A Study on Radiation Damage in PWO-II Crystals

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***Abstract*—We report in this paper an investigation on radiation hardness of 20 cm long PWO-II crystals produced at BTCP for the proposed PANDA experiment at FAIR. The optical and scintillation properties of PWO-II crystals, such as transmittance, light output and light response uniformity, were measured before and after γ-ray irradiations at different dose rates. PWO-II crystals are compared to PWO crystals used in the CMS experiment. Progresses are observed in the light output and the radiation hardness for PWO-II crystals. Its use for crystal calorimeters in future high energy physics experiments is discussed. To maintain crystal precision, a precision light monitoring system is mandatory to follow variations of crystal transparency *in situ*.**

***Index Terms*—Crystal, light output, photo-luminescence, radiation damage, scintillator, transmission.**

I. Introduction

BECAUSE of its high density (8.3 $\mathrm{g/cm^3}$) and short radiation length (0.89 cm), lead tungstate ($\mathrm{PbWO_4}$ or PWO) crystals have been used to construct electromagnetic calorimeters (ECAL) for high energy physics (HEP) experiments, such as the CMS experiment at the LHC [1]. It is also being considered for future HEP experiments, where precision measurements of photons and electrons are crucial for their physics mission. The use of large size scintillation crystals in future HEP experiments, however, faces a challenge from the severe radiation environment expected in these experiments. Recently, PWO crystals with improved radiation hardness were grown at Bogoroditsk Techno-Chemical Plant (BTCP), Russia for the proposed PANDA experiment at FAIR [2]–[4]. In this paper, we present a study on large size PWO-II crystals grown recently at BTCP and compare them to the CMS PWO crystals.

Fig. 1 shows PWO samples investigated in the paper. Two shorter samples are PWO-II crystals with dimension of $24.5 \times 24.5 \times 200\ \mathrm{mm^3}$. Two longer samples are typical PWO crystals grown at SIC and BTCP for the CMS experiment. A total of 53 CMS PWO samples were studied in our previous publication [5] in which 21 from BTCP and 32 from SIC. While PWO crystals are grown at BTCP by Czochralski method, they are grown by Bridgman method at SIC. All 21 BTCP samples are of the CMS endcap size with a slightly tapered shape: $28.5 \times 28.5\ \mathrm{mm^2}$ at the small end, $30 \times 30\ \mathrm{mm^2}$ at the large end and 220 mm long.

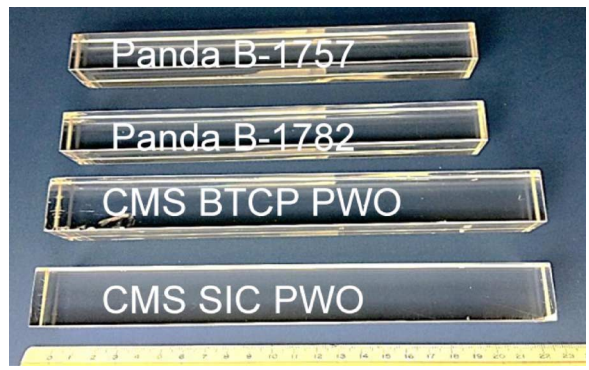

Fig. 1. PWO samples used in this study.

Among the 32 samples from SIC, 20 samples have a rectangular shape of $22 \times 22 \times 230\ \mathrm{mm^3}$ and the other 12 are of the CMS endcap size. All six faces of these PWO samples are polished.

The longitudinal transmittance (LT) was measured using a PerkinElmer LAMBDA 950 UV/Vis spectrophotometer with double beam, double monochromator and a large sample compartment equipped with an integrating sphere. The systematic uncertainty in repeated measurements is 0.15%. The scintillation light output (LO) was measured before and after irradiations by using a Hamamatsu R2059 PMT with a bi-alkali photo-cathode and a quartz window. For the LO measurement, the large (tapered samples) or one (rectangular samples) end of the samples was coupled to the PMT with Dow Corning 200 fluid, while all other faces of the sample were wrapped with Tyvek paper. A collimated $^{137}\mathrm{Cs}$ source was used to excite the samples. The γ-ray peak positions were obtained by a simple Gaussian fit. The LO of a crystal is defined as an average of the light output measured by aiming a γ-ray source at 7 (PWO-II) or 9 (CMS PWO) evenly distributed locations along the crystal axis. The systematic uncertainty in repeated measurements is 1%.

The γ-ray irradiations were carried out in two facilities at Caltech: a 50 curie $^{60}\mathrm{Co}$ source for irradiations at low dose rates (2, 8 and 30 rad/h) and a 7,000 curie $^{137}\mathrm{Cs}$ source for high dose rate at 7,160 rad/h. The dose rates were calibrated. During irradiations a long side of the sample faces the γ-ray source. The differences of dose rates between the middle and the end of samples are less than 10%. The main consequence of γ-ray induced radiation damage in PWO crystals is radiation induced absorption which causes degradation of LT and LO. It may also affect crystal's light response uniformity (LRU) and cause an unrecoverable degradation of the energy resolution if the light attenuation length (LAL) is reduced significantly [6], [7]. The γ-ray induced absorption may also spontaneously recover, leading to dose rate dependent radiation damage [8], [9].

