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Utilizing Transparent ZnO Thin Film as Permeation-Barrier to Assist Top Emission Polymer Light-Emitting Devices Light Outcoupling and Longevity

Yen-Hsun Lu¹, Yung-Hsin Liao¹, Cheng-Yang Shih¹, Yu-Chi Huang¹, Kou-Chen Liu¹

1. Institute of Electro-Optical Engineering, Chang Gung University, Taiwan, +886-3-2118800

1. Introduction

The full transparent polymer light-emitting diodes (PLEDs) emit light from both surfaces of devices, which have attracted much attention as the next-generation displays [1] such as wind shield [2]. However, these kind devices need to carefully reconsider traditional encapsulation structure and technique. The encapsulation layer using in top emitting surface not only provides a hermetic function [3] but also serves as refraction index matching layer to increasing light output[4]. According to reported, the inorganic layer was transmittance of 85~90% that SiNx, SiOx, SiOxNy, AlOx, and Al₂O₃: N films were generally employed as inorganic passivation layers for encapsulation of organic devices [5-7]. However, the above-mentioned inorganic films show lower refraction index than ZnO thin film. In this study, we reported the ZnO used as a passivation layer protected PLEDs against oxygen and moisture permeation. Using a ZnO layer as inorganic passivation layer was not reported before. It is noteworthy that the ZnO has high transmittance in the wavelength 490-540 nm region is up to 95% and the reflective index ($n=2$) of ZnO can easily match to conducting oxide such as IZO or ITO to improve internal light coupling out. A PLED device utilizing ZnO/UV-curable resin as encapsulation layer and their electrical characterization, luminance property, and lifetime have been detail studied in this paper.

2. Experiment

Organic layers were deposited by spin coater onto a glass substrate coated with a patterned indium-tin-oxide (ITO) electrode. A hole injection layer PEDOT:PSS was spin coated onto ITO glass substrate and baked in atmosphere at 120°C for 15min. Next, the active luminescent polymer film PFO was spin coated onto PEDOT:PSS layer, and baked in glove box at 120°C for 30 min. Thereafter, the samples were transferred into thermal evaporation chamber that evaporated Ag interlayer (1nm). Then, the IZO cathode was deposited by dc-sputtering 50 W at room temperature (~ 120 nm). Then the reference device passivated with glass lid that a thin UV-curable resin was applied from a syringe around the edge of the glass adhesively and exposed to UV light 120 seconds. Another passivation layer was ZnO/UV-curable resin directly passivated on the surface of the device by rf sputter and exposing UV-curable resin after coating, respectively. All measurements were carried out at room temperature under atmosphere.

3. Result and discussion

The X-ray diffraction (XRD) spectra show in Fig. 1 for ZnO, ITO and IZO thin films deposited on glass at room

temperature. The crystallinity is demonstrated in the X-Ray Diffraction measurements. The diffraction patterns of the ZnO thin film clearly displays a highly ordered structure with the distinctive peak at $2\theta=34.24^\circ$. In addition, the XRD measurement of the ITO film indicates more crystalline than IZO film. This data can explain our experimental works that a passivation layer of crystalline ZnO cannot directly deposit onto the ITO cathode. To prevent ZnO film crack, a thin Al₂O₃ layer has to be inserted between a ZnO and ITO layer. This thin Al₂O₃ layer cause light emitting from top surface lessening. On the contrary, a ZnO passivation layer aptly places onto the IZO cathode without any film delaminating and decreasing light output from top surface. In Fig. 2, the PLED device with IZO cathode encapsulated with glass and the ZnO/UV-curable resin films were examined by the current density-voltage (I-V) characteristics. Both two devices show similar electrical behavior for instance turn on voltage and leakage current. According to Kim et al. reported [8], the effect of sputtering damage can be observed from the leakage current at reverse bias. However, in our experiment data, all devices keep the same low leakage current density at reverse bias. It can be explained that one more process of ZnO layer sputtering does not cause further damage. Fig. 3 displays the total brightness and current efficiency obtained by summation of the top and bottom light outputs of the full transparent PLEDs. The light intensity increases linearly with current density. This PLED device encapsulated with ZnO/UV-curable resin has less current efficiency. In table 1, compared with the device encapsulated by glass, the light intensity emitting from top side illustrate 10% higher than that of the device encapsulated with ZnO/UV-curable resin but the light intensity emitting from bottom side indicate only 5% higher than that of the device encapsulated with ZnO/UV-curable resin. The difference of luminance emitting from bottom side of two devices can attribute to UV light damaged PFO layer during UV-curable resin curing process. However, the luminance emitting from top side difference clearly result from UV-curable resin layer absorption 10% light that consist of the result shown in Fig. 4. The normalized EL spectra of the PLEDs passivated with a ZnO/UV-curable resin films and reference device have been measured at current of 1 mA as shown in the Fig. 5. The EL spectra of the top and bottom side from both devices demonstrate almost same characteristic. This result indicates that the addition passivation layer do not influence the EL characteristic of the device. It means that the encapsulation layer will not produce series micro cavity effect. The rate of degradation with difference encapsulated layer for the full

