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OluwaseunAdedokun

Department of Pure and Applied Physics, Ladoke Akintola University of Technology, Ogbomoso, Nigeria. \\ Nanotechnology Research Group (NANO+), Ladoke Akintola University of Technology, Ogbomoso, Nigeria., oadedokun@lautech.edu.ng

Victoria Mofoluke Odebunmi

Department of Pure and Applied Physics, Ladoke Akintola University of Technology, Ogbomoso, Nigeria. \\ Department of Physics, Emmanuel Alayande College of Education, Oyo, Nigeria., oadedokun@lautech.edu.ng

Yekinni Kolawole Sanusi

Department of Pure and Applied Physics, Ladoke Akintola University of Technology, Ogbomoso, Nigeria. \\ Nanotechnology Research Group (NANO+), Ladoke Akintola University of Technology, Ogbomoso, Nigeria., oadedokun@lautech.edu.ng

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Effect of Annealing Temperature on Structural, Optical and Electrical Properties of Spin Coated Tin Oxide Thin Films for Solar Cells Application

Oluwaseun Adedokun^{1,3*}, Victoria Mofoluke Odebumi^{1,2} and Yekinni Kolawole Sanusi^{1,3}

¹ Department of Pure and Applied Physics, Ladoke Akintola University of Technology, Ogbomosho, Nigeria.

² Department of Physics, Emmanuel Alayande College of Education, Oyo, Nigeria.

³ Nanotechnology Research Group (NANO⁺), Ladoke Akintola University of Technology, Ogbomosho, Nigeria.

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Abstract: Transparent Conducting Oxide (TCO) thin films are materials of significance for their applications in optoelectronics and sun powered cells. This paper reports the impact of annealing temperature on structural, optical and electrical properties of tin oxide (SnO₂) thin films for solar cells application. The sol-gel was prepared from anhydrous stannous chloride, SnCl₂ as precursor, 2-methoxyethanol as a solvent and di-ethanolamine as stabilizer. The thin film was deposited on glass substrates by means of spin coater at the rate of 2000rpm for 40 seconds. After pre-heated at 200°C, the samples were annealed at 300, 400, 500 and 600°C for 2 hours respectively. The structural, optical and electrical characteristics of prepared films were characterized using X-ray diffraction (XRD) analysis, UV-visible spectroscopy and electrical measurement. X-ray diffraction pattern shows polycrystalline tetragonal-cassiterite structure with most extraordinary pinnacle having a grain size of 17.01nm. Annealing temperature increases the grain size. There is increment in the absorption of photon energy of the film as the annealing temperature decreases with increasing wavelength and the transmittance was basically increases with increasing annealing temperature as the wavelength increases. The energy band gaps were in the range of 4.116 to 4.106 eV. The sheet resistance was observed to decrease as the annealing temperature increases. In view of these outcomes, SnO₂ thin films prepared could have useful application in transparent conducting oxide electrode in solar cell.

Keywords: Tin oxide; Annealing temperature; Sol-gel spin coating; Grain size; Energy band gap; Sheet resistance.

1 Introduction

The most important electrical conductive material is the transparent conducting oxides (TCOs). These oxides have been analysed and discovered to absorb low quanta to light. TCOs have some distinct properties which outline their uniqueness; these include their method of deposition, conditions of preparation and the availability of semiconductors having band gap of approximately ≥ 3.1 eV [1]. Transparent conducting oxides are of group of materials with visible transparency and high electrical conductivity. These properties make TCOs to be applicable in opto-electric devices such as electrochromic

and solar cells devices [2,3]. Examples of TCOs are indium tin oxide (ITO), cadmium oxide (CdO), tin oxide (SnO₂), indium oxide (IO), aluminium doped zinc oxide (AZO), undoped zinc oxide (ZO) and fluorine doped tin oxide (FTO) [4]. SnO₂ is an n-type semiconductor. SnO₂ have various applications which include: transparent conducting electrodes, liquid crystal displays, photovoltaic cells, thin-film heaters, and solid-state gas sensors. Several methods such as chemical vapour deposition (CVD) [7], Atmospheric Photo CVD [2], Spray Pyrolysis [4], sol-gel dip coating [5], and sol gel spin coating [6], have been successfully tried to produce device of quality films. SnO₂ thin films have been achieved by a variety of deposition techniques such as

