The effect of an EML sequence and an interlayer on the performance of the phosphorescent-fluorescent mixed WOLEDs

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Abstract

We investigate the effect of a light emitting layer (EML) sequence and an interlayer on the performance of the phosphorescent-fluorescent mixed white organic light emitting diodes. Two types of phosphorescent-fluorescent mixed system were evaluated. The proper position of each primary color EML was crucial to obtain best performance in each system whereas the effect of an interlayer was found to be different in both systems.

1. Introduction

White organic light emitting diodes (WOLEDs) are studied because of their application in lightings [1], full-color displays [2] and sheet backlight units for liquid crystal displays. The combination of phosphorescent and fluorescent emitter is an important approach to the production of WOLEDs with good performance.

Main approach is using the fluorescent blue emitter together with the phosphorescent green and red emitters [3, 4]. Another combination is using the phosphor-sensitized-fluorescence [5]. The reason for higher efficiency is the efficient utilization of both singlet and triplet excitons by properly selecting and positioning each dopant doped layer [3].

In addition, they commonly required an interlayer (non-emitter doped layer) to separate the phosphorescent and the fluorescent emitting regions thereby preventing the mutual quenching [4]. However, the insertion of interlayers causes a higher operating voltage as well as additional fabrication steps. Therefore, the selection of proper EML sequence and an interlayer would be crucial to obtain the best performance from this kind of emitter combination.

In this study, we systematically evaluated the effect of an EML sequence and an interlayer on the performance of the phosphorescent-fluorescent mixed WOLEDs.

2. Experimental

We prepared largely two sets of OLED devices: devices with EML combination of the fluorescent blue emitter and the phosphorescent green and red emitters, and devices with EML combination of the phosphor-sensitized-fluorescent yellow and separate blue emitters.

The basic OLED device structure and the energy levels of the materials used in this study are shown in figure 1. The OLED device structure we used is as follows: Indium-Tin-Oxide (ITO) anode / poly(3,4-ethylenedioxy-thiophene):poly(styrene sulfonic acid) (PEDOT:PSS, 40 nm) hole injection layer (HIL) / 4,4'-bis[N-(1-naphthyl)-N-phenyl-amino]biphenyl (α-NPD, 50 nm) hole transport layer (HTL) / various combination of the primary color EMLs / 2,9-dimethyl-4, 7 diphenyl-1, 10-phenanthroline (BCP, 10 nm) hole blocking layer (HBL) / tris(8-hydroxyquinolinato)-aluminum (Alq_3, 40 nm) electron transport layer (ETL) / LiF(0.5 nm) / Al(100 nm) cathode.

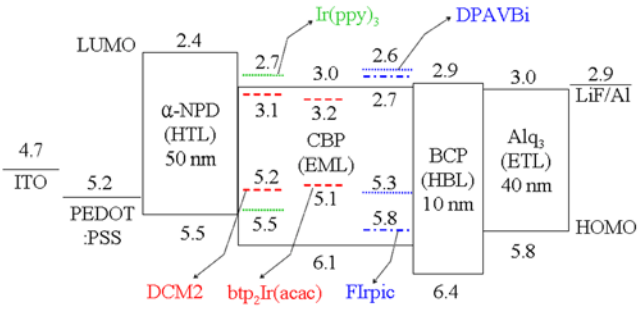

Fig. 1. The basic OLED structures used in this experiment and energy level diagram.

All layers were prepared with thermal evaporation onto UV-O_3 treated ITO substrates except the PEDOT:PSS layers, which were prepared by spin-coating at 4000 rpm and drying under vacuum of

about 10^{-3} Torr for 30 minutes. The substrates were ultrasonically cleaned by dipping them into various solvents such as isopropyl alcohol, acetone, and methyl alcohol prior to UV-O_3 treatment. Organic and metal evaporation were conducted under a base pressure of 5×10^{-6} Torr without breaking the vacuum, and the evaporation speeds were 1–2 Å/s for the organic materials and 4–5 Å/s for the metals. The doping concentration was adjusted by varying the relative evaporation speeds of the host and dopant materials and the evaporation speeds were monitored with a quartz-oscillator thickness monitor.

3. Results and discussion

3.1. The combination of phosphorescent red and green and fluorescent blue emitter in single host material

Four types of white OLEDs were prepared with various sequences of the primary color EMLs for device set-I: R/G/B in device W-1, G/R/B in device W-2, R/G/I/B in device W-3, and G/R/I/B in device W-4, in which a 10 nm 4,4′-*N*,*N*′-dicarbazole-biphenyl (CBP) layer doped with 8% bis(2-(2′-benzo[4,5-a]thienyl)pyridinato-*N*,C^{3}′)iridium(acetyl-acetonate) ($btp_2Ir(acac)$), a 5 nm CBP layer doped with 6% *fac*-tris(2-phenylpyridine) iridium ($Ir(ppy)_3$), a 10 nm CBP layer doped with 4% 4,4′-bis[2-{4-(*N*,*N*-diphenylamino)phenyl}vinyl]biphenyl (DPAVBi), and a 5 nm non-doped CBP layer were used as the red (R), green (G), and blue (B) EMLs and the interlayer (I), respectively.

