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Enhancement of Luminance Efficiency in Organic Light-Emitting Devices Utilizing Mixed Layers Embedded in Electron Transport Layers

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While the hole transport decreased in the mixed layer in the organic light-emitting devices (OLEDs) with an Al: lithium quinolate (Liq) mixed layers in the electron transport layer (ETL), the electron injection increased the Al:Liq mixed layer. The enhancement of luminance efficiency in the OLEDs with an Al:Liq mixed layer in the ETL originated from the more balance between the electrons and the holes in the emitting layer due to the decrease in the hole injection and the increase in the electron injection resulting from the existence of the Al:Liq mixed layer.

Keywords: electron injection; hole injection; luminance efficiency; mixed electron transport layer; mixed layer; organic light-emitting devices

I. INTRODUCTION

Organic light-emitting diodes (OLEDs) have been currently receiving considerable attention as potential candidates for applications in full-color flat-panel displays because of their unique advantages of low driving voltage, low power consumption, high contrast ratio, wide viewing angle, low cost, large flexibility, and fast response [1–4].

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Recently, OLEDs have been very attractive because of interest in their promising applications in large-area flexible displays. The enhancements of the efficiency in OLEDs utilizing various structures, such as a doped layer in a mixed host structure [5,6], a graded doping in a mixed host structure [7,8], multiple heterostructures [9,10], and an electron injection layer (EIL) [11–15] have been widely studied to improve the balance of the holes and the electrons in the emitting layer (EML). Recently, studies on the enhancement of the electron transport in a lithium (Li)-doped tris-(8-hydroxy quinoline)aluminum (Alq_3) electron transport layer (ETL) [16–20] and on the enhancement of hole blocking in an Al-doped organic layer [21] have become interesting due to their promising application in high-efficient OLEDs. Even though some works on enhancing OLED efficiency by using the doped double ETL have already suggested improvement of the balance of the holes and the electrons in the EML, systematic studies concerning the enhancement of efficiency in OLEDs with mixed layers embedded in the ETLs are still necessary to enhance the efficiency of OLEDs.

This article reports the electrical and the optical properties of OLEDs utilizing mixed layers embedded in the ETLs deposited by using organic molecular-beam deposition (OMBD). Current density-voltage (J-V) and luminance-voltage (L-V) measurements were carried out to investigate the electrical and the optical properties of the OLEDs without or with mixed layers embedded in the ETLs. The electrical properties of hole and electron only devices with mixed layers were investigated to clarify the carrier transport effect of the mixed layer in the OLEDs.

II. EXPERIMENTAL DETAILS

The sheet resistivity of the indium tin oxide (ITO) thin films coated on glass substrates used in this study was below $15\ \Omega/\text{square}$. The ITO substrates were cleaned using acetone and methanol at 60°C for 5 min, min, and then rinsed in de-ionized water thoroughly. After the chemically cleaned ITO substrates had been dried by using N_2 gas with a purity of 99.9999%, the substrates were treated with oxygen plasma for 10 minutes. Several kinds of samples used in this study were deposited on ITO-coated glass substrates by using OMBD with effusion cells and shutters. The detailed structures used in this study are summarized in Table 1. Device I is a reference OLED with a single ETL, and devices II and III are the OLEDs with an $\text{Al}:\text{Alq}_3$ mixed layer embedded in the ETL. Device III is an $\text{Al}:\text{Liq}$ mixed layer embedded in the ETL. The volume ratios of the $\text{Al}:\text{Alq}_3$ and the $\text{Al}:\text{Liq}$ mixed layers

TABLE 1 Device Structures of the OLEDs Used in this Work

Device	Structure
I	ITO (100 nm)/NPB (50 nm)/1% DCM-doped Alq ₃ (40 nm)/Alq ₃ (20 nm)/Al (100 nm)
II	ITO (100 nm)/NPB (50 nm)/1% DCM-doped Alq ₃ (40 nm)/Alq ₃ (15 nm)/Al:Alq ₃ (2:8) (5 nm)/Al (100 nm)
III	ITO (100 nm)/NPB (50 nm)/1% DCM-doped Alq ₃ (40 nm)/Alq ₃ (15 nm)/Al:Li _q (9:1) (5 nm)/Al (100 nm)
IV	ITO (100 nm)/NPB (50 nm)/Al (100 nm)
V	ITO (100 nm)/NPB (45 nm)/Al:Alq ₃ (2:8) (5 nm)/Al (100 nm)
VI	ITO (100 nm)/NPB (45 nm)/Al:Li _q (9:1) (5 nm)/Al (100 nm)
VII	ITO (100 nm)/BCP (5 nm)/Alq ₃ (45 nm)/Al (100 nm)
VIII	ITO (100 nm)/BCP (5 nm)/Alq ₃ (40 nm)/Al:Alq ₃ (2:8) (5 nm)/Al (100 nm)
IX	ITO (100 nm)/BCP (5 nm)/Alq ₃ (40 nm)/Al:Li _q (9:1) (5 nm)/Al (100 nm)

