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Solution-processed white organic light-emitting diodes with mixed-host structures

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ABSTRACT

Efficient white light-emitting diodes (WOLEDs) were fabricated with a solution-processed single emission layer composed of a molecular and polymeric material mixed-host (MH). The main host used was a blue-emitting molecular material of 4,4′-bis(2,2′-diphenylvinyl)-1,1′-biphenyl (DPVBi) and the assisting host used was a hole-transport-type polymer of poly(9-vinylcarbazole) (PVK). By co-doping 4,4′-bis[2-(4-(*N*,*N*-diphenylamino)phenyl)vinyl]biphenyl and 5,6,11,12-tetraphenylnaphacene into the MH, the performances of the fabricated devices made with different mixing ratio of host materials were investigated, and were to depend on the mixing ratios. Under the optimal PVK:DPVBi ratio (3:7), we achieved a maximum luminance of 14 110 cd/m^2 and a maximum current efficiency of 9.5 cd/A. These improvements were attributed to the MH structure, which effectively improved the thermal stability of spin-coated film and enhanced the hole-injection/transporting properties of WOLEDs.

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1. Introduction

White organic light-emitting diodes (WOLEDs) have highly promising applications in areas such as full-color flat-panel displays, liquid-crystal-display back-lighting and large area illumination [1–4]. In order to obtain white emission from organic light-emitting diodes, a general strategy is to use different organic materials, which emit at different primary colors to fully cover the whole visible spectrum range [2–4]. Currently, there are many approaches reported to realize WOLEDs, mainly by employing multiple emissive layers in which each layer emits a primary color [1–6], or a single emissive layer doped simultaneously with different color light-emitting materials [7–11]. The fabrication process of WOLEDs through multilayer structure is very complex, the emission colors of device are usually sensitive to the bias voltage or current density because the shift of exciton-recombination zone [12,13]. Single-layer WOLEDs consisting of only one active organic layer can emit in the entire visible range and can achieve higher Commission International de l'Eclairage (CIE) coordinates stability [14,15].

Single-layer small molecular WOLEDs are usually fabricated by the co-evaporation of different emitting dyes and host materials in a single emission layer [7–11]. The doping concentration of dyes must be controlled carefully to obtain balanced white emission, which is a difficult task to achieve in the co-evaporation process and usually leads to a fabrication process of great complexity, poor reproducibility and high cost. As an alternative approach, solution-processing is a more desirable technique for low-cost manufacture because it is easy to realize co-doping of several dopants and compatible with low-cost, large area manufacture technology such as spin-coating [14–16], ink-jet printing [17] and screen-printing [18,19]. In the past few decades, the research on solution-processed WOLEDs has predominantly focused on polymer blends or dyes-doped polymers [20–23]. Recently, solution-processed small molecule thin films have drawn much attention because it was found that highly efficient small molecular materials, which were usually used in vacuum-deposition, can be utilized in this process, thus having the aforementioned advantages [14–16,24–26]. Solution-processed WOLEDs employing small molecules as hosts show good performances that were comparable to the vacuum-deposited counterparts, with much improved process feasibility [15].

As a blue-emitting molecular material, 4,4′-bis(2,2′-diphenylvinyl)-1,1′-biphenyl (DPVBi) has been a widely used host material in WOLEDs [7,8]. However, we observed significant crystallization of DPVBi in spin-coated films using DPVBi [15], which is due to its low glass-transition temperature of DPVBi (64 °C) [27]. We also found that the crystallization of DPVBi can be suppressed by

