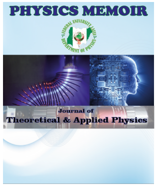


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Original Research

# Effects of Annealing Temperature on Optical and Electrical Conductivity of (80 nm ITO)/(100 nm ZnO) Transparent Conducting Oxide Multilayer Films

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**Abstract**

Conductive and transparent multilayer films consisting of two layers (ITO/ZnO) have been fabricated as Transparent Conducting Oxides (TCO). 100 nm ZnO film was deposited on 80 nm Indium Tin Oxide (ITO) film by thermal evaporation in a residual pressure of $10^{-5}$ Torr and the films were annealed at 400 °C and 500 °C for 60 minutes in Tubular furnace. The as-grown and the annealed films were characterized using Powder x-ray Diffractometer (PXRD) for the phase identification of the materials and UV-Visible Spectrophotometer for the photometric measurements. Beer's Lambert equation was employed to estimate the absorbance of each film under study. Four point probes based instrument was used to measure the average resistance of the films. The sheet resistance of the film was calculated from the electrical resistance of the material. The results revealed that the transmittance spectra of (80 nm ITO/100 nm ZnO) multilayer films depend strongly on the annealing temperatures and the wavelength. Optical transmittance of about 95 % was estimated for the 400 °C annealed film in the ultraviolet and visible region and relatively decreases towards the infrared region of the solar spectrum. The average electrical conductivity of 14000 $(\text{ohm.cm})^{-1}$ was also estimated for the 400 °C annealed film. Rhombohedral centered and hexagonal lattice structures were identified for the ITO and ZnO films respectively. 80 nm ITO/100 nm ZnO multilayer films annealed at 400 °C is hereby recommended as a good TCO for solar cells applications.

*Keywords:* TCO, ZnO, ITO, XRD, UV-Visible Spectrophotometer

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## 1. Introduction

Transparent Conducting Oxide (TCO) film is a significant component in different optoelectronic devices such as solar cells, light emitting diodes, photodiodes, and electro chromic devices. The resistivity of these electrodes should

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be minimized as much as possible with keeping its high optical transparency particularly over the visible region of the solar spectrum [1].

Indium Tin Oxide (ITO) thin films are wide gap semiconductors with a fairly low resistivity and generally used as transparent electrode in the visible region of the spectrum [2]. Due to this uniqueness, ITO films are widely employed for many applications. It is a well known n-type transparent conducting oxide material [3-5].

Zinc Oxide (ZnO) is a wide band gap semiconductor having high optical transparency and luminescence in visible and near ultraviolet region of spectrum. Therefore, it is usually used in light emitting diodes and solar cells [6-11]. It has unique chemical and physical properties, such as high chemical stability, high electrochemical coupling coefficient, broad range of radiation absorption and high photo stability. Infact, it is a multifunctional material. In materials science, Zinc Oxide is classified as a semiconductor in group II-VI, whose covalence is on the boundary between ionic and covalent semiconductors. Their broad energy band (3.37 eV), high binding energy (60 meV) and high thermal and mechanical stability at room temperature make it excellent for potential use in electronics, optoelectronics and laser technology [12-14]

It is usual that the low temperature processed ITO films have at least one order of magnitude higher electrical characteristics compared to those of the high temperature processed ITO electrodes. This reason limits the use of ITO films in optoelectronic applications. A possible method to overcome this problem is to employ multilayer thin films as substrate. Several papers have been reported on the use of ITO/metal/ITO type multilayer films with sheet resistance lower than the single layer ITO glass in which the metal layer included Copper (Cu), Silver (Ag) and Gold (Au) [1520].

Multilayer thin films showed different physical properties other than the conventional monolayer thin films [21]. In order to get higher conductivity with higher transparency and multifunctionality, Transparent Conducting Oxide (TCO) has been incorporated in multilayer stacks. In this work, 100 nm ZnO film was deposited on 80 nm ITO film via thermal evaporation to form multilayered TCO film structure to achieve both higher conductivity and transmittance in the visible region of the solar spectrum. The effects of annealing temperatures on the optical and electrical characteristics of each film were studied.

