

Efficient blue and green phosphorescent OLEDs with host material containing electronically isolated carbazolyl fragments



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ABSTRACT

Dry process-able host materials are well suited to realize high performance phosphorescent organic light-emitting diodes (OLED) with precise deposition of organic layers. We demonstrate in this study high efficiency green and blue phosphorescent OLED devices by employing 3-[bis(9-ethylcarbazol-3-yl)methyl]-9-hexylcarbazole based host material. By doping a typical green emitter of *fac* tris(2-phenylpyridine)iridium (Ir(ppy)₃) in the compound the resultant dry-processed green device exhibited superior performance with low turn on voltage of 3.0 V and with peak efficiencies of 11.4%, 39.9 cd/A and 41.8 lm/W. When blue emitter of bis[2-(4,6-difluorophenyl)pyridinato-C2,N](picolinato)iridium (III) was used, the resultant blue device showed turn on voltage of 2.9 V and peak efficiencies of 9.4%, 21.4 cd/A and 21.7 lm/W. The high efficiencies may be attributed to the host possessing high triplet energy level, effective host-to-guest energy transfer and effective carrier injection balance.

1. Introduction

Organic light-emitting diodes (OLEDs) have drawn great attention due to their increasing applications in flat-panel displays and solid-state lightings [1]. A wide variety of OLED based display devices is already presented in the market from the last few years, and lighting products are also developing rapidly [2,3]. Nowadays, phosphorescent materials become ideal for fabricating high-efficiency OLEDs, because they can theoretically approach a nearly 100% internal quantum efficiency by harvesting singlet and triplet excitons simultaneously through intersystem crossing [4–7]. In phosphorescent OLEDs, to reduce the quenching associated with relatively long excited-state lifetimes of triplet emitters and triplet–triplet annihilation, triplet emitters are normally used as guests emitters in a host material [8–10]. Effective host materials are thus of equal importance for efficient phosphorescent OLEDs (PhOLEDs). It is essential for efficient electro-phosphorescence that the triplet level of the host be larger than that of the triplet emitter to prevent reverse energy transfer from the guest back to the host, and to effectively confine triplet excitons on guest molecules. For blue phosphorescence, conjugation in the host molecules must be extremely confined to achieve a triplet energy level larger than photon energies of blue light (~2.7 eV) [11].

Carbazole moiety was the most widely used core structure of high triplet energy host materials due to high triplet energy and versatile chemical modification. Carbazole has a high triplet energy of 3.02 eV, which is high enough for blue host material, and 3,6,9-positions of carbazole can be easily modified with various functional groups. The most representative carbazole high triplet energy host material was 1,3-bis(*N*-carbazolyl)benzene (mCP) which has a high triplet energy of 2.9 eV [12]. It was used as the host material for deep blue emitting dopant and over 10% quantum efficiency value was realized [13]. However, mCP has a problem of poor thermal and morphological stability due to crystallization at high temperature. Dimers and trimers of carbazole, 3-(9-carbazolyl) carbazoles and 3,6-di (9-carbazolyl) carbazoles, were also effective to get high triplet energy as the conjugation of the carbazole core was not extended. The triplet energy of the carbazole dimers and trimers was in the range of 2.9–3.0 eV for efficient energy transfer to blue phosphorescent guest molecules [14]. Recently new efficient host materials having electronically isolated carbazole rings are described in literature [15–17].

In our previous work, we have reported synthesis and characterization of new carbazole based host material: 3-[bis(9-ethylcarbazol-3-yl)methyl]-9-hexylcarbazole. The host was used in red PhOLED device, which exhibited superior performance with peak efficiencies of 6.9%,

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3.2 cd/A and 4.5 lm/W [18]. Here, we report efficient green and blue PhOLEDs using the host material with electronically isolated carbazolyl fragments and high triplet energy level.

2. Experimental

The electro-phosphorescent devices were fabricated on glass substrates containing a bottom indium tin oxide (ITO) anode. Commercial organic materials and the ITO coated glasses with sheet resistance of $\sim 15 \Omega/\text{square}$ were purchased from Lumtec and Shine Materials Technology. All organic materials were subjected to temperature gradient sublimation. The organic and metal layers were deposited onto the ITO-coated substrate by thermal evaporation and the device fabrication was completed in a single cycle without breaking the vacuum. Shadow mask was used to define the active area ($2 \times 2 \text{ mm}^2$) of the device. Current density-voltage-luminance characterization was measured using a Keithley 238 current source-measure unit and a Keithley 6485 pico-ammeter equipped with a calibrated Si photodiode. The electroluminescent spectra were recorded using an Ocean Optics spectrometer.

3. Results and discussion

The synthesis of 3-[bis(9-ethylcarbazol-3-yl)methyl]-9-hexylcarbazole (**BCrMCr** in Fig. 1) was carried out by the synthetic route, which was published earlier [18]. The material demonstrated high thermal stability with glass transition temperature of 73°C and very high triplet energy level of 2.97 eV [18]. The wide triplet energy gap makes the derivative suitable for use in green as well as in blue PhOLEDs.

