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Performance enhancement of white-electrophosphorescent devices incorporating a mixed-transition layer

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The performance of white organic light-emitting devices has been improved significantly by incorporating a mixed-transition layer, consisting of the hole-transporting (or electron-transporting) layer doped with the emissive dye. These devices exhibit maximum current and power efficiencies of 13.3 cd/A and 11.3 lm/W, respectively (1.6 and 2 times higher than the corresponding values of devices without the mixed-transition layer), with slightly modified Commission Internationale De L'Eclairage coordinates, from (0.300, 0.371) to (0.312, 0.366). This is attributed to the enhancement of carrier transport by doping, thereby improving both the exciton formation probability and the charge transport balance. © 2008 American Institute of Physics. [DOI: 10.1063/1.2896606]

White organic light-emitting devices (WOLEDs) are currently attracting considerable attention for applications in flat panel displays and as solid state lighting sources. Various approaches have been used to optimize the efficiency of these devices, including the use of multiple-emissive-layer structures,^{1–3} the insertion of a hole-injecting layer,^{1,2} and the addition of an exciton-blocking layer.³ The balance of charge transport is a key feature in the operation of WOLEDs.⁴ Holes are normally the main charge carriers (as a result of the high hole mobility in organic materials).⁵ One popular method of improving the charge balance is to incorporate electron-transporting compounds. A number of such materials has now been synthesized, including 4,7-diphenyl-1,10-phenanthroline (BPhen).⁶ This compound has an electron mobility of 5×10^{-4} cm²/V s, similar to the hole mobility in *N,N'*-bis(naphthalene-1-yl)-*N,N'*-bis(phenyl)-benzidine (NPB) [$(3-6) \times 10^{-4}$ cm²/V s].⁷

A further approach is to exploit chemical doping.^{8,9} Although the electrical properties of the host and dye are very different, the carrier transport may be controlled by selecting appropriate doping concentrations. For instance, a dye can be chosen with energy levels within those of the host. Generally, if the dye concentration is low (less than 1 wt %), the dye molecules mainly act as traps, hindering the carrier transport.⁹ By increasing the dye concentration, the distance between the dye molecules is decreased until the carriers are effectively shared between the dye and the host.⁹ At high dye concentrations (more than 10 wt %), the carrier transport becomes dominated by hopping between sites associated with the dye molecules.⁹ However, at these high concentrations, the dye molecules may start to aggregate; the effect can be to reduce the device efficiency because of self-quenching processes.¹⁰

For some material combinations, the mismatch of energy levels can result in the formation of heterojunctions, which

reduce the current flow. Recently, Lai *et al.*¹¹ have shown that this problem can be alleviated by doping the emissive layer with the material used in either the electron-transporting layer (ETL) or the hole-transporting layer (HTL) to form “double hosts.” Tsai and Jou¹² have also revealed an improvement in the carrier transport by doping NPB into an emissive layer. Unfortunately, such double host devices lead to complications in the WOLED manufacture because of the difficulty in controlling accurately the doping concentration.

Here, we describe the introduction of a mixed-transition layer (MTL) between the emissive layer and the ETL (or HTL). The MTL is composed of the emissive dye and the material used in the ETL (or HTL). As a result, the MTL favors the transport of electrons (or holes). If the dye concentration of the MTL is judiciously chosen, it is anticipated that some carriers (holes or electrons) could hop between the dye molecules from the ETL (or HTL) to the subsequent emissive layer without encountering the large energy barrier associated with a heterojunction, leading to an improvement in the performance of the WOLED.

The configurations of WOLEDs and chemical structures of the various compounds used in our work are shown in Fig. 1. The blue fluorescent dye, *N*-(4-((*E*)-2-(6-((*E*)-4-(diphenylamino)styryl)naphthalen-2-yl)viny)phenyl)-

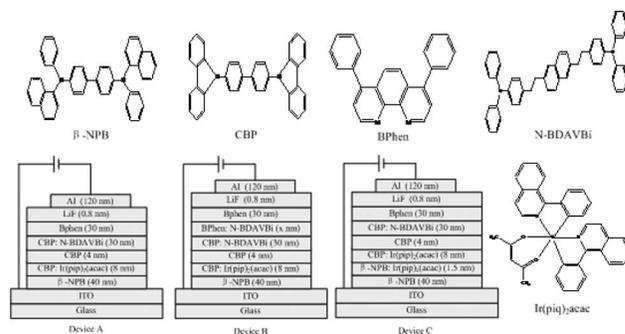


FIG. 1. Configurations of WOLEDs and chemical structures of the compounds used.

