Tuning the Topo-Morphological properties of ITO Films using Al-Ag interlayer for Low-Resistance Optoelectronics Devices

*1Isiyaku, A. K., 1Isah, M., 1Aliyu, M. M., 1Tanko, A. Y., 2Abdulkadir, A. and 3Ali, A. H.

1Department of Physics, Faculty of Science, Kaduna State University, Kaduna P.M.B 2339, Nigeria.
2Department of Physics, Umaru Musa Yar’adua University, Katsina, P. M. B. 2218, Nigeria
3Laser and Semiconductor Technology Research Group (LASERG), PDSR-COR, Department of Physics and Chemistry, Faculty of Applied Sciences and Technology, Universiti Tun Hussein Onn Malaysia (UTHM), Pagoh Educational Hub, KM 1 Jalan Panchor, 84600 Muar, Johor, Malaysia School of Physics, Petra University, Amman, Jordan.

*Corresponding author’s email: kabiru.aliyu@kasu.edu.ng  Phone: +2348060840640

ABSTRACT
Indium tin oxide (ITO) is recently attracting intense attention for application as transparent conducting electrode in different optoelectronic devices including solar cells, liquid crystal displays and organic light emitting diodes. This is attributed to their high optical transmittance in the visible region and good electrical conductivity. In this work, surface topological-morphological (topo-morphological) and electrical properties of ITO based multilayer films with aluminum-silver (Al-Ag) metal interlayer (ITO/Al-Ag/ITO) are investigated after post annealing treatment at 300-500°C by atomic force microscopic (AFM), field emission scanning electron microscopy (FESEM), four-point probe and hall-effect techniques respectively. The ITO/Al-Ag/ITO films are deposited by direct current and radio frequency magnetron sputtering techniques on p-type Si at room temperature. The results showed a smooth surface topology by as-deposited film with films smoothness improving after post annealing treatment as analyzed by AFM method. Sharp crystallities peaks were obtained by all the films with smaller peaks diffusing and recombining to form larger films with increasing post-annealing temperature. The films root means square roughness increased as the temperature increases with film annealed at 500°C showing a superior and enhanced microstructure. Compared to as-deposited film, careful observation shows that surface morphology smoothness increased with increase in temperature with films annealed at 400°C and 500°C showing highest increasing surface smoothness pattern as determined by FESEM technique. Similarly, higher grain sizes are observed especially by films annealed at 400°C and 500°C due to the heat absorption which causes the particles to expand thereby narrowing the grain boundaries and consequently improve the surface smoothness. These FESEM findings are consistent with AFM measurements confirming the high surface property and enhanced grain size with increasing post-annealing temperature. The films exhibit decreased electrical resistivity and sheet resistance while carrier concentration and Hall mobility increased with increasing post annealing treatment indicating highest corresponding values of 2.7 × 10⁻³ Ωcm, 3.92 Ω/sq, 16.1× 10⁻³ cm³ and 41.4 cm²/Vs at 500°C respectively. These highly enhanced topo-morphological multilayer films can be a promising contact for low-resistance optoelectronics devices.

Keywords:
Indium tin oxide, Topology, Morphology, Radio frequency, Magnetron sputtering.

INTRODUCTION
Transparent conducting oxides (TCOs) materials such as indium tin oxide (ITO), titanium oxide (TiO₂), zinc oxide (ZnO), or fluorine tin oxide (FTO) are recently attracting intense attention for application as transparent conducting electrode or anti-reflective coating (ARC) in different optoelectronic devices including solar cells, electrochromic, liquid crystal displays (LCDs) and organic light emitting diodes (OLEDs) (Isiyaku et al., 2019; Kumar et al., 2015; Pu et al., 2015).
This is accredited to their high optical transmittance in the visible region of the spectrum and good electrical conductivity (Cui et al., 2002; Isiyaku et al., 2020: Isiyaku et al., 2019a). ITO excellent properties such as high optical transmittance, chemical stability and good electrical conductivity have made it attractive for application in photonics and optoelectronics devices (Guillen and Herrero, 2011). ITO layers properties such as optical, electrical, mechanical, and structural have revealed an immense dependence on the types of deposition techniques used during preparation (Liu et al., 2015; Ali et al., 2015). Different techniques such as sol-gel, direct current (DC) and radio frequency (RF) magnetron sputtering, ultrasonic spray, chemical vapor deposition, thermal evaporation or electron beam evaporation have been used for ITO layers deposition (Isiyaku et al., 2022; Aijo et al., 2014; Bundesmann et al., 2014; Lee et al., 2012; Guillen and Herrero, 2011). Magnetron sputtering has the advantages of depositing uniform and high-quality ITO layers with good layers adhesion to substrate (Isiyaku et al., 2020; Isiyaku et al., 2021a).

