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SPUTTERING POWER DEPENDENCE PHYSICAL PROPERTIES OF NANOCRYSTALLINE DC MAGNETRON SPUTTERED SnO_2 THIN FILMS

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ABSTRACT

Nanocrystalline tin oxide (SnO_2) thin films were deposited on glass substrates by dc reactive magnetron sputtering technique under various sputtering powers from 20 W to 50 W. X-ray diffraction results showed that the films are polycrystalline nature with tetragonal structure of SnO_2. The predominant plane was changed from (110) to (101) with increasing the sputtering power. The films deposited at sputtering power of 40 W exhibited smooth and dense surface. From the photoluminescence spectra, the intensity of the emission peak was increased with increasing the sputtering power. The optical transmittance of the films increased from 72% to 93 % with increasing the sputtering power. The lowest electrical resistivity of 0.007 Ωcm was observed at the sputtering power of 50 W.

Keywords: Tin oxide, Nanocrystalline, Thin films, Sputtering, Sputtering power

1. Introduction

Tin dioxide (SnO_2) is an important compound semiconducting material due to its interesting properties such as low electrical resistivity, highly transparent, high chemical and mechanical stability and wide-band-gap [1-3]. Due to these properties, it finds applications in catalysis, displays, sensors [4-5], transparent conductor [6], low-emission glass and thermal insulation [7], light emitting devices [8] and solar cells [9]. SnO_2 is the most widely used metal oxide for gas sensors, and the sensitivity increases drastically with decreasing grain size from 20 to 5 nm [10]. SnO_2 have been fabricated by variety of approaches such as ultrasonic spray pyrolysis [7], sol-gel [1], spin-coating [11], reactive thermal evaporation [12], sputtering [13-15], plasma enhanced atomic layer deposition [16], and pulsed laser evaporation [5]. Compare to these techniques DC reactive magnetron sputtering is one of the best technique, due to it high deposition rates, easy control over the composition of the deposited films and maintained in homogenous manner over large area of substrates. In this present work we report the effect of the sputtering power on the structural, compositional, morphological, optical and electrical properties of SnO_2 films prepared by DC reactive magnetron sputtering technique.

2. Experimental

SnO_2 thin films were deposited on glass substrate by dc magnetron sputtering from home made circular planar magnetron sputtering system. The sputter chamber was pumped with diffusion pump and rotary pump combination. The pressure in the sputter chamber was measured using digital pirani and penning gauge combination. The magnetron target assembly was mounted at the top of the sputter chamber such that the sputtering could be done by down configuration. Both flow rates of sputter (argon) and reactive gases (oxygen) were controlled individual by Tylan mass flow controllers. Before deposition of each film, the target was sputtered in pure argon atmosphere for 10min to remove oxide layers if any on the surface of the target. The sputtering condition maintained during the growth of SnO_2 films are mentioned in Table 1.

Characterisation

The chemical composition of films determine by electron probe microanalysis (EPMA). The crystallographic structure of the films was analysed with X -ray diffraction (XRD). The surface morphology and microstructure of the films were analysed with atomic force microscope (AFM) and scanning electron microscope (SEM), respectively. Photoluminiscence (PL) spectra were measured with a LS55 fluorescence spectrometer at

room temperature. The transmittance of the films was monitored using the Hitachi U-3400 UV-Vis-NIR double beam spectrometer in the wavelength range of 300-1000 nm. The resistivity of the films was measured using vander Pauw method.

Table 1. Deposition Parameters Maintained During the Preparation of SnO_2 Films by DC Reactive Magnetron Sputtering

Parameter	Value
Sputtering target	: Sn (99.9%)
Target to substrate distance	: 60 mm
Ultimate pressure	: $2x10^{-6}$ mbar
Oxygen partial pressure (pO_2)	: $7x10^{-4}$ mbar
Sputtering pressure (Sp)	: $3x10^{-2}$ mbar
Substrate temperature (Ts)	: 473 K
Sputtering power (Wp)	: 20-50 W

3. Results & Discussion

Compositional and Structural Properties

The chemical composition of as deposited films at different sputtering powers was listed in Table 2. EPMA results reveal that the films exhibited nearly stoichiometery (Sn/O ratio=0.52) at the films deposited at sputtering power of 40 W. In remaining sputtering powers, the stiochiometry of the films was slightly deviated.

