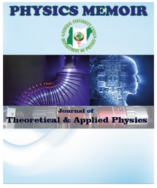

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Original Research

Characterization and Optimization of Transparent and Conductive ITO Films Deposited on n and p-types Silicon Substrates

A. K. Isiyaku[a,b,*], A. H. Ali[a], S. G. Abdu[b], M. Tahan[c], N. A. Raship[d], A. S. Bakri[c], N. Nayan[c]

[a] *Optical Fiber Laser Technology Group, Department of Physics and Chemistry, Faculty of Applied Sciences and Technology Pagoh Educational Hub, Universiti Tun Hussein Onn Malaysia, 84600 Pagoh, Johor Malaysia*
[b] *Department of Physics, Faculty of Science, Kaduna State University, P.M.B 2339, Kaduna State, Nigeria*
[c] *Microelectronic and Nanotechnology Shamsuddin Research Centre (MiNT-SRC), Universiti Tun Hussein Onn Malaysia*
[d] *Department of Electrical and Electronic Engineering, Universiti Pertahanan Nasional Malaysia, 57000 Kem Sungai Basi, Kuala Lumpur, Malaysia*

Abstract

The characteristics of enhanced transparent and conductive indium tin oxide (ITO) films on n-type and p-type Si substrates grown by radio frequency (RF) magnetron sputtering were investigated. The structural, optical and electrical properties of the films after annealing at different temperatures ranging from 300^{o}C to 600^{o}C in air were studied. X-ray diffraction (XRD) analysis reveals an amorphous structure for the as-deposited films of both the n-type and p-type Si. The annealed films exhibited a polycrystalline nature with preferential peaks orientation along (222) and (400) crystalline directions. Atomic force microscope (AFM) results indicate smooth surface morphology with increasing roughness as the annealing temperature increases. The surface roughness of the ITO films on the p-type Si was high (6.65 nm) at 500^{o}C and very good micro-structures on both Si types were obtained at 500^{o}C. Optical transmittance is enhanced from 89.1% for the as-deposited film to 95.7% for film annealed at 500^{o}C in the visible range. ITO films on n-type and p-type Si demonstrate a substantial reduction in their electrical resistivity and sheet resistance with increasing annealing temperature. The ITO/p-Si structure exhibits a low resistivity of $2.45\times10^{-5}\Omega$-cm compared to $7.46\times10^{-5}\Omega$-cm for ITO/n-Si structure. The films performance showed a figure of merit of $14.68\times10^{-3}\Omega$-cm for ITO/n-type Si and $44.74\times10^{-3}\Omega^{-1}$ for ITO/p-type at 500^{o}C indicating that the optimized ITO films on p-type Si can be a promising ITO/p-Si hetero-junction for silicon solar cells.

Keywords: ITO, Post-Annealing Treatment, Figure of Merit, n-type Si, p-type Si, Band Gap

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*Corresponding author tel. no:
Email address: kabiru.aliyu@kasu.edu.ng (A. K. Isiyaku)

1. Introduction

Transparent and conductive indium tin oxide (ITO) films are commonly used as contact layer in optoelectronic devices. This is because of their high optical transmittance in the visible region, good electrical conductivity and chemical stability [1-3]. The wide band gap nature of the ITO films (3.4 - 4.5 eV) is responsible for their high transmittance whereas the large number of intrinsic oxygen vacancies and extrinsic Sn^{4+} doping in the material are responsible for their excellent conductivity [4, 5]. Solar cells [6], liquid crystal displays (LCD) [7] and organic light-emitting diodes (OLED) [8], are some of the optoelectronics devices that incorporate ITO film as a contact. The incorporated ITO films need to be highly transparent in the visible region and of good electrical conductivity in order to improve the performance of device application. Thus, selecting an optimum ITO film thickness is crucial for simultaneously having high transmittance and good conductivity. Furthermore increasing the thickness of the ITO films above 300 nm results in a lower transparency though with increased conductivity, and because the resistivity value of the films is found to increase when a film of thickness less than 100 nm is used, there is need for more investigation [9, 10]. The choice of an appropriate thickness is therefore very vital to balance the trade-off between transparency and conductivity. Ghorannevis et al. [4] deposited ITO films of 100, 200 and 300 nm thickness, respectively using RF magnetron sputtering and reported that 100 nm film exhibited the lowest transmittance and produced the highest resistivity thereby buttressing the importance of thickness selection for optimum utilization of ITO films for various applications.

