

# A Radiation Damage and Recovery Study for Lead Tungstate Crystals from BTCP and SIC

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**Abstract.** This paper presents result of a study on radiation damage and recovery for lead tungstate crystals produced at BTCP and SIC. Correlations were observed between initial light output and initial longitudinal transmittance at 360 nm, between the loss of longitudinal transmittance at 440 nm and the loss of light output, and between radiation damages levels at different dose rates. No correlations, however, were found between crystal's initial optical properties and radiation hardness. Excellent linearity was observed between the variations of crystal's light output and its longitudinal transmittance at 440 nm in several cycles of irradiation followed by recovery, indicating these PWO crystals can be monitored *in situ* at LHC.

**Keywords:** Calorimeter; Lead Tungstate; Crystal; Radiation Damage; Correlation

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## INTRODUCTION

Because of its high density and fast decay time, lead tungstate ( $\text{PbWO}_4$  or PWO) crystal was chosen by the compact muon solenoid (CMS) experiment to construct a precision electromagnetic calorimeter (ECAL) of 76,000 crystals at the large hadron collider (LHC). After extensive R&D in the past decade, PWO crystals are now in mass production [1]. 40,000 crystals have been produced at Bogoroditsk Techno Chemical Plant (BTCP) in Tula, Russia, for the CMS experiment at CERN. Shanghai Institute of Ceramic (SIC) produced a few thousands crystals for the PrimEx experiment at the Jefferson Laboratory, and started to produce PWO crystals for CMS since 2003.

A total of 54 full size PWO samples were studied: 32 from SIC and 22 from BTCP. All samples went through a 200°C thermal annealing, which removes any residual absorption, followed by  $\gamma$ -ray irradiations at 15, 400 and 9000 rad/h until equilibrium [2]. Properties measured are longitudinal transmittance, emission and excitation spectra, light output, decay kinetics, light response uniformity and their degradation. Correlations between initial optical properties and with radiation damage were investigated for all 54 samples. In addition, two samples each from SIC and BTCP in a batch went through several irradiation and recovery cycles at 100 and 400 rad/h to study the linearity between the variations of the longitudinal transmittance and the variations of the light output.

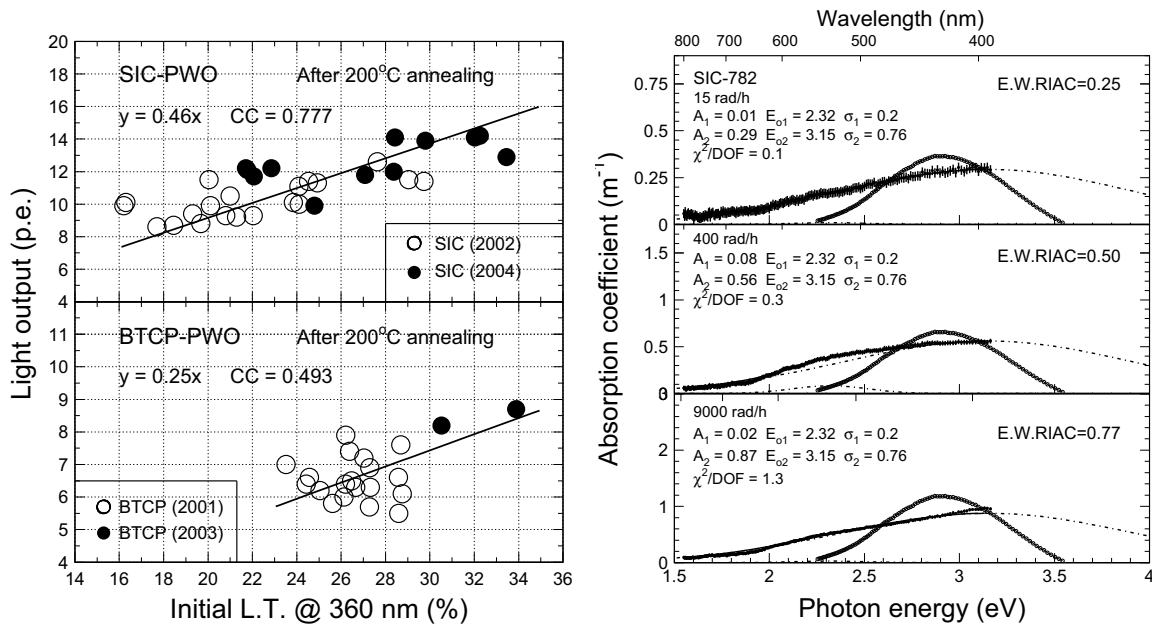
## CORRELATIONS

Correlations were observed between the initial light output and the initial longitudinal transmission at 360 nm, and samples from SIC and BTCP show different slopes, as shown in Fig. 1 (Left). This correlation may be attributed to the self-absorption of

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the very blue part of the emission spectrum, as evidenced by the cross-over of the transmittance and emission spectra at 360 nm. This correlation was also observed by others [3].