Table 2. The Chemical Composition of as Deposited Films at Different Sputtering Powers.

Sputtering power	Atomic percentage		
	Sn	O	Sn/O
20 W	31.8	68.5	0.46
40 W	34.1	65.9	0.52

Figure1. shows the X-ray diffraction pattern of SnO_2 films deposited at different sputtering powers. The as deposited films exhibits polycrystalline nature and represent tetragonal structure of SnO_2. The films deposited at low sputtering power, (110) plane was predominant peak and two additional planes (101) and (211) was appeared with low intensity. At optimum sputtering power of 40 W, the peak intensity and polycrystallinity nature of the films increased greatly. This was due to increasing of the grain size and decreasing the structural defects. Beyond this optimum sputtering power, the predominant plane was changed from (110) to (101) and intensity of the peaks was decreased. The changing of predominant plane and crystallinity of the films was related to the energy of the sputtered

particles reaching to the substrate surface [17]. From this observation, the sputtering power must be sufficiently low to obtain the better crystalline films and the (101) plane was predominantly growth at high sputtering powers.

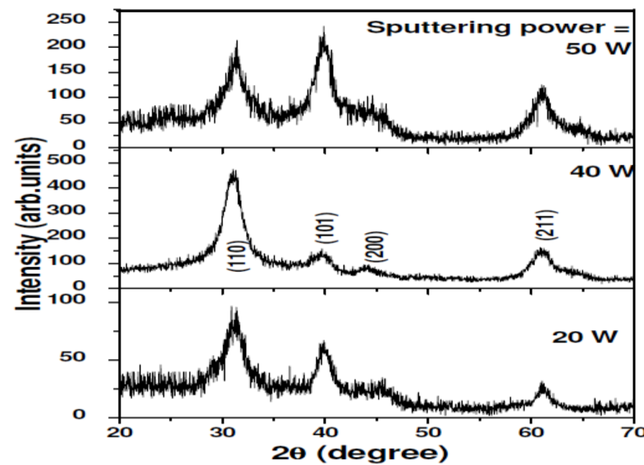


Figure1. XRD Pattern of SnO_2 at Different Sputtering Powers.

The average grain size of films was calculated by using Scherre's relation [18]. Grain size of the films was increased from 4.6 nm to 6.1 nm with increase of sputtering power from 20 W to 40 W and thereafter it decreased to 5 nm at higher sputtering power of 50 W. The lattice parameter a and c are calculated from (110) and (211) peaks, and are listed in Table 3.The lattice parameter a increases from 4.715 nm to 4.732 nm where as c decreases from 3.184 nm to 3.169 nm with the increase of sputtering power from 20 W to 40 W. The variation in the lattice parameter with increase of sputtering power was due to stress developed in the films.

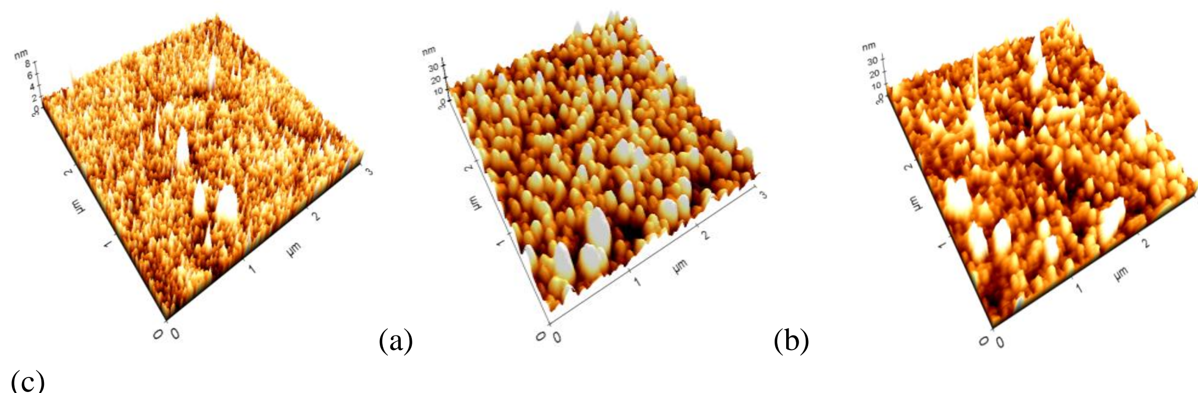
Table 3. Lattice Parameters Values of SnO_2 Films at Different Sputtering Powers.

