High figure of merit of the post-annealed Ti/Al/ITO transparent conductive contacts sputter deposited on n-GaN

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1. Introduction

Transparent conductive contacts (TCC) is very critical in determining the functional success of optoelectronic devices such as GaN-based light emitting diodes (LED) and laser diodes (LD). High electrical resistivity of contacts may reduce the performance of the light emitting devices, whereas low optical transmittance will reduce optical power output from the devices. Metal contacts such as Ti/Au [1], Ti/Cr/Al [2], Cr/Al [3] and Ti/Al [4] have been reported to have good ohmic contacts to n-GaN layer of the semiconductor devices. However in order to produce high external quantum efficiency, the output light from the active region to the outer space (such as air) need to be taken into detail consideration. Most of the multi-layer metal contacts show very low optical transmittance characteristics. The opaque properties of the metal contacts to the light make it unsuitable to be used as a window and current spreading layer on light emitting devices.

Transparent conductive oxides such as indium tin oxides (ITO) offer a good solution as a contact and current spreading layer on light emitting devices [5]. This is due to its unique properties of good electrical conductivity and has high optical transmittance in visible spectrum. Kim et al. reported on the ITO/n-GaN of LED structure which achieved electrical contact resistivity and optical transmittance at 460 nm of 3.78 × 10⁻⁴ Ω-cm and 98.6%, respectively after annealing in N₂ ambient at 300 °C for 7.5 min [5]. Since ITO has electrical conductivity lower than the metal contacts, inserting very thin metal layer between the n-GaN semiconductor and top ITO layer can improves the conductivity of the ITO. However, most of the as-deposited ITO is amorphous in nature which causing lower electrical conductivity and low light transmittance characteristics [6].

Post-annealing of the as-deposited TCC samples can improves the structural and morphological properties of the TCC and consequently enhanced the electrical and optical characteristics [7]. Koteswara and Rajagopal reported on the increasing of optical transmittance of ITO/n-GaN of more than 90% after annealing at 500 °C for 1 min under nitrogen ambient [8]. They also reported on the improvement of the polycrystalline and surface morphological quality after the annealing process. In this report, ITO-base transparent conductive contacts will be deposited on n-GaN with the insertion of Ti/Al thin metal under-layer. Post-annealing will be
performed on the TCC at designated parameter. Analysis based on the structural, morphological, electrical and optical will be perform on the TCC to determine its performance.

2. Methodology

A 2 μm thick commercial n-GaN template grown on sapphire substrates with carrier concentration of \(-10^{18}\) cm\(^{-3}\) is used in this study. Transparent glass substrate is also used in conjunction with the n-GaN in order to characterize the optical characteristics of the TCC samples. Ti thin metal films is deposited on the substrates by DC magnetron sputtering system at room temperature followed by Al thin metal films with individual thicknesses of 5 nm. After that, ITO thin films with thickness of 80 nm is deposited on the Ti/Al under-layer by RF magnetron sputtering. Based on the initial optimization study on post-annealing parameter including annealing temperature and duration, the Ti/Al/ITO TCC samples then were undergone post-annealing at 600 °C in N\(_2\) ambient for 15 min.

Sample analysis were conducted based on the structural, morphological, electrical and optical characteristics of the samples. X-ray diffraction (XRD) 2-theta analysis was performed on the TCC sample to determine the crystalline characteristics. Energy dispersive x-ray spectroscopy (EDXS) is used to investigate the elemental characteristics of the TCC samples. Surface morphological of the samples were scanned by atomic force microscope (AFM) and field emission scanning electron microscope (FESEM). The electrical studies in term of electrical resistivity, carrier mobility and carrier concentration were performed with the aid of Hall effect system. Ohmic characteristics was examined by current-voltage (I–V) system whereas specific contact resistance was determined by transfer length method (TLM). The optical studies was conducted using UV–visible optical spectrophotometer. The figure of merit is calculated based on the electrical resistance and the optical transmittance characteristics of the TCC samples.

