



Evaluating displacement damage in cerium-doped yttrium borate using photoluminescence lifetime



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ABSTRACT

The photoluminescence lifetime and spectrum of $\text{YBO}_3:\text{Ce}^{3+}$ are observed before and after proton bombardment. The optical properties change under irradiation, and thus the material can record information about the radiation exposure. As the fluence of 1 MeV protons increases beyond 10^{14} proton/cm², the lifetime of $\text{YBO}_3:\text{Ce}^{3+}$ decreases nearly exponentially. Furthermore, by changing the concentration of cerium, the sensitivity of the material to the radiation induced defects is controllable such that lower-doped $\text{YBO}_3:\text{Ce}^{3+}$ is more sensitive to damage.

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1. Introduction

Rare-earth orthoborates are particularly suited for applications such as flat panel displays [1] and fluorescent lighting [2] because of their VUV transparency [3] and exceptionally high damage threshold [4]. Studies of these materials are often focused on fabrication because the light output is strongly dependent on the bulk configuration [5] (powder vs. crystal, for example). Other studies have focused on the conversion efficiency useful for silicon solar cells [6] or properties and structure suitable for optoelectronic devices [7,8]. These materials have also been used as scintillators for radiation detection [9,10] where the luminescence intensity is measured as a function of radiation dose. In the present work, we are interested in the radiation effects, which leverage all the optical properties that make these materials good for other applications. However, we are interested in how the photoluminescence changes with radiation-induced displacement damage, not scintillation.

Phosphor luminescence intensity is primarily governed by the arrangement of luminescent centers in the host lattice and the local crystal environment of each luminescent center. Systematic modification of the local crystal environment of the luminescent

center using particle radiation changes the optical characteristics of the material.

Previous experiments demonstrate that proton irradiation of $\text{YBO}_3:\text{Ce}^{3+}$ and $\text{YBO}_3:\text{Eu}^{3+}$ causes the intensity of the photoluminescence emission to degrade [11]. Gaubas et al. [12] report similar results using CdS. Both results show the emission intensity remains unchanged until a critical fluence. As fluence increases beyond the critical amount, the emission intensity degrades like Birks and Black [13].

In the present work, analysis focuses on the emission lifetime for each peak observed from the emission spectrum upon exciting the material with a pulsed UV laser. Usually a decrease in lifetime accompanies a decrease in emission efficiency for materials with high activator concentrations [14]. This can be understood in terms of concentration quenching. At low activator concentrations, the luminescent centers are too far apart to transfer energy between each other and thus the lifetime of an ensemble of emitters is that of the single luminescent center. If the concentration of activators is increased, the average distance between two activators and between an activator and quenching center decreases so that the probability of energy transfer, which is a faster process than emission, is enhanced. Consequently, excited activators are more likely to lose energy to quenching centers than to emission, and the emission efficiency decreases. Similarly, because the transfer is faster than emission, the lifetime of emission decreases. That is, the

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same mechanisms responsible for decrease in emission efficiency also decrease the lifetime.

Yet no studies have examined specifically how the emission intensity and lifetime change as a result of defect concentration. In large part, the lack of study is due to the difficulty of producing samples with a known and well-characterized defect profile. However, particle radiation allows experimentation that systematically increases the defect concentration in a relatively well-controlled manner so the effects of non-activator defects can be studied independently of the activator concentration. Study of the damage caused by radiation is important for a variety of reasons. (1) An understanding of the interaction dynamics between materials and particle radiation can be critical to the health of microelectronics and the design of microelectronics resistant to radiation [15], (2) the degradation of scintillation detectors is intimately related to the amount of exposure of the material to particle radiation [16], and (3) a detailed understanding of the accumulation of damage can lead to cumulative detectors, which is beyond the capability of scintillation detectors.

Radiation effects in phosphors have been studied previously. In fact, experiments using scintillation have been performed to determine radiation-resistance in phosphors [17,18], and many phosphors have been used as radiation detectors because they are good scintillators [19]. This investigation, however, describes the effects of the radiation-induced displacement damage instead of scintillation. While scintillation data can be collected during the irradiation, results of this work query of the material photoluminescence stimulated by a UV/VIS excitation after irradiation as a means to examine changes in the photoluminescence due to the radiation.

Spectral methods of defect exploration in ceramic materials provide several advantages. For example, (1) information can be acquired about the material-radiation interactions without physical contact to the object of interest, which might otherwise disturb the quantity being measured (defect concentration). (2) The displacement damage in many ceramic materials is not time sensitive (i.e., it does not anneal or return to its original structural state), so the material retains a record of the radiation exposure that can be gathered any time after the radiation event. (3) The material can also be used for measuring cumulative damage (up until complete quenching) if the radiation exposure occurs intermittently. Ultimately, phosphor photoluminescence and lifetime provide new opportunities for radiation detection that utilize remote measurement, create a permanent record of radiation exposure, and could detect (and conceivably quantify) multiple types and fluences of radiation.

