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Characterization of optical properties on large-size $PbWO_4$:Y crystals grown by modified Bridgman method

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Abstract

The optical properties and radiation hardness of large-size $PbWO_4$:Y crystal grown with modified Bridgman method were investigated through characterization on the optical transmittance, X-ray excited luminescence, light yield and irradiation damage of series $24 \times 24 \times 24\,mm^3$ block crystals cut from a long as-grown ingot. From the measuring results of the sequential small block samples it can be concluded that large-size modified Bridgman grown $PbWO_4$:Y crystals have excellent uniformity of optical properties and radiation hardness.

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Keywords: A1. Doping; A2. Bridgman technique; A2. Single crystal growth; B1. Lead tungstate; B2. Uniformity

1. Introduction

Lead tungstate $PbWO_4$ (PWO) crystals have drawn wide attention as a promising scintillator for radiation detection in high energy physics and nuclear physics due to its high density, short radiation length, small Moliere radius, fast decay time, non-hygroscopicity and low production cost [1–3]. To meet with the requirement of severe application environment with unprecedented levels of radiation in the experiments for examples, CMS experiment in CERN [4], various ways to improve the uniformity of optical properties and radiation hardness of the large-size PWO crystals have been employed such as raw material purifying, heterovalent ions doping and high temperature annealing. Doping with trivalent and pentavalent ions, Y^{3+}, Gd^{3+}, La^{3+}, Lu^{3+} and Nb^{5+}, could significantly improve the optical properties and radiation hardness of PWO [5–8]. From the viewpoint of uniform dopant concentration along the crystal length, Y^{3+} may be more appropriate than other dopants for its effective segregation coefficient in PWO is about 0.91 [9]. In fact, the PWO crystals used in electromagnetic calorimeter of CMS experiment are mainly doped with Y^{3+}. To know more about the uniformity of optical properties and radiation hardness of large-size PWO:Y crystals, we characterized a series block crystals cut from a large-size PWO:Y ingot grown with modified Bridgman method. Based on the measurement of their transmission spectra, X-ray excited luminescence spectra, light yield (LY) and irradiation damage (radiation-induced absorption coefficient, μ_{ir} and LY loss), the results show that large-size PWO:Y crystals grown with modified Bridgman method have excellent uniformity of optical properties and radiation hardness along the whole length.

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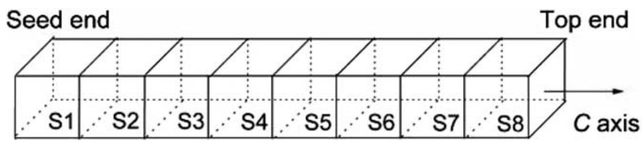

Fig. 1. Position of samples in crystal ingot. All samples are of $24 \times 24 \times 24$ mm^3.

2. Experimental

A PWO:Y ingot in the size of approximately $25 \times 25 \times 250$ mm^3 was grown with 5 N PbO and WO_3 powders by modified Bridgman method. The crystallization orientation was *c*-axis direction, and the concentration of Y^{3+} ion was about 150 at. ppm in the melt. The detailed growth technology can be seen in Ref. [10]. Fig. 1 shows the sequence of the block crystal samples cut from a large-size PWO:Y ingot. All these samples from S1 to S8 are of $24 \times 24 \times 24$ mm^3, with six faces optically polished.

The optical transmittance was recorded along the *c*-axis direction of each block sample by a SHIMAZU-2501PC spectrophotometer with an accuracy of ±0.002 absorbency. The irradiation experiment was carried out with ^{60}Co source at high irradiation dose (500 rad/h, 6×10^4 rad). The X-ray excited luminescence spectra were measured on a XEL spectrometer, FluorMain, where a F-30 movable clinical X-ray tube (W anticathode target) was used as the X-ray source, and operated at 80 kV, 4 mA. The scintillation spectra of the samples were obtained by 44 W plate grating monochromator and Hamamatsu R928-28 PMT with the data acquired by computer. Light yield at 20 °C was measured by using a ^{137}Cs γ-ray source and Philip XP2262B PMT on QVT Multichannel Analyzer test bench with a gate width of 100 ns.