Sample	Sputtering Power (W)	Lattice Parameters (Å)	
		a	*c*
SnO_2	20	4.715	3.184
	40	4.732	3.169
	50	4.718	3.173
SnO_2 (ICDD)	-	4.737	3.186

Surface Morphology and Microstructure

AFM images of SnO_2 films at different sputtering powers are shown Figure 2. At low sputtering power of 20 W the film exhibits rough surface (RMS roughness=2.9 nm) with small grains, this was due to poor crystallinity. As increasing the sputtering power to 40 W, the films represent uniform morphology, and grains become bigger and spherical in shape

with smooth surface (RMS roughness=1.3 nm). Further increasing the sputtering power, shape of the grains was changed and more clusters are appeared on the film surface and the RMS roughness increased to 2.2 nm. At higher sputtering powers the deposition rate is high and its effects on the nucleation and growth rate of the films, as a result films exhibits the different size of grains and clusters.



(c)

Figure 2. AFM Images of SnO_2 Films Deposited at Different Sputtering Powers: (a) 20 W, (b) 40 W and (c) 50 W.

Figure 3. shows the SEM images of SnO_2 films at different sputtering powers. The films exhibited fine grains with dense surface at low sputtering power of 20 W and the grain size increased greatly at sputtering power of 40 W. The size of the grains was decreased and non-uniform when the films deposited at sputtering power of 50 W.

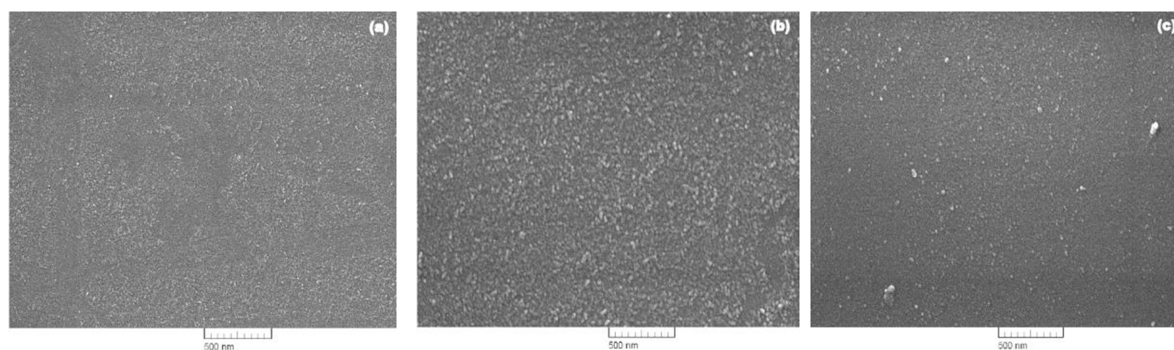


Figure 3. SEM Images of SnO_2 Films Deposited at Various Sputtering Powers: (a) 20 W, (b) 40 W and (c) 50 W.

Photoluminescence Properties

The PL spectra of SnO_2 films deposited at various sputtering powers are shown in Figure 4. The SnO_2 film deposited at sputtering power of 20 W, the films shows the emission peak at 534 nm with low intensity, it was due to structural defects and small crystallite size [19]. As increasing the sputtering power to 40 W the emission peak intensity was increased and a shoulder peak appears at 608 nm.

