

Pulsed laser deposition of ITO/AZO transparent contact layers for GaN LED applications

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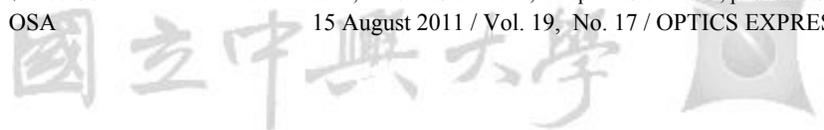
Abstract: In this study, indium-tin oxide (ITO)/Al-doped zinc oxide (AZO) composite films were fabricated by pulsed laser deposition and used as transparent contact layers (TCLs) in GaN-based blue light emitting diodes (LEDs). The ITO/AZO TCLs were composed of the thin ITO (50 nm) films and AZO films with various thicknesses from 200 to 1000 nm. Conventional LED with ITO (200 nm) TCL prepared by E-beam evaporation was fabricated and characterized for comparison. From the transmittance spectra, the ITO/AZO films exhibited high transparency above 90% at wavelength of 465 nm. The sheet resistance of ITO/AZO TCL decreased as the AZO thickness increased, which could be attributed to the increase in a carrier concentration, leading to a decrease in the forward bias of LED. The LEDs with ITO/AZO composite TCLs showed better light extraction as compared to LED with ITO TCL in compliance with simulation. When an injection current of 20 mA was applied, the output power for LEDs fabricated with ITO/AZO TCLs had 45%, 63%, and 71% enhancement as compared with those fabricated using ITO (200 nm) TCL for the AZO thicknesses of 200, 460, and 1000 nm, respectively.

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References and links

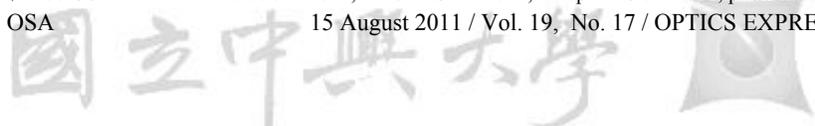
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1. Introduction

High efficiency GaN-based light-emitting diodes (LEDs) are used in a large range of potential applications such as full-color displays, traffic signals, automobiles, solid-state lighting, backlights of liquid-crystal displays, and so on [1–3]. However, the external efficiency of LEDs is mainly limited by the difficulty of light extraction due to the trap of light emitting inside the LED by the total internal reflection at the semiconductor-air interface, which resulted from the refractive index difference of GaN-based materials and air [4]. In order to solve the problem, several methods including surface roughening [5], photonic crystals [6], patterned sapphire substrates [7], and changing the chip shape [8] have been proposed. Additionally, the replacement of poorly transparent (60~70% in the visible region) Ni/Au alloy contact layer by a transparent contact layer (TCL) on p-GaN can also improve the light extraction. Recently, indium tin oxide (ITO) has emerged as one of most promising materials for the TCL in GaN-based LEDs due to its low resistivity and high transparency in the range of visible wavelength [9,10]. Unfortunately, indium is a rare metal with high price, and the low thermal stability of ITO leads to a poor device reliability. Therefore, many indium-free ZnO-based materials have increasingly studied as the alternative TCL in high efficiency LEDs due to their electrical and optical properties similar to ITO, better thermal stability, and smaller lattice mismatch to GaN [11–13]. ZnO-based thin films have been grown by several techniques such as sputtering [14], molecular beam epitaxy [15], metal-organic chemical vapor deposition [16], atomic layer deposition [17], E-beam evaporation [18], and pulsed laser deposition (PLD) [19]. Among these techniques, PLD is the ideal production technology for the growth of epitaxial ZnO-based thin films because it can keep the consistent composition of the original material which is preserved in the interaction and the oxygen plasma created by the pulsed laser is very energetic. Our previous research has reported that the 200-nm-thick Al-doped ZnO (AZO) films with highly (002)-oriented structure grown by PLD at the growth temperature of 100 °C exhibits low resistivity of 233 $\mu\Omega$ -cm and high transmittance above



90% in both visible and UV regions [20]. It indicates that the film has great potential in the TCL on LEDs.