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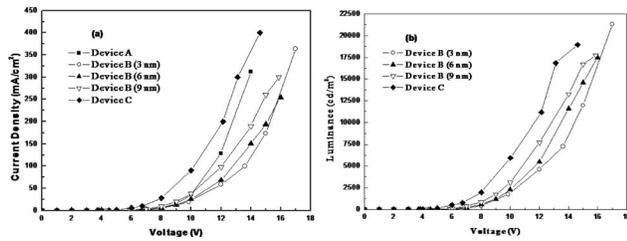


FIG. 2. (a) Current density (J) versus voltage (V) and (b) luminance (L) versus voltage characteristics of the different device configurations.

N-phenylbenzenamine (*N*-BDAVB_i), and the red phosphorescent dopant, bis[1-(phenyl)isoquinoline] iridium (III) acetylacetonate [$\text{Ir}(\text{piq})_2(\text{acac})$], were used to dope the same host, *N,N'*-dicarbazolyl-4,4'-biphenyl (CBP). The optimal doping concentration of these two dyes in CBP is 2 wt %¹³ and 8 wt %, ¹⁴ respectively. *N,N'*-Bis(naphthalene-2-yl)-*N,N'*-bis(phenyl)-benzidine (β -NPB) was used as the hole-transporting layer, and BPhen as the hole-blocking and electron-transporting layer. Device A is the reference device structure, in which the thicknesses of the organic layers and the LiF/Al are have been optimized, as indicated in Fig. 1. The 4 nm undoped CBP layer prevents direct energy transfer from the blue dye to the red phosphor and also transfers triplet excitons to the phosphorescent compound.^{15,16}

Detailed process of fabrication and measurement for WOLEDs have been described in our previous paper.^{12,13}

In order to investigate the effects of the MTL, we have fabricated device B structures (Fig. 1) by inserting an additional Bphen:*N*-BDAVB_i layer below the BPhen layer of device A. From the data shown in Fig. 2, the current density for device B structures is significantly lower than that of device A, measured at the same voltage. This suggests that the effect of carrier trapping, resulting from the mismatch of the energy levels between *N*-BDAVB_i and BPhen [Fig. 3(a)], is more significant than that of improved carrier transport in the dye molecules. It is also evident from Fig. 2 that the current density in device B structures increases with increasing thickness of the MTL. This can be explained as follows. Because of the electron-transporting property of BPhen, holes tend to accumulate at the CBP/BPhen interface and then hop between the dye molecules in the CBP: *N*-BDAVB_i layer to the MTL. In contrast, electrons are injected from the lowest unoccupied molecular orbital (LUMO) of BPhen to that of the blue dye. As a result, some electrons could be expected to hop directly between *N*-BDAVB_i molecules in the MTL to the CBP:*N*-BDAVB_i layer. By increasing the thickness of the MTL, the number of the dye molecules in the BPhen will increase, thereby improving the probability of electrons moving to the *N*-BDAVB_i molecules then to the

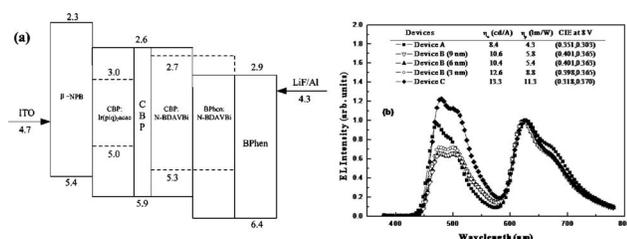


FIG. 3. (a) Energy level diagram of Device B; (b) EL spectra of the devices at a current density of 20 mA/cm² (for Device B, $x=3$ nm, 6 nm, 9 nm); the inset shows the performance of different devices (η_c : maximum current efficiency, η_p : maximum power efficiency).

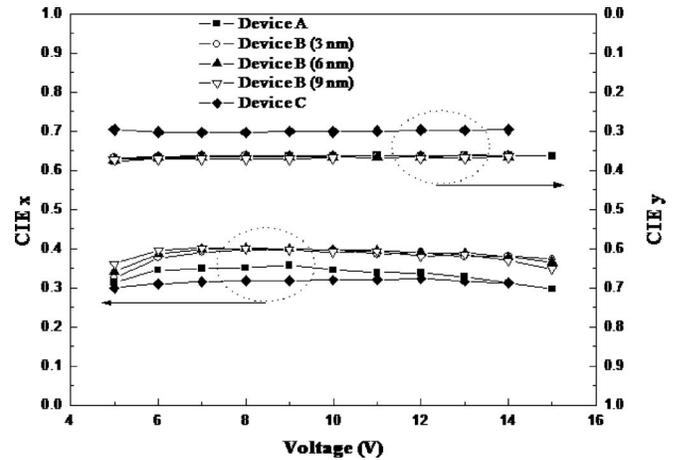


FIG. 4. CIE_{x,y} coordinates of the devices.

