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Flexible Encapsulation of Organic Light-Emitting Diode Using the LiF/polymer/LiF Buffer and Metal Foil

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New simple and reliable encapsulation using the inorganic/organic/inorganic buffer and metal foil was studied on a red phosphorescent organic light-emitting diode (PhOLED) for flexible electronics. The LiF/polymer(AZ1512-photoresist)/LiF and Cu foil were used as the materials of the inorganic/organic/inorganic layers and a metal cover in the configuration of encapsulation, respectively. To make the process simple in the fabrication of red PhOLED, bis(10-hydroxybenzo[h]quinolinato)beryllium (Bebq₂) was used in common as a host material of emissive layer and an electron transporting material of electron transport layer.

The fabricated device before encapsulation showed a maximum quantum efficiency of 15.3%, color coordinates of (0.67, 0.32) and a luminance of 5,100 cd/m² at 8V. Luminance change was little found in the process of flexible encapsulation. Luminance degradation of the device encapsulated with the buffer layers of LiF/AZ1512-photoresist/LiF and Cu foil according to the passage of time was very stable during a long term after one week and comparable to that of the device encapsulated with a conventional glass lid.

Keywords Flexible encapsulation; Cu foil; LiF; photoresist; red PhOLED; efficiency

Introduction

The lifetime of an organic light-emitting diode (OLED) is critically limited in the atmosphere of humidity and oxygen. In order to isolate an OLED from atmospheric oxygen and water, encapsulation is a key technology [1, 2]. Typical encapsulation of an OLED fabricated on glass uses a method of glass lid or metal cap with a desiccant inside [3, 4]. This method is quite simple and reliable, but it cannot be applicable to flexible displays and bendable solid-state lightings. As one of the solutions to realize a reliable and flexible OLED, thin film encapsulation using an inorganic/organic multi-barrier structure has been studied [5, 6]. Multilayer combination of polymer and inorganic dielectrics can be very effective in blocking the penetration of water and oxygen from the outside environment. However, the process of multi-barrier coatings is rather complicated and time-consuming because many of organic and inorganic layers should be contained in the thin film encapsulation. Thus, the

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development of a new encapsulation with excellent gas barrier property and high flexibility which can be easily realized by simple process is crucial for the wide applications of OLED.

Red light-emitting devices are used as an optical source for solid-state lighting or medical phototherapy as well as one of three basic pixels in the full-color display [7, 8]. Recently, a light in the red to the near infrared range was utilized to accelerate wound healing and relieve pain from injury [9]. As an OLED is basically a planar light source, its application is very wide. For the practical use of OLED in a variety of applications, however, the device structure should be as simple as possible with high flexibility. Moreover, high luminous efficiency and good reliability in the long term operation are required in the device performance.

In this study, a simple structural and red phosphorescent OLED with new flexible encapsulation was reported. Red light sources can be used in a wide range of applications and the internal quantum efficiency of phosphorescent OLED theoretically reaches 100% which is four times higher than that of fluorescent OLED [10, 11]. The device structure could be simplified by reducing the number of organic materials necessary in the device fabrication and introducing a new encapsulation method. The configuration of new flexible encapsulation was composed of the LiF/AZ1512-photoresist/LiF buffer layers and Cu foil. The process of this encapsulation was very easy and cost-effective due to low temperature deposition of buffer layers and simple attachment of metal foil.

Experimental Procedure

The substrates with an indium-tin-oxide (ITO) anode of $12 \Omega/\text{sq}$ were cleaned by ultrasonic cleaning process with acetone and isopropyl alcohol. The remaining solvent was removed by soft-baking for 10 minutes at 100°C . Then, the substrates were plasma-treated at 120 W for two minutes under 8 mTorr pressure of O_2/Ar . The plasma treatment reduces the energy barrier for hole injection from the anode to the next organic layer and also removes contaminants on the ITO surface [12]. All organic layers were deposited by an in-situ thermal evaporation method under the pressure of 10^{-7} Torr. Layer thicknesses of the deposited materials were measured using an oscillating quartz thickness monitor. As a sequence of process in the device fabrication, the N,N'-diphenyl-N,N'-bis-[4-(phenyl-m-tolyl-amino)-phenyl]-biphenyl-4,4'-diamine (DNTPD) with thickness of 400 Å was firstly deposited as a hole injection layer (HIL). Then, the 300 Å-thick film of 1,1-bis(di-4-polyaminophenyl)cyclohexane (TAPC) was formed as a hole transport layer (HTL). In the formation of emissive layer (EML), the 300 Å-thick film of bis(10-hydroxybenzo[h]quinolinato) beryllium (Bebq_2) doped with 5 vol.% RP411 (red phosphorescent dye provided from SFC Co.) was deposited. After that, the 500 Å-thick film of Bebq_2 as an electron transport layer (ETL) was additively deposited without doping. The common use of Bebq_2 in the EML and ETL makes the fabrication process simple. Finally, 10 Å-thick interfacial layer of LiF and 1200 Å-thick cathode of Al were successively deposited onto the ETL surface. The energy diagram of organic materials used in the PhOLED was shown in Fig. 1