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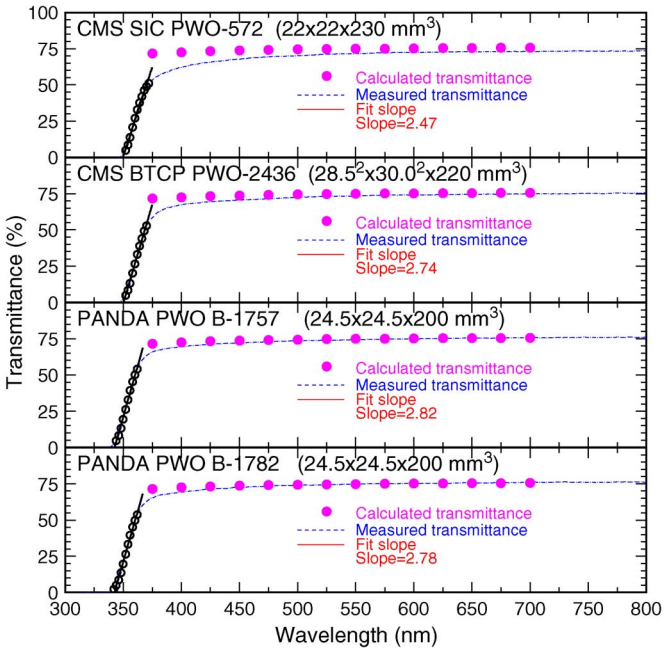


Fig. 2. The LT spectra (dashed lines) are shown as function of wavelength for two CMS PWO and two PWO-II samples. Also shown in the figure are the theoretical limit (solid dots) and the slopes obtained by a linear fit across the band edge.

II. Gamma Ray Induced Radiation Damage in PWO-II Crystals

Two PWO-II crystals were annealed at 200°C for 200 minutes to remove residual damage and then irradiated at 2, 8, 30 and 7,160 rad/h step by step until reaching equilibrium. It is well known that the γ-ray induced damage in PWO crystals recovers under room temperature, leading to dose rate dependent radiation damage [7], [8]. The recovery of radiation damage was measured after these samples were irradiated at 30 rad/h in equilibrium. The radiation damage data for two PWO-II samples are compared to CMS PWO crystals [5].

A. Initial Optical and Scintillation Properties

Fig. 2 shows the LT spectra for two CMS PWO crystals (top two) and two PWO-II crystals (bottom two). The measured transmittance spectra are compared to the theoretical limit (solid dots), which were calculated by using refractive indices assuming no internal absorption following [10]

$$\mathrm{T_s} = (1-\mathrm{R})^2 + \mathrm{R}^2(1-\mathrm{R}) + \cdots = \frac{(1-\mathrm{R})}{(1+\mathrm{R})} \tag{1}$$

where

$$\mathrm{R} = \left(\frac{n_{crystal} - n_{air}}{n_{crystal} + n_{air}}\right)^2 \tag{2}$$

A comparison of the LT data and the theoretical limit of transmittance reveals crystal's optical quality, e.g. preexisting internal absorption and scattering centers. Fig. 2 shows clearly that the measured LT data are very close to the theoretical limits, indicating that the internal absorption and scattering are negligible in these samples.

Also shown in Fig. 2 are the numerical values of the slope of the LT across the band edge, which were obtained by a linear fit (straight lines) to 10 data points (black dots) between two wavelengths: 350 nm and 370 nm for CMS BTCP PWO and PWO-II, and 352 nm and 372 nm for CMS SIC PWO respectively. The difference between these two fit ranges is due to the birefringent nature of PWO crystals and the different optical orientation between PWO crystals grown in these two producers [11]–[13].

Fig. 3 shows a comparison of the initial LT data at (a) 360 nm, (b) 440 nm and (c) 600 nm for all PWO samples. It is clear that the PWO-II crystals have a better LT, especially at 360 nm. We also notice that the effect of different crystal length between 20 cm for PWO-II, 22 cm for CMS BTCP and 23 cm for some CMS SIC samples is negligible in this comparison.

Fig. 4 shows a comparison of the initial LO. PWO-II samples have significantly higher LO than CMS BTCP PWO, but are compatible to CMS SIC PWO. Once again the effect of different crystal length between 20 cm for PWO-II, 22 cm for CMS BTCP and 23 cm for some CMS SIC samples is negligible in this comparison.

