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Scintillation Characteristics of Doped PbWO₄ Crystals *

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Emission spectra, longitudinal transmission and light output for the Sb-doped and Y-doped PbWO₄ crystals are presented. Scintillation decay kinetics has been measured by using gate integration and the single photon counting method with blue and green filters. Radiation hardness has also been measured by using ⁶⁰ Co γ -ray irradiation under different dose rates and different dose rate profiles. The light yield and radiation hardness of the Sb-doped and Y-doped PbWO₄ crystals are improved as compared to the undoped crystals and are suitable for the requirements of compact muon solenoid.

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PbWO₄ (PWO) crystals are chosen by compact muon solenoid (CMS) experiment at CERN (European Laboratory for Particle Physics) as the scintillation element for a electromagnetic calorimeter (ECAL). To construct a precision ECAL, the PWO crystals are required to have good optical transmittance, adequate light yield (larger than eight photoelectron/MeV measured by an XP2262B PMT), fast decay time (at least 90% of scintillation light is emitted within less than 100 ns) and good radiation hardness (a loss of light yield less than a few per cent of total light output) in situ at the large hadron collider (LHC).^[1]

The quality of PWO crystals depends on the purity of raw materials, stoichiometry tuning and growth parameters. However, a complete suppression of impurities and structure defects in full size PWO crystals is very difficult. Doping with trivalent or pentavalent ions is thus considered to be an effective alternative approach to compensate defects.^[2,3] Here, the results of measurements on several Sb-doped and Y-doped PWO samples, which were grown by the modified Bridgeman method at the Shanghai Institute of Ceramics, Chinese Academy of Sciences, are presented.

The Sb-doped and Y-doped PWO samples were annealed in air. Typical photoluminescence spectra, measured by a HITACHI 850 spectro-fluorometer with UV excitation, are shown in Fig. 1. The spectra of the Sb-doped and Y-doped PWO are peaked at 530 nm and 420 nm, respectively.

Optical longitudinal transmission of crystals, measured by using a SHIMADZU MPC-2200 spectrophotometer, is shown in Fig. 2. Transmission of both the doping PWO around 320–350 nm is improved as compared to the undoped crystals,^[4,5] but the Sb-doped sample still has significant absorption at 420 nm.



Fig. 1. Emission spectra of the Y-doped and Sb-doped PWO excited by UV.



Fig. 2. Longitudinal transmittance of the SIC Y-doped and Sb-doped PWO crystals.

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Fig. 3. Decay time of crystal SIC-278.

Table 1. Light yield (L1) of sample	Table 1.	Light	yield	(LY)	of sample
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Sample No	. Size	e(mm)	Doping	g LY
				$({\rm photoelectron}/{\rm MeV})$
SIC-674	$22^2 \times 2$	$26^2 \times 23$	0 Y	13.4
SIC-702	$22^2 \times 2$	$26^2 \times 23$	0 Y	13.5
SIC-S767	$22^2 \times 2$	$26^{2} \times 23$	0 Y	14.5
SIC-S722	$22^2 \times 2$	$26^2 \times 23$	0 Y	13.3
SIC-278	$22^2 \times 2$	$26^2 \times 23$	0 Sb	10.7
SIC-289	$25^2 \times 2$	$25^2 \times 18$	0 Sb	11.3
SIC-294	$22^2 \times 2$	$26^{2} \times 23$	0 Sb	13.1
SIC-295	$22^2 \times 2$	$26^{2} \times 23$	0 Sb	12.9

Table 2. Ratio of fast component.

Sample No.	Doping	LY(100 ns)/LY(1000 ns) (%)
SIC-S392	Y	97
SIC-S412	Υ	99
SIC-606	Υ	99
SIC-S643	Y	98
SIC-278	\mathbf{Sb}	96
SIC-289	\mathbf{Sb}	95
SIC-294	$^{\mathrm{Sb}}$	96
SIC-295	$^{\rm Sb}$	93

Light output was measured by using XP2262B PMT with silicon grease (n = 1.5) coupling. The samples were excited by a radioactive source. The photon current measured was integrated by using a LeCroy QVT 3001 with a gate width of 200 ns.^[6] A special Roma fit method ^[7] was used to determine the light yield. Table 1 lists the dimensions of samples and their light yield corrected to 18 °C. The light outputs of the Sb-doped and Y-doped samples are improved as compared to the undoped crystals and both exceed the requirement of CMS. The scintillating decay kinetics was measured by two methods. One is to measure the light output with different gate widths. The ratios between light outputs with 100 ns and 1000 ns gate widths are listed in Table 2. Another is to measure the single photon counting spectrum ^[8] by using a blue filter ($\lambda =$ 443 ± 16), a green filter ($\lambda = 535 \pm 15$) and without a filter. The decay kinetics can be fitted to a sum of three exponentials. Figure 3 shows the result for SIC-278 in the time range of 1200 ns. The numerical results obtained by using these two methods are consistent.



Fig. 4. Dose profile of lateral irradiation with and without lead mask: dots, LHC dose profile; solid line, Co-60 photons side; dotted line, Co-60 photons side, Pb masked; dashed line, Co-60 photons front.



Fig. 5. Light yield loss of SIC-278 under different dose distributions.

The barrel crystal of CMS ECAL will be exposed to a dose rate from 15 to 30 rad/h at shower maximum *in situ* at LHC. It is required that the loss of crystal light output is less than a few per cent with the LHC dose rate profile. Samples were irradiated laterally at the University of Science and Technology of China by using a 2.6 Ci 60 Co source. The top 18 cm length of full size samples was irradiated. The bottom 5 cm of the sample was shielded by 5 cm lead and 0.6 cm iron. The room temperature was controlled at 18 ± 1 °C. The sample temperature was measured and used for light output corrections. The light output was measured when irradiation was interrupted. The cycle time for light yield measurement was about 40 min including 20 min of stabilization time for the PMT. The irradiation time was longer than 24 h, so that light output of the sample reached the equilibrium.^[9] Results of some typical samples are listed in Table 3.

Table 3. Loss of light yield after side irradiation.

Sample No.	Doping	LY loss $(\%)$				
		$20 \ \rm rad/h$	$15~\mathrm{rad/h}$	$75~\mathrm{rad/h}$	$100 \ rad/h$	
SIC-695	Υ		-5.0		-16.6	
SIC-702	Υ		+4.0		-10.8	
SIC-S767	Y		-9.0		-15.7	
SIC-S754	Y		-6.3			
SIC-278	$^{\rm Sb}$	-12.5		-20.0		
SIC-289	$^{\rm Sb}$	-7.5		-11.5		
SIC-294	$^{\rm Sb}$	-12.0				
SIC-295	\mathbf{Sb}	-7.0				

It is not easy to exactly reproduce the LHC dose profile.^[10] To simulate the longitudinal dose rate distribution, we used a lead mask with different thicknesses. Figure 4 shows the dose rate profile. The loss of light output of SIC-278 under irradiation with and without a lead mask is shown in Fig. 5. The ratio of the loss of light output with mask to the 18 cm lateral irradiation is in the range 1:1.5-2.

In conclusion, it is considered that doping by Sb^{3+} or Y^{5+} may cause oxygen leakage in PWO and thereby reduce the density of Pb^{3+} and O^- defects, which are thought to be the main defects of PWO crystals. Our result shows the quality improvement by Sb or Y doping. Light output and decay kinetics of all samples are good enough for CMS. Significant improvement in radiation hardness is also found as compared to undoped crystals (normally around 20% light loss). Ydoped PWO has less radiation damage and more fast components than Sb-doped crystals.

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