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Nanocrystalline Au: SnO_2 Thin Films Grown by DC Reactive Magnetron Sputtering

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ABSTRACT
In this work, nanocrystalline gold doped tin oxide (Au:SnO_2) thin films were prepared on glass substrates by dc reactive magnetron sputtering at different substrate temperatures. The physical properties of as deposited films were characterized by different analytical techniques. From the XRD results, the polycrystallinity and grain size of the films increased with substrate temperature. The as deposited films exhibited smooth surface without cracks and voids. The films deposited at substrate temperature of 473 K exhibited highest optical transmittance of 88% with band gap of 3.30 eV.

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I. INTRODUCTION

Tin oxide (SnO_2) thin films have been extensively used in solar cells, flat panel displays, catalysis, batteries and gas sensors [1-3], due its excellent properties such as high optical transparency, high chemical and thermally stable, direct band gap (3.6 eV at 300 K), mechanically harder than ZnO, low cost, and non-toxic [4-6]. Recently, nanostructured SnO_2 and metal doped SnO_2 thin films have attracted extensive interest for development of gas sensors. The gas sensitivity of SnO_2 films mainly depends on electrical properties and large surface area of nanostructures [7]. Pure and doped SnO_2 thin films have been prepared by various deposition techniques such as sol-gel method [8], spray pyrolysis [9], pulsed laser deposition [10] electron beam evaporation [11] and sputtering [12-13]. Among these techniques dc magnetron sputtering is one of the best techniques due to its high deposition rates, good adhesion to the substrate, maintained in uniform manner over large scale of substrates and easy to control the chemical composition of the film. In this study, gold doped SnO_2 thin films were deposited by dc magnetron sputtering technique and investigated the substrate temperature influenced compositional, structural, microstructural, morphological, optical and electrical properties.

II. EXPERIMENTAL

Nanocrystalline gold doped tin oxide (Au:SnO_2) thin films were deposited on glass substrate by dc reactive magnetron sputtering from home made circular planar magnetron sputtering system. The sputter chamber was pumped with the combination of diffusion pump and rotary pump. The pressure in the sputter chamber was measured using digital pirani and penning gauge combination. A circular planar magnetron was used as the magnetron cathode. 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 Au:SnO_2 films are mentioned in Table 1. The chemical composition of films was analyzed by electron probe microanalysis (EPMA). The crystallographic structure of the films was characterized with X-ray diffraction (XRD). The microstructure and surface morphology of the films were analyzed with scanning electron microscope (SEM) and atomic force microscope (AFM). The quality of the films was determined by LS55 fluorescence spectrometer (perkin elmer) at room temperature. The transmittance of the films were monitored using the Hitachi U-3400 UV-Vis-NIR double beam spectrometer in the wavelength range of

300-1000nm. The resistivity of the films was measured by four-point probe method.

Table 1 Deposition conditions during the preparation of Au:SnO_2 films

Deposition method	: DC reactive magnetron sputtering
Power source	: DC power supply (1000 V and 1 Amp)
Sputtering target (Mosaic)	: Pure tin (99.9%), gold strips (99%)
Substrates	: Glass
Target to substrate distance	: 60 mm
Ultimate pressure	: $5x10^{-6}$ mbar
Oxygen partial pressure	: $7x10^{-4}$ mbar
Substrate temperature	: 303 K to 573 K
Sputtering power	: 50 W
Sputtering pressure	: $2x10^{-2}$ mbar
Deposition time	: 9-17 min
Substrate rotation	: 20 rpm

III. RESULTS AND DISCUSSION

3.1 Compositional result

The chemical composition of the Au:SnO_2 films was listed in Table 2. The Sn/O ratio was close to the stoichiometric compound of SnO_2 and the gold incorporation is close to 1 at.% in the films. The oxygen content in the films was slightly decreased with increasing of the substrate temperature from 303 K to 473 K.

