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Original research article

XRD, SEM, XPS studies of Sb doped ZnO films and electrical properties of its based Schottky diodes

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ABSTRACT

This work presents both the morphological and structural characterizations of ZnO depending on the Sb doping and the electrical characterization of ZnO based Schottky diodes grown on ITO substrates by sol gel dip coating method. In Schottky diode fabrication, undoped and Sb doped ZnO were used, which exhibit n-type and p-type behavior, respectively. For undoped and Sb doped ZnO, Pt and Al were used as a metal contact. For the surface morphology, scanning electron microscopy (SEM) has been carried out and the surface properties that play an important role on the Schottky diode performance were characterized by SEM. X-ray diffraction (XRD) measurements revealed that crystal quality got worse and crystallite size decreased with Sb incorporation. The presence of Sb in the ZnO was confirmed by X-ray photoelectron spectroscopy (XPS). Undoped and Sb doped ZnO based Schottky diodes were fabricated and their electrical properties were carried out in dark. The diode parameters such as ideality factor (n), barrier height (φ_B) and series resistance (R_s) were systematically analyzed by using thermionic emission theory and Cheung's method.

1. Introduction

Zinc oxide (ZnO) films have been studied with many researches due to intrinsic properties such as wide band gap (3.2 eV), large exciton binding energy (60 meV), low production cost and bio-safety. This material have attracted attention in scientific and technological communities because of their application in several devices as field effect transistors [1–3], heterojunction and Schottky diodes [4–6], sensors [7,8] and solar cells [9,10]. Among these devices, metal-semiconductor Schottky barrier diodes based on ZnO films are basic building blocks of electronic device.

The ZnO-based Schottky diode was investigated in 2002 [11] in first time. The authors reported that silver Schottky contacts were fabricated on n-ZnO epilayers, which were grown on R-plane sapphire substrates by metalorganic chemical-vapor deposition. The flat band barrier height was determined to be 0.89 and 0.92 eV by current-voltage and capacitance-voltage measurements, respectively. The ideality factor was found to be 1.33. There are a number of works with metal/ZnO Schottky diodes in available literature. Most of these diodes were used n-type ZnO. However, the p-type doping of ZnO is still a challenge, which becomes the main hindrance in the development of ZnO-based optoelectronic devices [12].

The doped of ZnO with group I (Li, Na, K) or group V (N, As, Sb) elements will change the conductivity of type. Theoretically, group-I elements as p-type dopants in ZnO are far better than group-V elements. Bagheri et al. [13] deposited Li doped ZnO by sol gel. Authors reported that the conductivity of the ZnO film doped with optimized Li ratio (15 at%) changed from

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n-type to p-type. Chelouche et al. [14] prepared Na doped ZnO thin films onto quartz substrate by sol gel deposition. They reported that p-type conductivity was observed only on the 3 at % Na-doped sample and with an increasing Na concentration it was observed a decrease in the average grain size and surface roughness. Tay et al. [15] deposited K defect in ZnO films grown using the aqueous solution route, which explains the switching between p- and n-type conductivities under different doping or thermal annealing conditions. They emphasized that the success of p-type doping depends on the aqueous growth conditions and the concentration ratio of K^{+} /Zn^{2+} and solution pH are important parameters in this success.

Sb doped ZnO films have been deposited by metal-organic chemical vapour deposition (MOCVD) [8], spray pyrolysis [9,10], rf sputtering [11], sol gel spin coating [12,13]. Among many techniques for preparing Sb doped ZnO films, sol gel dip coating has many advantages such as simple, inexpensive, and having large area applications.

To the best of our knowledge, there is only one study on the fabrication and characterization of Sb doped ZnO based Schottky diode in the available literature. In this work, Mandalapu et al. [16,17] investigated the electrical properties of Sb doped ZnO films grown on n-Si by molecular beam epitaxy using Al/Ti metal. In their investigations they used the current-voltage (*I-V)* and capacitance-voltage (*C-V)* characterizations. Used Al/Ti contacts exhibited Schottky diode behavior with a barrier height of 0.8 eV estimated from the *C-V* measurements. It was also reported that the ideality factor or precise barrier height was not determined because of not being known the actual area of device.