However, indium (In) metal that formed the larger part of ITO is expensive in nature and reducing the ITO layer thickness to smaller thickness (to less than 150 nm) usually increases the layer’s resistivity due to classical size effect (Isiyaku et al., 2020; Isiyaku et al., 2016; Rein, 2015; Ali et al., 2014). Hence, the use of thinner transparent ITO layers having low electrical resistance is critical. It is understood that multilayer ITO (ITO/metal/ITO) concept has been recently explored to reduce the cost of In metal consumption and enhance the optoelectronics properties of the layers (Isiyaku et al., 2021a; Ali et al., 2016; Liu et al., 2015). It is observed that both the ITO and metal films in this multilayer structure play distinct key roles. Metals films such as Al, Cu, Ag, Ni, Pd, Au, Cr or Mg have been applied and each one of them shows different properties which allow one to get layer electrode more attuned to the specific properties (Guillem and Herrero, 2011; Isiyaku et al 2020). In this structure, separation of charge carrier is accelerated due to the present of free electrons in the ITO/metal materials and therefore enhance the carrier transport of the device (Abdulkadir et al., 2022; Isiyaku et al. 2021a). Compare to many other metals, silver (Ag) film exhibits moderate high transparency in the visible region and has a low resistivity value (Isiyaku et al., 2020). Moreover, in addition to its low-cost effectiveness (4.5–5.1 USD/kg), this material become widely acceptable for use as interlayer in high performance and less expensive ITO based multilayer structure. Aluminum (Al) film good adhesiveness and low resistivity value (2.8 μΩ cm), coupled with non-corrosion and non-oxidation properties have made it appropriate for photonic devices (Isiyaku et al., 2021b; Ali et al., 2016). ITO properties have been extensively studied on various substrates including glass, silicon Si (n or p type), polyethylene terephthalate, gallium nitride (n or p type), or polyethylene naphthalate (Alvarez-Fraga et al., 2015; Isiyaku et al., 2021a; Zhou et al., 2012). ITO multilayer structure can either serve as a transparent conducting electrode, anti-reflecting coating, an ohmic contact or rectifying contact (Kumar et al., 2015; Balasundraprabhu et al., 2009).

Parameters such as annealing treatment, layer thickness, and sandwiched metal intermediate layer have been reported to have immense influence on the ITO/metal contact characteristics (Isiyaku et al., 2019b; Ali et al., 2016). Substantial enhancement in topological, morphological, optical and electrical properties have similarly been witnessed after post annealing treatments (Ali et al., 2016; Lien et al., 2010; Lee and Park, 2006). In this work, the effect of inserting Al-Ag interlayer on the top and bottom ITO layers (ITO/Al-Ag/ITO multilayer) on topological-morphological (topo-morphological) and electrical properties are investigated. The films are prepared using radio frequency (RF) and direct current (DC) magnetron sputtering techniques and post annealed at 300-600°C respectively. Additionally, films correlation between topo-morphological and optoelectronic properties are discussed while single ITO films are equally prepared for comparison analysis.