The structural, optical and electrical properties of the ITO films also depend on the deposition methods used. Several thin films deposition methods such as pulsed laser ablation [11], spray pyrolysis [12], thermal evaporation [13], electron beam evaporation [14] chemical vapour deposition [7] and direct current (DC)/ radio frequency (RF) magnetron sputtering [4, 15] have been used for the deposition of ITO films and uniform thin films over a large area of excellent quality can be obtained using DC/RF sputtering [16, 17]. Mostly, the as-deposited films produced by magnetron sputtering are amorphous in nature and it has also been observed that the as-deposited amorphous films can be transformed into polycrystalline films with better electrical and optical properties when subjected to post temperature annealing treatments [18]. Heat treatment method, temperature annealing ambient and other factors such as time, pressure and temperature play significant roles in the deposition of high-quality ITO films [19]. Some researchers reported on reduced structural defect in annealed ITO films with enhanced films crystallinity [19–21]. Furthermore, different groups of researchers have produced ITO/Si films with enhanced optical and electrical properties after post-annealing treatments at temperature above 250^{o}C [5,9,18–22] and changes on the optoelectronic characteristics of the ITO films are believed to originate from the variation on the local ordering of the ITO structure during crystallization and oxygen vacancy formation [19, 23, 24]. Thus this present work will report the structural, optical and electrical properties of improved ITO films deposited on n-type and p-type Si using RF sputtering method after post temperature annealing treatments from $300 - 600^{o}$C.

2. Materials and Methods

This work reports on the ITO films deposited using RF magnetron sputtering on n-type and p-type Si with resistivity of 0.1-1.0 Ω-cm respectively. The effects of varying annealing temperatures in the range $300 - 600^{o}$C on the structural, optical and electrical properties of 170 nm ITO films deposited using In_2O_3: SnO_3 (ITO) target of 90:10 weight ratio in a pure argon atmosphere were studied. Prior to the films deposition, Si substrates were cleaned in boiled acetone for 5 minutes. They were then rinsed in Isopropyl solution, deionised water and blown dry in a nitrogen environment. Furthermore, the corresponding glass substrates used for transmittance measurements were cleaned using D1DN steps (D1 decon90 glass cleaner, D deionized water, and N nitrogen gas atmosphere). Experimental conditions and sputtering parameters of this work is as shown in Table 1 and carbolite electric furnace was used for the post temperature annealing treatments in air at 300^{o}C, 400^{o}C, 500^{o}C and 600^{o}C respectively.

ITO films thickness was measured using optical reectometer Filmetrics F20 model. While their structural analysis was performed using X-ray diffraction (XRD), Atomic force microscopic (AFM) was used to examine the films surface morphology. Optical transmittance measurements in the wavelength range 300-700 nm were conducted using UV/Vis spectrophotometer and their electrical characteristics were determined by a four-point probe.

Table 1. Details of Sputtering Parameters

Vacuum Pressure	Working Pressure	RF Power	Gas Flow	Target-Substrate Distance
6.0×10^{-6}Torr	5.2mTorr	120W	50sccm	7cm

3. Results and Discussion

Fig. 1(a) and (b) shows an XRD structural pattern for the as-deposited and post annealed ITO films deposited on n-type Si (111) and p-type Si (111) substrates respectively. The as-deposited films on both the n-type and p-type Si indicate non-crystalline characteristics with a broad peak sandwich between 2 theta degree values 30^o and 35^o indicating the amorphous feature of the films. Unlike in p-type substrate, the as-deposited ITO film on an n-type substrate shows a narrow peak along (222) as the maximum intensity. For the annealed films (on n and p types) at 300^oC, a polycrystalline pattern indicating cubic Bixbyite structure was observed with sharp diffraction peaks along (222), (400), (440) and (622) directions. All the annealed films showed a strong and preferred peak orientation at (222) direction. It was also observed that with the exclusion of a crystal peak at (211), the ITO films structure and orientation at annealed temperatures 400^oC, 500^oC and 600^oC are similar to that of 300^oC film. This pattern indicates an enhancement in crystallinity with increasing annealing temperature as also reported by Balasundaraprabhu et al. [15].