3. Experimental results and discussion

The uniformity of optical properties and radiation hardness of large-size PWO:Y crystals can be evaluated by comparing the optical transmittance, X-ray excited luminescence spectra, LY, μ_{ir} and LY loss of the above sequential block samples.

3.1. Transmission

The transmission spectra of samples S1–S8 along the *c*-axis direction are shown in Fig. 2(a) and (b). From Fig. 2 it can be seen that the transmission spectra of all samples almost superposed into one curve, except sample S8 for which the transmittance at the 350–450 nm wavelength range was smaller than the other samples. The transmittance of sample S8 was 2% smaller than the sample S1 at the peak emission wavelength 420 nm of PWO.

3.2. X-ray excited luminescence spectra

Fig. 3 shows the X-ray excited luminescence spectra of all the samples with approximately same luminescence at peak wavelength near 425 nm, no significant differences were seen in the luminescence spectra. The blue luminescence of PWO at about 425 nm is attributed to self-trapped excited blue luminescence [11].

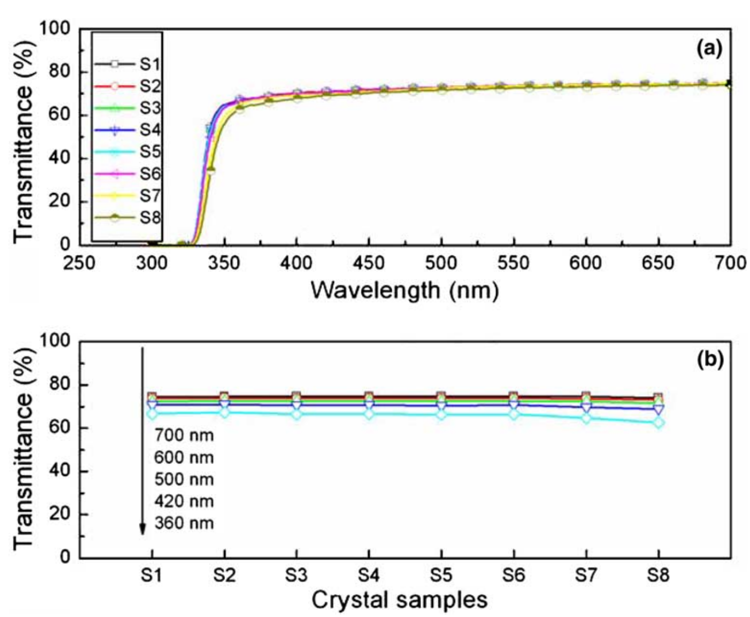

Fig. 2. The optical transmission spectra of samples S1–S8 before irradiation: (a) Wavelength: 300–700 nm, (b) at 360, 420, 500, 600, 700 nm respectively.

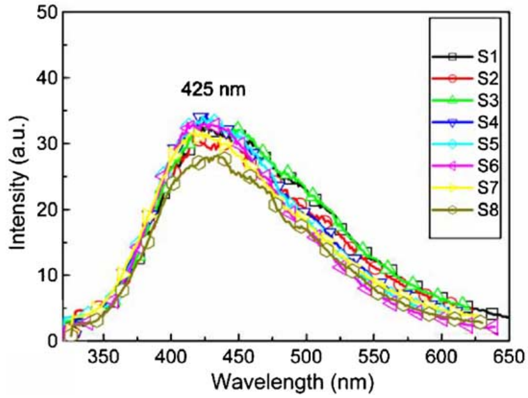

Fig. 3. X-ray excited luminescence spectra of samples S1–S8.

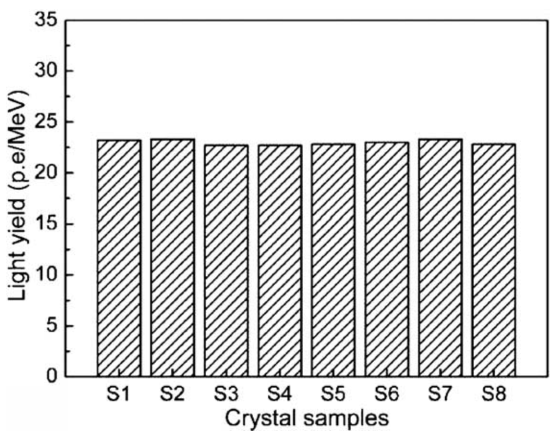

Fig. 4. The light yield of samples S1–S8 before irradiation.