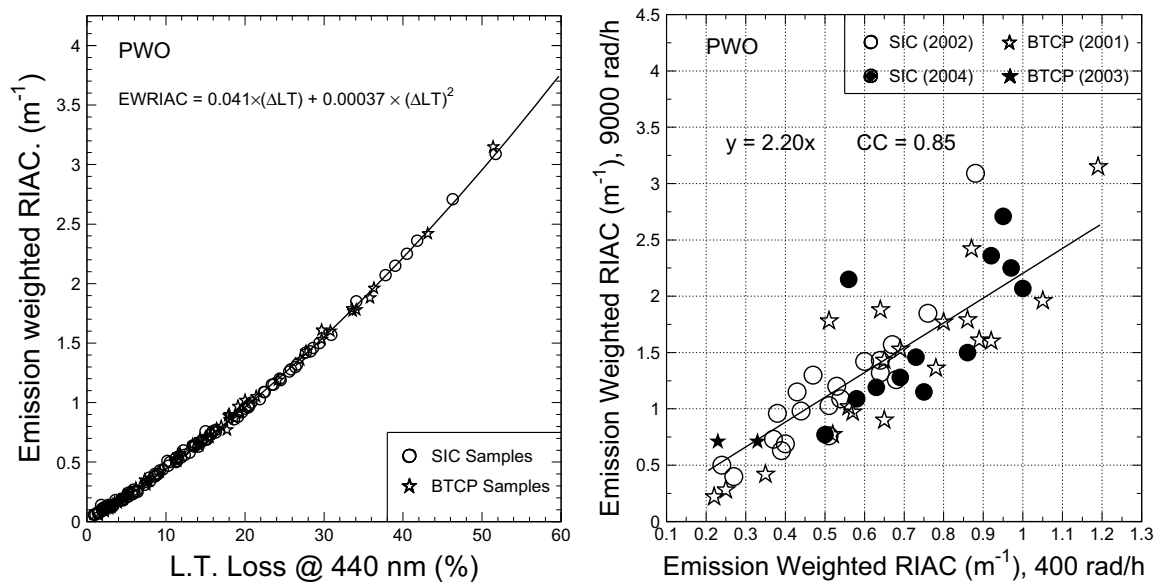


**FIGURE 1.** Left: Correlations are observed between initial light output and initial longitudinal transmission at 360 nm for BTCP and SIC PWO samples. Right: Radiation induced absorption coefficients are shown as a function of photon energy for SIC-782. Also shown in the figure are emission spectrum, decomposed color centers and EWRIAC in units of  $m^{-1}$ .

PWO crystals are known to suffer from radiation induced absorption which is dose rate dependent [2]. Fig. 1 (Right) shows radiation induced absorption coefficient (RIAC) as a function of photon energy for a sample SIC-782 under  $\gamma$ -ray irradiations with dose rate of 15, 400 and 9,000 rad/h in equilibrium [2]. Also shown in the figure is a decomposition of the RIAC to two color centers and crystal's emission spectrum with its peak between two color centers. The numerical values of the emission weighted radiation induced absorption coefficients (EWRIAC) [1] are also shown in the figure. Since the PWO scintillation mechanism is not affected by  $\gamma$ -ray irradiations, the loss of the light output is related to the EWRIAC. The linear correlation coefficient was measured to be 0.48 between the light output loss and the EWRIAC at 15 rad/h although samples have different preexisting absorption.

The EWRIAC values of all samples are less than  $1 m^{-1}$  up to 400 rad/h, indicating no damage to the light response uniformity [2] for PWO crystals used in the CMS ECAL barrel if the LHC luminosity is increased by a factor of ten, i.e. the SLHC. The EWRIAC values measured at 9,000 rad/h, however, are diverse. Some samples show  $3 m^{-1}$ , indicating damages in the light response uniformity and thus the energy resolution [2]. Rigorous quality control on radiation hardness thus is required for PWO crystals used in the endcaps. It is also found that SIC samples have relatively deeper color centers as compared to BTCP samples and thus longer recovery time constants.