It was observed that in both the Si types, an amorphous structure was formed when ITO film of 170 nm thickness was grown at room temperature using RF sputtering with the ITO film on n-type Si showing a slight growth of crystal peak at the (222) direction. This indicates that the sputtered particles kinetic energies (K.E) that tend to improve the movement of arriving particles on the substrate surface are weak hence the amorphous nature observed. In their report Ghorannevis et al. [4] under certain sputtering conditions, produced crystalline ITO films of thickness 200 nm and 300 nm at room temperature using RF sputtering, signifying the strong effect of K.E of the sputtered ITO atoms on the substrate surface for higher thicknesses. Furthermore, a polycrystalline structure of a dominant (222) preferential orientation was obtained when the as-deposited ITO films were subjected to furnace annealing from $300 - 600^o$C. This process enhanced the K.E of the sputtered atom resulting in an increased ad atom mobility on the substrate surface and hence improved electrical conductivity of the films. Moreover, none of Sn, SnO, SnO_2 element or compound characteristic peaks was detected in all the annealed samples, which shows a possibility that Sn atoms were incorporated into In_2O_3 lattice substitutionally [4, 15].

Fig. 2 shows AFM images of ITO films on n-type and p-type Si for as-deposited and annealed 400^oC and 500^oC. The scan done over 1000 nm x 1000 nm area shows a smooth surface morphology for both the Si types. The dependence of ITO films root-mean-square (RMS) roughness and grain size on post annealing temperature is as shown in Fig. 3a and 3b. The ITO films images which were analysed using AFM NanoScope software shows that surface morphology is dependent on annealing temperature of the samples. Root mean square roughness of both Si types increased as the temperature increases and results in larger grains. While the as-deposited film showed a fairly good microstructure the film annealed at 500^oC displayed a better and improved microstructure. The grain size of ITO films increased with increased annealing temperature, with films annealed at 400^oC and 500^oC having larger grain sizes of 43.6 nm and 47.5 nm on n-type Si and 86.3 nm and 97.9 nm on p-type Si with fewer grain boundaries in the latter compared to the former. Gulen et al. [19] and Hu et al. [25] also reported similar results of surface roughness and grain sizes of their produced ITO films. The AFM images of the ITO films in Fig. 2 shows that at higher temperatures smaller crystallites peaks diffusing and recombining to form larger grains produced films with enhanced optical transmittance.

Fig. 4a and 4b shows ITO films optical transmission and absorption spectra. In Fig. 4a, the as-deposited film exhibited the lowest optical transmittance of 89.1% at 600 nm wavelength. A substantial improvement in optical transmittance with increasing temperature annealing was observed. ITO film annealed at 500^oC shows the highest transmittance of 95.7 at 600 nm wavelength in the visible region. The enhanced optical transmission of the films is attributed to the particles growth, possible loss of light arising from oxygen vacancies and reduction in film defects [19, 26]. The interfering fringes observed at different wavelengths implies that the films are highly uniform [4]. Furthermore, the transmittance spectra peaks of the films annealed up to 500^oC were found to be shifted toward lower