*Corresponding author: oadedokun@lautech.edu.ng

chemical vapour deposition (CVD) [7], sol-gel processing [8], sputtering [9], and pulsed laser deposition (PLD) [10]. Among all these deposition techniques, sol-gel spin coating was used in this research. Sol-gel spin coating has been chosen to prepare SnO₂ thin films because it's numerous advantages over other techniques such as low cost, control of morphology and stoichiometry [6]. In this paper, SnO₂ thin films were prepared by sol-gel spin coating and the effect of annealing temperature on structural, optical and electrical properties of SnO₂ thin films were reported. Structural characteristics are verified using X-ray diffractometer and the effect of annealing temperature on SnO₂ thin film on some optical properties by UV-Vis spectroscopy and electrical properties were investigated by four point probe measurements, respectively.

2 Experimental Details

The glass substrates with dimension 25.4mm × 76.2mm were rinsed in de-ionized water. They were then cleaned in a breaker containing ethanol and the beaker is put on VWR ultrasonic cleaner at room temperature for 30 minutes. The cleaned glass slides were dried in a stream of nitrogen gas. All glass slides received the same cleaning process. Tin oxide precursor solutions were prepared from stannous chloride as metal source, 2-methoxyethanol as a solvent and di-ethanolamine as a stabilizer. The starting materials, stannous chloride and 2-methoxyethanol were ensured free of impurities. 0.2 M of stannous chloride was dissolved in 10mL of 2-methoxyethanol and was mixed for 30 minutes at 60°C until the mixture turns milky. Di-ethanolamine of 10mL was included little by little under steady mixing until milky solution was cleared. The resultant solution was stirred continuously for another 90 minutes until a colourless, homogeneous and transparent solution was obtained. The solution was kept in a closed container for 40 hours at 27°C for aging and also to obtain high thickness. The mixture prepared was deposited on a glass substrate using sol-gel spin coating method. Spin coating parameters were as follows: 2mL of solution per deposition with a speed of 2000 rpm for 40 seconds each by using LAURELL WS-400-6NPPSS spin coater. The films were pre heated in ambient atmosphere at 200°C for 10 minutes to evaporate the solvent. The SnO₂ thin films were annealed at 300, 400, 500 and 600°C for 2 hours using a furnace.

3 Results and Discussion

3.1 Structural Analysis of FTO Thin Films

SnO₂ thin films structure was investigated through X-ray diffraction (XRD) technique. It was observed that the XRD pattern of the as prepared thin film peaks are not clearly seen indicating that the film is almost amorphous.

As the annealing temperature increases, the XRD patterns show polycrystalline phase. X-ray diffraction pattern shows polycrystalline tetragonal structure with increase in annealing temperature and that grain size also increased. This result is in line with that of Kahattha et al. (2016) [6]. The more prominent diffraction peaks are (110), (101), (211) and (301), which exhibiting tetragonal-cassiterite structure reported for SnO₂ (JCDPS No. 21-1250) with a preferred orientation along (110) as shown in figure 1. The grain size (D) of the films was obtained from the XRD spectrum using Deybe Scherrer's formula [11].

$$D_{hkl} = \frac{k\lambda}{\beta \cos\theta} \quad (1)$$

Where k is a constant to be taken 0.9, β is Full Half Maximum (FWHM) in radian, λ is the wavelength of X-ray used and θ is Bragg's angle.

The grain size increases as the annealing temperature increases indicating that the crystallinity of the films was enhanced as shown in Table 1.

3.2 Optical Properties of FTO Thin Films

The optical transmittance spectra of undoped tin oxide thin films deposited on glass substrates are annealed at 300, 400, 500 and 600°C was exhibited in figure 2. The transmission values of the films were observed to be low in the infrared region and high visible region. This means that the films behaved as a translucent material at longer wavelengths. The result implies that the temperature of thermal annealing produce an optical transmittance enhancement due to the better crystallinity with the increasing of annealing temperature, leading to less light scattering loss in thin films [13].

