Materials Science in Semiconductor Processing 26 (2014) 395–403

Contents lists available at ScienceDirect

Materials Science in Semiconductor Processing

journal homepage: www.elsevier.com/locate/mssp

Effect of annealing atmosphere on the electrical properties of nickel oxide/zinc oxide p–n junction grown by sol–gel technique

M. Jlassi [a,*], I. Sta [a], M. Hajji [a,b], B. Ben Haoua [c], H. Ezzaouia [a]

[a] Laboratoire de Photovoltaïque, Centre de Recherche et des Technologies de l'Energie, Technopole de Borj-Cédria, BP 95, 2050 Hammam-Lif, Tunisia
[b] Ecole Nationale d'Electronique et des Communications de Sfax, Technopole de Sfax, BP, 1163, CP 3021, Tunisia
[c] Laboratoire VTRS, faculté des Sciences et Technologie, Université d'El oued BP 789, 39000 El oued, Algeria

ARTICLE INFO

Available online 29 May 2014

Keywords:
ZnO
NiO
Sol–gel
Spin-coating
p–n Junction
Annealing atmosphere

ABSTRACT

Zinc oxide (ZnO) and nickel oxide (NiO) thin films were prepared on glass substrates by a sol–gel method. Spin coating was used to fabricate a p-NiO/n-ZnO junction. The influence of the post annealing atmosphere (air or nitrogen) on the microstructure and surface morphology of NiO and ZnO thin films and the p-NiO/n-ZnO junction are examined. The structural properties are characterized by X-ray diffraction (XRD) and the surface morphology of the thin films and the p–n junction are investigated by atomic force microscopy (AFM). Optical properties are investigated by UV–visible spectroscopy and the electrical properties, such as *I–V* photocurrent, are characterized by a voltage source meter instrument. XRD patterns show that the films are polycrystalline with preferred orientation in the (002) direction for the ZnO films and the (200) direction for the NiO films. The AFM results indicate that the morphology of the ZnO and NiO films and the p-NiO/n-ZnO junction are mainly influenced by the annealing atmosphere. All films have a high optical transmittance of about 80% in the visible region and a sharp absorption edge. The optical band gaps of the two materials change with the annealing atmosphere (air or nitrogen). The p-NiO/n-ZnO heterojunction device has an average transmittance of over 80% in the visible region, which lies between the transmittance of the ZnO and NiO films separately. The ideality factor, barrier height, and series resistance of the heterojunction treated in different annealing atmospheres are determined by using conventional forward bias *I–V* characteristics and also Norde's and Cheung's methods.

1. Introduction

The requirements of the field of ultraviolet (UV) optoelectronics, such as UV detection and UV emission [1,2], have initiated an increased interest in transparent conducting oxides (TCOs), which are used for a wide range of applications [3–5]. The use of TCOs in the fabrication of optoelectronic devices has not been successful due to the lack of TCOs exhibiting p-type conductivity. The low carrier mobility and density associated with the narrow valence bands of TCOs make it even more difficult to obtain a good p-type conduction required for device applications. Most transparent oxide p–n junctions and light emitting diodes were fabricated using zinc oxide (ZnO) and $SrCu_2O_2$ [6]. ZnO is a promising semiconductor material for advanced optoelectronic applications because of its wide direct band gap (3.4 eV) and large exciton binding energy (60 meV) [7,8], therefore it has recently become one of

* Corresponding author.
E-mail address: mohamedjlassilpv@yahoo.fr (M. Jlassi).

http://dx.doi.org/10.1016/j.mssp.2014.05.008
1369-8001/

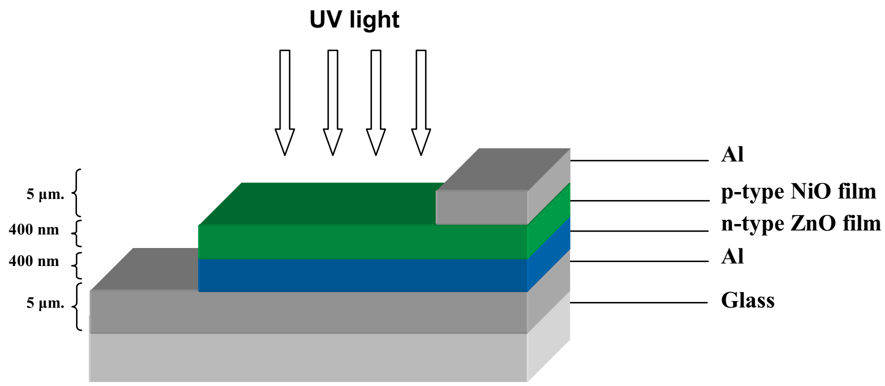

Fig. 1. Device structure of the NiO/ZnO heterojunction fabricated on an aluminum covered glass substrate.

the most common candidates for highly transparent optoelectronic devices, in the blue or ultraviolet range. Although the p–n junction is widely adopted for use in UV photodetectors [9], the growth of p-type ZnO films is still less studied. Over the last few years, many compact UV detectors have been prepared from p–n heterojunctions formed between n-type ZnO and other materials, such as silicon [10], NiO [11–13], and ZnMgO [14]. NiO is a wide band gap semiconductor of 3.6–4.0 eV at room temperature [15]. NiO is easily fabricated as a p-type semiconductor and is widely used for applications, such as transparent conductive films [16] and electrochromic devices [17,18].