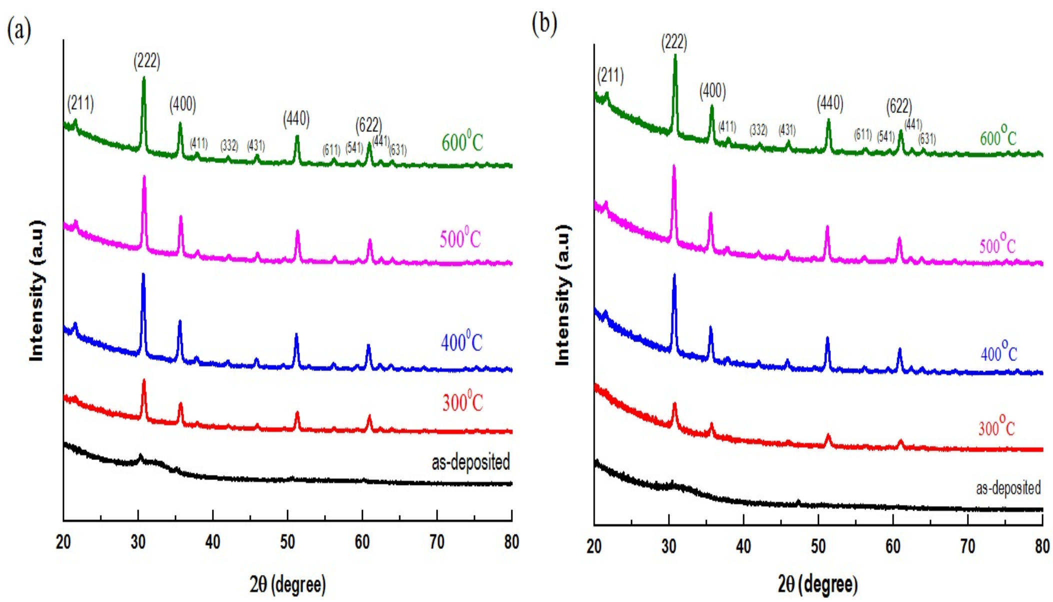

Figure 1. XRD patterns for the as-deposited and post annealed ITO films deposited on (a) n-type Si (111) and (b) p- type Si (111) respectively

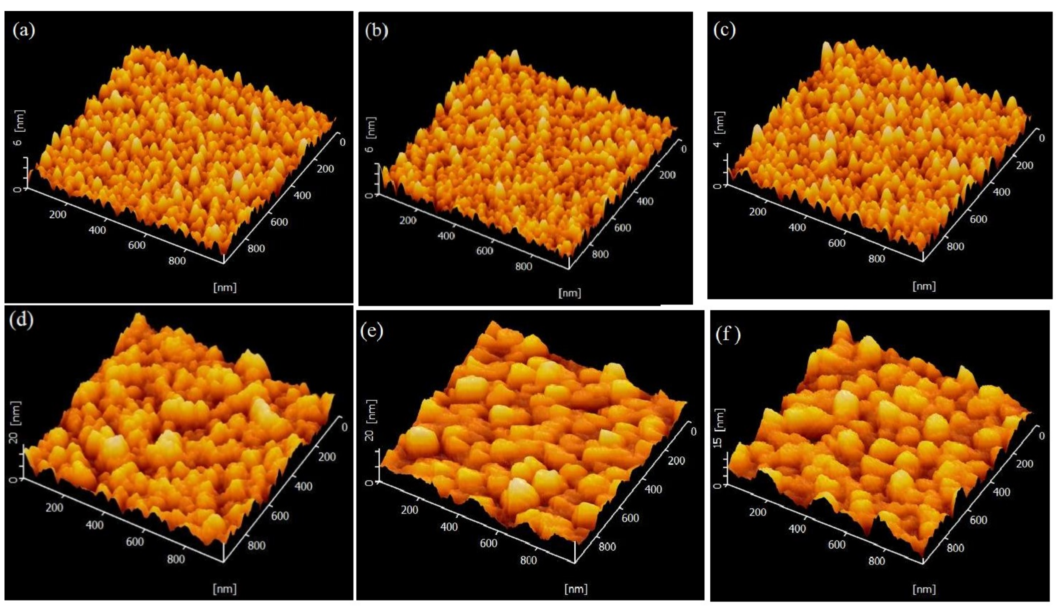

Figure 2. AFM images of ITO films (a) as-deposited on n-type Si (b) as-deposited on n-type Si annealed at 400^{o}C (c) as-deposited on n-type Si annealed at 500^{o}C (d) as-deposited on p-type Si (e) as-deposited on p-type Si annealed at 400^{o}C (f) as-deposited on p-type Si annealed at 500^{o}C

energy (blue shifted) resulting in an increase in ITO film carrier concentration and is well explained by a famous Burstein-Moss shift model [27–30]. The shift in optical absorption edges toward lower wavelengths (Fig. 4b) as the annealing temperature increases signifies a possible widening in the optical band gap energy with increasing temperature[4,30]. The increased charge carriers' concentration is responsible for the widening or increased band

