RADIATION DAMAGE MEASUREMENTS OF UNDOPED LEAD TUNGSTATE CRYSTALS FOR THE CMS ELECTROMAGNETIC CALORIMETER AT LHC

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Abstract

A measurement method is described which allows to accurately track the scintillation light yield changes of lead tungstate crystals. The method has been used to study crystals to which post-growth high-temperature annealing treatments in various atmospheres were applied; it has allowed to select the most radiation resistant crystals and thus to optimize the annealing process. We describe the irradiation facility, irradiation procedure and results obtained. This work has led to the production of radiation resistant 50mm long crystals and the process is now being extended to 230mm long crystals.

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1 INTRODUCTION

In the fall of 1994, The Compact Muon Solenoid Collaboration (CMS) at the CERN Large Hadron Collider made the final crystal selection for its electromagnetic calorimeter (ECAL) [1]. The collaboration, after rejecting BaF₂ and CsI early on (See table 1 in Ref.[2]), concentrated on CeF₃ and lead tungstate (PbWO₄) for further evaluation [3],[4]. Because the calorimeter will be operated in a 4T magnetic field, where space is at a premium and because of economics considerations, CMS finally preferred PbWO₄ to CeF₃, judging that the low scintillation light yield could be offset by the use of photodetectors with some internal gain such as avalanche photodiodes or vacuum phototriodes and that adequate temperature regulation and monitoring could take care of the temperature dependence of the light yield (-1.98 % per °C at 20°C, see Fig.6 in Ref. [5]). An early report [5] indicated that undoped crystals emit more than 90% of the light in less than 100ns and that niobium doped crystals were faster and emitted their light with time constants of 3ns (55% of the light) and 14ns (45% of the light); niobium-doped crystals were also reported to show a considerable improvement in radiation hardness over undoped crystals. Unfortunately, most of the crystals received by CMS for the 1995 evaluation of a prototype matrix in a high energy beam at the CERN-SPS accelerator did not come close to these performances, exhibiting a large slow component in the scintillation light emission as well as a significant amount of radiation damage [6],[7], often with some degree of rapid recovery. The slow component was attributed to molybdenum contamination [8], [9] and the low dose radiation damage, shown to affect only the transmission and not the scintillation mechanism, was believed to be due to crystal defects such as oxygen vacancies in the crystal lattice [10] rather than to contaminants.

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The light output degradation saturates at a level dependent on the dose rate [11]. The damage level at saturation is more severe the higher the dose rate used. This is visible in **Fig.1** for a relatively radiation-soft crystal. There, the relative light yield loss is plotted versus cumulated dose, for three different irradiations using dose rates of 0.15, 0.40 and 4.0 Gy/h respectively. The radiation damage from photon irradiation has been experimentally proven to be reversible, and the original light yield of a crystal can be restored after an irradiation, through heating up to 200°C for a few hours. Before each irradiation, the initial light yield of the crystal was restored through such a heating procedure.

The producers have worked systematically to decrease the number of defects in the crystal lattice. For undoped crystals, one way is to fine tune the stoichiometric ratio and to control it with precision during the growth process. Another one, possibly complementary, is to use an optimized post-growth treatment, as described in section 4, to compensate oxygen deficiencies. Feedback from studies such as the present one has helped the producers to fine-tune these processes.

In 1996, some newly delivered crystals showed an improvement but manufacturers lacked the facilities to adequately test slow component and radiation damage, depending mostly on feedback from the test beams; the collaboration decided to set up several irradiation facilities to accelerate the test process and to provide year-round feedback to the manufacturers. We describe here the test facility operated by our group at the Eichlabor of the swiss Paul Scherrer Institut, the test procedure and early results for undoped crystals produced by the Shanghai Institute of Ceramics (SIC).

2 IRRADIATION FACILITY

Irradiations are made with a commercial Siemens medical irradiator containing a 40Tbq ^{60}Co source. The source can be collimated and remotely activated; a light bulb inside the irradiation head illuminates the irradiated area to check its boundaries and position relative to the object to be irradiated. The object to irradiate is placed on a remotely controlled table whose position is accurately controlled to set the dose rate. The whole system is installed in an air conditioned bunker at the PSI Eichlabor [12], a swiss facility with ISO 9000 certification to act as a secondary standard for the calibration of radioactivity detectors. Practical doserates in the range 0.1-10 Gy/h are used in our setup. Usually, one crystal at a time is irradiated and its light yield is monitored in situ before, between and after irradiations to ensure good thermal and optical stability of the system. The total dose varies from 0.5 Gy to 1 kGy, the facility being used by our group typically over periods of four to five days, during which 80-100h of irradiation can be accumulated (this is about the maximum since the facility is shared with other activities, which cannot be interrupted for too long a time).

