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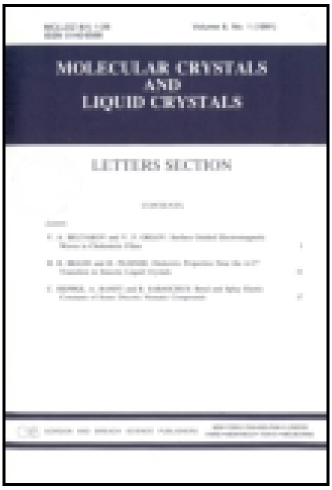

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Efficient Charge Balance of Green Organic Light Emitting Diodes Using Mixed Charge Transporting Materials as the Host

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Efficient Charge Balance of Green Organic Light Emitting Diodes Using Mixed Charge Transporting Materials as the Host

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The mixed layer for an emitting layer (EML) consists of two materials 4,4′,4″-tris(N-carbazolyl)-triphenylamine (TCTA) and 1,3,5-tris(N-phenylbenzimidazole-2-yl)benzene (TPBi) without host material. A series of EML structures were fabricated with three types of single mixed layers (mixing ratio 2:1, 1:2) and double mixed layers (mixing ratio 2:1/1:2), and the EML of using double mixed layer was optimized. The maximum luminous efficiency (LE) and quantum efficiency (QE) were 62.07 cd/A, 18.92%, respectively. Moreover, the roll-off ratio of LE was 20.42% at 20,000 cd/m^2. Interestingly, this result shows the roll-off ratio was reduced roughly 50% compared to the reference device.

Keywords Organic light emitting diode; mixed host; energy transfer; roll-off ratio; charge balance

Introduction

Phosphorescent organic light emitting diodes (PHOLEDs) have attracted interest due to their potential to achieve superior performance [1, 2]. In recent years, PHOLEDs needed to have high performance and low production costs because of the increasing demand in commercial applications. As a result, a lot of research has gone into high performance structures, such as double emitting layers (EML) and multiple quantum well structures [3, 4]. However, these structures have several problems, i.e., excessive charge accumulation at the organic interface can be generated, which quenches processes and increases driving voltage [5]. As a result, many groups have investigated other ways to reduce the number

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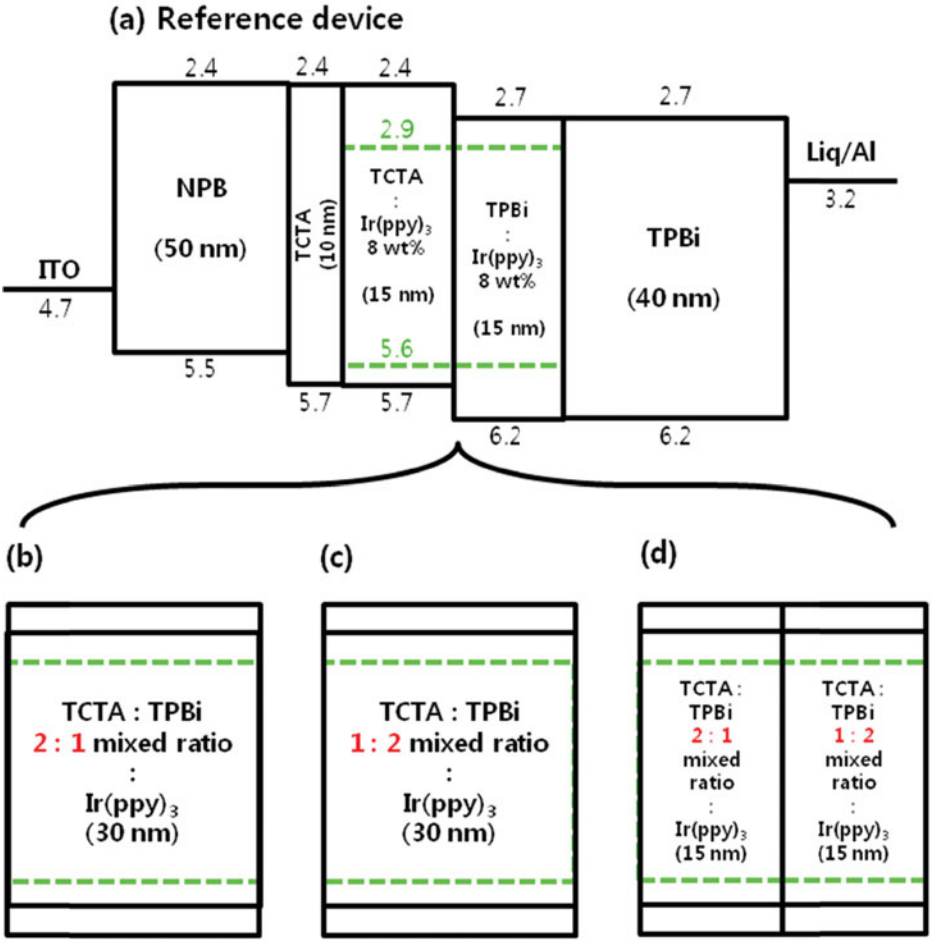


