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Letter

A hybrid white organic light-emitting diode with stable color and reduced efficiency roll-off by using a bipolar charge carrier switch

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ABSTRACT

A hybrid white organic light-emitting diode (WOLED) with an emission layer (EML) structure composed of red phosphorescent EML/green phosphorescent EML/spacer/blue fluorescent EML was demonstrated. This hybrid WOLED shows high efficiency, stable spectral emission and low efficiency roll-off at high luminance. We have attributed the significant improvement to the wide distribution of excitons and the effective control of charge carriers in EMLs by using mixed 4,4′,4″-tri(9-carbazoyl) triphenylamine (TCTA) and bis[2-(2-hydroxyphenyl)-pyridine] beryllium ($Bepp_2$) as the host of phosphorescent EMLs as well as the spacer. The bipolar mixed TCTA:$Bepp_2$, which was proved to be a charge carrier switch by regulating the distribution of charge carriers and then the exciton recombination zone, plays an important role in improving the efficiency, stabilizing the spectrum and reducing the efficiency roll-off at high luminous. The hybrid WOLED exhibits a current efficiency of 30.2 cd/A, a power efficiency of 32.0 lm/W and an external quantum efficiency of 13.4% at a luminance of 100 cd/m^2, and keeps a current efficiency of 30.8 cd/A, a power efficiency of 27.1 lm/W and an external quantum efficiency of 13.7% at a 1000 cd/m^2. The Commission Internationale de l'Eclairage (CIE) coordinates of (0.43, 0.43) and the color rendering index (CRI) of 89 remain nearly unchanged in the whole range of luminance.

1. Introduction

White organic light-emitting diodes (WOLEDs) are attracting significant attention due to its unique merits of fabrication of large-scale for solid-state lighting sources, full-color displays and backlights for liquid-crystal displays [1–5]. Recently, luminous efficiency of OLEDs has been greatly improved [6–16]. Thereinto, the phosphorescent WOLEDs showed outstanding efficiency since all electrically generated singlet and triplet excitons can be harvested by phosphorescent emitters [14,17–20]. However, the development of phosphorescent WOLEDs is severely limited by the absence of efficient deep-blue phosphorescent emitters with an operational lifetime suitable for commercial applications [21].

To get over the instability of the blue phosphoresce in WOLEDs, hybrid WOLEDs which combine phosphorescent green and red (or yellow) emitters with fluorescent blue one have been designed and it was found that the hybrid WOLEDs indeed showed improved operational stability [1,6,9,17]. Additionally, since the triplet excitons in the fluorescent EML can diffuse to the phosphorescent EMLs and be harvested by the phosphorescent dopants, achieving an internal quantum efficiency (IQE) of 100% in principle [11], the hybrid WOLEDs can also achieve high efficiency as well. However, the structure of separated phosphorescent emitters and fluorescent emitter in hybrid WOLEDs, which is a common approach for creating white light [1,9,11,16,22], also brings about one problem of color shift with voltage bias due to the shift of recombination region with voltage, representing a drawback of such an approach. Obviously, the reasonable control of charge carriers in EMLs in hybrid WOLEDs becomes a key factor in the structure

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design of hybrid WOLEDs with high efficiency and stable spectrum.

In this paper, a color stable hybrid WOLED with the structure of red phosphorescent EML/green phosphorescent EML/spacer/blue fluorescent EML was presented. Mixed hole-transporting 4,4′,4″-tri(9-carbazoyl)triphenylamine (TCTA) and electron-transporting bis[2-(2-hydroxyphenyl)-pyridine] beryllium ($Bepp_2$) were used as the host of phosphorescent EMLs as well as the spacer to regulate the distribution of charge carriers in EMLs. It can be concluded that the bipolar property of mixed TCTA:$Bepp_2$ contributes to the wide distribution of recombined excitons and the effective control of charge carriers in EMLs, resulting in the stable spectrum emission of the fabricated hybrid WOLED. At the same time, the hybrid WOLED also shows a high efficiency and low efficiency roll-off at high luminance.