Figure 2 shows the characteristics of the white devices. The inset shows the normalized EL spectra at 25 mA/cm^2 and the variation in the CIE 1931 chromaticity coordinates with the driving current density from 5 to 100 mA/cm^2.

Both higher external quantum efficiency (EQE) and stable chromaticity coordinate characteristics were obtained in devices with R/G EML sequence. A high external quantum efficiency of 8.5% (a current efficiency of 18.3 cd/A) at 100 cd/m^2 was obtained from device W-3. This can be attributed to the balanced charge carrier conduction in R/G EML sequence since uniform electron-hole recombination zone can be formed over a wide range of driving current density [6]. In contrast, the main recombination zone, which is positioned relatively toward the R layer due to its lower electron mobility, is shifted toward the G layer in the devices with G/R EML sequence at higher driving current density due to the increased electron conduction toward the G layer by increased electric field. Thus, relative intensity of green emission increases at high current density, resulting in large shift of the chromaticity coordinates for devices of W-2 and W-4 (see Fig. 2 (b)).

The emission from the phosphorescent EML positioned prior to the blue fluorescent EML was found to increase when the interlayer was used (green emission for W-3 and red emission for W-4), which indicates that triplet quenching by the non-emissive triplet level of the fluorescent dopant is prevented by the interlayer and thereby more higher efficiencies was obtained.

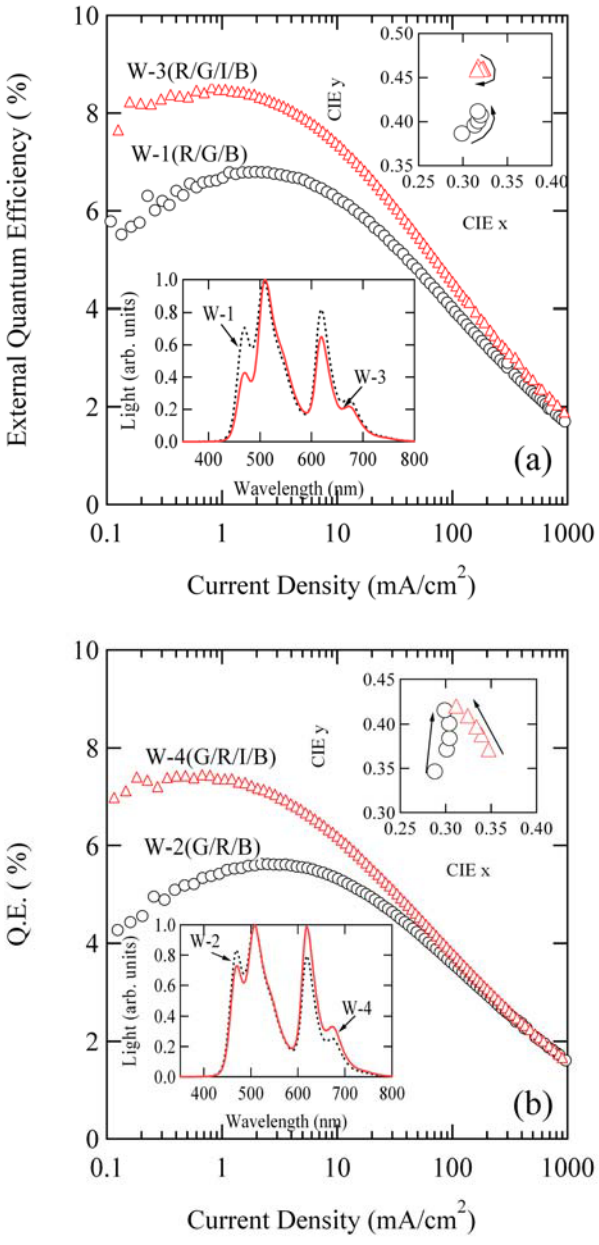

Fig. 2. The characteristics of the set-I white devices.

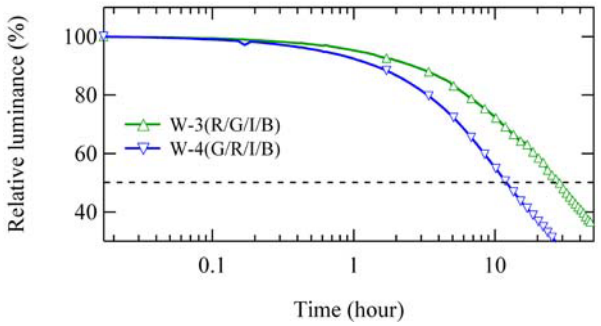

Fig. 3. The operating lifetime characteristics of selected white devices from set-I.

