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# **Improved Physical Properties of ZnO Films with a second Superposed SnO2 very Thin Films Deposited by Spray Pyrolysis Method**

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**Abstract:** ZnO Thin films were deposited on glass substrates at  $350 \pm 10$  OC by pyrolysis spray technique. A very thin layer of SnO2 was deposited under ZnO films for the purpose of getting a bilayer thin SnO2/ ZnO. A thermal annealing was carried out for the bilayer with a temperature of 350 0C for one hour. The structural, optical and electrical properties of monolayer ZnO films and SnO2/ZnO bilayer thin films were investigated. X-ray diffraction confirmed the hexagonal wurtzite structure of ZnO films, and the hexagonal tetragonal structure of SnO2/ZnO bilayer films. The optical measurement showed a rise of the average transmittance from 81 % to 94 % for SnO2/ZnO annealed bilayer films. The optical band gap varied between 3.22 to 3.31 eV for SnO2/ZnO annealed bilayer films. The maximum electrical conductivity of 17.85 ( $\Omega$  .cm-1) has been observed for SnO2/ZnO annealed bilayer films.

**Keywords:** Thin film, zinc oxide, spray pyrolysis, band gap.

## **1 Introduction**

Transparent conductive oxides (TCO) are currently of great commercial and scientific importance and are widely used in applications in electro-optical devices [1], gas sensors [2] and solar cells [3], because to the high transmittance in the ultraviolet visible (UV - Vis) range wavelength and electrical conductivity [4,5]. The TCO/Metal and TCO/TCO structure have been manufactured to enhance the TCO properties towards solar photovoltaic applications [6,7,8].

In recent years, zinc oxide (ZnO) has been of increasing interest in many research works because of its many potential applications. ZnO is a nontoxic (II-VI) binary semiconductor material [9] with very interesting characteristics such as piezoelectricity [10], photoconductivity [11], optical waveguides [12], gas probes [13], excellent electrical, optical, mechanical, and chemical sensing properties, as well as thermal stability. These materials are wide-band-gap (3.27 eV) semi-conductors with a large (60 meV) exciton binding energy [14]. ZnO thin films can be obtained by several techniques such as

pulsed laser deposition [15], thermal evaporation [16], sputtering [17], sol gel [18] and spray pyrolysis [19,20,21,22,23]. Several researchers have reported TCO characteristics of bilayer films involving ZnO and SnO2 films such as Vaezi et al. [24], Ravikumar et al [25] and Anandhi et al [26].

Enhancement physical properties of the ZnO films by second superposed SnO2 thin films and the thermal annealing of the SnO2/ZnO bilayer are studied in this work. To the best of our knowledge, the enhancements of the properties of SnO2/ZnO bilayer grown by spray pyrolysis technique have not yet been reported in the literature.

## **2 Experimental Details**

#### *2.1 Preparation of Thin films*

The  $ZnO$ ,  $SnO<sub>2</sub>$  and  $SnO<sub>2</sub>/ZnO$  thin films were prepared on glass substrates using the spray pyrolysis technique as shown in fig.1. The glass substrates were cleaned by diluted hydrochloric acid (HCl) and acetone, washed by double distilled water and dried. The sprayed solutions have been obtained by dissolving zinc nitrate  $(Zn(NO<sub>3</sub>)<sub>2</sub>)$  (50ml) and tin chloride (SnCl2) (20 ml), in doubly distilled water with a

$$
30 \leq \frac{1}{2}
$$

K. Salim, M. N. Amroun: Improved Physical Properties of ...

concentration of (0.1 M), respectively; were used as host precursors.

