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Highly efficient 7,8,10-triphenylfluoranthene-doped blue organic light-emitting diodes for display application

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We have demonstrated an organic light-emitting diode based on blue-fluorescent dopant 7,8,10-triphenylfluoranthene in a host of dipyrenylfluorene derivatives. The device shows pure blue emission with a peak wavelength of 456 nm and Commission International de L'Eclairage coordinate at (0.164, 0.188). An electroluminescence efficiency as high as 3.33 cd/A and external quantum efficiency of 2.48% can be achieved. Comparison of the photoluminescence and electroluminescence spectra reveals a nearly identical exciton relaxation and efficient energy transfer from the host to the dopant. © 2006 American Institute of Physics. [DOI: 10.1063/1.2167814]

Since the organic light-emitting diodes (OLEDs) were developed by Tang and VanSlyke,¹ a great deal of effort was made to produce full-color flat panel displays^{2,3} and highly efficient red, green, and blue emitters are essential to achieve this goal. On the one hand, highly efficient red⁴ and green⁵ electroluminescence devices have already been described; however, highly efficient pure blue light remains a challenge. Consequently, many different blue-fluorescent molecules, derivatives,⁶ anthracene oligoquinoline, such as tetraphenylsilane,⁸ and distyrylarylene derivatives⁹ have been proposed to solve this problem. Among these materials, external quantum efficiencies on average of about 2.4% can be achieved. Higher efficiencies up to 5% were reported by spirofluorene derivatives.^{10,11} Although current blue-emitting devices have not been completely optimized, there remains a strong demand to develop new molecules for highly efficient blue OLEDs.

In typical blue OLED configurations, the blue-emitting layer is often fabricated by doping a blue fluorophore into a wide band gap host. For example, distyrylarylene⁹ and anthracene derivatives¹² are often reported host materials that improve the carrier injection and transport over singlecomponent devices. However, blue dopants with low highest occupied molecular orbital (HOMO) energy levels still suffer from poor charge injection, if a suitable hole transporting layer (HTL) is not used. Several materials, such as poly(Nvinylcarbazole) (PVK) and 4,4'-dicarbazolyl-1,1'-biphenyl (CBP), can be used as HTL to match low HOMO levels.^{7,13} Here, we report the incorporation of two materials, 7.8.10-triphenylfluoranthene¹⁴ (TPF) and derivatives of dipyrenylfluorene¹⁵ (DPF) as an emission layer. By doping TPF into DPF, an efficient energy transfer from host to dopant affords highly efficient blue OLEDs. Details of different doping concentrations and the resulting device performance are also presented.

The devices had the following structure; ITO/PEDOT (30 nm)/NPB (15 nm)/CBP (15 nm)/blue-emitting layer (20 nm)/BCP (20 nm)/Ca (1 nm)/Al (80 nm). The hole transporting layer is а combination of 4, 4'-bis-(1-naphthyl-N-phenylamino)-biphenyl (NPB) and CBP. The electron transporting layer uses a 30 nm bathocuproine (BCP). The blue-emitting material uses DPF as blue host and TPF as fluorescent blue dopant. The molecular structure of DPF and TPF are shown in Fig. 1. The thin films are fabricated though thermal vacuum deposition in a vacuum smaller than 5×10^{-6} torr. This blue-emitting layer (BEL) is produced by coevaporating DPF and TPF in molar ratios of 1% and 6% TPF. Additionally, single layers of TPF and DPF for blue devices are also provided for comparison. The thickness of the films was monitored by quartz crystal monitors, which were calibrated with a Dektak profilometer. The device affords a light-emitting area of $\sim 11 \text{ mm}^2$. Upon device fabrication completion, it was transferred to a nitrogen-filled glove box for electrical and optical measurements. The current-voltage (I-V) curves were characterized with a Keithley 2400 source measurement unit. The luminescence current was collected by a calibrated silicon photodiode and electroluminescence (EL) spectra as well as brightness were later measured by a Photo Research PR650 spectrophotometer. The Commission International de L'Eclairage coordinates (CIE) were used to describe the color of OLEDs. The photoluminescence (PL) spectra were measured by a Jobin Yvon Spex Fluorolog-3 double grating spectrometer.

Figure 1 also shows the device structure and schematic energy level diagram. Since the blue-emitting materials (TPF) have a lower HOMO band, 6.3 eV, relative to the NPB (5.4 eV), there will be a 0.9 eV barrier, resulting in difficult hole injection from the NPB to (BEL). A second hole trans-

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FIG. 1. Molecular structures of TPF and energy diagram of blue lightemitting device of ITO/PEDOT/NPB/CBP/BEL/BCP/CaAl. The LUMO and HOMO energies of each organic molecule and directions of charge flow are indicated.

porting layer, CBP, which has a HOMO of 5.7 eV and is often employed as a phosphorescent dye host¹⁶ was inserted between NPB and BEL.¹³ In this case, it can both enhance the luminescence efficiency and lower the operation voltage. The electron-transporting layer of BCP with the same thickness as the combined HTLs was used to produce a symmetrical structure and balanced charges.