3. Results and discussion

XRD was employed to identify the phase evolution which resulted from the interfacial reaction of the TCC layer with the n-GaN after deposition and post-annealed as shown in Fig. 1. Significant ITO (222) and (411) peaks were observed at 30.67° and 37.67°, respectively for the 600 °C post-annealed samples. Very high intensity of GaN (002) and (004) peaks were observed at 34.58° and 72.89°, respectively. AlN is used as a buffer layer for the upper n- GaN template and the peaks can be clearly observed at 35.98° and 76.33° corresponding to the AlN (002) and (004), respectively. The TiN (111) and (222) peaks have the same 2-theta position as the AlN since they have almost similar lattice parameter, thus distinguishing between them using the lattice spacing measurement is challenging. The formation of TiN phase which is associated with the extraction of N by Ti from GaN upon post-annealing, creates high density of N vacancies. This excess of carriers resulting in the creation of donor states near the metal-semiconductor interface, pinning the Fermi level near the conduction band and lowering the Schottky barrier height (SBH), thus improving the carrier tunneling through the TCC-n-GaN layer. The presence of the TiN layer at the interface of the TCC-n-GaN is believed to promote the formation of low resistance ohmic contact, because of its lower work function (3.74 eV) with respect to pure Ti (4.33 eV). The use of Al layer also helps in lowering the electrical resistance of the TCC-n-GaN due to its low electrical resistivity and low work function characteristics.

Table 1 shows the elemental composition in wt% of the Ti/Al/ITO TCC layer on n-GaN scanned by EDXS. From the observation results, N and O were reduced after post-annealing as compared to the as-deposited samples. However the wt% of the metal based materials including Al, Ti, Ga, In and Sn were increased after the post-annealing process. Ga shows the highest composition as it serves as a base material for GaN. Sn has low composition as it is used as a dopant species to the ITO. In addition, the low composition of Ti is also due to the very thin under-layer of Ti metal deposited on the n-GaN. Fig. 2 shows the plotted elemental distribution of the as-deposited and post-annealed samples.

Fig. 3 shows the 3D AFM images of the Ti/Al/ITO TCC layer on n-GaN scanned over an area of 1 × 1 μm\(^2\). From the observation results, it is clearly shown that the crystalline sizes of the as-deposited sample are smaller than the post-annealed samples. Measurement by NanoScope Analysis reveals the sizes of the as-deposited and post-annealed samples are 53 nm and 103 nm, respectively. Surface roughness affects the uniformity of the TCC and often creates undesirable properties of the contacts. Therefore good quality surface roughness is essential to produce a good performance of TCC. Further analysis showed that the surface roughness \(R_s\) of the as-deposited and post-annealed samples are 3.61 nm and 1.84 nm, respectively. During post-annealing, the crystallites of the TCC layer were expanding due to the absorption of heat from surrounding, resulting in agglomeration of the crystallites and consequently improving the surface roughness. Moreover this improved surface profiles helps in improving the optical transmittance of the TCC layer as well as the electrical conductivity by reducing the light and carrier scattering by grain boundary effects.

Further morphological analysis was performed by FESEM using 15 kV high voltage and 300000 × magnifications as shown in Fig. 4. The surface morphology of the as-deposited sample is smooth with small grains distributed throughout evenly on the surface. After post-annealing at 600 °C, the surface becomes smoother as compared to the as-deposited sample. Although Ti/Al thin metal under-layer is easily deformed when exposed to the moderate temperature (600 °C) environment, but the top ITO layer helps to protect the surface changes due to the thermal changes besides produce a smoother surface morphology of the TCC layer. This FESEM results are in good agreement with the surface roughness

![Fig. 1. 2-theta XRD of the as-deposited and the post-annealed Ti/Al/ITO TCC on n-GaN.](image-url)
The electrical properties of the Ti/Al/ITO TCC contact layer on n-GaN is shown in Table 2 for the as-deposited and post-annealed sample. The electrical resistivity of the TCC layer decreases after post-annealing at 600°C to $8.607 \times 10^{-5}$ Ω·cm. The mobility of the sample after post-annealing is also increasing one order of magnitude as compared to the as-deposited sample. Although the carrier concentration of the post-annealed sample is lower than the as-deposited sample, the resistivity and the carrier mobility is better than the as-deposited sample. This may be due to the carrier concentration of the post-annealed is just enough to allow easy carrier flow through the TCC-n-GaN as well as improve current conductivity through the entire TCC layer. In addition, the carrier concentration of $\sim 10^{21}$ cm$^{-3}$ is sufficient for good electrical current conduction.

