to its soft hygroscopic surface which causes a low longitudinal transmittance value measured before irradiation and a reduced radiation

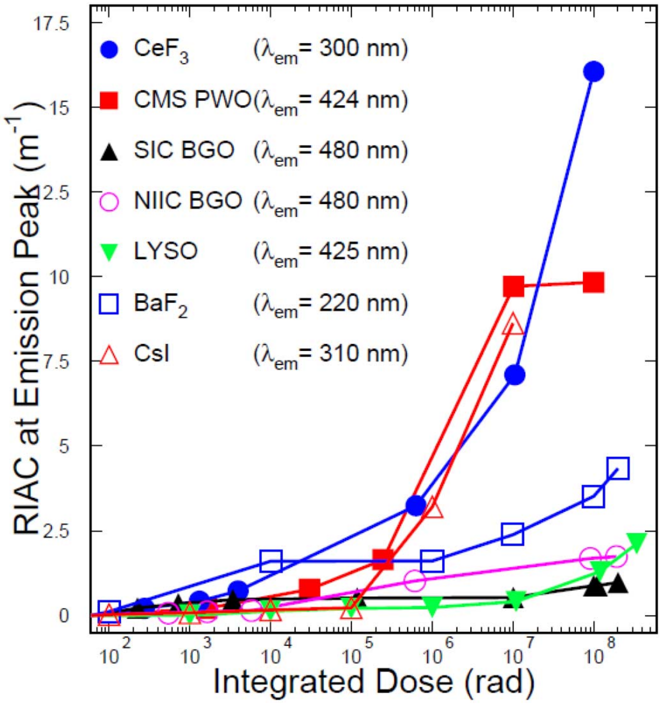

Fig. 20. The RIAC values at the emission peak are shown as a function of integrated dose for various crystals.

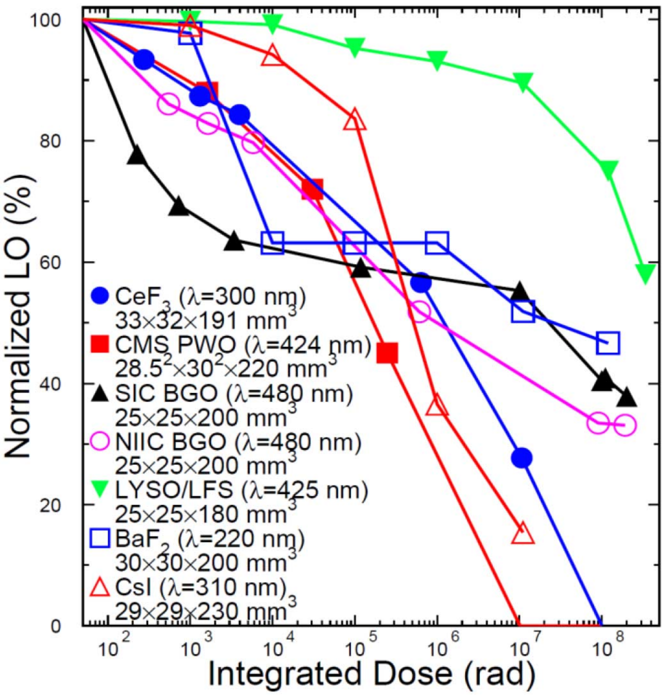

Fig. 21. Normalized LO as a function of integrated dose for various crystals.

induced transmittance loss. BaF_2, BGO and LYSO crystals show good radiation hardness beyond 1 Mrad.

Fig. 21 shows the normalized LO as a function of integrated dose for various long crystal samples. In terms of light output loss, LYSO clearly shows the best radiation hardness among all crystal scintillators. The best sample of this type maintains 75% and 60% LO respectively after 120 and 340 Mrad. On the other hand, BGO and BaF_2 crystals also maintain 35% and 45% LO respectively after 200 and 120 Mrad, so may be considered as cost-effective alternatives for future HEP experiments in a severe radiation environment. Pure CsI shows good radiation hardness below 100 krad. Because of its low cost, it is a good alternative for future HEP experiments with modest radiation environment.

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