Simulation of the Optical Properties of $Zn_{1-x}Co_xO$ Thin Films Grown Onto Glass Substrate by Ultrasonic Spray Pyrolysis

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Abstract: ***$Zn_{1-x}Co_xO$ thin films (x=0-22 at.%) were prepared using the ultrasonic pyrolysis sputtering technique. The films have a hexagonal wurtzite structure, confirmed by X-ray difraction, with an estimated average crystallite size in the range of 18-30 nm. Concerning the optical properties, by applying the Levenberg-Marquardt least squares method, the experimental transmittance data were perfectly fitted with the transmittance data calculated via a combination of the Wemple-DiDomenico model, the absorption coefficient of an electronic transition and the Tauc-Urbach model. Decreases in bandgap energy due to the presence of high concentrations of localized states in thin films were calculated from the Smakula formula***

Keywords: Thin films, X-ray diffraction, Optical properties

1. Introduction

Transparent Conductive Oxides (TCO) are remarkable materials in many fields. The existence of their dual properties, electrical conductivity and transparency in the visible range, makes them ideal candidates for applications in photovoltaics and optoelectronics. Zinc oxide ZnO is a semiconductor material belonging to this family of OCTs, it presents interesting electronic, electrical and optical properties for optoelectronic applications, especially in the photovoltaic field. The band gap is of direct nature, the value of its width varies from 3.3 eV to 3.4 eV and an excitonic binding energy of 60 meV [1]. The doping with cobalt is of great interest in many fields of application. Their particular properties make them suitable for use as optical components, integrated optical amplifiers,...

These films have been used in several electronic and optoelectronic fields such as: conductive gas sensors [2], light emitting diodes [3], photocatalytic reactors [4], optical windows in solar cells [5]. ZnO films can be made by several techniques such as: sputtering [6], chemical vapor deposition [7], sol gel and spray pyrolysis [8]. The aim of this work is to study the influence of Co-doping on the structural and optical properties of ZnO thi films.

2. Experimental Part

2.1 Film preparation

In this study, the ultrasonic pyrolysis spray technique was used to synthesize ZnO, $Zn_{1-x}Co_xO$ films. This technique is a relatively simple alternative that uses traditional and inexpensive means [9]. Its implementation is locally feasible. It also has the advantage of elaborating thin layers on large surfaces such as those of solar cells or flat screens [9,10]. From the advantages mentioned above, we have selected this elaboration process and we have opted for its use in our work. In fact 1.2g of zinc acetate [$C_4H_6O_4Zn.2H_2O$], We have as source material that we dissolved in50 ml of deionized water; 20 ml of CH3OH; 30 ml of C2H5OH. In addition to the undoped ZnO thin film, we prepared several series of layers, one of which is doped with Cobalt, 1-22% (Co, at. %) cobalt nitrate hexahydrate [Co $(NO_3)_2$-$6H_2O$] was used as the source of Co. To fix the pH value at about 4.8, a small amount of acetic acid was added, so as to prevent hydroxide evolution. The thin films were deposited on glass substrates (30 × 10 × 1.2 mm^3) at the temperature of 450 °C, weighting 30 min.

2.2 Characterization techniques

In order to study these parameters, different characterizations were performed. To do so, we characterized our films by X-ray diffraction in order to deduce the evolution of their microstructure by using a high resolution diffractometer (Rigaku Ultima IV powder) equipped with Cu-Kα radiation (λ=1.5418 Å). The optical properties were studied using UV-Visible transmission, with a Perkin Elmer UV-Vis-NIR spectrophotometer (Lambda 19) in the range 190-1800 nm.

3. Results and discussion

3.1 Structure analysis

The X-ray diffraction spectra show the presence of the wertzite phase of the hexagonal structure of ZnO according with (JCPDS card no. #00–036–1451) [10]. Deposits obtained with Co concentrations show a strong preferential orientation along the (002) axis in the set of films, without any other impurity phase at least within the detection limits of X-ray diffraction. This result clearly indicates that Co^{2+} can be incorporated into the ZnO lattice developed by Ultrasonic Spray method without any phase segregation taking place in these films. We also observe an increase in peak intensity as a function of doping rate. The results of the phases examined qualitatively and quantitatively by the Rietveld method are shown in Table 1.The value of the lattice parameter "a" rises with increasing Co content, while