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The Studies on effect of substrate Temperature on Synthesis and Properties of Doped & Undoped Transparent Conducting SnO_2 Thin Films

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Abstract

The undoped and Fluorine doped tin oxide thin films have been prepared by spray pyrolysis from SnCl2 precursor at the various substrate temperatures. The Fluorine doped tin oxide thin films belong to special class oxides that combine high electrical conductivity with high optical transparency. Such transparent Conducting Oxide (TCO) thin films thus an important component for optoelectronic applications. Spray pyrolysis deposition is a simple and relatively low cost technique for thin film deposition. This work comparers the fluorine doped and undoped SnO_2 *thin films. The effect of substrate temperature and fluorine doping on the structural, optical and electrical properties of thin films have been studies. The characterization of samples was carried out by X-ray diffraction, scanning electron microscopy, UV-VIS spectrophotometer and the four probe method. The as-prepared films are polycrystalline with a tetragonal crystal structure. The films have moderate optical transmission of the pure* SnO_2 *and fluorine doped* SnO_2 *deposited films is71.2% and 73.3%. The obtained results reveal that the properties of the films are greatly affected by substrate temperatures and doping levels.*

Keywords: *Pyrolysis; Tin oxide; X-Ray diffraction; Electrical properties and measurements.*

1. Introduction

The highly transparent and conducting thin films and its high transmittance and conductivity, have wide applications [1, 2]. In recent years, there has been a growing interest in the use of transparent conducting oxide thin films as conducting solar window materials in thin film solar cells [3–6], as heat reflectors for advanced in solar application[7,8] and as gas sensors [5–13]. Tin oxide is the first transparent conductor which is significantly commercialized [5, 9, 14]. Among the different transparent conductive oxides, SnO_2 films doped with fluorine are most appropriate for use in solar cells, owing to its low electrical resistivity and high optical transmittance. SnO_2 is chemically inert, mechanically hard, and can resist high temperatures [5]. Doped or undoped SnO_2 can be synthesized by numerous techniques such as thermal evaporation [3,9], sputtering [5,9–12,16,17], chemical vapor deposition [3,18–20], sol–gel coating [3,17,21], painting [3,17], spray pyrolysis [3,5,7,13],and hydrothermal method. Among the various deposition techniques, the spray pyrolysis is the most suitable method for the preparation of doped tin oxide thin films because of its simple and inexpensive experimental arrangement. It is easy of add to various doping materials, reproducibility, high growth rate and mass production capability for uniform large area coatings and is the essential characteristics of the simple spray pyrolysis technique [16]. Usually, the chemical spray pyrolysis method is used to deposit FTOs at optimized substrate temperatures at 600^0C [20].

In the present work, SnO_2 and fluorine doped SnO_2 thin films were prepared by the spray pyrolysis technique at various substrate temperature using dehydrate stannous chloride ($SnCl_2 \cdot 2H_2O$) and ammonium Bifluoride ($NH_4 HF_2$) as precursors. The aim of this work is to study the relationship between the doping and undoping levels and some physical properties of SnO_2 & SnO_2:F thin films such as the electrical, structural and optical properties.

2. Experimental details

The fluorine-doped and undoped tin oxide thin films in the present study were prepared using a homemade spray pyrolysis apparatus. The both thin films was deposited on an optical glass substrate at various substrate temperatures 300^0C, 400^0C, 500^0C and 600^0C. Dehydrate stannous chloride ($Sncl_2$:$2H_2O$) was used for making the precursor solution. This precursor was dissolved in 4 mL concentrated hydrochloric acid (HCl) and then added with methanol served as the starting solution. The solution molarities of 0.1 M were deposited and doping 0.1 M, ammonium Bifluoride ($NH_4 HF_2$) by ranging from 2 ml to 10 ml in step of 2 ml. Other preparative parameters like nozzle to substrate distance (45 cm), air flow rate (25 L/min) and substrate temperature (600^0C) were kept constant as the optimized values. The samples were analyzed with X-rays diffractometer using a monochromatic radiation Cu-Kα, λ=1.5406 Å at 30 kV, 10 mA in the range of scanning angle 20° < 2θ < 79.99°. Morphological analysis of the films was carried out by Scanning Electron Microscope (SEM). Optical transmittance spectra of the films were measured using a PC based UV-VIS Systronics spectrophotometer 119 model in the range 200 nm to 999 nm of wavelength with air as reference. Four probe set up was used for electrical measurements at room temperature.