Table 2 Chemical composition of Au-SnO_2 films at different substrate temperatures

Substrate temperature (K)	Atomic percentage		
	Sn	O	Au
303	32.6	66.5	0.9
473	34.2	64.9	0.9

3.2 Structural properties

Figure1 shows the XRD patterns of Au:SnO_2 films at different substrate temperatures. The films deposited at 303 K were amorphous and the crystallinity of the films started at 373 K and the (101) phase was predominate. As the increasing the substrate temperature the peak intensity and polycrystallinity of the films increased, due to reduction of the microstructural defects and improvement in the structural homogenity of the films. From the XRD patterns, the Au:SnO_2 films exhibited the SnO_2 phase only and no diffraction peak of metallic Sn or Au phase or oxide phase of Au were observed. The similar result was observed in dc magnetron sputtered Au:Ag:SnO_2 thin films [14], and sol-gel prepared noble metal doped nanocrystal SnO_2 films [15]. The integration of the noble metal within the SnO_2 lattice could be very probable since sputtering is a known deposition technique permitting a homogenous distribution of noble metal atoms in the SnO_2 matrix.

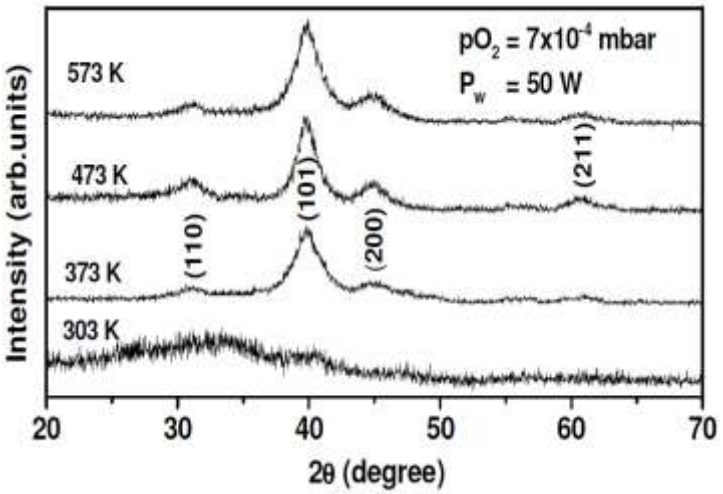
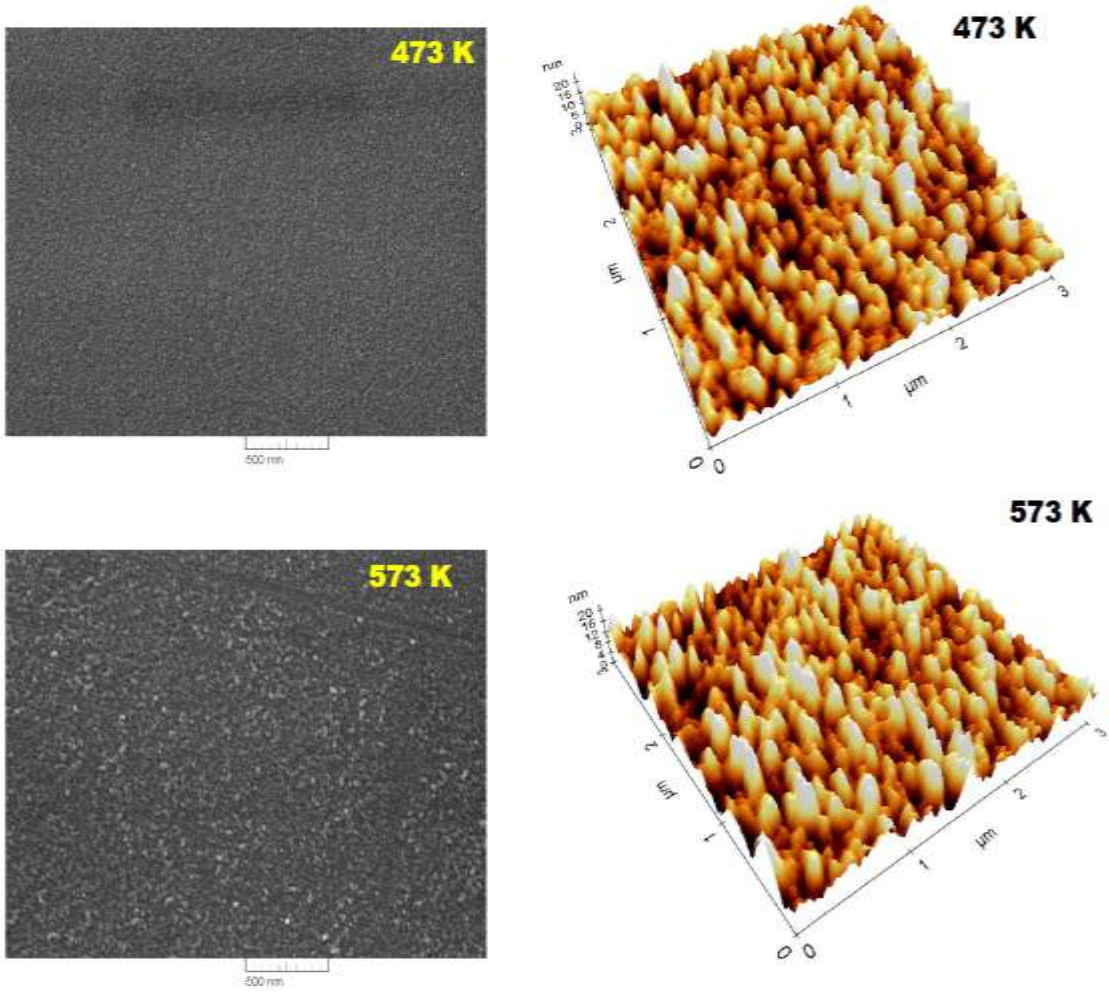


