Effect of Charge-transporting Molecules on Electrophosphorescence in a Device Fabricated Using Third-generation Dendrimer Encapsulated Tris[2-benzo[*b*]thiophen-2-ylpyridyl]iridium Complex

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We have synthesized a new deep-red light-emitting phosphorescent third-generation dendrimer with hole-transporting carbazole dendrons. The new dendrimer is highly efficient when used as a solution-processable emitting material and when used in an electrophosphorescent light-emitting diode system. We selected 4,4′-bis(9-carbazolyl)-1,1′-biphenyl (CBP) and 2-(1,1′-biphenyl-4-yl)-5-(4-*tert*-butylphenyl)-1,3,4-oxadiazole (PBD) as guest molecules in order to manipulate the carrier transport and carrier confinement in the emissive layer. We fabricated a dendrimer light-emitting diode (DLED) whose configuration is represented as ITO/PEDOT:PSS (40 nm)/dendrimer containing guest molecules (55 nm)/BCP (2,9-dimethyl-4,7-diphenylphenanthroline) (10 nm)/Alq_3 (40 nm)/LiF (1 nm)/Al (100 nm) and investigated the device performance. The luminance of the multilayered devices fabricated using the PBD-containing dendrimer was 381 cd/m^2 at 52.42 mA/cm^2 (20 V) for the dendrimer bearing PBD, which was slightly better than that of a device fabricated using dendrimer alone.

It is well known that phosphorescent materials are particularly attractive because of the unique light emission with a theoretical internal quantum efficiency of 100% from both singlet and triplet excitons.[1–3] In particular, homoleptic or heteroleptic cyclometalated Ir^{III} complexes are the most promising phosphorescent dyes for use in organic light-emitting diodes (OLEDs). This is because they have relatively high phosphorescent efficiencies and it is possible to modify their ligand structures in order to tune the Commission International de L'Eclairage (CIE) coordinate of the emitted light.[4–7]

In this paper, we report the electrophosphorescent properties of devices fabricated from third-generation dendrimer with or without guest molecules, which help in controlling carrier transport and carrier confinement in an emissive layer. The peripheral carbazolyl dendrons not only prevent self-aggregation but also donate energy to the core of the dendrimer. It has been reported that the core of the third-generation dendrimer is well protected by the surrounding carbazolyl dendrons and that intermolecular interaction and self-aggregation are effectively suppressed.[8] We used two guest molecules, namely, CBP and PBD, in an attempt to improve device efficiency. It must be mentioned here that we used small molecules such as CBP or PBD as guest molecules and not the host molecules used in conventional electrophosphorescent devices. The high triplet energy of the guest molecule facilitates efficient emission of red phosphorescence from the dendrimer. The triplet energies of CBP ($T_1 = 2.67$ eV)[9] and PBD ($T_1 = 2.48$ eV)[10] are also sufficiently high for efficient exciton confinement inside the Ir^{III} complex via energy transfer.

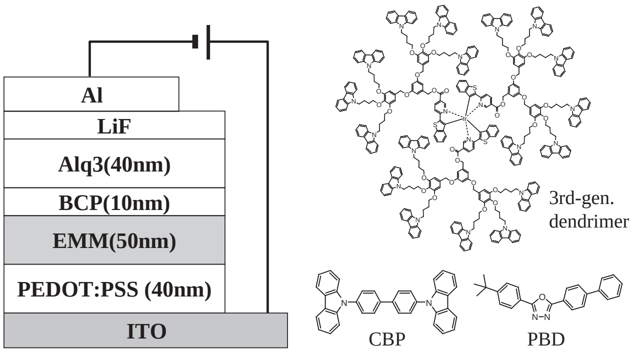

Figure 1. Structures of third-generation dendrimer with homoleptic Ir^{III} complexes, CBP, and PBD. The device configuration is also illustrated.

We fabricated the electroluminescence (EL) devices using third-generation dendrimer and charge-transporting molecules (see Figure 1).

The multilayer diodes that we fabricated can be represented as ITO/PEDOT:PSS (40 nm)/dendrimer:CBP or dendrimer:PBD/BCP (10 nm)/Alq_3 (40 nm)/LiF (1 nm)/Al (100 nm). We fabricated devices by using our conventional method.[8] We fabricated three different multilayered devices A, B, and C using the dendrimer alone, dendrimer with CBP (30 wt %), and dendrimer with PBD (30 wt %), respectively. The current density–voltage–luminance curves for the three devices are shown in Figure 2. The turn-on voltages of these devices are in the range 6.5–11.0 V and are similar to the typical voltages of Ir^{III} complex-based DLEDs. The third-generation dendrimer has a slightly high turn-on voltage, which is attributed to its high dielectric nature with a large extent of the nonconjugated spacer. Device B has a slightly high turn-on voltage. This may be due to the fact that the HOMO energy levels between CBP (6.3 eV) and PEDOT:PSS are higher than the energy level between PBD (6.0 eV) and PEDOT:PSS, because of which less facile hole injection into the emissive layer is expected. The maximum luminance of devices B and C are 541 cd m^{-2} (at 169.02 mA cm^{-2}) and 381 cd m^{-2} (at 52.42 mA cm^{-2}), respectively; the luminance of device C is comparable to or higher than that of device A. The plots of luminous efficiency and external quantum efficiency (EQE) of each device versus the current density are shown in Figure 3. The maximum external quantum efficiencies of device B and C are determined to be 4.21% (at 0.31 mA cm^{-2}, 0.40 lm W^{-1}, 1.58 cd A^{-1}) and 6.22% (at 0.33 mA cm^{-2}, 0.77 lm W^{-1}, 2.09 cd A^{-1}), respectively.