3. Results and discussion

3.1 XRD micro-structural characterization of tin oxide films

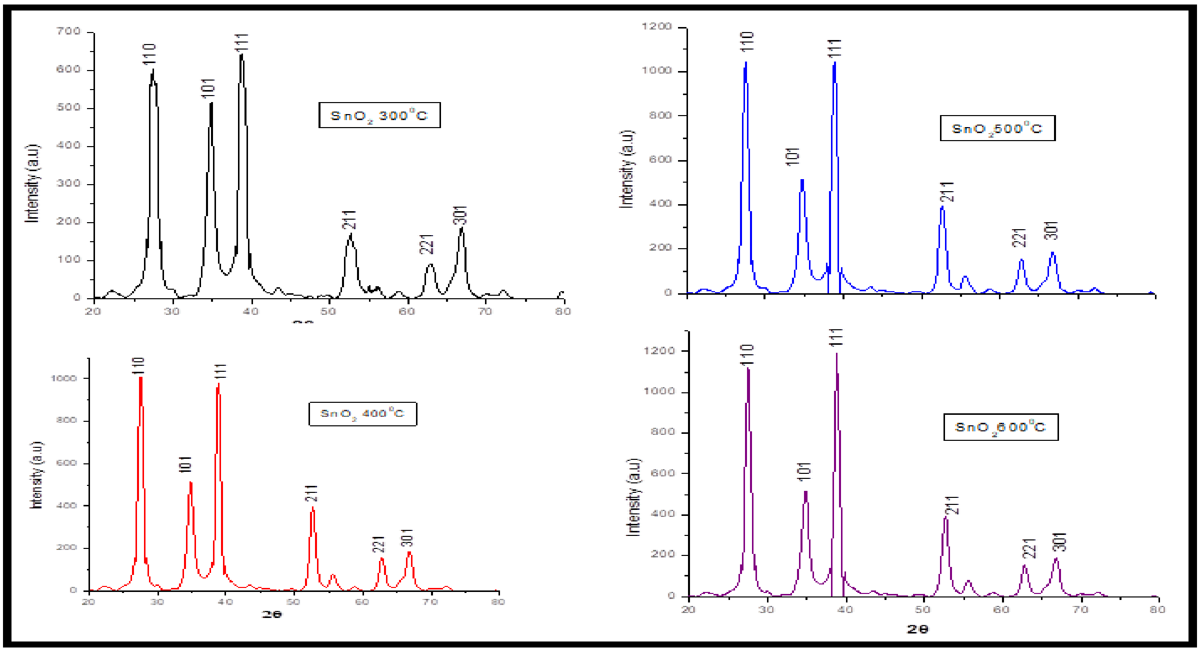

Fig. 1 XRD patterns of SnO_2 thin films.