Figure 1. Device structure and energy level diagrams of the green PHOLEDs (devices A, B, C, and D).

of organic materials that have PHOLEDs [6, 7]. Above all, the charge balance factor is the key point in OLEDs.

In this respect, we introduced a mixed layer system that will satisfy these conditions. Here, the interface is removed between the EML and other EMLs in the double EML structure by adjusting the mixing ratio for two charge transporting layers (CTL) without host material. Furthermore, we suggest an efficient charge balance and injection property can be achieved with an appropriate CTL mixing ratio.

Experimental

Fabrication of OLED

Indium-tin-oxide (ITO) coated glass was cleaned in an ultrasonic bath in the following sequence: acetone, methyl alcohol, deionized water, and ethyl alcohol. It was then dried with a nitrogen gas blower and baked in a convection oven. Next, a pre-cleaned ITO was

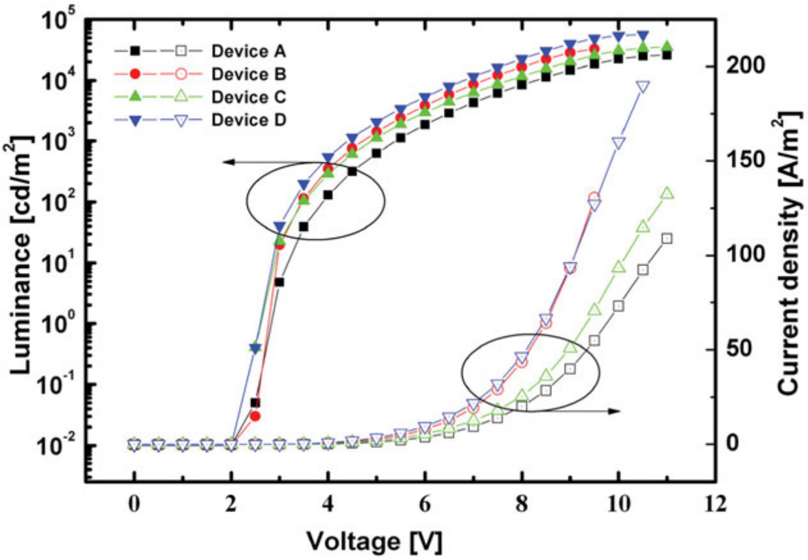


Figure 2. Luminance and current density versus voltage characteristics of devices A, B, C and D.

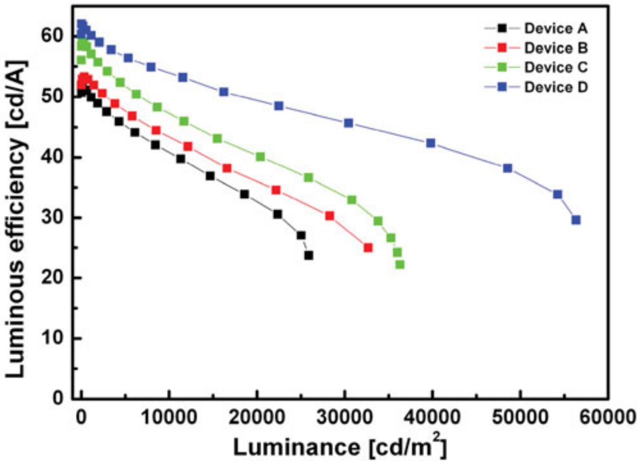
treated with an O_2 plasma with a power source of 125 W for 2 min under a low vacuum of 2×10^{-2} Torr. Green PHOLEDs were fabricated using the vacuum thermal evaporation method, and the organic layers were sequentially deposited onto an ITO-coated glass substrate (10 Ω/sq, emitting area 3 × 3 mm) without a vacuum break under a high vacuum (5×10^{-7} Torr). The deposition rate for all the organic materials was 1 Å/s and 0.1 Å/s for lithium quinolate (Liq). An aluminum (Al) electrode was then deposited at 10 Å/s. Finally, fabricated devices were sealed using a glass lid encapsulation method with barium oxide as desiccant in a nitrogen atmosphere to protect organic layers.