In this work, the ITO (50 nm)/AZO (200-1000 nm) films, which were grown by PLD, were characterized and used as the TCLs on GaN blue LEDs to improve light extraction efficiency. In the ITO/AZO TCLs, the 50-nm-thick ITO film is served as an ohmic contact layer with p-GaN layer. On the other hand, the AZO films with various thicknesses are applied for uniform current spreading, high transparent and thick window layer to achieve the highly efficient LEDs. Moreover, the conventional LED with ITO (200 nm) TCL grown by E-beam evaporation was fabricated as contrasted sample.

2. Experimental

In this study, two types of TCLs were fabricated. First, the ITO (200 nm) TCL was deposited by E-beam evaporation. The target material applied for this experiment was an ITO pellet which consisted of 95 wt % In_2O_3 and 5 wt % SnO_2 . The working pressure and substrate temperature were controlled to be 5×10^{-2} Pa and 250 °C, respectively. Second, the ITO (50 nm)/AZO (200-1000 nm) composite TCLs were grown by PLD (PLD/MBE-2000, PVD products) from the stoichiometric ITO and AZO ceramic targets (99.999% purity) of 3 in. diameter. The ITO target has the same composition used in E-beam evaporation, and the composition of AZO target is 98 wt % ZnO and 2 wt % Al_2O_3 . The growth temperature of ITO and AZO films prepared by PLD was fixed at 100 °C. In the PLD system, the substrates were kept at 80 mm away from the targets, and the substrates were heated using a resistive heater during films growth. A KrF excimer laser ($\lambda = 248$ nm) was employed as an ablation source with 1 Hz repetition rate and the energy fluence was 600 mJ/pulse. As the base pressure was smaller than 5×10^{-8} Torr, the Ar was introduced into the chamber, and the working pressure of films growth was fixed at 1×10^{-2} Torr. The ITO (50 nm)/AZO (200-1000 nm) TCLs were designated as ITO/AZO (200-1000 nm) TCLs.

The LEDs were prepared on c-plane sapphire using low-pressure metal-organic chemical vapor deposition. The LED structures consisted of a 3- μm -thick layer of u-GaN, a 2- μm -thick layer of n-type GaN:Si (n-GaN), 6 pairs of InGaN/GaN multiple quantum well (MQW) active layers, a 100-nm-thick p-type AlGaIn layer and a 0.2- μm -thick p-type GaN:Mg layer. For the device process, a mesa pattern of LED sample was defined with the size of 12×24 mm². Next, the ITO film was fabricated by PLD or E-beam evaporation on the LED. The ITO layer was partially etched in a diluted HCl solution, and the LED structure was etched by means of the inductively coupled plasma with Cl_2/Ar gas to expose the n-GaN layer. Then, the AZO film was grown by PLD at the growth temperature of 100 °C. Finally, the Cr/Au (20 nm/200 nm) was deposited by E-beam evaporation as the n- and p-pad electrodes. The sheet resistances of the samples were measured using a conventional four-point-probe method. The transmittances of TCLs were determined from the N&K analyzer (model: 1280, N and K Tech.). The contact resistances between TCLs and p-GaN layer were evaluated by the transmission-line model (TLM) method [21]. The current-voltage (I-V) characteristics of LED samples were determined using a semiconductor parameter analyzer (Keithley, 2400 sourcemeter), and the output powers of LEDs were measured with a calibrated integrating sphere. Trace-Pro software (commercial light-simulation software based on geometrical optics) is used to simulate the photon trajectories of LEDs with various TCLs to quantitatively analyze light-extraction efficiency. The films thickness and surface roughness were measured by the atomic force microscope (AFM).

3. Result and Discussion

Figure 1 exhibits the AFM images of ITO (200 nm) deposited by e-beam evaporation and ITO (50 nm) deposited by PLD. The root mean square (rms) roughness of the ITO (200 nm) and ITO (50 nm) were 2.6 and 3.8 nm, respectively. It can be expected that the optical and electrical properties of the LED samples were influenced by the surface roughness.