Fig. 2 (Left) shows a correlation between the EWRIAC and the variations of the longitudinal transmission at 440 nm for all samples at all three dose rates. This correlation



**FIGURE 2.** Left: Correlations are observed between the EWRIAC and the variations of the longitudinal transmission at 440 nm for all samples in equilibrium under all three dose rates. Right: Correlation between the EWRIAC values measured at 400 and 9,000 rad/h.

can be fit to a 2nd order polynomial, as shown in the figure. This result is consistent with previous investigation where 440 nm was chosen for the monitoring wavelength because of its best linearity [4].

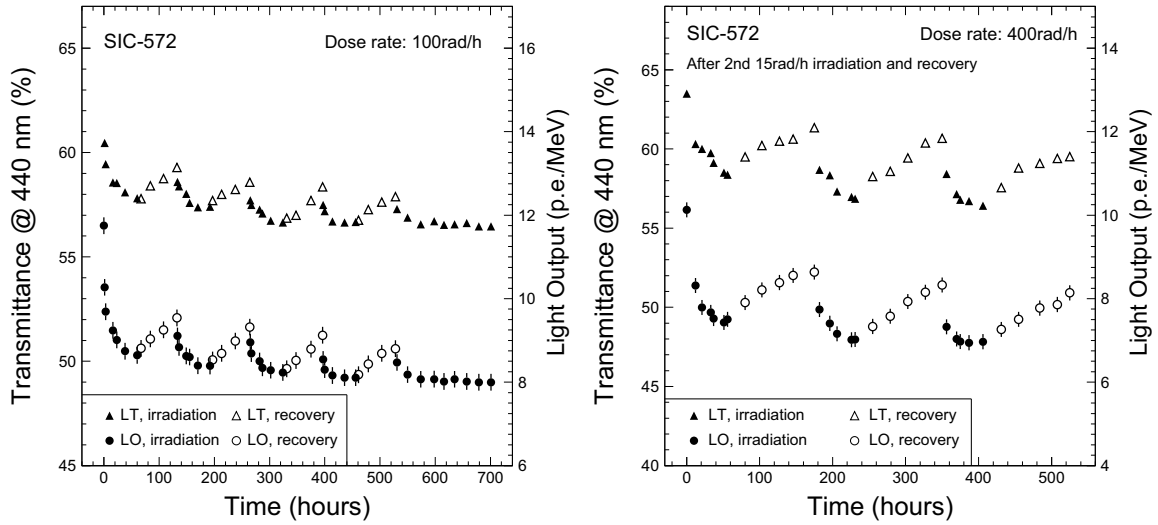
Fig. 2 (Right) shows correlations between the EWRIAC values measured at 9,000 rad/h and 400 rad/h. The linear correlation coefficient is 0.85, which is much larger than 0.69 observed between 400 and 15 rad/h. A weaker correlation observed between low dose rates can be explained by the influence of preexisting absorptions. The strong correlation observed between high dose rates provides a foundation to transfer or compare radiation damage data measured at different laboratories, so is a useful tool for crystal quality control. This correlation was also observed by others [3].

No correlations were observed between initial optical properties and the EWRIAC. The numerical values of the linear correlation coefficient measured between the initial longitudinal transmittance at 360 nm versus the light output loss at 15 rad/h, the EWRIAC at 15 rad/h and the EWRIAC at 9,000 rad/h were found to be 0.19, -0.19 and 0.04 respectively, indicating that preexisting absorption in PWO crystals has no effect on the radiation induced absorption.

