

Enhancing the lifetime of flexible organic light emitting diodes by encapsulation technique

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ABSTRACT: Flexible organic light emitting diodes (OLEDs) have been fabricated and their lifetime have been improved by encapsulation technique. We have also discussed the mechanism in lifetime decreasing of flexible OLEDs. The device structure is PET/ITO/NPB/Alq₃/LiF/Al. First, employing SiO_x and TeO₂ films thermally deposited on the devices as thin barrier film was investigated. Comparing with the bare device, the flexible OLED with SiO_x and TeO₂ as an encapsulation layer has a longer lifetime by 3 times and 7 times respectively. Then, we proposed an efficient encapsulation method by thermally evaporated SiO_x as a thin barrier film, following by a lamination process with an adhesive passivation layer (HBF-200UV). At an initial luminance of 100 cd/m², the lifetime of flexible OLED with encapsulation process has a longer lifetime by 26 times comparing to the bare device. The encapsulation technique combining lamination process and thin barrier film provided lower permeation rates of moisture and oxygen for flexible OLEDs more efficiently.

Key words: flexible, organic light emitting diodes, encapsulation

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I. INTRODUCTION

As a next generation display, the organic light emitting diode (OLED) has great performances such as potentially low manufacturing cost, high brightness, thin thickness, low power consumption, fast response time, wide viewing angle, fast response time, emission spectra cover the entire range of visible light [1-8]. In addition, the OLED can be fabricated onto flexible polymeric substrates instead of the rigid glass substrates for flexible display applications.

The flexible polymeric substrates have low ability to prevent moisture and oxygen pass through. Moreover, organic material is sensitive to moisture and oxygen from ambient air. It is critical to prevent the degradations induced by oxygen and moisture. In order to solve this problem, an active encapsulation technique using a glass or metal lid attached by a bead of UV cured epoxy resin has been extensively performed to protect OLEDs from oxygen and water vapors. However, these seals are large and heavy so that they severely limit the applications of OLEDs.

For this reason, thin and lightweight encapsulation method has been extensively studied. GiHeon etc. applied an adhesive multilayer of polyacrylate-based adhesive and Al as a passivation layer, which encapsulated flexible OLED by lamination process [9]. Seung Ho Kwon etc. used triple-layer passivation, which combine high-density

polyethylene (HDPE) and Al-Li to improve the lifetime of polymer light emitting diode [10]. Among these, the passivation layer should provide not only a good barrier against moisture and oxygen but also an insulator in order to prevent the short-circuit between anode and cathode.

In this study, the flexible OLEDs with different encapsulation materials, SiO_x and TeO₂, have been applied. A classic and developed material, SiO_x act as a barrier to oxygen and water vapor especially applied to food packaging and medical device industries. In addition, the TeO₂ content has significantly influenced the glass quality and the IR transmission, transparency and resistance toward moisture [11]. The water vapor transmittance rate (WVTR) and the packing density of thin barrier film are discussed.

II. EXPERIMENT

In this study, we report an encapsulation method by means of combine a thin barrier film formed by evaporation and an adhesive multilayer formed by a conventional lamination process. Figure 1 (a) depicts the flexible OLED configuration studied in this work. Prior to organic layer deposition by thermal evaporation, indium-tin-oxide (ITO) coated polyethylene terephthalate (PET) substrates, with a sheet resistance of 12 Ω/square, were solvent cleaned

by ultrasonic baths in acetone and isopropyl alcohol then in deionized water, followed by UV ozone treatment for ten minutes. Organic layers and cathode materials were sequentially without breaking vacuum through a shade mask. Deposition began with a 400 Å-thick film N, N'-diphenyl-N, N'-bis (1-naphthyl-phenyl)-(1, 1'-biphenyl)-4, 4'- diamine (NPB) as a hole transporting layer. Next, a 600 Å-thick film tris(8-hydroxyquinolino)- aluminum (Alq_3) was deposited as emitting and electron transporting layer, followed by a 7 Å-thick film electron buffer layer LiF and a 2000 Å-thick film cathode Al. The active area of the devices was 24 mm². The organic layers were evaporated at a pressure of below 5×10^{-6} torr, and the evaporation rate was controlled to be 1-2 Å/s. The thickness of the evaporation layers was controlled by oscillating quartz monitors and further calibrated by making ellipsometric measurements. The current density-voltage (J-V) and luminance-current density (L-J) were measured by the TOPCON SR-1 spectrometer with a Keithley 2400 programmable voltage-current source. All measurements were made at room temperature.