The efficiency of device C is found to be slightly higher efficiency than that of device A. It should also be noted that

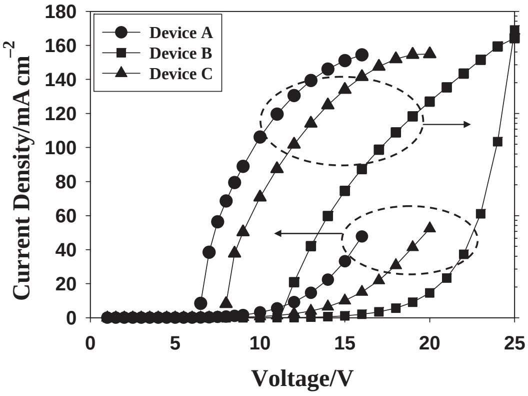

Figure 2. Dependence of current density and luminance on the applied voltage.

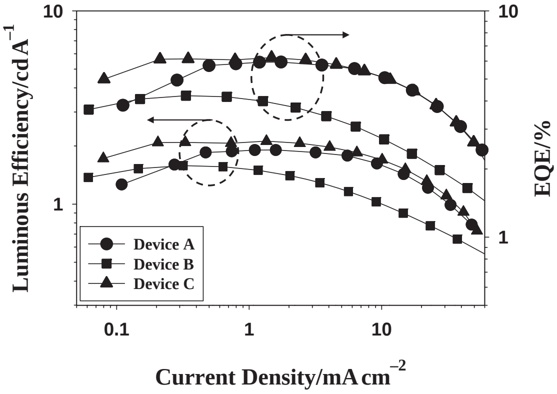

Figure 3. Dependence of luminous efficiency and external quantum efficiency on the current density.

Table 1. Measured and calculated parameters of the three devices fabricated using third-generation dendrimer

Device	Turn-on /V	Maximum luminance/cd m^{-2} (corresponding ***J***)	Maximum power efficiency/lm W^{-1} (corresponding ***J***)	Maximum luminous efficiency/cd A^{-1} (corresponding ***J***)	Maximum external quantum efficiency/η_{ext} (corresponding ***J***)	CIE Index (x, y)
Device A	6.5	373 (47.74) at 16 V	0.77 (0.22) at 7.5 V	1.95 (1.58) at 9 V	5.94 (1.58) at 9 V	$x = 0.70$ $y = 0.30$
Device B	11	541 (169.02) at 25 V	0.40 (0.31) at 13 V	1.58 (0.31) at 13 V	4.21 (0.31) at 13 V	$x = 0.64$ $y = 0.33$
Device C	8	381 (52.42) at 20 V	0.77 (0.20) at 8.5 V	2.09 (0.33) at 9 V	6.22 (0.33) at 9 V	$x = 0.70$ $y = 0.30$

the efficiency of device B, which is fabricated from the CBP-containing dendrimer, is the lowest among all the devices. CBP is an ambipolar charge-transporting molecule,[11,12] and it is expected that there will be greater balance between the different types of charge carriers in CBP than in PBD, which transports only electrons. However, in device B, the injected holes travel through the emissive layer so fast that exciton recombination occurs at or in the vicinity of the interface between the emissive layer and BCP layer. Therefore, the efficiency of device B is less than that of devices A and C. In addition, the BCP emission was slightly mixed with the emission of pure Ir$^{\text{III}}$ complex ($x = 0.70$, $y = 0.33$), displaying different CIE coordinates ($x = 0.64$, $y = 0.33$) (see Table 1).

The luminous efficiency and external quantum efficiency of all three devices decreased at high current densities, probably because of triplet–triplet annihilation. The luminous and external quantum efficiencies of device B, which has a relatively low value, decayed rapidly (Figure 3). It can be speculated that the balance between hole and electron transport was disturbed because hole mobility was enhanced to a greater extent than electron mobility. CBP is commonly used as a host material for phosphorescent dyes because it enables efficient transfer of triplet energy. However, it is easily crystallized in certain light-emitting matrices. Further, the deep LUMO energy level ($E_{\text{LUMO}} = 3.0\,\text{eV}$) in CBP results in poor electron transport, because of which charge-transport balance is disturbed.

To summarize, the conventional host molecule CBP and an electron-transporting molecule PBD were employed as guest molecules for fabricating electrophosphorescent devices. PBD was found to be more suitable for fabricating these devices than CBP, which is an ambipolar guest molecule in the dendrimer. Device C, which was fabricated using PBD, retained its CIE coordinates over a wide range of current densities and its efficiency was very encouraging.

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