In addition, accelerated lifetime tests were conducted for the four white devices capped with a glass lid in an Ar-filled glove box. A constant current bias stress corresponding to an initial luminance of 1000 cd/m^2 was applied. The relative luminances are shown as functions of the operating time in figure 3. The device with an R/G EML sequence was found to have longer half-lifetimes (the time for the luminance to fall by 50%), with W-3 exhibiting the best characteristics. The half-lifetimes of the white devices are proportional to their efficiencies (at the initial luminance are obtained) as generally believed: Higher efficiency leads to the longer lifetime of devices when the same electroluminescence materials are used.

3.2. The combination of phosphor-sensitized-fluorescence yellow and separate phosphorescent blue EML

In the previous section, we showed that an interlayer as well as the EML sequence is important to obtain higher efficiency in the combination of phosphorescent red, green and fluorescent blue emitter. However, the insertion of interlayers causes a higher operating voltage as well as additional fabrication steps. Therefore, it is important to design a simpler device structure by reducing the number of interlayers while maintaining the overall device performance as much as possible.

17 nm CBP layer co-doped with 8 % Ir(ppy)$_3$ and 0.2% [2-methyl-6-[2,3,6,7-tetrahydro-1H,5H-benzo [ij]quinolizin-9-yl)ethenyl]-4H-pyran-4-ylidene] propane-dinitrile (DCM2) was used as the yellows (Y) EML and 13 nm CBP layer doped with 8 % iridium(III)bis[(4,6-difluorophenyl)-pyridinato-N,C2'] -picolinate (FIrpic) was used as the B EML for the device W-5(Y/I/B) and W-6(Y/B) of the device set-II.

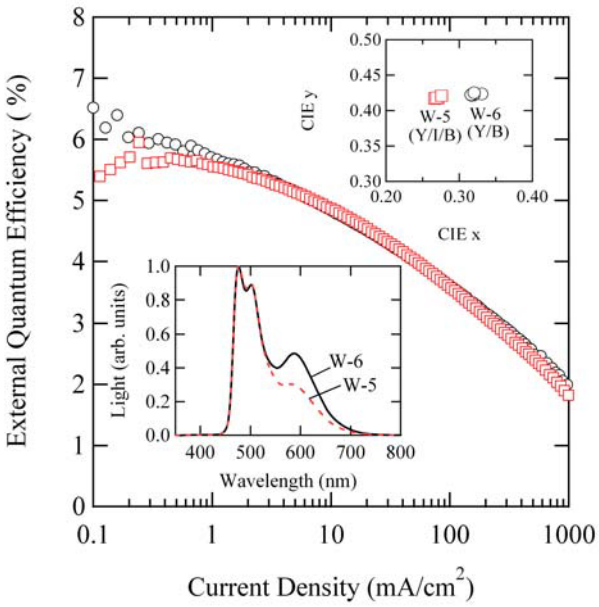

Fig. 4. The characteristics of the set-II white devices.

Differently from the results of the device set-I, there was no difference in the EQE of the device W-5 and W-6 as shown in figure 4 since there was no loss channel in this combination of EMLs even without an interlayer. The triplet energy level of FIrpic is higher than that of Ir(ppy)$_3$ and the singlet energy level of DCM2, this means energy transfer from the B to Y layer could be also utilized as the red (by DCM2) and green (by Ir(ppy)$_3$) emission. A slightly lower red emission of device W-5 compared to device W-6 could be attributed to the existence of an interlayer which prevents energy transfer from the B EML (FIrpic) to the Y EML (Ir(ppy)$_3$ and finally, to DCM2).

The device W-6 showed the peak EQE and current efficiency of 6.2 % and 14.3 cd/A respectively at 0.1 mA/cm^2 of driving current density. This is a reasonable figure when considering the peak EQE of each EML, because 30 nm CBP doped with 8 % FIrpic exhibited peak EQE of about 5 % in our prior experiment and 8 % peak EQE of phosphor-sensitized fluorescent Y EML is known [7].

In addition, the chromaticity coordinate change was very small; (0.32 ± 0.01, 0.42 ± 0.002) from 5 to 100 mA/cm^2 of the driving current density range were obtained in the device W-6. This well suppressed color coordinates shift characteristics mainly attributed to the relatively increased blue emission because almost all color shifts with the driving current density change was originated from the phosphor-

sensitized-fluorescent EML [8].

Consequently, we can remove an interlayer in this combination of the phosphor-sensitized-fluorescent yellow and the phosphorescent blue EMLs without sacrificing the overall performance of the device whereas the interlayer should be used in the previous section.

4. Summary

In the combination of phosphorescent red, green and fluorescent blue emitter in single host material, higher efficiencies, stable chromaticity coordinates and longer lifetimes were obtained for white OLEDs with an R/G/B EML sequence, which is optimal based on the charge carrier conduction properties of each EML, when compared to those with a G/R/B EML sequence. The use of an interlayer is also important to obtain higher efficiency.

In addition, we demonstrated that a simple and efficient white OLED could be achieved with the stack of the phosphor-sensitized-fluorescent yellow and phosphorescent blue EMLs without using an interlayer between them because the non emissive quenching among different emitters can be prevented in this combination of emitters.

5. Acknowledgements

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6. References

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