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Abstract

Highly efficient blue and white phosphorescent organic light-emitting diodes (PHOLEDs) with an exciton-confining structure were investigated in this study. Effective charge confinement was achieved by stacking two emitting layers with different charge-transporting properties, and blue PHOLEDs with a maximum luminance efficiency of 47.9 lm/W were developed by using iridium(III) bis(4,6-(difluorophenyl) pyridinato-N,C2')picolinate (FIrpic) as an electrophosphorescent dopant. Moreover, when the optimized green and red emitting layers were sandwiched between the two stacked blue emitting layers, white PHOLEDs (WOLEDs) with peak external and luminance efficiencies of 19.0% coupling technique and 54.0 lm/W, respectively, were obtained without the use of any out-coupling technique.

Keywords: organic light-emitting diodes, white phosphorescent, high efficiency, exciton

1. Introduction

Organic light-emitting diodes (OLEDs) have been actively investigated for both display and lighting applications [1-3]. In particular, white OLEDs (WOLEDs) are attractive candidates for future solid-state lighting sources for several reasons, including their low operating voltages and high power efficiencies as well as their suitability for processing on flexible substrates [4]. WOLEDs for solid-state lighting applications require high efficiency, a high color-rendering index (CRI), high color stability, and an appropriate color temperature [5, 6].

Many architectures for developing WOLEDs have been reported, such as the use of excimer or exciplex formed by one or two dopants, the stacking of several OLEDs (tandem OLEDs), the down-conversion of blue OLEDs, the doping of several dopants in a single emitting layer, and the doping of a multi-EML structure with different color-emitting dopants [4, 7]. Among these, the architecture of doping a multi-EML structure has advantages over the other architectures in terms of color controllability and efficiency because the recombination current, singlet and triplet energy transfer, and light-emitting performance of each layer can be easily controlled through the layer thickness and the doping concentration [7, 8].

There are various methods for developing highly efficient WOLEDs. In particular, the use of phosphorescent materials is an effective way of achieving high efficiency in the WOLEDs due to their ability to efficiently utilize both the singlet and the triplet excitons [3-7, 9-11]. Green or red phosphorescent OLEDs (PHOLEDs) with almost 100% internal quantum efficiency have been reported [3]. The light-emitting efficiency of the blue PHOLEDs, however, was reported to be much lower than those of the green and red PHOLEDs, thus restricting the performance of the WOLEDs [12-15].

In this work, high-efficiency WOLEDs with an exciton-confining structure were reported. To obtain highly efficient WOLEDs, high-performance blue PHOLEDs were
developed using a stacked emitting structure in advance. Consequently, very efficient WOLEDs with a maximum luminance efficiency of 54.0 lm/W at a luminance of 47 cd/m² was obtained, without the use of any out-coupling technique, through the insertion of green and red emitting layers sandwiched between two blue emitting layers.

2. Experiments

In this study, a series of PHOLEDs were fabricated using the following configuration: indium tin oxide (ITO) (70 nm)/1,1-bis[(di-4-tolyaminophenyl)cyclohexane (TAPC) (50 nm)/emissive layer/1,3,5-tri(m-pyrid-3-yl-phenyl) benzene (Tm3PyPB) (20 nm)/Tm3PyPB-Cs (30 nm)/LiF (1 nm)/Al (120 nm). Two emissive layers containing 7-wt% FIrpic co-deposited with the hole-transport-type host material 4,4′,4″-tri(N-carbazolyl)triphenylamine (TcTa, 10 nm) and 10-wt% FIrpic co-deposited with the electron-transport-type host material 2-(diphenylphosphoryl) spirofluorene (SPPO1, 10 nm) [16] were deposited successively. To fabricate a WOLED, a x-nm-thick layer of 7-wt% RD-doped TcTa and a y-nm-thick layer of 7-wt% tris-fac-(2-cyclohexenylpyridine)-iridium(III)-(Ir(chpy)₃)-doped TcTa were inserted between two blue emitting layers [17, 18] {device A: (x, y) = (0.5, 0.5); device B: (x, y) = (0.5, 1.5); and device C: (x, y) = (1.0, 1.0)}.

ITO was cleaned via the standard oxygen plasma treatment. The OLED-grade materials were purchased and used without further purification. All the organic layers were deposited in a high-vacuum chamber below 5 x 10⁻⁷ torr, and thin LiF and Al films were deposited as cathode electrodes. The OLEDs were transferred directly from vacuum into an inert environment glove-box, where they were encapsulated using 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 using a current/voltage source/measurement unit (Keithley 238) and a Minolta CS-100.

3. Results and Discussion

To develop efficient PHOLEDs, the effective confinement of both the charge carriers and the triplet excitons is necessary [13-16]. Therefore, TAPC and Tm3PyPB with a wider triplet energy level (T₁=2.9 and 2.78 eV, respectively, which are higher than that of FIrpic (T₁=2.62 eV)) as well as the low-lying highest-occupied molecular orbital (HOMO) and the high-lying lowest-unoccupied molecular orbital (LUMO) energy levels were used as a hole-transporting layer (HTL) and as an electron-transporting layer (ETL), respectively. The schematic diagrams of the structures of the WOLEDs, and the energy level diagram of the materials that were used in this study, are shown in Fig. 1.