## DAMAGE AND RECOVERY CYCLES

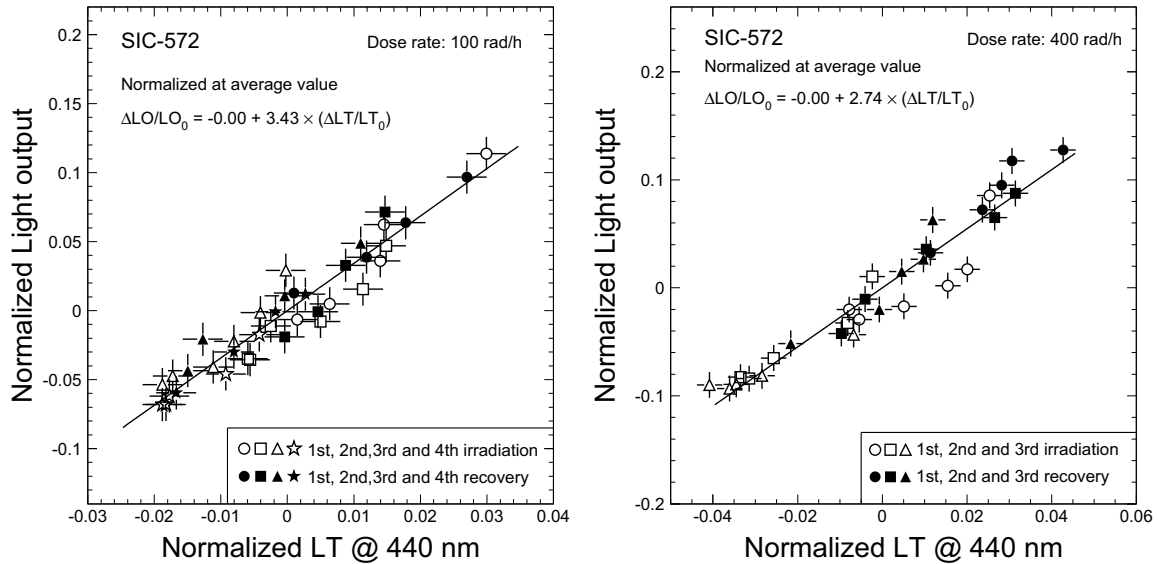
Two SIC samples (570 and 572) and two BTCP samples (2482 and 2531) went through long term irradiation and recovery cycles in a batch at 100 and 400 rad/h. Figs. 3 shows histories of the longitudinal transmission at 440 nm and the light output as a function of time for sample SIC-572.

The observed variations are consistent with our model on color center kinetics based



**FIGURE 3.** Variations of longitudinal transmission and light output are shown as a function of time for sample SIC-572 in multi-cycles of irradiation at 100 rad/h (Left) and 400 rad/h (Right) followed by recoveries.

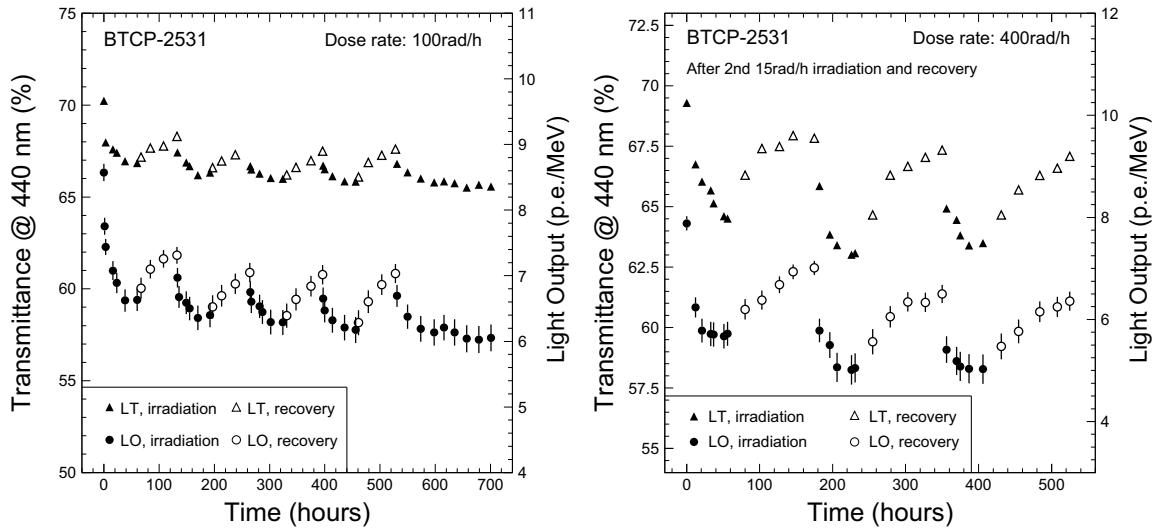
on color center formation (damage) and annihilation (recovery) [2]. Variations of the light output and the longitudinal transmittance are also observed to be correlated. In addition, a longer time is found to be needed to reach equilibrium under 100 rad/h as compared to 400 rad/h, which can be explained by a longer damage time constant at low dose rate [2].