Further analysis on the I–V characteristics of the Ti/Al/ITO TCC layer on n-GaN reveal that the 600°C post-annealed sample has better ohmic behavior than the as-deposited sample as shown in Fig. 5. This allows electrical current to flow easily through the TCC-n-GaN layer without high electrical barrier. The calculated electrical resistance of the Ti/Al/ITO from the I–V measurement for the as-deposited and post-annealed samples are 138.0 Ω and 32.8 Ω, respectively.

Besides the electrical resistivity of the TCC contact with good

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Fig. 2. EDXS properties of the (a) as-deposited (b) post-annealed Ti/Al/ITO TCC layer on n-GaN.

Fig. 3. 3D surface morphological of the (a) as-deposited and (b) post-annealed TCC samples scanned by atomic force microscope.
ohmic characteristics, specific contact resistance is used to describe the contact resistance between the contact and the n-GaN. The specific contact resistance is determined by TLM using metal mask to deposit the TCC layer on the n-GaN surface. Fig. 6 shows the graph of resistance, \( R_p \) versus pad distance, \( L_T \) of the TLM mask. The sheet resistance, \( R_{sh} \) of the semiconductor layer outside the contact region can be determined from the slope of the graph. The specific contact resistance, \( \rho_c \) then is determined from the calculation of the \( \rho_c = R_{sh} \left( L_x/2 \right)^2 \), where \( L_x \) is the graph intercept at x-axis. From calculation, the specific contact resistance of the Ti/Al/ITO on n-GaN after post-annealing is 2.39 Ω-cm². Although the calculated of \( \rho_c \) is higher than other reported value, but it is still acceptable since the TLM mask used in this study has pads distances in mm ranges whereas the pads distances used by other researchers are in μm ranges. As a comparison, there are reports on the metal contact that is using almost the similar TLM metal mask that were used in this study with reported specific contact resistance in the ranges of \( 10^{-4} \text{–} 10^1 \text{ Ω-cm}^2 \) [9].

Fig. 7 shows the optical transmittance characteristics of the TCC for the as-deposited and post-annealed samples. The post-annealed sample shows higher optical transmittance as compared to the as-deposited sample. The transmittance of the post-annealed and the as-deposited samples at 470 nm is 95% and 59%, respectively. The transmittance characteristics for the post-annealed sample is higher (~95%) at blue-green spectrum and reducing and almost constant (~90%) at red-infrared spectrum. The as-deposited transmittance characteristics slightly reducing as the wavelength gets longer. The transmittance is sharply decreasing as the wavelength reaching to ultraviolet region due to the light absorption by the TCC.

It is important to evaluate the efficiency of the TCC multilayer based on their transmittance and electrical properties for appropriate application in optoelectronic devices. Figure of meritallow evaluating the performance of transparent conductive electrodes based on electrical resistance and optical transmittance. FOM for
the as deposited and post-annealed samples at 470 nm wavelength are $2.39 \times 10^{-4}$ $\Omega^{-1}$ and $5.91 \times 10^{-2}$ $\Omega^{-1}$, respectively. FOM of the post-annealed TCC is the highest as compared to the as-deposited samples. Therefore, the post-annealed TCC shows the best efficiency and consequently helps in improving the performance of light emitting devices.

4. Conclusion

The 600 °C post-annealed samples shows better structural, morphological, electrical and optical properties as compared to the as-deposited sample. The measured electrical resistivity and optical transmittance of the Ti/Al/ITO TCC contact is $8.607 \times 10^{-5}$ $\Omega$-cm and 95%, respectively. FOM of the post-annealed samples is determined as $5.91 \times 10^{-2}$ $\Omega^{-1}$, which make it the best candidates as an ohmic TCC layer for light emitting devices application. The improvement in the crystalline quality of the post-anneal TCC helps in lowering the electrical resistivity. Light scattering by grain boundary effects was reduced after post-annealing with the reducing of grain boundary and smoother surface morphology.

Acknowledgement

The support from Universiti Tun Hussein Onn Malaysia, Universiti Sains Malaysia, Universiti Malaya and Ministry of Higher Education, Malaysia are gratefully acknowledged. This research was partially funded by USM RU Postgraduate Research Grant Scheme (RU-PRGS-845016), USM RU Grant, Exploratory Research Grant Scheme (ERGS) ER012-2011A, High Impact Research and University of Malaya Research Grant (UMRG) RG141-11AFR.

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