2. Background

The $\text{YBO}_3:\text{Ce}^{3+}$ phosphor is exposed to step-stress proton irradiation to quantify radiation-induced changes of the lifetime. Proton irradiation is expected to implant defects in the material that affect the probability of emission, resonant transfer, and quenching mechanisms. Similar to thermographic applications, the type of phosphor host lattice and dopant may provide different information or sensitivity to radiation-induced defects. For example, a phosphor doped with boron may be more sensitive to neutrons than to protons due to its large neutron radiation cross-section. Depending on the stability of the crystal lattice, some materials may be more tolerant to radiation than others, such as fluorites vs. pyrochlores [17]. Therefore, several samples with varying doping concentrations were prepared and tested.

The Hollerman group investigated the effects of proton irradiation on the scintillation efficiency of luminescent phosphors. The materials they tested, (Gd_2O_3 doped with Eu, Pb, and Tb; Y_2O_3 doped with Tb and Eu; $\text{Y}_3\text{Al}_5\text{O}_{12}$ doped with Ce; and ZnS doped with

Ag) are found in plasma screens and other applications that require radiation hardness so that the continued scintillation does not degrade over time with continued exposure. Similar to our previous work [11], they measured the effects of long-term radiation damage using the degradation of the luminescence emission as a proxy for damage. They tested samples for the fluorescence efficiency as well as the dose necessary to reduce the luminescence to half of the original intensity [20,21,16]. They claim that the resulting degradation is from displacement damage, but the mechanisms were not explored in detail. Unlike the present work, the source of the excitation was the radiation itself (i.e. scintillation), not a subsequent UV/VIS excitation. In addition, the Hollerman group measured emission intensity, not photoluminescence lifetime, to indicate radiation exposure. Finally, although the Hollerman group investigated numerous oxides and various dopants, YBO_3 was not among the materials considered. Therefore, the present work is the first study using photoluminescence lifetimes of $\text{YBO}_3:\text{Ce}^{3+}$ for displacement damage detection. Similar to the Hollerman group, Gaubas et al. [12] examined the proton-induced luminescence and the microwave photoconductivity of a 20 μm layer of CdS during proton irradiation. The protons in their beam had an energy of 1.6 MeV, and their work examined radiative and non-radiative recombination to determine damage effects.

The approach used in this work is unique because (1) measurements do not examine the scintillation produced by irradiation; instead the analysis focuses on the photoluminescence after irradiation. This is important because phosphor properties may differ depending on how the material is queried. For example, photoluminescence spectra of triboluminescent materials and triboluminescence emission are not always the same [22], which suggests that the scintillation during irradiation and the emission when excited by a UV/VIS source after irradiation may not be the same. (2) The material composition ($\text{YBO}_3:\text{Ce}^{3+}$) is unique among those tested for radiation hardness. (3) The samples investigated are powders—the product of the synthesis method without the addition of binders [23]. The product material is derived from inexpensive reactants and accessible fabrication techniques. However, phosphor properties can be sensitive to the fabrication method and powders are particularly variable compared to highly controlled crystalline samples. Perhaps most importantly, (4) we analyze the lifetime to investigate the changes caused by irradiation, which proves to be a more robust measure of the radiation effects compared to spectral methods.

3. Methods

The concentration quenching curves for $\text{YBO}_3:\text{Ce}^{3+}$ for both emission intensity (Fig. 4) and lifetime (Fig. 2) were determined with samples doped with 1–10 mol% cerium. The radiation-induced damage was tested with twelve samples of $\text{YBO}_3:\text{Ce}^{3+}$ powder exposed to proton irradiation. Three samples were doped at 0.5 mol%, three at 4 mol%, three at 6 mol%, and three at 8 mol% cerium. Each sample was characterized with photoluminescence spectroscopy and lifetime before irradiation and after each irradiation step. $\text{YBO}_3:\text{Ce}^{3+}$ was synthesized using a gel-combustion synthesis. Reagents ($\text{NH}_4\text{B}_5\text{O}_8$ (Sigma Aldrich, $\geq 99.5\%$), $\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (Sigma Aldrich, 99.9%), glycine ($\text{C}_2\text{H}_5\text{O}_2\text{N}$, Sigma Aldrich Reagent Plus, $> 99\%$), NH_4NO_3 (Macron Chemicals, ACS Grade), $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (Sigma Aldrich, 99.5%), and $\text{Eu}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (Pfaltz and Bauer, Inc., 99.9%) were combined in an alumina crucible and diluted with deionized water. The contents were stirred on a hot plate at 80 °C until a homogeneous gel formed. The precursor was placed in a 600 °C muffle furnace until reaching the flame temperature at which point the gel combusted and formed a solid foam. The final product was crushed into a powder and calcined at 1100 °C for

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