



Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/gmcl20>

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Version of record first published: 05 Oct 2009

To cite this article: Seok Jae Lee, Ji Hyun Seo, Gu Young Kim & Young Kwan Kim (2009): A Study on the Phosphorescent Blue Organic Light-Emitting Diodes Using Various Host Materials, *Molecular Crystals and Liquid Crystals*, 507:1, 345-352

To link to this article: <http://dx.doi.org/10.1080/15421400903054287>

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A Study on the Phosphorescent Blue Organic Light-Emitting Diodes Using Various Host Materials

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We investigated the electrical characteristics of three devices which were fabricated with phosphorescent blue emitter, iridium(III) bis(4,6-difluorophenylpyridinato- N,C^2)picolinate (FIrpic), doped in host materials, 4,4,N,N'-dicarbazolebiphenyl (CBP), N,N'-dicarbazolyl-3,5-benzene (mCP) of the carbazole-type and p-bis(triphenylsilyl)benzene (UGH2) of the wide-energy-gap, respectively. We demonstrated the efficient blue electrophosphorescence using exothermic energy transfer between mCP and FIrpic with triplet energy level of 2.90 eV and 2.65 eV, respectively. Device property for a mCP-based structure was found to be superior to that employing the endothermic host-guest combination consisting of the CBP and FIrpic with triplet energy level of 2.57 eV and 2.65 eV, respectively. In case the device with UGH2, despite of the very wide band gap (4.40 eV), it is inefficient given the large energy barrier (1.70 eV) for hole injection into the highest occupied molecular orbital (HOMO) of UGH2 as the drive voltage increases. The device with mCP host material shows a maximum luminous efficiency and power efficiency of 17.00 cd/A and 10.9 lm/W, respectively.

Keywords: blue phosphorescent organic light-emitting diodes; energy transfer; host materials; triplet state

1. INTRODUCTION

Phosphorescent emitters are appropriate for obtaining highly efficient organic light emitting diodes (OLEDs) because they enable an effective use of both triplet and singlet excitons, thus providing a 100% internal quantum efficiency for light emission [1–3]. A Princeton University group has demonstrated a way to break through the efficiency limitation by using certain organic materials which emit intense

This work was supported by Seoul R&D Program (10555).

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phosphorescence from triplet state at room temperature [2,4]. The phosphorescence materials, such as iridium (Ir), platinum (Pt) and europium (Eu) complexes with organic ligands, were doped as an emitter into a host material of emissive layer [5]. The phosphorescence from the device can be understood in terms of the energy transfer from both the singlet and triplet states of the host molecules to the triplet states of phosphorescence emitter molecules, and by direct excitation of the phosphorescence emitter molecules followed by charge trapping [6,7].

In designing phosphorescent blue organic light-emitting diodes, the use of a host material with both wide-band gap and higher triplet-excited-state (T^1) energy than that of a phosphorescent emitter is essential to ensure energy transfer from the host material to the phosphorescent emitter [8]. In other words, as the energy gap of the emitter becomes large, successively larger host energy gap is required to maintain efficient exothermic energy transfer from host to emitter. For the green or red phosphorescent organic light emitting diodes (PHOLEDs), a host with suitable triplet energy level is easy to obtain. However, for blue PHOLEDs, as the blue phosphorescent emitter in common use is iridium(III) bis(4,6-difluorophenylpyridinato- $N, C^{2'}$)picolinate (FIrpic) with the triplet energy level of 2.65 eV, it is very difficult to find a suitable high-energy hosts [8–10].

In this paper, we analyze and make a comparative study the electrical characteristics of three devices which were fabricated with phosphorescent blue dopant, iridium(III) bis(4,6-difluorophenylpyridinato- $N, C^{2'}$)picolinate (FIrpic), doped in host materials, 4,4, N, N' -dicarbazolebiphenyl (CBP), N, N' -dicarbazolyl-3,5-benzene (mCP) of the carbazole-type and p -bis(triphenylsilyl)benzene (UGH2) of the wide-energy-gap, respectively.

2. EXPERIMENTAL DETAILS

The sheet resistivity and thickness of the indium tin oxide (ITO) thin films coated on glass substrates used in this study were $30 \Omega/\square$ and 1000 \AA , respectively. The ITO cleaning procedure included sonications in acetone, methanol, and ionized water at 60°C for 15 min. The chemically cleaned ITO substrates were kept for 24 h in isopropyl alcohol. After the cleaned ITO substrates had been dried by using N_2 gas with a purity of 99.9999%. Thereafter, pre-cleaned ITO was treated by O_2 plasma with the conditions of 2×10^{-2} Torr, 125 W for 2 min. OLEDs were fabricated using the high vacuum (5×10^{-7} Torr) thermal evaporation of organic materials onto the surface of the ITO-coated glass substrate. As shown in Figure 1, three devices structure

