

Effects of Hole Transport and Injection Layers on the Efficiency of Flexible Organic Light-emitting Diodes

Fa-Ta Tsai, Ching-Kong Chao, Sheng-Da Cai, Rwei-Ching Chang

Abstract—The efficiency of organic light-emitting diodes (OLEDs) strongly depends on the mobility of electric holes in the organic materials. To improve the performance of flexible OLEDs, various thicknesses of hole injection layers and hole transporting layers in the flexible OLEDs were studied in this work. The PEDOT:PSS hole injection layer was deposited on the ITO coated PET substrate by spin coating at the first, and then deposited the NPB hole transporting layer, Alq₃ emitting layer, and aluminum cathode by thermal evaporation. The OLED component was covered with a PET film and packaged by UV glue in the final. The luminance, chromaticity coordinate, and life time of the flexible OLEDs were tested, where the thickness effects of the PEDOT:PSS and NPB on the OLED performance were also studied. The result showed that the flexible OLED stacked as PET /Al 200 nm /Alq₃ 100 nm /NPB 40 nm /PEDOT:PSS 130 nm /ITO /PET had the best performance with 2640.7 cd/m² at 10 V.

Keywords— OLED; flexible; hole injection layer; hole transport layer.

I. Introduction

Due to the advantages of high brightness and contrast, lower power consumption, wider viewing angle, faster response time, easy color tuning, thinner, and lighter, organic light emitting diodes (OLEDs) draw great attention. OLED displays based on glass substrates have been used in smart phones, smart pads, and TVs. Moreover, flexible OLEDs have attracted much attention because of high mechanical flexibility [1-3]. However, to achieve commercial standard, the device lifetime and luminance efficiency of the devices still needs to be improved.

To improve the performance of OLEDs, various attempts have been taken in the fabrication processes. One of the promising methods is to increase the efficiency of the electron transportation between the organic materials [4-6]. Due to its high optical transparency and electrical conductivity, ITO is the most widely used transparent anode for OLEDs. The performance of OLEDs is highly affected by the hole injection ability of the ITO anode. Hole injection layers (HIL) are inserted between the hole transporting layer (HTL) and the ITO anode to improve OLED performance by decreasing the energy level between the HTL and anode. Many HILs have been studied [7-13] in which Poly(3,4-ethylene-dioxythiophene)

poly(styrenesulfonate) (PEDOT:PSS) was reported as a good hole injection material.

In this work, the effects of various thicknesses of HIL and HTL on the performance of the OLEDs were studied. Various thicknesses of PEDOT:PSS films were deposited on ITO PET substrates by spin coating. And then, the other organic emitting layers were deposited on the top of the PEDOT:PSS films by thermal evaporation to finish the OLED devices. The luminance of the OLEDs with various HTL and HIL thicknesses were tested.

II. Specimen Preparation

The structure of the OLED is shown in Fig. 1, where a flexible PET substrate, an ITO anode, a PEDOT:PSS (Clevios AI4083) hole injecting layer (HIL), a NPB hole transporting layer (HTL), an Alq₃ emitting layer (EL), an aluminum cathode, and a PET cover were stacked in sequence. The ITO-coated PET substrate with size 300×200×0.175 mm was treated by standard cleaning procedure including ultrasonic bath and oxygen plasma before deposition. To fabricate the OLED, a PEDOT:PSS HIL was deposited on the ITO/PET substrate by spin coating at the first. The film thickness was controlled by the spin speed, where 110, 130, and 150 nm was spun with 6000, 5500, and 5000 rpm for 30 sec, respectively. The specimen was followed by drying process at 70 °C for 10 min. And then, the NPB HTL and Alq₃ EL were thermally evaporated with 1 Å/s deposition rate. And, an aluminum cathode was successively evaporated. Finally, the device was covered with a PET substrate, packaged by UV glue, and cured by UV light for 1 hr. Various thicknesses of the PEDOT:PSS HIL, NPB HTL, and Alq₃ EL were considered.

The thicknesses of each layer were confirmed by a Kosaka ET4000 surface profiler. The luminance and chromaticity of the OLEDs is measured by UV/VIS/NIR spectrophotometer (BWTEK BTC112) in the ambient air at room temperature.

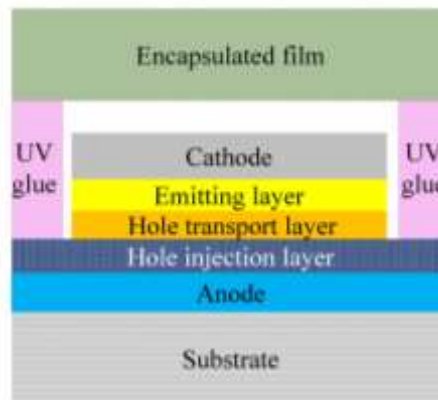


Figure 1. Structure of OLED.

