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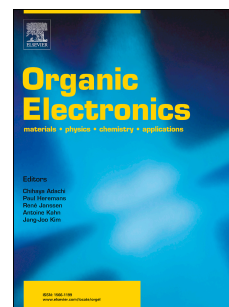
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Optimized trade-off between electroluminescent stability and efficiency in solution-processed WOLEDs adopting functional iridium(III) complexes with 9-phenyl-9-phosphafluorene oxide (PhFlPO) moiety

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Abstract

Employing complimentary color phosphorescent emitters of the well-known **FIrpic** and new unsymmetrical orange emitter **Ir-POB** with 9-phenyl-9-phosphafluorene oxide (PhFIPO) moiety, solution-processed white organic lighting-emitting diodes (WOLEDs) had been fabricated to cope with the variation of EL spectra at different working voltage through excitation of both **FIrpic** and **Ir-POB** with the same mechanism in the device. Through constructing a thin exciton formation zone by the energy level layout between host 4,4',4''-tris(carbazol-9-yl)triphenylamine (**TCTA**) and 1,3,5-tris-(*N*-(phenyl)-benzimidazole)-benzene (**TPBi**), both **FIrpic** and **Ir-POB** in the single emission layer can be excited by energy-transfer mechanism to bring stable white EL spectra in wide range of driving voltages with small CIE coordinate variation of $\Delta x = 0.0044$ and $\Delta y = 0.0197$. Furthermore, in order to overcome the disadvantage associated with energy-transfer mechanism, both **FIrpic** and **Ir-POB** have been excited by direct charge-trapping mechanism in WOLEDs by judiciously controlling the energy level layout between phosphorescent emitters and co-hosts polyvinyl carbazole (**PVK**) and 1,3-bis(5-(4-(*tert*-butyl)phenyl)-1,3,4-oxadiazol-2-yl)benzene (**OXD-7**). The concerned WOLEDs not only maintain stable white EL in even wider driving voltage rang with CIE coordinate variation of $\Delta x = 0.0078$ and $\Delta y = 0.0053$, but also furnish much higher EL efficiencies of maximum external quantum efficiency (η_{ext}) of 22.4%, a maximum current efficiency (η_L) of 58.4 cd A⁻¹ and a maximum power efficiency (η_P) of 41.3 lm W⁻¹. Hence, by unifying excitation mechanism of the phosphorescent emitters, optimized trade-off between white EL stability and high EL efficiencies have been successfully achieved. Definitely, these results can provide crucial information for developing WOLEDs with high performances.

Keywords: Phosphafluorene oxide; Unifying excitation mechanism; Functional phosphorescent

emitters; Stable white EL; High efficiency; White organic lighting-emitting diodes (WOLEDs)

1. Introduction

Organic light-emitting diodes (OLEDs) can convert electricity into light named electroluminescence (EL) by organic luminescent molecules[1]. In addition, the EL color or wavelength of the OLEDs is generally up to the band-gap of the involved organic luminescent molecules[2]. Hence, through changing the chemical structures of the organic emitters, the OLEDs can show various EL colors[3, 4]. Through employing three emitters of primary colors (Red, Green and Blue, RGB)[5, 6] or two complimentary color (Yellow and Blue)[7, 8] emitters, the OLEDs can even emit white EL, which is very suitable for lighting purpose, since the white EL from OLEDs can exhibit very broad spectral line-shape. This is very desirable feature of white OLEDs (WOLEDs), because the broad spectral line-shape of white EL should make it similar to the natural sun light, representing the most favorable lighting light.

To construct WOLEDs, primary- or complimentary-color emitters are typically employed to construct single or several individual emitting layers in one device to bring broad white EL spectra[9-11]. Obviously, WOLEDs with single emission layer are preferable due to their simple structures to make fabrication much easier. Unfortunately, WOLEDs with single emission layer generally exhibit a critical drawback of variation in the EL spectra at different working voltages[12-15]. For sure, this disadvantage will discourage the practical application of WOLEDs as new lighting sources. It had been shown that cascade energy transfer between different emitters or different excitation modes of the emitters have induced the variation of EL spectra at different working voltages[16, 17]. In order to cope with this problem, the emitters employed are set into different zones to fabricate WOLEDs with multi emission layers[18, 19]. However, this strategy

should definitely make fabrication of WOLEDs as a very complicated process. Hence, it is very necessary to construct WOLEDs with single emission layer at the same time maintaining stable white EL spectra.

