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New types of lead tungstate crystals with high light yield

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Abstract

Because of their high stopping power and fast scintillation, lead tungstate crystals have attracted much attention in the high energy physics and nuclear physics communities. The use of lead tungstate, however, is limited by its low light output. An effort has been made at the Shanghai Institute of Ceramics to improve this. The results indicate that a factor of ten increase of the light output, mainly in the microsecond decay component, may be achieved. The photo luminescence spectrum, light output and decay kinetics of new samples are presented. Longitudinal uniformity of a sample of 22 radiation lengths is studied. Possible applications for calorimetry in high energy and nuclear physics experiments are discussed.

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Keywords: Crystals; Scintillation; Lead tungstate; Light output

1. Introduction

In the last five years an extensive R&D program has been carried out by the Compact Muon Solenoid (CMS) experiment in developing lead tungstate ($PbWO_4$) crystals to be used in the Large Hadron Collider (LHC). As a result of this development program, $PbWO_4$ crystal is now a mature material in market with low cost. It, however, is interesting to note that the yttrium doped $PbWO_4$ crystals chosen by CMS have limited light output, about 10 p.e./MeV for full size samples measured with a photo multiplier (PMT) of bi-alkali cathode. This limits their application in areas other than high energy and nuclear physics. There are also issues not fully understood in the nature of scintillation for tungstate family. For example, why trivalent doping, such as lanthanum and yttrium, at less than 100 ppm level changes scintillation to blue while oxygen compensation changes scintillation of undoped $PbWO_4$ crystal to green? Why trivalent doped $PbWO_4$ crystals have only fast scintillation while most members of tungstate family have extensive slow scintillation? And, is there any special processing which would make $PbWO_4$ crystals providing higher light yield than CMS crystals? The answer to the last question is positive.

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The fundamental approach to modify scintillation property of a material is two-folds. One is through modifying crystal structure by changing growth parameter. Doping during crystal growth is another approach which may compensate structure defects, eliminate unwanted impurities and change scintillation properties [1–5]. Early Glow Discharge Mass Spectroscopy (GDMS) analysis revealed that contaminations of certain cation, especially Mo, were responsible for the slow scintillation component in $PbWO_4$, as reported by Kobayashi et al. [6] and Zhu et al. [7]. On the other hand, Mo doping introduces a significant fraction of the slow component and thus increases the light output in $PbWO_4$ crystals. Following this line, $PbWO_4$ samples doped with various dopant were grown and were found with significant increase of light yield [8,9]. In this paper we present scintillation and other optical properties of $PbWO_4$ crystals doped with two special dopant A and B.[1] It is found that light output of up to ten-folds of that of the CMS yttrium doped $PbWO_4$ crystal, mainly in the microsecond decay component, can be achieved. $PbWO_4$ crystals of this type may find applications in high energy and nuclear physics experiments, such as crystal calorimeters in future electron linear colliders or in heavy ion colliders, where interaction cross-section allows an integration time of a few μs.

2. Samples

A total of ten samples, grown by a modified Bridgman method at Shanghai Institute of Ceramics (SIC), China, were studied. Table 1 lists their ID number, dimension, dopant and peak of the photo luminescence. As a comparison, a standard CMS yttrium doped sample S762, which is a 23 cm long tapered from $2.2 \times 2.2\,cm^2$ to $2.6 \times 2.6\,cm^2$, is also listed in this table. Two different dopants (A and B) were introduced at different levels in the melt during growth.

All samples have rectangular shape with all surfaces polished. No further treatment, other than simple cleaning with alcohol, was carried out before measurements. The as-grown samples are transparent, colourless without visible defects, such as cracking, inclusions, scattering particles and growth striation. Both A and B dopings do not change crystal structure, no other phase was observed in the XRD spectra [8].

[1] Pending on patent application, the chemical nature of particular dopant is not released at present.

