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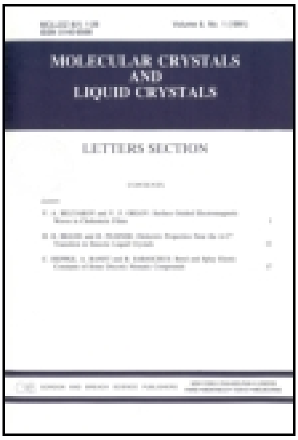

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Low Driving Voltage and High Efficiency Blue Phosphorescent OLEDs with Mixed Host System

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Low Driving Voltage and High Efficiency Blue Phosphorescent OLEDs with Mixed Host System

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We report both low driving voltage and high efficiency characteristics in blue phosphorescent organic light-emitting diodes with a simple device structure. A mixed host system consisting of narrow band-gap host material of 4,4',4"-tris(N-carbazolyl)triphenylamine (TCTA) and bipolar carrier transporting material of 2,6-bis(3-(carbazol-9-yl)phenyl)pyridine (26DCzPPy) shows low driving voltage performances with high luminance stability. A very low onset voltage of 3.0 V and a driving voltage of 3.8 V to obtain a brightness of 1000 cd/m^2 are achieved with a current efficiency of 44.2 cd/A.

Keywords Blue phosphorescent; mixed host system; organic light-emitting diode

Introduction

Since the first report of organic light emitting diode (OLED) in 1987 [1], this very simple organic device has evolved dramatically over the last decades thanks to several scientific efforts in both materials science and device physics, for example, introduction of phosphorescent emitters [2], host-guest system [3], multi-layer structure [4–6], and so on. All these attempts have resulted in high quantum efficiency and low driving voltage characteristics, which leads to the current OLED production. Especially electro-phosphorescence devices have been regarded as the key technology in high efficiency OLED products due to theirs theoretical 100% internal quantum efficiency which is four times higher than conventional electro-fluorescent devices. Up to date, highly efficient red, green, and blue phosphorescent OLEDs (PHOLEDs) with over 20% external quantum efficiency (EQE), an almost ideal value, have been reported [7–9].

In order to facilitate the exothermic energy transfer from the host to the dopant molecules, the general PHOLEDs demand high triplet energies of host materials at least over 2.8 eV. In addition this, about 0.5 ∼ 1.5 eV exchange energy of singlet to triplet is also considered for host materials. Therefore, general current blue phosphorescent host materials have wide band-gap characteristics over 3.5 eV. As the results, most reported blue PHOLEDs based on these wide band-gap host materials show high driving voltage characteristics over 5 V [10–12]. The main reason of such high driving voltage performances

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Table 1. Key performance characteristics of fabricated five blue PHOLEDs.

	Device A	Device B	Device C	Device D	Device E
Turn-on Voltage	3.4 V	3.6 V	2.8 V	3.2 V	3.0 V
Operating Voltage	7.0 V	4.7 V	3.5 V	4.0 V	3.8 V
Efficiency	32.3 cd/A 14.4 lm/W	28.0 cd/A 15.7 lm/W	29.1 cd/A 26.7 lm/W	36.4 cd/A 28.3 lm/W	44.2 cd/A 36.6 lm/W
Maximum Efficiency	36.6 cd/A 25.6 lm/W	31.0 cd/A 20.3 lm/W	34.4 cd/A 36.0 lm/W	41.3 cd/A 38.2 lm/W	47.1 cd/A 45.0 lm/W
Maximum Luminance	13930 cd/m^2	11400 cd/m^2	5144 cd/m^2	10760 cd/m^2	8413 cd/m^2
CIE (x,y)	(0.157, 0.313)	(0.161, 0.333)	(0.155, 0.320)	(0.156, 0.318)	(0.160, 0.332)

in current blue PHOLEDs is carrier trapping phenomena arisen from the big energy differences between wide band-gap of host and narrow band-gap dopant materials. The direct charge carrier movement and recombination at dopant molecules lead to high driving voltage performances. Hence, high dopant concentration in such host dopant systems could help to reduce driving voltage through the connection of dopant molecules [13]. However, we should sacrifice the device efficiency due to concentration quenching between dopant molecules [14]. So, it is very difficult to achieve both high efficiency and low driving voltage performances simultaneously in blue PHOLEDs with current wide band-gap host materials.