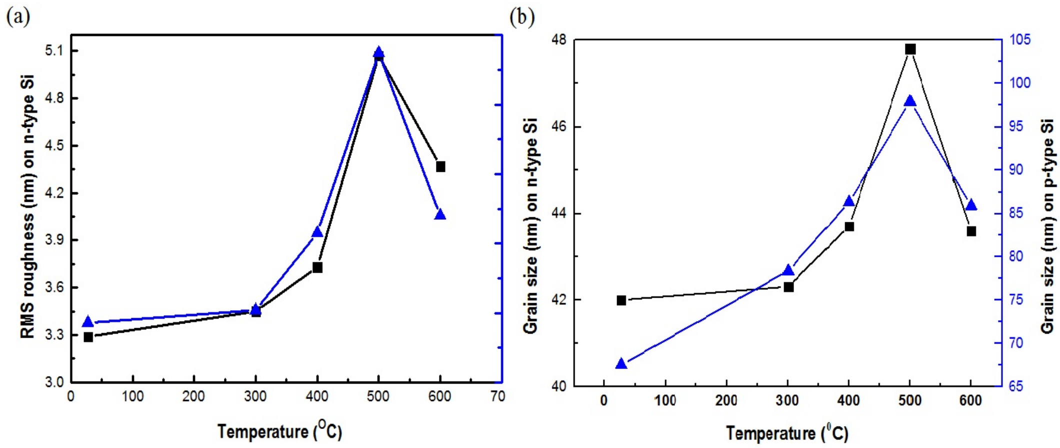

Figure 3. ITO films variation of (a) RMS roughness on n-type and p-type Si with annealing temperature (b) Grain size on n-type and p-type Si with annealing temperature