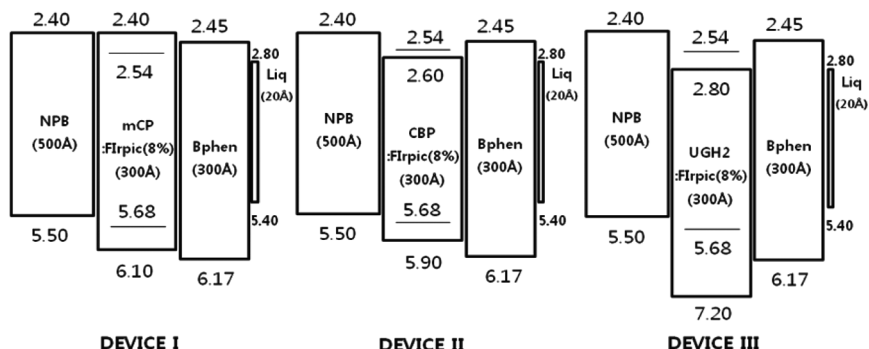


FIGURE 1 Energy level diagrams of blue phosphorescent devices with mCP (DEVICE I), CBP (DEVICE II) and UGH2 (DEVICE III).

(DEVICE I, II, III) used in this experiment were ITO (100 nm)/N,N'-bis-(1-naphyl)-N,N'-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB, 50 nm) as hole transporting layer/Bis(3,5-Difluoro-2-(2-pyridyl)phenyl-(2-carboxypyridyl) iridium III (FIrpic) of the triplet energy level 2.65 eV doped in HOST materials (30 nm, 8% doping) as blue emitting layer/4,7-diphenyl-1,10-phenanthroline (Bphen, 30 nm) as hole blocking and electron transporting layer/lithium quinolate (Liq, 2 nm) as electron injection layer/aluminum (Al, 100 nm). The deposition rates were 1.0–1.1 Å/sec for organic materials and 0.1 Å/sec for lithium quinolate (Liq), respectively. After the deposition of the organic layers and without a vacuum break, the aluminum (Al) cathode was deposited at a rate of 10 Å/sec. The deposition rates were controlled by using a quartz crystal monitor and the doping concentrations of the dopants were optimized. After organic and metal depositions, devices were encapsulated in a glove box with O₂ and H₂O concentrations below 1 ppm. A desiccant material consisting of barium-oxide powder was used to absorb the residue moisture and oxygen in the encapsulated device. The emission area of devices was 3 mm × 3 mm. The voltage – current density – luminous efficiency – power efficiency characteristics and electroluminescence (EL) spectra were measured and recorded at once with chroma meter CS-1000A (Minolta) and current and voltage were controlled with measurement unit (model 2400, Keithely).

3. RESULTS AND DISCUSSION

Figure 1 shows the energy level diagrams of blue phosphorescent devices with mCP (DEVICE I), CBP (DEVICE II) and UGH2 (DEVICE

III) as the host materials for the present study. Three devices (DEVICE I~III) were fabricated to investigate the effect of different energy band gap and triplet energy level of the host materials on device performances. The lowest unoccupied molecular orbital (LUMO) of mCP, CBP, UGH2 as host materials and FIrpic as emitter were 2.40 eV, 2.60 eV, 2.80 eV and 2.54 eV, and the highest occupied molecular orbital (HOMO) of mCP, CBP, UGH2 and FIrpic were 6.10 eV, 5.90 eV, 7.20 eV and 5.68 eV, respectively. While the LUMO level of FIrpic falls outside of that of CBP and UGH2, the both HOMO and LUMO level of FIrpic fall within of that of mCP. In the UGH2 host materials case, because of ultrahigh energy gap (4.40 eV) between HOMO and LUMO level, exciton formation occurs by direct charge injection from the adjacent hole and electron transporting layers onto the emitter doped into the host matrix.

Figure 2 shows the chemical structures and energy level diagram of the lowest triplet state (T^1) of host materials; mCP, CBP as carbazole-based material, UGH2 as ultrahigh energy gap organosilicon compounds used, and FIrpic as phosphorescent emitter. Carbazole is known to be a potential host material for blue electrophosphorescence because of its wide-band gap energy, high triplet energy, and good charge-transporting ability [11–13]. The ultrahigh singlet and triplet energies associated with kinds of UGH materials effectively