2. Experimental

The device structure of the proposed hybrid tricolor WOLED is shown in Fig. 1. All the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) values of the organic materials (TCTA [14], $Bepp_2$ [23], Ir(MDQ)$_2$(acac) [9], Ir(ppy)$_2$(acac) [24] and DSA-ph [25]) are obtained from the representative works. Here, TCTA serves as the hole transporting and electron/exciton blocking layer. $Bepp_2$ is as the electron transporting and hole/exciton blocking layer. As we know, TCTA is a good hole transporting layer (HTL) material with high hole mobility [26], wide band-gap with high triplet energy (2.8 eV) [6], and a fitting PL peak around 390 nm which overlaps well with absorption peak of most of phosphors [8,27], while $Bepp_2$ is a good electron transporting layer (ETL) material with a high electron mobility of $\sim 10^{-4}$ cm^2/V s [28] and wide band-gap with high triplet energy (2.6 eV) [23], which is high enough for red and green phosphors. Therefore, mixed TCTA and $Bepp_2$ was used as the host of phosphorescent EMLs as well as the spacer, exhibiting bipolar transporting property. For red and green emission, Iridium(III)bis(2-methyldibenzo-[f,h]quinoxaline) (acetylacetonate) [Ir(MDQ)$_2$(acac)] and bis(2-phenylpyridinato-N,C$^{2'}$)iridium(acetylacetonate) [Ir(ppy)$_2$(acac)] are employed as red and green dopants, respectively. For blue EML, p-bis(p-N,N-diphenylaminostyryl) benzene (DSA-Ph) is as the fluorescent dopant and $Bepp_2$ as the host. The devices were fabricated on MoO_3-modified ITO substrate, and LiF/Al was deposited through a shadow mask as cathode. All devices were prepared by thermal evaporation in a high-vacuum system with pressure less than 5×10^{-4} Pa.

Current–voltage-luminance characteristics were measured by using a Keithley source measurement unit (Keithley 2400 and Keithley 2000) with a calibrated silicon

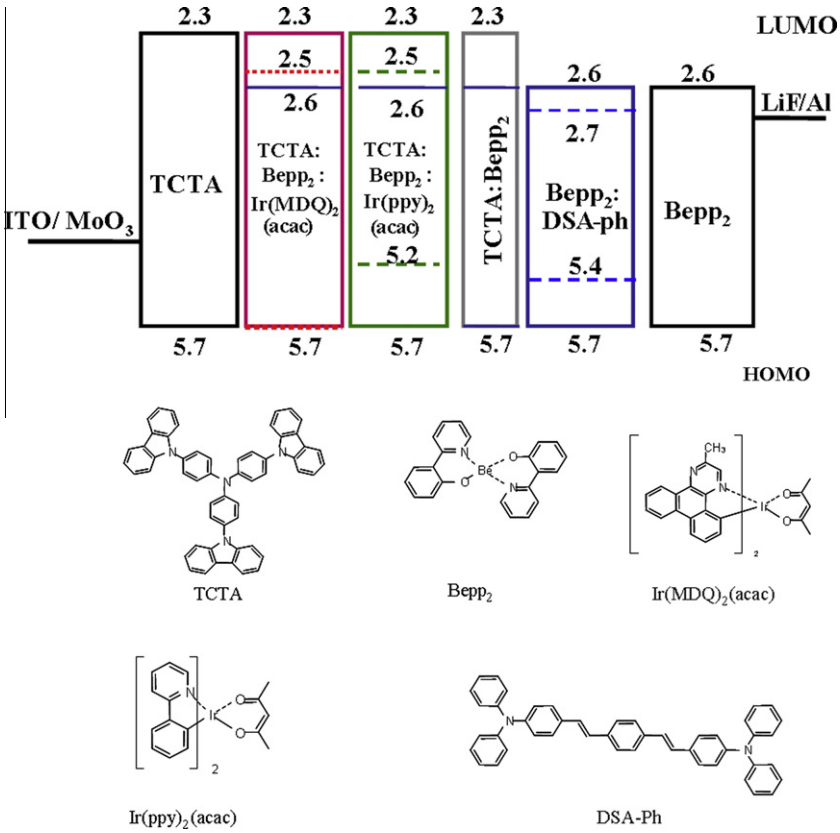

Fig. 1. Energy diagrams of the proposed hybrid WOLED, showing the highest occupied and lowest unoccupied molecular orbital energies (HOMO and LUMO) relative to vacuum (Top) and chemical structures of the organic materials used (Bottom).

photodiode. The EL spectra were measured by spectrascan PR650 spectrophotometer. All the measurements were carried out in ambient atmosphere.