The transmission spectra with wavelength from 200 to 1000 nm of ITO (200 and 1000 nm) and ITO/AZO (200, 460, and 1000 nm) films were measured as shown in Fig. 2. The ITO (1000 nm) film shows the transmittance about 70-75% at the wavelength between 400 and 700 nm. It indicates that the thicker ITO layer is not suitable for TCL in LED. Except for 1000-nm-thick ITO film, all films exhibit the transmittance higher than 85% at wavelength range of 400-700 nm. Transparent thick films with a few microns in thickness, such as 1000-nm-thick ITO in this work, usually have lower optical transmittance because of light scattering from longer optical path. However, we can found that the thicker AZO films still have high transmittance, even for the thickness increased to 1000 nm. This can be attributed to the strong absorption in UV-blue range and a smooth decrease in the extinction coefficient with wavelength for the AZO films, resulting from the band gap of 3.37 eV and its polycrystalline/partially amorphous nature. We also found that the absorption of ITO (50 nm)/AZO (200-1000 nm) almost does not change with the AZO thickness. It can be explained by the previous study by B. Z. Dong *et al* [22]. In the previous study, the carrier concentration of thinnest AZO film (15 nm) is the smallest one, thus the occupied states in valence band are less and cause the small optical band gap. As the film thickness increases to larger than 56 nm, the carrier concentration becomes stable, resulting in the stable absorption and optical band gap. Therefore, we speculate that the carrier concentration of AZO film has become stable as the thickness is above 200 nm, leading to the stable absorption in our work.

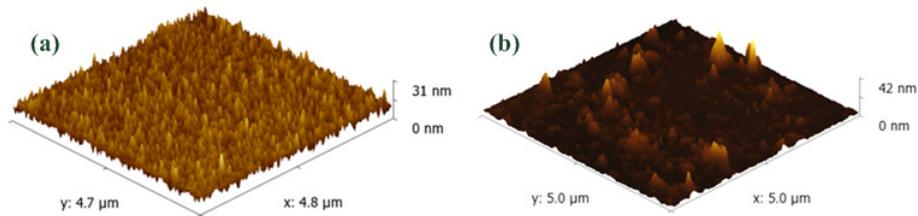


Fig. 1. AFM images of (a) ITO (200 nm) deposited by e-beam evaporation and (b) ITO (50 nm) deposited by PLD.

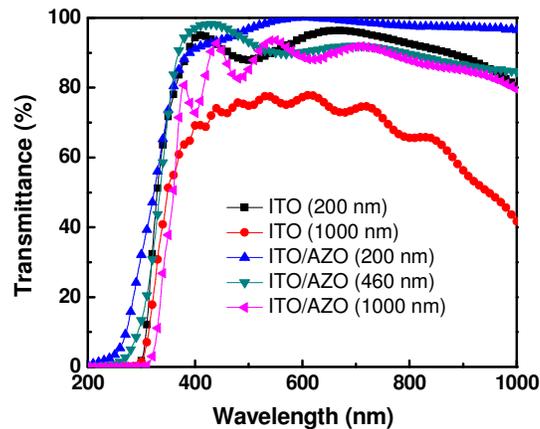


Fig. 2. Transmittance spectra of ITO (200 and 1000 nm) and ITO/AZO films with various AZO thicknesses from 200 to 1000 nm.

Table 1. The Transmittance at Wavelength of 465 nm and Sheet Resistance of ITO/AZO (200-1000 nm), and the Contact Resistance between ITO/AZO (200-1000 nm) and p-GaN Layer Evaluated from TLM

Various thicknesses of ITO/AZO films	Transmittance at 465 nm (%)	Sheet resistance (Ω/\square)	Contact resistance from TLM ($\Omega\text{-cm}^2$)
ITO/AZO (200 nm)	94	28.1	1.33
ITO/AZO (460 nm)	96	10.8	3.00×10^{-1}
ITO/AZO (1000 nm)	90	3.5	1.32×10^{-3}

