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Long-lifetime, high-efficiency white organic light-emitting diodes with mixed host composing double emission layers

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A long-lifetime, high-efficiency white organic light-emitting diode was fabricated with a mixed host in one of double emission layers. The first layer comprised yellow rubrene doped in a mixed host consisting of 50% *N*,*N*′diphenyl-*N*,*N*′-bis-(1-naphthyl)-1,1′-biphenyl-4-4′-diamine (NPB) and 50% 2-(*t*-butyl)-9,10-bis(2′-naphthyl)anthracene (TBADN). The second layer comprised blue 4,4′-bis[2-{4-(*N*,*N*-diphenylamino)phenyl}vinyl] biphenyl doped in TBADN. This device exhibited the longest lifetime, five times that of its pure NPB counterpart. The resulting efficiency was 6.0 lm/W (10.9 cd/A) at 10 mA/cm^2, 33% better than that of the NPB counterpart. These improvements were attributable to the mixed-host structure, which effectively dispersed carriers and gave a good charge balance. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2408663]

Organic light-emitting diodes (OLEDs), particularly white OLEDs, are attracting interest because of their potential as flat-panel displays and for liquid-crystal-display backlighting and solid-state lighting applications.[1–4] These applications require white OLEDs with high brightness, high efficiency, high-color stability, and long lifetime.[2–4] Numerous bright, efficient, and color-stable white OLEDs have been reported.[5–10] Applications such as illumination and high-quality displays demand relatively long lifetimes. For example, a lifetime of at least 100 000 h is required for televisions.[11] Numerous device-lifetime studies have therefore been performed.[11–15] The use of a mixed host in a single emission layer significantly increased the lifetime, but reduced the efficiency.[16–18] Double emission layers frequently result in an unstable color because of the probability of a shift in the recombination zone.[19,20] However, Ma *et al.* used mixed layers for the blue emission layer and electron-transport layer (ETL). This improved the performance of white OLEDs with double emission layers.[21]

This report presents a white OLED with improved lifetime, efficiency, and color stability. The device comprises two emission layers. The first one emits yellow light obtained by doping 1 wt. % rubrene in a mixed host consisting of *N*,*N*′diphenyl-*N*,*N*′-bis-(1-naphthyl)-1,1′-biphenyl-4-4′-diamine (NPB) and 2-(*t*-butyl)-9,10-bis(2′-naphthyl)anthracene (TBADN). The second layer emits blue light by doping 2 wt. % 4,4′-bis[2-{4-(*N*,*N*-diphenylamino)phenyl}vinyl] biphenyl (DPAVBi) in TBADN. A device with a 50% NPB and 50% TBADN mixed host has the longest lifetime, two or five times that of the counterparts with a pure TBADN or NPB host. The resulting efficiency is 6.0 lm/W or 10.9 cd/A at 10 mA/cm^2, 20% or 33% better than that of the TBADN or NPB counterpart.

Figure 1 shows energy-level diagrams of white OLED structures studied and the chemical structures of the materials used. Each device comprises 10 Ω/sq indium tin oxide glass, 70 nm hole-injection layer of 4,4′,4″-tris(*N*-(2-naphthyl)-*N*-phenylamino)-triphenylamine, 20 nm hole-transport layer (HTL) of NPB, 7 nm yellow-emission

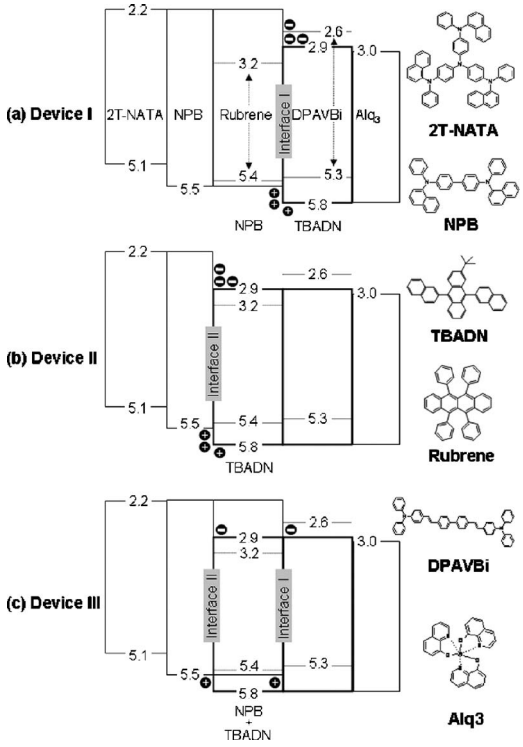

FIG. 1. Schematic energy-level diagrams of white OLEDs with two emissive layers. The first layer is yellow rubrene doped in three different hosts: (a) NPB (device I), (b) TBADN (device II), and (c) NPB/TBADN mixed host (device III). The second layer consists of blue DPAVBi doped in TBADN. Also shown are the chemical structures of the organic materials used.