The blue and green phosphorescent iridium complexes were used as dopants to investigate the application of **BCrMCr** as the host material for the organic PhOLEDs. Herein, bis [2-(4,6-difluorophenyl)pyridinato-C2,N](picolinato)iridium (III), Firpic, and *fac*-tris(2-phenylpyridine)iridium, *fac*-Ir (ppy)₃, are the most commonly used emitters in the host studies because of their nearly 100% photoluminescent quantum yield (PLQY) [19,20]. Furthermore, the absorption spectra of both emitters, as well as the PL spectra of **BCrMCr**, are shown in Fig. 2. As indicated, both emitters exhibited adequate spectral overlap with the host emission. In addition, 1,1-bis [(di-4-tolylamino)phenyl] cyclohexane (TAPC), which possesses wide triplet energy bandgap of 2.87 eV, was selected as the hole transport layer (HTL) because of its high carrier transport capability [21]. Furthermore, two wide triplet energy bandgap materials, 1,3,5-tri [(3-pyridyl)-phen-3-yl]benzene (TmPyPB) and 1,3-bis [3,5-di (pyridin-3-yl)phenyl]benzene (BmPyPB), were respectively used as the electron transport layer (ETL) to determine effective exciton confinement and the carrier balance [22,23]. We tried to adopt a simplified tri-layer architecture of blue or green OLEDs to examine the potential of **BCrMCr** host [24]. We have also

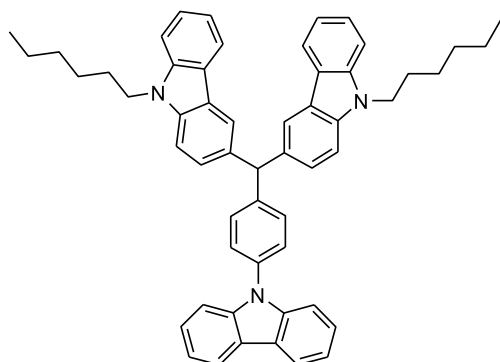


Fig. 1. Chemical structure of 3-[bis(9-ethylcarbazol-3-yl)methyl]-9-hexylcarbazole.

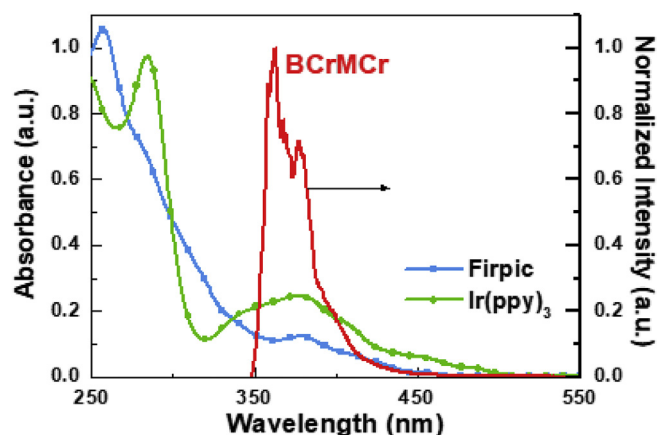


Fig. 2. Absorption spectra of both emitters and the PL spectra of **BCrMCr**.

tested different doping concentration of Firpic and *fac*-Ir (ppy)₃. It was found that optimal doping concentration is wt.8%.

Consequently, the OLED architectures of the blue (B) and green (G) devices consisted of ITO (120 nm)/TAPC (30 nm)/**BCrMCr** doped with 8 wt.% Firpic or *fac*-Ir (ppy)₃ (20 nm)/ETL (50 nm)/LiF (0.8 nm)/Al (150 nm), for which LiF and aluminum were respectively used as the electron injection layer and reflective cathode. Two types of the ETL were employed: TmPyPB (50 nm) for devices B1 and G1 and BmPyPB (50 nm) for the second set of devices B2 and G2. Fig. 3 shows the chemical structures of the material, the schematic architecture, and the energy level diagram of the devices B and G.

The EL characteristics of all the tested devices and the associated numeric data are depicted in Fig. 4 and Table 1. Fig. 4(a) exhibits the EL spectra of the devices, which were recorded at 1000 cd/m^2 . As shown in Fig. 4(a), the electroluminescence (EL) spectra of devices B and G respectively presented pure emissions of Firpic and *fac*-Ir (ppy)₃, supporting the presence of effective energy transfer between **BCrMCr** and the guest [25]. Furthermore, no additional emission peaks were observed, indicating that the carrier recombination zone is located within the EML and that the exciton diffusion to the adjacent layers is avoided even with this simplified tri-layer architecture [26]. In addition, according to the results of time-of-flight measurements, the hole mobility of **BCrMCr** reaches only about $10^{-5} \text{ cm}^2/\text{V}\cdot\text{s}$. In contrast, the electron mobilities of TmPyPB and BmPyPB are respectively estimated to be about 10^{-3} and $10^{-4} \text{ cm}^2/\text{V}\cdot\text{s}$ [19,20]. Thus, one can expect that the exciton formation zone might locate at HTL/EML interface in the devices B and G, leading the nearly identical EL spectra. In other words, the difference in electron transport capability does not significantly influence the location of the exciton formation zone.

Fig. 4(b) shows the current density-voltage-luminance (*J-V-L*) curves. Although the electron mobility of BmPyPB is lower than that of TmPyPB, the use of BmPyPB as ETL in devices B2 and G2 resulted in much higher current densities, which might contribute to the improved carrier injection from the cathode to the organic layer. Therefore, compared to the devices with TmPyPB, devices B2 and G2 with BmPyPB ETL respectively showed higher maximum luminance levels of 21560 cd/m^2 and 72526 cd/m^2 with correspondingly lower operation voltages.

As shown by the efficiency curves, devices B1 and B2 exhibited similar peak EL efficiencies of 9.4% (21.5 cd/A and 21.4 lm/W) and 9.4% (21.4 cd/A and 21.7 lm/W) in the forward direction. Furthermore, at a practical luminance of 10^2 cd/m^2 , the efficiency of device B1 remained high at around 9.4% (21.3 cd/A and 14.9 lm/W) while the forward efficiency of device B2 was also maintained at 9.1% (20.5 cd/A and 17.7 lm/W). Nevertheless, device B2 demonstrated superior performance than B1 at much higher luminance levels ranging from 10^3 to 10^4 cd/m^2 , and similar behavior could be observed in the G devices.

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