Figure 1 (b) illustrates the skeleton of a method for measurement the permeation, and the permeation sensors are prepared as follows. The electronic pads were produced from etching ITO/glass substrate by lithography as tradition. First, the center 0.2×0.4 cm² section is covered by a 100 nm layer of calcium with common evaporation techniques. Then the calcium was encapsulated with a thin barrier film, SiO_x and TeO_2 by thermal evaporation. The permeation rate is calculated from the advancing degradation of a calcium layer. Finally, the samples are connected to a Keithley 2400 to measure the resistance. The measurement voltage was set at only 0.5 V in order to avoid an electrochemically enhanced corrosion at the contacts. The method introduced a resistance measurement to determine the degree of Ca corrosion.

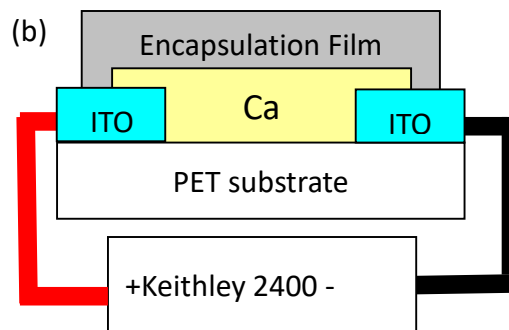
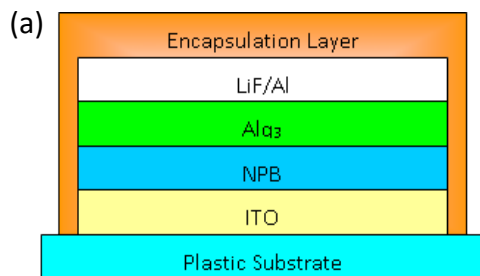


Figure 1: (a) Fundamental structure of a flexible OLED with an encapsulation layer. (b) Layout of the permeation sensor. The PET is covered with a calcium layer that is contacted by ITO pads.

III. RESULTS AND DISCUSSION

Figure 2 shows lifetimes of flexible OLED devices encapsulated by a thin film TeO_2 , SiO_x , and without encapsulation. All measurements of lifetime were made at room temperature and the initial luminance was controlled at 100 cd/m². The bare device to 50% of initial luminance had a very short lifetime of about 8.5 hrs in ambient conditions. It was well known that organic materials were sensitive to moisture and oxygen. On the other hand, the flexible OLED cannot endure ambient conditions for long time. The devices encapsulated by a thin film of TeO_2 and SiO_x have half-life were 30.6 hrs and 17 hrs, respectively. From this result, the use of evaporation deposited TeO_2 thin film for the barrier layer can not only reduce the total thickness of barrier layer also increase the lifetime of a flexible OLED device based on a PET substrate significantly. It suggested that the thin film of TeO_2 resist humidity and oxygen are better than SiO_x .

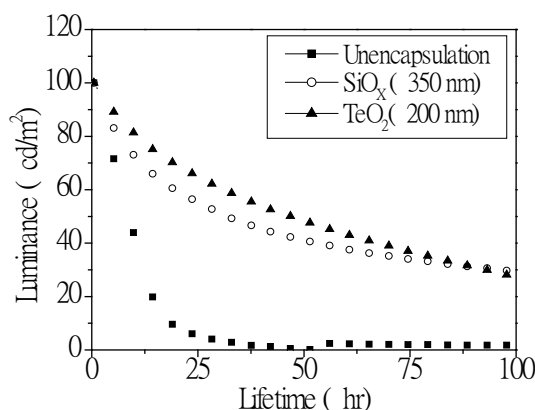


Figure 1: Luminance decay of the flexible OLED operated at a constant dc current of 10 mA/cm².