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TABLE I. Effect of NPB:TBADN mixing ratio on the EL characteristics of white OLEDs studied.

Mixing ratio		At 10 mA/cm^2				CIE 1931 (x,y) chromatic coordinates		At initial 5000 cd/m^2
NPB (%):	TBADN (%)	Voltage (V)	Luminance efficiency (cd/A)	Power efficiency (lm/W)	EQE (%)	At 100 cd/m^2	At 10 000 cd/m^2	Half lifetime (hour)
100	0	6.1	8.7	4.5	3.7	(0.320, 0.347)	(0.279, 0.300)	18
75	25	5.9	9.9	5.3	4.3	(0.321, 0.347)	(0.289, 0.316)	25
50	50	5.8	10.9	6.0	4.7	(0.329, 0.353)	(0.316, 0.341)	100
25	75	5.7	10.4	5.8	4.5	(0.328, 0.351)	(0.327, 0.346)	86
0	100	6.1	9.7	5.0	3.8	(0.386, 0.411)	(0.379, 0.394)	51

layer, 23 nm blue-emission layer, 20 nm ETL of tris-(8-hydroxyquinoline) aluminum (Alq_3), 1 nm lithium fluoride, and 150 nm thick aluminum cathode. Five NPB:TBADN mixing weight ratios, 100:0, 75:25, 50:50, 25:75, and 0:100, were investigated, and the resulting electroluminescent (EL) characteristics are summarized in Table I. The devices were encapsulated with desiccant incorporated. The EL characteristics, spectra, and lifetimes were obtained using a Keithley 2400 electrometer and a Minolta CS-1000S spectrophotometer. The external quantum efficiency was then calculated directly from these measured properties.[22]

Figure 2 shows the effect of mixed-host ratio on the EL spectra of the devices at 10 000 cd/m^2. The device with the pure NPB host, device I, yielded a bluish-white EL spectrum with stronger intensity of the blue emission, while the device with the pure TBADN host, device II, emitted yellowish-white light. The emission from the device with the mixed host, device III, exhibited pure white, with Commission International de L'Eclairage (CIE) coordinates (0.316, 0.341), for a 50% TBADN with 50% NPB mixture.

Figure 3 shows the effect of mixed-host ratio on the device lifetime. The lifetime was measured under constant current with an initial brightness of 5000 cd/m^2 at room temperature. The lifetime of device I was 18 h, while that of device II was 51 h. The lifetime of device III was improved by the introduction of TBADN into NPB. Mixing 50% TBADN with 50% NPB yielded a maximum lifetime of 100 h, which was nearly two or five times that of the pure TBADN or NPB counterpart, respectively.

The marked lifetime improvement may be attributed to the following reasons. First, the carriers were effectively dispersed in the vicinity of two interfaces in device III, but only one in device I or II (Fig. 1), thus preventing damage by excessive charge accumulation at the emissive interface.[14,15] Specifically, the recombination zone of device I was localized mainly at the interface (interface I in Fig. 1) between the yellow-emission and blue-emission layers. However, recombination occurred mostly in the blue-emission layer. This is because the barrier to the transport of holes from the yellow-emission layer to the blue-emission layer is 0.3 eV, which is much lower than the barrier to the transport of electrons from the blue-emission layer to the yellow-emission layer, which is 0.7 eV. This can also be evidenced by the blue emission dominant in device I in Fig. 2.

Similarly, the recombination zone of device II was localized at the interface (interface II in Fig. 1) between the HTL and yellow-emission layer, and recombination occurred mostly at the side of the yellow-emission layer. Therefore, strong yellow emission was observed from the resulting device, as revealed by the EL spectra in Fig. 2.

In device III, some holes could be transported freely from the HTL to interface I via the NPB in the mixed host. Some electrons could be transported freely from the blue-emission layer to interface II via the TBADN in the mixed host. Typically, the lifetime of a blue OLED is shorter than that of a yellow one. Effectively protecting the blue-emission layer can frequently prolong the lifetime of a given white OLED. One major degradation mechanism is the damage caused by excessive charge accumulation.[14,15] Device III, with the mixed host in the yellow-emission layer, effectively dispersed the carriers into the two interfaces, interfaces I and II, thus preventing excessive charge accumulation. Consequently, both the yellow-emission and blue-emission layers were protected and this resulted in a longer device lifetime.

Secondly, the holes were effectively consumed by recombination in device III, so fewer unrecombined holes en-

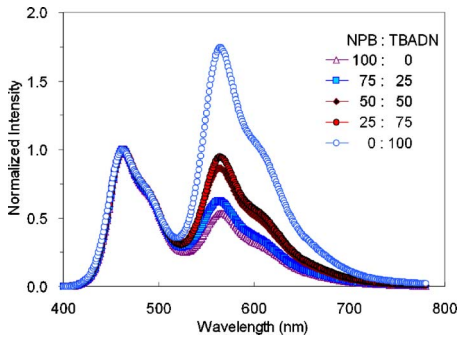

FIG. 2. (Color online) Effect of NPB:TBADN mixing ratio on the EL spectra of the resultant white OLEDs at 10 000 cd/m^2.

