Improved Performance of White Phosphorescent Organic Light-Emitting Diodes through a Mixed-Host Structure

Jonghee Lee, Jeong-Ik Lee, and Hye Yong Chu

Highly efficient white phosphorescent organic light-emitting diodes with a mixed-host structure are developed and the device characteristics are studied. The introduction of a hole-transport-type host (*N*, *N*'-dicarbazolyl-3-3-benzen (mCP)) into an electron-transport-type host (m-bis-(triphenylsilyl)benzene (UGH3)) as a mixed-host emissive layer effectively achieves higher current density and lower driving voltage. The peak external quantum and power efficiency with the mixed-host structure improve up to 18.9% and 40.9 lm/W, respectively. Moreover, this mixed-host structure device shows over 30% enhanced performance compared with a single-host structure device at a luminance of 10,000 cd/m^2 without any change in the electroluminescence spectra.

Keywords: Organic light-emitting diodes (OLED), white OLED, phosphorescent, mixed-host structure.

I. Introduction

Organic light-emitting diodes (OLEDs), especially phosphorescent OLEDs, have been actively investigated for both display and lighting applications due to their ability to efficiently utilize both singlet and triplet excitons [1], [2]. In particular, white OLEDs have been attractive candidates for future solid state lighting sources since their power efficiencies have surpassed those of incandescent bulbs [3].

There have been many reports on the development of efficient white OLEDs (WOLEDs) including the use of excimer formed by single dopant, a multi-elastic multilayer (EML) structure doped with different color emitting dopants, stacked OLEDs, a down-conversion structure, and the doping of several emitters in a single emitting layer [3]-[5]. The single EML structure makes it possible to obtain higher power efficiency, and color-stable WOLEDs prevent mismatches of energy levels in multiple EMLs; therefore, higher power efficiency and color-stable WOLEDs can be obtained [6], [7].

One effective method to obtain a lower driving voltage and higher efficiency in phosphorescent OLEDs (PHOLEDs) is to use a mixed-host structure instead of a single-host structure [8]-[12]. NPB:Alq3 or another fluorescent mixed-host structure was found to be effective in fluorescent OLEDs with improved efficiency and higher stability [8], [9]. More recently, a mixed-host structure for green PHOLEDs with various hole- or electron-transport-type hosts was proposed [10], [11]. However, several challenges still remain for adjusting mixed-host structures in blue and white PHOLEDs because there are few electron-transport-type wideband gap hosts, while there are

Manuscript received Apr. 1, 2009; revised May 18, 2009; accepted June 4, 2009.

This work was supported by the future technology development program of MOCIE/ITEP [2006-10028439, OLED Lighting].

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doi:10.4218/etrij.09.1209.0005

many hole-transport-type wideband gap hosts. M-bis-(triphenylsilyl)benzene (UGH3) is a weak electron-transport-type ultra-wideband gap material which has a good hole blocking ability. It is reported to be highly efficient in sky blue PHOLEDs with 20% peak external quantum efficiency [12]. We have taken advantage of the characteristics of UGH3 to develop blue and white PHOLEDs with a mixed-host structure.

In this work, we present high-efficiency phosphorescent white PHOLEDs which we developed with a mixed-host structure of a hole-transport-type mCP host and an electron-transport-type UGH3 host as an emissive layer. We have studied the device characteristics by changing the composition ratio of the two host materials in the emissive layer.

II. Experimental

As shown in Fig 1, a series of organic light-emitting devices in the current study were made using the following configuration: indium tin oxide (ITO) (70 nm)/4, 4'-bis[*N*-(1-naphthyl)-*N*-phenyl-amino] biphenyl (NPB) (40 nm)/*N,N*'-dicarbazolyl-3-3-benzene (mCP) (7.5 nm)/emissive layer (30 nm)/m-bis-(triphenylsilyl)benzene (UGH3) (5 nm)/ bathocuproine (2, 9-dimethyl-4, 7-diphenyl-1,10-phenanthroline (Bphen) (50 nm)/LiF (1 nm)/Al (120 nm). A blue light emitting iridium(III)bis(4, 6-difluorophenyl)-pyridinato-N,$C^{2'}$)picolinate (FIrpic) and an orange phosphorescent emitter, bis(2-phenylbenxothiozolato-N, $C^{2'}$)iridium (III) (acetylacetonate) (Bt_2Ir(acac)) were doped in a UGH3 or UGH3:mCP mixed host. The doping concentrations of FIrpic and Bt_2Ir(acac) were 14% and 0.3%, respectively. The standard device with a UGH3-only host was also prepared (device A), and mCP was used as a hole-transport-type host in the mixed structure devices (devices B to D) because it has high hole mobility as well as high triplet energy of 2.9 eV. The composition ratio of the UGH3 and mCP materials in the mixed-host structure