**MATERIALS AND METHODS**

In this work, ITO/Al-Ag/ITO multilayer films were deposited using both SNTEK model of RF and DC magnetron sputtering systems for ITO and metals films respectively. Target of ITO with 90%:10% weight ratio of In2O3 and SnO2 was used to prepare the ITO films top and bottom layers using RF sputtering method in an uncontaminated argon environment at normal room temperature. The multilayer films are prepared on substrates; p-type Si wafer [p-type (111), boron, 0.1–1.0 Ω cm] and glass respectively. Table 1 presents the sputtering parameters adopted during the preparations. 13 cm is the target-substrate distance used for all depositions. Before the deposition, 1 x 1 cm size of Si wafer was cut, cleaned at 55°C in boiled acetone for 5 minutes. The wafer then rinsed in Isopropyl solution, Deionized (DI) water and later blown dry in nitrogen (N2) gas environment. Also, 2x 2 cm size of glass substrates were cut, and Decon-90 glass cleaner was used to clean, rinsed in DI water and then blown dry in N2 atmosphere. Al and Ag targets of 99.999% purity are used to prepare Si/ITO/Al-Ag/ITO films using DC sputtering process using parameters shown in Table 1. Optical reflectometer Filmetric F20 was used to measure separate thickness of ITO, Al, Ag and ITO (single) films recorded as 40 nm, 6 nm, 9 nm and 90 nm separately. The multilayer samples were subjected to
post annealing treatments at temperature 300-500°C respectively.

Table 1: ITO, Al and Ag sputtering parameters adopted during the preparation

<table>
<thead>
<tr>
<th>Sputtering parameters</th>
<th>ITO (top and bottom)</th>
<th>Al</th>
<th>Ag</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pre-sputtering time (sec)</td>
<td>900</td>
<td>900</td>
<td>900</td>
</tr>
<tr>
<td>Vacuum pressure (pa)</td>
<td>$7.99 \times 10^{-4}$</td>
<td>$7.73 \times 10^{-4}$</td>
<td>$7.47 \times 10^{-4}$</td>
</tr>
<tr>
<td>RF/DC power (watt)</td>
<td>120</td>
<td>80</td>
<td>90</td>
</tr>
<tr>
<td>Working Pressure (pa)</td>
<td>$6.93 \times 10^{-1}$</td>
<td>$6.81 \times 10^{-1}$</td>
<td>$6.71 \times 10^{-1}$</td>
</tr>
<tr>
<td>Deposition time (sec)</td>
<td>240</td>
<td>30</td>
<td>45</td>
</tr>
</tbody>
</table>

Atomic force microscope (AFM, Model: AFM5010 Hitachi) was used to determine films surface topology and it was operated under contact mode and NanoScope analysis software. Films morphology was examined by a field emission scanning electron microscope (FESEM, Model: JEOL JSM-7600F-SM17600053, equipped with an energy-dispersive X-ray spectrometer EDS, OXFORD X-MAX, Japan) with 5 kV acceleration voltage and 100,000 × magnification respectively. Electrical characteristics measurements were determined using both Four-point probe technique (Pro 4 Lucab Lab model) and Hall Effect measurement (Lake Shore 8400 Series model) systems respectively. The set-up of the characterization techniques used in this work is shown in Figure 1.

Figure 1: Set-up characterization models for (a) AFM (Topological analysis), (b) FESEM (morphological analysis), (c) Four-point probe (Electrical analysis) and (d) Hall Effect system (Electrical analysis).

RESULTS AND DISCUSSION
The performance of TCO films are enhanced by the improvement in optoelectronics properties in which surface topology plays a huge part. Figure 2 shows AFM images (three-dimension (3D)) of ITO/Al-Ag/ITO multilayer for as-deposited and post annealed films scanned over 1000 nm × 1000 nm respectively. It can be seen that, the as-deposited film exhibited a smooth surface topology. The films smoothness improved with increasing post annealing temperature. The dependence of this multilayer root means square (RMS) roughness and grain size (D) at different post-annealing temperature is displayed in Table 1 as deduced from AFM Nanoscope analysis software. Single ITO film annealed at 500°C is presented for comparison. From Table 2, it is clearly observed that the surface topology parameters of the multilayer films increased with increase in post-annealing temperature. Sharp crystallites peaks were obtained by all the films with smaller peaks diffusing and recombining to form larger films with increasing post-annealing temperature (Figure 2). The RMS of the multilayer films increased as the temperature increases with even the as-deposited film showing a good microstructure while the film annealed at 500°C showed a superior and improved microstructure due to improvement in the films...
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structural property after post annealing treatment at high temperature (Isiyaku et al., 2020). The increase in RMS is due to rapid growth in grain size (Bundesmann et al., 2014; Lee & Park, 2006; Lien et al., 2010).