Furthermore, the transmittance spectra show a sharp absorption edge in the wavelength around 350 nm, suggesting semiconductor behaviour with the existence of direct optical band gap [6]. It was observed that there is increase in absorption of photon energy as the annealing temperature decreases. For each samples, there is an absorption level that do not differ from one another, their sharp absorption edge is located at 350 nm. The optical conductivity spectra of undoped tin oxide tin films deposited on glass substrates and annealed at 300, 400, 500 and 600°C was shown in figure 3. It was observed that the annealing temperature increases the optical conductivity with the annealed film at 400°C having the value of $3.5 \times 10^{17} \text{ cm}^{-1}$ at 4.4eV which is the highest followed by the annealed film at 600°C having the value of $2.75 \times 10^{17} \text{ cm}^{-1}$ at 4.3 eV. From the graph of $(\alpha hv)^2$ against photon energy, the energy band gap, E_g of the prepared FTO thin films can be determined.

The optical band gap of SnO₂ thin film was approximated from the calculation of the linear relationship between $(\alpha hv)^2$ and hv according to the equation 2 [11,12].

$$\alpha hv = A(hv - E_g)^{1/2} \quad (2)$$

The energy band gap for as prepared, 300°C, 400°C, 500°C and 600°C of SnO₂ was determined to be 4.116eV, 4.121eV, 4.120eV, 4.117eV and 4.106eV, respectively. Annealing temperature reduced the energy band gap of SnO₂. This applies to the growth of grain size and the increase in defect states near the bands and this is in turn reduced the value of the energy gap [14].

3.3 Electrical Properties of SnO₂ Thin Films

The resistivity of the samples was measured by using four

Table 3 shows sheet resistance for as prepared tin oxide thin film at different annealing temperatures. The sheet resistance increase directly with increment in annealed temperature. There are two determinate of sheet resistance namely carrier concentration and mobility [16]. For optimum performance of the thin films, it must have low sheet resistance and high conductivity. The sheet resistance at annealed at 600°C show the minimum value of 1.602 Ωsq⁻¹ which may be due to plenty of conducted deposited particles on the glass substrate after an annealed process.

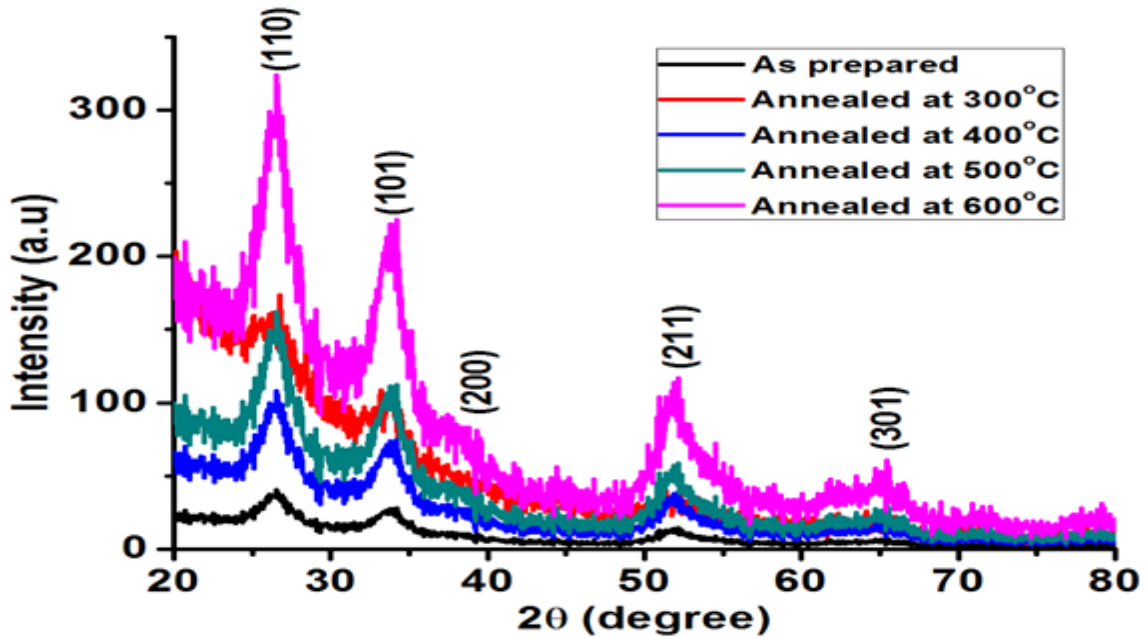


Fig. 1: XRD patterns of SnO₂ thin films annealed at different temperature

Table 1: Variation of average grain size of SnO₂ thin films with annealing temperature.