Fig. 5 shows correlations between the numerical values of the initial LO and the initial LT at 360 nm. Also shown in the plot are the linear fits and the corresponding linear correlation coefficients (CC), defined as

$$\mathrm{CC} = \frac{\sum(x-\bar{x})(y-\bar{y})}{\sqrt{\sum(x-\bar{x})^2 \sum(y-\bar{y})^2}} \tag{3}$$

Positive correlation coefficients of 78% and 92% are observed between the initial LO and initial LT at 360 nm for PWO crystals from SIC and BTCP respectively. This correlation may partly be attributed to the self-absorption effect of the very blue part of the emission spectrum, as evidenced by the cross-over of the transmittance and emission spectra at 360 nm [5]. A simple linear fit shows different slopes between the initial LO (y) and the initial LT at 360 nm (x) for the SIC (0.48) and BTCP (0.25) samples, which may be caused by their different optical orientations [5].

B. Recovery of Radiation Induced Damage

Fig. 6 shows the recovery of the LT after irradiation at 30 rad/h in equilibrium for two PWO-II samples. Also listed in the figure are the numerical values of the emission weighted longitudinal transmittance (EWLT) which is defined as:

$$EWLT = \frac{\int LT(\lambda) Em(\lambda) d\lambda}{\int Em(\lambda) d\lambda} \tag{4}$$

The EWLT value provides a numerical presentation of the LT data across the emission spectrum. It is a direct measure of the transparency of crystal's scintillation light. Similar to the LT the systematic uncertainty of the EWLT value is 0.15%.

Fig. 7 shows the EWLT values as a function of recovery time for two PWO-II samples. Consistent recovery time constants of a few days were determined by an exponential fit for both samples. The initial EWLT values (70.0% and 70.1% for sample B-1757 and B-1782), however, are not reached after 900 hours

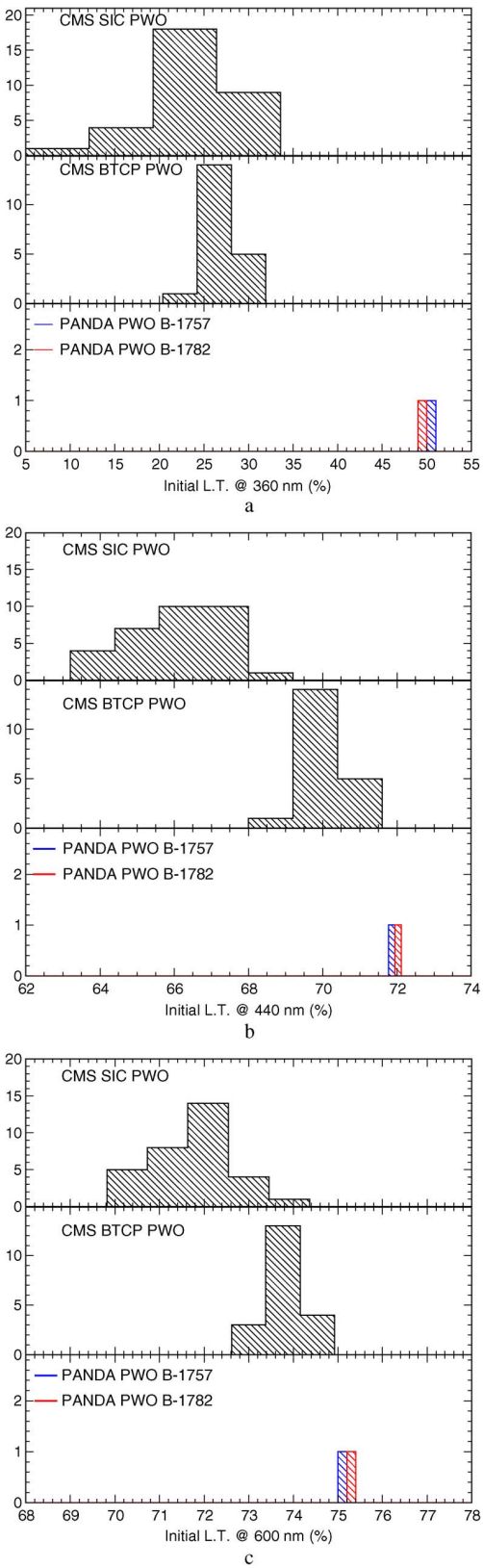


Fig. 3. Comparison of the initial LT at (a) 360 nm, (b) 440 nm and (c) 600 nm for CMS PWO and two PWO-II samples.

recovery, indicating the existence of deep color centers in these crystal samples.

