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LIGHT OUTPUT AND RADIATION DAMAGE IN A LEAD TUNGSTATE CRYSTAL

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ABSTRACT

The optical absorption, light output and effects of radiation damage have been studied in a $2.2 \times 2.2 \times 12.8 \text{ cm}^3$ crystal of lead tungstate. The crystal exhibits a longitudinal attenuation length which varies from roughly 13 cm to 1.5 m over the region where the scintillation emission occurs (400-800 nm). The light yield produces approximately 10 photoelectrons per MeV on a photomultiplier tube with a bialkali photocathode. The scintillation and light transmission properties of the crystal were characterized before and after exposure to 1 Mrad of ^{60}Co gamma rays. Significant radiation induced optical absorption was observed in this crystal, along with a considerable amount of phosphorescence after irradiation. Partial spontaneous recovery from the damage occurred at room temperature, and it was possible to induce additional recovery by optical bleaching.

INTRODUCTION

Lead tungstate (PbWO_4) is a relatively new crystal scintillator [1-4] which has a number of properties that make it an attractive candidate for use in electromagnetic calorimetry for high energy and nuclear physics. It has a high density ($\rho=8.3 \text{ gm/cm}^3$), short radiation length ($X_0 = 0.9 \text{ cm}$), small Moliere radius ($R_M = 2.2 \text{ cm}$), and weak scintillation emission in the visible wavelength range. However, for many high energy physics applications, such as at the CERN LHC, calorimeters are required to operate in a high radiation environment, and therefore the radiation damage properties of the calorimeter materials are important. This paper reports on the results of an investigation of the optical absorption, light output and radiation damage in a PbWO_4 crystal of the type which could be used in a high energy physics calorimeter.

The lead tungstate sample used in this study was produced at the Institute of Nuclear Problems in Minsk, Belarus. The sample is rectangular shaped, with dimensions $2.2 \times 2.2 \times 12.8 \text{ cm}^3$. To the eye, the sample is transparent with a yellowish tint. All of the surfaces are well polished. Two principle characteristics of the crystal were measured: the optical absorbance and the scintillation light output. The crystal was exposed to a high intensity ^{60}Co radiation source in order to study the effects of radiation damage. The absorbance and scintillation light output were measured initially, and then after incremental radiation doses, up to a cumulative total dose of 1 Mrad. Following the last irradiation, the natural recovery of the crystal was measured as a function of time at room temperature after irradiation, and optical bleaching was used to try to induce additional recovery from damage.

Figure 1. a) Longitudinal absorption coefficient (L) and transverse absorption coefficients measured at 2.0 cm (T2), 6.0 cm (T6), and 11.0 cm (T11) along the length of the crystal. b) Attenuation lengths computed using the longitudinal and three transverse scans.

ABSORBANCE AND LIGHT OUTPUT MEASUREMENTS

The absorbance was measured as a function of wavelength using an Hitachi model 3210 double-beam spectrophotometer. Since crystal growth often results in varying levels of impurities along the length of a crystal, absorbance scans were taken through the 12.8 cm longitudinal axis of the sample, and through the 2.2 cm dimension at several points along the length. Figure 1a shows a longitudinal scan and several transverse scans, plotted as absorption coefficient (in units of cm⁻¹) uncorrected for surface reflections, as a function of wavelength.

The attenuation length was calculated by comparing the longitudinal absorbance to the transverse absorbance and assuming the difference is due to internal absorption. This method assumes that the contribution to the measured absorbance from the surface reflections is the same for both the transverse and longitudinal scans, and neglects multiple reflections from each surface. Curves for the attenuation length (in meters) as a function of wavelength are plotted in Figure 1b. The difference between the attenuation length curves is due to the slight differences between the transverse absorbance scans at the different points along the crystal.

The light output of the crystal was measured in terms of the number of photoelectrons produced on the bialkali photocathode of a 2" diameter quartz window photomultiplier tube (Hamamatsu R2059) per MeV of energy deposited in the crystal. One of the 2.2 × 2.2 cm² surfaces of the crystal was coupled to the window of the phototube using Viscasil 600M optical couplant. The other surfaces of the crystal were wrapped in approximately

4 layers of .005" UV-reflecting teflon. A ^{60}Co source with 1.17 and 1.33 MeV gamma ray emissions was used to deposit energy in the crystal and the phototube signal was digitized using a Lecroy 2249W ADC with a 100 ns integration time. The peak of the pulse height distribution was determined from a gaussian fit and used along with the gain of the phototube (in ADC channels per photoelectron) to determine the number of photoelectrons per MeV produced by the crystal. Although the resolution was not sufficient to distinguish between the 1.17 and 1.33 MeV emissions, the average value obtained was approximately 10 photoelectrons per MeV.