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mixing other materials with higher glass-transition temperatures [15]. Following this idea, we introduced a polymeric assisting host of poly(9-vinylcarbazole) (PVK) with higher glass-transition temperature of 210 °C to a molecular main host of DPVBi with a high energy transfer capability [28,29]. Using this mixed-host (MH), we fabricated single emissive layer WOLEDs, which is doped with blue dye of 4,4′-bis[2-(4-(*N*,*N*-diphenylamino) phenyl)vinyl] biphenyl (DPAVBi) and yellow dye of 5,6,11,12-tetraphenylnaphacene (Rubrene). These WOLEDs show high luminance and stable white-emitting behavior under a driving voltage. We also found that the MH can significantly improve the luminance and current efficiency of the devices than those made of single host. We further investigated the optimal PVK:DPVBi ratio by fabricating devices with different MH ratios. At a weight ratio of 3:7 (PVK:DPVBi), the WOLED exhibited a maximum current efficiency of 9.5 cd/A, which is 1.56 times as large as that of the single host device (DPVBi). The significant improvement was attributable to the mixed-host structure, which effectively improved the thermal stability of spin-coated films and enhanced the hole-injection/transporting properties of WOLEDs.

It is clearly demonstrated in our work that MH is one important factor to consider in building WOLEDs, and an optimal mixing ratio should be sought in order to design WOLEDs with high efficiency. It is also demonstrated that another advantage of solution-processing is that the ratio of MH and the dopant concentration can be easily manipulated. Furthermore, the molecular and polymeric materials mixed-host can be applied in solution-involved printing technologies (such as ink-jet printing and screen-printing) to fabricate WOLEDs [18].

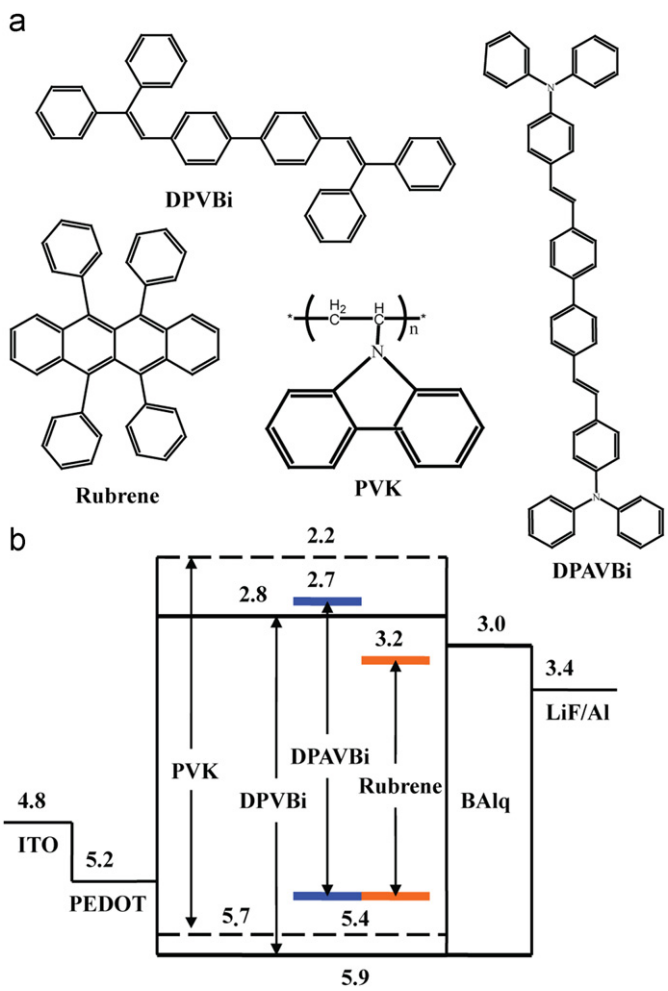

Fig. 1. (a) Chemical structures of the emissive materials used, and (b) schematic energy–level diagrams of white OLEDs.

2. Experimental

The devices based on the MH structure were fabricated on indium tin oxide (ITO)-coated glass with the structure, ITO/PEDOT:PSS(30 nm)/EML(50 nm)/BAlq(30 nm)/LiF(0.5 nm)/Al(100 nm). Poly (3,4-ethylenedioxythiophene):Poly (styrenesulfonate) (PEDOT:PSS) and bis(2-methyl-8-quinolinato)-4-phenylphenolate aluminum (BAlq) were used as the hole-injection layer (HIL) and electron-transport layer (ETL), respectively. DPAVBi and Rubrene co-doped PVK:DPVBi MH was used as white-emitting layer (EML). Fig. 1(a) shows the chemical structures of the emissive materials used for the WOLEDs.