Figures 3(a) and 3 (b) show packing density of the encapsulation film varied with (a) evaporation rate and (b) thickness of SiO_x and TeO_2 . The packing

density can be expressed by the equation

$$\text{packing density} \equiv \frac{n_f - n_v}{n_s - n_v} \text{-----(1)}$$

where n_f is the refractive index of the film, n_s is the refractive index of the solid, and n_v is the refractive index of the void which the value is set by 1. First, the evaporation rate varying with packing density was measured under the same thickness of thin barrier film at 100 nm. From the results, as the evaporation rate changes from 1 Å/s to 10 Å/s the packing density of TeO₂ from 0.95 to 0.9, SiO_x from 0.87 to 0.66, respectively. In addition, the thickness varying with packing density was measure under the same evaporation rate of thin barrier film at 1 Å/s. As the thickness change from 50 nm to 150 nm the packing density changes from 0.87 to 0.9, SiO_x from 0.81 to 0.88, respectively. In the measure, the influence of evaporation rate and the quality of material on packing density are evident. From this result, evaporation rate is an important parameter of packing density for same material, and the value of packing density critically affect the lifetime of flexible OLED. It may attribute to that organic layer in the flexible OLED absorb water and oxygen from voids of SiO_x and TeO₂ films.

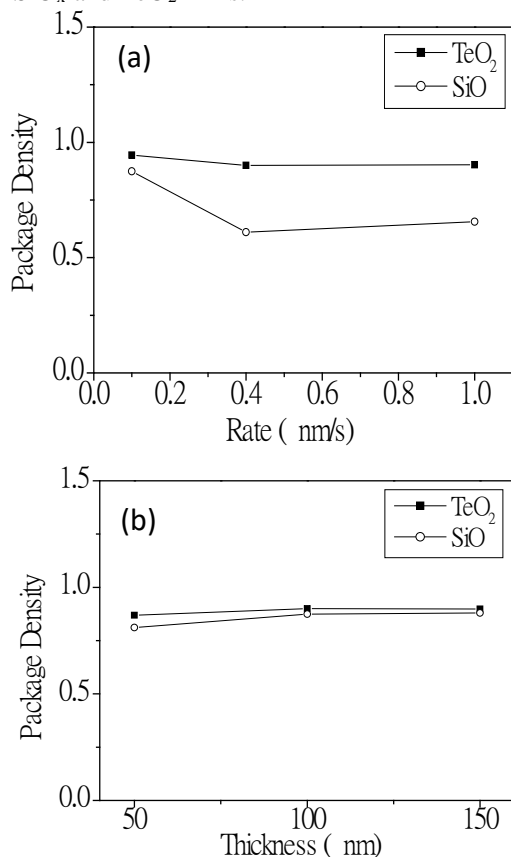


Figure 3: Packing density of the encapsulation film varied with (a) evaporation rate and (b) thickness of SiO_x and TeO₂.

Moreover, the permeation for a PET substrate coated with an encapsulation film of 350 nm SiO_x and 200 nm TeO₂ is shown in Fig. 4. The permeation for PET without encapsulation film is also shown for comparing. The permeation can be expressed by the equation

$$P = -n \frac{M(\text{reagent})}{M(\text{Ca})} \delta \rho \frac{l}{b} \frac{d(1/R)}{dt} \text{-----(2)}$$

where n is the molar equivalent of the degradation reaction; $M(\text{reagent})$ and $M(\text{Ca})$ are the molar masses of the permeating reagent and Ca; t is the measurement time, $1/R$ is the slope of the conductance curve, δ , ρ , l , and b are the density, resistivity, length and width of Ca layer respectively. The slopes of the TeO₂ and SiO_x in the curve can be directly related to the permeation rate, which are calculated to be 2.399×10^{-1} and 3.99×10^{-1} g/m²-day at 38 °C and 90% RH. As described above, the packing density and permeation rate are the important factors in lifetime of flexible OLED.

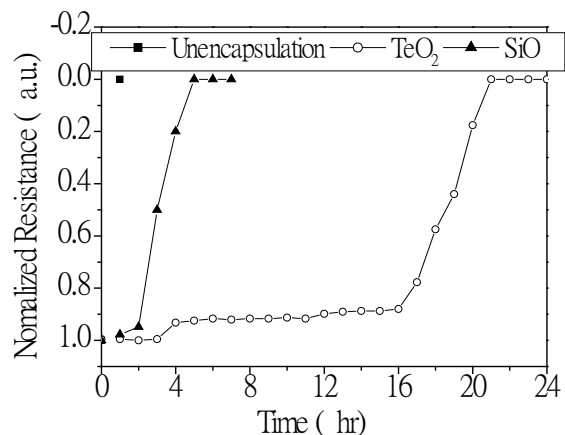


Figure 4: The permeation for a PET substrate coated with an encapsulation film of 350 nm SiO_x and 200 nm TeO₂.