are 2:8 and 9:1, respectively. Devices IV, V, and VI are hole only devices for OLED devices I, II, and III, and devices VII, VIII, and IX are electron only devices for devices I, II, and III. The organic layers were deposited at a substrate temperature of 27°C and a system pressure of 5×10^{-8} Torr. The deposition rates of the organic layers and the metal layers were approximately 0.1 and 0.5 Å/s, respectively. The deposition rates were controlled by using a quartz crystal monitor. The size of the emitting region in the pixel was 3 mm × 3 mm. The current-voltage characteristics of the OLEDs were measured on a programmable electrometer with built-in current and voltage measurement units (model 236, Keithely). The brightness was measured by using a brightness meter, chromameter CS-100A (Minolta).

III. RESULTS AND DISCUSSION

The current density-voltage and luminance efficiency-current density characteristics of devices I, II, and III are shown in Figures 1 and 2. While the current densities of devices II and III utilizing mixed layers embedded in the ETL at the same voltages are lower than that of devices I, their luminance efficiencies are higher than that of device I. The efficiencies for OLEDs with mixed layers embedded in the ETL are enhanced because of the balance between the holes and the electrons in the EML due to a decrease of the hole transport resulting from the existence of the mixed layers. The hole or electron only devices were fabricated to investigate the variation of the electron or

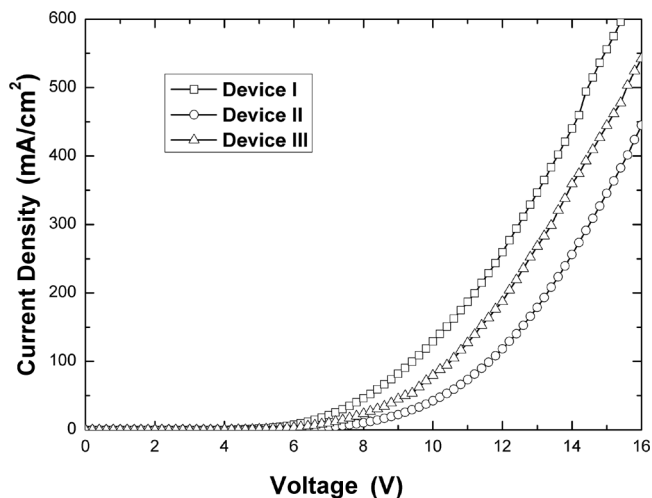


FIGURE 1 Current density-voltage characteristics of devices I, II, and III.

the hole transport current by using the mixed layers embedded in the ETL. The energy band structure of the electron only device decreases the hole injection due to an increase in the hole injection barrier, and that of the hole only device decreases the electron injection. Devices

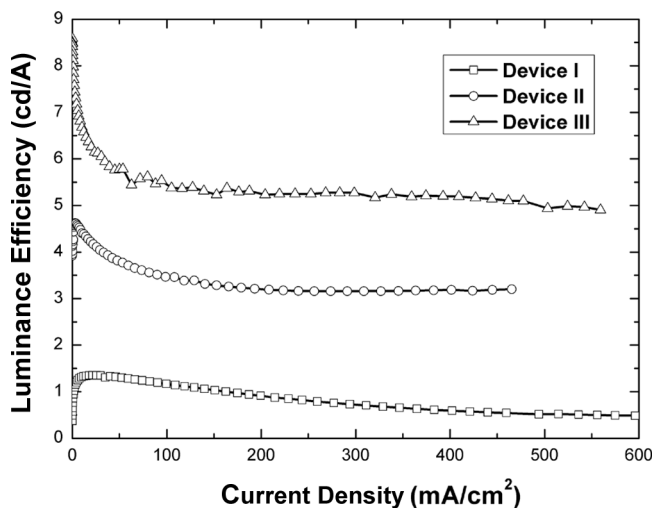


FIGURE 2 Luminance efficiency-current density characteristics of devices I, II, and III.

IV, V, and VI contain hole-only devices in comparison with OLEDs for devices I, II, and III. While the hole injection barriers in the hole only devices are below 0.5 eV, the electron injection barriers in the electron only devices are above 1.5 eV. The electron only devices corresponding to the OLED devices I, II, and III are denoted by devices VII, VIII, and IX.

Figure 3 shows the current densities as a function of the voltage characteristics of the hole-only devices without or with mixed layers embedded in the ETL. The hole only devices corresponding to devices I, II, and III are IV, V, and VI, respectively. The mixed layers embedded in devices II and III correspond to hole only devices V and VI. The current density of the device IV is higher than those of devices V and VI. Because the total current of hole only devices is almost equal to the hole currents due to the suppression of electron injection, the mixed layers existing in devices II and III decrease the hole current due to the formation of the hole capture sites in the doped Al atoms [21].