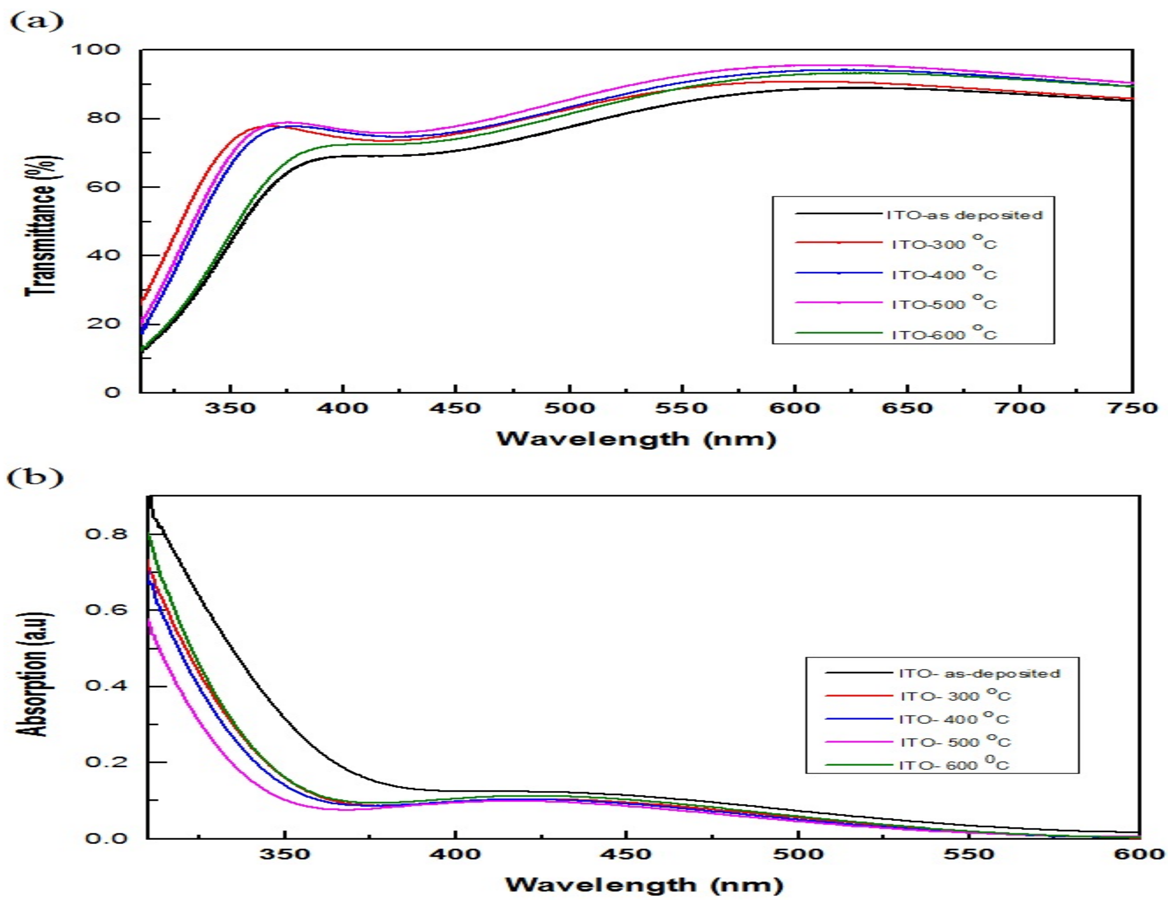

Figure 4. ITO films (a) optical transmission spectra and (b) absorption spectra for as-deposited, and post annealed 300^{o}C, 400^{o}C, 500^{o}C and 600^{o}C respectively

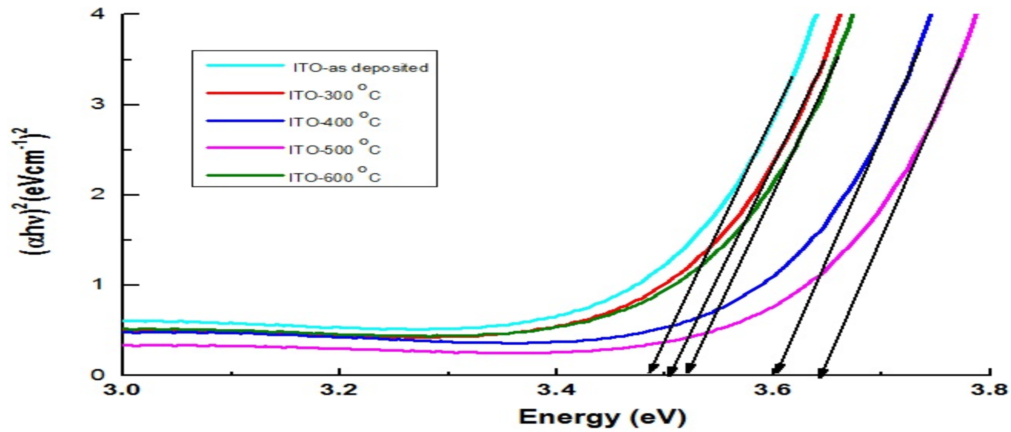

Figure 5. ITO films optical absorption coefficient (α) against band gap energy (Eg)

gap energy as in Burstein-Moss model. In this case, the electron concentration in the annealed ITO films at higher temperature causes a partial filling of the lowest states of the conduction band thereby obstructing the lowest levels and then forcing the conduction band edge to lower wavelengths [30, 31].