As new lighting sources, one of the priorities for WOLEDs is energy-saving, since many traditional lighting sources cannot convert electricity into light efficiently and cause terrible waste of energy. Conventional organic fluorescent emitters cannot fulfill this purpose well due to the fact that they can just harness *ca.* 25% of all the electrically generated excitons[20]. Importantly, phosphorescent emitters, especially 2-phenylpyridine-type (ppy-type) Ir(III) complexes, can harvest both singlet and triplet excitons in the EL process to furnish 100% internal quantum efficiency in the OLEDs[21-23]. Accordingly, the EL performances of the phosphorescent ppy-type Ir(III) complexes are very crucial for furnishing highly efficient WOLEDs[24-28]. Based on these advanced phosphorescent emitters, many WOLEDs had been constructed to show very high EL efficiencies[28-33]. Even for the polymer-based WOLEDs with simple structures, EL efficiencies as high as of 12.34 cd A⁻¹, 9.59 lm W⁻¹ had been achieved[34], showing great potential of these phosphorescent ppy-type Ir(III) complexes in WOLEDs. Recently, new functional phosphorescent ppy-type Ir(III) complexes with unsymmetrical structures can show very high EL efficiencies in monochromic OLEDs[35-37]. Reasonably, introducing these novel phosphorescent emitters into WOLEDs should furnish impressive EL performances.

So, in this research, together with well-known sky-blue phosphorescent emitter **FIrpic**, one unsymmetrical phosphorescent ppy-type Ir(III) complex bearing a 9-phenyl-9-phosphafluorene oxide (PhFIPO) moiety has been employed to construct solution-processed complimentary color WOLEDs with single emission layer. Through controlling excitation mode of the complimentary color

phosphorescent emitters, the solution-processed WOLEDs with single emission layer can show very stable white EL spectra at the same time maintaining high EL efficiencies. The concerned results will definitely provide crucial information for optimizing the EL performances of WOLEDs.

2. Experimental

Chemical structures of the key materials employed for making WOLEDs are shown in Fig. 1. Well-known phosphorescent emitter **FIrpic** is commercial available and the orange phosphorescent emitter **Ir-POB** with unsymmetrical structure had been prepared by our reported strategy[37]. It had been shown that **Ir-POB** can exhibit high EL efficiencies in monochromic OLEDs[37]. Accordingly, it should possess the potential to bring attractive EL performances in WOLEDs as well. In addition, its orange emission can complement with the sky-blue emission from **FIrpic** to bring white emission in the devices. Poly (3, 4-ethylenedioxythiophene):polystyrene sulfonate acid (PEDOT:PSS) had been adopted to deposit hole injection layer (HIL). 1,3,5-tris-(*N*-(phenyl)-benzimidazole)-benzene (**TPBi**) layer serves purpose of both hole-blocking and electron-transporting. Host materials involved in the WOLEDs are 4,4',4''-tris(carbazol-9-yl)triphenylamine (**TCTA**), 1,3-bis(5-(4-(*tert*-butyl)phenyl)-1,3,4-oxadiazol-2-yl)benzene (**OXD-7**) and polyvinyl carbazole (**PVK**). The chemical structures of the materials aforementioned are shown in Fig. 1.

