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A STUDY OF THE OPTICAL AND RADIATION DAMAGE PROPERTIES OF LEAD TUNGSTATE CRYSTALS

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ABSTRACT

A study has been made of the optical and radiation damage properties of undoped and niobium doped lead tungstate crystals. Data were obtained on the optical absorbance, the intensity and decay time of the scintillation light output, and the radioluminescence and photoluminescence emission spectra. Radiation damage was studied in several undoped and niobium doped samples using ^{60}Co gamma ray irradiation. The change in optical absorption and observed scintillation light output was measured as a function of dose up to total cumulative doses on the order of 800 krad. The radiation induced phosphorescence and thermoluminescence was also measured, as well as recovery from damage by optical bleaching and thermal annealing. An investigation was also made to determine trace element impurities in several samples.

INTRODUCTION

Lead tungstate ($PbWO_4$) is a new scintillating material which is of great interest for use in high energy electromagnetic calorimeters. It has a very high density (ρ = 8.3 g/cm^3), short radiation length (X_o = 0.9 cm) and small Moliere radius ($R_M \approx$ 2 cm), and has a scintillation light output which peaks between 450-550 nm with a decay time in the range from 5-15 ns. It is presently being considered for use in two large, high resolution electromagnetic calorimeters, one for the CMS experiment and the other for the ALICE experiment, at the Large Hadron Collider at CERN. In order to meet the stringent demands of these two experiments, the crystals are required to be of high purity, produced uniform light output, and, in the case of CMS, be resistant to radiation damage up to several megarads.

We have carried out a study of the optical and radiation damage properties of a number of undoped and niobium doped crystals obtained from sources in the former Soviet Union. A total of seven samples, measuring typically 2 × 2 × 21 cm^3, were measured in all. In this paper, we will mainly discuss the results for two particular samples (number 767 and 768), both of which are Nb doped, and were part of a group of crystals prepared for study by CMS. These two samples, which were grown under similar conditions, showed very different properties, particularly with respect to radiation damage. Other samples, both undoped and Nb doped, have shown similar behavior, some exhibiting good radiation hardness and others not. We will compare a number of differences observed in samples 767 and 768 in order to attempt to identify which of these properties could be relevant in determining the important optical and radiation damage properties of $PbWO_4$. Additional information on an undoped sample can be found in ref.[?].

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OPTICAL AND RADIATION DAMAGE PROPERTIES

The initial (pre-irradiation) transmission spectra for samples 767 and 768 are shown in Figure ??. Sample 767 showed a distinct absorption band in the region around 430 nm, while sample 768 exhibited only a shoulder. Sample 767 also contained a small ($\sim$ 2 cm long) region of inhomogeneity near the center of the crystal at one end which produced much higher absorbance near the center than near the edges.

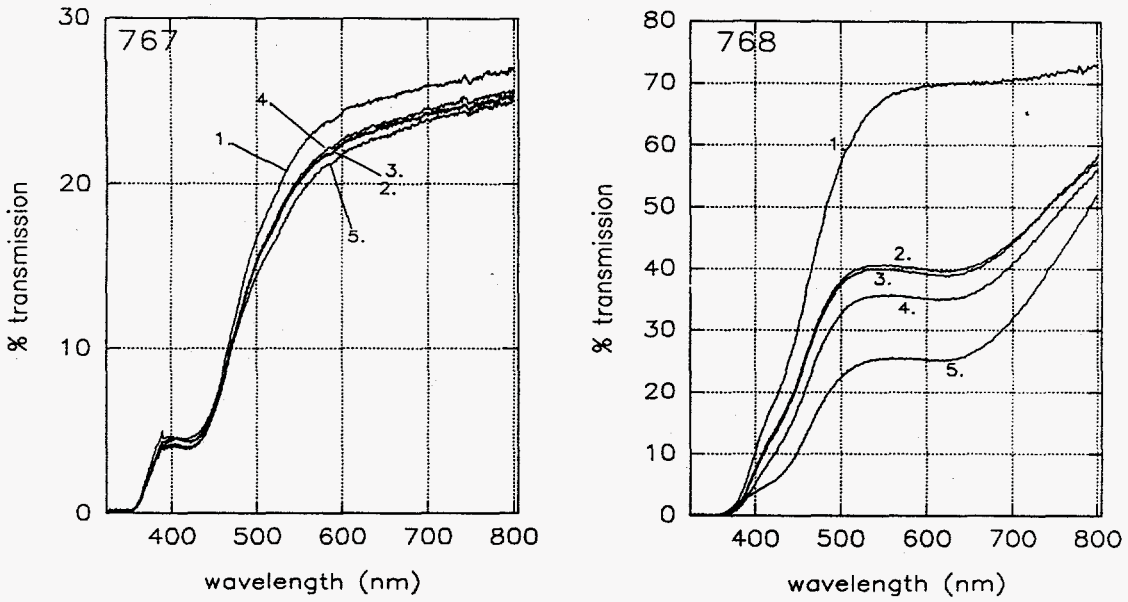

Figure 1: Transmission spectra as a function of radiation dose for samples 767 and 768. Cumulative doses for 767 are 1 - initial, 2 - 2.6 Krad, 3 - 7.6 Krad, 4 - 18.2 Krad, 5 - 769 Krad. Dose rate was 120 rad/hr for doses 2 through 4 and 3.5×10^4 rad/hr for dose 5. Cumulative doses for 768 are 1 - initial, 2 - 2.4 Krad, 3 - 11 Krad, 4 - 20 Krad, 5 - 834 Krad. Dose rate for dose 2 through 4 was 132 rad/hr and 3.6×10^4 for dose 5.

