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Effect of Substrate Temperature on Fluorine Doped Tin Oxide Thin films (FTO) by Chemical Spray Pyrolysis Method

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Abstract. Fluorine doped tin oxide (FTO) thin films were prepared by chemical spray pyrolysis method on glass substrates for different substrate temperatures and their structural, optical and electrical studies were investigated. X-ray diffraction (XRD) study showed polycrystalline nature of the films with mixed phase of both tetragonal and orthorhombic structures of tin oxide. The grain size (D) of the thin films calculated using Scherrer's formula and W-H plot are in the range 13 nm to 21 nm. The FESEM images revealed that the films have smooth and homogeneous surface morphology with thin granular grains distributed throughout the surface. The sample synthesized at 420^0C has maximum values for transmission percentage in the visible region, conductivity and mobility of 95%, 631.6 Siemens /cm and 4.54 $cm^2V^{-1}s^{-}1$ respectively.

INTRODUCTION

Flourine doped tin oxide (FTO) is a good candidate for applications requiring TCO because of its ability to adhere strongly to glass, chemical stability, resistance to physical abrasion, high optical transparency in the visible region and electrical conductivity. High-quality FTO could be prepared via physical vapor deposition (PVD), [1] chemical vapor deposition (CVD), [2] electron beam evaporation, [3] magnetron sputtering, [4] pulsed laser deposition, [5] chemical spray pyrolysis etc. [6]

In the present work, FTO thin films have been prepared by spray pyrolysis technique at substrate temperatures in the range 360- 420 °C using stannous chloride ($SnCl_2.2H_2O$) and ammonium fluoride (NH_4F) as precursors. The aim of this work is to study the effect of substrate temperature on the crystal growth orientation and some physical properties of FTO thin films deposited by spray pyrolysis technique, such as the structural, optical and electrical properties.

EXPERIMENTAL

The FTO thin films were deposited on ultrasonically cleaned glass substrate. Required amount of $SnCl_2.2H_2O$ to make 0.2 molar solution was initially dissolved in 5 ml of concentrated hydrochloric acid (HCl) followed by heating at 90 °C for 10 minutes. This mixture was then diluted by adding distilled water to make 50 ml solution. For fluorine doping, 10wt.% ammonium fluoride (NH_4F) (99% purity, Merck) dissolved in doubly distilled water (50 ml) was added to the starting solution. The spray solutions were magnetically stirred for 1 h before spraying on the substrate. Samples were prepared for substrate temperatures 360 °C, 380 °C, 400 °C and 420 °C. Samples were also synthesized on quartz substrate for optical analysis.

The structural characterization of the FTO thin films were carried out using X-ray diffraction analysis (XRD) on a Rigaku D-Max Geigerflex X-ray diffractometer using CuKα radiation source (λ=1.5418 Å) for 2θ values between 20° and 70° at room temperature. The optical characterization of the FTO thin films were studied using Shimadzu

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UV-Vis spectrophotometer model-UV 1800. The morphology of the prepared films was studied by field emission scanning electron microscopy (FESEM) using JEOL JSM 7600F microscope. Hall measurements were done using ECOPIA HMS-5000 in Vander Pauw configuration.

RESULTS AND DISCUSSIONS

To determine the crystal structure of the FTO thin films, X-ray diffraction technique was employed. The XRD pattern of FTO thin films is shown in Fig.1(a). All samples showed preferential orientation in (112) plane which correspond to orthorhombic phase of SnO_2 according to JCPDS file no: 78-1063. There are other reflection planes corresponding to orthorhombic and tetragonal phases (JCPDS file no: 41-1445) of SnO_2. The average grain size of sprayed FTO thin films for different substrate temperatures was calculated using the Scherrer's formula. [7] Figure1. (b) shows W-H plot of FTO thin films for different substrate temperatures. It was observed that the average grain size increased with increasing substrate temperature, and it is in the range of 15-21nm.