In the samples preparation process, the following conditions have been considered: substrate temperature 350  $\pm$  10 °C, solution flow of 2 ml /min and spray nozzle to heating plaque distance of 30 cm. We took two minutes interval time between spraying periods in order to avoid excessive cooling of the glass substrates resulting from continuous spray. One also noted that the prepared solutions were immediately sprayed to avoid any possible chemical changes with time. The structural characterization was performed at room temperature using a Bruker X-ray diffractometer model D2 Phaser with CuKα radiation ( $\lambda$ =1.5406 Å). The optical transmittances have been recorded between 200 and 2500 nm wavelength using a JASCO 570 type UV-visible-NIR double-beam spectrophotometer. The electrical parameters were measured by using the ECOPIA HMS-5000 Hall Effect measurement system at room temperature.

**3 Results and Discussions** 

# *3.1 Structural Parameters*

The XRD patterns of  $ZnO$ ,  $SnO<sub>2</sub>$ ,  $SnO<sub>2</sub>/ZnO$  and  $SnO<sub>2</sub>/ZnO$ annealed bilayer films are shown in Fig. 2. The XRD patterns clearly show five well defined peaks for ZnO (100), (002), (101), (102) and (103) and three peaks for SnO<sub>2</sub> (110), (200) and (211). The SnO<sub>2</sub>/ZnO bilayer thin films exhibited a The XRD patterns of ZnO, SnO<sub>2</sub>, SnO2/ZnO and SnO2/ZnO annealed bilayer films are shown in Fig. 2. The XRD patterns clearly show five well defined peaks for ZnO (100), (002), (101), (102) and (103) and three peaks for  $SnO<sub>2</sub>(110)$ , (200) and (211). The  $SnO<sub>2</sub>/ZnO$ bilayer thin films exhibited a presence of the two phase of hexagonal wurtzite structure of ZnO (JCPDS N◦: 36-1451) [27,28] and the tetragonal structure of  $SnO<sub>2</sub>$  (JCPDS N∘: 041-1445) [29].



Fig.1: Sample models for ZnO, SnO<sub>2</sub>/ZnO bilayer, and SnO<sub>2</sub>/ZnO bilayer annealed films.



**Fig.2:** XRD patterns of ZnO, SnO2, SnO2/ ZnO Bilayer and SnO2/ ZnO Bilayer annealed films.

tetragonal structure were determined from XRD results using the following equation [28,29,30]:

$$
\frac{1}{d_{hkl}^2} = \frac{4}{3} \left( \frac{h^2 + hk + k^2}{a^2} \right) + \frac{l^2}{c^2}
$$
 (1)

$$
\frac{1}{d_{hkl}^2} = \left(\frac{h^2 + k^2}{a^2}\right) + \frac{l^2}{c^2}
$$
 (2)

Where: d is interplanar spacing, (hkl) are the miller indices. The lattice constant (a, c) and values are tabulated in (Table 1)**.** 

The calculated values 'a' and 'c' of all the films are in good agreement with those provided by the standard card of ZnO hexagonal phase (JCPDS N◦: 36-1451) with lattice parameters a=3.249A $\degree$  and c=5.20 A $\degree$ ) and of SnO<sub>2</sub> tetragonal phase (JCPDS No. 41-1445) with lattice parameters a=4.738 A $^{\circ}$  and c=3.187 A $^{\circ}$ ). A slight deviation has been observed from the bulk lattice parameters indicate the presence of stress/strain in the films, which might be caused by the defects, stoichiometric deviations and/or mismatch of thermal expansion coefficients of both the substrate and film [31].

The crystallite size (D) was calculated for preferential orientations using Scherer's equation. In addition, structural defects such as micro-strain (ε) and



dislocation density (δ) were calculated using the following relations [32]: The calculated crystalline parameters of the films are regrouped in (Table 1)**.** 

$$
D = (0.9. \lambda) / (\beta \cdot \cos \theta) \tag{3}
$$

$$
\varepsilon = \frac{\beta \cos \theta}{4} \tag{4}
$$

$$
\delta = \frac{1}{n^2} \tag{5}
$$

The crystallite size decreased from 34.67 nm for pure ZnO to 25.03 nm for bilayer SnO2/ZnO, which can be attributed to increasing structural defects [33].

