Highly efficient tandem PHOLEDs with lithium-doped BPhen/NDP-9-doped TAPC as a charge generation layer

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ABSTRACT

The development of large-area organic light-emitting diode (OLED) displays requires a highly efficient tandem device architecture and an easily processable charge generation layer (CGL) with a low voltage drop and high optical transparency. In this study, we investigated and applied a doped organic n-CGL/p-CGL using thermal vacuum deposition in tandem OLED devices. A doping concentration of 1.0 wt.% for Li in 4, 7-Diphenyl-1, 10-phenanthroline (BPhen) was optimal for the n-CGL with 8 wt.% for 2-(7-dicyanomethylene-1,3,4,5,6,8,9,10-octfluoro-7H-pyrene-2-ylidene)malononitrile (NDP-9)-doped N,N-bis(4-methylphenyl)benzenamine (TAPC) as a p-CGL. Maximum luminous efficiencies of 42.5 and 63.4 cd/A and a 4,000 cd/m² current density for the target luminance values of 11.2 and 6.5 mA/cm² were demonstrated for double-stack and triple-stack tandem blue phosphorescent OLED devices, respectively. Implementing these highly efficient tandem device structures will improve the overall lifetime of OLED displays by lowering their operating current density at the target luminance.

1. Introduction

Since their development in 1987, organic light-emitting diodes (OLEDs) have become the next-generation display technology due to the remarkable recent advances in their device performance and mass-production technology. For small- and medium-sized OLED display applications, such as smart phones, tablets, and laptops, a full-color OLED display can be fabricated using vacuum deposition through fine shadow metal masks (FMMs) to form red-green-blue (RGB) sub-pixels. Currently, this mass-production technology is almost exclusively used to fabricate full-color OLED displays that implement RGB sub-pixels, but it is not applicable to relatively larger-sized OLED displays, such as TV applications. Alternatively, conventionally large OLED displays implement RGB color filters to a white OLED or apply red and green color conversion layers to a blue OLED. However, the addition of color filters or conversion layers leads to transmission and conversion loss, requiring OLED devices to emit more light under the same level of current density to reach a suitable luminescence level. Tandem OLEDs were first demonstrated in 2004 to overcome the limitations of conventional OLED architectures with a single electroluminescent (EL) unit [1].

Tandem OLEDs have multiple EL units connected electrically in series. Thus, they can emit more light than conventional OLEDs under the same current density, which means they have a much higher luminous efficiency (LE) and can be operated at a relatively low current density for the same level of target luminescence. As a result, the tandem structure can significantly improve the OLED lifetime and LE simultaneously. Theoretically, the LE of an N-fold (N being the number of EL units) tandem device can be enhanced N-fold, as N-fold luminance can be achieved from each EL unit with the same current density as that of a single EL-unit OLED.

For large-sized TV applications, state-of-the-art full-color OLED display technologies implement RGB color filters in conjunction with a white tandem OLED or quantum dot (QD) color conversion layers with a blue tandem OLED [2–4]. Fabrication of RGB OLED devices that implement highly efficient phosphorescent emitters for all RGB pixels is arguably the biggest challenge in the display industry because the lifetime of a blue emitter is significantly shorter than the lifetimes of red and green emitters. However, a phosphorescent blue emitter with two or more stacks of tandem structures could be implemented to circumvent the challenges associated...
with lifetime imbalance in full-color OLED displays that use QD color conversion layers.

The key enabler in tandem OLEDs is the intermediate connector, also known as the charge generation layer (CGL), which provides the electron and hole carriers for the EL units. CGLs, as fundamental interconnecting layers between EL units, must have low electrical resistance for a low operating voltage with less power loss, and LUMO and HOMO levels that are well-matched with the adjacent charge transport layers to prevent charge accumulation in the CGL. A high energy barrier in the junction interface between an n-CGL and a p-CGL causes a serious accumulation of charge carriers and increases the device operating voltage, which further degrades the device performance.

