

## Correlation between host materials and device performances of phosphorescent white organic light-emitting diodes with blue/orange/blue stacked emitting structure

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### Abstract

A mixed host structure of TCTA and TPBI was used in orange emitting layer and host composition was critical to device performances of PHWOLEDs. PHWOLEDs with TPBI host in orange emitting layer showed high quantum efficiency of 10.3 % at 1000 cd/m<sup>2</sup> with little change of CIE coordinates of (0.32, 0.34) from 100 cd/m<sup>2</sup> to 10,000 cd/m<sup>2</sup>.

### 1. Introduction

In recent years, many device architectures of phosphorescent white organic light-emitting diodes (PHWOLEDs) have been developed to improve their efficiency, lifetime, and color performances. In particular, multi-stacked emitting layers structure has been known to give good color stability, good color tuning and high efficiency due to the control of charge distribution

The change of number of stacked emission layers and/or thickness of emitting layers were popular ways to control color performances of PHWOLEDs even though doping concentration management and energy level matching of organic layers have been developed as an approach to tune device performances of PHWOLEDs. Many different device architectures with different stacked emitting structures have been studied to tune CIE color coordinates and color stability in

PHWOLEDs. It was reported that stacked emitting structures with two adjacent emitting layers with an exciton blocking layer could control the white color coordinates<sup>1</sup>. Three multilayer structures consisted of three primary phosphorescent dopants doped in separate layers were effective to manage white color performances<sup>2</sup>. Another way to adjust white color coordinates was to change the layer thickness of the device<sup>3-4</sup>.

In this work, we developed PHWOLEDs with blue/orange/blue stacked emitting structure and investigated the relationship between host materials in orange emitting layer and device performances of PHWOLEDs. In addition, device performances of PHWOLEDs were correlated with recombination behavior of blue/orange/blue PHWOLEDs.

### 2. Experimental

Device configuration used in this work was indium tin oxide (ITO, 150 nm)/N,N'-diphenyl-N,N'-bis-[4-(phenyl-m-tolyl-amino)-phenyl]-biphenyl-4,4'-diamine (DNTPD, 60 nm)/N,N'-di(1-naphthyl)-N,N'-diphenylbenzidine (NPB, 20 nm)/N,N'-dicarbazolyl-3,5-benzene (mCP, 10 nm)/mCP: iridium(III)bis(4,6-(di-fluorophenyl)-pyridinato-N,C2') picolinate (FIrpic) (12 nm, 12 %)/4,4',4''-tris(N-carbazolyl)triphenylamine (TCTA):1,3,5-tris(N-phenylbenzimidazole-2-yl)benzene (TPBi):bis(1-phenylquinoline)

acetylacetonate ( $\text{Ir}(\text{pq})_2\text{acac}$ ) (6 nm, 10 %)/mCP:FIrpic (12 nm, 12 %)/2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP, 5 nm)/tris(8-hydroxyquinoline) aluminium (20 nm)/LiF (1nm)/Al (200 nm). Five different mixed host devices with different host composition were fabricated to investigate the correlation between mixed host composition and white color performances of the PHWOLEDs. The relative compositions of TCTA:TPBI in the red emission layer were 0:100, 25:75, 50:50, 75:25, and 100:0, which were denoted as 0 TCTA, 25 TCTA, 50 TCTA, 75 TCTA and 100 TCTA, respectively.

Red sensing devices with  $\text{Ir}(\text{pq})_2\text{acac}$  as a red sensing layer in blue emitting layer were also fabricated to investigate recombination zone of blue/orange/blue PHWOLEDs.  $\text{Ir}(\text{pq})_2\text{acac}$  was deposited at a thickness of 0.5 nm at the center of blue emitting layer and deep red intensity was monitored.

Current density-voltage-luminance (I-V-L) characteristics and electroluminescence (EL) spectra of PHWOLEDs were measured with Keithley 2400 source measurement unit and CS 1000 spectrophotometer.

### 3. Results and discussion

Blue/orange/blue stacked PHWOLEDs is advantageous in that good color stability can be obtained because the shift of recombination zone in orange light emitting layer can be compensated by two blue emitting layers. A mixed host emitting structure of TCTA and TPBI was applied in orange emitting layer and white device performances were correlated with host structures. TPBI has poor hole injection properties in spite of good electron injection properties and low current density was obtained in TPBI based PHWOLEDs. In the case of 100 TCTA device, poor electron injection properties of TCTA limited overall current density. Rather high current density in TPBI rich host structure among mixed host devices is due to the fact that electron is a minor carrier in mCP host structure. As electron injection is rather limited in mCP host structure, more electron injection from mCP to orange emitting layer can improve current density of blue/orange/blue stacked PHWOLEDs. Therefore, TPBI rich host structure was advantageous for current density due to efficient electron injection through TPBI.