The crystallite size of bilayer films increased with annealing temperature from 25.03 nm to 26.597 nm, which can be attributed to activate atom diffusion and hence, facilitate to repairing the dislocated atomic occupancies and even promote the coalescence of adjacent particles [34-35]. The bilayer annealed films exhibited a less structural disorder (strain and dislocation density), which indicate improvement in the crystallinity of the films.

#### *3.2 Optical parameters*

The optical transmittance spectrum as a function of the wavelength for  $ZnO$ ,  $SnO<sub>2</sub>$ , the bilayer  $SnO<sub>2</sub>/ZnO$  and SnO2/ZnO annealed films deposited on glass substrates is

showed in Figure 3. The SnO2/ZnO bilayer are highly transparent with an average transmission of 78-85% in the visible-IR region. The transmittance achieved 94 % for the bilayer films after annealing, which is requested for satisfactory optical window in visible range in photovoltaic systems [36]. Improving in the transmittance of the films could be explained by the reduction of surface roughness [36], to a low scattering effects resulting from the structural homogeneity of the films [39]. The deficiency of interference fringes in transmission spectra is due the surface roughness caused by the spray pyrolysis technique. This is maybe due to very small droplets resulting from this technique that vaporize above the glass substrates and condense as clusters [37]. The formula used to calculate the optical band gap energy of the films is [38]:

$$
(ahv)^n = A(hv - Eg) \tag{6}
$$

Where  $\alpha$ : Absorption coefficient;  $hv$ : Photon energy, A: relation constant, *Eg*: optical band gap. For: n=2 direct band gap semiconductors and n=0.5 indirect band gap semiconductors. The absorption coefficient  $\alpha$  is derived from transmittance T using the beer-lambert law [28]:

$$
\alpha = \frac{1}{t} \ln \frac{1}{T} \tag{7}
$$

Where t: is the film thickness.



**Table 1:** Structural parameters of the deposited films.

**Table 2:** Optical parameters of the deposited films.

samples	$Eg$ (eV)	Eu (eV)
ZnO	3.22	0.081
SnO <sub>2</sub>	3.91	0.326
Bilayer $SnO_2/ZnO$	3.28	0.155
Bilayer $SnO_2/ZnO$ annealed	3.31	0.142

The obtained optical band gap of the ZnO films is 3.22 eV and  $3.91$  eV for the SnO<sub>2</sub> films, which agrees with the literature [40]. The optical band gap increases from 3.22 eV for pure ZnO films to 3.28 eV SnO2/ ZnO Bilayer. This gap blue shifting could be explained by the contribution of the Moss–Burstein effect of the degenerate energy levels are created with band filler which make the level of Fermi moves over the edge of the conduction band [41,42,43]. The optical band gap increases after annealed to 3.31 eV due to the oxygen diffusion with annealing temperature [44].



The disorder measure is characterized by Urbach tail. The Urbach energy is estimated from the variation of the



**Fig. 3:** Optical transmittance of the prepared films.



Fig. 4: Optical gap of ZnO, SnO<sub>2</sub>, SnO<sub>2</sub>/ ZnO Bilayer and SnO<sub>2</sub>/ ZnO Bilayer annealed films versus  $(\alpha h v)^2$ .

absorption coefficient. This absorption coefficient of the films shows a tail for the photon energy sub-band [38]:

$$
\alpha = \alpha_0 \exp(\frac{h\nu}{E_U}) \tag{8}
$$

where  $\alpha_0$  is a constant and *Eu*: is the Urbach energy. The Urbach energy is found to increase from 0.081 eV for pure ZnO films to  $0.155$  eV of the SnO<sub>2</sub>/ZnO bilayer (Table 2), due to an increase in structural disorder. This is consistent with the results from XRD analysis. The Urbach energy decreased from 0.155 for SnO<sub>2</sub>/ZnO bilayer to 0.142 eV with annealing, which indicate the decrease in the structural disorder [44].