Measurements

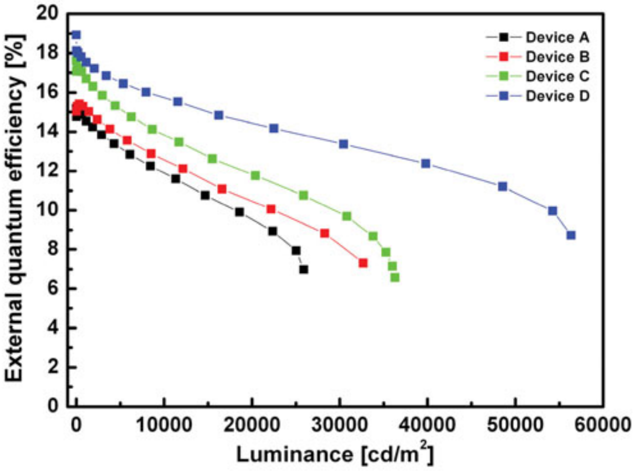
The DC voltage bias, the optical and electrical properties of the green devices, including current density, luminance, luminous efficiency (LE), external quantum efficiency (EQE), Commission International de L'Éclairage coordinates, and electroluminescence (EL) spectra characteristics, were measured with Keithley 236 and Chroma Meter CS-1000A instruments. All the measurements were carried out under ambient conditions at room temperature.

Results and Discussion

The structures and energy level diagrams of two CTL materials that were used for EMLs in the Green PHOLEDs are shown Fig. 1 [8, 9]. They have ITO (180 nm)/4,4′-bis[N-(naphthyl)-N-phenylamino]biphenyl (NPB, 50 nm) as a hole transporting layer, (HTL)/N,N′-dicarbazolyl-3,5-benzene and 4,4′,4″-tris(N-carbazolyl)triphenylamine (TCTA 10 nm) as exciton confinement layers, (ECL)/four types of EMLs (device A, B, C, and D, 30 nm) with single (S-EML) or double (D-EML) structures, 1,3,5-tris(N-phenylbenzimidazole-2-yl)benzene (TPBi, 40 nm) as an electron transporting layer, and (ETL)/Liq (2 nm) as an electron injection layer (EIL)/Al (100 nm). Device A, which was used as a reference device, consisted of 8 wt% [tris(2-phenylpyridine)iridium(III)]



(a)



(b)

Figure 3. (a) Luminous efficiency versus luminance characteristics. (b) External quantum efficiency versus luminance characteristics of green PHOLEDs (devices A, B, C and D).

(Ir(ppy)$_3$) as a phosphorescent green emitter. It has double EMLs, which used TCTA and TPBi as host materials. Devices B and C have a S-EML, which was formed as TCTA:TPBi with mixing ratios of 2:1 and 1:2 for the division of p-type and n-type, respectively. Device D has mixed double EMLs, and EML-1 and -2 hosts were used as TCTA:TPBi with mixing ratios of 2:1 and 2:1, respectively.