Table 1
List of samples investigated in this paper

ID	Dimension (cm)	λ_{pho} (nm)
Samples doped with dopant A		
S25	$2.9 \times 9.5 \times 2.9$	560
S27	$2.0 \times 12.0 \times 2.0$	560
Z9	$2.0 \times 19.8 \times 2.0$	560
Z14	$2.0 \times 17.9 \times 2.0$	560
Z22	$2.0 \times 16.0 \times 2.0$	560
Z23	$2.0 \times 9.7 \times 2.0$	560
Z24	$2.0 \times 3.0 \times 2.0$	560
Z25	$2.0 \times 12.0 \times 2.0$	560
Samples doped with dopant B		
Z20	$2.0 \times 14.0 \times 2.0$	560
Z21	$2.0 \times 10.3 \times 2.0$	560
A standard CMS yttrium doped sample		
S762	$2.2 \times 23.0 \times 2.6$	420

3. Properties of the new type $PbWO_4$ crystals

Photoluminesence, longitudinal transmittance, light output, decay kinetics as well as longitudinal uniformity were measured at Caltech, more detailed discussions on the equipment and technique used for the crystal characterization can be found in ref. [7].

Fig. 1 shows the photo luminescence spectra for an A doped sample Z24, a B doped sample Z20 and a standard CMS yttrium doped sample S762, both A and B doped samples have similar photo luminescence peaked at 560–600 nm, while that from sample S762 is peaked at 420 nm. This shows that the scintillation of these new types of $PbWO_4$ crystals is mainly in green, contrary to the blue of standard CMS yttrium doped $PbWO_4$ crystals.

Table 2 lists the light output integrated in five different gate widths for all $PbWO_4$ samples listed

in Table 1. Also listed in the table is the ratio of light outputs between 50, 100 and 2,000 ns. Significant increase of light output, especially in slow scintillation component, is observed for samples doped with A and B.

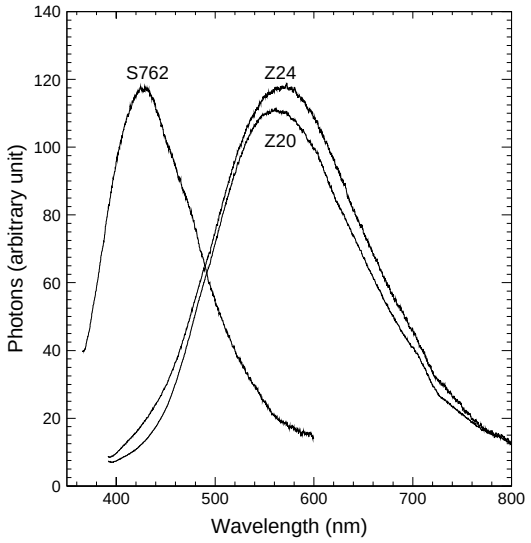

Fig. 1. Photo luminescence spectra for samples Z24, Z20 and S762.

With light output measured as a function of integration time, the scintillation decay kinetics of the samples was determined. Fig. 2 shows a comparison of light outputs, in photoelectron per MeV, as a function of the integration time for samples Z24, Z20 and S762. The A and B doped samples have significant additional slow component.

Z20 and Z24 provide photo electron yield of 5 to 8 times of that of the yttrium doped CMS crystal with tail end and seed end coupled to the PMT respectively, as shown in Table 2. Fig. 3 shows distributions of the quantum efficiency of the R2059 PMT and corresponding emission spectra for a CMS choice of yttrium doped $PbWO_4$ sample S762 and sample Z24. The corresponding emission weighted quantum efficiencies are $(13.7\pm0.3)\%$ and $(5.3\pm0.1)\%$, respectively, for S762 and Z24. The difference of the PMT response thus is a factor 2.6 for these two types of crystals. Calculation by using emission of sample Z20 shows the same result. The light output of sample

Table 2
Summary of $PbWO_4$ light output (p.e./MeV)

Sample ID	Gate width (ns)					Fraction (%)	
	50	100	200	1000	2000	a[a]	b[b]
S25[s c]	10.2	14.8	22.3	49.2	55.4	18	27
S25[t d]	10.5	13.8	17.7	29.8	31.8	33	43
S27[s]	11.3	15.2	20.4	40.5	46.1	25	33
S27[t]	12.5	15.7	17.0	18.9	19.4	64	81
Z9[s]	6.1	8.3	11.1	22.4	26.0	24	32
Z9[t]	6.0	7.9	8.7	9.0	9.1	66	87
Z23[s]	21.0	27.3	31.5	40.4	41.8	50	65
Z23[t]	20.3	25.4	27.4	29.7	30.2	67	84
Z24[s]	22.3	28.4	36.5	71.0	82.5	27	34
Z24[t]	22.0	27.5	34.5	63.1	72.4	30	38
Z20[s]	8.2	9.5	9.7	9.8	9.9	83	96
Z20[t]	9.9	13.7	19.9	46.0	54.3	18	25
Z21[s]	21.3	28.0	31.3	34.5	35.1	61	80
Z21[t]	20.5	28.5	34.4	42.0	42.4	48	67
S762	9.3	10.3	10.4	10.4	10.4	89	99

[a] 50 ns/2 μs.
[b] 100 ns/2 μs.
[c s] Represent sample's seed end coupled to the PMT.
[d t] Represent sample's tail end coupled to the PMT.