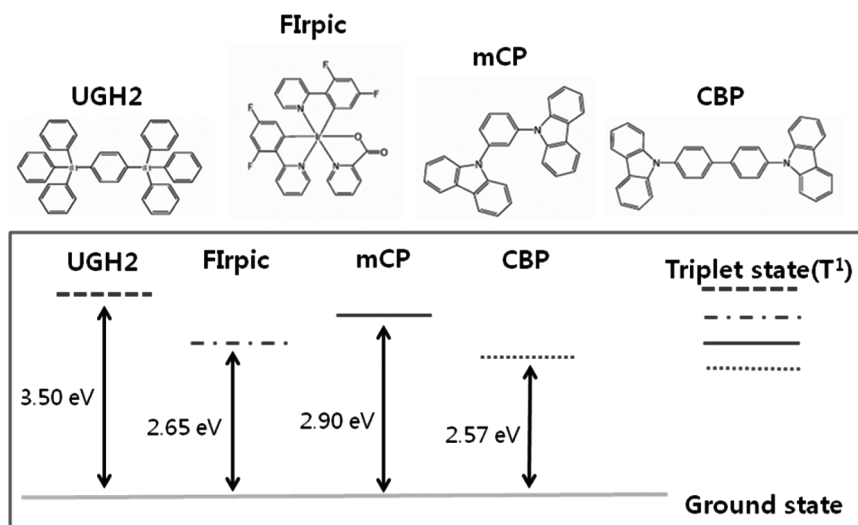


FIGURE 2 Chemical structures and energy level diagram of the lowest triplet state (T^1) of hosts (UGH2, mCP, CBP) and emitter (FIrpic), respectively.

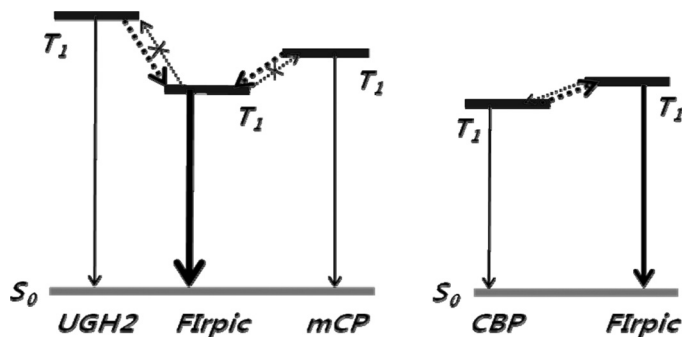


FIGURE 3 The endothermic and exothermic transitions between the hosts and FIrpic are shown by broken arrows, while phosphorescence from the T^1 state to the ground state is shown by solid arrows.

suppress both the electron and energy transfer quenching pathways between the emissive emitter and the host material [14]. The triplet energy band gap of mCP, CBP, UGH2, and FIrpic were 2.90 eV, 2.57 eV, 3.50 eV, and 2.65 eV, respectively [14,15].

Figure 3 shows the endothermic and exothermic transitions between the host materials (mCP, CBP, UGH2) and the emitter (FIrpic). The both transitions are shown by broken arrows, while phosphorescence from the T^1 state to the ground state is shown by solid arrows. Since the triplet energy level of FIrpic is 2.65 eV, we infer that the energy transfer from mCP and UGH2 to FIrpic is exothermic. However, since the triplet energy level of CBP is slightly less than that of FIrpic, endothermic energy transfer from CBP to FIrpic is inferred. This efficient confinement of triplet energy must be the reason for the efficient phosphorescence from the FIrpic triplet state. Thus, high triplet energy of the host material is very important with regard to the emission efficiency of blue phosphorescent OLEDs.

The luminous efficiency of DEVICES I, II and III was compared in Figure 4(a) and the inset of Figure 4(a) shows the current density-voltage property of the three devices with different host materials. The maximum luminous efficiencies of DEVICES I, II and III are 17.00, 11.00 and 2.02 cd/A, respectively. DEVICE I with mCP shows the best luminous efficiency value of 17.00 cd/A at 0.78 mA/cm^2 . The luminous efficiency of DEVICE I with mCP was quite higher than DEVICE II with CBP, while the current density of DEVICE I and DEVICE II were almost same, as shown in inset of Figure 4(a). In case the device with UGH2, despite of the very wide band gap (4.40 eV), it is inefficient given the large energy barrier