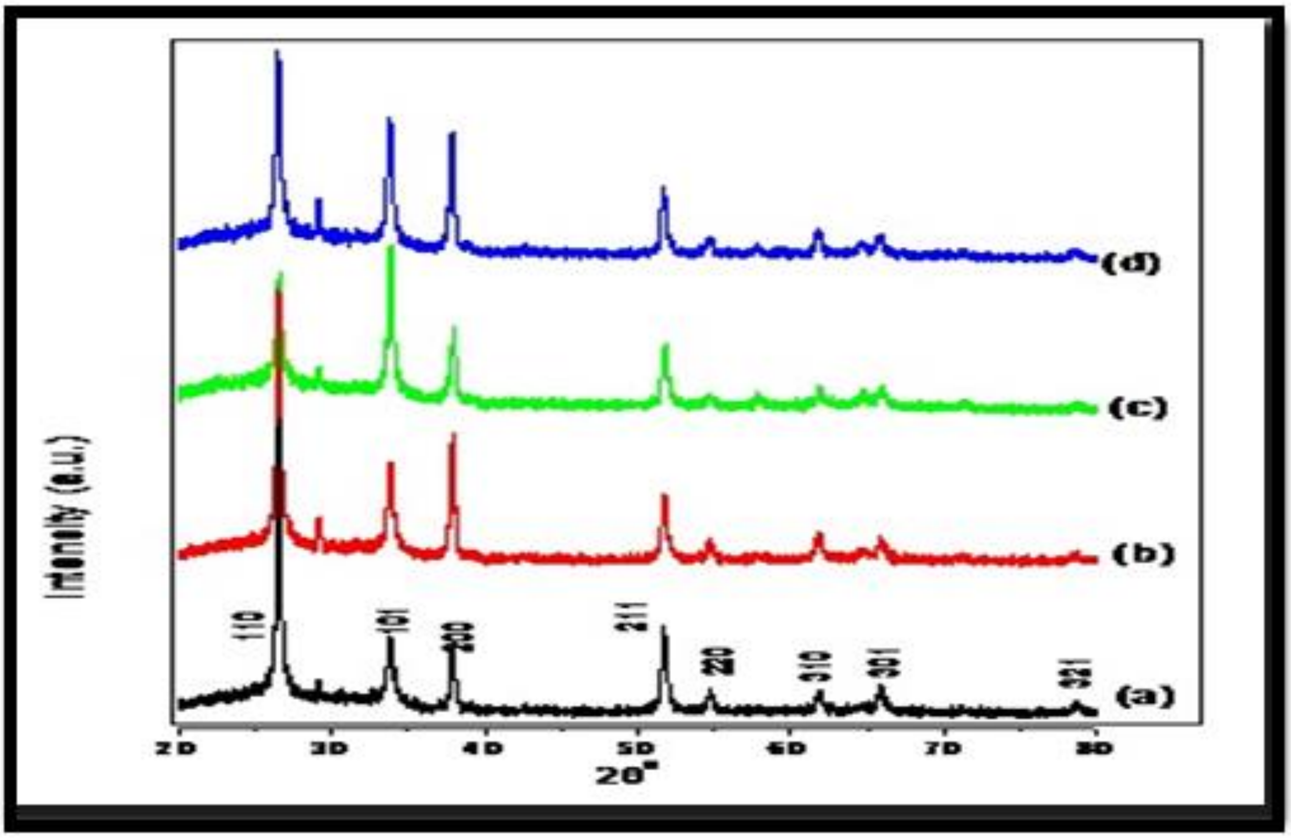

Fig. 2 XRD patterns of Fluorine doped SnO₂ thin films.,

Figure 1 shows the XRD patterns of the Tin oxide thin films at various substrate temperatures 300^0C, 400^0C, 500^0C and 600^0C. The tin oxide thin film can be tetragonal rutile structured because its diffraction peaks can be clearly observed at 2θ = 26.6°, 33.5°, 37.8° and 51.7°, which corresponding to (110), (101), (200) and (211) directions of the rutile structured SnO_2. The XRD pattern of Fig.1 suggests that average grain sizes of SnO_2 thin film synthesized are approximately 3-4nm, based used on the Scherer formula:

$$D= \frac{0.9\lambda}{\beta\cos\theta}$$

Where D is the mean grain size, λ is the wavelength (1.54056nm for Cu Kα1), β is the full width at half maximum, θ is the scanning angle. Also the height of peak increases with increase in substrate temperature. Similarly, Fig 2. Shows the XRD patterns of the Fluorine doped Tin oxide thin films at substrate temperature 600^0C for total spray solution 20ml and fluorine doped by ranging from 2 ml to 10 ml in step of 2 ml. Here *SnO_2*: F thin film can be observed tetragonal rutile structured because its diffraction peaks can be clearly observed that the increase in the fluorine doping Concentration a new direction of crystal growth appears corresponding to the reflection from the (1 1 0), (1 0 1) (2 1 1) and (2 0 0) planes at 2θ = 26.6°, 33.5°, 37.8° and 51.7°. The presence of other peaks such as (1 0 1), (2 2 0), (3 1 0), (3 0 1) and (3 2 1) have also been detected but with substantially lower intensities at the higher doping concentration.

3.2 Surface morphology of as-prepared films

Fig.3 shows a typical SEM micrograph of the tin oxide film deposited at 20ml solution flow rate. The SEM microstructures reveal that all the films have a homogeneous surface morphology with nanocrystalline grains, also all the films are without any cracks and holes. Patterns indicate that films are polycrystalline in nature. Comparison of interplaner distance (d) values with JCPDS data show that phase present in the deposited films belongs to pure tetragonal structure. The EDAX spectra of the same film reveal the elemental analysis, which shows that Sn and O were found to be 89.55: 11.45 in percent.

