THE INFLUENCE OF INDIUM DOPING ON STRUCTURAL, OPTICAL AND ELECTRICAL PROPERTIES OF SnO₂:IN THIN FILMS DEPOSITED BY SPRAY TECHNIQUE

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ABSTRACT

In this present work, undoped and Indium doped tin dioxide were deposited on glass substrate by Ultrasonic spray method. We investigated the effect of deposition conditions to obtain In doped SnO2 thin films with various concentration (1 to 8 wt.%). XRD analysis confirmed that SnO_2 thin films crystallize in the tetragonal structure of SnO2. The grain size average decreases with In content increase. We found that the maximum films transmittance varies from 65-93% in the visible range of the spectrum. The films optical gap varies between 3.48 and 3.80 eV. However, we have noticed that the sheet resistance increases up to $880*10^3$ (Ω /sqr) with increasing the in doping concentration. Owing to their high optical gap and high sheet resistance; the prepared films can be employed in optoelectronic devices.

KEYWORDS: Tin dioxide; thin films; semiconductor doping; ultrasonic spray method.

1 INTRODUCTION

Based on latest researches, transparent conductive oxides (TCO) films of semiconductor materials have used in various applications in science and technology. However, tin dioxide is most studied semiconductor compounds, it is has attracted much attention due to their transparency and good conductivity [1-3]. Based on these characterics, SnO₂ can be used in various potential applications such as in UV photodetectors, transparent diodes, UVtransparent transistors, solar cells, sensors, memory devices n-type oxide semiconductors, touch screens, displays and defrosting windows [1-8]. SnO₂ has been intensively studied as a promising material for cell solar due to its good optical transparency, high electrical conductivity and high stability, with a wide band gap of about 3.6 eV [2,9]. Moreover, the SnO₂ semiconductor films were prepared by various techniques such as magnetron sputtering technique [10], pulsed laser deposition (PLD) [11], chemical vapour deposition [12], sol-gel process [13], electrochemical deposition [14] and spray pyrolysis [7,15].

In this present work, indium doped tin dioxide was deposited on glass substrate at 450 $^{\circ}$ by Ultrasonic spray method. We studied effect of deposition condition to obtain SnO_2 thin films of indium concentration (1 to 8 wt.%) on structural, optical and electrical properties.

2 EXPERIMENTAL DETAILS

The deposition of In: SnO_2 thin films require a starting solution containing tin and indium, for that we used as precursors tin chloride dihydrated ($SnCl_2$, H_2O) and indium chloride ($InCl_3$). SnO_2 :In solution were prepared by dissolving 0.1 M ($SnCl_2$, H_2O) in the solvent containing equal volumes absolute methanol solution (99.995%) purity. The Indium doping was used ratio of In/Sn = 0.01, 0.02, 0.04 and 0.08.

The crystal phase of the thin films were characterized by X-ray diffraction (XRD, Bruker D8 Advanced X-ray diffractometer) with Cu K α radiation (λ = 1.541 Å), the analysis the samples were scanned from 20° to 80°. The optical transmission spectra of the films were measured in the range of 300–1000 nm using a double-beam Lambda 35UV/ visible spectrophotometer and the electrical resistivity was determined using the four point method.

3 RESULTS AND DISCUSSIONS

The crystalline quality of In doped SnO_2 thin films was carried out by analyzing the X-ray diffraction is show in Figure 1. Overall, several growth directions are observed: (110), (101), (200), (211), (310), (301). We notice that the deposited films have a polycrystalline structure, with the preferred orientation (200) to substrate. The film doped with 1% In have a high crystallinity. The high increases in In doping allowed us the emergence of a new preferred

orientation is (301).

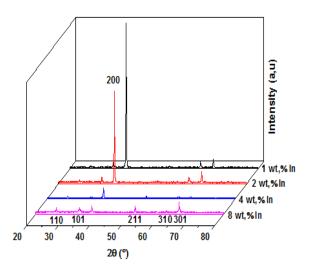


Figure 01: X-ray diffraction spectra of In doped SnO₂ thin films prepared at different indium concentrations

The grain size is determined from the width at half peak height of (200) and (301) plans using the Scherrer formula [16]:

$$G = \frac{0.9\lambda}{\beta \cos \theta} \tag{1}$$

Where G is the grain size, λ is the X-ray wavelength ($\lambda = 1.5406 \text{ A}^{\circ}$), β is the full width at half-maximum (FWHM), and θ is Bragg angle of the diffraction peaks.

