



Improved light yield of lead tungstate scintillators

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Abstract

The application at medium and low energies of lead tungstate scintillators, so far optimized for the ECAL calorimeter of CMS for the future LHC, is strongly limited by their poor light yield. Suitable dopants like molybdenum and terbium can help to overcome this problem. Concepts, results, advantages and drawbacks of this approach are discussed. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Acceptance of lead tungstate (PWO) as scintillator material for high-energy physics experiments at LHC [1] is based on a number of unique properties of this compound, on successful beam tests with high-energy electrons [2] and several attempts to improve its properties [3]. Optimization of crystal growth conditions led to samples with very small radiation-induced changes in their transmission spectrum [4] and light yields of 15–17 phe/MeV in the fast (10 ns) scintillation component out of a volume of a few cm³. This corresponds to 10–12 phe/MeV for scintillator elements having the full size of the CMS Electromagnetic Calorimeter (ECAL) specification (total length ~ 25 radiation lengths).

At medium energies far below 1 GeV very promising energy and time resolutions ($\sigma_E/E = 1.54\%/\sqrt{E(\text{GeV})} + 0.30\%$, $\sigma_t = 130$ ps) have been achieved using monoenergetic photons and electrons between 50 and 850 MeV, respectively [5,6]. These experiments show that the energy response below 200 MeV is significantly limited by the photon statistics of the detected scintillation yield. Until now, however, properties of PWO were especially optimized for application in LHC detectors where radiation hardness and fast scintillation play an important role, but light yield remains less critical. Therefore, we started first attempts to improve the light yield of PWO scintillators by various additional dopants with the aim to make this material suitable for applications not only in CMS ECAL, but also in ALICE experiment at LHC where relatively low rate is needed and other experiments requiring detection of γ -quanta of relatively low energy.

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2. Experimental conditions

Six lead tungstate single-crystals grown at the Bogoroditsk Techno-Chemical Plant (Bogoroditsk, Russia) under conditions described elsewhere [4] have been investigated: One series doped with Mo (Mo I: 200 ppm, Mo II: 500 ppm) and Mo plus La (Mo III: 500 + 100 ppm, respectively), and one series doped with Tb (Tb I: 100 ppm, Tb II: 500 ppm and Tb III: 2000 ppm in the melt). Samples with dimensions of $1 \times 1 \times 0.5 \text{ cm}^3$ have been prepared for the spectroscopic measurements from the bottom parts of ingots of 240 mm length. These samples were also irradiated with a ^{60}Co -source (100 Gy/min) to determine their radiation hardness by measuring the induced absorption at 400 nm. Light yield measurements have been done on cube-shaped samples 1.5 resp. 2 cm in size and wrapped in Teflon tape. The light output at room temperature has been collected by a 2" photomultiplier tube (Hamamatsu R2059-01, fused silica window, bialkali photocathode) and integrated in a charge-sensitive ADC varying the gate width between 0.25 and 4 μs .

3. Results

Using a PWO crystal which fulfills the CMS/ECAL specifications as reference (Nb/Y-PWO in Fig. 1), the response to low-energy

γ -sources (^{137}Cs (662 keV)) has been determined. The obtained pulse height spectra (integration gate 2.5 μs) show a significant increase of light output and improvement in resolution due to photon statistics for samples doped with Tb and Mo. An energy resolution (FWHM) of $\Delta E/E < 50\%$ ($\sigma/E < 21\%$) can be achieved for the latter crystal. In order to compare the primary light yields, a scaling factor of 1.8 has to be considered to correct for the reduced quantum efficiency of the photocathode at the Mo emission wavelength. Further results of our investigation regarding important parameters of a scintillator are presented in Table 1 in comparison with those of the crystal obtained

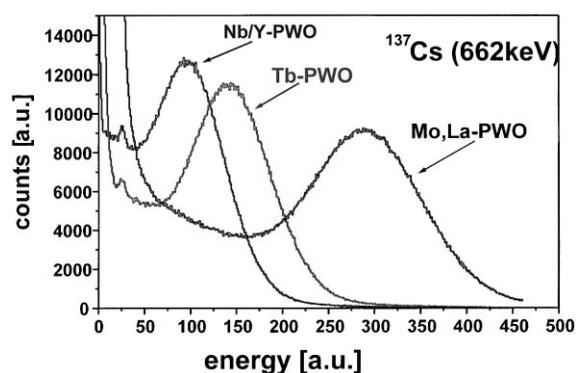


Fig. 1. Response to low-energy photons measured for Tb- and Mo-doped PbWO_4 -samples in comparison to a crystal which fulfills CMS/ECAL specifications (Nb/Y-PWO).

Table 1

Scintillation properties of several doped PWO crystals measured at room temperature

Sample	Light yield measured within 1500 ns, (phe/MeV)	Fraction of the LY, emitted within 30/100/1000 ns, (%)	Maximum wave-length of radio-luminescence, (nm)	Induced absorption at 400 nm, (m^{-1})
Regular	16	85/95/99	420	0.3
Mo I	33	56/68/95	500	– 7
Mo II	38	46/61/94	500	– 6
Mo III	56	44/60/93	500	1.1
Tb I	27	98/99.5/99.9	348–587 (Tb^{3+}) 420	2.2
Tb II	17	97/98/99.2	348–587 (Tb^{3+}) 420	0.8
Tb III	8	91/92/93.5	382–587 (Tb^{3+}) 420	0.5

from the regular production for LHC experiments. They show the following.

