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Effect of capillary tube on structural and Optical Properties of SnO₂ Thin Films Prepared by APCVD

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Abstract : Tin oxide SnO2 films were prepared by atmospheric chemical vapor deposition (APCVD) technique. Our study focus on prepare SnO_2 films by using capillary tube as deposition nozzle and the effect of these tubes on the structural properties and optical properties of the prepared samples. X-ray diffraction (XRD) was employed to find the crystallite size. (XRD) studies show that the structure of a thin films changes from polycrystalline to amorphous by increasing the number of capillary tubes used in sample preparation. Maximum transmission can be measured is (95%) at three capillary tube. (AFM) where use to analyze the morphology of the tin oxides surface. Roughness and average grain size for different number of capillary tubes have been investigated. The optical properties of the SnO₂ thin films were determined using UV-Visible spectrum.

Keywords : APCVD ; SnO₂ thin film; optical properties; capillary tube.

I. Introduction

SnO₂ is chemically inert, mechanically hard, and can resist high temperatures [1]. tin Oxide thin films are n-type semiconductors with low electrical resistance with high optical transparency in the visible range[3]. It is used as a window layer in solar cells [2-3], heat reflectors in solar cells [4], various gas sensors [5], Tin oxide is a wide band gap ($\approx 4 \text{ eV}$) and indirect band gap (of about 2.6 eV) nonstoichiometric semiconductor.

Thin oxide films have been deposited using different techniques, such as thermal evaporation [6,7], sputtering [7–13], chemical vapour deposition [6,14–16], sol–gel dip coating [6,13,17], painting [6,13,18], spray pyrolysis [6,8,16,19-24], hydrothermal method [25] and pyrosol deposition [6,16,26–28]. In this research, SnO2 thin film was preparing by using atmospheric chemical vapor deposition (APCVD) technique. A SnO₂ thin film CVD process normally consist of four basic ingredients: vapor of the so-called 'precursor' which is a tincontaining molecule (i. e. SnCl₄), an oxygen source (i. e. H₂O vapor and/or O₂), a carrier gas (i. e. N₂ or Ar) and a substrate. When these four ingredients are brought together inside a CVD reactor a thin film can be synthesized.

II. Experimental details

The substrate which are glass were cleaned first by ultrasonically cleaned in distilled water to remove the dust and then they were ultrasonically cleaned in ethanol (purity 98%) for at least 15 min. APCVD is a chemical process which consists of heating hydrated tin dichloride (SnCl₂. 2H₂O) under oxygen flow. The vapor of the precursor reacts with oxygen then carried on the glass substrate through different nozzle (1, 3, 5, 7 capillary tubes) by the O₂ gas. N₂ gas use to prevent the oxidation of substrate during heating. The substrate temperature (working temperature) was 450°C.

Deposition parameters of tin oxide film				
Thin film	SnO2			
Substrate	Glass			
Temperature (°C)	450			
O_2 gas flow rate	0.5 L/min			
N ₂ gas flow rate	0.5 L/min			
No. capillary tube	1,3,5,7			
Time deposition	1 sec			

 Table -1: The operating parameters for (APCVD) system

III. Results And Discussion

A. Structural properties

The XRD patterns recorded for SnO_2 thin films deposited by the APCVD technique as a function of the different no. of capillary tubes (1, 3, 5, 7 capillary tubes) that use to deposition thin film. The SnO_2 films deposited on glass substrate at temperature (450)°C. The structure of a thin film changes from polycrystalline to amorphous by increasing the number of capillary tubes used as shown in Fig. (1). The particle size (D) was estimated using the Scherrer equation:

$$D = 0.9\lambda / \beta \cos \theta \qquad (1)$$

where D is the crystallite size, λ is the X-ray wavelength, β is the full width at half maximum of the diffraction peak, and θ is the Bragg diffraction angle of the diffraction peaks. The particle (grain) size decrease with increase the no. of capillary tubes as shown in figure (2)

The use of a different no. of capillary tubes in the deposition process change the shape of the thin film due to the fact that the number of particles attached to the glass base will double and therefore the thickness of the thin film will increase with increase the no. of capillary tubes as shown in figure (2). This interpretation is consistent with the results (76.4, 195.8, 123.5, and 355.7 nm) for (1, 3, 5, and 7) capillary tubes respectively.

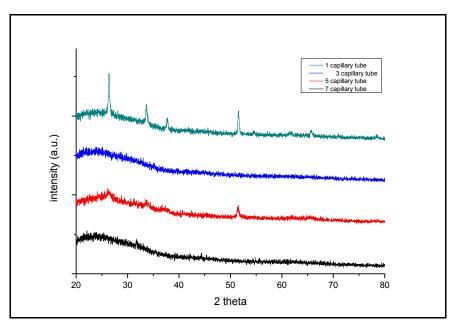
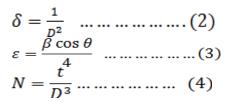


Figure 1. X-ray diffraction pattern for SnO₂ film deposited on glass substrate at different nozzle

The structural parameters such as dislocation density (δ), strain (ϵ), and the number of crystallites (N) per unit area were calculated using the relations[1]:



where λ is the wavelength of the radiation used (0.154184 nm), β the full width at half maximum, and θ the angle of diffraction.