The transmittance at wavelength of 465 nm and sheet resistance of ITO/AZO (200-1000 nm), and the contact resistance between ITO/AZO (200-1000 nm) TCL and p-GaN layer evaluated from TLM were summarized in Table 1. It is found that the transmittances of ITO/AZO films with various AZO thicknesses of 200, 460, and 1000 nm are 94, 96, and 90%, respectively. The results of sheet resistance show that the resistance of ITO/AZO films decreased with an increase of AZO film thickness from 200 to 1000 nm. The TLM results exhibit that the contact resistances between ITO/AZO TCL and p-GaN are 1.33, 3.00×10^{-1} , and 1.32×10^{-3} $\Omega\text{-cm}^2$ as the AZO thicknesses are 200, 460, and 1000 nm, respectively. It indicates that the contact resistance also reduced as the thickness of AZO layer increased, which results from the decrease in the sheet resistance due to the increment in carrier concentration.

Trace-Pro Simulation results are exhibited in Figs. 3(a)–3(d). Here, a 60 mW power (10000 light rays) was assumed to emit randomly from the MQW active layer. The simulated results reveal that the fluxes and rays are 17.16 mW and 1360 rays, 23.11 mW and 1373 rays, 25.57 mW and 1504 rays, and 33.51 mW and 1899 rays for the LEDs with various TCLs of ITO (200 nm), ITO/AZO (200 nm), ITO/AZO (460 nm) and ITO/AZO (1000 nm), respectively. In our simulation, the epilayer absorption and the effect from the silver cup in the epoxy lamp are not considered; however, the results also can demonstrate the influence of optical scattering at LEDs with various TCLs. The simulation results indicate that the LEDs with ITO/AZO (200-1000 nm) TCLs lead to more light extraction than that with ITO (200 nm) TCL. Moreover, the light extraction of LED with ITO/AZO TCL is increased with an increase of AZO thickness. It can be explained that the thicker AZO window layer can improve the light extraction by allowing additional light to escape through the side facets. Additionally, the larger surface roughness could decrease the refractive index, which improves light extraction of LED. From the results of surface roughness in Fig. 1, it reveals that the more light extraction in the LEDs fabricated with ITO (50 nm)/AZO (200-1000 nm) TCLs than that fabricated with ITO (200 nm) is partially attributed to the larger surface roughness in ITO (50 nm) film.

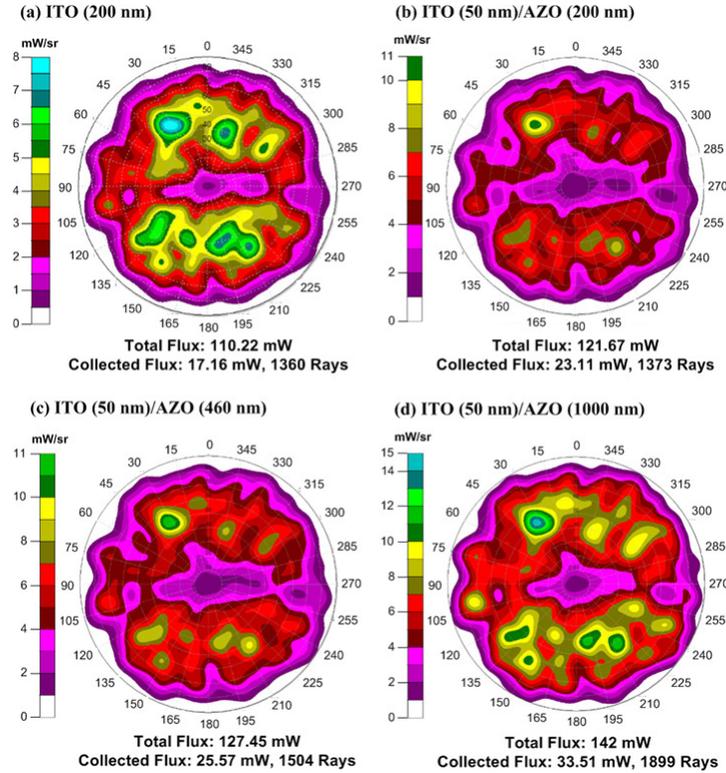


Fig. 3. Trace-Pro simulation taken from GaN-based LEDs with (a) ITO (200 nm), (b) ITO/AZO (200 nm), (c) ITO/AZO (460 nm), and (d) ITO/AZO (1000 nm) TCLs.