For all of the MH devices, the doping concentrations of DPAVBi and Rubrene were 6 % and 0.36 %, respectively. The composition of the EMLs used in five devices (labeled as A, B, C, D, and E) is summarized in Table 1. The five devices A, B, C, D, and E have different PVK:DPVBi weight ratios, which are 0:10, 2:8, 3:7, 4:6, and 10:0, respectively. In the experiment, the EML materials were dissolved in chlorobenzene to prepare solution with concentration of 15 mg/ml. The water-dispersed PEDOT:PSS mixture was spin-coated on top of the ITO substrate at a spin speed of 2000 rpm for 60 s. The PEDOT:PSS layer was baked at 120 °C for 30 min. It has a thickness of ~30 nm and acts as the HIL. The EML was then prepared on this HIL by spin-coating, and then it was baked at 120 °C for 5 min. The thickness of the light-emitting layer is found to be about 50 nm. After baking, the BAlq layer (30 nm) and the cathode LiF (0.5 nm)/Al (100 nm) were deposited by thermal evaporation in a vacuum chamber at a pressure of 5×10^{-4} Pa, respectively. The schematic energy-level diagram of the devices is shown in Fig. 1 (b). Following the same procedure as the above, hole-only devices were also fabricated. The structures of the hole-only devices are ITO/PVK (80 nm)/NPB (20 nm)/Al (100 nm) and ITO/DPVBi (80 nm)/NPB (20 nm)/Al (100 nm).

Ultraviolet-visible (UV–vis) absorption and photoluminescence (PL) spectra were measured with UV–vis spectrophotometer (U-3010, Hitachi Inc) and fluorescence spectrophotometer (FluoroMax-4, HORIBA Jobin Yvon Inc). The film thickness was measured with an ellipsometer. The current–voltage-luminescence

Table 1
The composition of the EML and the EL characteristics of WOLEDs, maximum luminance L_{max}, maximum current efficiency $\eta_{c,max}$ and maximum power efficiency $\eta_{p,max}$.

No	Weight ratio				Performances			
	PVK	DPVBi	DPAVBi (x%)	Rubrene (y%)	L_{max} (cd/ m^2)	$\eta_{c,\ max}$ (cd/A)	$\eta_{p,\ max}$ (lm/W)	CIE(x,y) 5 V/10V
A	0	10	6	0.36	8000	6.1	3.5	(0.328.0.458)/(0.295.0.428)
B	2	8	6	0.36	10300	8.0	4.6	(0327.0.419)/(0.309.0.400)
C	3	7	6	0.36	14110	9.5	5.1	(0.323,0.408)/(0.318,0.409)
D	4	6	6	0.36	9210	6.5	3.7	(0.309,0.415)/(0.303,0.402)
E	10	0	6	0.36	840	2.2	1.3	(0.268,0.420)/(0.208,0.353)

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characteristics were measured with a Keithley 2602 source meter and Luminance meter. Electroluminescence (EL) spectra and CIE coordinates of the devices were analyzed with a spectrometer (PR650). All measurements were carried out under ambient conditions at room temperature.