The CGL is typically a p-n junction that consists of a metal oxide/metal oxide bilayer [5], an organic doped n-type layer/p-type layer [6], or organic photo-voltaic heterojunction [7,8] structures. Of these, doped n-type layer/doped p-type layer CGLs composed of organic materials are commonly used because they are easy to fabricate through thermal deposition without a vacuum break and they have excellent electrical and optical properties. Most doped n-type layers (n-CGLs) are prepared by doping an electron transport layer (ETL) material with a low-work-function metal [9,10] or low-work-function metal salts [10,11] to enhance electron transport. Typical n-CGLs are tris(8-hydroxyquinolinato) aluminum (Alq3) doped with Mg, Ca, or Cs2CO3, and 2,9-Dimethyl-4,7-Diphenyl-1,10-Phenanthroline (BCP) doped with Li or Cs2CO3 [12,13]. BPhen doped with Li or Cs2CO3 is another type of n-CGL [14–16]. The most commonly used doped p-type layers (p-CGLs) are co-deposited metal oxides with a hole transport layer (HTL) material or a hole transport material doped with a strong electron acceptor, such as N,N0-bis-(1-naphthyl)-N,N0-diphenyl-1,10-biphenyl-4-40-diamine (NPB) doped with WO3, MoO3, or 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F4–TCNQ) [17–19]. The development of large-area OLEDs that implement a tandem architecture requires reliable and easily processable CGLs with a low excess voltage drop and high optical transparency. In this study, an organic n-type layer/p-type layer CGL doped using thermal vacuum deposition was investigated and applied to double- and triple-stack tandem OLED devices.

## 2. Experiment

Blue phosphorescent OLED devices with a single EL unit (SB), a double-stack EL unit (BB), and a triple-stack EL unit (BBB) were fabricated by implementing a doped organic n-type layer/p-type layer CGL. All the devices were fabricated on 50nm-thick ITO-deposited glass substrates. Using a patterned ITO as the anode electrode, a 5μm-thick polyimide film was coated and patterned to define an emission area of 2mm × 2mm. Organic layers and thin metal layers were fabricated using a thermal vacuum deposition system (SUNICEL Plus 200) under an ultra-high base vacuum pressure of <10−7 Torr. The wet-cleaned substrate was placed on a hot plate (250°C) for at least 30 min to remove residual moisture, and then treated with plasma (oxygen and argon mixture) for 10 s to reduce thehole injection barrier by increasing the work function of the ITO surface. For all the devices in this experiment, a 5nm-thick TAPC doped with 5wt.% of NDP-9 was utilized as the hole injection layer (HIL), and a 2nm-thick 8-Hydroxyquinolinolato-lithium (Liq) was utilized as the electron injection layer (EIL).

Approximately 100nm-thick Al was used as a cathode electrode for all (bottom emission) devices. The individual phosphorescence EL units consisted of a stacked structure of the HIL, the emitting material layer (EML), and the ETL. A 40nm-thick TAPC and 1,3,5-Tri(m-pyridin-3-ylphenyl)benzene (TmPyPB) were used as the HTL and ETL materials, respectively. The EML host material was tris(4-carbazoyl-9-ylphenyl)amine (TCTA), and the phosphorescence dopant was bis[2-(4,6-difluorophenyl)pyridinato-C2,N](picolinato)iridium (Flrpic). The thickness and doping concentration of the EML were 30nm and 12wt.% respectively, for all devices. A 10nm-thick Li-doped BPhen and a 10nm-thick NDP-9-doped TAPC were used as the n-CGL and the p-CGL, respectively. To investigate how different Li doping ratios in the n-CGL affect the overall performance of tandem devices, we fabricated a double-stack (BB) tandem device, a triple-stack (BBB) tandem device, and a single-EL (SB) reference device, as shown in Table 1 and Figure 1, and we compared their performance using different doping ratios of Li in BPhen from 0.5 to 4.0wt.%. We also measured the light transmittance values of the SB, BB, and BBB devices and of the CGL using a spectrophotometer (Konica Minolta, CM3500D).