## 2. Materials and Method

Zinc Oxide (99.99 % pure) powder obtained from Labtrade, Ilorin was evaporated from molybdenum boat and deposited on the commercially available 80 nm ITO film via thermal evaporation to form the multilayer of (80 nm ITO)/(100 nm ZnO) film in a residual pressure of $10^{-5}$ Torr. The ITO substrates used were cleaned with acetone and methanol, and washed in an ultrasonic bath with de-ionized water and dried in a dust free atmosphere. The substrates temperature was kept fixed at room temperature. The fabricated films were respectively annealed in the furnace at 400 °C and 500 °C for sixty minutes. The as-grown and the annealed films were characterized using Powder X-Ray Diffractometer (PXRD) for the phase identification of the materials, UV-Visible Spectrophotometer for the photometric measurements (transmittance and reflectance). Beers Lambert equation was employed to estimate the absorbance of each film under study.

$$Absorbance = 2 - \log_{10} \%T. \tag{1}$$

Four point probe based instrument was employed to measure the average resistance of the films by passing current through the outside two points of the probe and measuring the voltage across the inside two points and the sheet resistance of each film was calculated.

$$R_s = 4.53\frac{V}{I}, \tag{2}$$

where $V/I$ is the electrical resistance of the material $R_s$ is the sheet resistance of the film. The thickness of the film (in cm) and its resistivity (in ohm cm) are related to $R_s$ by:

$$R_s = \frac{\text{Resistivity}}{\text{Thickness}}. \tag{3}$$

The resistivity of the film can be calculated if the thickness is known or the thickness can be estimated if the resistivity is known. The conductivity ($\delta$) of the film can be calculated using equation (4):

$$\delta = \frac{1}{\rho} \tag{4}$$

### 3. Results and Discussion

Figure 1 shows the PXRD of ZnO powder. The hexagonal wurtzite structure of Zinc Oxide was confirmed by several peaks of $2\theta$ at around 32, 34, 36, according to International Centre Diffraction Data (ICDD) card no: 89-1397. This assignment is in agreement with [6]. The most prominent peak at $2\theta$ of 32 degree in the diffractogram was indexed to (100) plane with inter planar spacing of 2.8171Åand the space group P63mc (183). Figure 2 shows the PXRD of as grown ITO film with Rhombohedral centered structure confirmed by the ICDD card no: 89-4599. The observed peaks at $2\theta$ angles are 30, 35, 51 and 61 degrees which correspond to the diffraction angles of ITO. The most prominent peak at $2\theta$ of 30 degree was indexed to (121) plane with inter planar spacing of 2.9231Å. The annealing temperatures have no effect on the inter-planar distance and lattice constants of the multilayer films. The multilayer films become more polycrystalline at higher annealing temperatures without phase shift.

