Lifetime Improvement of Organic Light-Emitting Diodes Using Cyclo-Olefin Polymer Film as Passivation for Flexible Display

Ki-Su Kim, Byung-Min Park, Kwan-Young Han

Department of Display Engineering, Dankook University, 119, Dandae-ro, Dongnam-gu, Cheonan-si, Chungcheongnam-do, 31116, Korea

Keywords: Organic Light-Emitting Diodes, Lifetime, Cyclo-Olefin Polymer, Passivation, Encapsulation

ABSTRACT

In this study, we have optimized the passivation method of the new concept to improve the lifetime and reliability of organic light-emitting diode (OLED) devices. A cyclo-olefin polymer (COP) film was used as the passivation layer and its properties were investigated. In OLED device, COP films observed to prevent dark spot of emitting area. COP film is suitable as a passivation layer for flexible displays because of its excellent optical properties and flexibility. These advantages can replace the conventional encapsulation methods that are not applicable to flexible display.

1 INTRODUCTION

Recently, organic light-emitting diodes (OLED) is replacing liquid crystal display (LCD) and expanding its application area as next generation display. Compared with LCD, OLED has no backlight unit, so it is possible to manufacture a thin panel. It has excellent contrast ratio, color reproduction, and viewing angle. In addition, research is being actively conducted since it can be used as a flexible display such as foldable, rollable, and stretchable, which is expected as the next generation display. However, despite these many advantages, due to the limitations of organic and electrode materials used in OLED, they are very sensitive to oxygen and moisture in the atmosphere. Currently commercially OLED displays require a water vapor transmission rate (WVTR) of about 10⁻⁶g/m²/day. In general, conventional glass encapsulation is used, and recently, thin film encapsulation (TFE) technology has been applied to devices. However, this method is not suitable for encapsulation of flexible displays, and therefore requires a new encapsulation technique. Therefore, encapsulation (or passivation) that can block moisture in the air is a very important core technology. In order to solve this problem, this study reports a cyclo-olefin polymer (COP) film having excellent optical properties, flexibility, and gas barrier property as a passivation layer of OLED.

2 **EXPERIMENT**

Patterned ITO/glass of the size 2.5x2.5 cm were used

as substrates to prepare green OLED device with glass /ITO/HAT-CN/NPB/CBP:Ir(ppy)3[10%]/TPBi/Alq3/LiF/Al structure. The substrates were washed with acetone, methanol and isopropyl alcohol (IPA) in the ultrasonic bath at 30 °C for 5 min, respectively. The remaining solvent was removed in oven at 100 °C for 20 min. And then, the substrates were treated by UV-Ozone for 15 min.

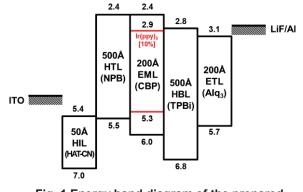


Fig. 1 Energy band diagram of the prepared green OLED device.

All organic layers of OLED device were deposited by thermal evaporation in a vacuum chamber with a base pressure of 5×10⁻⁷ Torr. As a sequence of process in the fabrication. the device 1,4,5,8,9,11-Hexaazatriphenylene hexacarbonitrile (HAT-CN) with thickness of 50Å was firstly deposited as a hole injection layer (HIL). And the N,N'-bis(naphthalene-1-yl)-N,N'bis(phenyl)-benzidine (NPB) with thickness of 500Å was firstly deposited as a hole transport layer (HTL). Then, the 200Å-thick film of host material 4,4'-Bis(Ncarbazolyl)-1,1'-biphenyl (CBP) and 10% doping dopant Tris[2-phenylpyridinato-C2,N]iridium(III) material (Ir(ppy)3) was formed as an emission layer(EML). And then, the 500Å-thick film of 2,2',2"- (1,3,5-benzinetrivI)tris(1-phenyl-1-H-benzimidazole) (TPBi) as an hole blacking layer (HBL) was additively deposited. After that, the 200Å-thick film of Tris(8-hydroxy-quinolinato) aluminum (Alq₃) was formed an electron transport layer (ETL). Finally, 8Å-thick interfacial layer of lithium fluoride (LiF) and 1000Å-thick cathode of aluminum (AI) were

deposited onto the ETL. The energy diagram of organic materials used in the green OLED was shown in fig. 1.