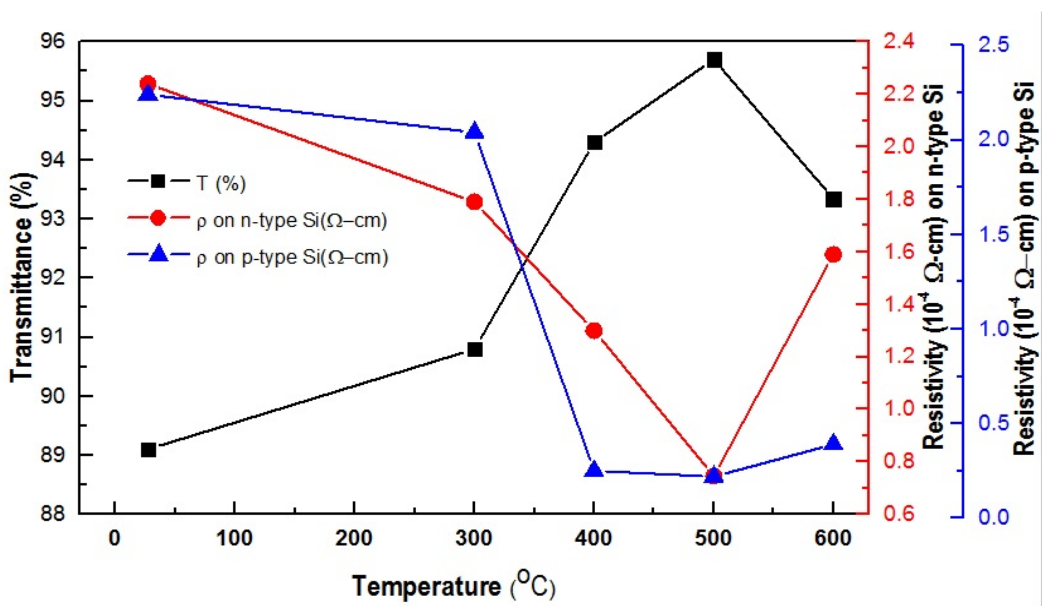

Figure 6. Transmittance and resistivity of the ITO films as a function of post-annealing temperature

Tauc plot for the as-deposited and annealed ITO films is presented in Fig. 5. This plot represents the relationship between optical absorption coefficient (α) and band gap energy (Eg) as expressed in Eqn. 1. In this work, direct allowed transition is assumed hence [19]:

$$(\alpha hv)^2 = A(hv - Eg) \tag{1}$$

where α stands for absorption coefficient, A is the edge width parameter (constant), hv denotes the photon energy and Eg is the energy band gap.

The ITO band gap energies are deduced by extrapolating the linear plots of Fig. 5 at $\alpha = 0$. The derived optical band gap values for the as-deposited ITO films are 3.49, 3.51, 3.60, 3.64 and 3.52 eV for annealed temperatures 300^{o}C, 400^{o}C, 500^{o}C and 600^{o}C respectively. The as-deposited film has the smallest band gap value while the film annealed at 500^{o}C has the largest band gap value indicating an increase in the band gap energy with post annealing temperature. At a very high temperature (500^{o}C), the band gap value decreased to less than 3.60 eV as a result of increased grain size caused by quantum confinement effect [19, 32–34].

Variation of ITO films transmittance peaks and electrical resistivity (ρ) (on n-type and p-type Si) with the annealing temperature is displayed in Fig. 6. The resistivity values decrease with increasing post temperature annealing. Lowest resistivity values of 7.46×10^{-5} Ω cm for n-type Si and 2.45×10^{-5} Ω cm for p-type Si were obtained at 500^{o}C. The reduction in resistivity is attributed to the increase in grain size at a high temperature resulting in decreased grain boundary scattering [1, 25] as well as an enhancement in electrical conductivity [1, 15]. For film annealed at 600^{o}C, it is resistivity increased to 1.59×10^{-4} Ω cm for n-type and 3.9×10^{-5} Ω cm for p-type. This behaviour is due to the film weak electrical property as a result of the oxidation of the film oxide vacancy [1]. Likewise, ITO films transmittance increases with increasing post annealing temperature with only that of film annealed at 600^{o}C dropping slightly due to increased electrical resistivity [1].