Doping with Mo shifts the maximum of the scintillation towards green (maximum of luminescence at 500 nm) due to the emission of MoO_4^{2-} oxy-complex [7], whereas the blue luminescence (peaking at 420 nm) of regular WO_4^{2-} groups overlapped by narrow Tb^{3+} emissions (at 348, 382, 410, 440, 490, 548 and 587 nm) originating from the terms $^5\text{D}_2$, $^5\text{D}_3$ and $^5\text{D}_4$ is observed in samples Tb I–III. Their relative contributions change with activator content. At low Tb^{3+} concentration radiative transitions $^5\text{D}_3 \rightarrow ^7\text{F}_{0-6}$ located in the blue region dominate whereas green–red $^5\text{D}_4 \rightarrow ^7\text{F}_{0-6}$ luminescence prevails at increasing activator concentration, and luminescence of the regular tungstate groups gets strongly suppressed.

In both series of crystals a remarkable increase of light yield is observed. This is due to the existence of additional channels for radiative recombination at suitable luminescence centers. As was already stated [8], the first excited state of such a center has to have an energy close to or little below that of the lowest zero phonon transition of a regular anionic oxy-complex (estimated value is $27\,000\text{ cm}^{-1}$) of lead tungstate. For crystals doped with Mo this condition is fulfilled, the energy of the first excited level of $(\text{WO}_4)^{2-}$ being 0.45 eV above that of the $(\text{MoO}_4)^{2-}$ complex. Additional doping with La creates two shallow electron centers [9] with energy levels close to the conduction band, but above the radiating level of $(\text{MoO}_4)^{2-}$. Therefore, an additional source for electrons feeding the Mo emission is created and the light yield is further increased. Tb^{3+} provides excited f levels ($^5\text{D}_3$) close to the first excited level of $(\text{WO}_4)^{2-}$, too, but also one ($^5\text{D}_4$) deeper in the gap and some (like $^5\text{D}_2$) near the conduction band. There additional luminescence practically doubles the light yield at low activator concentration, but at higher concentration quenching processes (like the up-conversion $^5\text{D}_3 \rightarrow ^7\text{F}_1 + h\nu$, $^7\text{F}_6 + h\nu \rightarrow ^5\text{D}_4$) lead to a decrease of the light yield again.

Both doping approaches suffer from a reduced radiation hardness compared to the standard PWO crystals. The only exception is Tb III, which on the other hand shows an increased contribution of a slow scintillation component stemming from

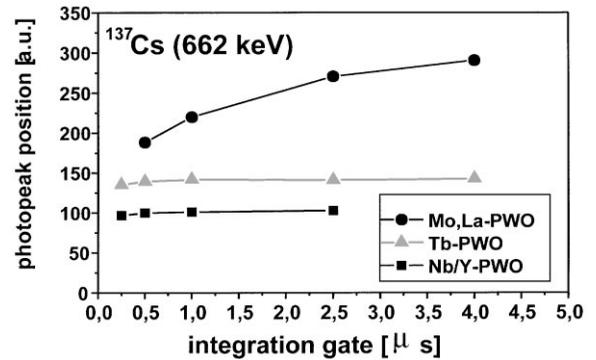


Fig. 2. Measured position of the photopeak (pedestal not subtracted) of the ^{137}Cs (662 keV) source as a function of the integration width for the three samples used in Fig. 1.

the $^5\text{D}_4$ luminescence. The fraction of such slow components is especially high in Mo-doped samples due to the longer decay times of the $(\text{MoO}_4)^{2-}$ complex as well as a contribution of shallow $(\text{MoO}_4)^{3-}$ traps [10] and also cannot be reduced by co-doping with La. Fig. 2 illustrates the difference in light output and decay time by comparing the measured position of the photopeak of the ^{137}Cs (662 keV) source as a function of the width of the integration gate. Whereas nearly the full light yield can be collected within far less than 500 ns in the case of the reference and the Tb-doped PWO, the Mo-doped scintillator requires an integration over several microseconds. Therefore, future applications of PWO:Mo have to cope with limited count-rate capabilities to avoid pile-up, but for the advantage of a very compact scintillator with light yield sufficient for applications at lower energies.

Finally, it should be noted that radiation hardness of both types of crystals certainly can be improved by the methods described earlier [11].

4. Conclusion

A new optimization has been found which significantly increases the light yield of lead tungstate crystals. Depending on the applications, an improvement of nearly a factor of 2 can be obtained as compared to the best crystals obtained so far for the CMS experiment at CERN. In this case, the crystals are doped with Terbium and the emission

is as fast as for the standard Y + Nb doping (about 99% of the light is collected in the first 100 ns). An improvement of nearly a factor of 4 can be obtained with co-doping (molybdenum plus lanthanum) at the price of a longer decay time. This work opens new perspectives for application of lead tungstate crystals in medium- and low-energy experiments.

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