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Original research article

Green photoluminescence, structural and optical properties of Nd-TiO_2 thin films

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ABSTRACT

Thin films of pure and neodymium doped titanium oxide with various concentrations were deposited on crystalline silicon and onto glass substrates by spin coating technique. The influences of the concentration of neodymium on structural, morphological and optical properties were studied by x-ray diffraction (XRD), scanning electron microscopy (SEM), AFM microscopy, UV-Visible spectroscopy and photoluminescence spectroscopy. It was found that the obtained films are well crystallized with high density; moreover, all films exhibit high transmittance in the visible and near infra-red regions. The optical band gap was found in the range of 3.37 - 3.65 eV. The surfaces of the dense films are homogeneous with the presence of small grains at the surface. Better and strong green photoluminescence was observed for the 3% Nd doped TiO_2 thin film around 520 nm and which shows a relationship with oxygen vacancies.

1. Introduction

Nano-structured semiconductors materials have attracted a lot of attention due to their unusual optical and electrical properties [1,2]. Semiconductor research is a very important field in the ongoing research activity across the world, and the interest on optical properties of thin films is increasing this last years for the development of photonic and electronic devices [3,4]. Recently, intensive efforts have been made to develop a semiconductor metal oxide having typical optical properties, which can lead to new optoelectronic integrated devices with high performances. In addition, these materials offer the possibility of integrating their magnetic and electronic properties in spintronic devices [5,6]. For the moment, the titanium oxide known also as titania (TiO_2) thin films are the most used in industry because of their remarkable coating properties such as high transparency in the visible and near infrared, good stability against chemical and mechanical attacks, high dielectric constant and refractive index, their reasonable cost, non-toxicity, good electrical and thermal properties, simple technology of their fabrication, high photo-conversion efficiency and photo-stability [7–9]. As a result, they have a wide range of applications such as solar cells, photocatalysis, optical coatings, gas sensors, planar wave guides, biomedical devices, capacitors, high-brightness transmitters, memoristors … etc. [10–13]. Photoluminescence is generally controlled by the doping with the rare earth elements. Lanthanides are usually famous for their unique and luminescence properties when incorporated in semiconductors. The motivation is that it can absorb and emit a wide range of wavelengths from near infra-red (NIR) through the visible to UV. Among the various rare earth elements, neodymium has attracted a great interest in

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recent years for the manufacture and the development of laser diodes and photonic integrated devices due to its sharp and intense luminescence in the near infra-red [4,14,15] and among the methods of depositing thin layers, the sol-gel method is widely used because of several advantages; low temperature development, high homogeneity and good control of the chemical composition, besides the possibility to produce thin layers on large surfaces as well as its low cost [16–18]. The objective of the present work is to study the effect of the incorporation of neodymium on the structural, morphological, optical and photoluminescence of thin films of titanium dioxide TiO_2.

2. Experimental procedure

Thin layers of pure TiO_2 and that doped with neodymium were prepared from the precursor of titanium isopropoxide (Ti (OCH $(CH_3)_2)_4$) 99.99% produced by Aldrich, which was added to isopropanol (CH3CHOHCH3) to be diluted under magnetic stirring later, pure methanol (CH_3OH) wa added to make the solution less viscous and nitric acid to stabilize the final solution. The doping is made by the precursor nitrate hexa-hydrate neodymium (Nd $(NO_3)_3{\cdot}6H_2O$) which is added directly with the methanol. The atomic ratio of dopant in the solution varied from 1 to 7%. Thin films of Nd-TiO_2 are deposited on glass and silicon substrates by spin coating technique using a rotating speed of 3000 rot/min for 15 s. After the deposition of each layer, and to improve the crystal quality, these films are annealed at 525 °C for 90 min. Annealing of TiO_2 films causes crystals rearrangement leading to stable TiO_2 phase of anatase. Various analytical techniques were used to characterize thin films; the X-ray diffraction was made by an MPD diffractometer X'pert Pro with (θ–2θ) configuration and copper anticathode, the surface of the films were observed using a Philips XL brand SEM-FEG scanning electron microscope. The optical transmittance measurements were made using a spectrophotometer Perkin Elmer Lambda 950-Precisely, and the photoluminescence spectra using a spectrophotometer Perkin Elmer B50. The samples were tested at 77 K under an excitation of 266 nm and 420 nm at room temperature, using a xenon lamp.

3. Results and discussion

3.1. X ray diffraction

Fig. 1 shows the X-ray diffraction spectra of the pure and doped samples with 1%, 3%, 5% and 7% Neodymium. XRD spectra show good crystallization of the samples and the presence of the anatase phase (peak attributed to the (101) plane) for both pure and doped samples until 3%. For the sample doped with 5% Nd, the coexistence of both anatase (101) and rutile (110) is noted. When increasing doping to 7%, the diffraction peak becomes broader and less intense; indicating that the high doping creates defects in the crystalline structure and deforms the crystal structure.

The average crystallite size was calculated from the Scherrer formula [19]:

$$D = \frac{0.94\lambda}{\beta\cos\theta} \tag{1}$$

Where: D is the crystallite size, λ is the wavelength of the X-ray radiation, β is the width at half-height and θ is the diffraction Bragg angle.

Fig. 2 shows the average crystallites sizes of different samples. For pure TiO2 layer, the average crystallite size is about 23 nm.

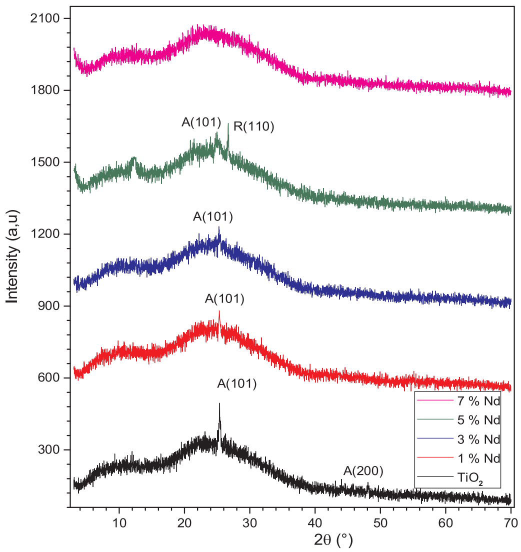

Fig. 1. X-ray diffraction spectra of pure and Nd doped TiO_2 thin films.