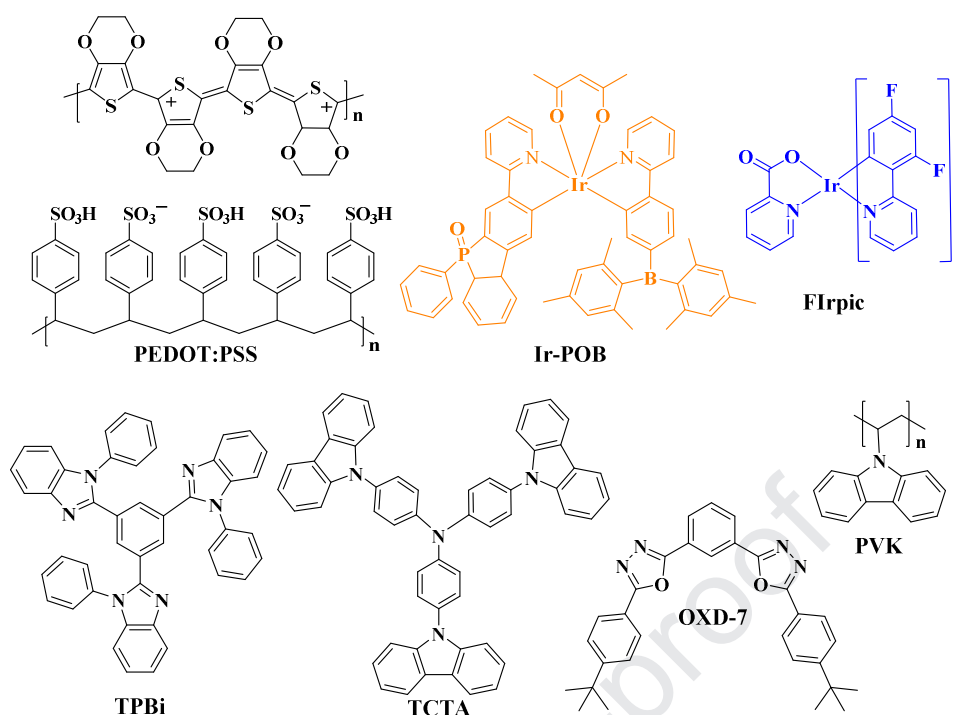


Fig. 1. Chemical structures for the materials used to fabricate WOLEDs.

3. Results and discussion

3.1. Achieving stable white EL through excitation of phosphorescent emitters by energy-transfer

In the electrical excitation process of WOLEDs, the involved emitters have great chance to be excited by different mechanisms, typically energy-transfer and direct charge-trapping, due to the layout pattern of the energy-levels between host materials and phosphorescent emitters[17]. It had been shown that different excitation mechanisms involved in WOLEDs generally induce the variation in white EL spectra at different driving voltages[16, 38]. Adopting structure **A** shown in Fig. 2, three solution processed WOLEDs (Devices **W1**, **W2** and **W3**) had been fabricated with different ratio between complimentary color phosphorescent emitters **FIrpic** and **Ir-POB**.