Although many researchers have studied the preparation and characterization of Sb doped ZnO, as mentioned above there is only one report about their Schottky diodes applications. Of course, a reason of this can be the challenge of obtaining successfully p-type ZnO and its fabrication as an Schottky diode. In the work of Mandalapu et al. [17], both Schottky and ohmic contacts were taken from the top and as a result, the detailed investigations of diode parameters were not made due to the unknown of actual area of the device. But, in our work, ITO substrate was used and there was no such problem. Therefore, the importance of this study is to contribute to the literature in this subject, where there is little work and to consist of detailed and systematic characterizations presenting the effect of Sb incorporation. In this paper, it has been investigated how Sb incorporation influences the structural, morphological and electrical properties of the ZnO films which are grown on ITO by a sol gel dip coating technique. And also, it has been carried out the electrical characterization of *Pt/n-ZnO/ITO, Al/p-ZnO:0.2%Sb/ITO and Al/p-ZnO:0.4%Sb/ITO* Schottky diodes by using measurements.

2. Experimental

The undoped and Sb doped ZnO films were deposited by sol gel dip coating method onto ITO substrates. Zinc acetate dihydrate [$Zn(CH_3COO)_2{\cdot}2H_2O$] (ZnAc) and antimony chloride [$SbCl_3$] (SbCl) were used as a starting material and dopant source. 2-Methoxyethanol [$C_3H_8O_2$] and ethanolamine [C_2H_7NO] (MEA) as a solvent and stabilizer, respectively. The molar ratio of MEA to ZnAc was maintained at 1:1 and the concentration of ZnAc and SbCl was 0.35 M. These solutions were mixed together in different volume proportions 0.2 and 0.4%. In order to yield a clear, homogeneous and transparent solution, it was stirred for 2 hours at 60 °C using a magnetic stirrer. After stirring process, the solution was infiltrated using filter paper. The ITO substrates were cleaned in methanol, acetone using an ultrasonic cleaner. The cleaned ITO substrate was placed on the sample holder and was dipped at withdrawn speed of 8 mm/min. The dip coated films were preheated at 300 °C for 10 min in a furnace. This coating/drying procedure was repeated ten times, before the films were annealed at 600 °C in air for 60 min. Then, Schottky contacts were made onto surface of the film, patterned with a shadow mask by circular dots of 0.5 mm in diameter. Evaporation and sputtering methods were used to be formed aluminum and platinum contacts on the surface, respectively. The fabricated *Pt/n-ZnO/ITO, Al/p-ZnO:0.2%Sb/ITO and Al/p-ZnO:0.4%Sb/ITO* Schottky diodes are named SD0, SD2 and SD4 respectively. The schematic structure of these Schottky diodes is illustrated in Fig. 1.

X-ray diffraction patterns (XRD) were obtained with a BRUKER D2 Phaser Series X-ray automatic diffractometer using the $Cu_{K\alpha}$ radiations (λ = 1.54059 Å) in the 2θ range between 30° and 60° at room temperature with a scanning step size of 0.02°. The diffractometer reflection was taken at room temperature. Surface morphology was studied using ZEISS Ultraplus model field emission scanning electron microscopy (SEM). VAKSIS PVD-HANDY/MT/101T thermal evaporation and VAKSIS Midas PVD-MT/1M3T, Turkey sputter systems were used for the evaporation of Al and sputtering of Pt contacts, respectively. The

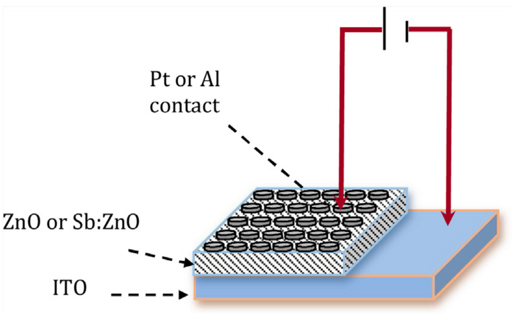

Fig. 1. The schematic diagram of the fabricated Schottky diodes.