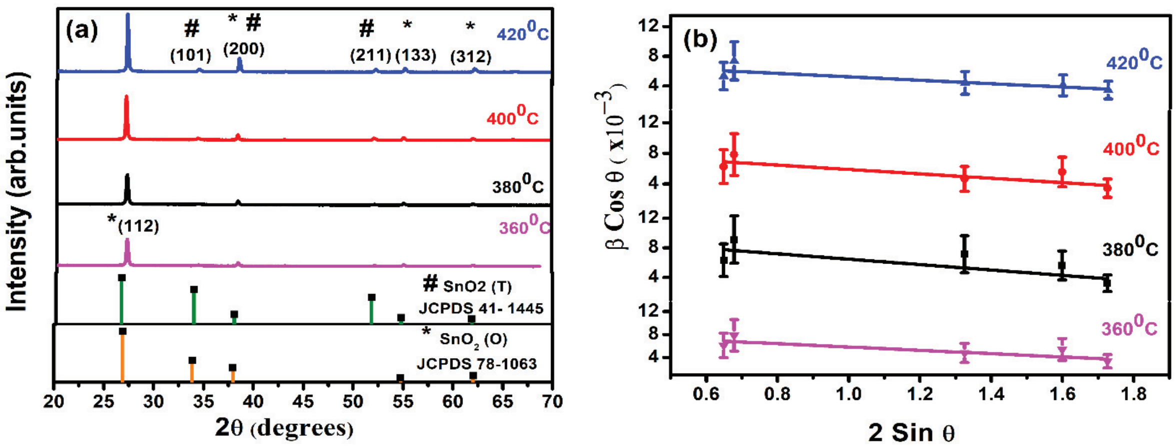

FIGURE 1. (a) XRD pattern, (b) W-H plot of FTO thin films for different substrate temperatures. The "*" sign denotes the orthorhombic SnO_2 phase and "#" sign denotes tetragonal phase of SnO_2

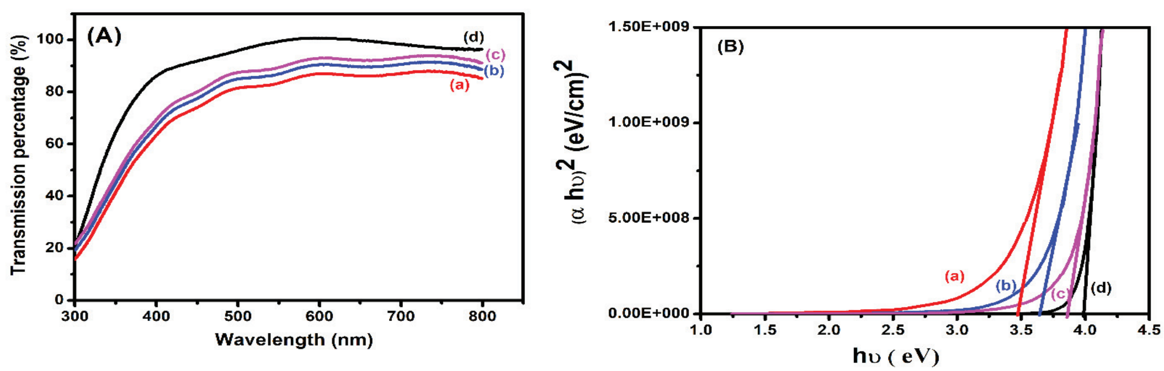

FIGURE 2. (A)Transmission spectrum and (B) Tauc Plot of FTO thin films on quartz substrate for (a) 360°C, (b) 380°C, (c) 400°C and (d) 420°C

The variation of transmission percentage with respect to wavelength of FTO thin films for different fluorine doping levels is shown in Fig.2(A). It was observed that the transmission percentage increases with increasing substrate temperature. The transmission percentage is in the range of 82-95%. Here, Tauc plot is used for the band gap determination using the relation proposed by Tauc, Davis and Mott [7] shown in Fig.2 (B). The band gap of FTO thin films for 360°C, 380°C, 400°C and 420°C are found to be 3.47 eV, 3.75 eV, 3.87 eV, and 4.0 eV, respectively. The band gap is increases with substrate temperature. The sample synthesized at 420 °C have maximum transmission percentage of 95% along with wide band gap.