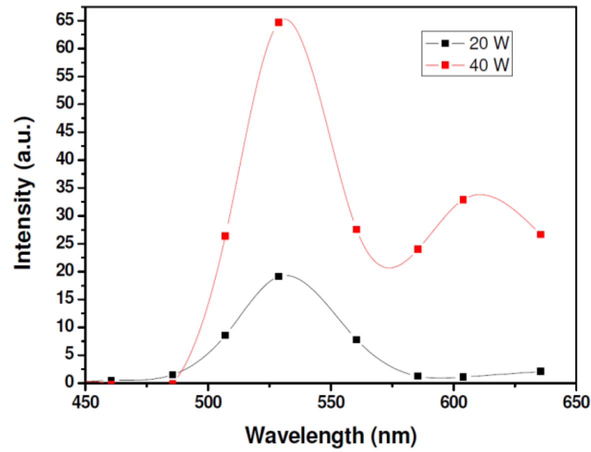


Figure 4. PL Spectra of SnO_2 at Different Sputtering Powers.

Optical and Electrical Properties

The optical transmittance spectra of SnO_2 films deposited under various sputtering powers was shown in Figure 5. The transmittance of the films was strongly influenced by the sputtering power. The films showed low optical transmittance of 72% at sputtering power of 20 W and it was sharply increases to 93% at the sputtering power of 40 W. Beyond this sputtering power the transmittance of the films decreased to 80%. The high transmittance of the films was due to bigger grain size and smooth surface of the films.

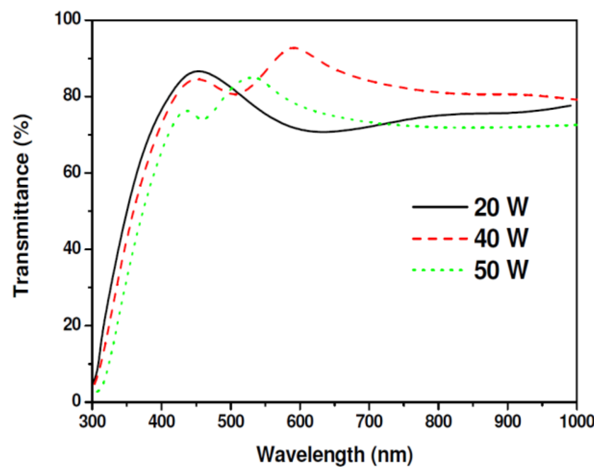


Figure 5. Optical Transmittance Spectra of SnO_2 Films at Different Sputtering Powers

The optical band gap (E_g) of the films was evaluated from the extrapolation of the linear portion of the plots of $(\alpha h\nu)^2$ versus $(h\nu)$ (α is the absorption coefficient, $h\nu$ is the photon energy). The absorption edge of the films shifted towards higher wavelength side and the optical band gap of the films decreased from 3.74 eV to 3.54 eV with increasing of sputtering power from 20 W to 50 W.

The obtained optical and electrical values of SnO_2 films at different sputtering powers were listed in Table 4. The electrical properties of the films were highly influenced by the microstructure and composition of the films. The films exhibited high electrical resistivity of 7 Ωcm at low sputtering power of 20 W and it was decreased to 0.007 Ωcm with increase of sputtering power to 50 W. The high electrical resistivity at low sputtering powers was due to poor crystallinity of the films. The low electrical resistivity of the films at higher sputtering powers was may be due to Sn rich in the deposited films.

Table 4. Optical and Electrical Values of SnO_2 Films at Different Sputtering Powers.

Sample	Resistivity (Ωcm)	Transmittance (%) at λ=590 nm	Band gap (eV)
20 W	7	72	3.74
40 W	0.01	93	3.62
50 W	0.007	80	3.54

4. Conclusions

Nanocrystalline SnO_2 films were deposited on glass substrate under various sputtering powers using DC reactive magnetron sputtering technique. The deposited films were nearly stoichiometry and XRD results show that the films were in polycrystalline nature. The grain size of the films was increased from 4.6 nm to 6.1 nm with increase of sputtering power from 20 W to 40 W. From microsturcture and surface morphology images, the shape and sized of the grains was changed with increasing of the sputtering power. The films showed high optical transmittance of 93% and band gap of 3.62 eV with low electrical resistivity of 0.01 Ωcm at sputtering power of 40 W.

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