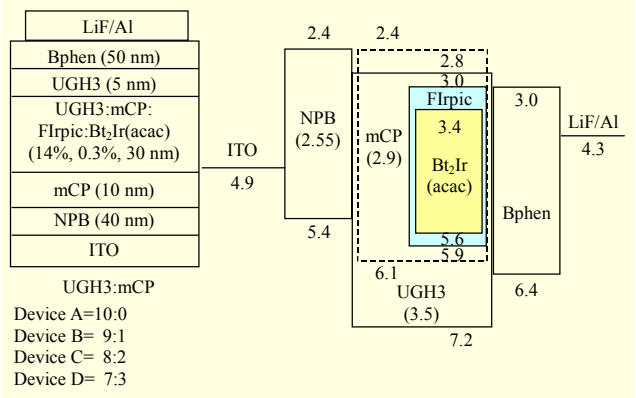

Fig. 1. Schematic diagrams of the structures of the white PHOLEDs and energy level diagrams for the materials tested in this study.

devices B to D were 9:1, 8:2, and 7:3, respectively. NPB and Bphen were used as a hole-transporting layer (HTL) and an electron transporting layer (ETL), respectively. The mCP interlayer at the HTL/EML and the undoped UGH3 interlayer at the EML/ETL interfaces were incorporated to enhance OLED performance by blocking triplet exciton quenching and reducing charge overflow. An energy level diagram of materials used in this study is shown in Fig. 1 [7], [13]. ITO was cleaned by the standard oxygen plasma treatment. OLED grade materials were purchased and used without further purification. All organic layers were deposited in a high vacuum chamber below 5×10^{-7} torr, and thin films of LiF and Al were deposited as a cathode electrode. The OLEDs were transferred directly from the vacuum chamber to an inert environment glove-box, where they were encapsulated using a UV-curable epoxy, and a glass cap with a moisture getter. The electroluminescence spectrum was measured using a Minolta CS-1000. The current-voltage (I-V) and luminescence-voltage (L-V) characteristics were measured with a current/voltage source/measure unit (Keithley 238) and a Minolta CS-100.

III. Results and Discussion

Efficient phosphorescent blue OLEDs of 20% peak external quantum efficiency have been reported which use the blue light emitting FIrpic doped ultra-wide bandgap UGH3 and adequate interlayers [13]-[15]. Based on these highly efficient blue OLEDs, we designed a white OLED by co-doping of a blue light-emitting FIrpic and an orange phosphorescent emitter, Bt_2Ir(acac), in a mixed-host structure of UGH3 and mCP (devices A to D). UGH3 is known as a weak electron-transport-type host material [13]-[15]; however, its charge carrier mobility, especially that of the hole carrier, is quite low. Therefore, most of the recombination process in the UGH3 emissive layer was carried out through direct trapping at the dopant site and positioned at the shallow region of the HTL/EML interface. Accordingly, it is expected that the introduction of a hole-transport-type mCP would result in a distributed recombination region and increased charge carrier mobility within the EML.

Figure 2 shows the current density-voltage-luminance curves of four devices according to the relative composition ratio of mCP and UGH3 host materials. In devices A to D, as the mCP composition ratio was increased, the current density of the device also greatly increased. The driving voltages of devices A to D at 5000 cd/m^2 reduced consequently and were 8.5, 8.0, 7.7, and 7.5 V, respectively. These higher current densities and reduced driving voltages in UGH3:mCP mixed-host devices can be explained as follows. As shown in Fig. 1, the highest occupied molecular orbital (HOMO) level of mCP is 6.1 eV

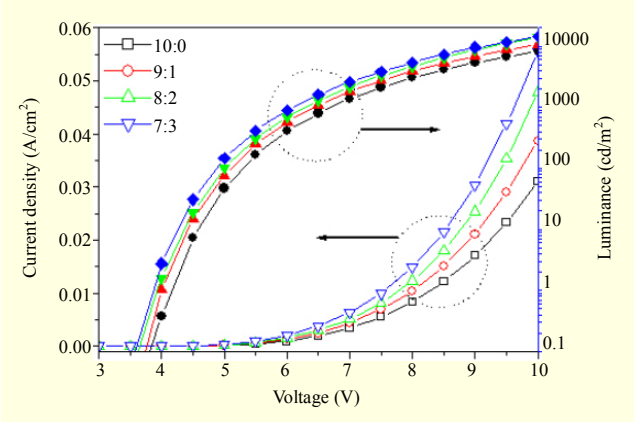

Fig. 2. Current density versus voltage (I-V) and voltage versus luminance (V-L) characteristics of the devices in this study.