In this paper, we report both low driving voltage and high efficiency characteristics in blue PHOLEDs with a mixed host system. A very low onset voltage of 3.0 V and a driving voltage of 3.8 V to reach 1000 cd/m^2 brightness are achieved with a current efficiency of 44.2 cd/A.

Experimental

To make blue PHOLED devices, sublimated grade di-[4-(N,N-ditolyl-amino)-phenyl] cyclohexane (TAPC) from Luminescence Technology and 1,3,5-tri[(3-pyridyl)-phen-3-yl]benzene (TmPyPB) from Daejoo Electronic Materials were used as a hole transport layer and an electron transport layer, respectively. Bis(3,5-difluoro-2-(2-pyridyl)phenyl-(2-carboxypyridyl)iridium(III) (FIrpic) from Luminescence Technology was used as a blue phosphorescent dopant material. 1,3-Bis(carbazol-9-yl)benzene (mCP), 2,6-bis(3-(carbazol-9-yl)phenyl)pyridine (26DCzPPy), and 4,4’,4”-tris(N-carbazolyl)triphenylamine (TCTA) from Luminescence Technology were used as phosphorescent blue host materials. In this work, we have fabricated several blue PHOLEDs using the following simple device structure (Fig. 1): ITO/ TAPC (45 nm)/ host: FIrpic (15 nm)/ TmPyPB (40 nm)/ LiF (0.5nm)/ Al (100nm). To fabricate blue PHOLEDs, we use clean glass substrates coated with a 150 nm thickness of ITO layer which having a sheet resistance 10 Ohm/square as an anode. The active patterns size of 2 × 2 mm^2 were formed by the photolithography and wet etching processes. The ultrasonic cleaned glass substrate in an isopropyl alcohol,