3. Results and discussion

Structural optimization of the proposed hybrid WOLED including systematically varying the thickness and the doping concentrations of EMLs and spacer has been carried out. As shown in the supplementary data, the EL spectra can be effectively controlled by modulating the concentration of $Bepp_2$ to TCTA in the spacer. Firstly we optimized the concentration of $Bepp_2$ in spacer by 100:0, 100:10, 100:15 and 100:20. It is found that the ratio of 100:15 makes the fabricated WOLED for the best spectral emission with high CRI of 89. As we see, the device without spacer deviates from the white emission with the absence of blue emission. The ratio of TCTA:$Bepp_2$ in the red EML has also been optimized with 2:1, 1:1, and 1:2 by fixing the ratio in spacer of 100:15 and in green EML of 1:1, and then 1:1 of TCTA:$Bepp_2$ was selected for the best spectra. The final power efficiency-external quantum efficiency-brightness characteristics and the electroluminescence (EL) spectra at different brightness are shown in Fig. 2. The EL performances at different brightness are also summarized in Table 1. It can be seen that the hybrid WOLED shows high efficiency and low efficiency roll-off. A current efficiency of 30.2 cd/A, a power efficiency of 32.0 lm/W and an external quantum efficiency of 13.4% at luminance of 100 cd/m^2 can be obtained. And the device still keeps a current efficiency of 30.8 cd/A, a power efficiency of 27.1 lm/W and an external quantum efficiency of 13.7% at luminance of 1000 cd/m^2. From 100 to 1000 cd/m^2, the external quantum efficiency and the current efficiency increases instead of decreasing, and the power efficiency decreases by only about 15%. As shown in the inset of Fig. 2, the hybrid WOLED exhibits excellent white emission with three separated emissive peaks, which are clearly originated from red, green and blue EML, respectively. The color-rendering index (CRI) reaches 89 in the whole range of brightness. More importantly, the hybrid WOLED is rather stable that the Commission Internationale de l'Eclairage (CIE) coordinates are (0.43, 0.43), hardly changing from 100 cd/m^2 to 10000 cd/m^2.

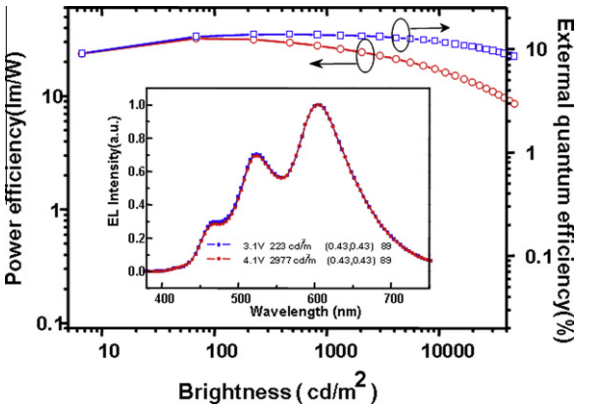

Fig. 2. Power and external quantum efficiencies as a function of brightness for the hybrid WOLED with the structure of ITO/MoO_3(8 nm)/TCTA(60 nm)/ TCTA:Bepp2(1:1):Ir$(MDQ)_2$(acac)(6%, 5 nm)/ TCTA:$Bepp_2$(1:1):Ir$(ppy)_2$(acac)(6%, 5 nm)/TCTA:$Bepp_2$(100:15, 3 nm)/ $Bepp_2$:DSA-ph(2.5%, 10 nm)/$Bepp_2$(30 nm)/LiF(1 nm)/Al. The normalized EL spectra at various voltages are shown in the inset, as well as the corresponding brightness, CIE coordinates, and CRI of the device.