### Table 1. Schematic layer structure of the OLED devices.

| Device | Layer structure | Remark |
|--------|----------------|--------|
| SB     | ITO (50 nm) / HIL / EL-B / Al (100 nm) | single EL |
| BB     | ITO (50 nm) / HIL / EL-B / CGL / EL-B / EIL / Al (100 nm) | double-stack EL |
| BBB    | ITO (50 nm) / HIL / EL-B / CGL / EL-B / CGL / EIL / Al (100 nm) | triple-stack EL |

*aNote aHIL:TAPC:NDP-9 (5 wt.%, 5 nm). bEL-B: TAPC (40 nm) / TCTA:Flrpic (12 wt.%, 30 nm) / TmPyPB (40 nm). cEIL: Liq (2 nm). dCGL: BPhenLi (x wt.%, 10 nm) / TAPC:NDP-9 (8 wt.%, 10 nm).
3. Results and discussion

The characteristics of the organic materials used in this experiment are listed in Table 2. The HOMO, LUMO, and triplet energy levels ($T_1$) were well aligned for the SB device, as shown in Figure 2. The carrier mobilities of the HTL/EML/ETL materials were those of the ideal device structure. NDP-9 is one of the most widely used and most effective p-type dopants due to its strong electron affinity. It has a deep LUMO level of 5.4 eV, which is energetically in the vicinity of the HOMO level of TAPC.

Table 2. Electrical characteristics of organic materials.

| Material | LUMO (eV) | HOMO (eV) | Hole mobility (cm$^2$/V·s) | Electron mobility (cm$^2$/V·s) | Remark |
|----------|-----------|-----------|-----------------------------|------------------------------|--------|
| Liq      | 3.15      | 5.58      | –                           | –                           | EIL    |
| TmPyPB   | 2.75      | 6.75      | $\sim 5.0 \times 10^{-6}$   | $\sim 1.0 \times 10^{-3}$  | ETL    |
| TCTA     | 2.45      | 5.83      | $\sim 3.0 \times 10^{-4}$   | $\sim 1.0 \times 10^{-8}$  | blue host |
| Flrpic   | 3.1       | 5.8       | –                           | –                           | blue dopant |
| BPPhen   | 3.0       | 6.4       | –                           | $\sim 5.4 \times 10^{-4}$  | n-CGL host |
| Li       | 2.93 (work function) | – | – | – | n-CGL dopant |
| TAPC     | 2.0       | 5.5       | $\sim 1.0 \times 10^{-3}$  | $\sim 5.0 \times 10^{-6}$  | HTL, p-CGL host |
| NDP-9    | 5.4       | 8.0       | –                           | –                           | p-CGL dopant |

Figure 2. Energy band diagram and triplet energy levels with energy transfer routes of the single EL unit device (SB).
Therefore, NDP-9 doping in TAPC is facilitated by electron transfer from the HOMO level of TAPC to the LUMO of NDP-9. The hole injection barrier height from the HTL to the EML is 0.55 eV. Although it may seem significant for transporting holes to the EML, holes can be injected directly into the HOMO level of the EML dopant, which is 5.8 eV for FIrpic. The HOMO level of the ETL (TmPyPB) is 6.75 eV, which is ideal for blocking excess holes escaping from the EML. TCTA is a common phosphorescent host material because of its large band gap ($E_g = 3.4$ eV) and high $T_1$ energy (2.86 eV). The $T_1$ levels of the blue dopant (Flrpic), ETL (TmPyPB), and HTL (TAPC) are 2.65 eV, 2.78 eV, and 2.87 eV, respectively. Hence, the diffusion of exciton energy to the HTL or ETL is prevented, as shown in Figure 2. Consequently, excitons are well confined in the EML. The doping concentration of FIrpic in TCTA was optimized to 12 wt.%. This doping ratio was used for all the devices in this experiment. As a result, we obtained a relatively low turn-on voltage and high luminous efficiency characteristics from the SB device.