Figure 1(a) shows the current density-voltage curves of PHWOLEDs with different host structures in orange emitting layer. The high current density in mixed host devices can be explained by efficient hole and electron injection through TCTA:TPBI mixed host structure. TCTA has the HOMO of 5.7 eV which is suitable for hole injection, while TPBI has the LUMO of 2.8 eV for electron injection. The combination of hole transport type TCTA and electron transport type TPBI can facilitate holes and electrons injection from mCP, increasing current density of blue/orange/blue stacked PHWOLEDs. TPBI has poor hole injection properties in spite of good electron injection properties and low current density was obtained in TPBI based PHWOLEDs. In the case of 100 TCTA device, poor electron injection properties of TCTA limited overall current density. Rather high current density in TPBI rich host structure among mixed host devices is due to the fact that electron is a minor carrier in mCP host structure. As electron injection is rather limited in mCP host structure, more electron injection from mCP to orange emitting layer can improve current density of blue/orange/blue stacked PHWOLEDs. Therefore, TPBI rich host structure was advantageous for current density due to efficient electron injection through TPBI.

Luminance-voltage characteristics of the PHWOLEDs are also shown Figure 1(b) Luminance was also increased in PHWOLEDs with mixed host emitting structure and showed the same tendency as current density. Quantum efficiency-luminance curves of the PHWOLEDs were plotted in Figure 1(c) PHWOLEDs with TPBI rich host structure exhibited high quantum efficiency, while other devices showed rather low quantum efficiency. In general, quantum efficiency is determined by holes and electrons balance in light emitting layer. Holes are major carriers in mCP based devices due to hole transport character of mCP, while electrons are minor carriers<sup>5</sup>. Therefore, holes and electrons balance can be improved if more electrons are injected from electron transport layer into emitting layer. Electron injection can be improved by electron transport type host materials and TPBI host in orange light emitting layer is better than other host materials in terms of electron injection. Therefore, high quantum efficiency was obtained in PHWOLEDs with TPBI rich host structure, while other devices showed rather low quantum efficiency. Best quantum efficiency of 10.7 %

at 1,000 cd/m<sup>2</sup> was achieved in PHWOLEDs with TPBI host in orange light emitting layer.

Figure 1(d) shows the EL spectra of the PHWOLEDs with the mixed host structure in orange light emitting layer. All EL spectra were normalized to compare relative contribution of blue and orange emission in PHWOLEDs. Strong orange emission with peak maximum at 602 nm was observed in PHWOLEDs with TPBI rich host, while PHWOLEDs with TCTA rich host structure showed rather weak orange emission. The intensity of orange emission in PHWOLEDs depends on recombination efficiency in orange emitting layer and triplet exciton density in orange emitting layer. In terms of recombination efficiency in orange emitting layer, 25 TCTA device would be better than other devices. In our previous work, optimum host composition for high efficiency in TCTA:TPBI mixed host devices with orange dopant was TCTA:TPBI (25:75) compared with TCTA:TPBI (50:50) of green phosphorescent devices<sup>6</sup>. Electrons are strongly trapped in orange or red devices with phenylquinoline type phosphorescent dopant and charge balance is optimized in TPBI rich mixed host structure. Similar results were also obtained in PHWOLEDs with mixed host emitting structure in orange emitting layer. Orange light emission was strong in PHWOLEDs with TCTA:TPBI (25:75) host structure, indicating optimized charge balance in orange emitting layer. Other than charge balance, exciton density which is closely related with recombination zone of PHWOLEDs is also important for the intensity of orange emission. Hole is a major carrier in mCP based phosphorescent devices and hole transport type host structure in orange emitting layer would shift recombination zone of PHWOLEDs from hole transport layer side to electron transport layer side. recombination zone of PHWOLEDs was close to hole transport layer side in PHWOLEDs with TPBI in orange light emitting layer, but it was shifted to electron transport layer side in PHWOLEDs with TCTA host in orange light emitting layer. In PHWOLEDs with TCTA host, holes and electrons recombination mostly happens in blue emitting layer near electron transport layer due to strong hole transport properties of TCTA, decreasing the intensity of orange light emission. Therefore, blue shift of color coordinate was observed in PHWOLEDs with TCTA rich host structure. In PHWOLEDs with TPBI host, recombination zone is rather concentrated inside orange emitting layer due to electron blocking

properties of mCP light emitting layer. Therefore, strong orange emission was observed in PHWOLEDs with TPBI host in orange emitting layer in spite of poor charge balance in TPBI emitting layer. CIE coordinate of 0 TCTA device was (0.39, 0.35), while it was (0.33, 0.34) in 100 TCTA devices. Except for 25 TCTA, blue shift of color coordinate according to TCTA content in mixed host structure was observed. Rather exceptional color coordinate of (0.41, 0.35) in 25 TCTA is due to strong orange emission originated optimized charge balance in TCTA:TPBI (25:75) mixed host structure.