Next, we studied the rate of degradation of the fabricated devices. First, the devices were encapsulated by SiO_x and TeO₂ as a thin barrier film respectively. Then the two devices were covered by the same lamination films, HBF-200UV, in lamination process. The measurements of lifetime were identical in the same condition as described above. Figure 5 shows luminance versus driving time for three devices. Results from the lifetime of the initial luminance of the devices encapsulated by SiO_x (350 nm)/HBF-200UV and TeO₂ (200 nm)/HBF-200UV decays to 50% of its initial luminance are 213 hrs and 78 hrs respectively. When it comes to the ability of prevention of destruction of the moisture and oxygen, SiO_x (350 nm)/HBF-200UV is preferable to TeO₂

(200 nm)/HBF-200UV. The thin barrier film of SiO_x may be a factor of improvement within the flexible OLED lifetime. In Fig. 6, the result was confirmed by the observation of AFM: the two thin barrier films influence the adhesive behavior of HBF-200UV. In addition, the surfaces of thin films affect the lifetime of flexible OLED device as well. The RMS surface roughness of the 100 nm thick SiO_x and TeO_2 film was 2.12 nm and 2.76 nm respectively. The result implies smooth surface of thin barrier film is suitable for the flexible OLED encapsulation.

Figure 7 plots the electrical characteristics of the fabricated diodes classified by processing step was examined to compare the electrical properties both before and after forming the lamination film. From this result, there was no significant difference in the characteristics of devices. On the other hand, when the thin barrier film and lamination film were formed on the device affect organic layer and metal cathode slightly.

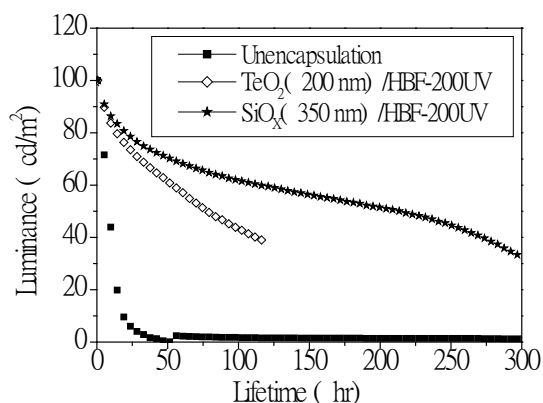


Figure 5: Luminescence decay of the flexible OLEDs operated at a constant dc current of 10 mA/cm².

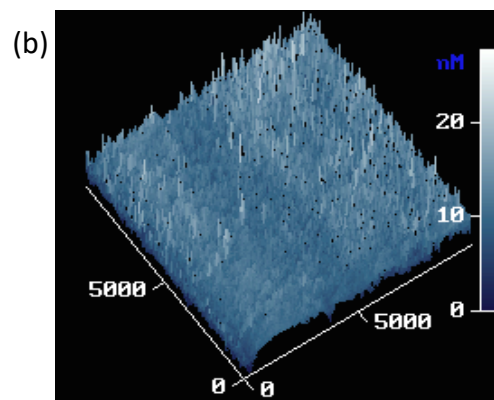
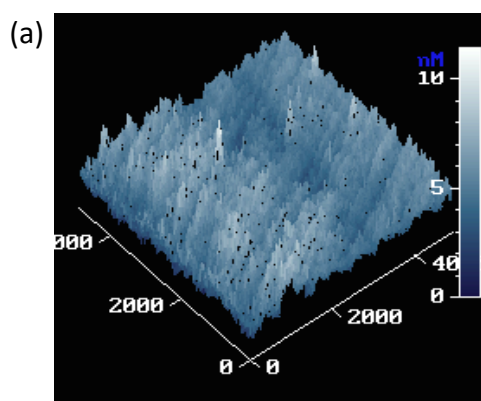


Figure 6: AFM images of (a) 100 nm SiO_x and (b) 100 nm TeO_2 coated glass substrate.