3. Results and discussion

Fig. 2 shows the luminance-current density, current efficiency–voltage, and power efficiency–voltage characteristics of devices A–E. Among all the devices, device C shows the best performance. It presents a maximum current efficiency ($\eta_{c,\ max}$) of 9.5 cd/A at 6.7 V and a maximum power efficiency ($\eta_{w,\ max}$) of 5.1 lm/W at 5.7 V and a maximum luminance (L_{max}) of 14,110 cd/m^2 at a current density of 373 mA/cm^2. The luminance and current efficiency of the devices with MH (B–D) are higher than that with only DPVBi (device A) or PVK (device E). As we can see, device A shows a $\eta_{c,\ max}$ of 6.1 cd/A and a L_{max} of 8000 cd/m^2, while device E shows a $\eta_{c,\ max}$ of 2.2 cd/A and a L_{max} of 840 cd/m^2. The EL characteristics of the WOLEDs employing the MH were summarized in Table 1. In comparison with previously reported

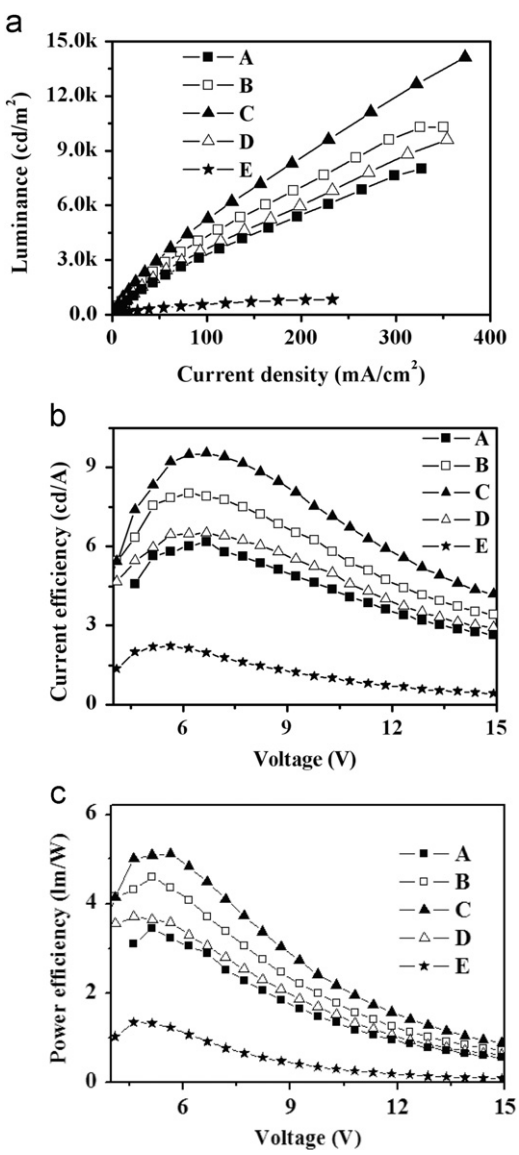

Fig. 2. (a) The luminance–current density, (b) current efficiency–voltage, and (c) power efficiency–voltage characteristics of device A–E.

WOLEDs with MH structure, the efficiency of device C was comparable to that of the vacuum-deposited multilayer WOLEDs based on the DPAVBi and rubrene as dopants (10.9 cd/A) [1]. In addition, the performances of our devices were better than that of solution-processed WOLEDs with MH structure in previous reports, in which the reported best current efficiency is 6.0 cd/A (1000 cd/m^2) [14] and 8.3 cd/A (1000 cd/m^2) [15].

The higher efficiency and luminance of devices B–E can be attributed to the blend of PVK and DPVBi used as MH. We think two factors are responsible for the improvement of the device performance. First, with the adding of PVK into the blends, the thermal stability of spin-coated film of the blends was improved. Fig. 3 shows optical microscope images of spin-coated films of (a) DPVBi, (b) PVK:DPVBi=2:8, (c) PVK:DPVBi=3:7, and (d) PVK:DPVBi=4:6. All the organic films were spin-coated on the ITO substrate and annealed for 30 min at temperature 80 °C. Fig. 3 (a) shows the spin-coated film of DPVBi, where significant crystallization of DPVBi was observed during the annealing process. This effect is due to the low glass–transition temperature of DPVBi (64 °C) [27]. With the increasing PVK concentration in the blends, the crystallization of spin-coated film of the blends was reduced as can be seen from Fig. 3(b). As the PVK concentration is increased to 30 wt.%, the crystallization and coagulation can no longer be observed (see Fig. 3(c)). Since PVK has a glass–transition temperature of about 210 °C [28], it is effective on preventing the crystallization from happening in spin-coated films and thus improves the thermal stability of these films. This situation illustrates the ability of the PVK to enhance the processing properties of the materials for spin-coating: the active PVK:DPVBi blend layer deposited by spin-coating can give uniform thin films. Furthermore, the molecular and polymeric materials mixed-host can be applied in solution-involved printing technologies (such as ink–jet printing and screen–printing) to fabricate WOLEDs [18]. It is known that polymeric hosts can markedly improve the thermal stability and mechanical properties of printed thin films [18].