emissive layer. Further evidence of improved electron transport is provided by examining the EL spectra of the devices at 20 mA/cm² shown in Fig. 3(b). As the thickness of the Bphen:*N*-BDAVB_i layer increases from 3 to 9 nm, the blue emission is slightly reduced. This suggests that more electrons have transferred to the red layer and then recombined with holes in $\text{Ir}(\text{piq})_2\text{acac}$. In summary, the electron transport can be improved by inserting a MTL which consists of the dye (*N*-BDAVB_i) doped into an electron transport material (BPhen).

From data shown in the inset to Fig. 3(b), the maximum current and power efficiency of device B with 3-nm-thick MTL is 12.6 cd/A and 8.8 lm/W at 0.01 mA/cm², respectively, which is 1.5 and 2 times higher than the corresponding values for device A (8.4 cd/A at 2.2 mA/cm² and 4.3 lm/W at 0.5 mA/cm²). In general, the exciton formation probability is proportional to $n \times p$, where n and p denote the density of electrons and holes, respectively. If n is increased, more excitons will form, leading to the higher efficiency that is observed. In addition, a pronounced efficiency roll-off at high current densities, due to triplet-triplet annihilation,¹⁴ is noted. The decreased efficiencies of device B are observed on increasing the thickness of mixed layer. This may be attributed to the strong exciplex formation between *N*-BDAVB_i and BPhen. In the MTL, holes are mainly located in the highest occupied molecular orbital (HOMO) of the *N*-BDAVB_i because of the preferred electron-transporting property of BPhen, while the electrons are predominantly found in the LUMO level of BPhen owing to the 0.2 eV energy barriers between the LUMO of these two materials [Fig. 3(a)]. As a consequence, the 2.4 eV energy gap, between the HOMO of *N*-BDAVB_i and the LUMO of BPhen, matches the green emission band and exciplexes form. Unfortunately, these exciplexes usually reduce the efficiency,^{17,18} accounting for the decreased efficiency with increasing thickness of the MTL. When the thickness of the MTL is greater than 6 nm, the maximum efficiencies are almost the same. The detailed reasons for this are unclear. A probable explanation is that the number of the exciplexes has saturated with the 6-nm-thick MTL. The exciplex emissions broaden the electroluminescence spectrum by adding the green emission, resulting in tricolor emission.

The improvement of the device with the additional 2 wt % Bphen:*N*-BDAVB_i layer is useful, but rather limited. We could not increase the dye concentration further as we

have noted a reduction in efficiency for single layer BPhen:N-BDAVBi device structures (data not shown in this paper). The concentration of phosphorescent dyes used in WOLEDs is usually higher than that of fluorescent dyes, because of the short-range process of Dexter transfer.¹⁹ Therefore, we have inserted a 1.5-nm-thick 8 wt % NPB:Ir(piq)₂acac layer as the MTL to form device C. Increased hole transport is suggested by the *J-V* characteristics depicted in Fig. 2 and the enhanced blue emission shown in Fig. 3(b). This device exhibits a maximum in the current and power efficiency of 13.3 cd/A and 11.3 lm/W at 0.008 mA/cm² [data inset in Fig. 3(b)], respectively, which is 1.6 and 2.6 times higher than these of device A. No exciplex emission has been observed between NPB and Ir(piq)₂acac. This may be one reason why the maximum current efficiency of device C is higher than that of device B. Overall, inserting the MTL doped with the the 8 wt % Ir(piq)₂acac is an effective approach to improve the carrier transport and circumvent the effects of carrier trapping.

Figure 4 depicts the variation of CIE coordinates with the applied voltage for the different devices. The colors of devices B and C are more stable than that of device A. For example, the CIE coordinates of device C change only slightly, from (0.300, 0.371) to (0.312, 0.366), as the voltage is increased from 6 to 14 V. This suggests that the balance of hole and electron currents has been improved within the recombination zone.

In conclusion, improved performance of WOLEDs has been obtained by inserting a MTL of Bphen:N-BDAVBi [or NPB:Ir(piq)₂acac]. This is attributed to the enhancement of carrier transport by the doping approach. Carriers can hop directly between dye molecules in the MTL to the subsequent layer, resulting in an increase in the exciton formation probability and the device efficiency. This work suggests a simple and effective means of improving the performances of white organic light-emitting devices.

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