The spectral light output of both samples was compared by measuring the radioluminescence and photoluminescence emission spectra. The radioluminescence was measured using ^{60}Co gamma ray excitation, and the photoluminescence was measured using UV excitation at a wavelength of 315 nm. For sample 767, the photoluminescence was also measured for other excitation wavelengths from 250-350 nm, and no dependence of the emission spectrum on the excitation wavelength was found. Figure ?? shows the spectra measured for the two crystals after correcting for phototube quantum efficiency and monochrometer grating efficiency. The radioluminescence peaks at around 515 nm for sample 767, while sample 768 shows a lower peak at around 486 nm. Although the photoluminescence appears to peak at longer wavelengths (527 nm for 767 and 513 nm for 768), at least part of this shift can be attributed to self absorbance in the crystal. Decay times measured for the radioluminescence showed two major components in the range of 2-3 ns and 15-20 ns, along with a third slow component with a decay time of several hundred nanoseconds.

Both samples were irradiated using ^{60}Co gamma rays up to a maximum total dose of 769 krad for sample 767 and 834 krad for sample 768. In both cases, the initial irradiation was carried out at a low dose rate ($\sim$ 120-130 rad/hr), in order to study any rapid changes

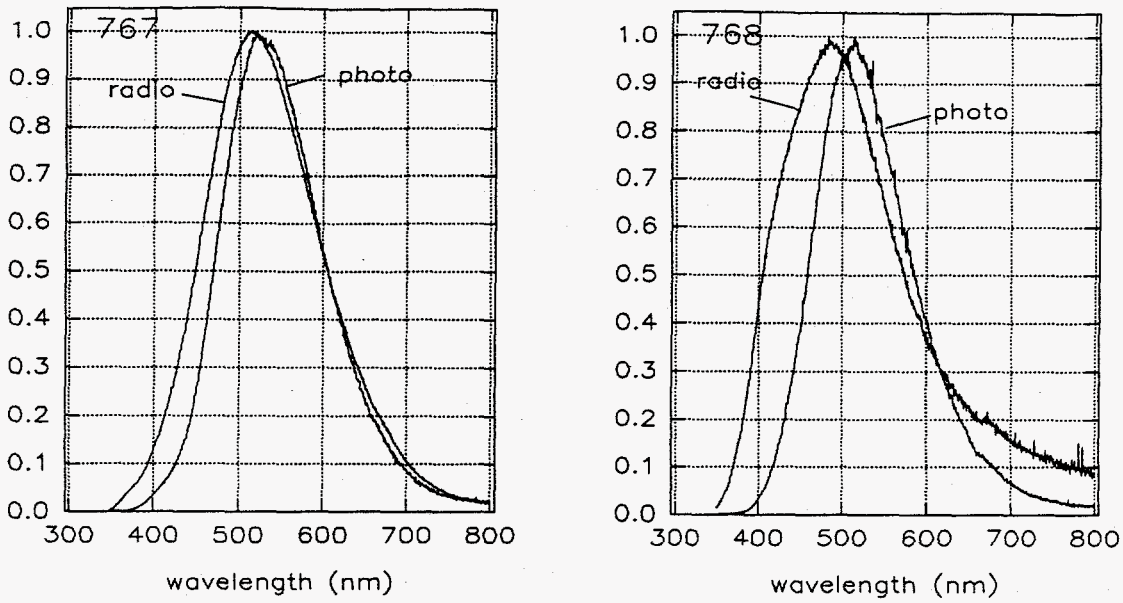

Figure 2: Radioluminescence and photoluminescence emission spectra for samples 767 and 768. All spectra have been corrected for phototube quantum efficiency and monochrometer efficiency and normalized to one at their peak.

which may occur in either sample. As shown in Fig. ??, sample 768 damaged quickly, exhibiting a series of strong, broad absorption bands in the region between 400 and 700 nm, and continued to damage with increasing dose. This damage was similar to that which was observed in undoped samples [?]. On the other hand, sample 767 showed very little damage, both at the initial low dose rate, and at the higher dose rate up to the maximum dose. The light output of both samples measured before irradiation on a 2" dia. phototube was $\sim$ 8 photoelectrons per MeV. After the initial dose, the light output from sample 768 was not measurable due to the increased self absorbance and interference from radiation induced phosphorescence. However, the light output from sample 767 was measurable even up to the maximum dose, as shown in Figure ??. The light output measured approximately 30 minutes after the final dose showed a slight decrease of about 15%, but recovered to its original value within 5.5 hours. Sample 768 showed a natural recovery in its transmission with a time constant of $\sim$ 160 hours. Optical bleaching using light in the 600-700 nm range produced significant recovery when applied immediately after irradiation, and thermal annealing for several hours at a temperature of 200 °C produced slightly more recovery.