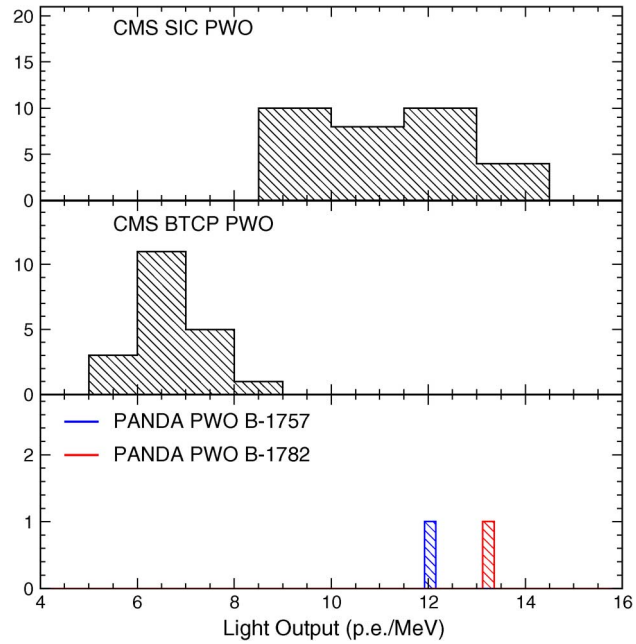


Fig. 4. Comparison of the initial LO for CMS PWO and PWO-II samples.

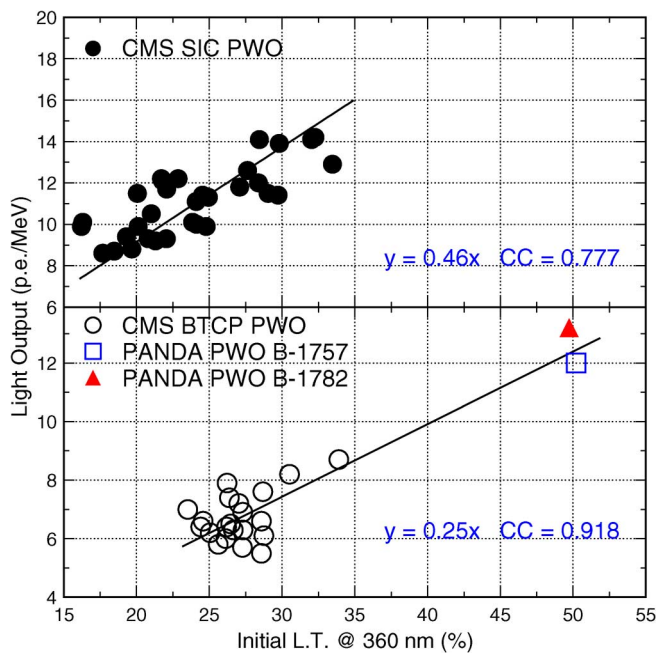


Fig. 5. Correlations between the initial LO (y) and the initial LT (x) at 360 nm.

C. Radiation Damage in Equilibrium at Different Dose Rates

Both PWO-II crystals were irradiated at 2, 8, 30 and 7,160 rad/h step by step until reaching equilibrium. Fig. 8 shows the LT spectra for two PWO-II crystals in equilibrium at difference dose rates. The numerical EWLT values were also listed in these plots.

The light response uniformity of PWO-II samples was measured by aiming a collimated γ-ray source at several points

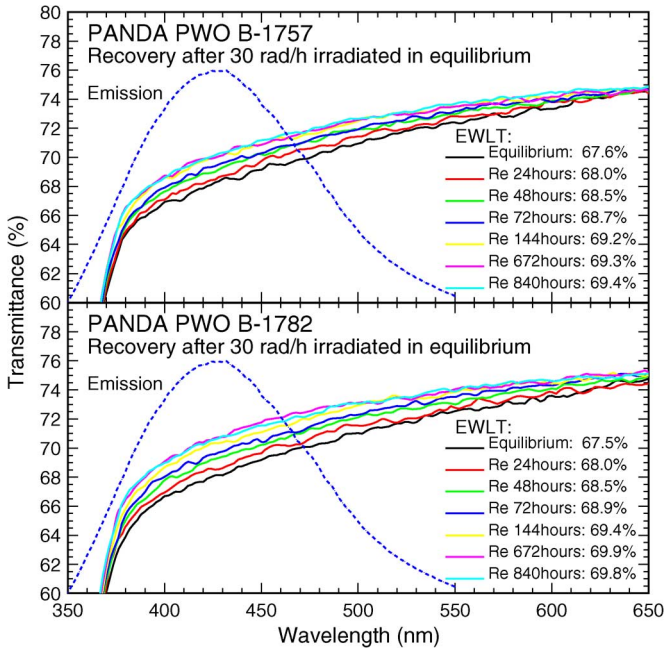


Fig. 6. The recovery of LT after irradiation at 30 rad/h in equilibrium is shown for two PWO-II crystals.