Annealing Temperature	Crystalline Size (nm)
As prepared	10.90
300 °C	12.83
400 °C	14.62
500 °C	16.15
600 °C	17.01

point probe set-up. The sheet resistance was calculated by using equation 3 [15].

$$R_s = \frac{\rho}{t} \tag{3}$$

Where ρ is the resistivity, R_s is the sheet resistance and t is the thickness of the films.

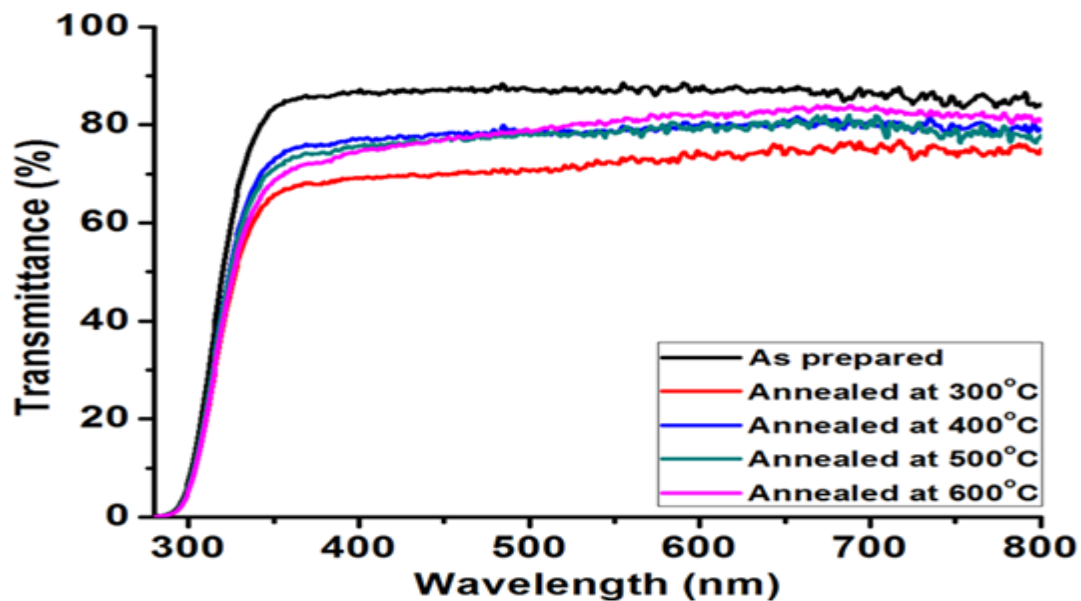


Fig. 2: UV-Vis transmittance spectra for un-doped SnO₂ thin films annealed at different temperature.

Table 2: Optical energy band of annealed SnO₂.

Sample	Energy band gap (eV)
As prepared	4.116
Annealed 300°C	4.121
Annealed 400°C	4.120
Annealed 500°C	4.117
Annealed 600°C	4.106

Table 3: Sheet resistance (Rs) and transmittance of SnO₂ annealed at different temperature.

Temperature	Rs (Ωsq^{-1})	Transmittance
As prepared	8.448	85%
300°C	5.247	68%
400°C	2.068	75%
500°C	1.796	73%
600°C	1.602	80%