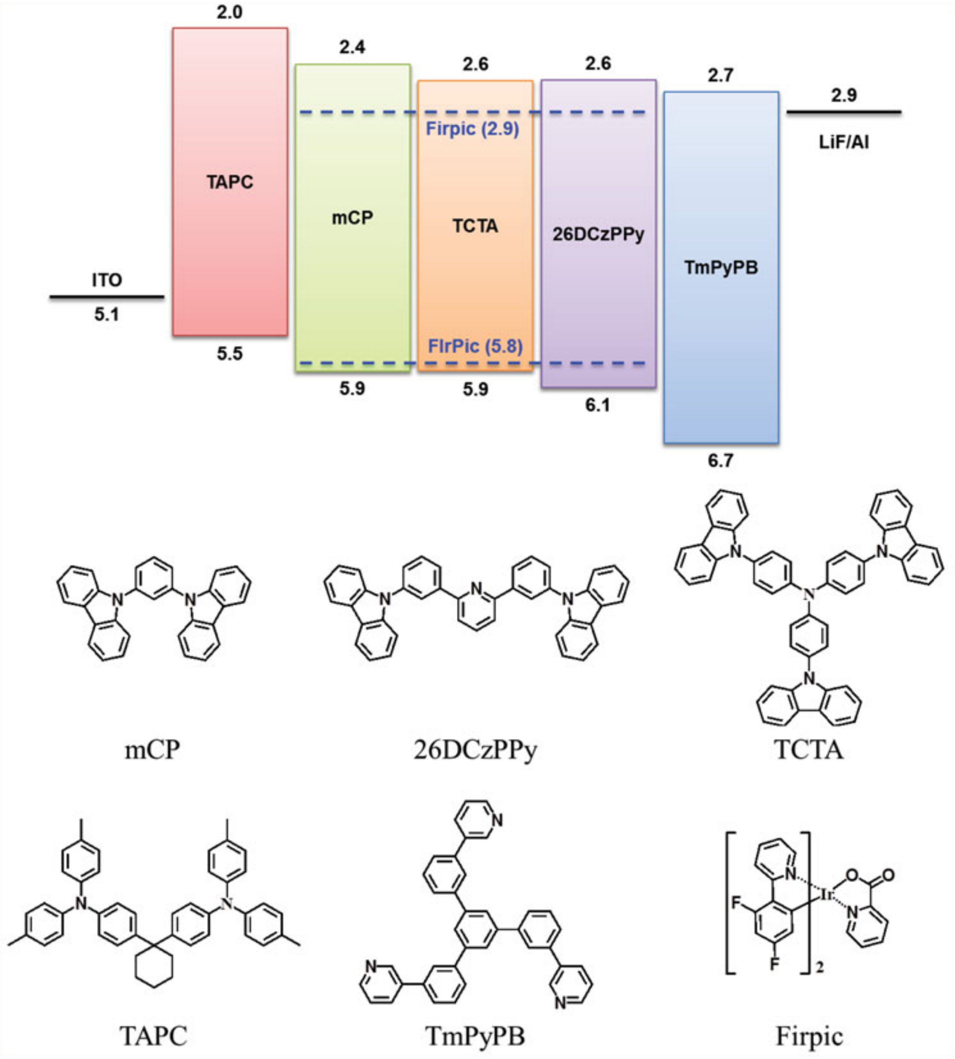

Figure 1. The device structure with the energy band diagram and molecular structures of used materials.

acetone, and methanol was rinsed in deionized water, and finally was treated in ultraviolet (UV)-ozone for 3 min. The ozone gas inside the chamber was generated by using UV light to excite the oxygen in the air. Each organic layer was deposited under a pressure of $\sim 10^{-7}$ torr with a deposition rate of ~ 0.5 Å/s. Subsequently, 0.5 nm thickness of lithium fluoride (LiF) and 100 nm thickness of aluminum (Al) were deposited in vacuum chamber without breaking the vacuum and used as a cathode. The current density-voltage (J-V) and luminance-voltage (L-V) data of blue PHOLEDs were measured employing a Keithley SMU 238 and a Minolta CS-100A. Electroluminescence (EL) spectra and Commission Internationale de I'Eclairage (CIE) color coordinate were obtained using a Minolta CS-1000A.

Results and Discussion

Four blue PHOLED devices were fabricated by using mCP, 26DCzPPy, TCTA, and 26DCzPPy: TCTA as phosphorescent blue host materials. These three host materials contain carbazole structures which are well-known hole transporting chemical moiety [15]. Hence, the mCP and TCTA show mainly hole transporting characteristics. On the other hands, 26DCzPPy was reported as a bipolar carrier transporting material attributed to combining carbazole hole transporting and pyridine electron transporting moieties [16]. Four blue PHOLEDs were fabricated by varying host materials with a following structure: "ITO/ TAPC (45 nm)/ host (mCP, 26DCzPPy, TCTA, 26DCzPPy: TCTA (1:1 ratio)): 3 % FIrpic (15 nm)/ TmPyPB (40 nm)/ LiF (0.5 nm)/ Al (100 nm)". Device A, B, and C are assigned for mCP, 26DCzPPy, and TCTA single host system, respectively. Device D has 26DCzPPy: TCTA mixed host system with the same emissive layer (EML) thickness. The optimized device with 26DCzPPy: TCTA mixed host system is assigned as the Device E: "ITO/ TAPC (50 nm)/ 26DCzPPy : TCTA (1:1 ratio) : 3 % FIrpic (10 nm)/ TmPyPB (40 nm)/ LiF (0.5 nm)/ Al (100 nm)". The Device E has thinner EML thickness from 15 nm to 10 nm and thicker TAPC thickness from 45 nm to 50 nm. Figure 1 shows the device structures with the energy band diagram for five fabricated blue PHOLED devices and each molecular structure of used materials. Figure 2 shows the J-V-L characteristics of fabricated blue PHOLEDs. In single host system devices, mCP host based device (Device A) shows very high onset voltage and driving voltage of 3.4 V and 7.0 V, respectively, due to wide bandgap characteristics of mCP which cause charge trapping in dopant molecules. Especially, deep electron traps at lowest unoccupied molecular orbital (LUMO) energy level of FIrpic molecule are expected to occur due to the big difference between the LUMO energy of mCP and FIrpic, 0.5 eV. Because electron mobility is relatively lower than hole mobility in this case, these electron traps seems to give high onset and driving voltage characteristics. Device A shows the current and power efficiency at driving voltage of 32.3 cd/A and 14.4 lm/W at 1000 cd/m^2, respectively (see Fig. 3). The 26DCzPPy host based device (Device B) shows lower onset and driving voltages as 3.6 V and 4.7 V, respectively. In spite