Fa-Ta Tsai, Ching-Kong Chao
Dep. of Mechanical Engineering, National Taiwan University of Science and Technology, Taiwan

Sheng-Da Cai, Rwei-Ching Chang
Dep. of Mechanical and Computer-Aided Engineering, St. John's University, Taiwan

III. Results and Discussion

The luminance of OLEDs at applied voltage 0 ~ 10 V are shown in Fig. 2, where the Alq₃ EL and PEDOT:PSS HIL thicknesses are fixed at 100 and 130 nm, and three thicknesses of the NPB HTL, 20, 40, 60 nm, are considered. The results show that the OLEDs work as the applied voltage higher than 4 V, and the luminance increases quickly as the voltage increases. The results also show the case of 40 nm NPB presents the highest luminance 2640.7 cd/m² at 10 V, the case of 60 nm NPB is 1948.5 cd/m², and 20 nm NPB has the lowest luminance 1752.6 cd/m² at 10 V. It indicates the optimal thickness arrangement of HIL, HTL, and EL in the OLED is needed to improve the electron transportation by decreasing the energy gap between the stacked layers.

Fig. 3 shows the corresponding images of OLEDs at 10 V, where NPB is (a) 20, (b) 40, and (c) 60 nm with fixed Alq₃ and PEDOT:PSS. Each luminant area is 2×2 mm, which indicates the case (b) with 40 nm NPB presents the highest luminance.

Fig. 4 shows the corresponding current density of OLEDs at applied voltage 0 ~ 10 V. It shows the OLED with 40 nm NPB presents the highest current density, and that with 20 nm NPB is the lowest. The results of current density are consistent with the luminance results shown in Fig. 2.

The life time of the OLEDs are shown in Fig. 5, where the OLED with Alq₃ 100 nm/NPB 40 nm/PEDOT:PSS 130 nm is discussed. Fig. 5 compares the luminance of the OLEDs between 0 hour and after 24 hours usage. It shows the luminance is 2640.7 cd/m² @10 V at the begin (0 hour), and the luminance decays to 1860.9 cd/m² @10 V after 24 hours usage. The result indicates the luminance decays 70.47% after 24 hrs.

The luminance at 10 V and chromaticity coordinate for the above three cases are listed in Table 1. It illustrates the case (a) has the lowest luminance 1752.6 cd/m² at 10 V, and case (b) presents the highest 2640.7 cd/m². It also indicates the chromaticity coordinate of case (b) is (0.34, 0.49), and cases (a) and (c) are (0.33, 0.51), which are consistent with the original color of the Alq₃ emitting layer.

For various thicknesses of PEDOT:PSS HIL, NPB HTL, and Alq₃ EL, similar patterns of luminance, current density, and luminance decay were found. In summary, the luminance of the OLEDs at 10 V with various thicknesses of PEDOT:PSS, NPB, and Alq₃ are listed in Table 2. The luminance wildly ranges from 121 to 2640 cd/m², which indicates the luminance of OLEDs strongly depend on the thickness of the organic layers. It illustrates that the OLEDs with 130 nm PEDOT:PSS and 100 nm Alq₃ presents the higher luminance than other case, where 40 nm NPB is the higher one than the other thickness. Overall, the combination of OLED with 130 nm PEDOT:PSS, 100 nm Alq₃, and 40 nm NPB presents the highest luminance of 2640.7 cd/m².

The summary of current density of the OLEDs at 10 V with various thicknesses of PEDOT:PSS, NPB, and Alq₃ are listed in Table 3. The results consist with the luminance distribution listed in Table 2. Similarly, the highest current density is

249.829 mA/cm²@10 V occurred at 130 nm PEDOT:PSS, 100 nm Alq₃, and 40 nm NPB.

The summary of luminance decay of the OLEDs with various thicknesses of PEDOT:PSS, NPB, and Alq₃ are listed in Table 4. In this work, the luminance decay is defined as the ratio of the luminance after 24 hours usage to the luminance at the OLEDs finished moment (0 hour). A narrow range from 65 to 74% are shown, which indicates the life time of the OLEDs is more dependent on the encapsulation than the electric hole mobility.