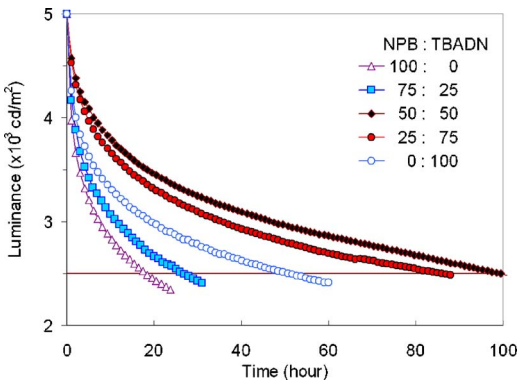

FIG. 3. (Color online) Effect of NPB:TBADN mixing ratio on the lifetime of white OLEDs driven under constant current with an initial brightness of 5000 cd/m^2 at room temperature.

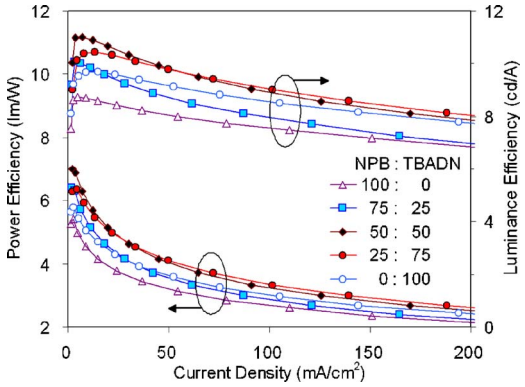

FIG. 4. (Color online) Effect of NPB:TBADN mixing ratio on the power efficiency and luminance efficiency of white OLEDs.

tered the ETL (Alq_3), preventing damage caused by the formation of the unstable Alq_3 cationic species.[12,15]

Thirdly, the power efficiency of device III was higher than that of the others, as shown in Fig. 4. More efficient devices generated less heat upon emission, preventing damage caused by the generation of excess heat during operation.[15] The efficiency at 10 mA/cm^2 was 4.5 lm/W (8.7 cd/A) for device I and 5.0 lm/W (9.7 cd/A) for device II. Device III with a 50% NPB and 50% TBADN mixed host also had a significantly higher efficiency of 6.0 lm/W (10.9 cd/A). Specifically, the major recombination zone of device I was localized at interface I. Hence, there were more holes but fewer electrons in the yellow-emission layer and fewer holes but more electrons in the blue one. In device II, the major recombination zone was localized at interface II, so there were fewer holes but more electrons in both layers. Hence both devices would exhibit comparatively poorer efficiency due to their poorer charge balance. The introduction of the mixed host in device III provided a path free of energy barriers for some holes to be transported further into the electron-rich blue-emission zone and a similar path for some electrons to be transported further into the hole-rich yellow one. This resulted in a better charge balance and hence a higher efficiency was obtained.

Device III had a longer lifetime because of its mixed-host structure. Interestingly, the lifetime of unmixed device II was nearly three times longer than that of unmixed device I. There are three reasons for this. First, device II possessed comparatively better thermal stability because it employed a host, TBADN, whose glass-transition temperature (T_g) is 126 °C, significantly higher than that of the pure NPB host in device I (T_g=99 °C). Secondly, device II exhibited comparatively higher power efficiency, so that less heat would be generated during its operation. Thirdly, in device II, the recombination zone was localized mainly at interface II, far away from the ETL, and, moreover, rubrene was an efficient hole trapper so that comparatively fewer holes could be transported to the ETL. Consequently, the amount of damage caused by the formation of the unstable Alq_3 cationic species would be less.

Besides its long lifetime and high efficiency, device III also exhibited excellent color stability. As shown in Table I, the chromatic variation was (0.013, 0.012) for device III with a 50% NPB and 50% TBADN mixed host for brightness between 100 and 10 000 cd/m^2. The color stability was even better, its chromatic variation being (0.001, 0.005) when a 25% NPB and 75% TBADN mixed host was used. However, the power efficiency and lifetime decreased slightly.

In conclusion, white OLEDs with a mixed host in one of the double emission layers exhibited marked improvements in lifetime and efficiency. Doping 1 wt. % yellow rubrene in a 50% NPB and 50% TBADN mixed host and 2 wt. % blue DPAVBi in TBADN yielded a device with the longest lifetime of 100 h, at an initial brightness of 5 000 cd/cm^2, which was five times that of a device with pure NPB. The resultant efficiency was 6.0 lm/W or 10.9 cd/A at 10 mA/cm^2, 33% better than that of the pure NPB counterpart. The improvements are attributable to the mixed-host architectures, which effectively dispersed the carriers into the two interfaces and gave a good charge balance. Additionally, the diode exhibited excellent color stability.

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