In order to elucidate the origin of high efficiency, reduced efficiency roll-off and stable spectrum of the proposed hybrid WOLED, Device 1, 2, 3, and 4 with structure of ITO/MoO_3(8 nm)/TCTA(60 nm)/TCTA:Ir$(MDQ)_2$(acac) (6%, 5 nm)/TCTA:Ir$(ppy)_2$(acac) (6%, 5 nm)/TCTA:$Bepp_2$(100:*x*, 3 nm)/$Bepp_2$:DSA-ph(2.5%, 10 nm)/$Bepp_2$(30 nm)/LiF(1 nm)/Al (*x* = 10, 15, 20, and 25 in Device 1, 2, 3, and 4, respectively) were fabricated. Here only TCTA was used as the

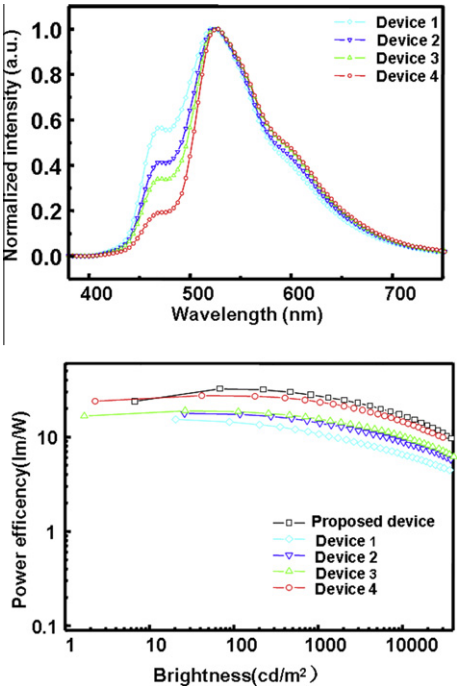

Fig. 3. Normalized EL spectra of Device 1, 2, 3, and 4 at 4 V bias (top), as well as their power efficiencies compared to that of the proposed WOLED at different brightness (bottom).

Table 1
EL characteristics of the hybrid WOLED at different brightness.

Brightness (cd/m^2)	Voltage (V)	PE (lm/W)	CE (cd/A)	EQE (%)	CIE (*x*, *y*)	CRI
100	2.94	32.0	30.2	13.4	(0.43, 0.43)	89
500	3.31	29.4	31.1	13.8	(0.43, 0.43)	89
1000	3.56	27.1	30.8	13.7	(0.43, 0.43)	89

host of red and green phosphorescent EML. Fig. 3 shows the EL spectra of Device 1, 2, 3, and 4 at 4 V bias, as well as their power efficiency compared to that of the proposed WOLED at different brightness. Although three emissive peaks still exist in the EL spectrum, the green emission becomes dominating with respect to red and blue emission and the red emission hardly changes even increasing the ratio of $Bepp_2$ in spacer, which only leads to more blue emission. In addition, the EL efficiencies of Device 1, 2, 3, and 4 are also decreased relative to that of proposed WOLED. Furthermore, obvious variation with the operational voltage appears in the EL spectra of Device 1, 2, 3, and 4, as shown in Fig. 4.

For Device 1, 2, 3, and 4 with only TCTA as the host of the red and green phosphorescent EML, the exciton recombination region should be obviously located at both sides of spacer due to the good hole-transporting property of TCTA [29], resulting in the green and blue emission by excitons in green and blue EML, respectively. The red emission is then originated from the energy transfer of partial excitons in green EML. Furthermore, partial triplet excitons in blue EML are also transferred into the green EML, leading to more green emission. This also explains the reason why more green emission can be observed in Device 1, 2, 3, and 4. It can be seen that the exciton formation in green EML is also dependent on the ratio of $Bepp_2$ to TCTA in spacer. The green emission increases with the growing concentration of $Bepp_2$ because in this case there are more electrons injecting into the green EML, leading to more electrons and holes recombining in green EML. Actually, the narrow exciton recombination region induces exciton quenching easily by the electrical field of accumulated space charges, reducing the efficiency and aggravating the efficiency roll-off. The change of EL spectra with bias voltage in Device 1, 2, 3, and 4 should also be related to the distribution of excitons in the narrow recombination region. As the bias voltage increases, there are more green light and red light emitting with respect to the blue light, which means that there are more excitons recombining in green EML with the increase of voltage bias, resulting in the instability of the emissive spectrum.