In general, HIL are inserted between the HTL and the anode to improve OLED performance by decreasing the energy level between the HTL and anode. Moreover, thicker HTL and EL can increase the luminance of OLEDs. However, the device applied voltage will increase as the HTL and EL thickness increase, resulting in higher energy consumption. Therefore, it is needed to design an optimal arrangement of the stacked layer thicknesses of OLEDs.

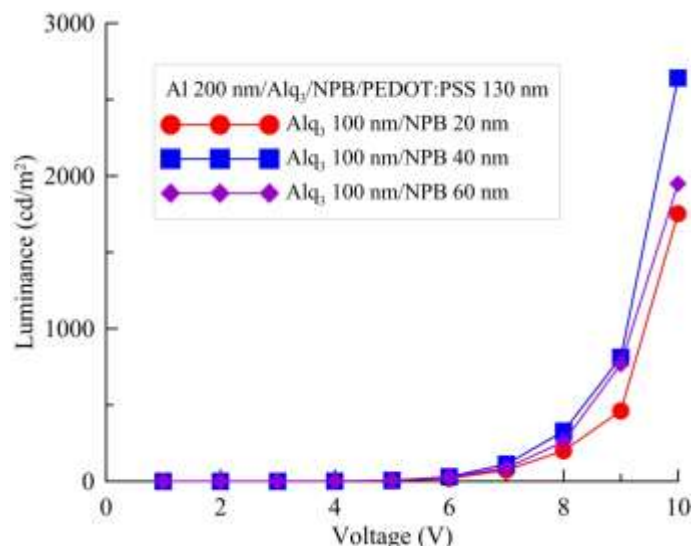


Figure 2. Luminance of OLEDs with various thicknesses of NPB.

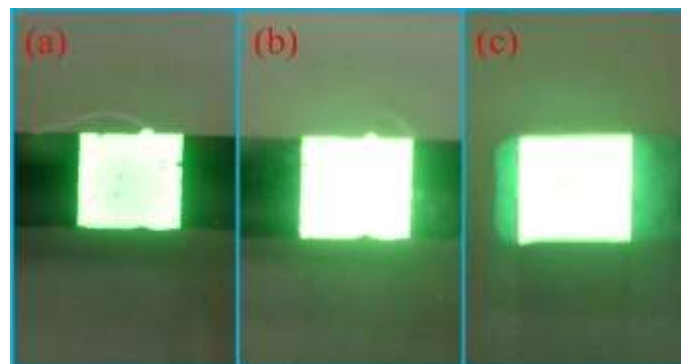


Figure 3. Images of OLED (a) Alq₃ 100 nm/NPB 20 nm/PEDOT:PSS 130 nm (b) Alq₃ 100 nm/NPB 40 nm/PEDOT:PSS 130 nm (c) Alq₃ 100 nm/NPB 60 nm/PEDOT:PSS 130 nm.

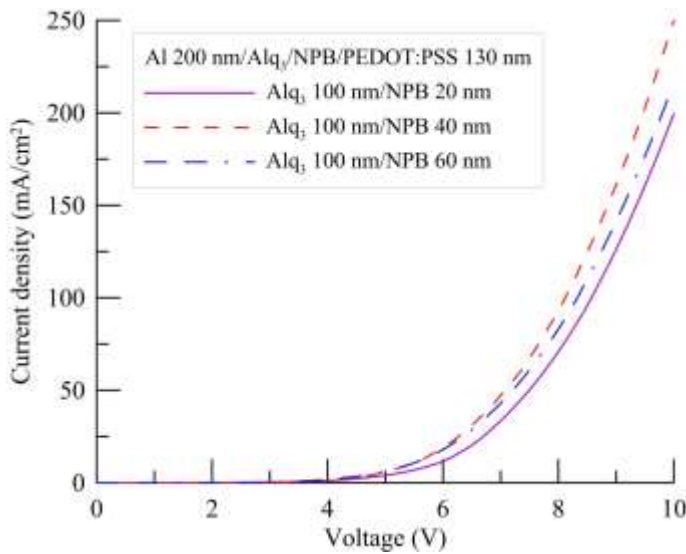


Figure 4. Current density of OLEDs with various thicknesses of NPB.