3.3. Light yield

Fig. 4 displays that the light yields of all samples before irradiation at 20 °C with a gate width 100 ns are about 23 p.e./MeV on the average. The non-uniformity of light yield (dLY) is defined as [12]:

$$\mathrm{dLY} = 100\% \times (\mathrm{LY}_{\max} - \mathrm{LY}_{\min})/\mathrm{LY}_{\mathrm{mean}}, \quad (1)$$

where $\mathrm{LY}_{\max}$, $\mathrm{LY}_{\min}$ and $\mathrm{LY}_{\mathrm{mean}}$ are the maximum, minimum and mean values of LY measured along the *c*-axis direction. The light yield of small samples is less divergent from each other. With the LY value of small samples, the calculated dLY value of long PWO crystal grown with modified Bridgman method was about 2.6% that is better than dLY value 3.8% for large size PWO:Y crystals grown with Czochralski method [13].

3.4. Radiation damage

From the transmittance of sample before and after irradiation we calculated the radiation-induced absorption coefficient (μ_{ir}) spectra of these samples. The μ_{ir} is defined as [8]:

$$\mu_{\mathrm{ir}} = (1/d) \times \ln(T_{\mathrm{initial}}/T_{\mathrm{ir}}), \quad (2)$$

where d stands for the thickness of the sample, T_{initial} and T_{ir} are the transmittance before and after irradiation respectively. Fig. 5(a) shows the spectra of μ_{ir} versus wavelength of samples S1 and S8. The largest μ_{ir} of the spectra in UV/Visible range is below 1.5 m^{-1} at the accumulated dose of 6×10^4 rad. To see the induced absorption development at 420 nm, the emission peak

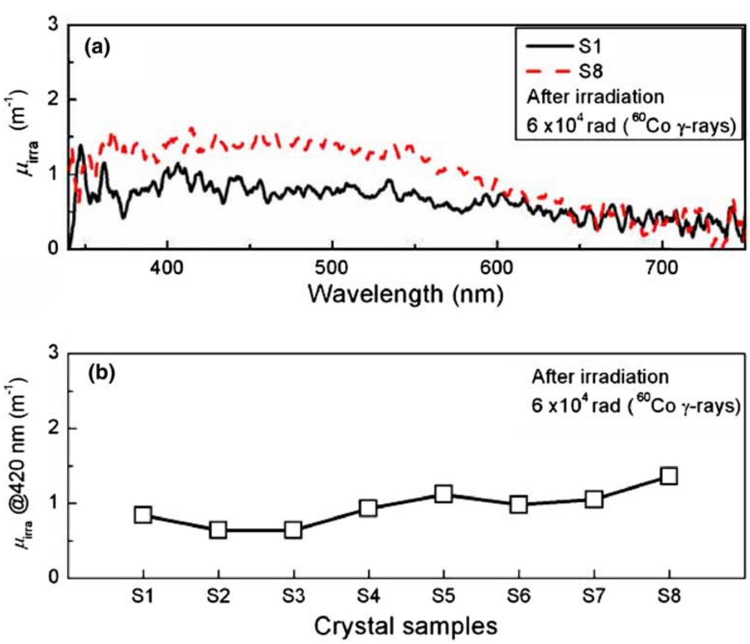

Fig. 5. Radiation-induced absorption coefficient: (a) Radiation-induced absorption coefficient μ_{ir} versus wavelength in samples S1 and S8. (b) Radiation-induced absorption coefficient at 420 nm in samples S1–S8 (500 rad/h, 6×10^4 rad, ^{60}Co γ-ray).