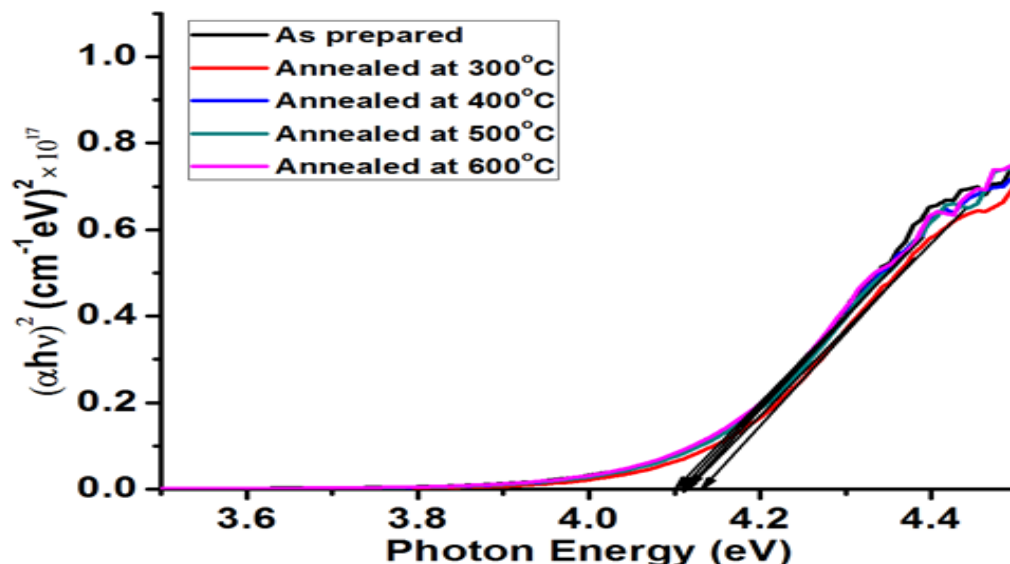


Fig. 3: Energy band gap for un-doped SnO₂ thin films annealed at different temperature.

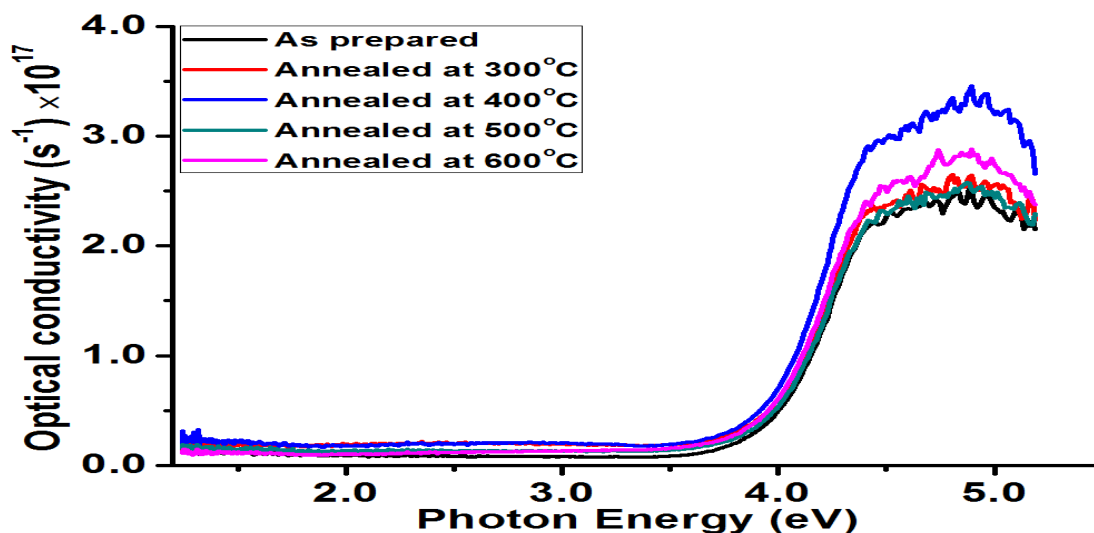


Fig.4: Optical Conductivity Spectra for un-doped SnO₂ thin films annealed at different temperature.

4 Conclusions

Effect of annealing temperature on structural, optical and electrical properties of tin oxide thin films which were prepared by sol-gel spin coating technique has been studied. X-ray diffraction patterns confirmed the proper phase formation of the SnO₂ thin film. Optical transmittance spectra of the films showed considerable high transparency (80%) in the visible region and the transparency decreases with the decrease in annealing temperature. The optical studies revealed that increasing the annealing temperature decreased the energy band gap

from 4.121eV to 4.106eV, this implies that the growth of grain size and the increase in defect states near the bands. The SnO₂ (annealed at 600°C) film has lowest sheet resistance (1.602 Ωsq⁻¹) making it to be highly conductive. The thin films properties observed confirmed that they are good materials for solar cell application.

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