The two main CGL features are charge generation and charge separation. Hence, upon charge generation at the n-p junction, electrons and holes should be rapidly injected into nearby EL units. To guarantee such charge separation, CGL materials with high charge mobility or conductivity are ideal. In addition, the energy barrier between the CGL and the ETL or HTL should be minimized, increasing the charge injection into the EL unit and reducing the charge accumulation. Figure 3 shows the energy band diagram and the carrier transfer route of the CGL implementing Li-doped BPhen as an n-CGL and NDP-9-doped TAPC as a p-CGL. Due to the high electron affinity of Li (2.93 eV), effective n-type doping can be easily achieved through co-deposition with BPhen. This n-type doping induces band bending in an n-CGL. Consequently, the effective electron injection barrier ($\Delta E_e$) at the n-p junction interface can be reduced. As $\Delta E_e$ depends on the doping concentration of Li and the thickness of the n-CGL, the accumulation of charge carriers in the CGL can be markedly suppressed by Li doping in the n-CGL, which is correlated with the shift of the light turn-on voltages to the lower bias and the enhanced LE and PE for tandem OLEDs. NDP-9 is the most effective p-type dopant due to its strong electron-accepting ability. Charges are generated in the NDP-9-doped TAPC layer, after which electrons are transferred to the ETL (TmPyPB) through BPhen, and holes are transferred to the HTL (TAPC). In this experiment, a 10 nm-thick Li-doped BPhen was tested as an n-CGL. To determine the optimal doping conditions for an n-CGL, the Li concentration in BPhen was split from 0.5 to 4.0 wt.%. A 10nm-thick TAPC doped with 8 wt.% of NDP-9 was used as a p-CGL for all tandem devices.

The effects of the Li doping in BPhen can be attributed to convenient electron transfer and limited gap state formation. The injection and transporting of electrons to the n-CGL from the p-CGL improved significantly as the Li doping concentration was increased from 0.5 wt.% to the saturation point at 1.0 wt.% as shown in Figure 4.

As shown in Figure 4(a), the operating voltages at the current density of 0.1 mA/cm² for the tandem devices were 7.7 ~ 9.5 V depending on the Li doping condition in the n-CGL. The tandem device with 1.0 wt.% Li doping in BPhen as the n-CGL showed the lowest operating voltage and the highest luminance characteristics, as shown in Figure 4(b). The operating voltages for 4,000 cd/m² were

![Figure 3. Energy band diagram and carrier transfer route of the CGL unit.](image-url)
14.5 ∼ 17.0 V depending on the Li doping condition in the n-CGL.

By implementing this optimized doping condition, 1.0 wt.% Li-doped BPhen, the BB and BBB devices and the SB device were fabricated simultaneously, without a vacuum break between the deposition processes.

The peak emission intensities of the SB, BB, and BBB devices under the same current density of 10 mA/cm² were almost double and triple the peak emission intensities of the SB device. However, as shown in Figure 5(a), the spectra shapes, such as the full-width half-maximums (FWHMs), the intensity ratios between the main and shoulder peaks of the spectra from the SB, BB, and BBB devices, differed. As the locations of the emissions and the gaps between the electrodes or cavities (117 nm for the SB device, 248 nm for the BB device, and 372 nm for the BBB device) were not equal, there was a slight change in the shoulder peak at 498 nm and at the FWHM due to the difference in the microcavity resonance effects. This microcavity resonance effect changes the measured luminescence intensities of devices in the normal direction, so it is used to evaluate device performance. As a result, the interpretation of the performance metric must reflect this effect to avoid misinterpretation of the results. In this experiment, however, this microcavity resonance effect was ignored in the evaluation of the device performance because the bottom emission device showed a weaker microcavity effect than the top emission device.