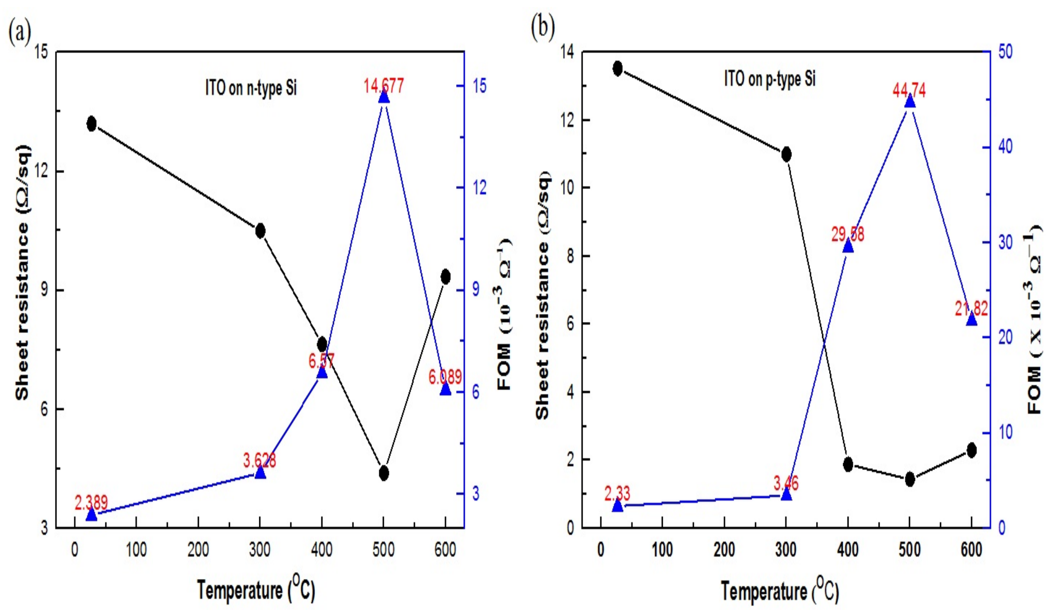

Figure 7. ITO films sheet resistance and figure of merit as a function of post-annealing temperature on (a) n-type Si (b) p-type Si

To determine the performance of ITO films deposited on different substrates, the figure of merit (FOM) is commonly used. The FOM as developed by Haacke, [35] has optical transmittance peak T and sheet resistance R_{sh} as the measuring parameters:

$$FOM = \frac{T^{10}}{R_{sh}} \tag{2}$$

Parameters such as film thickness d, electrical resistivity ρ, and optical absorption α also play a huge role in evaluating the performance of ITO films as expressed in Eqn. 3. This is obtained after $T = e^{-\alpha d}$ and $R = \rho/d$ have been replaced in Eqn. 1:

$$FOM = (d/\rho)e^{-10\alpha d} \tag{3}$$

Fig. 7(a) and (b) shows the ITO films sheet resistance on n-type and p-type Si and calculated FOM as a function of annealing temperature. It can be seen that the FOM value of the films increases with increasing temperature. On n-type Si, maximum FOM value of $14.68 \times 10^{-3}\ \Omega^{-1}$ was obtained whereas a FOM value of $44.74 \times 10^{-3}\ \Omega^{-1}$ was obtained for p-type Si by films annealed at 500^oC. The improvement in FOM values was as a results of simultaneous enhancement of optical and electrical properties after post annealing treatment [3]. Similarly, the sheet resistance value decreases with increasing annealing temperature. The minimum R_{sh} value of the films occurred for the film annealed at 500^oC with 4.39 Ω/sq for n-type Si and 1.44 Ω/sq for p-type Si respectively. The decrease in R_{sh} is attributed to the increase in grain sizes coupled with strong films crystallization due to thermal energy absorption as indicated in XRD results with increasing post annealing temperature [36].

4. Conclusion

This work compared the role of post-annealing treatment of an optimised ITO films on n-type Si and p-type Si deposited by RF magnetron sputtering. The structural, optical and electrical properties of 170 nm ITO films were characterized after subjected to the post-annealing temperature at 300^oC, 400^oC, 500^oC, and 600^oC respectively. Structural analysis reveal polycrystalline films in all the annealed films on n-type and p-type Si with preferential grain orientation strongly along the (222) and (400) crystal directions. The films show a smooth surface roughness with larger grain size on p-type Si compared to n-type Si. The film annealed at produced the best microstructures with a dense and porous network of nanoparticles by both Si types. The optimized ITO film annealed at 500^oC shows the highest transmittance of 95.7% at 600 nm wavelength and lowest sheet resistance of 4.39 Ω/sq on n-type Si and 1.44 Ω/sq on p-type Si respectively.The results indicate that the post annealing treatment enhances the film crystallinity, optical transmittance and electrical conductivity. The optical absorption edges blue shifted with increasing temperature causing an increase in band gap energy. The calculated figure of merit indicates a higher performance of enhanced ITO films on p-type Si with FOM value of $44.74{\times}10^{-3}\Omega^{-1}$ making it a promising candidate for use as ITO/p-Si heterojunction for silicon solar cell.

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