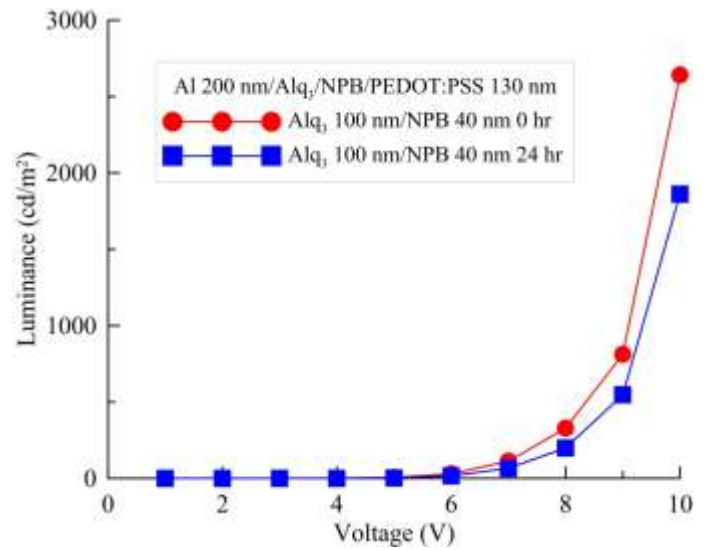


Figure 5. Luminance of OLEDs with Alq₃ 100 nm/NPB 40 nm/PEDOT:PSS 130 nm at 0 and 24 hrs.

Table 1. Luminance and chromaticity coordinate of OLEDs with various NPB thicknesses, where Alq₃=100 nm, PEDOT:PSS=130 nm

| | (a) | (b) | (c) |
|-------------------------|---------------------------------|---------------------------------|---------------------------------|
| OLED type | | | |
| Luminance | 1752.6 cd/m ² @ 10 V | 2640.7 cd/m ² @ 10 V | 1948.5 cd/m ² @ 10 V |
| Chromaticity coordinate | (0.33 , 0.51) | (0.34 , 0.49) | (0.33 , 0.51) |

Table 2. Summary of luminance of OLEDs with various thicknesses of HIL, EL and HTL (cd/m²@10 V)

| PEDOT:PSS HIL (nm) | Alq ₃ EL (nm) | NPB HTL (nm) | | |
|--------------------|--------------------------|--|--|--|
| | | 20 | 40 | 60 |
| 50 | 20 | 121.18 ^{+19.35} _{-21.62} | 173.94 ^{+20.41} _{-16.97} | 127.75 ^{+18.84} _{-18.26} |
| 70 | 40 | 465.75 ^{+10.72} _{-16.08} | 559.45 ^{+25.41} _{-20.49} | 434.59 ^{+21.43} _{-27.85} |
| 90 | 60 | 783.75 ^{+42.71} _{-32.76} | 1097.9 ^{+44.0} _{-52.5} | 651.23 ^{+30.42} _{-54.29} |
| 110 | 80 | 1125.9 ^{+30.5} _{-19.4} | 2123.4 ^{+33.7} _{-43.4} | 1224.3 ^{+31.3} _{-23.0} |
| 130 | 100 | 1752.6 ^{+38.8} _{-29.6} | 2640.7 ^{+52.9} _{-34.5} | 1948.5 ^{+48.8} _{-51.0} |
| 150 | 120 | 1455.0 ^{+32.7} _{-13.2} | 1688.2 ^{+25.2} _{-28.7} | 1210.0 ^{+43.7} _{-54.3} |

Table 3. Summary of current density of OLEDs with various thicknesses of HIL, EL and HTL (mA/cm²@10 V)

| PEDOT:PSS HIL (nm) | Alq ₃ EL (nm) | NPB HTL (nm) | | |
|-----------------------|-----------------------------|---|---|---|
| | | 20 | 40 | 60 |
| 50 | 20 | 9.803 ^{+0.203} _{-0.690} | 12.126 ^{+0.763} _{-0.047} | 6.997 ^{+0.885} _{-0.827} |
| 70 | 40 | 58.484 ^{+2.588} _{-0.637} | 76.251 ^{+0.084} _{-1.049} | 55.764 ^{+0.948} _{-0.567} |
| 90 | 60 | 119.939 ^{+0.871} _{-1.292} | 132.039 ^{+0.588} _{-7.502} | 98.878 ^{+1.174} _{-1.858} |
| 110 | 80 | 134.988 ^{+1.902} _{-0.513} | 237.228 ^{+1.202} _{-1.393} | 145.598 ^{+0.169} _{-1.201} |
| 130 | 100 | 199.404 ^{+1.298} _{-1.962} | 249.829 ^{+0.166} _{-0.784} | 216.024 ^{+0.116} _{-0.474} |
| 150 | 120 | 156.690 ^{+0.847} _{-0.743} | 193.773 ^{+2.359} _{-1.043} | 142.039 ^{+0.668} _{-1.449} |

Table 4. Summary of luminance decay of OLEDs with various thicknesses of HIL, EL and HTL (% @ 10 V).