## *3.3 Electrical Parameters*

The electrical properties were measured by measuring ECOPIA HMS-5000 Hall Effect measurement system at room temperature. The negative values of carrier concentrations showed that as deposited films are n-type conductivity. It observed from Figure 5 that the conductivity of the films rises from  $3.37 \times 10^{-03}$  ( $\Omega$  .cm)<sup>-1</sup> for ZnO films to 17.85 ( $\Omega$  .cm)<sup>-1</sup> for SnO<sub>2</sub>/ZnO bilayer annealed films. The increase in the electrical conductivity of the films could be assigned to increase in the carrier concentrations of the films from  $3.66 \times 10^{+16}$  for pure ZnO films to  $2.28.10^{+20}$  (cm<sup>-3</sup>) for SnO2/ZnO bilayer annealed films.



**Fig.** 5: (a) conductivity and (b) Carrier Concentrations of ZnO, SnO<sub>2</sub>, SnO<sub>2</sub>/ ZnO Bilayer and SnO<sub>2</sub>/ ZnO Bilayer annealed films and SnO2/ ZnO Bilayer annealed films.

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Samples	Sheet Resistance (Rsh) $(\Omega/\text{sq})$	Mobilite $\text{cm}^2/\text{V}_\text{S}$ )	Carrier Concentration $s$ (cm <sup>-3</sup> )	Conductivity $(\Omega.cm)$
ZnO	$1.01\times10^{-07}$	$5.78 \times 10^{-01}$	$-3.66\times10^{+16}$	$3.37.10^{-03}$
SnO <sub>2</sub>	$5.93\times10^{-03}$	$5.61\times10^{-01}$	$-6.33\times10^{+19}$	5.69
Bilayer $SnO_2/ZnO$	$1.36\times10^{-04}$	$8.68\times10^{-01}$	$-1.89\times10^{+19}$	2.63
Bilayer SnO2/ZnO annealed	$2.01\times10^{-03}$	$4.59\times10^{-01}$	$-2.28\times10^{+20}$	17.85

**Table 3:** Electrical parameters of the deposited films.

#### *3.4 Optical Electrical Performance*

The good TCO material should exhibit a low resistivity and a good transmittance spectral at the same time. For that, we applied the same idea of Ref. [45] and constructed a 2D diagram composed of two contradictory TCO characteristics: the sheet resistance *R<sub>sh</sub>* and the optical loss defined as  $1-(at 400-800 nm)$ . For an optimal comparison, we calculated  $(1-T)$  and  $R_{sh}$  for our SnO<sub>2</sub>/ZnO annealed films (Figure 6). We also included other experimental values from 16 different references; SnO2/ZnO annealed surpassed all our samples in terms of optical loss. From the figure 6, our sample  $SnO_2/ZnO$ annealed dominates the 16 experimental works in terms of optical loss and 9 in terms of sheet resistance. The subset, i.e. Refs [33,47] constitute the so called "Pareto front" or "Pareto set", it contains points that aren't dominated by the other points. Our greatest sample  $(SnO_2/ZnO)$  bilayer annealed films) belongs to the present sub-optimal set.



Fig. 6: A pareto front of SnO<sub>2</sub>/ZnO and SnO<sub>2</sub>/ZnO annealed thin films prepared in this work and literature.

#### **4 Conclusions**

In this study, good quality of SnO2/ZnO bilayer grown by spray pyrolysis technique spray technique. The XRD profiles; the bilayer films have several peaks associated of the picks ZnO and SnO2 films. The optical and electrical structural characterization demonstrate that the annealed bilayer films show good optical and electrical parameters, such as high transmittance in the visible range  $> 90\%$ , wide optical gap  $(Eg=3.31 \text{ eV})$ , and high electrical conductivity (17.85  $\Omega$ .cm), asserting it is potential window layer for obtaining a high efficient solar cell.

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