The difference exhibited in luminance and current density versus voltage curves is shown in Fig. 2. The driving voltage was dramatically reduced in the case of the TCTA:TPBi mixed ratio of 2:1 due to the TCTA. This differs from device B because it has more mixed TCTA, which has the fastest hole mobility of HTL materials [8]. It has a P-type host like neat TCTA. On the other hands, the driving voltage of device C was reduced slightly in spite of a higher TPBi ratio. However, LE and EQE are higher than in devices A and B, as shown in Fig. 3. Devices A, B, and C had a maximum LE value of 51.93 cd/A, 53.30 cd/A, and

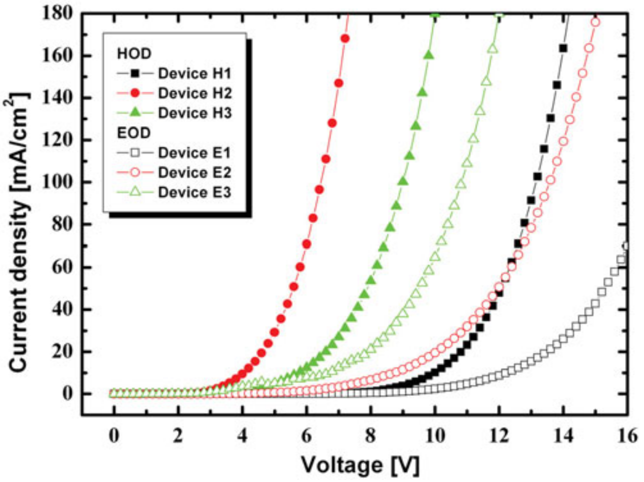


Figure 4. Current density versus voltage characteristics of devices H1∼H3 (HOD) and E1∼E3 (EOD).

59.02 cd/A, respectively, and EQE was 15.17%, 15.41%, and 17.62%, respectively. This difference could be explained by charge balance. In particular, carrier injection property and mobility are the key factors that determine charge balance efficiency in OLEDs. In order to clarify a charge balance factor, we fabricated additional devices to investigate carrier injection properties. The detailed device structures are as follows:

Device H1: ITO (180 nm)/NPB (50 nm)/TCTA (10 nm)/TCTA (15 nm)/TPBi (15 nm)/NPB (10 nm)/Al (100 nm)

Device H2: ITO (180 nm)/NPB (50 nm)/TCTA (10 nm)/TCTA:TPBi (2:1, 30 nm)/NPB (10 nm)/Al (100 nm);

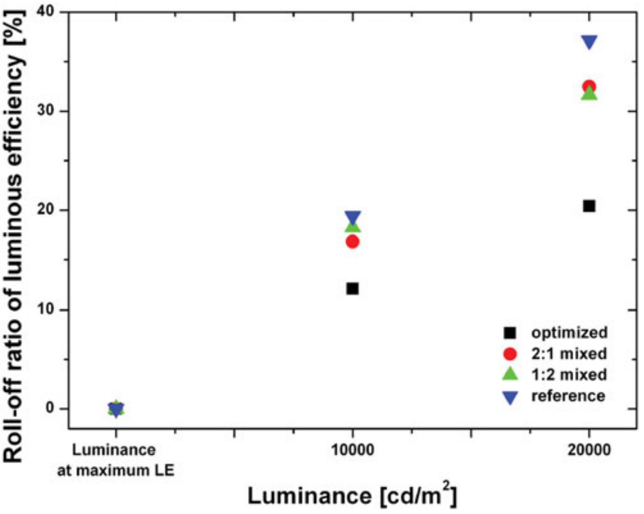


Figure 5. Roll-off ratio of luminous efficiency versus three point of luminance (devices A, B, C and D).

Device H3: ITO (180 nm)/NPB (50 nm)/TCTA (10 nm)/TCTA:TPBi (1:2, 30 nm)/NPB (10 nm)/Al (100 nm)

Device E1: ITO (180 nm)/TPBi (10 nm)/TCTA (10 nm)/TCTA (15 nm)/TPBi (15 nm)/TPBi (40 nm)/Liq (2 nm)/Al (100 nm)

Device E2: ITO (180 nm)/TPBi (10 nm)/TCTA (10 nm)/TCTA:TPBi (2:1, 30 nm)/TPBi (40 nm)/ Liq (2 nm)/Al (100 nm)

Device E3: ITO (180 nm)/ TPBi (10 nm)/TCTA (10 nm)/TCTA:TPBi (1:2, 30 nm)/ TPBi (40 nm)/Liq (2 nm)/Al (100 nm);