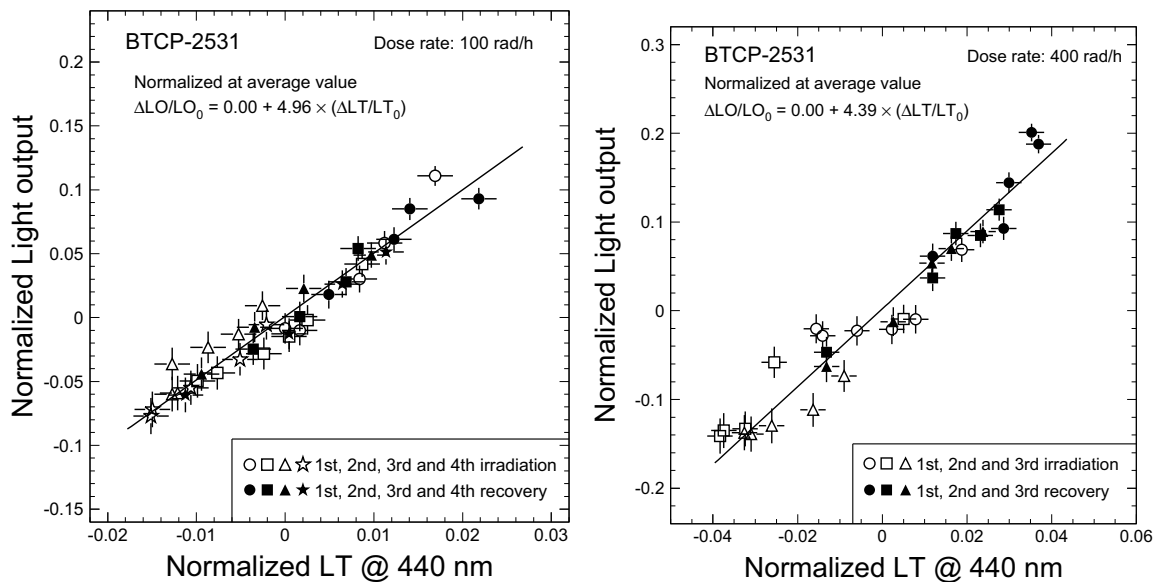
**FIGURE 4.** Normalized variations of the light output are shown as a function of normalized variation of the longitudinal transmission at 440 nm for sample SIC-572 under 100 rad/h (Left) and 400 rad/h (Right).

Fig. 4 presents linear fits to the data shown in Fig. 3 (Left) and (Right), respectively. The excellent linearity indicates that the variation of the light output may be calculated

by using the variations of the longitudinal transmittance at 440 nm. The CMS ECAL monitoring system will use laser pulses at 440 nm to provide crucial monitoring information *in situ* at LHC [5]. It is also interesting to note that the slopes are 3.43 and 2.74 for dose rates of 100 rad/h and 400 rad/h respectively, indicating that this slope is damage level dependent, in addition to the wavelength dependence observed in our previous investigation [4]. The other SIC sample shows similar result.



**FIGURE 5.** Variations of longitudinal transmission and light output are shown as a function of time for sample BTCP-2531 in multi-cycles of irradiation at 100 rad/h (Left) and 400 rad/h followed by recovery (Right) followed by recoveries.



**FIGURE 6.** Normalized variations of the light output are shown as a function of normalized variations of the longitudinal transmission at 440 nm for sample BTCP-2531 under 100 rad/h (Left) and 400 rad/h (Right).

Similarly, Fig. 5 (Left) and (Right) show histories for sample BTCP-2531. Since SIC

samples have relatively deeper color centers as compared to BTCP samples they take a longer time to reach equilibrium at low dose rates. Fig. 6 presents the linearity fits for sample BTCP-2531, showing slopes of 4.96 and 4.29 at 100 rad/h and 400 rad/h respectively. Again, the slope depends on the damage level. The result for the other BTCP sample is also consistent.

The BTCP samples have a slope of a factor of 1.5 larger as compared to the SIC samples. This observation is in a good agreement with the CMS beam test data. Based upon this observation, the variation amplitude of the light output for BTCP crystals *in situ* at LHC thus would be a factor of 1.5 larger as compared to the SIC crystals with the same variation amplitude in the longitudinal transmittance.

## CONCLUSION

By using 54 PWO samples, correlations were observed between initial longitudinal transmittance at 360 nm and initial light output, between the loss of transmittance at 440 nm and EWRIAC, and between EWRIAC measured at different dose rates. No correlations, however, were found between crystal's initial optical properties and radiation hardness.

Excellent linearity was observed between the variations of crystal's light output and its longitudinal transmittance at 440 nm in several cycles of radiation at 100 and 400 rad/h followed by recoveries, indicating PWO crystals can be monitored *in situ* at LHC.

It is also noticed that the ratio between variations of the light output and the longitudinal transmittance is damage level dependent, indicating a necessity to extract this ratio *in situ* at LHC. The BTCP samples have also found to have these ratios of a factor 1.5 of SIC samples, indicating that the former is relatively more difficult to be monitored *in situ* at LHC.

## ACKNOWLEDGMENTS

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