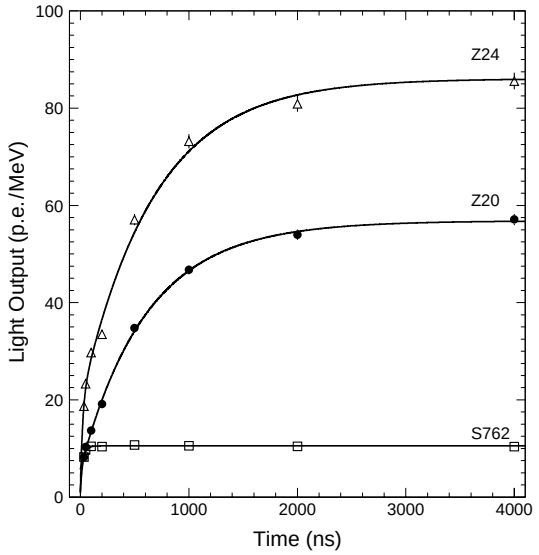

Fig. 2. The light output is shown as a function of integration time for samples Z24, Z20 and S762.

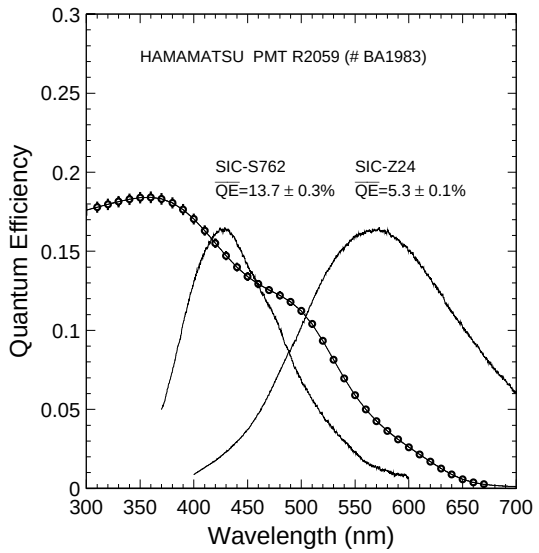

Fig. 3. Quantum efficiency of the R2059 PMT is shown as function of wavelength together with emission spectra of sample Z24 and S762.

Z20 and Z24 in photons received by PMT per MeV energy deposition thus is 13 to 21 times of that of the standard yttrium doped CMS $PbWO_4$. This number, however, has not taken into account the difference of the light path or crystal size. Taking into account the difference of the light path, 3 cm of Z24, 14 cm of Z20 and 23 cm of S762, our estimation is that up to a factor of ten increase in light output, mainly in μsec decay component, is expected for new types of samples as compared to that of the standard yttrium doped CMS $PbWO_4$ samples.

One important technical issue for doping is the uniformity. Our measurement shows that both dopants A and B are not uniformly distributed in $PbWO_4$. Table 2 shows that all A doped samples provide significant more slow component when the seed end is coupled to the PMT, while all the B doped samples provide significant more slow component when the tail end is coupled to the PMT. This indicates that the dopant A is concentrated at the tail end, and dopant B is concentrated at the seed end. In other words, the segregation coefficient of dopant A in $PbWO_4$ is less than one, and that of dopant B is larger than one.

4. Summary

In the last two years SIC has made an effort in developing new types of $PbWO_4$ crystals with high light yield. It is encouraging to find both dopant A and B are effective in increasing $PbWO_4$ light output, and light output increase of up to ten-fold is observed for these doped samples as compared to that of the standard yttrium doped CMS sample. We, however, have not be able to observe a dopant which cause significant more fast component. This increase of slow component is encouraging for users in high energy and nuclear physics field, but may still fall short for medical applications.

Both A and B doppings show bad longitudinal uniformity. One interesting approach thus is to double dope $PbWO_4$ crystals with both A and B. Since these two dopants have similar function but with rather different segregation coefficients, it is hoped that they would compensate each other by double doping and make large size, longitudinally uniform crystals.

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