Fig.1 XRD patterns of Au:SnO_2 films at different substrate temperatures.

The average crystallite size of films was calculated by using Scherrer's equation [16]. The obtained crystallite size values are 4.6 nm, 6.9 nm and 8.7 nm for substrate temperature of 373 K, 473 K, and 573 K respectively.

3.3 Microstructure and Surface morphology

The SEM images of Au:SnO_2 films at different substrate temperatures are shown in Fig.2(a). The films exhibit smooth surfaces with a fine microstructure without cracks, voids and aggregation of the particles. It observed from the SEM images that the grains became larger and denser with increasing of the substrate temperature. These results are consistent with the changes observed by XRD.



(a) (b)

Fig.2 (a) SEM and (b) AFM images of Au:SnO_2 films at different substrate temperatures.

From the AFM images (Fig.2(b)) the grains become bigger with increasing the substrate temperature. The surface roughness was 3.2 nm, 2.1 nm and 1.6 nm for 303 K, 473 K and 573 K deposited films respectively. Reddy et al. [17] observed that the RMS roughness of the DC reactive magnetron sputtered Au:SnO_2 films decreased from 3.1 nm to 2.8 nm with increase in the annealing temperature from 303 K to 723 K, due to enhancement of migration of atoms at the surface.

3.4 Photoluminescence properties

Fig.3. shows the room temperature PL spectra of the Au:SnO_2 films. The films deposited at 303 K exhibited a broad emission peak at 531 nm (2.33 eV). As increasing the substrate temperature to 473 K the intensity of the emission peak increases and an additional shoulder peak appeared at 605 nm (2.04 eV). At higher substrate temperature of 573 K, intensity of main peak (531 nm) increases and another shoulder peak appears at 625 nm (1.98 eV) along with 605 nm peak. The improvement of the peaks intensity with substrate temperature can be attributed to the decreasing of the structural defects and increasing the crystallite size

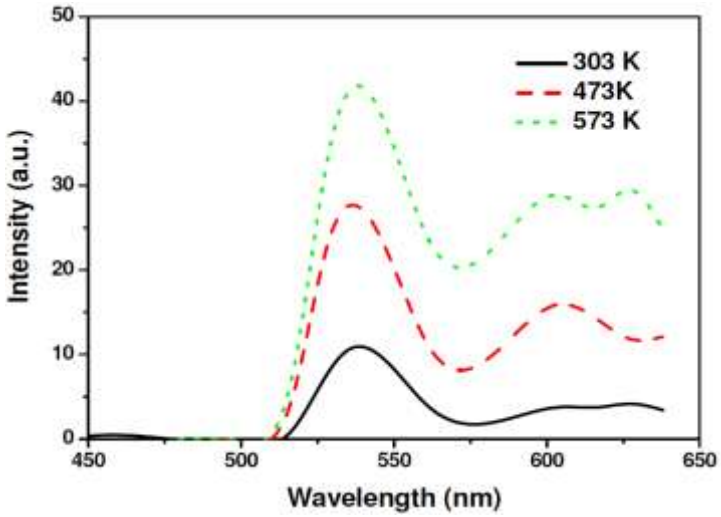


Fig. 3 Photoluminescence spectra of the Au:SnO_2 films at different substrate temperatures.