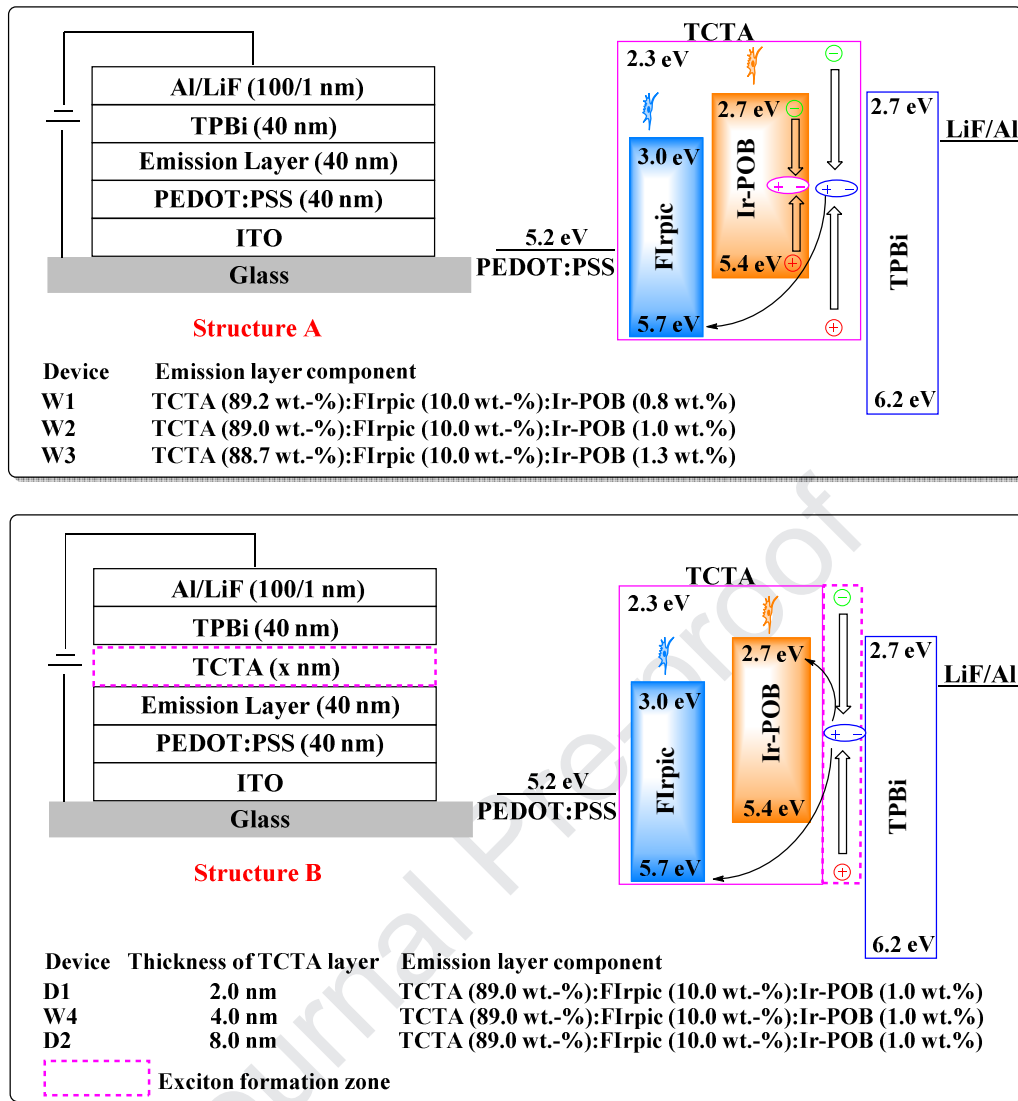
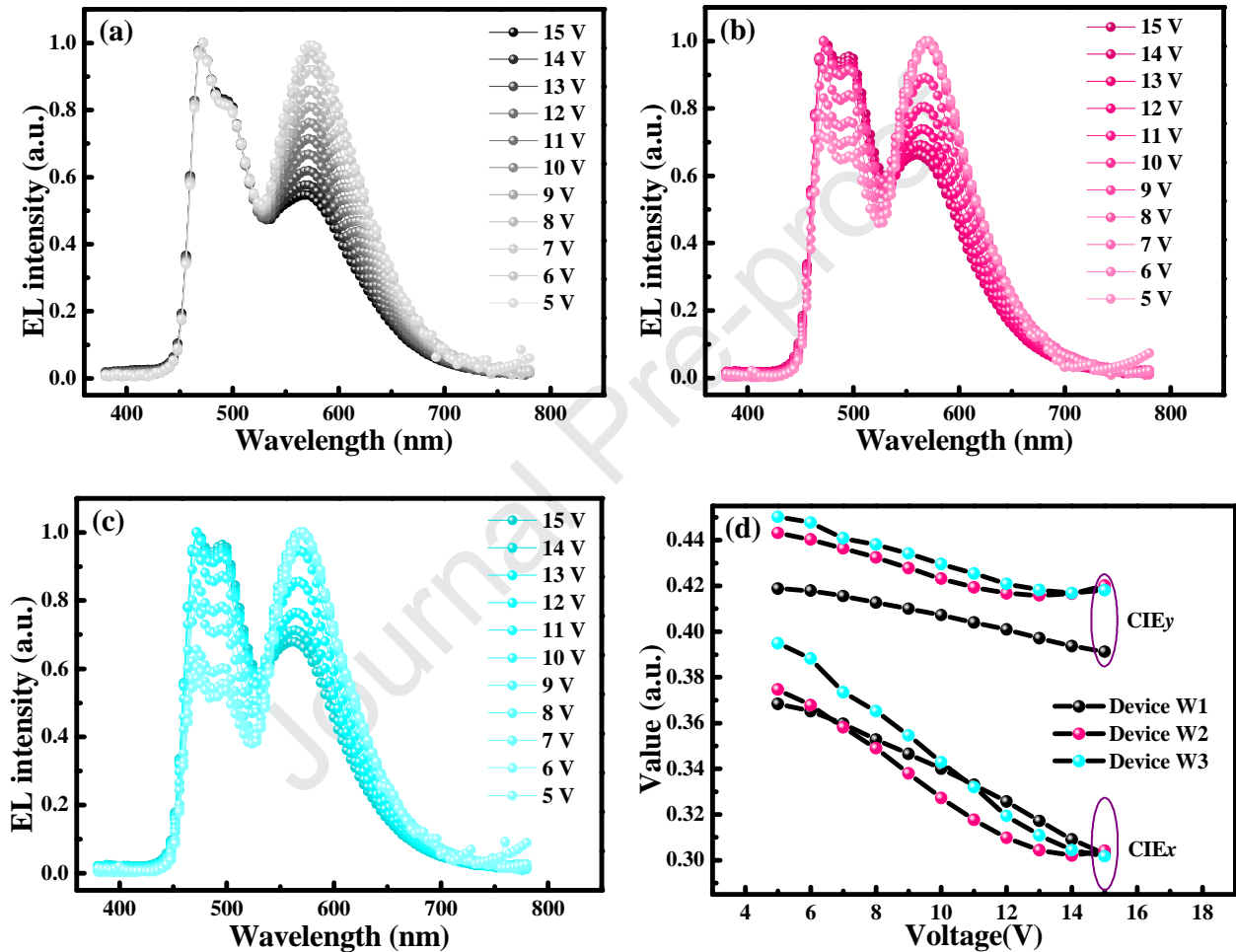