However, for the proposed hybrid WOLED, the utilization of bipolar host in red and green EML extends the width of exciton recombination region [29,30], even covering the red, green and blue EML because the injected electrons are easily transported into red EML by doping electron transporting molecule $Bepp_2$ into the EML. The wide exciton recombination region, on the one hand, reduces the exciton quenching caused by space electrical field, thus improving the EL efficiency and efficiency roll-off, on the other hand, stabilizes the exciton distribution in EML so that the change of spectra with voltage bias can be greatly reduced.

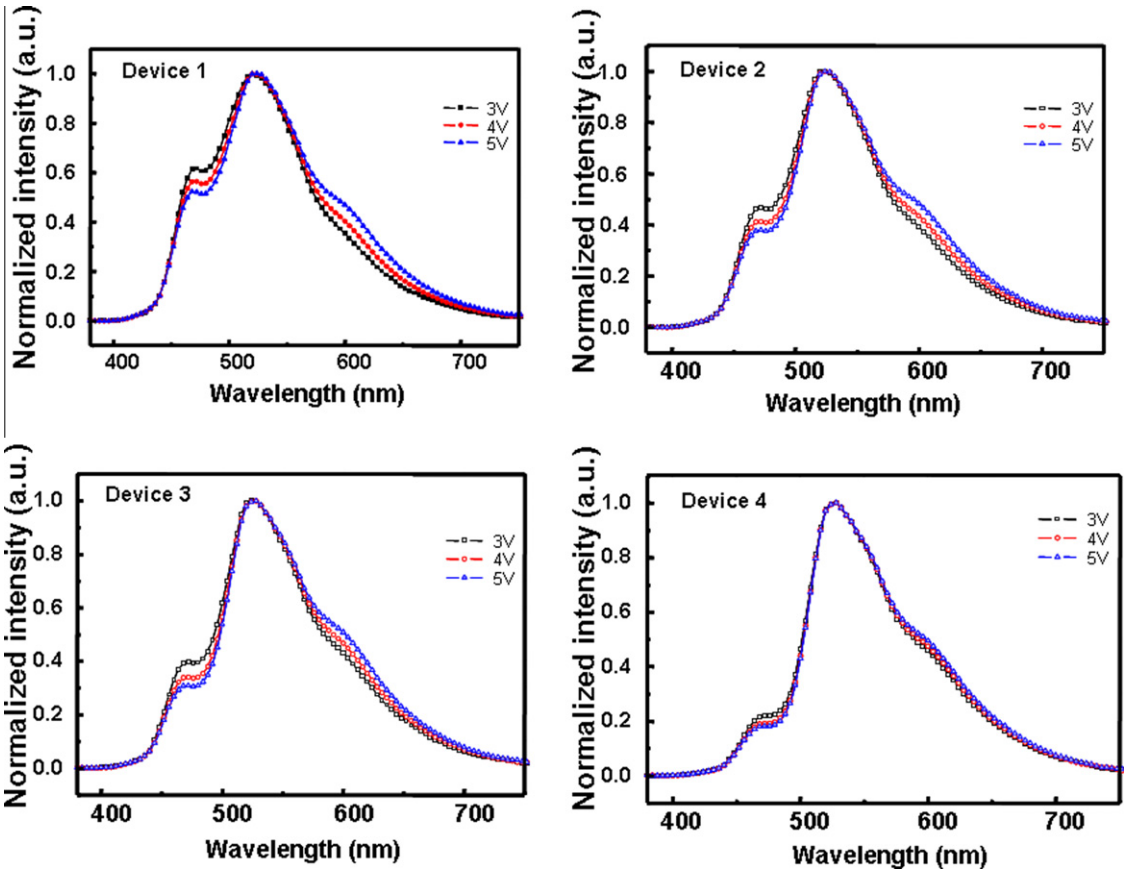

Fig. 4. Normalized EL spectra of Device 1, 2, 3, and 4 at different operational voltages.