The values of Strain (ϵ), Grain size (D), and Dislocation density (δ) are calculated from eq. (4) are given in table (1) respectively.

Table (1) Microstructural parameter of SnO_2 deposits for different nozzle and film thickness. Deposition time: 1 sec.

No. capillary tubes	Thickness (t)nm	Grain size nm(avr)	strain (ϵ) × 10 ⁻	dislocation density (δ) lines/nm ²	number of crystallites (N) per unit area(nm ⁻²)
1	76.4	13.43378	2.697	0.005541	0.031514
3	195.8	11.70254	3.096	0.007302	0.122172
5	123.5	12.50748	2.897	0.006392	0.063119
7	355.7	12.08329	2.999	0.006849	0.201618

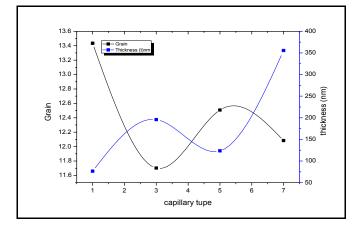
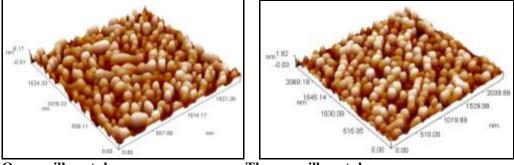


Figure (2) Change the Grain size and thickness of the thin film with the number of capillary tubes

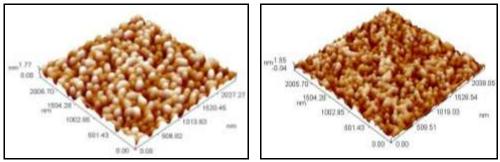
B. Surface topography properties -

The study of surface morphology of SnO_2 thin films deposited by chemical vapor deposition method has been carried out using atomic force microscopy (AFM). It can be seen from fig. (3) that with increasing no. of capillary tube the degree of surface roughness decreases from 1.81nm to 0.259 nm.



One capillary tubes

Three capillary tubes



five capillary tubes

seven capillary tubes

Figure 3. AFM images of samples of different no. capillary tubes

C. Optical properties -

Figure (4) shows the variation of transmittance (T) with respect to the wavelength of SnO₂ thin films with different no. Capillary tubes. The maximum transmittance is 95 % (in visible range nm) by using 3 capillary tubes. The visible transmittance decrease to 80% with increase the no. capillary tubes above 3.

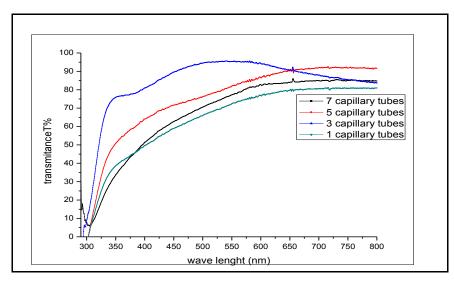


Figure 4. Optical transmittance for SnO₂ films for a different capillary tubes

The theory of optical absorption gives the relationship between absorption coefficient (α) and the photon energy *hv* [29,30]:

$$\alpha = \frac{B(hv - Eg)^2}{hv} \tag{2}$$

where *B* is a constant, *Eg* is the optical band gap, *h* is the Planck's constant, x = 0.5 for the directly allowed transitions, and α can be calculated from the transmission spectrum using ($\alpha = \frac{1}{d} \ln \frac{1}{t}$) Then the incident photon energy is related to the direct band gap Eg by equation:

$$(\alpha hv)^2 \alpha (hv - Eg)$$
 (3)

The *Eg* values were obtained by extrapolating the linear part of the plot of $(\alpha hv)^2$ vs. *hv* to α =0, as shown in Fig.(5). The values of the energy gap are significantly decreased from 3.89 eV to 3.6 eV with the increase of the number of capillary tube from 1 to 7. This may be ascribed to an effect of change in the grain size of polycrystalline films.

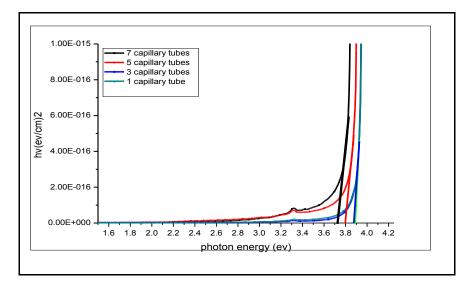


Figure 5. The variation of $(\alpha hv)^2$ vs. hv for determining the direct band gap Eg of the APCVD coated SnO2 films for a different nozzle.

IV. Conclusion

In this work, a facile and simple APCVD technique was successfully used to deposit SnO_2 thin films. We studied the effect of using capillary tubes on the structural and optical properties of the deposited samples. Surface roughness decreases from 1.81nm to 0.259 nm with increasing no. of capillary tubes. Transparency is increasing by increasing the number of capillary tubes used in the deposition process to three tubes, but starting to decrease by increasing the number of tubes above the three and the optical band gap of the films are decreased by increasing the number of capillary tubes.

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