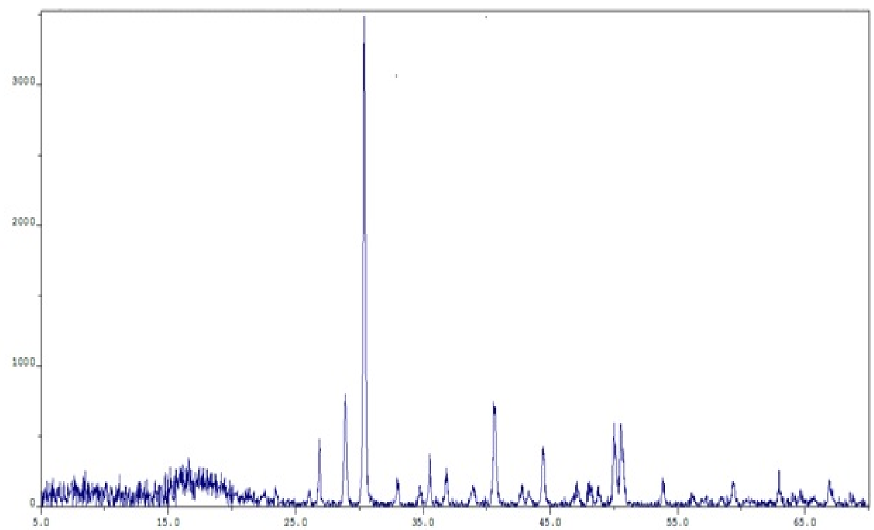


Figure 1. PXRD of ZnO powder

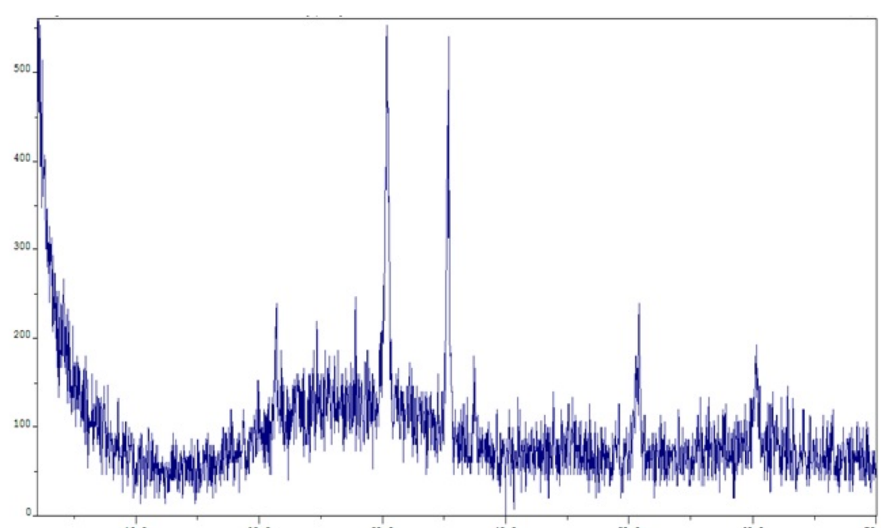


Figure 2. PXRD of as grown ITO film

The percentage reflectance of as-grown, 400 and 500 °C annealed (80 nm ITO)/(100 nm ZnO) multilayer films are shown in Figure 3. It was found that the magnitude of reflectance of multilayer films vary periodically with wavelengths. Multiple oscillations occur on the reflectance curves due to interferences among multiple reflected waves. As the wavelength increases, oscillation period of these films changes. Thus, the reflectance characteristics of (80 nm ITO)/(100 nm ZnO) multilayer films are strongly dependent on the annealing temperatures and wavelength of electromagnetic spectra. Highest peak value of percentage reflectance of 20, 55 and 65% were estimated at 550 nm wavelengths for the as-grown, 400 and 500 °C annealed films.

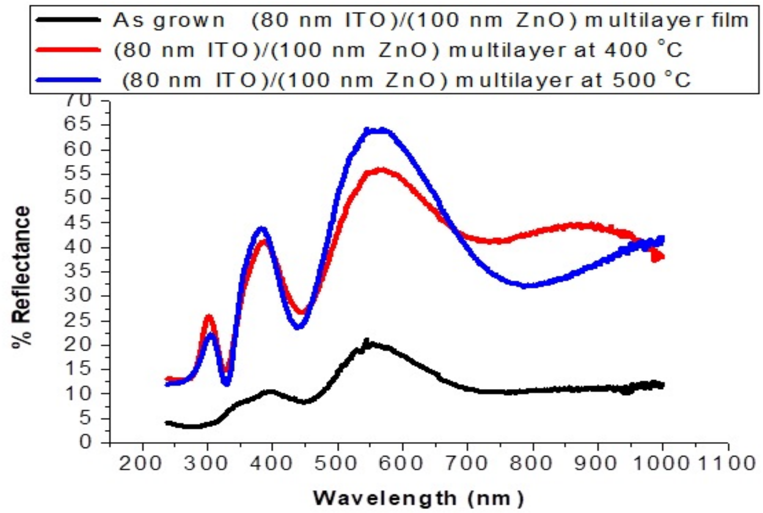


Figure 3. % Reflectance of (80 nm ITO)/(100 nm ZnO) multilayer films at different temperatures

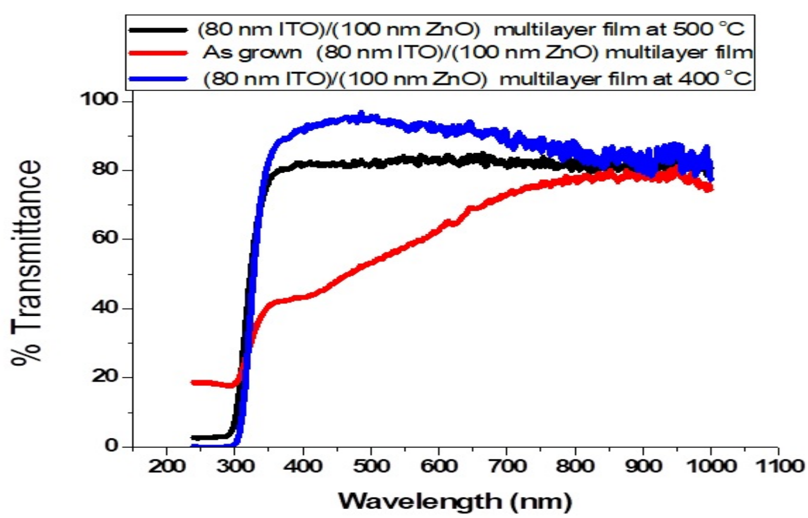


Figure 4. % Transmittance of (80 nm ITO)/(100 nm ZnO) multilayer films at different temperatures