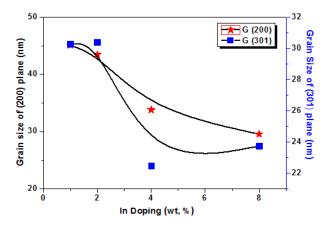


Figure 02: The variation of grain size of of (200) and (301) plans

In doped SnO_2 thin films prepared with indium doping

The variation of grain size of Indium doped SnO_2 thin films according to (200) and (301) plans as a function of Indium concentration is shown in Figure 2. We notice a decrease in grains size with increasing indium concentration, which implies deterioration in the crystallinity of the films. This is good in agreement with X-Ray results. This reduction of the grain size is probably caused by nucleation centers enhancement when indium ratio increases.

The Optical properties of Indium doped SnO₂ thin films deposited at different Indium concentrations were performed by measuring the transmittance in the wavelength region 300 to 1000 nm. Figure 3 shows the optical transmission spectrum of Indium doped SnO2 thin films, as it can be seen, the transmittance decreases with increasing In doping, the film doped with 1% In have a high transmission in the longer wavelengths (>400 nm) around 93 %, and present a sharp ultraviolet cut-off.

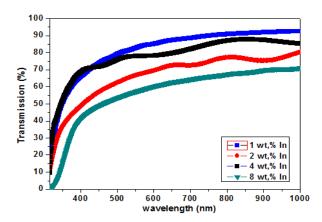


Figure 03: Optical trasmission spectra of In doped SnO₂ thin layer preapared at different indium concentrations.

The optical gaps energy, was calculated from the tauc formula [17,18] wich is given by following relation

$$(\alpha h \nu)^n = B(h \nu - E_g) \tag{2}$$

Where B is a constant, hv is the incident wavelength, α is the absorption coefficient and Eg is the optical band gap. On the other hand, we have used the Urbach energy (E_u), which is related to the disorder in the film network, as it is expressed follow [19, 20]:

$$\alpha = \alpha_0 \exp\left(\frac{h\upsilon}{E_u}\right) \tag{3}$$

Where α_0 is a constant hv is the photon energy and E_u

is the Urbach energy.

Figure 4 shows the variation of the optical gap and disorder depending on the concentration of indium doped SnO_2 thin films. It is observed that the disorder and the optical gap vary inversely. The lowering of the band gap is possibly due to the presence of In impurities in the SnO_2 structure, which induces the formation of new recombination centers. This leads to more disorder formation in film network.

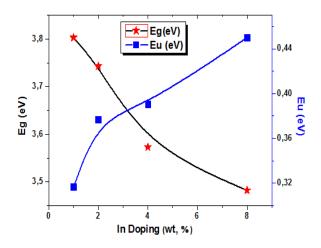


Figure 04: The variation of optical band gap and Urbach energy of In doped SnO₂ thin films with indium doping

Figure 5 show the variation of sheet resistance of indium doped SnO₂ thin films as a function of indium doping. The sheet resistance of these films increases with increasing In content. This rise may be due to the decrease of charge carriers concentration depending on the substitution of Sn⁴⁺ by In³⁺ ion. The grain boundaries arise attributed to the development of small size grains as consequence of In incorporation, and also by the incomplete bonding between the layers of the atoms. The boundaries act as charge carriers traps causing then an increase in films sheet resistance [21].

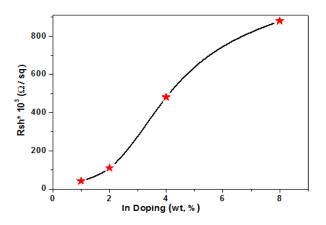


Figure 05: The variation of the sheet resistance with deposition In content of SnO₂ films

4 CONCLUSION

In summary, high-quality transparent of undoped and Indium doped tin dioxide thin films were grown on glass substrates at 400°C by ultrasonic spray chemical technique (SPC). The influence of Indium concentration (1 to 8 wt.%) on structural, optical and electrical properties was studied. XRD analysis of SnO₂ thin films reveals that films have a tetragonal structure of SnO₂. The average grain size decreased with increase of In content. The optical gap varies between 3.48 and 3.80 eV. Moreover, the sheet resistance is enhanced up to $880*10^3 \, (\Omega/\text{sqr})$ with in doping increase.

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