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Photosensitive defects in Gd_2O_3 – advanced material for solar energy conversion

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Abstract

The photo- and thermally- stimulated luminescence of Gd_2O_3 and $Gd_2O_3:Zn$ nanostructured powders has been investigated at 295 and 90 K. Optically active impurities and intrinsic defects in the host lattice have been detected. The kinetics parameters of the charge trapping centers (order of kinetics, activation energy, frequency factor) were determined. The nature of defect states and their role in the processes of the energy relaxation are discussed.

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1. Introduction

The phosphors based on lanthanides represent a perspective class of materials for photovoltaic devices. One of the main features of rare earth (RE) ions is a large number of discrete energy levels that form the $4f$ electronic shell [1]. The variety of optical transitions in a wide spectral range has the potential to be used for down- and up-conversion processes. These energy transfer mechanisms are the basis for the functioning of the so-called luminescent solar concentrators that are placed on the front or rear side of traditional Si solar cell as additional layers. Such structures allow one to use the additional fraction of solar spectrum in UV and NIR region, therefore, enhancing the conversion efficiency [2, 3].

Among the compounds containing the RE elements, the gadolinium oxide (Gd_2O_3) is a good candidate as host

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matrix of luminescence layers. In fact, it has remarkable optical and chemical properties, such as the transparency in the visible spectral region ($E_g = 5.6$ eV), low energy of phonons (≈ 600 cm⁻¹), high refractive index, good chemical durability and thermal stability [4, 5]. In addition, the similarity between the Gd³⁺ and RE³⁺ ionic radii and the electronic shell structures provide the possibility to create different pairs of rare earth ions (for example, Yb-Er, Yb-Ho, Yb-Tm) [6]. All these features make Gd₂O₃ a promising material for down- and up-conversion applications in solar energy.

The enhancement of the Si solar cells efficiency by means of the luminescence solar concentrators is directly determined by energy transfer mechanisms of the donor-acceptor couples in the optically active layers. Matrix imperfections are one of the main responsible for the energy migration processes. In general, one supposes that the structural defects in the host lattice lead to the decrease of the overall luminescence efficiency due to the creation of non-radiative relaxation channels [7]. However, there is information about improving the luminescence efficiency of RE-doped phosphors containing the structural distortions [8]. Thus, the investigation of the effect induced on the spectral-optical properties of RE-doped materials by intrinsic host lattice defects could be of great interest for improving the functional characteristics.

2. Samples and methods

The microstructured nominally pure Gd₂O₃ powder was prepared at Ural Federal University. Gadolinium nitrate (Gd(NO₃)₃) with a purity of at least 99.99 % was used as precursor and was dissolved in a mixture of water and isopropyl alcohol at concentration of 50 g/l. The suspension was firstly kept under stirring for 30 minutes and then was filtrated, dried and calcinated at 1000° C for 2 hours. The Gd₂O₃:Zn compacted powder was synthesized by sol-gel method at the Institute of Physics Polish Academy of Sciences. The gadolinium (Gd₂O₃) and zinc (ZnO) oxides as starting materials were kept in 1M HCl solution for 24 h, the molar concentration being 80 and 20 % respectively. The solution was firstly centrifuged for 15 min at 22° C and the deposited material, separated by supernatant, was repeatedly washed with a mixture of water and ethanol by centrifuging at the same conditions. The obtained gel powder was dried in air and annealed at 1000° C for 2 hours.

The XRD profile of Gd₂O₃ sample showed single cubic phase (Ia-3 space group) with lattice constant $a = 10.81$ Å. The Gd₂O₃:Zn sample has a monoclinic crystal phase (space group C2/m) with lattice parameters $a = 14.09$ Å, $b = 3.57$ Å and $c = 8.76$ Å. The average particle sizes of both powders were estimated to be 10-40 nm by using the Debye-Scherrer's method [9].

The photoluminescence (PL) and PL excitation studies were performed using a 400 W deuterium lamp, a R6358-10 Hamamatsu photomultiplier and two prismatic DMR-4 monochromators with the spectral resolution of 1 nm in the visible spectral region. Low-temperature measurements were carried out in a cryostat at $P = 10^{-4}$ Pa. The temperature of the sample was controlled by a platinum resistance thermometer HRTS-5760-B-T-1-12. The thermally stimulated luminescence (TL) spectra were recorded in the spectrally integrated mode in the region 300-600 nm at a constant heating rate of 0.3 K/s. The samples were previously subjected to X-ray irradiation with exposure dose of 50 kR at room temperature.

3. Results

In Fig. 1(a) the PL spectrum emitted by Gd₂O₃ sample is reported. It is acquired at ambient temperature under interband excitation at 5.4 eV. It evidences strong and sharp lines at 2.4, 2.6 and 2.8 eV and a weaker band at 4.1 eV. The observed emission in the visible region is characteristic of 4*f*-4*f* intraconfiguration optical transitions in europium (Eu³⁺), terbium (Tb³⁺) and praseodymium (Pr³⁺) ions. One can assume that these ions are in the Gd₂O₃ matrix as micro impurities. The band observed at 4.1 eV can be attributed to the ⁶P_J → ⁸S_{7/2} optical transition in Gd³⁺ ion. Generally, the luminescence of host ions is not typical and can indicate the structure distortions in lattice. In order to obtain detailed information about the emission centers and luminescence mechanisms we performed low-temperature measurements.

As can be seen from Fig. 1(b) broad non-elementary band is observed under the excitation of 5.4 eV at 90 K. As shown by the black dotted lines it can be decomposed into two Gaussian components with maximums at 2.7 eV (FWHM ≈ 0.8 eV) and 3.3 eV (FWHM ≈ 0.9 eV).

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