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Reassessment of the potential applications of Eu^{3+}-doped Y_2O_3 photoluminescent material in ceramic powder form

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Abstract

The preparation of Y_2O_3 doped with different amounts of Eu^{3+} ions obtained via a precipitation method using oxalic acid as precursor is reported here. The precursors were calcined at different temperatures to obtain the polycrystalline ceramic materials in order to verify the influence of temperature on the photoluminescence emission properties of the materials. The efficient incorporation of Eu^{3+} ions in Y_2O_3 host matrix was verified by X-ray diffraction. Based on the diffractograms obtained, using Scherrer's equation the increase of the crystallite size as a function of annealing temperature was verified. All materials obtained showed intense photoluminescence emission in the red region (~611.5 nm), assigned to the f–f intraconfigurational transition of the Eu^{3+} ions. The CIE chromaticity diagrams were built based on the photoluminescence emission spectra with *X* and *Y* coordinates being 0.672 and 0.325, respectively, indicating the pure color emission of the material. The materials obtained are excellent absorbers in the UV–vis region (bellow 250, around ~394 and ~463 nm) with intense emission in the red region (~611.5 nm). Two lifetime values are observed indicating that the Eu^{3+} ions are located in at least two sites of symmetry, C_2 and S_6. The lifetime values obtained are on the order of ms, being relatively long and interesting for contribution to solar cell efficiency improvement. The fact that these materials absorb ultraviolet radiation very well, resulting in intense emission in the red region, make them excellent candidates to for applications in solar cells.

Keywords: Photoluminescence; Europium; Yttrium oxide; Ceramic; Solar cells

1. Introduction

There has been significant development of materials for technological applications such as photonics, electronics, displays, and optical amplification, lasers, and solar cells [1–5]. These materials must provide appropriate physical and chemical characteristics for efficient devices [6,7]. With the need for clean and renewable energy sources that can reduce dependence on fossil fuel, along with the growing energy demands of modern society, there has been a push to implement new technologies for generating energy. Because of this, the use of solar energy to produce electricity has emerged as a renewable, clean and plentiful energy source, capable of bringing electricity to millions of people in isolated areas. Beginning in the 1970s, the need for renewable energy sources, amid the oil crisis, stimulated the research and development of devices based on converting solar energy into electrical energy. Later, due concern for the environment, to prevent damage caused by intense exploitation and consumption of fossil fuels, efforts were increased to seek cheaper materials, lower production costs and increase efficiency of energy conversion of these solar cells.

Even with progress in manufacturing solar cells, traditional devices feature a characteristic loss of efficiency as they are unable to absorb photons from all regions of the solar spectrum. Low

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energy photons, corresponding to the infrared region, are not absorbed by the solar cell and high-energy photons in the ultraviolet region, transferred to the cells and are not efficiently converted to electrical current. Thus, in a silicon solar cell, for example, approximately only 45% of the photons emitted by solar radiation is absorbed and used to convert to electrical energy. When the sunlight hits a traditional single junction solar cell, only photons corresponding to the visible wavelengths are absorbed. Thus, these solar cells have intrinsic efficiency losses. Approximately 65% of the solar energy is lost. Photons with a higher energy, like ultraviolet radiation is absorbed by the solar cell, but undergo a process of thermalization and some of the energy is lost as heat in the material network. In this sense, materials which absorb energy in the ultraviolet region and convert it into emission in the visible region may be associated with these types of solar cells, contributing to increased energy conversion efficiency.

The dye sensitized solar cells (DSSC) have received much attention from researchers all over the world due to its readily available, low cost and significant efficiency in energy conversion [8]. On the DSSC, the addition of the dye contribute to the enhance of the efficiency, since these dyes absorb in the visible region occurring the photoexcitation. When visible solar radiation focuses on this type of solar cells, electrons located in the HOMO of the dye absorb photons and are excited to the LUMO. The electrons localized in the LUMO are injected into the conduction band of the semiconductor in question, such as TiO_2, resulting in a current flow. Thus, one of the ways to increase the efficiency of solar cells is to increase the injection of electrons in the LUMO of the semiconductor. For this reason, the use of materials that convert the ultraviolet energy into visible region, which in most cases are not absorbed by the solar cells, it is of great importance to contribute to the increased efficiency of these types of devices.