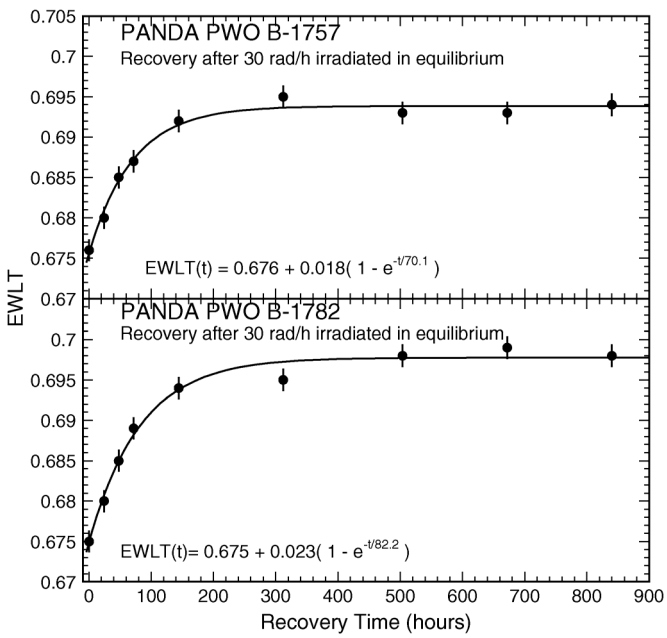


Fig. 7. The EWLT values are shown as a function of recovery time for PWO-II crystals after irradiation at 30 rad/h in equilibrium.

evenly distributed along the crystal axis. The corresponding light response was fitted to a linear function

$$\frac{y}{y_{mid}} = 1 + \delta\left(\frac{x}{x_{mid}} - 1\right) \tag{5}$$

where y_{mid} represents the light output at the middle of the crystal, δ the light response uniformity and x the distance to the end coupled to PMT. LO losses are observed for two PWO-II crystals at 2, 8, 30 and 7,160 rad/h in equilibrium. The light

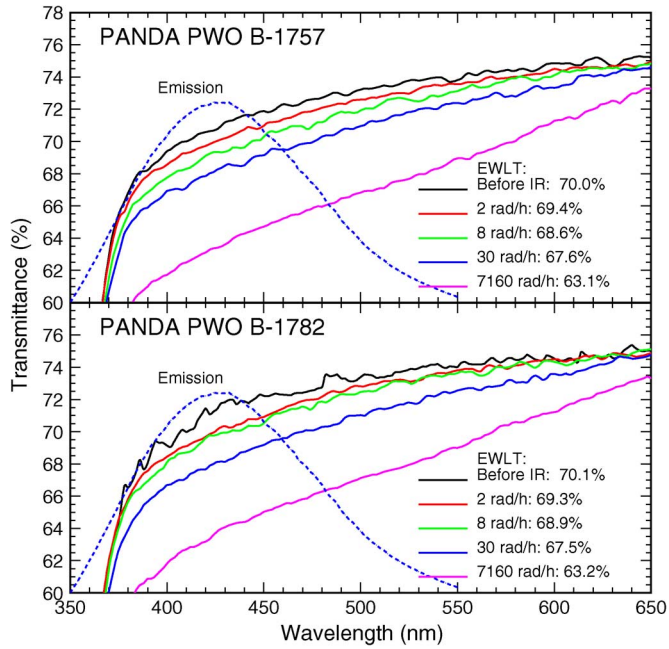


Fig. 8. The LT spectra are shown for two PWO-II crystals in equilibrium at 2, 8, 30 and 7,160 rad/h.

response uniformity of both samples was not changed up to 7,160 rad/h, indicating the constant term in energy resolution is under control. This is due to the fact that the degraded light attenuation length is long enough to maintain the LRU [6].

Fig. 10 shows the normalized EWLT and LO measured in equilibrium as a function of the dose rate for two PWO-II samples. The corresponding data of EWLT and LO loss are listed in the Table I. The EWLT loss of PWO-II crystals is about 10% at 7,160 rad/h while the LO loss is more than 40%. The data of two PWO-II crystals are consistent.

D. Comparison With CMS PWO Crystals

The emission weighted radiation induced absorption coefficient (EWRIAC) is a measure of the radiation damage, which is defined as

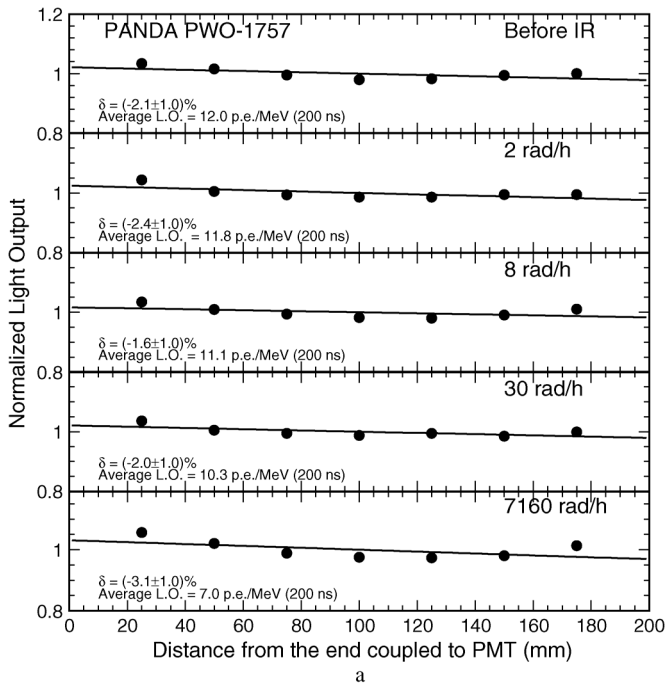
$$EWRIAC = \frac{\int RIAC(\lambda)Em(\lambda)d\lambda}{\int Em(\lambda)d\lambda} \tag{6}$$