The maximum LE of the BB device increased from 22.5 to 42.5 cd/A, and of the BBB device, to 63.4 cd/A, almost double and triple, respectively, the LE of the SB device, as shown in Figure 5(c). Consequently, the current density for 4,000 cd/m² decreased dramatically from 28.5 mA/cm² to 11.2 and 6.5 for the BB and BBB devices, respectively. However, as shown in Figure 5(d), because of the power loss in the CGLs in the BB and BBB devices, the maximum PEs of the two devices dropped from 18.3 lm/W to 13.7 and 12.5, respectively.

As the emissions of the SB device are essentially independent of those of the other EL units in the tandem structure, and as the EL units are simple connections of two or three identical EL units in series, the voltage across the CGL can be calculated by subtracting the voltage sum of two or three single devices from the voltage of the tandem devices composed of two or three single devices at the same current density. Assuming that the voltages across the EL units in the tandem devices under the same current level were the same as the voltage of the single EL device, the voltage across the CGL was calculated from the voltage applied to the tandem device. Figure 6(a) shows the calculated CGL voltage of the BB device versus the current density characteristics.

As shown in Figure 6(a), when the tandem devices were operated at a low current density (< 5 mA/cm²), the current flow through the CGL unit increased exponentially with the voltage across the CGL unit. This suggests that the charge injection from the n-CGL/p-CGL interface to the EL units follows the exponential relationship of a typical p-n junction device, explained by the following equations:

\[ J = J_0 e^{(V/V_a) - 1} \]  
\[ \ln(J) = \frac{V}{V_a} + A, \text{ for } V \gg V_a. \]

However, as shown in Figure 6(a), this trend deviates from the exponential characteristics at a relatively high current density (> 5 mA/cm²). This is attributable to the increased carrier density due to the impact ionization under the relatively high electric field in the CGL units.
Figure 5. (a) Normalized EL spectra, (b) current density and luminance versus applied voltage, (c) luminous efficiency versus current density, and (d) power efficiency and luminance versus current density characteristics of the single EL (SB), double-stack tandem (BB), and triple-stack tandem (BBB) devices.

Figure 6. (a) Calculated CGL voltage versus current density characteristics of the triple-stack tandem device (BBB) and (b) light transmittance characteristics of the CGL, single (SB), double-stack (BB), and triple-stack tandem (BBB) devices.
The light transmittance loss in the CGL was negligible, whereas the light transmittance decreased considerably when the device became thicker. The organic deposited layers in the SB, BB, and BBB devices were 117, 248, and 372 nm thick, respectively. As shown in Figure 6(b), the light transmittance of the BBB device in the short-wavelength region slightly increased due to the microcavity resonance effect. As mentioned earlier in this paper, along with the CGL, this microcavity effect must be optimized to further improve the device performance.

4. Conclusion

We demonstrated highly efficient blue light-emission tandem PHOLED devices using a doped organic n-CGL/p-CGL architecture. By implementing this triple-stack tandem device architecture, the current density for a target luminance can be reduced dramatically, ultimately extending the device lifetime. Therefore, blue phosphorescent emitters with triple-stack tandem OLEDs in conjunction with QD color conversion layers can be used to manufacture large full-color OLED displays. The significance of the Li doping concentration in BPhen as an n-CGL revealed that the performance of tandem OLED devices could be optimized to nearly theoretical limits by implementing a 10 nm-thick layer of 1.0 wt.% Li-doped BPhen and a 10 nm-thick 8.0 wt.% NDP-9-doped TAPC as the n-CGL and the p-CGL, respectively. Compared with a single EL device, the performance of the double- and triple-stack tandem devices with optimized CGL units significantly improved: their maximum luminous efficiency increased from 22.5 cd/A to 42.5 and 63.4 cd/A, their current densities for 4,000 cd/m² decreased from 28.5 mA/cm² to 11.2 and 6.5 mA/cm², and their operating voltages at 0.1 mA/cm² increased from 3.1 V to 7.7 and 12.3 V, respectively.

Disclosure statement

No potential conflict of interest was reported by the author(s).

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