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Luminescence study of some yttrium tantalate-based phosphors

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ABSTRACT

Yttrium tantalate (YTaO₄), yttrium niobium-tantalate (YTaNbO₄), yttrium niobate (YNbO₄), europium and terbium activated yttrium tantalate (YTaO₄:Eu, YTaO₄:Tb) and europium activated yttrium niobate (YNbO₄:Eu) phosphors were prepared by solid-state reaction, from a homogeneous mixture consisting of Y₂O₃, Eu₂O₃, Tb₄O₇, Ta₂O₅, Nb₂O₅ and Na₂SO₄ as flux. Photoluminescence (PL), thermally stimulated luminescence (TSL), X-ray diffraction (XRD) and scanning electron microscopy (SEM) of yttrium tantalate based phosphors are reported. In the present study TSL characteristics of these phosphors were first investigated and reported after exposure to beta, UV (311 nm) and X-ray radiations.

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1. Introduction

There are major interests from both scientific and application point of view for Eu and/or Tb activated yttrium tantalate, yttrium niobium-tantalate and yttrium niobate phosphors, Y(Ta, Nb)O₄:Eu/Tb [1–7]. Both host-lattice emission centers and rare-earth emission centers in these phosphors may give contribution to the overall luminescence. Rare-earth activated yttrium tantalate-type phosphors are promising materials for optoelectronics since they have variable luminescence chromaticity [5].

The growing interest in luminescence spectroscopy of rare-earth ions in ultraviolet (UV) and X-ray excitation is due to industrial demands for new applications. Yttrium tantalate (YTaO₄) and yttrium niobate (YNbO₄) phosphors are a perspective class of efficient materials that are generally used in X-ray intensifying screens. These phosphors display satisfying luminescence whenever excited by UV light, cathode radiation or X-rays. As it is well known, a material emitting light when excited by X-ray radiation is called an X-ray phosphor. Different energy sources such as electrons or UV light may stimulate the phosphors. The phosphors have a variety of application areas in films/screen cassettes, and also in electronic detector systems such as computed radiography, computed tomography and fluoroscopy [4,7].

Yttrium tantalate (YTaO₄) based phosphors seem to be promising media for preparation of up-conversion luminescence (UPL) materials since yttrium orthotantalate is physically and chemically stable, and it has favorable luminescence properties under UV or X-ray excitations [2,3].

Composition, crystalline structure, particle dimensions and luminescence properties of these powders could affect the performances of these phosphors. The different luminescence characterizations of Eu³⁺ or Tb³⁺ in tantalates and niobates have been previously reported in Refs. [8–11].

More recently, detailed studies of PL and TSL spectra of Tb doped YTaO₄ crystals showed phosphorescence with the peak wavelengths observed at 492, 543, 590 and 624 nm in PL emission spectra. For TSL measurements, the specimens were excited using He–Cd laser radiation, and then heated from room temperature up to 470 K. Glow peak of the material was observed at 330 K [1].

In Y(Ta,Nb)O₄ phosphors, the blue-light emission is associated with TaO₄ and NbO₄ groups from the host crystalline lattice. Luminescent emission could be shifted toward longer wavelengths when rare-earth ions such as Eu³⁺ or Tb³⁺ are used to partially replace the yttrium ion in the host crystalline lattice. In this case, Eu or Tb emission centers are created that give rise to specific red and green luminescence, respectively [7].

In this study, the thermally stimulated luminescence (TSL) properties of some yttrium tantalate/niobate phosphors, prepared without or with europium or terbium as doping ions were

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investigated under beta radiation, UV (311 nm) light and X-ray excitations. As far as we know there is no earlier work on thermally stimulated luminescence properties of these phosphors in literature. Moreover, the opportunity for using rare-earth doped yttrium tantalate-based systems was therefore an attractive system to explore the possibility of detecting novel features in a different crystal structure.