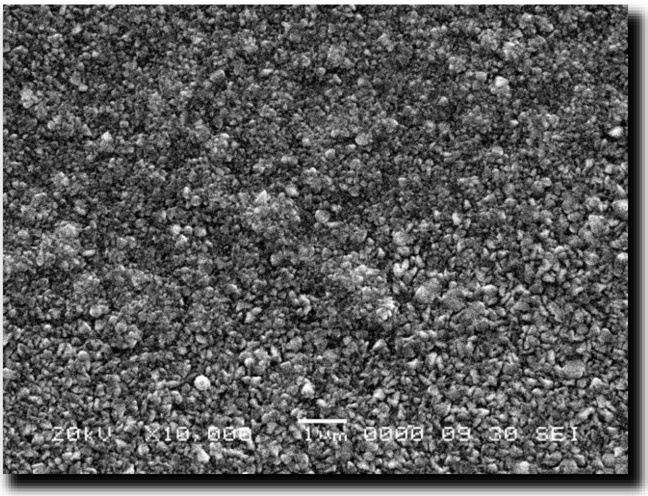

Fig 3. Typical SEM micrograph of SnO_2 thin films.

Fig.4 shows that SEM micrograph of the SnO_2: F thin film, fluorine doped by ranging from 2 ml to 10 ml in step of 2 ml. The SEM microstructures reveal that all the films have a homogeneous surface morphology with nanocrystalline grains, also all the films are without any cracks and holes. Patterns indicate that films are polycrystalline in nature. The Grain size ranges from 50 nm-250 nm.

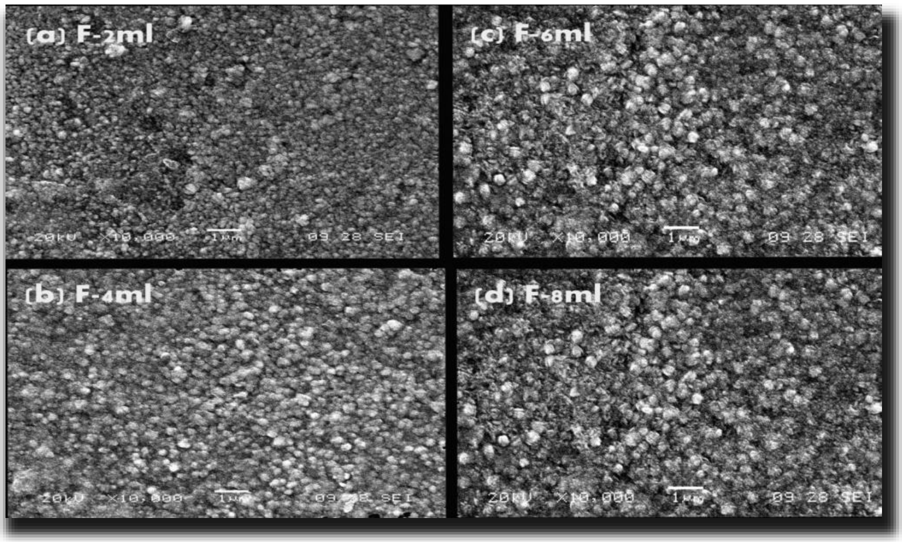

Fig.4: XRD Pattern for SnO_2: F thin film at (a) 2 ml (b) 4 ml(c) 6 ml and (d) 8 ml fluorine concentrations.

3.2 Optical properties

Fig.5 shows the variation of optical transmittance spectra in the region (200 nm - 999 nm) for the SnO_2 & SnO_2: F thin films. From the graph, the films deposited at the substrate temperatures 600^0C exhibits highest transmittance in the range of 744nm-760nm which maximum transmittance is 71.2% and 73.3%.

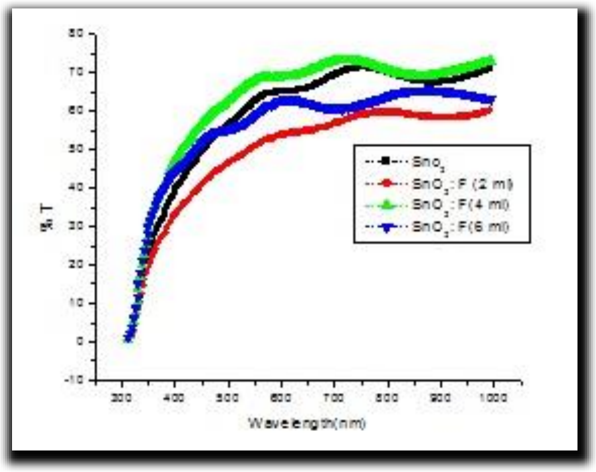

Fig5: Optical Transmittance against wavelength at different Fluorine concentration of the SnO_2 thin films.

Analysis on Optical Band-Absorption Coefficient:-

The analyzing the optical data with the expression for optical absorption coefficient 'α' and photon energy 'hυ' determine the optical band gap 'Eg'. Fig.6 & 7 shows the plot of $(\alpha h\upsilon)^2$ vs 'hυ' .Extrapolation of the linear portion of the plots to energy axis yielded the direct band gap values of SnO_2 & SnO_2: F are 3.6eV & 3.80eV.