The surface roughness, which was checked by atomic force microscope, was about 1.6 nm after multilayer organic evaporations. The smooth surface insured a reliable electrode contact for measurements. The current-voltage-luminescence characteristics of the device with a 6% TPF doped DPF OLED is shown in Fig. 2. The current turns on at 2.4 V and the light later turns on at 5 V. The delay of light emission turn on suggests that the hole and electron carriers are not perfectly balanced. Since there is still 0.6 eV energy mismatch of the HOMO levels between CBP and TPF, the hole injection is more difficult than the electron injection. The device shows low leakage current under a reverse bias condition. A maximum luminescence of 6210 cd/m² at 269 mA/cm^2 was measured. The device achieves maximum luminous efficiency as high as 3.33 cd/A with an external quantum efficiency of 2.48% at 11.66 mA/cm² and pure blue light with CIE coordinates of 0.164 and 0.188 (x, y, y)respectively) are observed. The luminous efficiency is comparable to that of other reported OLEDs with a fluorescent blue dopant, such as perylene¹² and anthracene derivative,⁶ but not as high as monostyrylamine derivative doped diphenyanthracene.17

Since the energy transfer can be achieved between DPF and TPF, different doping concentrations were studied; their PL (excited at 300 nm) and EL spectra are shown in Fig. 3. In the PL spectra [Fig. 3(a)], the band at 456 nm corresponds



FIG. 2. Current-voltage-luminescence characteristics of blue light-emitting device with 6% TPF doped in DPF. Forward (0 to 13 V) and reverse (0 to -5 V) bias scans are applied, whereas the current and electroluminescence are measured at the same time.

to pure DPF, while pure TPF emits at 456 nm but with a shoulder at 481 nm. Overall, pure TPF shows a broader emission than pure DPF. An increase in TPF doping concentration results in higher PL intensity and broadens the emis-



FIG. 3. (a) PL spectra and (b) EL spectra of different TPF doping concentrations. The concentrations range from 0%, 1% to 6% TPF in DPF host and pure TPF. The PL spectra are measured with excitation at 300 nm. The EL spectra are measured while biasing at constant current of 3 mA.

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FIG. 4. (a) The device electroluminescence efficiency and (b) device luminescence-current characteristics for different doping concentrations ranging from 0%, 1% to 6% TPF in DPF and pure TPF.

sion spectrum due to its 481 nm shoulder. The EL spectra [Fig. 3(b)] show a very similar effect as the same doping conditions used in PL study. Pure DPF shows an EL spectrum with a peak at 452 nm. With a doping concentration of 1% TPF, the EL spectrum is also centered at 452 nm but again with some broadening of the emission spectrum. A 480 nm shoulder also grows in with the enhanced TPF emission at 6% doping. The pure TPF EL spectrum shows two peaks at 456 and 480 nm. The corresponding peak broadening is also observed in the CIE y coordinate as it moves from 0.157 to 0.172 to 0.188 and finally 0.24 from pure DPF, 1% TPF, 6% TPF, and pure TPF, respectively. Comparing the PL and EL spectra in this doping scheme, the emission peak position and peak broadening effect are nearly identical. This indicates the exciton relaxation occurred in TPF, after an efficient energy transfer from the host to the dopant due to similar lowest unoccupied molecular orbital (LUMO), HOMO energy levels in TPF and DPF. An optimum device efficiency is obtained with 6% TPF doped in DPF.

Luminous efficiency and brightness based on the aforementioned doping levels are presented in Fig. 4. The pure TPF device is more efficient than the pure DPF device,

TABLE I. The efficiencies of various devices used in this letter.

Device	Luminous efficiency (cd/A)	External quantum efficiency (%)	CIE
TPF	2.89	1.75	(0.177, 0.24)
DPF+6%TPF	3.33	2.48	(0.164, 0.188)
DPF+1%TPF	1.29	1.04	(0.168, 0.172)
DPF	0.84	0.72	(0.168, 0.157)

which is consistent with the comparison of PL intensity. The doping of 6% TPF in DPF compared to that of pure DPF increases the efficiency from 0.84 to 3.33 cd/A with a corresponding luminescence enhancement from 794 to 2987 cd/m² at 100 mA/cm². The devices performance in terms of luminous efficiency, external quantum efficiency, and CIE at different doping levels are summarized in Table I.

In conclusion, a high efficient blue-electroluminescence device based on doping of TPF into DPF is demonstrated. A maximum luminous efficiency of 3.33 cd/A and external quantum efficiency of 2.48% can be achieved at 11.66 mA/cm² at an optimal 6% TPF doping. Blue emission with CIE coordinates of (0.164, 0.188) is observed originating from the energy transfer and decay mechanism from DPF to TPF. This is the demonstration of triphenylfluoranthene doped dipyrenylfluorene leading to very efficient blue light-emitting thin film devices for applications in OLED displays.

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