Among many published papers, those containing rare earth ions have important spectroscopic features [9,10]. Currently, a large number of studies reporting the use of rare earths to increase the efficiency of solar cells have been presented. To improve efficiency, a great combination of a host matrix, such as Y_2O_3, Gd_2O_3, ZnO, TiO_2, CeO_2, $Y_3Al_5O_{12}$, [11–13] among others, with an activator trivalent ion like as Eu^{3+}, Yb^{3+}, Er^{3+}, Ho^{3+}, Tm^{3+} ions, is required [14,15]. A good relationship between the matrix and the dopant ion involves the crystal structure, composition, morphology and optical properties as well as a synthetic route for obtaining these materials. In particular, one can cite yttrium oxide (Y_2O_3), which has some mechanical, and chemical [16] properties as well as low phonon energy, high melting point, low vapor pressure, and optical transparency to a broad range wavelength, which makes it an interesting host to trivalent ions for use in optical devices that emit light and [17–19]. The relative similarity of the radius and coordination number of rare earth (RE) ions with ions that compose the host matrix is very interesting for good relation between RE ions and the host matrix [20]. Eu^{3+}-doped Y_2O_3 is an example of a photoluminescent material, phosphor, comprising of a host lattice (Y_2O_3) and a luminescent center [21]. The electromagnetic radiation emitted by these photoluminescent materials usually occurs in the visible region, with the Y_2O_3:Eu^{3+} emission at 611 nm (red region) due to the electronic transition $^5D_0 \rightarrow {}^7F_2$ of the Eu^{3+} ion [22]. Phosphors exhibit important features, such as good luminous efficiency, long luminescence lifetimes, reactivity, stability and powder morphology, i.e., the more homogeneous the better the performance of phosphorus [23–26]. Materials that have a long lifetime of the excited state may become very interesting which can contribute to a longer time of absorption of visible radiation by the dye. Consequently, there is an increase in the time of transfer of the energy between HOMO and LUMO and longer time in the injection in the conduction band of the semiconductor.

Accordingly, the article presents some results related to the synthesis of Eu^{3+}-doped Y_2O_3 obtained at different annealing temperatures by a precipitation method using oxalic acid as a precursor. The structural and photoluminescent properties are present as a function of Eu^{3+} concentration in the Y_2O_3 host structure.

2. Experimental procedure

Initially 0.5 g of Y_2O_3 was dissolved in 20 mL of deionized H_2O and 3.0 mL of HCl, under stirring and heating at 60 °C. The Eu^{3+} ions were added as a dopant in the solution containing Y^{3+} ions in concentrations of 1, 3, 5, 7 and 10 mol% in relation to the amount of Y^{3+} ion. The solution of Eu^{3+} was previously standartized with EDTA 0.01 mol.L^{-1} at room temperature. A saturated oxalic acid solution was added in the solutions containing the metals and a white precipitate was collected. The precipitate was washed with water, centrifuged and kept in an oven at a temperature of approximately 100 °C for 3 h for drying. After, the precipitate was annealed at 900, 1000 and 1100 °C in an oven for 4 h to obtain final products. The precipitates were submitted to thermogravimetric analysis and differential thermal analysis (TGA/DTA) using a Shimadzu, model DTG-60H with a heating rate of 10 °C/min under air atmosphere from room temperature up to 1000 °C. The crystalline structure information was acquired by X-ray diffraction (XRD) using a Shimadzu diffractometer with $K\alpha$ radiation of Cu^0, λ=1.5418 Å, graphite monochromator, 15°–80°, 2θ. Based on the diffractograms obtained, the crystallite sizes were calculated using Scherrer's equation. The photoluminescence emission and excitation spectra were obtained at room temperature using a Fluorolog FL3-221-Horiba Jobin-Yvon with excitation fixed at 394 nm. The emission spectra between 575 and 725 nm, were obtained using the emission and excitation slit of 3 and 5 nm, respectively. The excitation spectra between 275 and 500 nm were obtained using the emission and excitation slit of 5 and 3 nm, respectively. Based on the emission spectra, the chromaticity diagram was obtained. Kinetic measurements were realized at room temperature, and were obtained operating the same spectrofluorometer equipped with xenon pulsed lamp and a phosphorimeter 1934 with emission and excitation slits fixed at 0.2 and 0.5 mm, respectively. The emission and excitation were fixed at 611.5 and 394 nm, respectively. In order to evaluate the morphology of the material obtained, the samples were characterized by