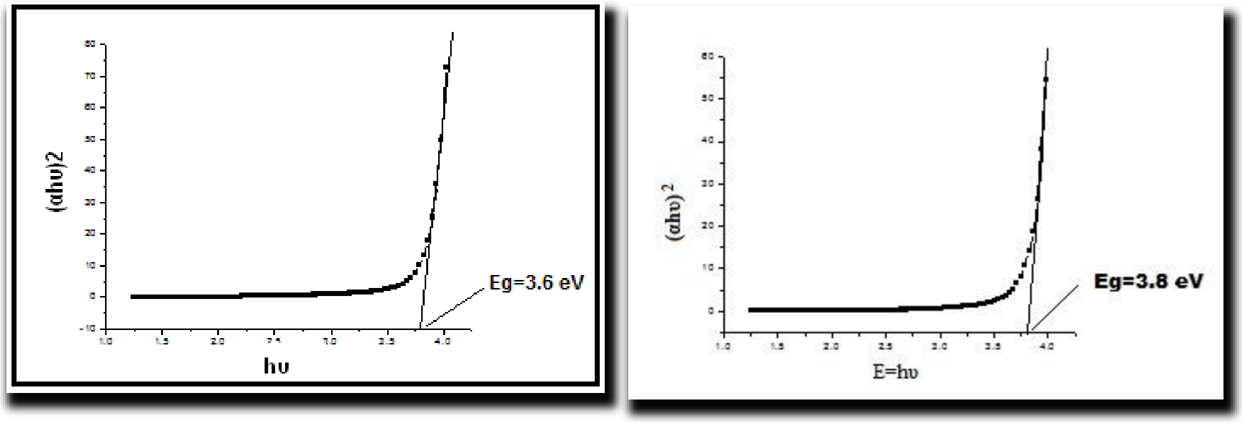

Fig.6 &7 typical plot of absorbance coefficient Vs. Photon energy for SnO_2 & SnO_2 : F thin films.

3.3 Electrical properties

Fig .8 shows the typical plot of voltage vs current for SnO_2 & SnO_2: F thin film at room temperature. From the analysis of the I-V curves, it is observed that SnO_2 resistivity is 0.07 x 10^{-3} Ω-cm and for SnO_2: F decrease with increase of doping volume and after increases doping volume increase the resistivity. Values are 0.06 x 10^{-3}, 0.024 x 10^{-3} and 0.024 x 10^{-3} Ω-cm for films deposited with doped Fluorine volume is 2ml, 4ml and 6ml solution is respectively.

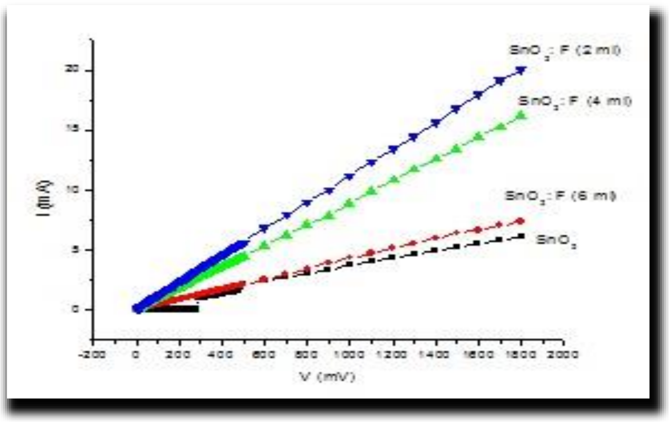

Fig. 8 I-V characteristics for SnO_2 & SnO_2: F thin films.

4. Conclusions

Transparent conducting SnO_2 and SnO_2: F thin films grown successfully by spray pyrolysis method. The structural investigation revealed that the both films are crystalline in nature with corresponding to (110), (101), (200) and (211) directions of the rutile structured SnO_2.as a preferred orientation with a tetragonal crystal structure. As deposited films was uniform and strongly adherent to substrate. The average transmittance of the deposited SnO_2 and SnO_2: F thin films were about 71.2 % and 73.3 % and optical band gap 3.60 eV & 3.8 eV respectively. The electrical studies conclude that resistivity decrease with increase fluorine doped and after increases doping volume increase the resistivity, Values are 0.06×10^{-3}, 0.024×10^{-3} and 0.024×10^{-3} Ω-cm. These films are found to be highly conducting.

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