The insertion of Al-Ag thin metals films sufficiently improved the grain size as compared to single ITO. Increase in post-annealing temperature led to increase in grain size (Figure 2). The highest measured grain size is 61.9 nm as obtained by multilayer film annealed at 500°C. Heat absorption as a result of post annealing treatment led to this substantial enhancement in grain sizes which subsequently enhanced both surface roughness and smoothness respectively. Grain boundaries reduces with increase in grain size by reducing the carrier and light scattering factors and hence enhanced the films optoelectronic properties (Isiyaku et al., 2021b). The multilayer films grain size showed an improvement in comparison with single ITO layer, the works of Ali et at. (2014) and Kumar et al. (2015) respectively.

![Figure 2: ITO/Al-Ag/ITO films AFM images for (a) the as-deposited (b) annealed films at 300°C (c) annealed films at 400°C and (d) annealed films at 500°C.](image)

Table 2: RMS roughness and grain size of ITO/Al-Ag/ITO films for as-deposited and annealed films and single ITO annealed at 500°C

<table>
<thead>
<tr>
<th>ITO/Al-Ag/ITO</th>
<th>RMS Roughness (nm)</th>
<th>Grain Size (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>As-deposited</td>
<td>1.26</td>
<td>52.8</td>
</tr>
<tr>
<td>300°C</td>
<td>1.56</td>
<td>55.6</td>
</tr>
<tr>
<td>400°C</td>
<td>1.67</td>
<td>60.4</td>
</tr>
<tr>
<td>500°C</td>
<td>1.71</td>
<td>61.9</td>
</tr>
<tr>
<td>Single ITO (at 500°C)</td>
<td>1.22</td>
<td>42.1</td>
</tr>
</tbody>
</table>

Surface morphological analysis of ITO/Al-Ag/ITO multilayer films was conducted by FESEM technique and the images are displayed in Figure 3. The as-deposited film exhibits smooth and continuously connecting surface morphology with dense particles evenly distributed throughout the film surface. Compared to as-deposited film, careful observation shows that surface smoothness increased with increase in temperature with films annealed at 400°C and 500°C showing an increasing surface smoothness feature. Similarly, higher grain sizes are observed especially by films annealed at 400 °C and 500 °C due to the heat absorption which causes the particles to expand thereby narrowing the grain boundaries and subsequently enhance the surface smoothness. These results are consistent with AFM measurements confirming the high surface property and grain size with increasing post annealing temperature. Even at moderate 500°C post-annealing temperature, no significant Al-Ag films agglomeration was observed on the multilayer films surface to allow for unwanted microstructural changes and that shows a good stability property of the films. The observations of the surface morphology features by FESEM and AFM techniques are in good agreement.
The structural property in terms of elemental compositions of the ITO/Al-Ag/ITO multilayer films was analyzed using EDX technique. The as-deposited and post-annealed multilayer films' elemental composition in weight percent is shown in Table 3. Element like oxygen O shows a reduction in weight after post-annealing treatment. Whereas metal and semiconductor materials; Ag, Al, In, Sn and Si increased with increasing temperature. Si element constitutes the highest composition because it serves as a base material. Similarly, Al shows the least elemental composition due to its very thin layer. However, the Sn low composition is attributed to it being a dopant element to the ITO composition. The elemental distribution of the as-deposited and 500°C post-annealed films are shown in Figure 4. The results confirm the present and improvement in ITO composition after post-annealing treatment which is in agreement with the work of Isiyaku et al. (2020).