Second, the MH effectively improved the hole-injection/transporting of devices by varying the ratio of the MH [1,30,31]. PVK is used as a hole-transport material, whose hole mobility (10^{-6} cm^2 V^{-1} s^{-1}) is much higher than that of DPVBi (10^{-8} cm^2 V^{-1} s^{-1}) and the hole-injecting barrier of PVK from PEDOT:PSS (0.5 eV) is lower comparing to DPVBi (0.7 eV) [32–36] (see Fig. 1(b)). To distinguish the hole-injection/transporting ability of two hosts, the hole-only devices, ITO/PVK (80 nm)/NPB (20 nm)/Al (100 nm) and ITO/DPVBi (80 nm)/NPB (20 nm)/Al (100 nm), were fabricated. As can be seen from Fig. 4, the hole-only device of PVK has a higher current density compared to device of DPVBi. So the blend of PVK and DPVBi used as MH play the roles of both hole-transport layer and EML in WOLEDs, and the hole-injection and transport in WOLEDs can be adjusted by changing the ratio of PVK and DPVBi in EML, which could improve the balances of carriers in EML [1,30,31].

The efficiency of dye–doped single emissive layer WOLEDs relies on energy transfer from the hosts to the guests, which can be estimated using the overlapping area of the absorption spectra of the guests and the PL spectra of the hosts. Fig. 5(a) shows the absorption spectra of DPAVBi and Rubrene in films, and PL spectra of DPVBi, PVK and DPAVBi in films. In Fig. 5(a), there is a partial spectral overlap between the fluorescent band of the hosts (PVK and DPVBi) and the absorption bands of both DPAVBi and Rubrene, which indicates that the lowest singlet–excited states in the MH can be transferred to the dyes (DPAVBi or Rubrene) via forster energy transfer. In addition, the spectra overlap between the PL of DPAVBi and the absorption of Rubrene suggests that the energy transfer process between them can happen. Fig. 5 (b) shows the PL spectra of the mixed PVK:DPVBi film (3:7), the

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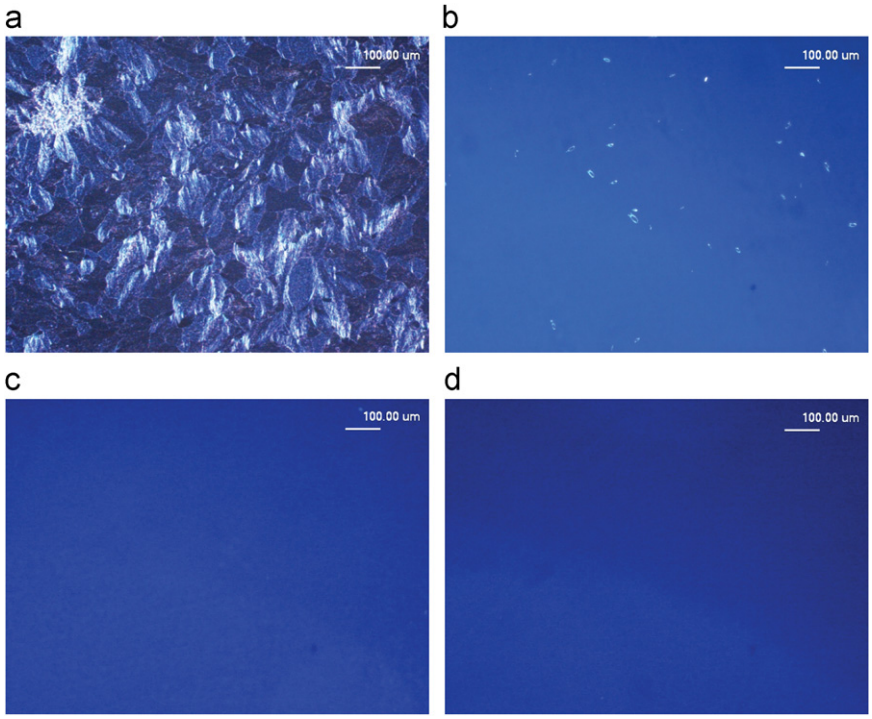