Figure ?? also shows the radiation induced phosphorescence in samples 767 and 768 measured as a function of time shortly after irradiation. The apparent lower phosphorescence intensity in sample 768 was at least partly due to its higher induced absorption. The time constant for the phosphorescence decay was approximately 6-7 minutes for both samples. Both samples also exhibited a modest thermoluminescence signal which showed several peaks in the temperature range from 50-300 °C. However, in absolute terms, the thermoluminescence signal was several orders of magnitude less than for many other materials, such as BaF_2 or undoped CsI.

A trace element analysis was performed on both samples using Particle Induced X-ray Emission (PIXE) and Glow Discharge Mass Spectroscopy (GDMS) in order determine

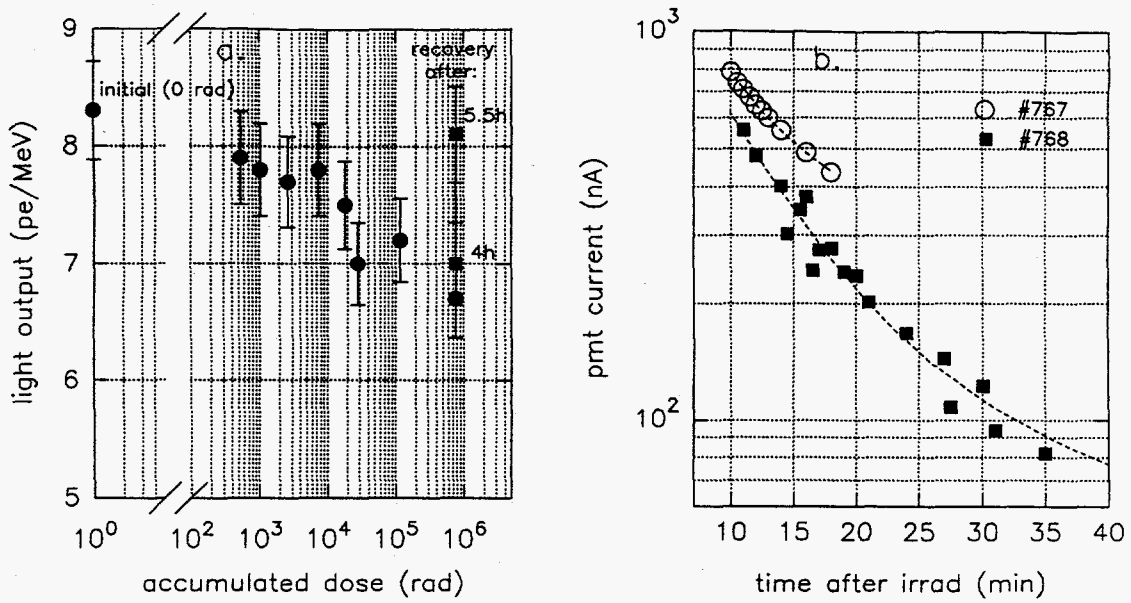

Figure 3: (a) Light output vs accumulated dose for sample 767. Except where indicated, all points were measured approximately 30 minutes after irradiation. (b) Phosphorescence intensity vs time for samples 787 and 768. A fit to spectra give decay times of 6.0 and 6.7 minutes for samples 767 and 768, resp.

the stoichiometry of the crystals and to search for trace element impurities. The results showed that both crystals were essentially stochoimetric $PbWO_4$, although there was an indication that the W/Pb ratio varied by $\sim 5\%$ from one end of the crystal to the other. A survey of 77 elements show similar levels of impurities in the few ppm to sub ppm level in both samples, although sample 767 showed a significantly higher level of sodium than 768 (9.6 ppm vs. 1.8 ppm).

CONCLUSIONS

A study of a number of lead tungstate crystals has revealed a large sample to sample variation in many of their properties. The optical transmission varies considerably, and some samples show sizable regions of visible inhomogeneities. The radioluminescence spectra also vary from crystal to crystal, with the peak occurring in the 480-520 nm range. The scintillation decay time shows two major components in the 2-3 ns and 15-20 ns range, along with a third slow component in the several hundred nanosecond range. Some samples, which do not necessarily show the best optical transmission before irradiation, exhibit particularly good radiation hardness. Initial trace element analyses carried out on samples having very different characteristics have shown no high level impurity content and similar levels of trace element impurities. Further work is under way to attempt to correlate these results with the optical and radiation damage properties of the crystals.

REFERENCES

[1] J.Kierstead et.al., Proceedings of the *Symposium on Scintillator and Phosphor Materials*, Materials Research Society, Vol. 348 (1994) 475.

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