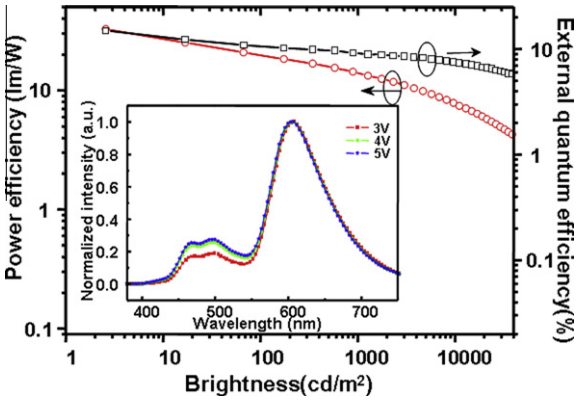

Fig. 5. Power and external quantum efficiencies as a function of brightness for Device 5. The normalized EL spectra at various voltages is shown in the inset.

Actually, the sequence of red and green phosphorescent EMLs in the hybrid Wields is also very important for obtaining white emission with high efficiency. For the fabricated device of ITO/MoO_3(8 nm)/TCTA(60 nm)/TCTA:$Bepp_2$(50:50): $Ir(ppy)_2(acac)$(6%, 5 nm)/TCTA:$Bepp_2$(50:50): $Ir(MDQ)_2(acac)$ (6%, 5 nm)/TCTA:$Bepp_2$ (100:15, 3 nm)/$Bepp_2$:DSA-ph(2.5%, 10 nm)/$Bepp_2$(30 nm)/Life(1 nm)/Al (Device 5), where the only difference from the proposed hybrid WOLED is the position exchange of red and green phosphorescent EML, Fig. 5 shows the EL efficiency as a function of brightness and EL spectra at different operational voltages. It can be seen that not only the efficiency is reduced and the efficiency roll-off becomes severe, dropping from the EQE of 10.6% and power efficiency of 19.7 lm/W at 100 cd/m^2 to 9.1% and 14.0 lm/W at 1000 cd/m^2, but also the emissive spectrum is far from white emission. Although three emissive peaks of red, green and blue can be recognized, and the spectra still shows good stability after 4 V bias, the red emissive intensity is obviously much stronger than that of blue and green, leading to the emission far from white light.

The direct exciton recombination in red and green EML and the energy transfer between different EMLs also exist in Device 5, and likewise, it has a wide charge carrier distribution and extended exciton recombination region. The only difference in EL processes of Device 5 from that of the proposed hybrid WOLED is that, besides the energy transfer from green EML to red EML, the excitons in blue and green EML are directly transferred into the red EML, resulting in more red emission in Device 5. As shown in Fig. 5, the wide exciton recombination indeed stabilizes the EL spectrum. However, the formation of more excitons in the red EML in Device 5 also reduces the EL efficiency