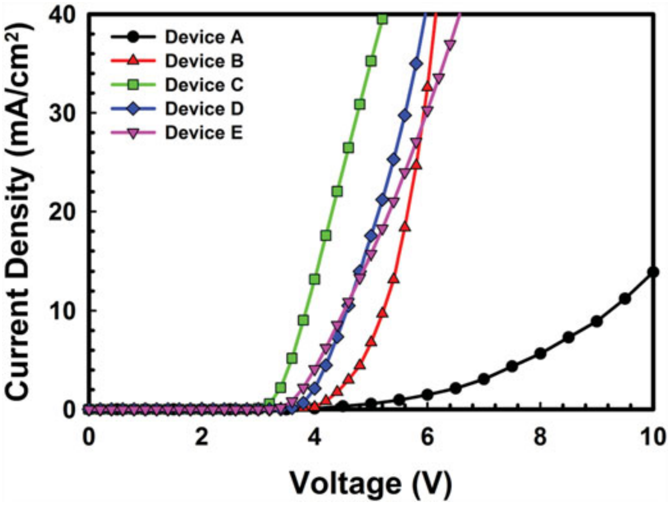

Figure 2. Current density-voltage characteristics of fabricated blue PHOLEDs.

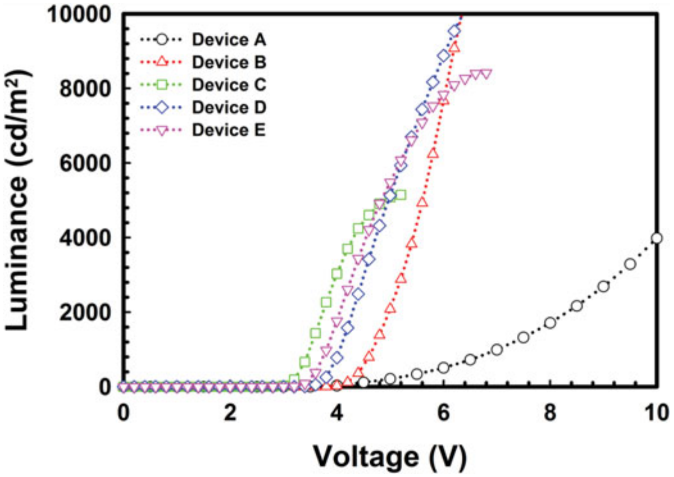

Figure 3. Luminance-voltage characteristics of fabricated blue PHOLEDs.

of a wide band-gap characteristic of 26DCzPPy, onset and driving voltages of Device B are improved significantly as compared with those of Device A, which may be attributed to the bipolar carrier transporting characteristic and reduced electron traps. However, these voltage values are still high to compare with those of red and green PHOLEDs due to still existence of hole trap sites in 26DCzPPy host device. Device B shows the current and power efficiency at driving voltage of 28.0 cd/A and 15.7 lm/W, respectively. Even though current efficiency is lower than that of mCP host device, power efficiency is improved thanks to reduction of driving voltage. The narrow band-gap host material of TCTA (Device C) shows very low onset voltage of 2.8 V which is very close to triplet energy level of FIrpic (2.7 eV), indicating both hole and electron trapping by dopant molecules is minimized. Moreover, Device C shows relatively good efficiency characteristics of of 29.1 cd/A and 26.7 lm/W at the 1000 cd/m^2 brightness value, respectively. Thanks to the significant driving voltage reduction of 3.5 V from Device A to Device C, power efficiency is drastically improved from 14.4 lm/W to 26.7 lm/W. However, significant role-off characteristics are observed in efficiency curves. These strange efficiency characteristics are caused by unstable luminance characteristic of Device C. The maximum luminance of Device C shows only 5144 cd/m^2. It is a relatively low value to compare with the maximum luminance characteristic of 13930 cd/m^2 of Device A. This indicates poor electrical stability of TCTA molecule itself. As shown in Fig. 4, efficiency roll-off phenomenon in mCP and TCTA device are very poor, however, 26DCzPPy device shows low efficiency roll-off characteristics, especially in the power efficiency curve. When bipolar host material of 26DCzPPy and narrow band-gap host material of TCTA are mixed in one emissive layer as the host, Device D and E show both low driving voltage and high efficiency characteristics. Device D shows the onset voltage and driving voltage of 3.2 V and 4.0 V, respectively, and the current and power efficiency of 36.4 cd/A and 28.3 lm/W at 4.0 V driving voltage, respectively. The bipolar characteristics of the mixed host materials and minimized both hole and electron traps help to form wide exciton recombination zone over entire emissive layer. Improved maximum luminance and reduced efficiency roll-off characteristics of Device D in comparison with Device C prove that exciton recombination zone is widen. Especially, under optimized EML thickness condition (Device E), the lowest onset voltage and driving voltage of 3.0 V