The percentage transmittance characteristics of (80 nm ITO)/(100 nm ZnO) multilayer TCO films at different annealing temperatures are shown in Figure 4 as functions of wavelength. The effect of annealing temperatures was pronounced in the percentage transmittance spectra of the multilayer films. The transmittance spectra also depend on annealing temperatures and the wavelength. Highest optical transmittance of about 95% was estimated for the 400 °C annealed multilayer films at 300 nm wavelength in the ultraviolet through visible region and relatively decreases towards the infrared region of the solar spectrum. The transmittance value of 400 °C is relatively higher than the as grown and 500 °C annealed films and relatively the same in the infrared region of the solar spectrum.

It was observed from the absorbance spectra in Figure 5 that the absorbance of (80 nm ITO)/(100 nm ZnO) multilayer film was relatively higher in the UV region than in the visible and infrared regions of the solar spectrum. In the visible region, the absorbance of as grown was relatively higher than the 400 °CC and 500 °C due to mass density and annealing temperature effects. This observation of nearly zero absorbance for 400 °CC and 500 °C annealed films in the visible region of the solar spectrum proved higher optical transparency of the films. The near zero absorption of 400 °C and 500 °C annealed films favours higher transmission of the incident photons. This observation is excellent for solar cells applications.

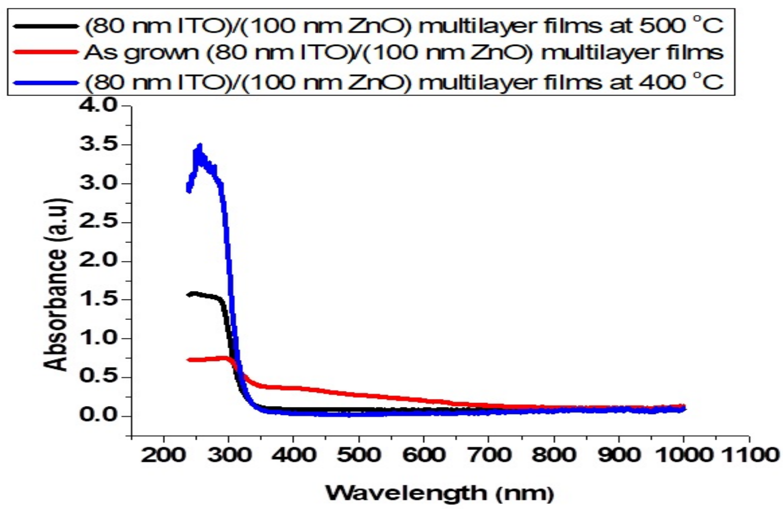


Figure 5. Absorbance spectra of (80 nm ITO)/(100 nm ZnO) multilayer films at different annealing temperatures

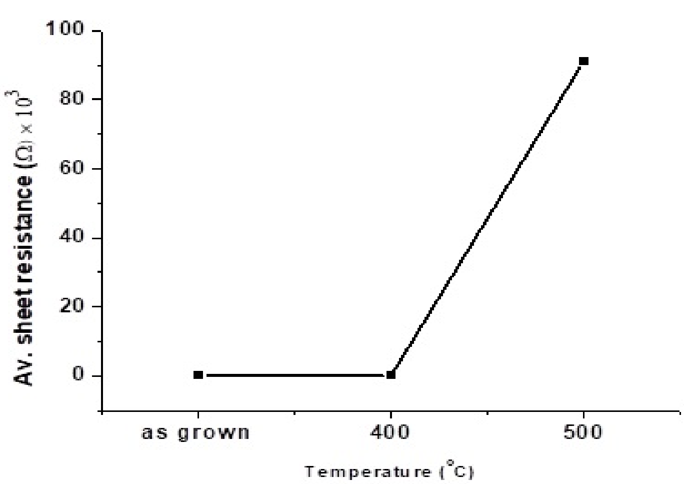


Figure 6. Average sheet resistance (Ω) against Temperature (°C) of (80 nm ITO)/(100 nm ZnO) multilayer films

Figure 6 shows the variations of the average sheet resistance against the annealing temperatures of (80 nm ITO)/(100 nm ZnO) multilayer films. The average sheet resistance of 7.878 and 8.85 ohms was estimated for the