2. Materials and methods

2.1. Sample preparation

Yttrium tantalate, yttrium niobium-tantalate and yttrium niobate phosphors doped with europium and/or terbium, $Y(\text{Ta,Nb})\text{O}_4:\text{Eu,Tb}$, were prepared by solid-state reaction, from a homogeneous mixture consisting of Y_2O_3 (Fluka, 99.9%), Ta_2O_5 (Alfa Aesar, 99.85%) and Nb_2O_5 (Merck, Optipur) as the oxide precursors for the host lattice, Eu_2O_3 and/or Tb_4O_7 (both Sigma–Aldrich, 99.9%) as activator system and $\text{Li}_2\text{SO}_4 \cdot \text{H}_2\text{O}$ (Alfa Aesar, 99%) as flux. The stoichiometric amounts of Y_2O_3 , Ta_2O_5 and/or Nb_2O_5 , Eu_2O_3 and/or Tb_4O_7 together with 30 wt% alkaline sulphate were ball-milled with acetone and dried at 70 °C. The phosphor samples were baked at 1200 °C for 4 h and slowly cooled to room temperature. Finally, the samples were water washed, dried and then sieved.

Several yttrium tantalate-based phosphors, i.e. YTA samples were prepared as follows:

- YTA-1 \Leftrightarrow YTaO_4 phosphor;
- YTA-2 \Leftrightarrow $Y(\text{Ta,Nb})\text{O}_4$ phosphor, with formula $\text{YTa}_{0.60}\text{Nb}_{0.40}\text{O}_4$
- YTA-3 \Leftrightarrow YNbO_4 phosphor,
- YTA-4 \Leftrightarrow $\text{YTaO}_4:\text{Eu}$ phosphor, with formula $\text{Y}_{0.90}\text{Eu}_{0.10}\text{TaO}_4$
- YTA-5 \Leftrightarrow $\text{YNbO}_4:\text{Eu}$ phosphor, with formula $\text{Y}_{0.90}\text{Eu}_{0.10}\text{NbO}_4$
- YTA-6 \Leftrightarrow $\text{YTaO}_4:\text{Tb}$ phosphor, with formula $\text{Y}_{0.93}\text{Tb}_{0.07}\text{TaO}_4$

2.2. Sample characterization

Particle morphology and sizes were evaluated using scanning electron microscopy (SEM) images taken with Jeol-JSM 5510LV Microscope (Au-coated samples).

Crystalline structure was determined by X-ray diffraction (XRD). The characteristic XRD patterns were registered with Bruker D8 Advance X-Ray Diffractometer ($\text{CuK}\alpha$ radiation, Ni filter and alumina/corundum powder as standard for instrument broadening correction). XRD quantitative phase analysis was performed using the PowderCell program for Windows. The crystalline phases were retrieved after a peak search semi-automatic routine using the data base PDF-2 [12].

Photoluminescence emission (PL) spectra were registered in identical experimental conditions, using Jasco FP-6500 Spectrofluorimeter Wavel equipped with photomultiplier PMT R3377 (Farbglasfilter WG 320, phosphors $\text{Zn}_2\text{SiO}_4:\text{Mn/N40}$ and $\text{CaWO}_4/\text{N61}$ were used as control samples).

Thermally stimulated luminescence (TSL) glow curves were recorded using thermoluminescence dosimeter reader (Harshaw Model 3500) with a linear heating rate of 2 K s^{-1} . Irradiation is performed by a ^{90}Sr – ^{90}Y beta source (650 MBq) and supplied Machlett OEG-50A a X-ray tube adjusted to 30 kV and 15 mA; UV (311 nm) illumination was performed by a Philips TL-01 narrow band UV lamp, which has an emission centered at 311 nm. All samples were irradiated at room temperature using different radioactive sources for 5 min. The experiments were carried out immediately after irradiation in order to overcome any fading effect which influence the glow curves [13].