In order to encapsulate the as-fabricated PhOLED, the 6000 Å-thick film of LiF was deposited on the Al cathode by high vacuum thermal evaporation as the first inorganic buffer layer. The LiF can be a good inorganic buffer material between an OLED and a final cover because it is an insulating material with a band gap of 14 eV [13]. Furthermore, the low melting temperature of 845°C makes the deposition of LiF very easy [14]. Then, the AZ1512 (a photoresist from AZ Electronic Materials) was deposited by a spin coating method with

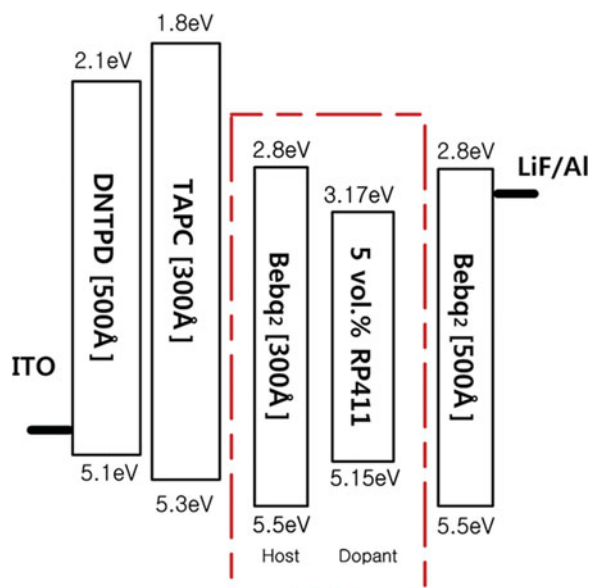


Figure 1. Energy level diagram of the organic layers used in the red PhOLED fabrication.

the spin speed of 2500 rpm as an organic buffer layer. A polymerized photoresist can be a good polymer for flexible encapsulation. After removal of the remaining solvent of photoresist by baking at 80°C for 25 minutes, the 6000 Å-thick film of LiF was deposited once again as the second inorganic buffer layer. Finally, the encapsulation process was completed by capping the buffer layers with the 0.06mm-thick Cu foil and sealing the edge of encapsulation with a sealant (Dow Corning® 3140). The cross-sectional view of a flexibly encapsulated device was shown in Fig. 2.

Electroluminescent characteristics of the fabricated devices such as current density, luminance, luminous efficiency, electroluminescence spectra, and CIE color coordinates

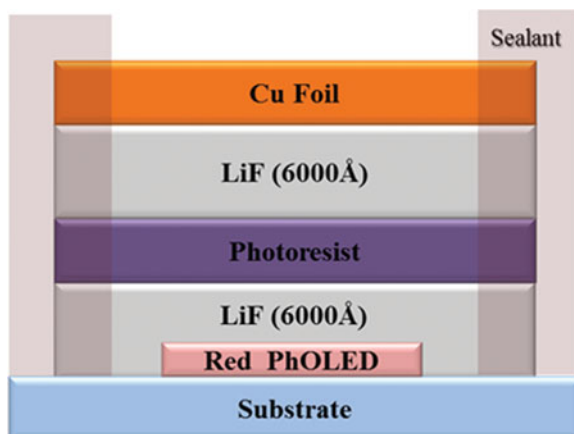


Figure 2. Cross-sectional view of a flexibly encapsulated device.