Fig. 3. Optical microscope images of organic films spin-coated on top of ITO substrates: (a) DPVBi, (b) PVK:DPVBi=2:8, (c) PVK:DPVBi=3:7, and (d) PVK:DPVBi=4:6.

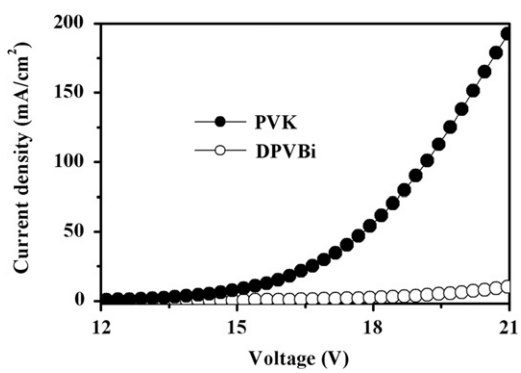

Fig. 4. Current density–voltage characterics of the hole-only devices.

DPAVBi (6 wt.%), Rubrene (0.36 wt.%) and both DPAVBi (6 wt.%) and Rubrene (0.36 wt.%) doped films. The excitation peak wavelength was at 350 nm for all the PL measurement. The DPAVBi and Rubrene show a peak at 468 nm and 550 nm, respectively. In Fig. 5 (b), the efficient energy transfer happens between the PVK and DPVBi, and the MH and DPAVBi, respectively. On other hand, a strong emission from MH is observed when the MH is doped with Rubrene, suggesting the energy transfer from the MH to Rubrene is incomplete with the doping concentration of 0.36 wt.%. When we dope both DPAVBi (6 wt.%) and Rubrene (0.36 wt.%) in the MH, the blue and yellow–emitting band are observed, which implies that the energy in the MH is shared by both dyes.

Fig. 6(a) shows the effect of mixed–host ratio on the EL spectra of the devices at 10 V bias. The EL spectra of the devices show two main peaks at 468 nm and 552 nm originated from the emission of DPAVBi and Rubrene, respectively. The device with the pure DPVBi host, device A, emitted yellowish–white light, while the device with the pure PVK host, device F, yielded a bluish–white EL spectrum with stronger intensity of the blue emission. The intensity of the Rubrene emission gradually decreases with the increasing concentration of PVK. This can be understood from the extent of spectral overlap between the absorption spectrum of