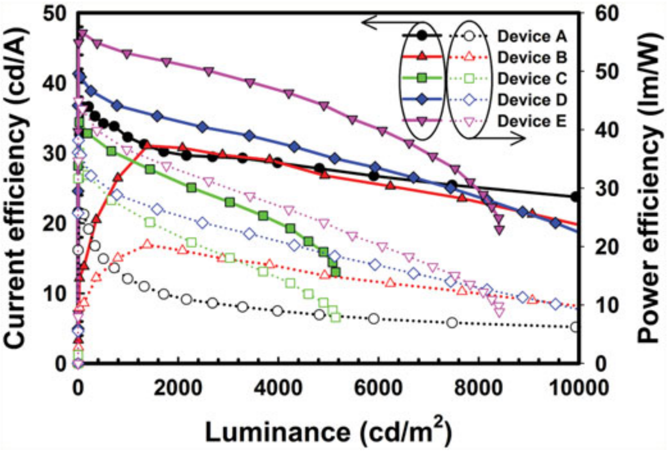

Figure 4. Current efficiency-luminance-power efficiency characteristics of fabricated blue PHOLEDs.

and 3.8 V, respectively, and the highest current and power efficiency of 44.2 cd/A and 36.6 lm/W at 4.0 V driving voltage are observed. The device performances of all devices are summarized in Table 1.

The electroluminescent spectra of Device A~D are shown in Fig. 5. The FIrpic based blue devices generally show a strong peak at about 470 nm with a shoulder peaks at about 495 nm. In our previous studies, it was reported that the intensity of shoulder peak is varied depending on emission zone location in FIrpic based blue PHOLEDs [17]. The emission shoulder around 495 nm in the EL spectrum is enhanced when charge recombination zone shifts to hole transport layer (HTL) side. The exciton recombination zone of Device A may be close to the electron transport layer attributed to both electron tapping situations and hole dominated transporting property of mCP, leading to not only weak shoulder peak but also

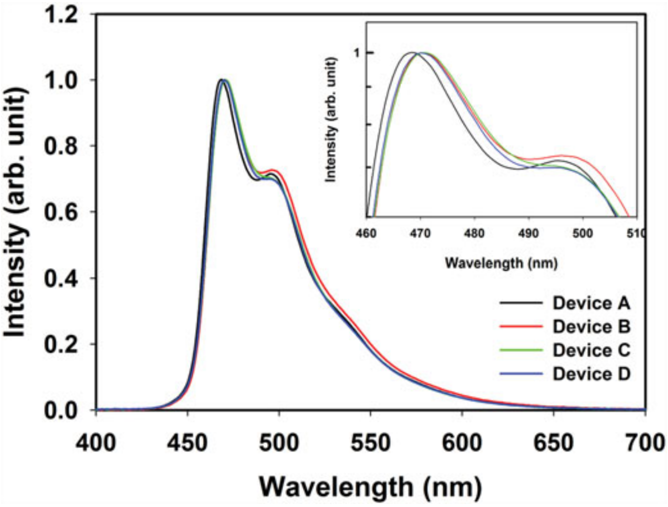

Figure 5. Electroluminescent spectra of Device A~D.

the main emission peak centered at 468 nm. In contrast, Device B shows stronger shoulder peak than that of Device A because Device B has HTL-sided emission zone due to both hole trapping situations and the better electron injection and transportation. On the other hand, when the charge trapping of both holes and electrons are minimized in the Device C and D, gradual shoulder peak is observed maybe due to broad emission zone. The color coordinates of the Devices A, B, C, D, and E show (0.157, 0.313), (0.161, 0.333), (0.155, 0.320), (0.156, 0.318), and (0.160, 0.332) at 1,000cd/m^2 brightness.

Conclusions

In summary, both low driving voltage of 3.8 V and very high efficiencies of 44.2 cd/A and 36.6 lm/W at 1000 cd/m^2 are realized in blue PHOLEDs by using a mixed host system consisting of narrow band-gap and bipolar carrier transporting host materials. Our mixed host system could minimize not only charge trapping but also enhance bipolar transport behavior in the emissive layer. We think that both low driving voltage and high efficiency blue PHOLEDs with high luminance stable characteristics could be a promising way for display and lighting production.

Acknowledgments

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