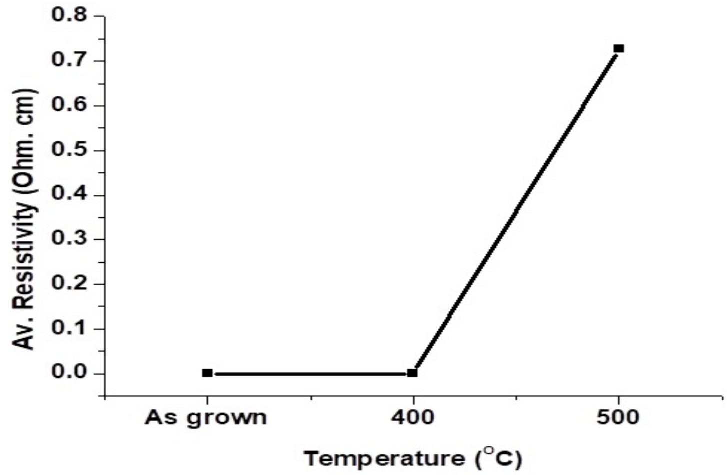


Figure 7. Average resistivity (Ω.*cm*) against Temperature (°C) of (80 nm ITO)/(100 nm ZnO) films

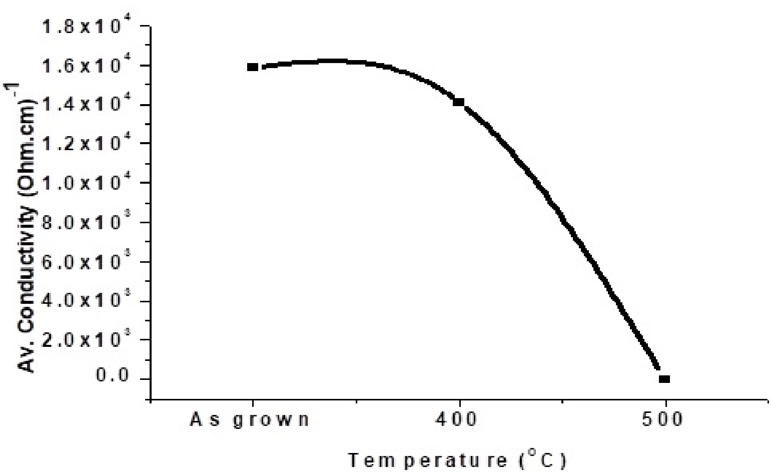


Figure 8. Average conductivity $(\Omega.cm)^{-1}$ against Temperature (°C) of 80 nm ITO/100 nm ZnO multilayer films

as grown and 400 °C annealed multilayer films respectively. The unusual behaviour was observed for the 500 °C annealed (80 nm ITO)/(100 nm ZnO) multilayer film with the average sheet resistance of 90880 ohms. The higher sheet resistance of the multilayer film at 500 °C is not suitable for solar cell application.

The average resistivity of (80 nm ITO)/(100 nm ZnO) multilayer films at different annealing temperatures under study is shown in Figure 7. The effect of annealing temperatures was clearly shown on the multilayer films. The average resistivity of the films showed symmetrical patterns with the average sheet resistances obtained for the as grown and the 400 °C annealed films. The unusual behaviour was also observed for the 500 °C annealed (80 nm ITO)/(100 nm ZnO) multilayer films with the average resistivity of 0.727 (ohms.cm) and cannot be employed for solar cells application.

Figure 8 showed the average conductivity of (80 nm ITO)/(100 nm ZnO) multilayer films at different annealing temperatures. The average conductivity of as grown film was maximum 16000 $(\text{ohm.cm})^{-1}$ and decreases as the annealing temperatures increases. The average conductivity of (80 nm ITO/100 nm ZnO) multilayer films at 400 °C was found to be 14000 (ohm.cm)$^{-1}$ and drastically becomes zero at 500 °C. The annealing temperature up to 500 °C for the multilayer of (80 nm ITO/100 nm ZnO) is not good as TCO for solar cells applications.

## 4. Conclusion

The transmittance spectra of (80 nm ITO/100 nm ZnO) multilayer films depend strongly on the annealing temperatures and the wavelength. Optical transmittance of about 95% was estimated for the 400 °C annealed multilayer film at 300 nm wavelengths in the ultraviolet and visible regions and relatively decreases towards the near infrared region of the solar spectrum. The average conductivity of 14000 (ohm.cm)$^{-1}$ was estimated for the 400 °C annealed film which drastically becomes zero at 500 °C. Rhombohedral centered and hexagonal lattice structures were identified for the multilayer films of ITO and ZnO respectively at different annealing temperatures under study. The annealing temperature of 400 °C is recommended for the treatment of (80 nm ITO/100 nm ZnO) multilayer films as a good TCO for solar cells applications.

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