RADIATION DAMAGE

The crystal was irradiated with ^{60}Co gamma rays at the Gamma Ray Irradiation Facility at Brookhaven National Lab. The sample was exposed to incremental doses totaling: 10^3 , 10^4 , 3×10^4 , 10^5 , 3×10^5 , and 10^6 rad, at a dose rate of 3.7×10^4 rad/hr. The sample was irradiated and stored in the dark, and handled only under dim red light. A longitudinal absorbance scan was taken 30 minutes after each incremental dose. Transverse absorbance scans were taken at points 2.0, 6.0, and 11.0 cm along the length, all within 60 minutes of each dose increment. Data from these measurements are shown in Figure 2. The transverse curves indicate that the damage was concentrated in one end (2.0 cm) of the crystal. No apparent saturation was reached in this end, even after the full dose of 1 Mrad, while the middle and other end did show signs of saturation after only 10^3 rad. This indicates a large variation in the concentration of impurities and/or defects along the length of the crystal. The total induced absorbance, measured down the longitudinal axis in the wavelength region where most of the scintillation emission occurs (400-500 nm), was ~ 0.07 . This corresponds to a transmission loss of $\sim 59\%$ along the 12.8 cm length of the crystal.

Following the last irradiation, periodic absorbance scans were taken to measure the recovery in transmission while the crystal was kept in the dark at room temperature. After 14 days of "natural" recovery, the crystal was optically bleached with UV and visible light from a medium pressure mercury light lamp. The sample was bleached with cumulative exposures of 1 minute, 10 minutes, and 1 hour. An absorbance scan was taken immediately after each incremental exposure. Figure 3 shows the natural and optically induced recovery for the longitudinal and transverse measurements of the crystal. Significant recovery was most obvious at the end that showed the more pronounced radiation damage. Optical bleaching accelerated the recovery, but did not remove all of the residual damage.

Phosphorescence from the crystal after irradiation was measured using the same phototube used for the light output measurements. The phosphorescence was measured after each incremental dose, and periodically after the last dose. The sample showed significant phosphorescence, apparently reaching a saturation level, even after the first dose. Following the fifth cumulative radiation dose (0.3 Mrad), the phosphorescence was measured at one minute intervals, starting at 20 minutes after irradiation. These results are shown in Figure 4. An exponential fit of the data to the form $y = a \cdot e^{-t/\tau}$ yields a value for the time constant of $\tau = 625$ sec.

Due to the induced transmission loss and the amount of phosphorescence, it was not possible to measure any scintillation light output from the crystal during the 14 days following the irradiation.

Figure 2. Change in absorption coefficient as a function of wavelength due to radiation damage for cumulative doses of: 1) 0 rad (initial), 2) 10³ rad, 3) 10⁴ rad, 4) 3 × 10⁴ rad, 5) 10⁵ rad, 6) 3 × 10⁵ rad, 7) 10⁶ rad. Figures represent scans taken: a) longitudinally, b) transversely at 2.0 cm, c) transversely at 6.0 cm, and d) transversely at 11.0 cm.

Figure 3. Recovery in absorption coefficient as a function of wavelength. 1) initial, 2) after 1 Mrad, 3) recovery after 4 hrs., 4) 22 hrs., 5) 5 days, 6) 14 days, 7) 1 minute of bleaching, 8) 10 minutes of bleaching, and 9) 1 hour of bleaching. Figures represent scans taken: a) longitudinally, b) transversely at 2.0 cm, c) transversely at 6.0 cm, and d) transversely at 11.0 cm.

Figure 4. Phosphorescence intensity as a function of time after 0.3 Mrad irradiation.

CONCLUSIONS

A study has been made of the optical absorption, light output and effects of radiation damage in a $2.2 \times 2.2 \times 12.8 \text{ cm}^3$ crystal of lead tungstate. The light yield gave approximately 10 photoelectrons per MeV, measured on a photomultiplier tube with a bi-alkali photocathode, and the attenuation length varied between 13 cm and 1.5 meters in the wavelength range from 400-800 nm. The effects of radiation damage were measured after exposure to 1 Mrad of ^{60}Co gamma rays which resulted in a significant amount of induced optical absorbance in the region of the scintillation emission, and a strong phosphorescence signal was observed. Partial natural recovery from the damage occurred with time at room temperature, and additional recovery was induced by optical bleaching. Although these studies represent only the measurements of one particular sample, the possible effects of radiation damage in this material has been demonstrated.

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