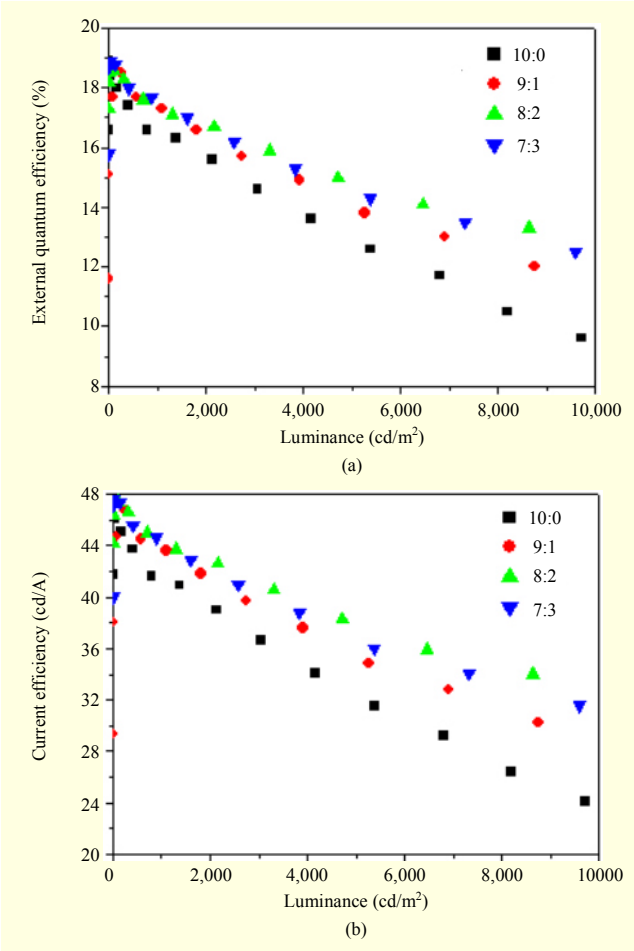

Fig. 3. Results for the white PHOLEDs: (a) external quantum efficiency versus luminance characteristics and (b) current efficiency versus luminance characteristics in this study.

(that of UGH3 is 7.2 eV), facilitating hole injection from the HTL side to the emissive layer. Moreover, mCP is also better than UGH3 in terms of hole mobility [13],[16],[17]. Therefore, proper combination of a hole-transport-type host and an electron-transport-type host can enhance charge carrier density and reduce the driving voltage in the emissive layer.

The external quantum efficiency and current efficiency of the mixed-host structure devices are plotted against luminance in Fig. 3 and summarized in Table 1. The peak quantum and current efficiency of the mixed host structure devices (devices B to D) compared with the UGH3-only host structure device (device A) were similar, while there were big differences in high luminance regions. As shown in Table 1, there was more than 30% enhancement of external quantum and current efficiency in UGH3:mCP (8:2) mixed-host device C at the high luminance region of 10,000 cd/m^2 compared with the UGH3-only standard device which showed almost the same performance at the low luminance region. At 10,000 cd/m^2, the external quantum efficiency and current efficiency of device A were 9.4%, and 22.6 cd/A. Those of device C were 12.8% and 32.9 cd/A, respectively.

In addition, efficiency *roll-off*, that is, the decrease of efficiency with increasing current density [18], [19] of mixed-host devices was less severe than that of the single-host device. The current density of the half-external quantum efficiency (J_0) for the UGH3 single-host device A and device C (UGH3:mCP=8:2) were 43 and 77 mA/cm^2, respectively. The J_0 of the mixed-host white PHOLEDs is almost two times that of the single-host device, exhibiting a reduced roll-off in efficiency.

Here, we discuss the reasons for the improved device performance and the reduced roll-off in the UGH3:mCP mixed-host devices at the high-luminance region. The recombination zone of the standard device A with only UGH3 is positioned at shallow region of the HTL/EML interface due to host (UGH3) properties such as HOMO/LUMO level and

Table 1. Device characteristics of white PHOLEDs in this study.