Table 3: Elemental composition of ITO/Al-Ag/ITO multilayer films.

<table>
<thead>
<tr>
<th>Elemental (weight %)</th>
<th>O</th>
<th>Al</th>
<th>Ag</th>
<th>In</th>
<th>Sn</th>
<th>Si</th>
<th>Total %</th>
</tr>
</thead>
<tbody>
<tr>
<td>As-deposited</td>
<td>12.31</td>
<td>0.31</td>
<td>5.63</td>
<td>11.01</td>
<td>1.23</td>
<td>69.51</td>
<td>100</td>
</tr>
<tr>
<td>300 °C</td>
<td>11.28</td>
<td>0.39</td>
<td>5.76</td>
<td>11.07</td>
<td>1.28</td>
<td>70.22</td>
<td>100</td>
</tr>
<tr>
<td>400 °C</td>
<td>10.85</td>
<td>0.48</td>
<td>5.78</td>
<td>11.12</td>
<td>1.30</td>
<td>70.47</td>
<td>100</td>
</tr>
<tr>
<td>500 °C</td>
<td>10.70</td>
<td>0.51</td>
<td>5.81</td>
<td>11.15</td>
<td>1.32</td>
<td>70.51</td>
<td>100</td>
</tr>
</tbody>
</table>
The multilayer and single ITO (at 500°C) films electrical characteristics in terms of electrical resistivity sheet resistance, carrier concentration $n$ and Hall mobility $\mu$ are determined using a four-point probe and Hall-Effect systems respectively. Table 4 shows the electrical resistivity of ITO/Al-Ag/ITO and ITO (at 500°C) as a function of post-annealing temperature. From Table 4, it is understood that the resistivity value decreased when the post-annealing temperature is increased. The insertion of Al-Ag bilayer metal films between top and bottom ITO layers drastically reduced the resistivity (ITO resistivity $\leq 10^{-4}$ $\Omega$ cm) even without heat treatment (Guillén & Herrero, 2011). The resistivity value decreases further after post heat treatment with multilayer films showing the least electrical resistivity of $2.70 \times 10^{-5}$ $\Omega$ cm at 500°C. Furthermore, sheet resistance $R_{st}$ of multilayer and single ITO films, as a function of post annealing temperature were evaluated as shown in Table 4. The as-deposited multilayer film shows a lower sheet resistance of 7.82 $\Omega$/s compared to single ITO film at 500°C (13.20 $\Omega$/s). The $R_{st}$ further decreases with increasing annealing temperature. The significant reduction in the $R_{st}$ is due to insertion of low resistivity metal films (of order $10^{-6}$ $\Omega$ cm) between the top and bottom ITO layers. Even though, the metal films insertion resulted in decrease in optical transmittance, post heat treatment at moderate temperature has reported an improvement in the optical transmittance due to reduction of films structural defects as reported by Ali et al., (2014) and Isiyaku et al., (2020). This improvement narrows the transmittance-conductivity trade-off of the films. Also, multilayer films $R_{st}$ can be obtained mathematically as expressed in Equation 1.

$$\frac{1}{R_{MSR}} = \left(\frac{1}{R_{M1}} + \frac{1}{R_{M2}} \right) + \frac{2}{R_{ITO}}$$