#### 4. Summary

In summary, device performances of blue/orange/blue stacked PHWOLEDs could be controlled by changing host materials in orange light emitting layer. High quantum efficiency was obtained in PHWOLEDs with electron transport type TPBI host in orange light emitting layer, while high current density was obtained in the device with TCTA:TPBI (25:75) host structure. In addition, excellent color stability was observed in the device with TCTA or TPBI host and it could be correlated with the change of recombination zone according to voltage. Therefore, device performances of PHWOLEDs could be effectively managed by controlling host structure of PHWOLEDs.

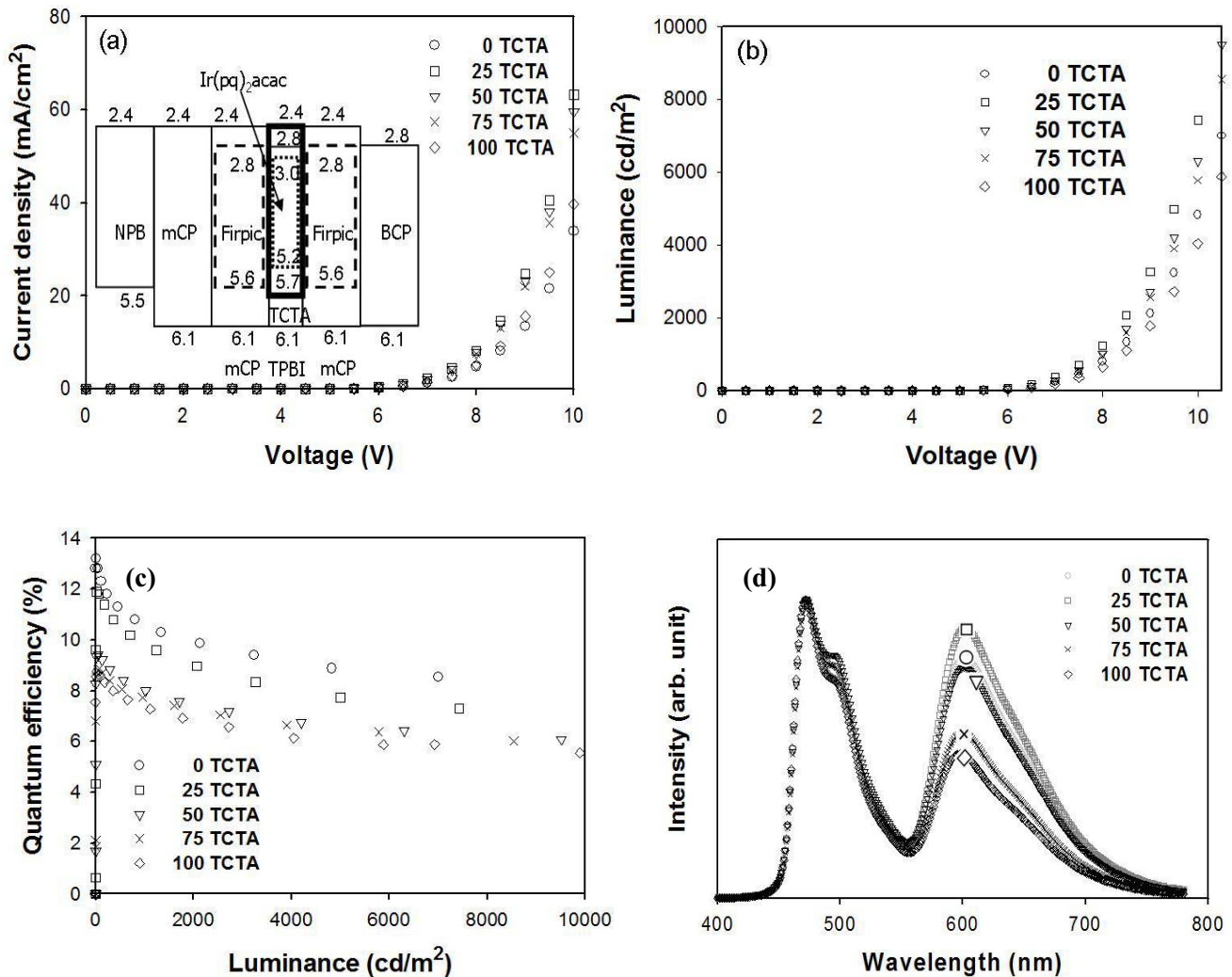
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**Figure 1. Characteristics of five different PHOLEDs with various compositions of mixed host (a) Current density-voltage curves (inset : energy level diagram) (b) Luminescence-voltage curves (c) Quantum efficiency-luminance curves (d) Electroluminescence spectra.**