Figure 4 shows the current–voltage characteristics of the InGaN/GaN MQW LEDs with the ITO (200 nm) and ITO/AZO (200-1000 nm) TCLs measured at room temperature. It exhibits that the turn-on voltages of LEDs with ITO/AZO (200-1000 nm) TCLs are just a little higher than that with ITO (200 nm) TCL, indicating that the thin ITO film with thickness of 50 nm plays a role of ohmic contact layer successfully. According to the past research, the GaN-based LEDs with ZnO-based TCLs grown on the p-GaN directly led to the poor electrical properties [23,24]. This was attributed to the higher resistivity of ZnO-based films, which caused a schottky contact to p-GaN. However, as a thin ITO layer with lower resistivity was inserted between ZnO-based film and p-GaN, the turn-on voltage was reduced due to the formation of ohmic contact [24]. On the other hand, the larger surface roughness in ITO (50 nm) as shown in Fig. 1 would cause a reduction in the contact area and an increment in contact resistance. This also leads to the higher turn-on voltage in the LEDs fabricated with ITO (50 nm)/AZO (200-1000 nm) TCLs than that with ITO (200 nm) TCL. We also found that the turn-on voltage of LEDs fabricated with ITO/AZO films decreased with increasing the AZO thickness. This could be attributed to the decrease in sheet resistance of the ITO/AZO TCL from 28.1 to 3.5 Ω/\square as the AZO thickness increased from 200 to 1000 nm, as shown in Table 1. The thicker AZO layer leads to a lower series resistance along the path of current spreading from the p-pad electrode, therefore, the lower turn-on voltage can be obtained.

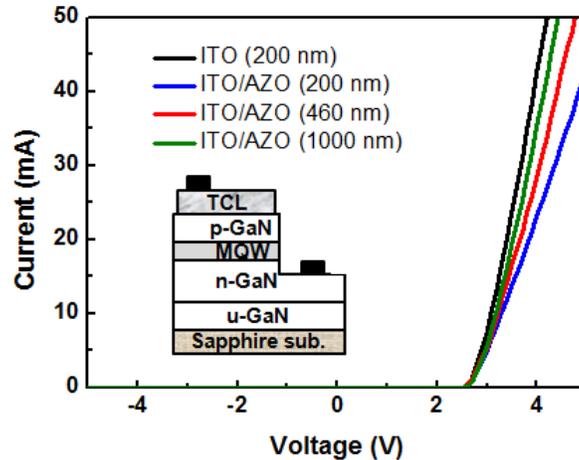
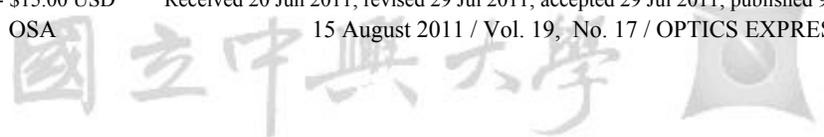


Fig. 4. Current–voltage characteristics of InGaN/GaN MQW LED samples with the ITO (200 nm) and ITO/AZO (200-1000 nm) TCLs measured at room temperature.

Figures 5(a)–5(d) show the optical microscope images of these blue LEDs with various TCLs of ITO (200 nm) and ITO/AZO (200-1000 nm) operated at a low drive current of 0.5 mA. It can be found that the brightness of LED fabricated with ITO/AZO (200 nm) TCL is almost the same as that with ITO (200 nm) TCL. For the LED with ITO/AZO (460-1000 nm) TCL, the higher brightness can be observed, especially as the AZO thickness is 1000 nm. It reveals that the thick AZO window layer can be used to provide current spreading and light extraction of LED sample efficiently, resulting in a great enhancement in brightness.