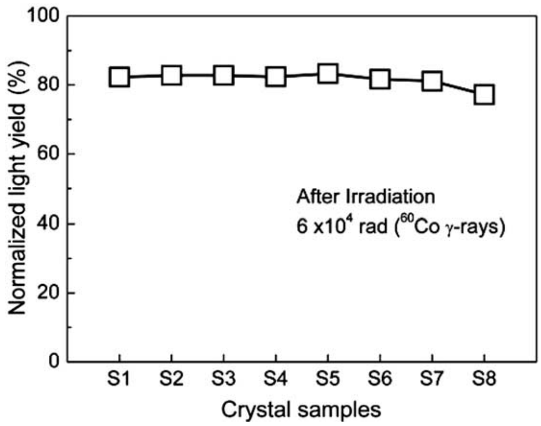

Fig. 6. Normalized light yield of samples S1–S8 after irradiation (500 rad/h, 6×10^4 rad, ^{60}Co γ-ray).

wavelength of PWO quantitatively, the μ_{ir} value for each sample was plotted in Fig. 5(b). From this figure, it can be seen that they are within the range of 0.6–1.3 m^{-1} at the accumulated dose of 6×10^4 rad. Compared to the induced absorption coefficient at 420 nm of large size Czochralski grown PWO:(Y, Nb) crystals irradiated at 5×10^4 rad dose [14], all the samples in this paper also show an induced absorption coefficient at 420 nm below 1.5 m^{-1} even at 20% larger irradiation dose.

The corresponding result of the measurement of LY loss after irradiation at 6×10^4 rad accumulated dose is given in Fig. 6. It shows that after irradiation the LY decreased by about 18% on the average in all samples, except sample S8 for which the LY decreased by about 23%.

From the results of above experiments of measuring optical transmittance, X-ray excited luminescence, LY, μ_{ir} and LY loss for all samples, it can be concluded that large-size PWO:Y crystals grown with Bridgman method have excellent uniformity of optical properties and radiation hardness except top end part. Compared with other samples, the sample S8 has relatively worse optical properties and radiation hardness, it may be related to the crystal growth method. First of all, the crystal growth process was not carried out in a completely sealed environment, especially at the top part of ingot. Therefore the excessive evaporation of PbO seems inevitable in such an oxygen-rich open environment, leading to the occurrence of large amount of lead vacancies (V_{Pb}) in top part of as-grown PWO ingot. Secondly, during the crystal growth, some impurities in material with effective segregation coefficient smaller than 1 such as K^+, Na^+, Mg^{2+}, Si^{4+} etc. may concentrate at the top end of PWO:Y ingot. The relatively excessive V_{Pb} and enrichment of impurities would increase the density of the permanent defect centers (Pb^{3+}, O^-, etc.) which jeopardize the optical properties and radiation resistance of the crystal. In order to obtain large-size PWO:Y crystals with excellent uniformity of optical properties and radiation hardness for CMS experiment, it may be indispensable and effective by purifying raw material, pre-crystallization, adding the dopant Y_2O_3 and growing long enough ingot to cut off the imperfect top part.

4. Conclusion

According to the characterization of optical properties, LY and radiation hardness for a series small block samples cut from a large-size PWO:Y ingot ($25 \times 25 \times 250$ mm^3) grown Bridgman method, we summarized the obtained results as follows:

(1) The optical transmission spectra of all the small block samples almost superposed into one curve except sample S8 for which the transmittance was 2% smaller than the sample S1 at the peak emission wavelength 420 nm of PWO.
(2) The LY_{mean} is about 23 p.e./MeV for all the small block samples. The non-uniformity of light yield in all samples was about 2.6%. Compared to the 3.8% of dLY in the large size Czochralski grown PWO:Y crystals [13], the Bridgman method grown PWO:Y crystals show an better uniformity of light yield.
(3) The radiation damage after ^{60}Co γ-rays irradiation were measured. The radiation-induced absorption coefficient at the emission peak wavelength 420 nm was as small as 0.6–1.3 m^{-1} after 6×10^4 rad dose irradiation. The LY loss after the same high dose irradiation was about 18% on the average. Compared to large size Czochralski grown PWO:(Y, Nb), PWO:La crystals [8,14], the measured series small block samples cut from a large-size Bridgman grown PWO:Y ingot show an induced absorption coefficient at 420 nm below 1.5 m^{-1} even at the 20% larger irradiation dose than that used in Ref. [14].

The obtained results from all the small block samples may indicate that large-size PWO:Y crystals grown with modified Bridgman method have excellent uniformity of optical properties and radiation hardness.

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