3. Results and discussions

The present study is focused on the thermally stimulated luminescence properties of some rare-earth doped or undoped yttrium tantalate/niobate phosphors with good photoluminescence and morphostructural characteristics, as reported in our previous papers [14–16].

3.1. Photoluminescence properties

Yttrium tantalate YTaO_4 , yttrium niobium-tantalate $Y(\text{Ta,Nb})\text{O}_4$ and yttrium niobate YNbO_4 samples, nominated YTA-1, YTA-2, and YTA-3 respectively, are undoped phosphors that, under different exciting radiations show UV-blue emission due to the charge transfer transition into TaO_4 and/or NbO_4 groups (recombination luminescence). The specific broad-band emission spectra obtained under 254 nm excitation are depicted in Fig. 1. The undoped YTaO_4 phosphor shows very weak luminescence in the UV-blue domain, in agreement with the literature data [17,18]. When tantalum atoms are partially or totally substituted by niobium ones, the sensitivity of the host lattice to UV excitation is increased and the luminescence becomes significant in the blue spectral region, at about 400–407 nm. TaO_4 and/or NbO_4 groups are responsible for the blue recombination luminescence, associated with some charge transfer transitions involving the tetrahedral MO_4 groups.

For $Y(\text{Ta,Nb})\text{O}_4$ mixed lattice, the emission peak is shifted toward longer wavelengths, the peak intensity is slightly smaller and the band is relatively broader in comparison with YNbO_4 sample, thus illustrating that energy transport through the lattice seems to be disturbed by the possible disordered matrix.

Europium doped yttrium tantalate $\text{YTaO}_4:\text{Eu}$, europium doped yttrium niobate $\text{YNbO}_4:\text{Eu}$ and terbium doped yttrium tantalate $\text{YTaO}_4:\text{Tb}$, noted YTA-4, YTA-5 and YTA-6, respectively, show the

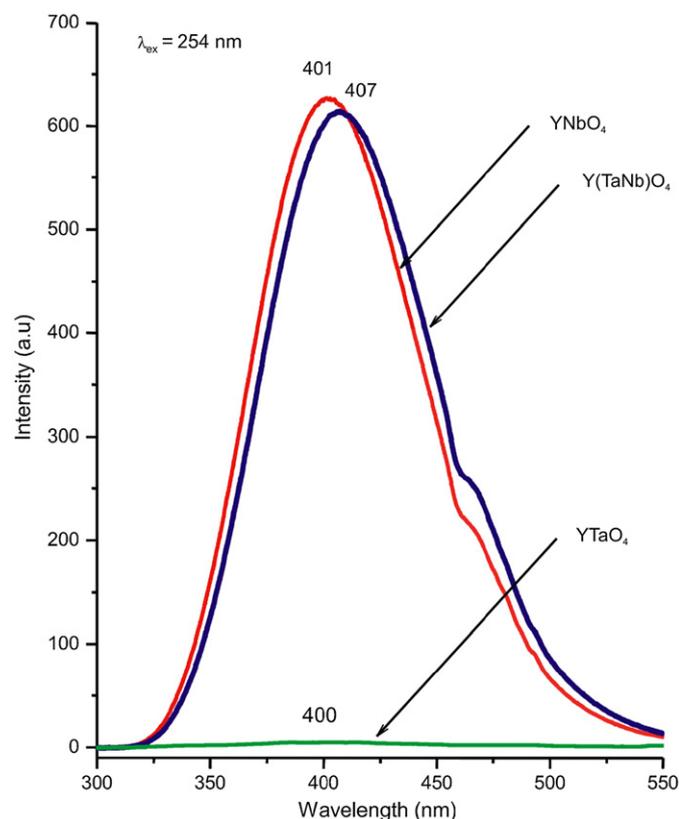


Fig. 1. PL spectra of YTaO_4 (YTA-1), $Y(\text{Ta,Nb})\text{O}_4$ (YTA-2) and YNbO_4 (YTA-3) samples.

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