3 TEST SETUP AND TEST PROCEDURE

The way of irradiating crystals has to take into account the radiation levels expected in the CMS ECAL calorimeter at LHC, where the environment is particularly hostile [13]. At the highest design luminosity of $10^{34} \text{cm}^{-2}\text{s}^{-1}$ one expects at the electromagnetic shower maximum

dose rates around 0.25 Gy/h in the ECAL barrel at $\eta=0$, values around 0.30 Gy/h at the barrel-endcap interface and 15 Gy/h at $\eta=3$. At the back end of the crystals, the dose rates are typically ten times smaller than at the shower maximum. These values lie well above what is expected from electromagnetically interacting particles and are due to the electromagnetic part of the hadronic shower cascades [13].

The irradiation is performed along the side of the crystal rather than from the tip; the exact length being irradiated is adjustable: short crystals are irradiated along their full length whereas full size crystals are irradiated over 2/3 of their length to crudely simulate the actual radiation dose distribution expected in the ECAL. A weak source, usually 3MBq of ^{60}Co , at the tip of the crystal excites its scintillation to serve for light yield measurements. This arrangement guarantees that more than just the tip of the crystal is tested for radiation damage. The crystal is mounted on a Philips XP2262B bialkali photocathode photomultiplier equipped with a standard CERN base type 4238 and either full length or shortened steel housing, held in place by centering rings and pushed against the photocathode by springy foam; optical coupling is achieved with Corning grease Q2-3067. In all cases, a full length μ -metal shield is installed, the photocathode being recessed by 8cm into the μ -metal shield. The whole assembly has its steel shield tip flush mounted into a 5cm thick cylindrical lead shield to protect the photomultiplier; collimators are also adjusted to minimize irradiation of the phototube. The photomultiplier is operated at a gain of around 10^8 ; the high voltage stability is monitored to $\pm 0.05\%$. The anode signal is split, one half of the signal goes via a 100ns delay to a Camac charge integrating LeCroy ADC type 2249, whereas the other half goes to a threshold discriminator, whose outputs are fed into a scaler and into a Camac-controlled precision gate generator which defines the charge integrating time, usually 200ns; the discriminator is set so that the valley below the single photoelectron peak is visible. The scintillation spectra usually show one, two and sometimes three photoelectrons peaks, allowing absolute calibration [Fig.2]. Two Pt₁₀₀ platinum resistors with 4 wire readout are installed in the vicinity of the crystal. The readout electronics is located outside the irradiation room.

Camac is controlled by a Pentium 90 PC running a LabView 4.0 virtual instrument via a HYTEC crate controller, allowing a single ADC channel to be read in at about 2kHz. The Pt₁₀₀ signals are read by the PC via a Keithley 2000 ohmmeter and a National Instruments GPIB controller, giving a relative temperature monitoring accuracy of a few 10^{-3} °C. The ADC data are accumulated into a 1024 channels histogram, temperature data are added and each histogram corresponding to a run of typically 10^6 individual readings is stored as a file onto the PC disk. Periodically, these histograms are transferred via Ethernet to CERN for analysis.

Analysis consists in pedestal verification, single and two photoelectron peaks monitoring for absolute charge calibration, temperature correction and a fit to determine the position of the photoelectric peak from the calibration source. If an accurate light measurement was needed, the fitting procedure would have to precisely model the underlying physics in the observed spectrum, as it has been done in [14]. In the present measurement however, rather

than an accurate absolute light yield, the analysis has to determine precisely the relative shifts in light yield. Thus, an empiric parametrization has been used for the high side of the spectrum, as the sum of an exponential plus a Gaussian function, the position of the Gaussian being used as a measure of the light yield. To obtain the best relative stability of the fitting, both spectrum boundaries for the fit and initial values are kept the same throughout the test of one crystal, consisting of a stabilization phase before irradiation, irradiation and recovery after irradiation. The accuracy of the method can be appreciated in **Fig.3**. There, triangles represent the expected modifications in light yield due to the temperature changes. The dots are the changes in light yield of the crystal. One can observe that, since the crystal was brought into the room from the outside and thus had a different temperature, it took a few hours to thermalize with respect to the room temperature. From there on, its light yield follows the temperature drifts of the room with a slight lag. From the light yield measurements, the stability of the procedure is apparent. The resulting light yield data are sent back to the irradiation facility to help fine tune the irradiation program. An accuracy of better than 3 % is obtained for crystals yielding at least 7 photoelectrons/MeV and around 1 % for 10 photoelectrons/MeV.