The performance of light output was measured by calibrating an integrating sphere with a Si photodiode on the package device, which can collect the light emitted in any direction from the LED. Figure 6 plots the light output powers as a function of injection current for the LEDs fabricated with 200-nm-thick ITO and ITO/AZO (200-1000 nm) TCLs. The light output powers of the LEDs with ITO/AZO (200-1000 nm) TCLs are higher than that with ITO (200 nm) TCL in the whole measured currents from 0 to 100 mA. The output powers for the LEDs fabricated with ITO/AZO (200 nm), ITO/AZO (460 nm) and ITO/AZO (1000 nm) TCLs had 45%, 63%, and 71% enhancement compared to that fabricated with ITO (200 nm) TCL at a 20 mA operating current, respectively. The improvement of light output power could be due to the increase of light extraction efficiency by substituting ITO/AZO (200-1000 nm) TCLs for ITO (200 nm) TCL. Furthermore, the increased light output power of LEDs with ITO/AZO TCLs as the thickness of AZO film increased from 200 to 1000 nm can result from the increment of light extraction through sidewalls in thicker AZO layer. Moreover, the improvements in brightness and optical output power of the LEDs with ITO (50 nm)/AZO (200-1000 nm) TCLs were also affected by the larger surface roughness in the ITO films, as discussed in the Fig. 3.



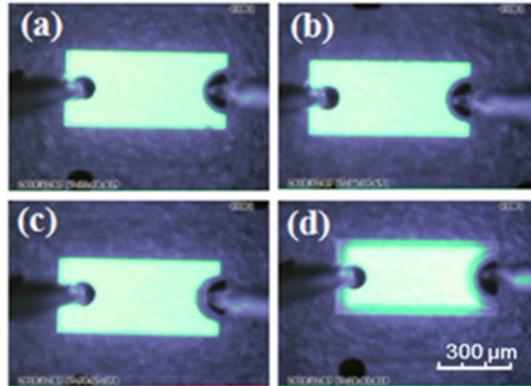


Fig. 5. Photographs of LEDs fabricated with (a) ITO (200 nm), (b) ITO/AZO (200 nm), (c) ITO/AZO (460 nm), and (d) ITO/AZO (1000 nm) TCLs, driven at a certain operation current of 0.5 mA.

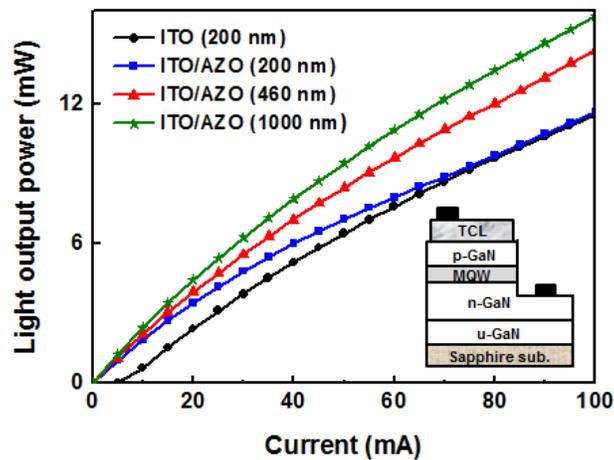


Fig. 6. Light output powers as a function of injection current for the LEDs fabricated with ITO (200 nm) and ITO/AZO (200-1000 nm) TCLs.

4. Conclusion

In conclusion, the ITO/AZO films with various AZO thicknesses of 200-1000 nm were deposited by pulsed laser deposition at 100 °C and utilized as TCLs for the fabrication of bright InGaN/GaN MQWs blue LEDs. The ITO/AZO (1000 nm) film shows high transmittance of 90% at wavelength of 465 nm and low contact resistance of $1.32 \times 10^{-3} \Omega\text{-cm}^2$ to p-GaN. Trace-Pro simulation results reveal that the LEDs with ITO/AZO (200-1000 nm) TCLs bring out more light extraction compared to that with ITO (200 nm) TCL. Moreover, the light extraction of LED with ITO/AZO TCL is increased as the AZO thickness increased. This could be explained that the light extraction is improved through the side facets in thicker AZO film. Compared to the conventional LED with ITO (200 nm) TCL, it was found that the light output power of the LEDs fabricated with ITO/AZO TCLs can be improved, especially as the thickness of AZO layer is 1000 nm.

Acknowledgments

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