As the passivation layer of the OLED device, the COP-B (188 μ m) films and the COP-D (25, 50 μ m) film with desiccant components were used. In order to laminate the as-fabricated OLED device, the COP-D films were previously dried at 130 °C for 1hour. And then, the COP-D films were laminated to the OLED device at 90 °C for 5 min with a rubber pad. Finally, the evaluation sample was produced by laminating a COP-B film on the COP-D film. All lamination process of COP-D film was performed in a nitrogen atmosphere.

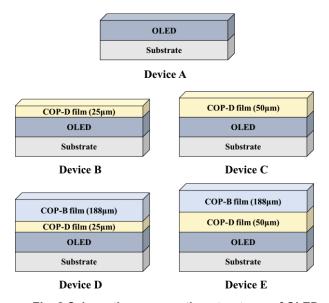


Fig. 2 Schematic cross-section structures of OLED devices with various passivation structures of COP film.

Fig. 2 exhibits the schematic cross-section structures of OLED devices without/with passivation structures: OLED device (a) without passivation, (b) with only COP-D film, (c) with only COP-D film, (d) with COP-D/COP-B film. (e) with COP-D/COP-B film.

The OLEDs without passivation were used as primitive reference device sample. The passivation structures of each device are as follows:

(a) glass/OLED (Device A), (b) glass/OLED/COP-D(25 μ m) (Device B), (c) glass/OLED/COP-D(50 μ m) (Device C), (d) glass/OLED/COP-D(25 μ m)/COP-B(188 μ m) (Device D), and (e) glass/OLED/COP-D(50 μ m)/COP-B(188 μ m) (Device E). The electrical and optical properties of the fabricated all OLED devices were measured at room temperature.

The electrical properties of the devices were investigated using an I-V-L measurement system (Polaronix M6100, McScience, Korea). The measurements of the optical properties, such as luminance, emission spectrum, and Commission Internationale de l'Eclairage (CIE) color coordinates of the samples were measured and evaluated using a spectro-radiometer (CS-1000, Minolta, USA). All electrical and optical measurements were carried out in a dark room.

3 RESULTS and DISCUSSION

In order to evaluate the lifetime characteristics of the OLED device according to the various passivation structure fabricated above, the growth of dark spots was examined over time.

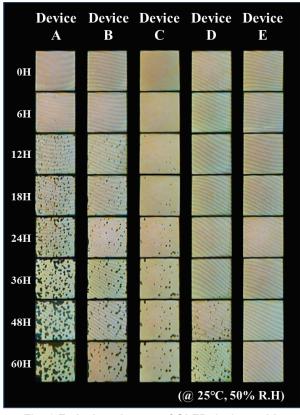


Fig. 3 Emission pictures of OLED devices with various passivation structures of COP film.

Fig. 3 shows the emission picture of OELD devices without passivation layer or with various passivation layers of COP film for up to 60 hours at 7V applied voltage. In the Device A, dark spots were observed after 6 hours of exposure, and after 12 hours, dark spot growth proceeded rapidly. On the other hand, although dark spots were observed after Device B and C exposure for 12 hours, the growth rate of dark spots was lower than that of Device A. In addition, it was confirmed that the dark spot growth of the Device C with 50 um COP-D film is slower than Device B with 25 um COP-D film. This is because the desiccant component inherent to the COP-D film absorbs the moisture present between the OLED device and the COP film and blocks moisture from penetrating into the OLED device from the atmosphere. As a result, the lifetime of Device C is relatively improved because the content of desiccant is increased by

increasing the thickness of COP-D film. Both Devices D and E with COP-D and COP-B film showed no dark spots and exhibited better electrical and optical properties than devices A, B, and C until 60 hours. This is because the COP-B film as barrier property compensates for the relatively poor gas barrier properties of the COP-D film. However, Device E, which has passivation structure of COP-D (50 um)/COP-B (188 um) film on the OLED device, exhibits more stable properties than Device D with passivation structure of COP-D (25 um)/COP-B (188 um). Therefore, as the thickness of the COP-D film increases, the content of the desiccant increases, thereby effectively absorbing or blocking moisture, thereby greatly improving the growth of the dark spot in the Device E.