where the $RIAC$ is the radiation induced absorption coefficient or the overall color center density, and Em is the scintillation emission. Both are function of wavelength. The radiation induced absorption coefficient (RIAC), or the color center density, were calculated according to the following equation:

$$RIAC = \frac{1}{LAL_{equilibrium}} - \frac{1}{LAL_{before}} \tag{7}$$

where the subscripts "equilibrium" and "before" refer to "in equilibrium" and "before irradiation" respectively. The light attenuation length was calculated by using the LT data according to [10]:

$$\mathrm{LAL} = \frac{1}{\ln\left\{\frac{[\mathrm{T}(1-\mathrm{T}_s)^2]}{\left[\sqrt{4T_s^4+T^2(1-T_s)^2}-2T_s^2\right]}\right\}} \tag{8}$$



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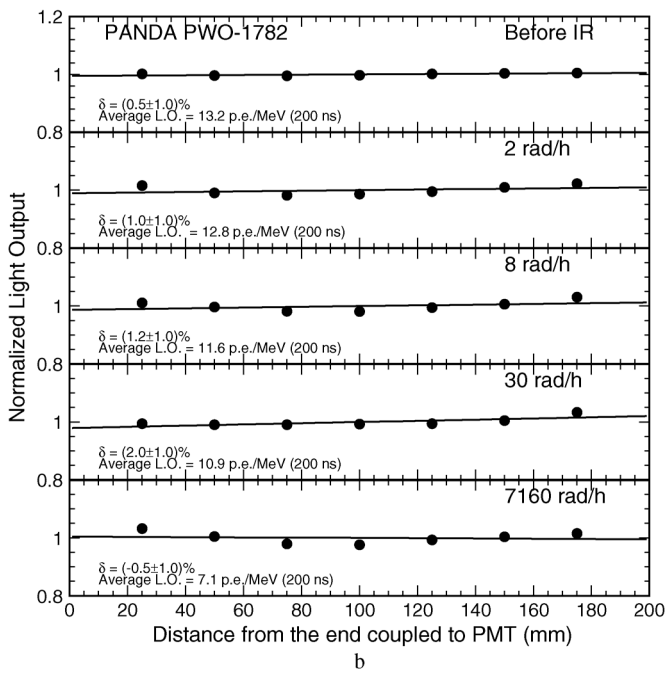

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Fig. 9. The LRU is shown for PWO-II crystals (a. B-1757, b. B-1782) h in equilibrium under irradiation at 2, 8, 30 and 7,160 rad/.

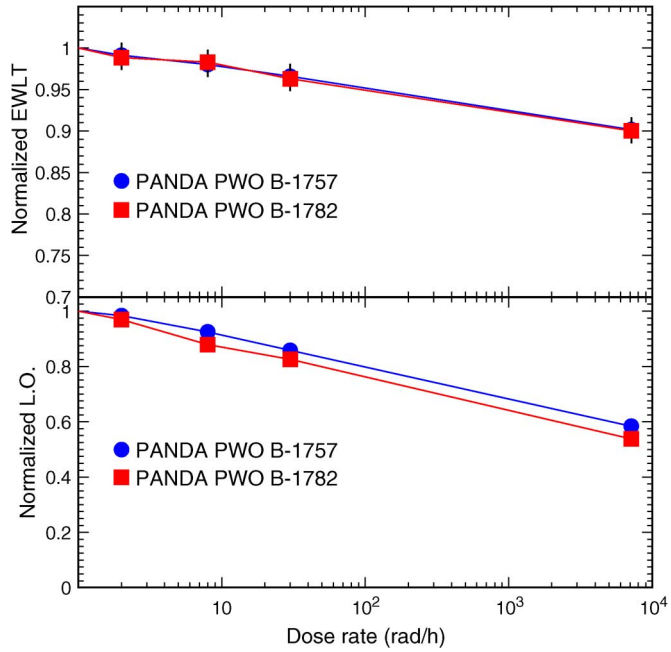

Fig. 10. Normalized EWLT and LO as a function of the dose rate.

TABLE I
SUMMARY OF LO AND EWLT LOSS

Samples		PWO B-1757	PWO B-1782
Initial EWLT (%)		70.0	70.1
Initial LO (p.e./MeV)		12.0	13.2
EWLT Loss (%)	2 rad/h	0.8	1.1
	8 rad/h	2.0	1.7
	30rad/h	3.4	3.7
	7,160rad/h	9.9	10.0
LO Loss (%)	2rad/h	2	3
	8rad/h	8	12
	30rad/h	14	17
	7,160rad/h	42	46

where T is the transmittance measured along crystal length l and T_S is the theoretical limits without internal absorption.