In this paper, we report the effects of different annealing atmospheres on the electrical characteristics of p-NiO/n-ZnO heterojunctions fabricated on glass substrates covered by thin films of aluminum. The heterojunctions were synthesized by the sol–gel method and subsequently annealed in two different atmospheres including air or nitrogen. The effect of annealing atmosphere on the device performance was characterized along with the structural, optical, and electrical properties. The structural properties of the p-NiO/n-ZnO heterojunction are characterized by X-ray diffraction (XRD) and atomic force microscopy (AFM); the optical properties are characterized by UV–visible spectroscopy, whereas the electrical properties of the p-NiO/n-ZnO heterojunction diode were characterized in detail by *I–V* characteristics depending on ambient temperature.

2. Experimental procedure

The p-NiO/n-ZnO heterojunctions were fabricated on patterned aluminum electrodes using the sol–gel spin coating method. The active area of the devices was 1 mm × 1 mm. The ZnO and NiO chemical solutions were prepared separately. To obtain the sol, the precursor zinc acetate dehydrate [Zn $(CH_3COO)_2 \cdot 2H_2O$] (98%, Sigma Aldrich) and nickel acetate tetra hydrate ($C_4H_6NiO_4 \cdot 4H_2O$) (98%, Sigma Aldrich) were separately dissolved in 2-methoxyethanol ($C_3H_8O_2$) (99.8%, Sigma Aldrich). After 30 min of stirring at room temperature, the hot plate temperature was ramped up to 60 °C and the diethanolamine (MEA) (99%, Merck) was added drop-wise as a stabilizer under constant magnetic stirring. The molar ratio of (MEA/Zn) was maintained at 1:1. The concentration of ZnO and NiO sol was fixed at 0.5 M. The solutions were stirred for 2 h at 60 °C until transparent and homogenous sols were obtained. The as-prepared solution was aged for 24 h. The ZnO and NiO films used in this work were fabricated by a sol–gel method using a spin coating technique onto glass substrates (microscope slides) covered by a thin evaporated aluminum layer which served as a metallic contact. All ZnO and NiO gel films were coated onto the substrates at a speed of 3000 rpm for 35 s. The as-coated films were pre-heated at 300 °C for 10 min. An oxide layer thickness of 1 µm was obtained after repeating the coating procedure four times in the same conditions. The films were subsequently annealed for one hour at 600 °C under two different atmospheres including air and nitrogen separately. In the same way, a p–n junction having the structure of Al/n-ZnO/p-NiO/Al, as it is shown in Fig. 1, was fabricated on a glass substrate.

The crystallographic structure was studied by X-ray diffraction using a Bruker D 8 advance (both Bragg–Brentano) X-ray diffractometer with Cu K_α radiation ($\lambda_{CuK\alpha}$ = 1.5405 Å) for 2θ values in the range of 20–90°. An atomic force microscope (Nanoscope III) was used in tapping configuration (Veeco AFM head RTESP silicon pur) to scan an area of 1 µm × 1 µm. The AFM results were used to estimate the surface roughness and the grain size in the films. A standard software was used to calculate RMS roughness and the grain size. The grain size was defined as the diameter of the grain in the middle of the height. The optical characterization was carried out by a UV–vis–NIR spectrophotometer (Lambda 950), equipped with an integrating sphere, in the wavelength range from 300 to 800 nm. The *I–V* photocurrent measurements of the p–n junction on the glass substrate were performed with a Keithley Model 2400 low voltage source meter instrument. All measurements were carried out at room temperature.

3. Results and discussion

3.1. Microstructural properties

The structural properties of ZnO and NiO produced by the spin coating method and treated under air or nitrogen were investigated by XRD. To perform structural identification and study the changes in the crystallinity of the samples, we regrouped all the results for ZnO and NiO in the same figure for those treated under air atmosphere (Fig. 2a) and under nitrogen atmosphere (Fig. 2b). As it is shown in Fig. 2(a), the ZnO sample is a polycrystalline consisting of ZnO hexagonal phase and presents a strong