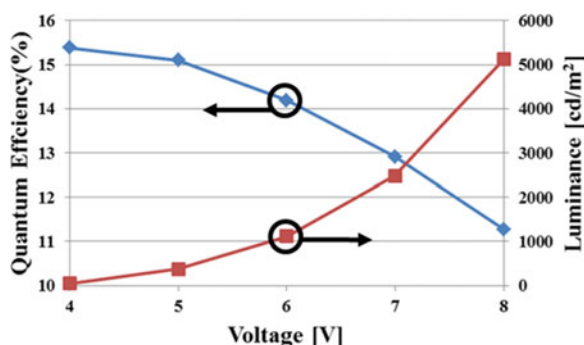


Figure 3. The quantum efficiency-luminance-voltage characteristics of the fabricated device before encapsulation.

were measured using a Polaronix M6100 system (McScience) and a CS-1000A spectroradiometer (Konica Minolta) in a dark condition.

Results and Discussion

In the energy diagram of organic layers shown in Fig. 1, the lowest unoccupied molecular orbital (LUMO) offset barrier of 1 eV at the interface of TAPC and Beq₂ is large enough to confine the electrons injected into the emissive layer. The hole mobility and triplet energy of TAPC are 0.01 cm²/V.s and 2.87 eV, respectively [15]. Thus, the TAPC can be a good hole transport material in the red PhOLED due to the high hole mobility and triplet energy as well as the good electron confinement. The Beq₂ as a host material has an energy gap of 2.7 eV and the RP411 as a phosphorescent dye has a triplet energy of 1.98 eV in the emissive layer. Therefore, the exciton energy of Beq₂ can be easily transferred to the triplet energy of RP411 in the host-dopant system. The Beq₂ can be also used as an electron transporting material in the ETL besides a host material of emissive layer owing to a high electron mobility of about 10⁻⁴ cm²/V.s [16]. The common use of Beq₂ in the EML and ETL results in reducing the number of organic materials necessary in the device fabrication and makes the fabrication process simple.

Figure 3 showed the quantum efficiency-luminance-voltage characteristics of the PhOLED before encapsulation. As shown in Fig. 3, the luminance at an applied voltage of 8V and the maximum quantum efficiency were 5,100 cd/m² and 15.3%, respectively.

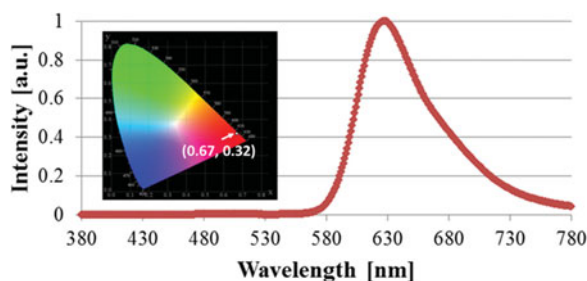


Figure 4. Electroluminescence spectra and CIE color coordinates.

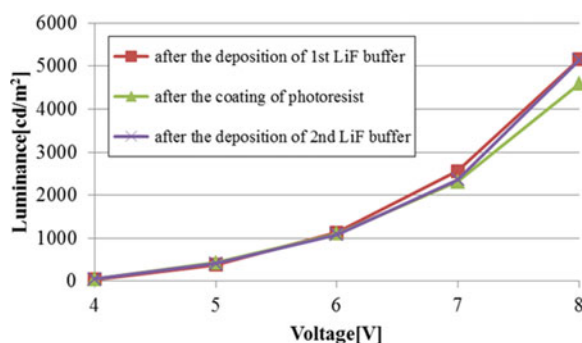


Figure 5. The variation of luminance-voltage characteristics according to the deposition of each buffer layer.

Though the structure of organic layers was rather simple without using an additional material in the ETL, the fabricated device showed good electroluminescent characteristics in the luminance and quantum efficiency. The electroluminescence spectra were shown in Fig. 4 in which a diagram of Commission Internationale de l'Eclairage (CIE) color coordinates was inserted. The peak wavelength of electroluminescence spectra and the color coordinates on the CIE chart were 627 nm and (0.67, 0.32), respectively. The color coordinates of CIE(0.67, 0.32) is similar to the NTSC (National Television System Committee) red coordinates of (0.67, 0.33) with a color purity of nearly 100%.