The test of a crystal takes approximately 24h. After being placed in the irradiation room, in its position, ready for irradiation, the crystal is let to thermalize for 6h to 8h as explained above. Throughout this time, its light yield is tracked with automated data taking where a run (yielding one accumulated spectrum) of 10^6 events is taken every 15mn. For every run, the average room temperature is recorded as well. After this stabilization period, without any access to the room nor modifications of the setup, the irradiation procedure begins. Every 30mn (or later into the irradiation, every 1h or 2h) the irradiation is interrupted and the light yield is measured in a sequence including a pedestal measurement run and three subsequent light yield measurement runs, each lasting approximately 7mn. The sequence is intended to allow the study of a possible short-term light yield recovery and to detect system instabilities. After several hours of irradiation, the crystal damage typically saturates, (**Fig.1**) and thus the irradiation is stopped. From there on, data taking is continued, in the same mode as for the thermal stabilization, to observe the short and medium-term recovery of the crystal.

4 RESULTS ON UNDOPED CRYSTALS FROM SIC

The test procedure described above was applied to several batches of undoped lead tungstate crystals produced by the Shanghai Institute of Ceramics under contract with ETH. Those crystals, typically cut in pairs from the same “father” ingot, underwent a post-growth treatment where they were annealed in a gaseous atmosphere at temperatures close to their melting point. Such a treatment is believed to allow compensation of oxygen deficiency.

- In a first round of optimizations, air and oxygen were used. **Fig.4** shows such a pair where one crystal was air-annealed and the other one oxygen-annealed; clearly, annealing in an oxygen atmosphere gives better results in term of crystal radiation hardness.
- In a second optimization round, other pairs of crystals were annealed in an oxygen atmosphere using different annealing cycles, whose details are confidential to the crystal pro-

ducers. From the results, an example of which can be seen in **Fig. 5**, the best annealing cycle in oxygen was selected.

- In a third round, the reproducibility of production of radiation resistant, 5 cm long crystal samples was proven, as is demonstrated in **Fig. 6**. All three crystals represented withstand doses rates of 0.25 Gy/h without losing more than 2 % in light yield.
- Although the radiation hardness of these crystals satisfies our early criteria for CMS, we decided to carry out some tests at higher dose rates and integrated dose. An early result is shown in **Fig. 7**, where the dose rate is about 13 times higher than the one expected in the barrel ECAL during normal LHC operation and of the magnitude expected in the end caps. Some of the crystals were then passed to our PSI and Caltech colleagues to undergo higher dose irradiation both with pions and with photons. Crystal SIC153 (**Fig. 6**) has now been subjected at Caltech to 200 kGy at 157 Gy/h with <3 % light yield loss [15].

5 RECOVERY

Fig. 8 shows the light yield change over almost three days, after the irradiation of a relatively radiation-soft crystal has ended. The light yield there is normalized to the value before the start of the irradiation, and this particular example corresponds to the data taken after the irradiation with a dose rate of 0.40 Gy/h of crystal 69 from SIC. A recovery is observed with a time constant of several hours, while no recovery is observed with time constants less than 1 h.

6 CONCLUSIONS

Significant progress has been made in developing undoped PbWO_4 crystals free of the radiation damage problems encountered in earlier crystals, paving the way for the CMS-ECAL construction. The technology has been demonstrated on 50 mm long crystals to allow an easy comparison between crystals from the same ingot and is now being extended to full size, 230 mm long crystals.

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References

- [1] CMS Collaboration, **Compact Muon Solenoid Technical Proposal**, CERN/LHCC 94-38, LHCC/P1 (1994) CERN, Geneva (Switzerland)
- [2] R.Y. Zhu et al., **Crystal Calorimeters for particle physics**, Nucl. Phys. B (Proc. Suppl.) 44 (1995) 88
- [3] E. Auffray et al., **Performance of a Cerium Fluoride crystal matrix measured in high-Energy particle beams**, Nucl. Instr. Meth. A 378 (1996) 171-178.