Device		EQE (%)	PE (lm/W)	CE (cd/A)	CIE coordinate $(x, y)^*$
A (10:0)	At peak	18.9	36.6	47.5	(0.431, 0.483)
	At 10,000 cd/m^2	9.4	7.7	22.6	(0.396, 0.466)
B (9:1)	At peak	18.7	41.1	47.4	(0.434, 0.487)
	At 10,000 cd/m^2	11.3	10.0	28.5	(0.399, 0.468)
C (8:2)	At peak	18.2	40.3	47.4	(0.436, 0.488)
	At 10,000 cd/m^2	12.8	11.9	32.9	(0.409, 0.474)
D (7:3)	At peak	18.9	40.9	47.6	(0.435, 0.486)
	At 10,000 cd/m^2	12.3	12.2	31.1	(0.396, 0.466)

* CIE coordinate (x, y) is determined from EL spectra.
EQE: external quantum efficiency, PE: power efficiency, CE: current efficiency

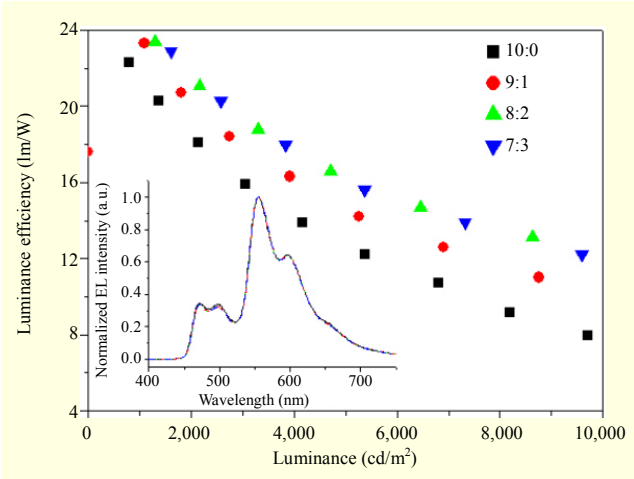

Fig. 4. Power efficiency versus luminance characteristics of the devices in this study (inset: electroluminescence spectra).

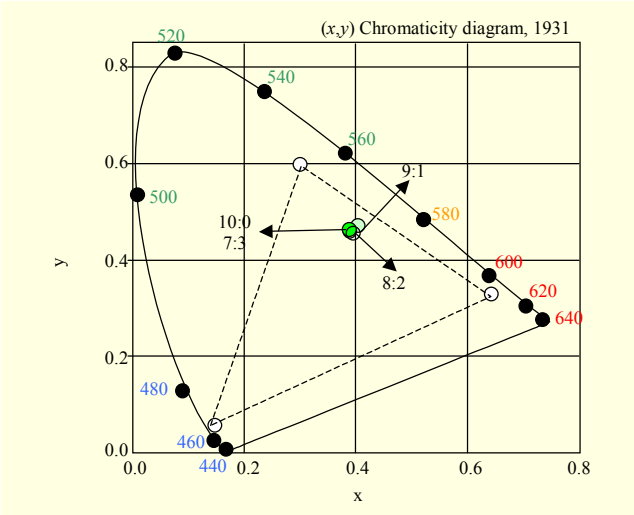

Fig. 5. CIE coordinates (x, y) at a luminance of 10,000 cd/m^2 of the devices in this study.

preferred charge transporting ability. Accordingly, as the current density of device A increases, triplet excitons lead to an inferior triplet quenching process such as the triplet-triplet annihilation (TTA) [18] or triplet-polaron annihilation (TPA) [19] in the emissive layer. However, a UGH3:mCP mixed-host structure resulted in effective hole and electron balance as well as a recombination zone shift from the HTL/EML interface to the center of the EML due to the introduction of mCP components into UGH3. Thus, the recombination zone was distributed over the entire emissive layer, and exciton quenching at high luminance was reduced. Additionally, the other electro-phosphorescent process, in which energy is transferred from added mCP material to dopants, could be beneficial to obtain high efficiency.

The power efficiency versus luminance of the devices in this study is shown in Fig. 4. As expected from improved external quantum efficiency and reduced driving voltage, the power efficiency of the UGH3:mCP mixed-host device C reached up to 40.9 lm/W. In particular, device C with a UGH3:mCP (8:2) mixed-host structure showed almost 60% improved power efficiency compared with device A at a luminance of 10,000 cd/m^2. We also found that this WOLED system exhibited little change in color coordinates from 100 to 10,000 cd/m^2, giving CIE coordinates from (0.43, 0.48) to (0.41, 0.47), (Δx = 0.02, Δy = 0.01) due to the single emissive layer structure (see inset of Figs. 4 and 5).

IV. Conclusion

In summary, the device characteristics of white PHOLEDs were investigated by using the mixed-host structure of a hole-transport-type host and electron-transport-type host materials. Adequate combination of host materials and combination ratio was effective to reduce the driving voltage; thus, the performance of white PHOLEDs was greatly improved by over 30% at a luminance of 10,000 cd/m^2.

Acknowledgement

We gratefully acknowledge Ms. K.-I. Song and Ms. S.J. Lee (ETRI) for assistance with EL measurements.

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