The current density versus voltage curves of each device is shown in Fig. 4. Device H1∼H3 and E1∼E2 were fabricated as a hole-only device (HOD) and electron-only device (EOD), respectively. The characteristic of current density versus voltage curve between HOD and EOD indicates that TCTA:TPBi with 1:2 mixed ratio is more charge balanced than other devices. As a result, device C exhibits higher efficiencies than device B. However, device D, which has a double EML (EML-1, EML-2), exhibits the highest LE and EQE despite device B has an efficient charge balance. Because, TPBi has a low hole mobility and a deep highest occupied molecular orbital level, whereupon hole injection was inefficient and accumulated at the HTL/EML interface. Therefore, we assume that the EML-1, which has a mixing ratio of 2:1 near p-type, was able to inject into the hole from an adjacent layer more easily, and the already injected hole was balanced with an electron at the EML-2, which has a mixing ratio of 1:2.

On the other hand, devices B, C, and D achieved higher performance compared with device A because device A's interface is a compulsory charge accumulation site, whereas the other devices have ambiguous interfaces due to mixed layers. Thus, excessive accumulation was reduced, which suppressed the quenching system. Consequentially, device D was optimized in this study, and it achieved a maximum LE of 62.07 cd/A and a maximum EQE of 18.92%, as shown in Fig. 3. In addition, the driving voltage is the same for device B. This indicates that the EML of device D has two advantages: efficient carrier injection property and charge balance. However, we needed to investigate the performance of an optimized device, such as the roll-off phenomena, in more detail.

Table 1. The value of luminous efficiency, external quantum efficiency and roll-off ratio (devices A, B, C, and D)

	Device A	Device B	Device C	Device D
LE [a] (cd/A)	51.93	53.30	59.02	62.07
LE [b] (cd/A)	41.85	44.33	48.24	54.55
LE [c] (cd/A)	32.64	36.00	40.33	49.39
EQE [a] (%)	15.17	15.41	17.62	18.92
EQE [b] (%)	12.19	12.84	14.12	15.91
EQE [c] (%)	9.53	10.46	11.83	14.43
Roll-off ratio of LE [b] (%)	19.41%	16.84%	18.26%	12.12%
Roll-off ratio of LE [c] (%)	37.13%	32.46%	31.66%	20.42%
Roll-off ratio of QE [b] (%)	19.64%	16.69%	19.86%	15.88%
Roll-off ratio of QE [c] (%)	37.13%	32.15%	32.83%	23.73%

[a] maximum value, [b] value at 8,700 cd/m^2, [c] value at 20,000 cd/m^2.

We considered another approach that investigated the roll-off ratio of LE at three points of luminance of the maximum LE, 8,700 cd/m^2 and 20,000 cd/m^2—as shown in Fig. 5 and Table 1. In the case of device D, the roll-off ratio was 12.12% and 20.42% at 8,700 cd/m^2 and 20,000 cd/m^2, respectively [11, 12]. Roll-off occurred using the quenching sequence, such as triplet-triplet or triplet-polaron annihilation, due to excessive generation of a triplet exciton in the narrow recombination zone (RZ) and mass energy transfer to the dopant. The triplet exciton has relatively long lifetime compared to the singlet exciton, even though the lifetime of the triplet exciton was shortened by using a metal complex. The interaction of triplet excitons results in the acceleration of a high electric field [13, 14]. However, a mixed host system extended the RZ due to the formation of bulk hetrojunction and an enhanced charge distribution ability [15]. Therefore, the roll-off ratio of tuned devices was reduced overall. In this respect, device D has an appropriate charge balance compared to other devices. In short, an EML-1 like device B could reduce driving voltage, and an EML-2 like device C could improve efficiency. Introducing both these EMLs has advantages. Furthermore, we expect this study to have commercial uses because all devices were fabricated using CTL without host material. In addition, this method will tune devices easily with a simple adjustment to the mixing ratio.

Conclusions

We demonstrated the electrical and optical properties of OLEDs with a simple mixing method and two mixing ratios. Device D was optimized with a D-EML structure, which has a TCTA:TPBi 2:1 mixed layer and a 1:2 mixed layer. It achieved a maximum LE and EQE of 62.07 cd/A and 18.92%, respectively. In addition, the roll-off ratio of the LE decreased in this device by 12.12% at 8,700 cd/m^2 and 20.42% at 20,000 cd/m^2 respectively. This was due to the RZ being extended by bulk heterojunction in the mixed layer and reaching an appropriate charge balance compared to the reference device.

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