New Types of Lead Tungstate Crystals with High Light Yield for Future Particle Physics Experiment

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Because of their high stopping power and fast scintillation, lead tungstate crystals have attracted much attention in the high energy and nuclear physics community. The use of lead tungstate crystals, however, is limited by its low light output. An effort has been made to improve this. The result indicates that a factor of ten increase of the light output, mainly in the microsecond decay component, may be achieved. Possible applications for calorimetry in high energy and nuclear physics experiments are discussed.

In the last six 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, large size (25 X_0) yttrium doped $PbWO_4$ crystals with fast scintillation light are produced in Bogoroditsk Techno-Chemical Plant (BTCP) in Tulla, Russia, and in Shanghai Institute of Ceramics (SIC) in Shanghai, China. Because of this development, $PbWO_4$ 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.

Early Glow Discharge Mass Spectroscopy (GDMS) analysis revealed that contaminations of certain cation, especially molybdenum, were responsible for the slow scintillation component in $PbWO_4$, as reported by Kobayashi et al. [1] and Zhu et al. [2]. Following this line, $PbWO_4$ samples doped with various dopant were grown and were found with significant increase of light yield [3, 4]. In this note we present scintillation properties of $PbWO_4$ crystals grown at SIC which were doped with two special dopant A and B [5]. It is found that light output of up to ten folds of that of the yttrium doped $PbWO_4$ crystal, mainly in the microsecond decay component, can be achieved.

Samples grown by a modified Bridgman method. Measurements were made for these samples, and were compared to a yttrium doped sample S762. While samples S25, S27, Z9, Z23, Z24 and Z25 are doped with dopant A at different levels in the melt during growth, samples Z20 and Z21 are doped with dopant B. All samples have rectangular shape of 3 to 20 cm long with all surfaces polished, except sample S762, which is 23 cm long tapered from 2.2×2.2 cm^2 to 2.6×2.6 cm^2.

Photo luminescence, longitudinal transmittance, light output, decay kinetics as well as longitudinal uniformity were measured at Caltech and cross checked at Brookhaven National Laboratory. Detailed discussions of the equipment and technique used in these measurements can be found in [2].

The left side of Figure 1 shows that both A and B doped samples have similar photo luminescence peaked at 560 nm, while that from sample S762 is peaked at 420 nm, indicating that the scintillation of these new types of $PbWO_4$ crystals is mainly in green, contrary to the blue of yttrium doped $PbWO_4$ crystals. The right side of Figure 1 shows that both A and B doped samples have significantly more light than CMS sample, and the A doped sample Z24 has more light than the B doped sample Z20. Table I lists the light output integrated in five different gate widths. 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.

With light output measured as a function of integration time, the scintillation decay kinetics

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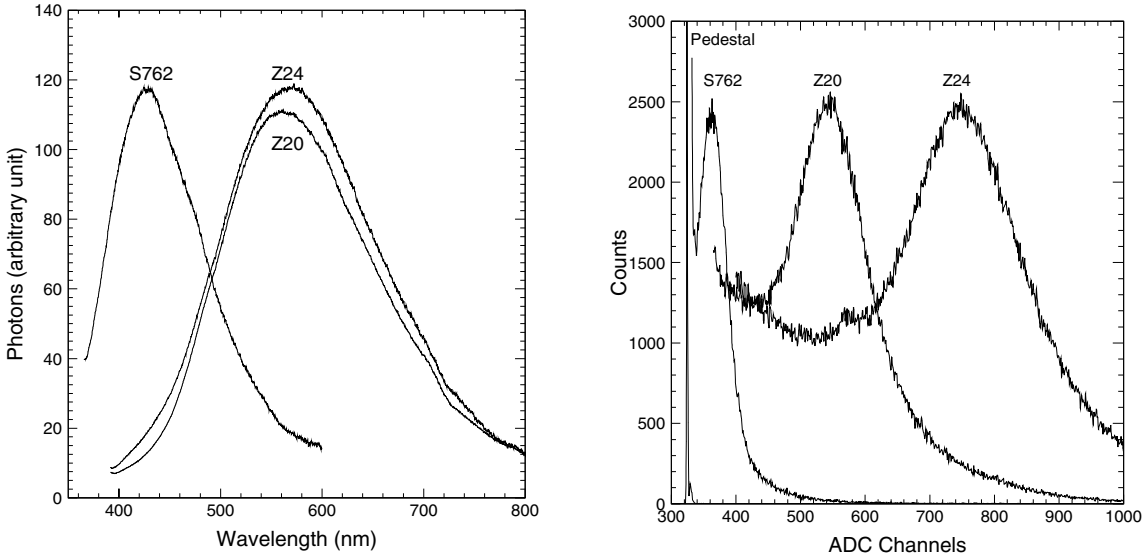

Figure 1: Left: Photo luminescence spectra for samples Z24, Z20 and S762. Right: ^{137}Cs spectra integrated in 2μs for samples Z24, Z20 and S762.