The current densities as functions of the voltage characteristics of the corresponding electron-only devices are shown in Figure 4. The devices VII, VIII, and IX correspond to the devices I, II, and III, respectively. The thin 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) layers between the ITOs and Alq₃ layers are inserted to prevent the hole injection in the electron only devices. While the current density of the device VIII with mixed layers of Al and Alq₃ is smaller than

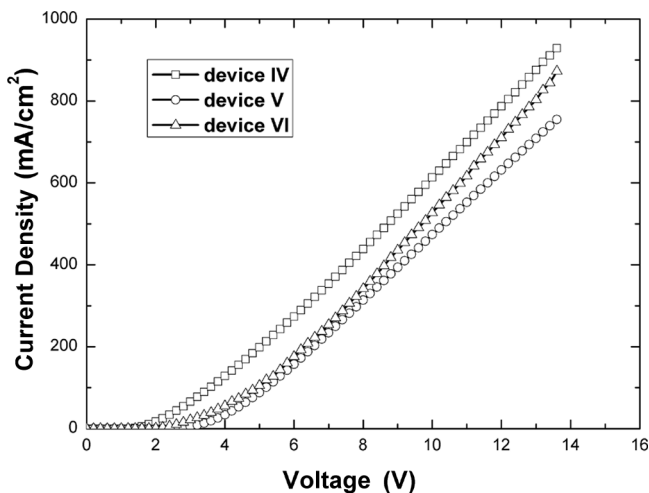


FIGURE 3 Current density-voltage characteristics of devices IV, V, and VI. The devices IV, V, and VI are the hole only devices in devices I, II, and III.

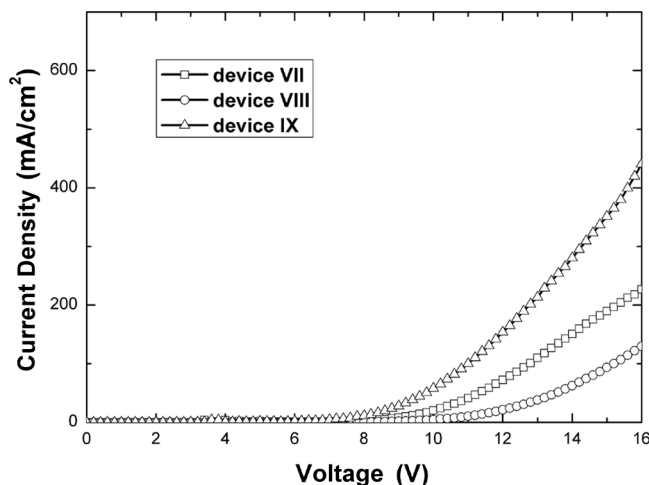


FIGURE 4 Current density-voltage characteristics of devices VII, VIII, and IX. The devices IV, V, and VI are the electron only devices in devices I, II, and III.

that of device VII, the current density of device IX with mixed layers of Liq and Al is the highest. The Li atoms in the mixed layer consisting of Al and Liq create a chemical reaction between the Al and the Liq layer. After the generated Li atoms form charge transfer complexes or Alq_3 anions, a better electron injection is achieved [15–16,19]. The trap sites in OLEDs are formed in the $\text{Al}:\text{Alq}_3$ mixed layer resulting from the chemical bonding of Al and Alq_3 [22], and the formed sites might affect the charge transport and the luminance mechanisms of OLEDs. The luminance efficiency is improved due to a decrease in the hole injection in device II. Because an increase in the electron injection together with a decrease in the hole injection occur in device III, the luminance efficiency of device III is the highest among the three devices.

IV. SUMMARY AND CONCLUSIONS

The electrical and optical properties of OLEDs utilizing mixed layers embedded in the ETL were investigated. The mixed layers consisted of $\text{Al}:\text{Alq}_3$ or $\text{Al}:\text{Liq}$. The electrical transport properties of the electron or hole only devices showed that the $\text{Al}:\text{Alq}_3$ mixed layer decreased the hole current and this induced a more balance rate of electrons and holes in the EML. The $\text{Al}:\text{Liq}$ mixed layer increased the electron

current, and the layer decreased the hole current, resulting in the enhancement of the more balanced rate of the electrons and the holes in the EML. The Li atoms generated from the chemical reaction of Al and Liq formed the charge transfer complexes or Alq_3 anions, resulting in the improvement of the electron injection in the Al:Liq mixed layer. These results can help improve understanding of the carrier transport mechanisms in the OLEDs with a mixed layer embedded in the ETL.

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