The FESEM images of the FTO thin films for different substrate temperatures are shown in Fig.3, which disclose that the films have even and consistent surface morphology with thin granular grains distributed throughout the surface.

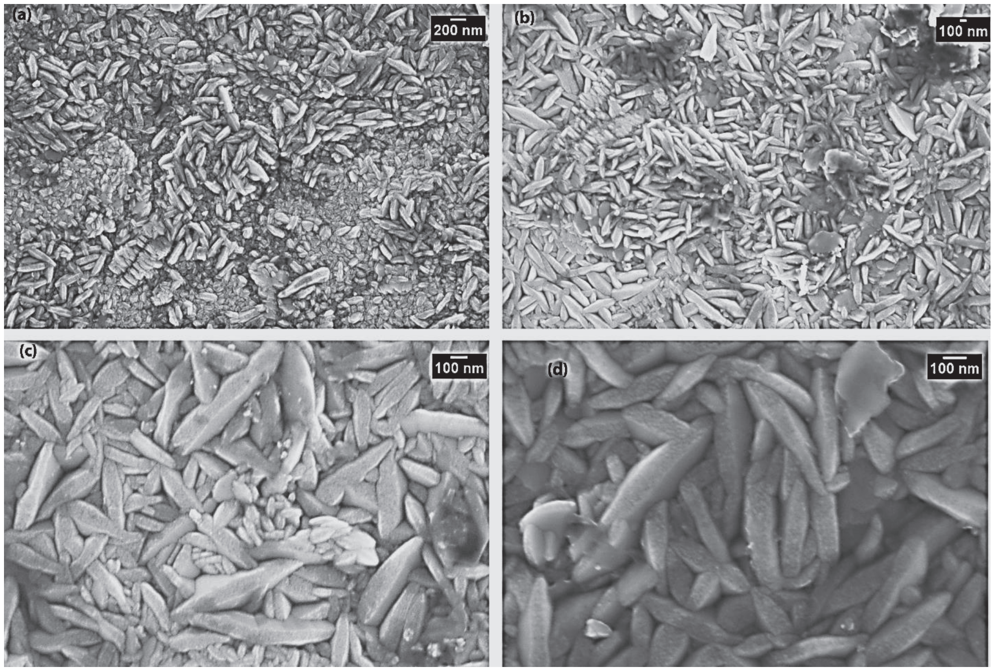

FIGURE 3. FESEM images of FTO thin films for (a) 360°C, (b) 380°C, (c) 400°C and (d) 420°C substrate temperatures

The FTO films exhibited n-type conduction with the mobility, hole concentration, and the resistivity values as shown in Table.1. It was observed that the carrier concentration, mobility and conductivity increases with increase in substrate temperature.

TABLE 1. Electrical studies of FTO thin films for different substrate temperatures

Substrate temperature (°C)	Carrier Concentration (/cm^3)	Resistivity (Ω.cm)	Conductivity (S/cm)	Mobility (cm^2/V.s)	Avg.Hall coefficient (cm^3/C)	Conductivity Type
Undoped SnO_2 film	2.989×10^{19}	4.03×10^{-2}	24.86	5.2	-8.159×10^{-1}	n
360	1.16×10^{20}	2.62×10^{-2}	38.08	1.97	-5.19×10^{-2}	n
380	1.20×10^{20}	1.91×10^{-2}	52.32	2.80	-5.3×10^{-2}	n
400	2.69×10^{20}	5.01×10^{-3}	170.2	4.54	-2.31×10^{-2}	n
420	8.67×10^{20}	1.58×10^{-3}	631.6	4.61	-7.19×10^{-3}	n

CONCLUSIONS

Effect of substrate temperature on structural, optical and electrical properties of FTO thin films was studied in this work. The sample deposited at 420°C exhibited maximum conductivity of 631.6 Siemens/cm and maximum transmission of 95% in the visible region. The conductivity of the sample can be improved by varying the percentage doping of fluorine for application in optoelectronic devices.

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