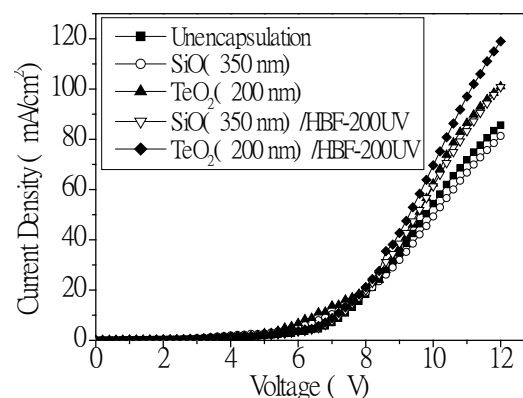


Figure 7: Current density-voltage characteristics of the flexible OLEDs with and without the encapsulation layers.

IV. CONCLUSION

In this study, we improve the lifetime of flexible OLED by encapsulation technique successfully. The devices encapsulated by a thin film of TeO_2 and SiO_x have half-life were 30.6 hrs and 17 hrs, respectively. Compare TeO_2 with SiO_x as thin barrier films, TeO_2 shows higher packing density and low permeation ratio. However, the lifetime of device with SiO_x (350 nm)/HBF-200UV can reach 213 hrs, and the lifetime of the device with TeO_x (200 nm)/HBF-200UV is only 78 hrs at an initial luminance of 100 cd/m². It due to the surface of SiO_x is smoother than TeO_2 , and the smooth surface avoids moisture and oxygen in the interface between thin barrier film and lamination film. Comparing the bare device, the lifetime of flexible OLED with SiO_x (350 nm)/HBF-200UV has a longer lifetime by 26 times.

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REFERENCES

- [1]. C.W. Tang, and S.A. Vanslyke, Organic electroluminescent diodes, *Appl. Phys.*, *51*, 1987, 913-915.
- [2]. Y. Yu, W.Y. Lin, X. Peng, Z. Wu, Y. Jin, X. Li, X. Zhang, H. Yang, and G. Xie, Realizing an efficient warm white organic light-emitting device possessing excellent color-stability and color rendering index, *Organic Electronics*, *68*, 2019, 129-134.
- [3]. F. Liu, H. Liu, X. Tang, S. Ren, X. He, J. Li, C. Du, Z. Feng, and P. Lu, Novel blue fluorescent materials for high-performance nondoped blue OLEDs and hybrid pure white OLEDs with ultrahigh color rendering index, *Nano Energy*, *68*, 2020, 104325-104334.
- [4]. T. Xu, J.G. Zhou, M.K. Fung, and H. Meng, Simplified efficient warm white tandem organic light-emitting devices by ultrathin emitters using energy transfer from exciplexes, *Organic Electronics*, *63*, 2018, 369-375.
- [5]. J.H. Jou, Z.K. He, Y.T. Su, Y.F. Tsai, and C.H. Wu, Healthy light with visual quality and energy-saving character, *Organic Electronics*, *38*, 2016, 396-400.
- [6]. G.T. Chen, S.H. Su, C.C. Hou, and M. Yokoyama, Effects of thermal annealing on performance of organic light-emitting diodes, *J. Electrochem*, *153*, 2007, 159-162.
- [7]. Y. Xiong, F. Deng, S. Xu, and S. Gao, Performance analysis of multi-primary color display based on OLEDs/PLEDs, *Optics Communications*, *398*, 2017, 49-55.
- [8]. D. Pereira, A. Pinto, A. Califórnia, J. Gomes, and L. Pereira, Control of a white organic light emitting diode emission parameters using a single doped RGB active layer, *Materials Science and Engineering: B*, *211*, 2016, 156-165.
- [9]. G.H. Kim, J. Oh, Y.S. Yang, L.M. Do, and K.S. Suh, Lamination process encapsulation for longevity of plastic-based organic light-emitting devices, *Thin Solid Films*. *467*, 2004, 1-3.
- [10]. S.H. Kwon, S.Y. Paik, O.J. Kwon, and J.S. Yoo, Triple-layer passivation for longevity of polymer light-emitting diodes, *Appl. Phys. Lett.*, *79*, 2001, 4450-4452.
- [11]. A.H. Khafagy, A.A. El-Adawy, A.A. Higazy, S. El-Rabaie, and A.S. Eid, Studies of some mechanical and optical properties of $(70 - x)\text{TeO}_2 + 15\text{B}_2\text{O}_3 + 15\text{P}_2\text{O}_5 + x\text{Li}_2\text{O}$ glasses, *Journal of Non-Crystalline Solids*. *354*, 2008, 3152-3158.

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