Corresponding Author

K.C. Liu, TEL: +886-3-2118800 ext: 3152 E-mail: jacobliu@mail.cgu.edu.tw

transparent PLED devices was shown in Fig. 6. Total three devices were used to realize the encapsulated layer effect. The first one without any encapsulated layer has very short lifetime and sharply decreasing in the luminance. The device performance seriously decay related to organic layers direct intrusion by moisture and oxygen and resulting larger operating voltage ($\sim 8.8\text{V}$). The second device encapsulated a glass (reference device) and the third device passivated with ZnO/UV-curable resin both show similar life time approximately 100 hours in atmospheric condition under dc constant current density of 6.6 mA/cm^2 (an initial luminance of 190 cd/m^2) and operating voltage kept around 7.5V . This indicates that using ZnO/UV-curable resin as a passivated layer has the same capability to prevent oxygen and moisture permeation. Fig. 7 shows optical images of the electroluminescence with time for all devices. We can clearly find the dark spots were formed after two hours in Fig. 7(a). The non-encapsulated device shown the moisture or oxygen permeation progress through the edge structure and the performance was poor when the device was stored in air condition. The device was glass encapsulated shown no dark spots formation after 100 hours in Fig. 7(b). However the ZnO/UV-curable resin encapsulated device, the pixel has been kept almost clear over 100 hours as shown in Fig. 7(c). This observation is consistent with our lifetime results.

4. Conclusion

In summary, we demonstrated the ZnO/UV-curable resin passivation layer which could effectively protect the device that showed similar electrical behavior to the glass encapsulated device, indicating that its fabrication process for forming the passivation layer did not influence the performance of the device apparently. The lifetime of both devices was almost same and the optical images of the electroluminescence with time did not find dark spots formed. However, ZnO/UV-curable resin (inorganic/organic multilayer) performs the characteristics of flexible and light which develop the applications of PLEDs in the field of flexible flat panel displays.

Reference

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Corresponding Author

K.C. Liu, TEL: +886-3-2118800 ext: 3152 E-mail: jacobliu@mail.cgu.edu.tw

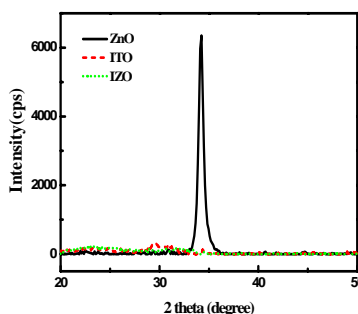


Fig.1. XRD pattern of ZnO,ITO and IZO on a glass substrate.

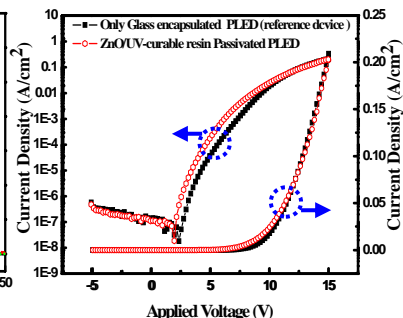


Fig.2. I-V characteristics of only encapsulated glass PLEDs and ZnO/UV-curable resin passivated PLEDs.

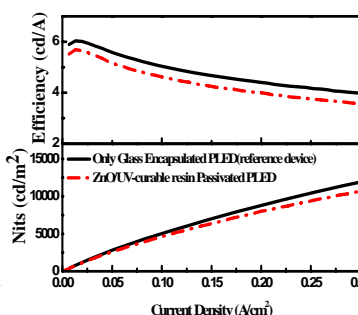


Fig.3. L-I characteristics and efficiency of only encapsulated glass PLEDs and ZnO/UV-curable resin passivated PLEDs, which were measured from top and bottom side.

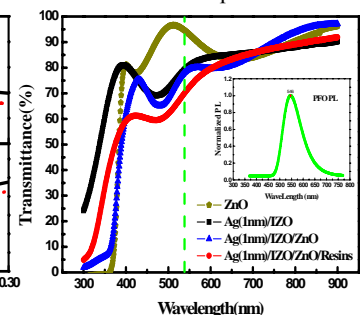


Fig.4. The transmittance of reference cathode and encapsulated passivation layer with ZnO/UV-curable resin on a glass. The inset is the PFO PL spectra.

Table 1. The composition of the light from top and bottom side. Unit: cd/m^2 .

Sample Structure	0.05A/cm ²		0.3 A/cm ²	
	TOP	BOTTOM	TOP	BOTTOM
Encapsulated Glass(reference device)	1444	1510	5810	6130
Encapsulated Passivation Layer Device	1240	1490	4860	5850

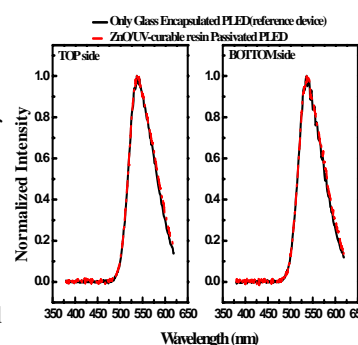


Fig.5. The comparison of the normalized EL spectra of the TEPLDs passivated with ZnO/UV-curable resin and reference device which were measured from top side and bottom side.

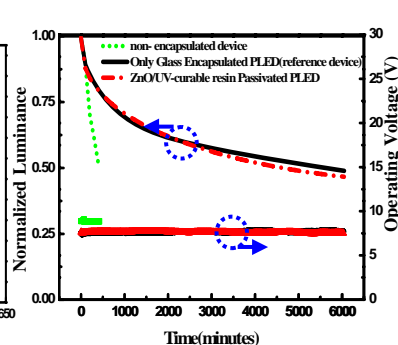


Fig.6. The comparison of the normalized luminance and operating voltage vs operating time of PLEDs passivated with ZnO/UV-curable resin, non-encapsulated device and a reference device.

Fig.7. Photographs of the emitting areas of the devices (a) non- encapsulated device, (b) encapsulated with glass lid (reference device), and (c) passivated with ZnO/UV-curable resin.