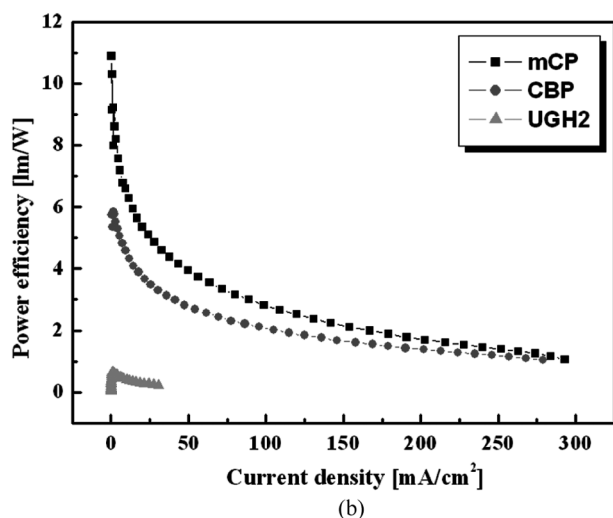
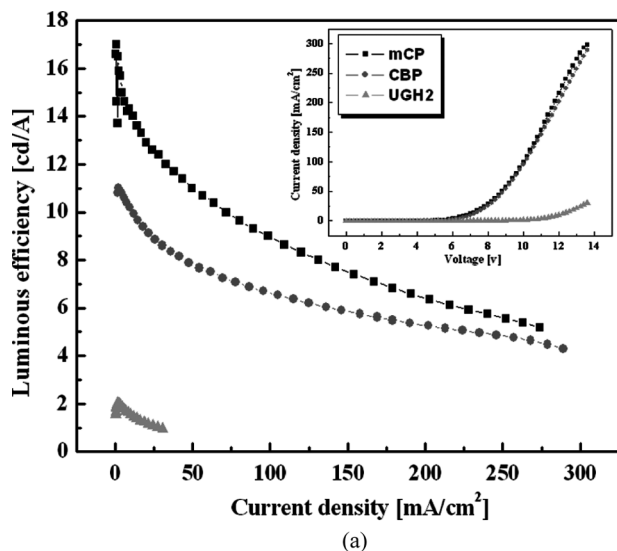


FIGURE 4 (a) Luminous efficiency-current density (inset : current density vs voltage) curves and (b) Power efficiency-current density curves of blue phosphorescent devices with host material of various energy band gap.

(1.70 eV) for hole injection into the highest occupied molecular orbital (HOMO) of UGH2 as the drive voltage increases. It is proved in inset of Figure 4(a) that the hole injection into UGH2 is difficult, where the current density of device with UGH2 is 10.00 mA/cm² at applied

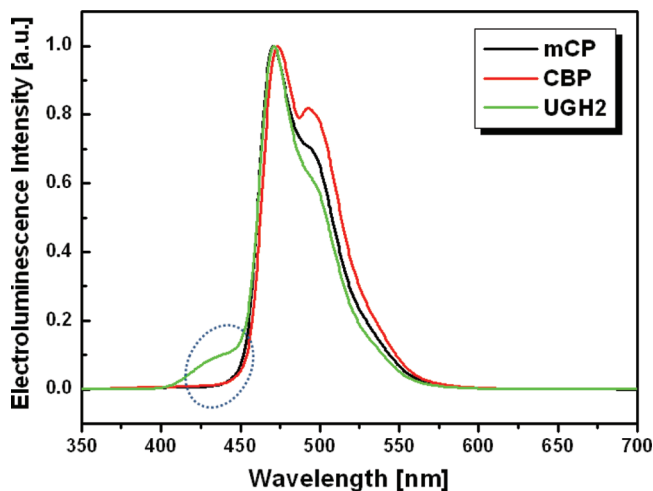


FIGURE 5 Electroluminescence (EL) spectra of blue phosphorescent devices with host material of various energy band gap.

voltage of 12 V. Figure 4(b) shows the power efficiency-current density property of the three devices. The maximum power efficiencies of DEVICES I, II and III are 10.90, 5.85 and 0.63 lm/W, respectively.

A typical electroluminescence spectra of three devices at 10 V bias is shown in Figure 5. Blue emission with peak position of 471 nm and subpeak around 495 nm were observed in all devices which originated from FIrpic triplet emission. In addition to blue emission, DEVICE III also showed another broad peak at 443 nm which is assigned to NPB emission, while DEVICE I and DEVICE II exhibited no emission in deep blue wavelength range. The NPB emission in DEVICE III may be due to hole accumulation at the interface between NPB and UGH2 and electron overflow from UGH2 to NPB. In general with UGH2 host material, holes are strongly trapped by emitter materials in FIrpic doped devices and holes can be accumulated between NPB and UGH2 because hole transport through FIrpic is limited. The accumulated holes can recombine with some electrons, giving rise to NPB emission.

4. CONCLUSIONS

In summary, we have demonstrated the efficient blue electrophosphorescence using exothermic energy transfer between mCP and FIrpic with triplet energy level of 2.90 eV and 2.65 eV, respectively.

Device property for a mCP-based structure was found to be superior to that employing the endothermic host-guest combination consisting of the CBP, UGH2 and Flrpic with triplet energy level of 2.57 eV, 3.50 eV and 2.65 eV, respectively. The device with mCP host material exhibits a maximum luminous efficiency and power efficiency of 17.00 cd/A and 10.9 lm/W, respectively.

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