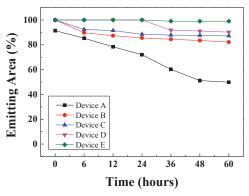


Fig. 4 Change in emission area over time of OLED devices with various passivation layers of COP film.

Fig. 4 shows the change in emission area over time of OLED devices with various passivation layers of COP film. This exhibits the ratio of the area of dark spots generated in the emission area of the OLED device shown in Fig. 3. The result was calculated by the following equation (1):

$$Emitting Area (\%) = \frac{None Dark Spot Area}{Total Emission Area} \ge 100$$
(1)

The Device B and C, which only forms the COP-D film, has a slower decrease in the emission area over time than the Device A without the passivation layer. In devices D and E in which the COP-D and COP-B films were formed, no reduction in the emission area was observed until 60 hours because no dark spot was produced in the emission area of the OLED device. Passivation structures of Device D and E using the COP-D and COP-B effectively block external oxygen and moisture. As a result, the Device B, C, D, and E of various passivation structures with the COP film compared to the non-passivated device A prevented the growth of dark spots. In addition, the lifetime of Device D and E with hybrid passivation structure using COP-D film and COP-B was significantly improved.

4 CONCLUSIONS

This study proposed a new passivation process to

improve the disadvantages of conventional rigid glass encapsulation or TFE applied to OLED display devices and evaluated its characteristics. To solve this problem, a passivation layer was formed using COP film, which is significantly thinner than conventional glass, and prevented growth of dark spots and improved lifetime of device. From these basic results, it is expected that it can be used as a next generation passivation layer for flexible displays if it has excellent gas barrier properties and flexibility through various structures and materials with COP film. In addition, it will be possible to use it widely in various OLED wearable devices and automotive related industries.

REFERENCES

- C. W. Tang and S. A. VanSlyke, "Organic electroluminescent diodes", Appl. Phys. Lett., 51, 913 (1987).
- [2] S. W. Seo, E. Jung, H. Chae and S. M. Cho, "Optimization of Al₂O₃/ZrO₂ nanolaminate structure for thin-film encapsulation of OLEDs", Org. Electron, 13, 2436 (2012).
- [3] J. C. Jeong, H. S. Kim and J. G. Jang, "High Efficient Green Phosphorescent OLED with Emission Layer of TPBI Selectively Doped with Ir(ppy)₃", Mol. Cryst. Liq. Cryst., Vol. 530, 130 (2010).
- [4] J. Wu, F. Fei, C. Wei, X. Chen, S. Nie, D. Zhang, W. Su and Z. Cui, "Efficient multi-barrier thin film encapsulation of OLED using alternating Al₂O₃ and polymer layers", RSC Adv, 8, 5721 (2018).
- [5] E. G. Jeong, Y. C. Han, H. G Im, B. S. Bae and K. C. Choi, "Highly reliable hybrid nano-stratified moisture barrier for encapsulating flexible OLEDs", Org. Electron, 33, 150 (2016).
- [6] P. Weijer, H. B. Akkerman, Piet. C. P. Boutten, P. Panditha, P. J.M. Klaassen and A. Salem, "Side leakage into the organic interlayer of unstructured hybrid thin-film encapsulation stacks and lifetime implications for roll-to-roll produced organic lightemitting diodes", Org. Electron, 53, 256 (2018).
- [7] Y. C. Han, E. G. Jeong, H. C. Kim, S. I. Kwon, H. G Im, B. S. Bae and K. C. Choi, "Reliable thin-film encapsulation of flexible OLEDs and enhancing their bending characteristics through mechanical analysis", RSC Adv, 6, 40835 (2016).
- [8] S. H. Yong, S. J. Kim and H. Chae, "Surface Planarization of Low-Temperature Flowable Silicon Oxide for Atomic Layer Deposition Al₂O₃ Thin Film Encapsulation", J. Nanosci. Nanotechnol, Vol 19, 2882 (2019).
- [9] H. Zhang, H. Ding, M. Wei, C. Li, B. Wei and J. Zhang, "Thin film encapsulation for organic light-emitting diodes using inorganic/organic hybrid layers by atomic layer deposition", Nanoscale. Res. Lett., 10, 169 (2015).