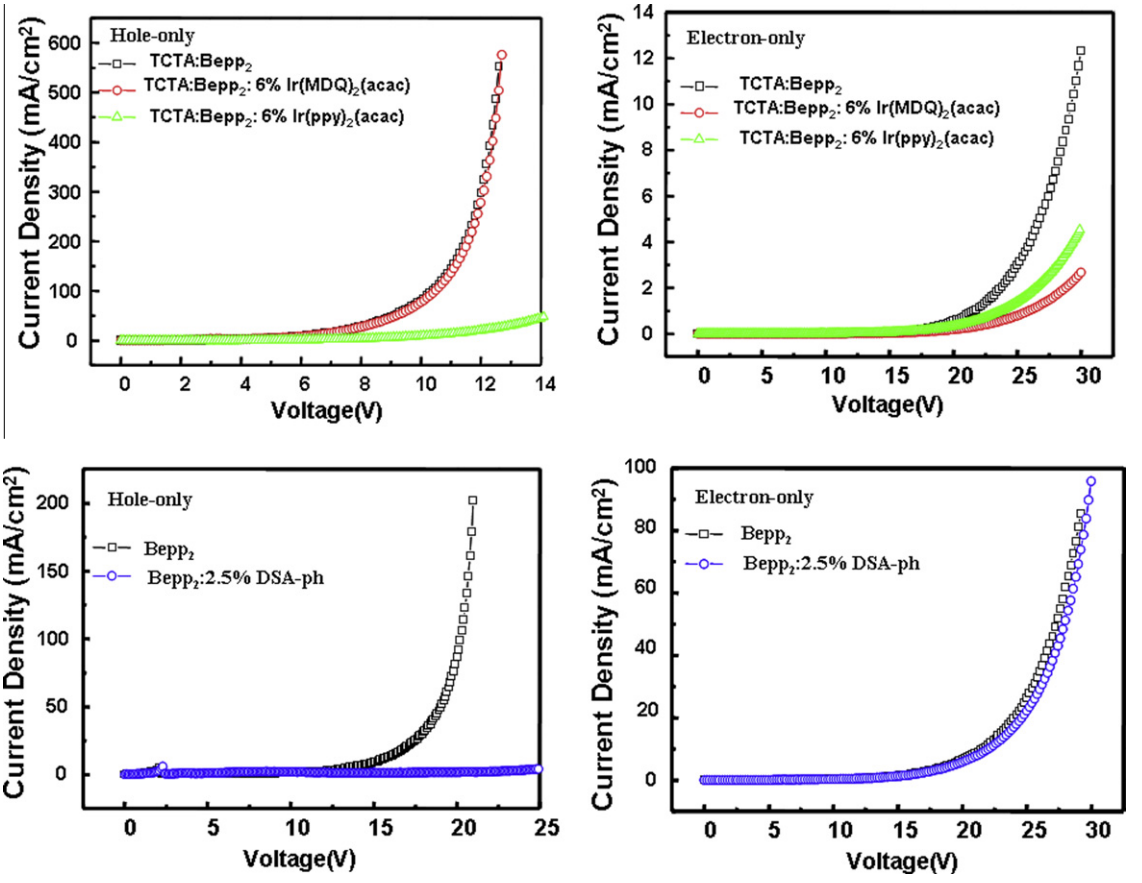

Fig. 6. *J–V* characteristics of the hole-only and electron-only devices.

and causes the efficiency roll-off at high brightness due to exciton quenching.

All above means that the bipolar host in red and green phosphorescent EML and bipolar spacer in proposed hybrid WOLED serve as a charge carrier switch by regulating charge carrier distribution and then regulating exciton recombination, which are directly related to efficiency and spectral stability.

To demonstrate the processes of exciton recombination in red, green and blue EMLs in the proposed hybrid WOLED, hole-only and electron-only devices with the structures of ITO/MoO_3(10 nm)/NPB(40 nm)/X(40 nm)/NPB(40 nm)/MoO_3(10 nm)/Al and ITO/LiF(1 nm)/TPBi(40 nm)/X(40 nm)/TPBi(40 nm)/LiF(1 nm)/Al, respectively, were prepared. Here X are TCTA:$Bepp_2$(1:1), TCTA:$Bepp_2$(1:1):$Ir(MDQ)_2$(acac)(6%), TCTA:$Bepp_2$(1:1):$Ir(ppy)_2$(acac)(6%), $Bepp_2$ and $Bepp_2$:DSA-ph(2.5%). The current–voltage properties of these devices are shown in Fig. 6.

As shown, electrons are captured by the $Ir(MDQ)_2$(acac) molecules under the applied electric field in red EML and then recombine with the directly injected holes, leading to the red emission, while the green emission from green EML is due to the recombination of captured electrons and trapped holes on $Ir(ppy)_2$(acac) molecules. For the blue emission from blue EML, the holes are firstly trapped by the DSA-ph molecules and then recombine with directly injected electrons on the DSA-ph molecules. These processes can also be well understood from the energy diagram shown in Fig. 1.

4. Conclusions

We have demonstrated a highly efficient fluorescent/phosphorescent hybrid WOLED based on EMLs consisting of red phosphorescent EML/green phosphorescent EML/spacer/blue fluorescent EML. Mixture of a hole transporting material TCTA and an electron transporting material $Bepp_2$ is used as the host of phosphorescent EMLs as well as the spacer. As a result, the width of the exciton recombination region is greatly extended. The designed hybrid WOLEDs not only shows high efficiency and low efficiency roll-off at high brightness, but also exhibits excellent spectral stability in a wide range of voltage. The current efficiency and power efficiency reach 30.8 cd/A and 27.1 lm/W at luminance of 1000 cd/m^2. The Commission Internationale de l'Eclairage (CIE) coordinates keep (0.43, 0.43) and the color rendering index (CRI) is about 89 in the whole range of luminance. We have attributed the high performance to the reasonable control of the distribution of charge carriers and the efficient utilization of the formed excitons in EMLs by the bipolar host and spacer.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.orgel.2012.03.005.

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