Table I Summary of $PbWO_4$ Light Output (p.e./MeV)

Sample	Gate width (ns)					Fraction(%)	
ID	50	100	200	1,000	2,000	$\frac{50ns}{2\mu s}$	$\frac{100ns}{2\mu s}$
S25^s	10.2	14.8	22.3	49.2	55.4	18	27
S25^t	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

^{s} represents sample's seed end coupled to the PMT.

^{t} represents sample's tail end coupled to the PMT.

of the samples was determined. The left side of Figure 2 shows a comparison of light outputs, in photoelectron per MeV, as a function of the integration time for samples Z24, Z20 and S762. On the face value samples Z20 and Z24 provide photo electron yield of 5 to 8 times of that of the yttrium doped $PbWO_4$ crystals of CMS size. The right side of Figure 2 shows distributions of the quantum efficiency of the R2059 PMT and corresponding emission spectra for a yttrium doped $PbWO_4$ sample S762 and sample Z24. The corresponding emission weighted quantum efficiencies are $(13.7 \pm 0.3)\%$ and $(5.3 \pm 0.1)\%$ 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 Z20 and Z24 in photons received by PMT per MeV energy deposition thus is 13 to 21 times of that of the yttrium doped $PbWO_4$

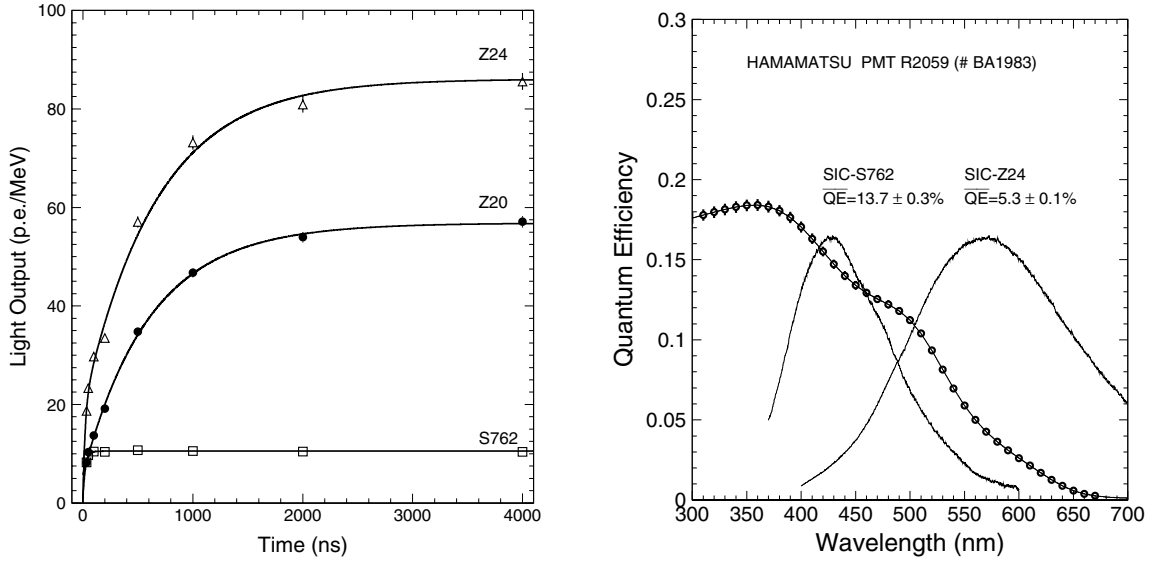

Figure 2: Left: The light output is shown as a function of integration time for samples Z24 ($2 \times 2 \times 3$ cm), Z20 ($2 \times 2 \times 14$ cm) and S762 ($2.2^2 \times 2.6^2 \times 23$ cm). Right: Quantum efficiency of the R2059 PMT is shown as function of wavelength together with emission spectra of sample Z24 and yttrium doped sample S762.

crystals of the CMS size. 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 $PbWO_4$ samples as compared to that of the yttrium doped samples of CMS size.

One important technical issue for doping is the uniformity. Since the segregation coefficient of a dopant in $PbWO_4$ crystals is usually not equal to one, the dopant tends to distribute not uniformly in the crystal. This would in turn cause bad longitudinal uniformity for large size samples. This bad longitudinal uniformity, if beyond some limit, may affect calorimeter performance. Our measurement shows that both dopants A and B are not uniformly distributed in $PbWO_4$. Table I shows that all A doped samples provide significant more slow component when the seed end is coupled to the PMT, while it is the tail end coupled to the PMT for the B doped samples. 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. This conclusion was also confirmed by measurement of emission and transmittance spectra.

In a brief summary, dopant A and B are effective in increasing light output, and up to ten folds increase, as compared to that of the yttrium doped $PbWO_4$ crystals of CMS size, is observed. Both A and B dopings has 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 when double doped for samples of large size. When this is achieved, we will have a new type of $PbWO_4$ crystals. $PbWO_4$ 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.

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