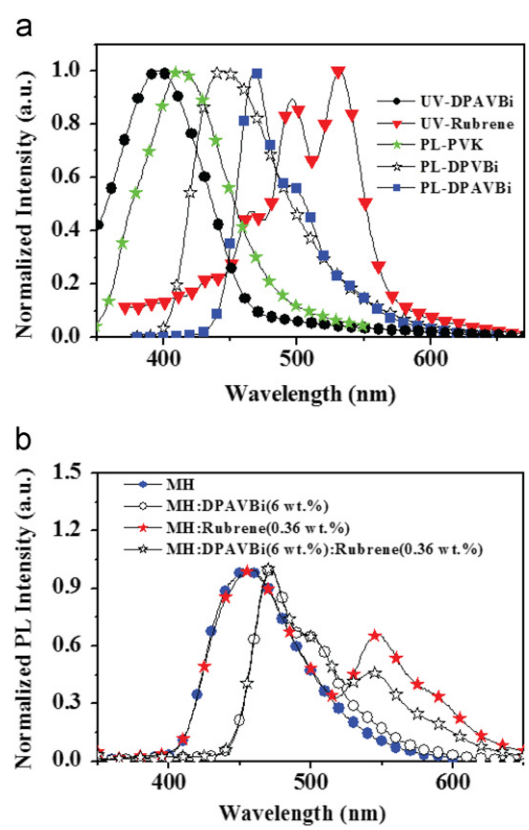

Fig. 5. (a) The absorption spectra of DPAVBi and Rubrene in films and PL spectra of pure PVK, DPVBi and 4 wt.% DPAVBi doped into PMMA film, (b) PL spectra of MH (PVK:DPVBi=3:7) film, DPAVBi (6 wt.%) doped MH, and both DPAVBi (6 wt.%) and Rubrene (0.36 wt.%) doped MH film.

Rubrene and the PL spectra of hosts in Fig. 5(a). There is a larger spectral overlap between the fluorescent band of DPVBi and the absorption bands of Rubrene than that of PVK and Rubrene. At the

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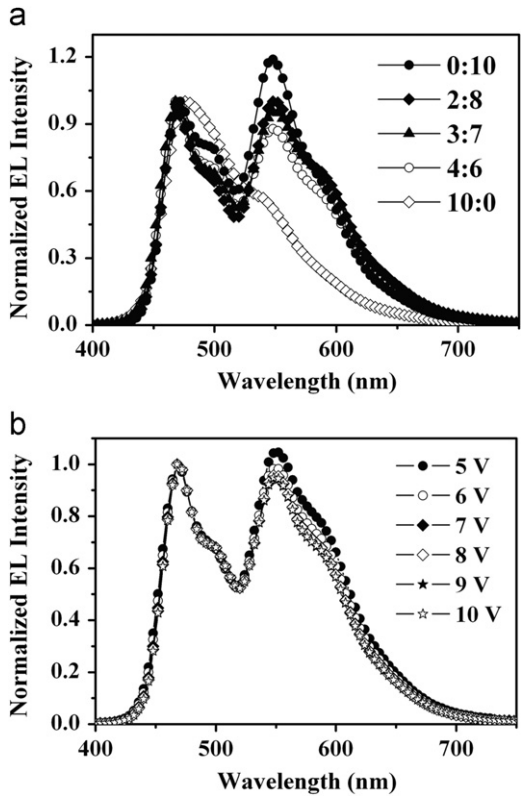

Fig. 6. Normalized EL spectra of (a) device A–E at 10 V bias and (b) device C at different bias voltages.

same doping concentration, the efficiency of energy transfer increases with the extent of spectral overlap between the emission of host and the absorption of dopant. The EL spectra of the device C (PVK:DPVBi=3:7) at different bias voltages are shown in Fig. 5(b). The EL spectra are almost independent of the driving voltage. The CIE coordinates change from (0.323, 0.408) at 5 V to (0.318, 0.409) at 10 V, which is only a slight change.

4. Conclusion

Efficient white light-emitting diodes were fabricated with a solution-processed single emission layer composing of a molecular and polymeric materials mixed-host. It is found that the device performances depend on the mixed-host ratio, and under the optimal PVK:DPVBi ratio (3:7), the WOLEDs show a maximum luminance of 14 110 cd/m^2 and a maximum current efficiency of 9.5 cd/A. These improvements were attributed to the mixed-host structure, which effectively improved the thermal stability of spin-coated film and enhanced the hole-injection/transporting properties of WOLEDs. Furthermore, the molecular and polymeric materials mixed-host can be applied in solution-involved printing technologies (such as ink-jet printing and screen–printing) to fabricate WOLEDs.

Acknowledgments

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