- [4] G.Alexeev et al., **Studies of lead tungstate crystal matrices in high energy beams for the CMS electromagnetic calorimeter at the LHC**, Nucl. Instr. Meth. A385 (1997) 425-434.
- [5] P.Lecoq et al., **Lead Tungstate (PbWO₄) Scintillators for LHC EM-Calorimetry**, Nucl.Instr.Meth. A365 (1995) 291
- [6] C.Seez, **Test Beam Studies of Lead Tungstate Crystal Matrices for the CMS Electromagnetic Calorimeter**, Proc. of the 6th International Conference on Calorimetry in High Energy, Eds.A.Antonelli et al., Frascati Physics Series (1996) 765-775
- [7] Y.Benhammou, **Test Beam Results of Lead Tungstate Matrices**, Proc. of the 5th International Conference on Advanced Technology and Particle Physics, Como, Italy (1996), Eds.E.Borchi et al., Nucl.Phys.B.Proc.Suppl. 61B (1998) 493-497
- [8] M.Kobayashi et al., **Scintillation and Phosphorescence of PbWO₄ Crystals**, Nucl.Inst.Meth. A373 (1996) 333
- [9] R.Y.Zhu et al., **Lead Tungstate Radiation Damage and Cure**, Proc.of the 6th Int.Conf. on Calorimetry in High Energy Physics, Ed.A.Antonelli et al., Frascati Physics Series (1996) 577
- [10] R.Y.Zhu et al., **A study of the properties of lead tungstate crystals**, Nucl.Instr.Meth. A376 (1996) 319
- [11] R.Y.Zhu, **Precision Crystal Calorimetry in Future High Energy Colliders**, IEEE Trans. Nucl. Sci. NS44 (1997) 468
- [12] K.Gmür and R.Reischi, **Das Eichlabor für Strahlenschutzmessgeräte am PSI**, PSI Bericht 95-04 (1995) ISSN1019-0643, PSI, CH-5232 Villigen, Switzerland
- [13] CMS Collaboration, **The Electromagnetic Calorimeter Project**, Technical design report, CERN/LHCC 97-33. CMS TDR 4 (1997) CERN, Geneva (Switzerland)
- [14] S.Baccaro et al., **Precise determination of the light yield of scintillating crystals**, Nucl.Inst.Meth. A385 (1997) 69-73
- [15] R.Y.Zhu, **Radiation Damage in Scintillating Crystals**, to be published in Proceedings of the 7th International Conference on Calorimetry in High Energy Physics (1997), Tucson, USA

Figure 1: Relative light yield loss for a radiation-soft crystal, under γ -irradiation at dose rates of 0.15 Gy/h (triangles), 0.40 Gy/h (squares) and 4.0 Gy/h (dots). Each data set is well fitted by the combination of an exponential decrease and a constant.

Figure 2: Scintillation spectrum with ^{60}Co for a low light yield crystal, showing one-, two- and a hint of three-photoelectrons peaks, as well as the valley below the single photoelectron peak.

Figure 3: Thermal stabilization plot for a 50mm long crystal before irradiation. **Triangles:** theoretically expected light yield changes due to the temperature drifts of the surrounding air. **Dots:** Light yield of the crystal, as a function of time. Both, data and effect due to temperature are normalized to unity at the end of the stabilization period.

Figure 4: Relative light yield loss of two undoped 50mm long crystals from the same ingot, one air-annealed and the other one oxygen-annealed. The crystals were irradiated laterally along their full length with ^{60}Co at 0.15 Gy/h.

Figure 5: Relative light yield loss of two undoped 50mm long crystals from the same ingot, annealed using different cycles in an oxygen atmosphere. The same measurement method is used as for crystals in the previous figure but at a slightly higher dose rate of 0.25 Gy/h.

Figure 6: Reproducibility of relative light yield loss for three different 50mm long oxygen-annealed crystals. Same measurement method and doserate as in the previous figure. Crystal 153 has since then withstood much higher dose and doserate, as explained in the text.

Figure 7: Relative light yield change for a 50mm long oxygen-annealed crystal irradiated at a higher doserate of 4Gy/h.

Figure 8: Change in light yield observed for a radiation-soft crystal (SIC 69), after gamma-irradiation at a dose rate of 0.40Gy/h up to an integrated dose of 3.2Gy. The values are normalized to the light yield the crystal had at the start of irradiation.