| PEDOT:PSS HIL (nm) | Alq ₃ EL (nm) | NPB HTL (nm) | | |
|-----------------------|-----------------------------|--------------|-------|-------|
| | | 20 | 40 | 60 |
| 50 | 20 | 66.81 | 65.14 | 69.52 |
| 70 | 40 | 70.95 | 65.75 | 68.18 |
| 90 | 60 | 67.24 | 72.55 | 73.45 |
| 110 | 80 | 70.20 | 71.15 | 65.65 |
| 130 | 100 | 67.35 | 70.47 | 74.73 |
| 150 | 120 | 74.93 | 69.69 | 66.47 |

iv. Conclusions

Effects of HIL and HTL on organic light-emitting diodes coating on flexible PET substrates were studied. Considering various thickness PEDOT:PSS hole injection layer and NPB hole transporting layer, the luminance of the OLEDs were tested. The results showed that the flexible OLED stacked as Alq₃ 100 nm /NPB 40 nm/PEDOT:PSS 130 nm has the best performance with 2640.7 cd/m² at 10 V. To balance the emitting performance and energy consumption, it is needed to design an optimal arrangement of the stacked layer thicknesses of OLEDs. Moreover, the 24 hours luminance tests indicated that the life time of the OLEDs is more dependent on the encapsulation than the electric hole mobility.

Acknowledgment

The support from the National Science Council through Grant NSC 102-2221-E-129-002-MY3 is gratefully acknowledged.

References

- [1] L. Ke, H. Liu, M. Yang, Z. Jiao, X. Sun, Degradation analysis of Alq₃-based OLED from noise fluctuations with different driving modes, *Chemical Physics Letters* 623 (2015), p. 68–71.
- [2] X. Wu, J. Liu, G. He, A highly conductive PEDOT:PSS film with the dipping treatment by hydroiodic acid as anode for organic light emitting diode, *Organic Electronics* 22 (2015), p. 160–165.
- [3] A.R. Cho, E.H. Kim, S.Y. Park, L.S. Park, Flexible OLED encapsulated with gas barrier film and adhesive gasket, *Synthetic Metals* 193 (2014), p. 77–80.
- [4] G. Liu, J.B. Kerr, S. Johnson, Dark spot formation relative to ITO surface roughness for polyfluorene devices, *Synthetic Metals* 144 (2004), p. 1–6.
- [5] H. Ohta, M. Orita, M. Hirano, H. Hosono, Surface morphology and crystal quality of low resistive indium tin oxide grown on yttrium-stabilized zirconia, *Journal of Applied Physics* 91 (2002), p. 3547–3550.

- [6] Y. Leterrier, L. Medico, F. Demarco, J.A.E. Manson, U. Betz, M.F. Escola, Mechanical integrity of transparent conductive oxide films for flexible polymer-based displays, *Thin Sol Films* 460 (2004), p. 156–66.
- [7] T.P. Nguyen, P. Le Rendu, P.D. Long, S.A. De Vos, Chemical and thermal treatment of PEDOT:PSS thin films for use in organic light emitting diodes, *Surface & Coatings Technology* 180 (2004), p. 646–649.
- [8] T. Aernouts, P. Vanlaeke, W. Geens, J. Poortmans, P. Heremans, S. Borghs, Printable anodes for flexible organic solar cell modules, *Thin Sol Films* 451 (2004), p. 22–5.
- [9] S.A. Carter, J.C. Scott, P.J. Brock, Enhanced luminance in polymer composite light emitting devices, *Applied Physics Letter* 71 (1997), p. 1145–1147.
- [10] B.W. Xiao, Y.F. Shang, M. Meng, C.N. Li, Enhancement of hole injection with an ultra-thin Ag₂O modified anode in organic light emitting diodes, *Microelectronics Journal* 36 (2005), p. 105–108.
- [11] F.S. Li, Z.J. Chen, C.L. Liu, Q.H. Gong, Improvement in performance of organic light-emitting diodes by adjusting charge-carrier mobility in organic/inorganic hybrid hole transporting layer, *Chemical Physics Letters* 412 (2005), p. 331–335.
- [12] G.F. Wang, X.M. Tao, R.X. Wang, Fabrication and characterization of OLEDs using PEDOT:PSS and MWCNT nanocomposites, *Composites Science and Technology* 68 (2008), p. 2837–2841
- [13] T.C. Li, R.C. Chang, Improving the performance of ITO thin films by coating PEDOT:PSS, *International Journal of Precision Engineering and Manufacturing–Green Technology* 1 (2014), p. 329–334.