where $R_{MSR}$, $R_{M1}$, $R_{M2}$, $R_{ITO}$ are sheet resistance for multilayer, first interlayer metal, second interlayer metal and ITO respectively. A minimum $R_{st}$ of 3.92 $\Omega$/sq for multilayer film at 500 °C and 13.2 $\Omega$/sq single ITO film at 500 °C are obtained. Furthermore, significant improvement in carrier concentration and Hall mobilities are observed with respect to post-annealing temperature. From Table 4, it is observed that the multilayer films carrier concentration and mobilities are considerably higher than that of single ITO films. The highest achieved carrier concentration of multilayer films at 500°C are $16.1 \times 10^{21}$ cm$^{-3}$ whereas that of ITO achieved at same temperature was $7.1 \times 10^{21}$ cm$^{-3}$. The ITO/Al-Ag/ITO multilayer films carrier mobilities increased from 22.2 cm$^2$/Vs to 41.4 cm$^2$/Vs at 500°C while single ITO at 500°C stands at 10.2 cm$^2$/Vs. Group of scientists such as Meshram et al., (2015), Kumar et al., (2015) and Ali et al., (2014) have reported similar findings such that the increase in the effective charge carrier for conduction is attributed to improved structural ordering that led to grain growth and reduction in electron scattering as a result of heat treatment effects. Oxygen vacancy creation which donates two electrons for each vacancy during annealing also contributed to the increase in carrier concentration and mobility (Balasundaraprabhu et al., 2009).
It is noted equally, that the observed grain growth and enhanced crystallinity after post annealing process which equally overcome the impact of scattering light effect can be accredited to the increased carrier mobilities (Isiyaku et al., 2021b). The single ITO film’s low carrier mobility even at 500°C can be connected to the films thickness size (Isiyaku et al., 2021a). In general, films electrical resistivity is found to be inversely proportional to carrier concentration and mobility as shown in Equation 2.

\[ \rho = \frac{1}{n\mu} \]  

(2)

charge of the carriers is represented by e sign. The results from Table 4 confirms this relation (Equation 2), which indicates that the electric resistivity decreased with increasing carrier concentration and mobility respectively. Lowest resistivity value of 2.70 × 10^{-5} \text{Ω} \text{cm} at 500°C was obtained at highest n and \( \mu \) values of 16.1 x 10^{-3} \text{cm}^2/\text{Vs} and 41.4 cm/Vs. In this regard, the films high electrical conductivity may now be linked to low resistivity as well as high carrier mobility and this result in hitch-free flow of uniform electrical current across the films into next various layers of the low resistance optoelectronic devices (Ali et al., 2016). In all-purpose, it can be observed that the inclusion of metal thin films together with heat treatment (strong films crystallization) effectively enhances the surface topo-morphological and electrical properties which in turn improved the performance of the films.

**CONCLUSION**

Topo-morphological and electrical properties of ITO/Al-Ag/ITO/p-Si multilayer films prepared by RF and DC sputtering techniques are investigated before and after post annealing treatment at 300-500°C respectively. The insertion of Al-Ag in-between the ITO top and bottom films together with post annealing treatment significantly improved the films topo-morphological as well as electrical properties correspondingly. The multilayer surface topology parameters increased with increase in post-annealing temperature. The as-deposited film exhibited a smooth surface topology and the smoothness improved with increasing post annealing temperature. Sharp crystallites peaks were obtained by all the films with smaller peaks diffusing and recombining to form larger films with increasing post-annealing temperature. Highest multilayer films RMS roughness and grain size of 1.71nm and 61.9 nm are obtained compared 1.22 nm and 42.1nm for single ITO films both 500°C respectively. Also, the films surface morphology of as-deposited film shows smooth and continuously connecting surface morphology with dense particles evenly distributed throughout the film surface. Compared to as-deposited film, surface smoothness increased with increase in temperature with films annealed at 400°C and 500°C showing an increasing surface smoothness property. This is attributed to the heat absorption which causes the particles to expand thereby narrowing the grain boundaries and subsequently improve the surface smoothness. The topological AFM and morphological FESEM results are in good agreement. The inclusion of Al-Ag metals interlayer films drastically reduced the films resistivity even without heat treatment. The resistivity value decreases further after post heat treatment with multilayer films at 500°C. showing the least electrical resistivity of 2.70 × 10^{-5} \text{Ω} \text{cm}. highest carrier concentration and carrier mobility of 16.1 x 10^{-3} \text{cm}^2/\text{Vs} and 41.4 cm/Vs are achieved at 500°C, the films good surface topo-morphology, low resistivity and high carrier concentration and mobility properties which in turn correspond to good microstructural and high electrical conductivity properties can be considered as favorable candidate in low resistance optoelectronic applications, since it can result in hitch-free flow of uniform electrical current across the films into next various layers of the low resistance optoelectronic devices.

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