3. 5 Optical and Electrical Properties

Fig.4. shows the optical transmittance spectra of Au:SnO_2 films at different substrate temperatures. The optical transmittance of the films was strongly influenced by the substrate temperature. The optical transmittance of the films increases from 79 % to 88 % with increasing substrate temperature and then decreases to 80 % at higher substrate temperature of 573 K. The enhancement of transmittance in the visible region with substrate temperature may be due to decrease in optical scattering caused by the densification of grains followed by the grain growth and the reduction of the grain boundary density [18]. However, Reddy et al. [19] observed the decreasing of the optical transmittance with increasing of annealing temperature in pulsed magnetron sputtered Ag: SnO_2 films.

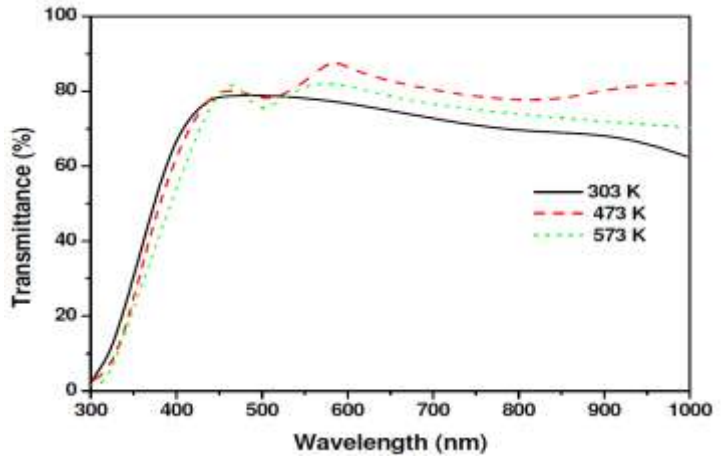


Fig. 4 Optical transmittance spectra of Au:SnO_2 films at different substrate temperatures.

The absorption edge of the Au:SnO_2 films was shifted to longer wavelengths with increase of substrate temperature, as a results, the band gap of the films was decreased. 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 obtained E_g values of Au:SnO_2 films are listed in Table 3. The decreasing of the band gap with increasing of substrate temperature was may be due to quantum confinement.

It was found out that the electrical resistivity of the films decreased significantly with substrate temperature (Table 3). The films deposited at lower substrate temperature of 303 K exhibited higher value of resistivity and it sharply decreased with increase of the substrate temperature. This was due to the increase of grain size and reduction of scattering of carriers at the grain boundaries with substrate temperature. Additionally, the Au depletion from the SnO_2 lattice and subsequent clusterization at the grain boundaries should also contribute for decrease in the resistivity of the film.

Table 3 Electrical and optical properties of Au:SnO_2 films at different substrate temperatures

Substrate temperature (K)	Resistivity (Ωcm)	Transmittance (%) at λ=590 nm	Band gap (eV)
303	1.8	79	3.45
373	0.1	81	3.39
473	0.005	88	3.30
573	0.001	80	3.26

IV. CONCLUSIONS

Nanocrystalline Au:SnO_2 thin films were prepared on glass substrates by dc reactive magnetron sputtering at different substrate temperatures. The crystallinity of films increased with increasing of the substrate temperature. The average crystallite size of films was around 8.7 nm with rms roughness of 1.6 nm at the substrate temperature of 573 K. The intensity of the emission peak increases, and an additional shoulder peak appeared at 605 nm (2.04 eV) with increasing the substrate temperature increases to 473 K. The electrical resistivity of films sharply decreased from 1.8 Ωcm to 0.001 Ωcm with increasing the substrate temperature from 303 K to 573 K.

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