![Fig. 1. Schematic diagrams of the structures of the WOLEDs, and energy level diagrams of the materials tested in this study.](image)

To improve the light-emitting efficiency of the WOLEDs, efficient blue PHOLEDs with the device architecture of a stacked emitting structure were developed and optimized. The blue PHOLED with a stacked emitting structure consists of two emissive layers with a hole-transport-type host of TcTa and an electron-transport-type band-gap host of SPPO1, both doped with a blue electrophosphorescent dopant of FIrpic. The optimized efficiency was obtained at FIrpic concentrations of 7 and 10 wt% in the TcTa and SPPO1 layers, respectively. The detailed investigation of this blue PHOLED will be reported elsewhere. By stacking two blue emitting layers with different charge-transporting properties, the expansion of the hole/electron recombination zone and the reduction of the triplet-quenching process in the emissive layer were achieved. Consequently, the blue PHOLED with a high luminance efficiency of 47.9 lm/W at a luminance of 33 cd/m² was achieved without any out-coupling enhancement, as shown in Fig. 2.

Based on this high-efficiency blue OLED, WOLEDs were established through the insertion of thin red and green emitting layers between two blue emitting layers. Fig. 3 shows the electroluminescence (EL) spectra for the WOLEDs. The EL spectra exhibited the peak wavelengths at 472, 536, and 620 nm in devices A, B, and C. These emissions correspond to the peak wavelengths of the EL spectra.

Fig. 2. Results for the blue PHOLEDs: the luminance efficiency vs. luminance characteristics (inset: electroluminescence spectra).

Fig. 3. The electroluminescence spectra of devices A, B, and C.

of the single-color OLEDs using the FIrpic, Ir(chpy)3, and RD dopants, respectively. The Commission Internationale de L’Eclairage (CIE) coordinates of these WOLEDs are (0.44, 0.48), (0.46, 0.50), and (0.51, 0.45) for devices A, B, and C, respectively. These slight spectral changes indicate that the insertion of thin red/green light-emitting layers at the TcTa/SPPO1 interface can effectively control the overall emission spectra, color temperature, and CRI within the WOLEDs. The CRIs and color temperatures of devices A, B, and C are 71 (3477 K), 70 (3244 K), and 76 (2371K), respectively, measured at the applicable luminance of 3000 cd/m². The CIE coordinates of the current WOLEDs may be yellowish white and not pure white. This can be resolved in this study by using a deep-blue emitter instead of a sky-blue lighting FIrpic, which is currently under investigation in related works.

The current density-voltage-luminance (J-V-L) characteristics of the WOLEDs are shown in Fig. 4. To obtain more efficient WOLEDs, particularly to achieve high luminance efficiency for solid-state lighting application, it is desirable that the driving voltage of the WOLEDs be lowered. As shown in Fig. 4, the driving voltages of all the WOLEDs at 1000 cd/m² were around 3.8 V and were almost identical. These low driving voltages even at a high luminance of 1000 cd/m² produced notable results and could be understood through thin emissive layers, a stepped progression of the HOMO of HTL and of the LUMO energy levels of ETL.

The quantum and luminance efficiencies of the emitted light in the forward direction of the WOLEDs vs. luminance are plotted in Fig. 5. The external quantum efficiencies of the WOLEDs in this study had very high values compared with those in the previous reports, indicating a successful charge balance and confinement in the light-emitting layer. The stacking of two blue light-emitting layers with different charge-transporting properties adjacent to the green/red emissive layers was employed to effectively confine the carriers and triplet excitons. The peak external quantum efficiencies of devices A, B, and C are 18.8, 19.0, and 17.6%, respectively, and the efficiencies of these WOLEDs had among the highest values reported to date. Moreover, the external quantum efficiency of device B remained high (10.2%) at a luminance of 17500 cd/m² and at a current density of about 70 mA/cm².

Furthermore, high external quantum efficiencies along
Fig. 5. Results for the WOLEDs: (a) the external quantum efficiency vs. luminance and (b) luminance efficiency vs. luminance characteristics.

with low driving voltages provided high luminance efficiencies in the WOLEDs. The maximum luminance efficiencies of devices A, B, and C are 50.4, 54, and 40.6 lm/W, respectively, which were achieved without the use of any out-coupling technique. Especially, device B exhibited a luminance efficiency of 52.6 lm/W at a luminance of 100 cd/m², which rolls off slightly to 37 lm/W at a luminance of 1000 cd/m². At the higher luminance values of 5000 and 10000 cd/m², the luminance efficiency remained 25.1 and 19.6 lm/W, respectively, still exceeding that of incandescent lamps.

4. Conclusions

Highly efficient WOLEDs for solid-state lighting application were developed through the exciton-confining emissive-layer structure, which can effectively confine the excitons within the light-emitting layer. By controlling each emissive-layer property, WOLEDs with a luminance efficiency of 37 lm/W at a luminance of 1000 cd/m² (the typical brightness for lighting applications) were achieved without using any out-coupling enhancement technique.

References

[18] RD is phosphorescent red dopant and the detailed information about this proprietary materials is disclosed later.