In order to investigate the adaptability of the LiF/photoresist/LiF as the buffer layers for flexible encapsulation, the variation of luminance-voltage characteristics was observed after the formation of each buffer layer as shown in Fig. 5. Luminance change according to the formation of buffer layers was not nearly observed at low voltage, though there was a little change in luminance at high voltage after the deposition of photoresist. A small degradation of luminance after the deposition of photoresist was recovered after the formation of the following LiF layer. A little change in luminance after the deposition of photoresist may be attributed to the residual solvent in the polymerized photoresist as an organic buffer. Considering the overall little change of luminance according to the deposition of buffer layers, it was thought that the configuration of LiF/photoresist/LiF was

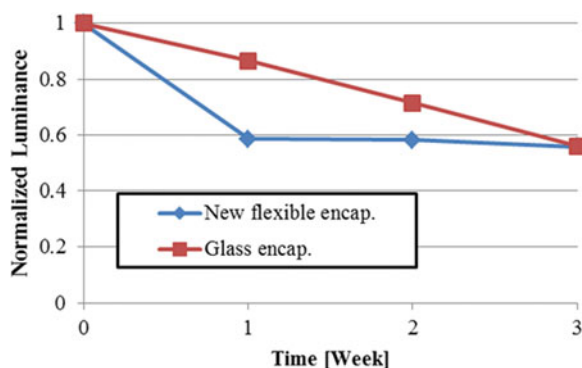


Figure 6. The luminance normalized to an initial value right after encapsulation according to the aging time.

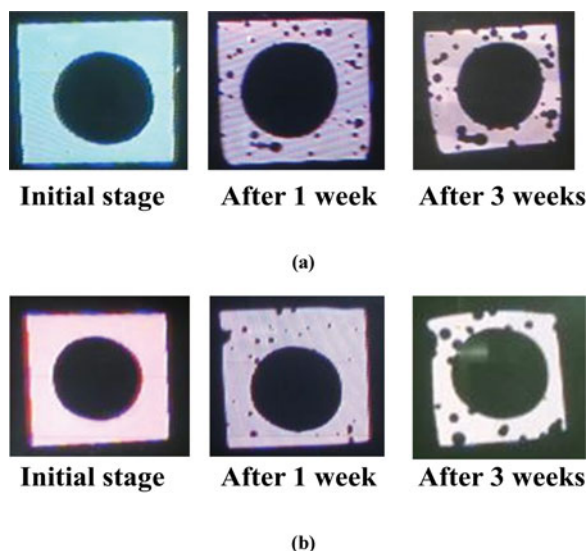


Figure 7. Images of dark spots according to the aging time: (a) new encapsulation using the LiF/polymer/LiF buffer and Cu foil, (b) conventional encapsulation using a glass lid.

suitable as a buffer system for flexible encapsulation because of excellent chemical stability and electrical insulating property. The luminance normalized to the initial value right after encapsulation according to the aging time was shown in Fig. 6. The aging test shown in Fig. 6 was implemented under a constant applied voltage of 9 V for three weeks. To examine the relative performance of flexible encapsulation in our experiments, the aging effect of the device encapsulated with the buffer layers of LiF/photoresist/LiF and Cu foil was compared to that of the device encapsulated with a glass lid. The structure of the device encapsulated with a glass lid was identical with that of the flexibly encapsulated device before they were encapsulated. Though the device encapsulated with the buffer layers of LiF/polymer/LiF and Cu foil showed a rapid degradation of luminance in the early stage, its luminance after one week was very stable as shown in Fig. 6. The rapid degradation of luminance in the early stage may be caused by some residual solvent in the photoresist layer or impurities from the adhesive of Cu foil. On the other hand, the long term degradation of luminance after three weeks was not inferior to that of the device encapsulated with a glass lid. Figure 7 showed the images of dark spots according to the passage of time in the differently encapsulated devices. No remarkable difference in the images of dark spots from the aging effect in the differently encapsulated devices could not be seen in Fig. 7.

As a result, the stability for long term operation of the new flexible encapsulation was compatible with that of a conventional glass encapsulation. Furthermore, the easy process due to low temperature deposition of LiF, spin-coating of photoresist and simple attachment of Cu foil makes the new flexible encapsulation more attractive in the practical applications.

Conclusions

A red PhOLED flexibly encapsulated with the buffer of LiF/polymer/LiF and Cu foil was reported in this study. The fabricated device showed the initial electroluminescent characteristics with a maximum quantum efficiency of 15.3% and color coordinates of (0.67,

0.32) before encapsulation. The device structure was rather simple due to use of the same material in the ETL with the host of EML, which resulted in reducing the number of organic materials necessary in the device fabrication. The process of our flexible encapsulation was much easier than the process of a conventional organic/inorganic multi-barrier method. The stability of the device encapsulated with the buffer layers of LiF/polymer/LiF and Cu foil was compatible with that of the device encapsulated with a glass lid.

Owing to the advantages of simple structure and easy process, the red PhOLED and new encapsulation introduced in this study can be usefully applied in the wide organic electronics.

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