Fig. 11 shows the correlations between the losses of the LO and the losses of the LT at 440 nm (top) and the EWRIAC (bottom) for the CMS PWO samples in equilibrium under γ-ray irradiations at 15 rad/h and the PWO-II samples at 30 rad/h. Positive correlations are observed with the linear correlation coefficient at a level of 50%. The data of PWO-II crystals fit well with the data of CMS PWO crystals. The correlation between the LO losses versus the losses of LT at 440 nm indicates that the LT at 440 nm is a good measure of the radiation damage. This result is consistent with our previous investigation that 440 nm was chosen for the monitoring wavelength because of its best linearity [15].

Fig. 12 shows the radiation induced absorption coefficients (data points with error bars) as a function of the photon energy for the samples CMS BTCP-2455 and PWO-II B-1757 in equilibrium under dose rates of 15, 400, 9,000 rad/h and 30, 7,160 rad/h respectively. Also shown in these plots are the emission spectra (open circles) and the corresponding EWRIAC values calculated according to the (6). In these plots the radiation induced color center densities were decomposed to a sum (solid lines) of two Gaussian shaped color centers (dashed lines):

$$Riac = \sum_{i=1}^{2} A_i e^{-\frac{(E-E_i)^2}{2\sigma_i^2}} \tag{9}$$

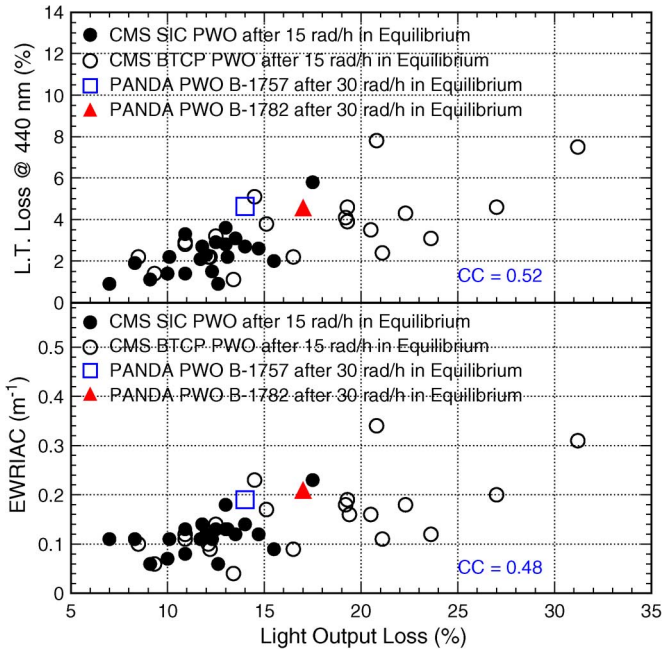

Fig. 11. Correlations between the LO losses versus the losses of LT at 440 nm (top) and the EWRIAC (bottom) for all PWO crystals.

where A_i, E_i, σ_i represent the amplitude, energy and width of the color center i. Consistent peak/width values of two color centers of 2.30/0.19 eV and 3.07/0.70 eV are obtained for both CMS BTCP-2455 and PWO-II B-1757. This indicates a similar nature of color centers in these two samples. The amplitudes of color centers in PWO-II B-1757, however, are lower than that of sample CMS BTCP-2455, indicating that this PWO-II sample has better radiation hardness.

Fig. 13 shows the distributions of the EWRIAC values as a function of the γ-ray dose rate for all samples. It is clear that two PWO-II crystals are in the better portion of the CMS PWO crystals. The EWRIAC values of PWO-II crystals are less than 0.5 m^{-1} up to 7,160 rad/h, indicating their excellent radiation hardness.

III. Summary

The Panda PWO-II crystals have excellent initial optical and scintillation properties. They have a better LT at 360 nm and LO as compared to CMS PWO crystals. Correlations are confirmed between: the initial LO and the initial LT at 360 nm, which can be attributed to a weak self-absorption.

The Panda PWO-II crystals have excellent radiation hardness, which is in the better portion of the CMS PWO crystals. They are radiation hard up to 7,160 rad/h. Correlations are confirmed between (1) the LO losses and the losses of LT at 440 nm and (2) the LO losses and the EWRIAC, indicating that the LO losses can be corrected by measuring LT at 440 nm.

The results presented in the paper are consistent what reported in previous publications [3], [4], [16], [17]. The results also show that the PWO crystals grown by Czochralski method have the same LO as compared to PWO crystals grown by the Bridgman method.

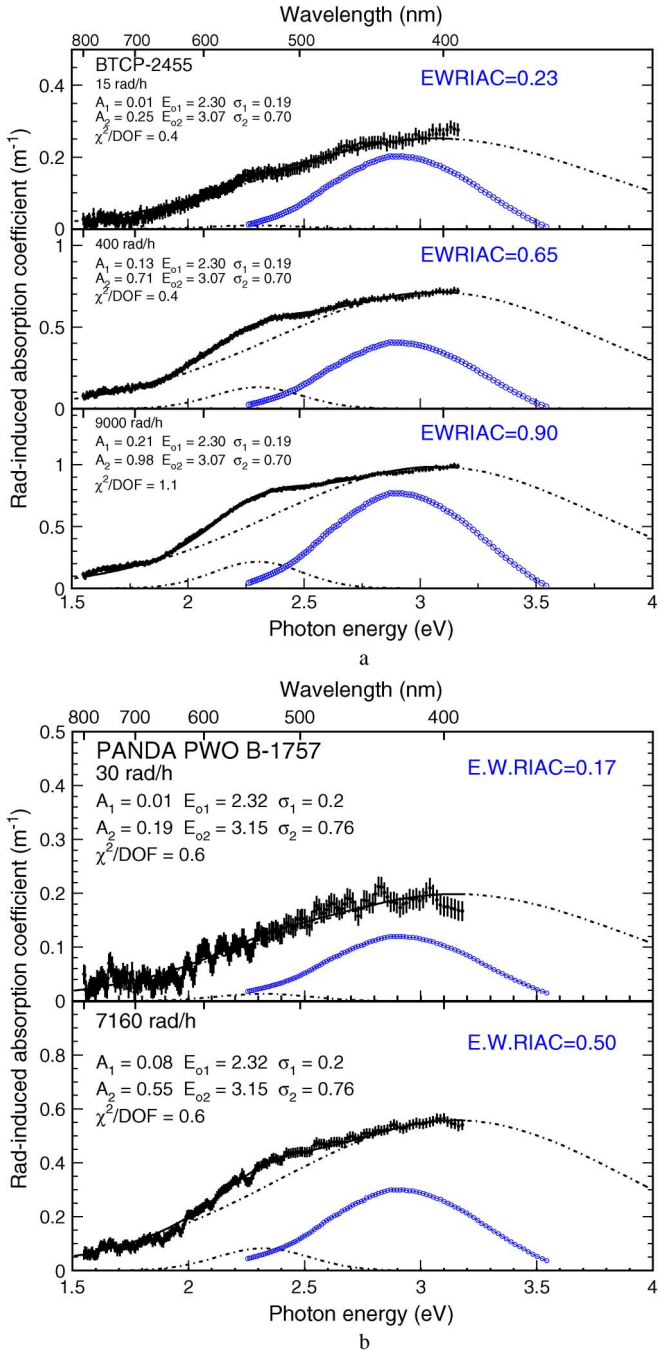

Fig. 12. Radiation induced absorption coefficients (data points with error bars) are shown as a function of the photon energy for (a) CMS BTCP-2455 in the equilibrium under dose rates of 15, 400 and 9,000 rad/h and (b) PWO-II B-1757 in the equilibrium under dose rates of 30 and 7,160 rad/h.

Radiation damage in PWO-II crystals recovers under room temperature, leading to a dose rate dependent damage level. Because of the recovery, a precision light monitoring system is mandatory for a PWO crystal calorimeter to trace variations of crystal transparency and thus the light output *in situ*.

Acknowledgment

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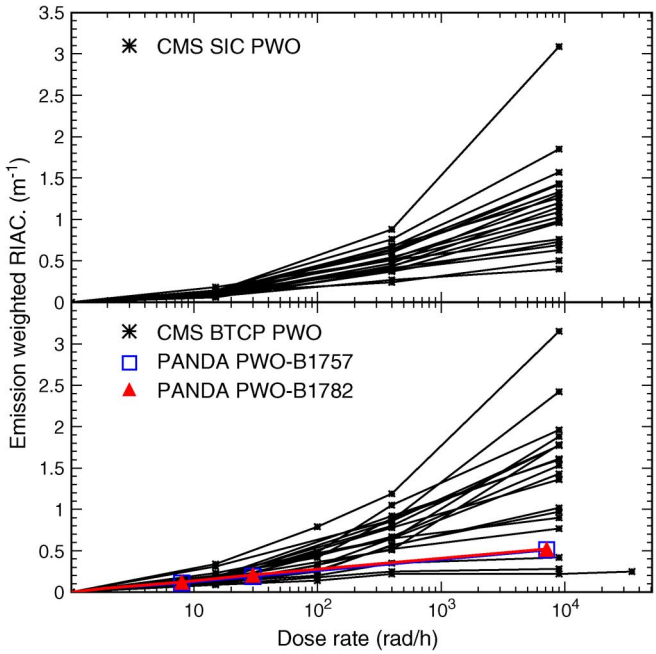

Fig. 13. Distributions of the EWRIAC as a function of the dose rate.

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