Fig. 2. Structures and energy-level layout for WOLEDs.

Despite the decent EL performances (Table 1, Fig. S1 and S2), all the WOLEDs exhibit variable white EL spectra under different driving voltages (Fig. 3). With increasing driving voltage, the orange component is weakened in the white EL spectra, while blue component is enhanced. The variation of the white EL spectra can be clearly indicated by the CIE coordinates (Fig. 3d). For device **W1**, CIE coordinate variation with $\Delta x = 0.0655$ and $\Delta y = 0.0276$ can be observed. Unfortunately, even larger CIE coordinate variation is undesirably detected with $\Delta x = 0.0627$ and $\Delta y = 0.0274$ for device **W2** and $\Delta x = 0.0932$ and $\Delta y = 0.0333$ for device **W3**.

Table 1 Maximum EL performances of devices **W1**, **W2** and **W3**.

Device	W1	W2	W3
$V_{\text{turn-on}}$ (V)	4.5	4.4	4.1
Luminance L (cd m ⁻²)	28060	27431	23524
η_{ext} (%)	13.1%	7.7%	8.9%
η_L (cd A ⁻¹)	33.5	21.9	24.5
η_P (lm W ⁻¹)	20.7	13.6	17.9

**Fig. 3.** EL spectra at different voltages for (a) Device **W1**, (b) Device **W2** and (c) Device **W3** together with (d) corresponding CIE(x,y) coordinates.

In order to find the origin of the variation in the white EL spectra of devices **W1**, **W2** and **W3**, the excitation mechanisms for **FIrpic** and **Ir-POB** should be clarified. To fulfill this purpose, current density (J)-voltage (V) curves for the devices with emission layer composed by different ratio of **FIrpic** to **Ir-POB** have been obtained (Fig. 4). When setting **Ir-POB** content of 0.8 wt.-% and

varying **FIrpic** content of 6.0 wt.-%, 8.0 wt.-% and 10.0 wt.-%, it has been found that the *J-V* curves of the concerned devices remain nearly unchanged (Fig. 4a), indicating the energy-transfer excitation mechanism of **FIrpic**. Differently, the current density can be effectively increased in the devices by setting **FIrpic** content of 10.0 wt.-% and varying **Ir-POB** content of 0.8 wt.-%, 1.0 wt.-% and 1.3 wt.-% (Fig. 4b), indicating the direct charge-trapping excitation mechanism of **Ir-POB**. In addition, **Ir-POB** can possess much higher HOMO level than that of **TCTA** and comparable LUMO level to that of **TPBi**, favoring its direct charge-trapping excitation as well. In addition, the HOMO levels of **Ir-POB** and **PEDOT:PSS** are quite close to form shallow well, which cannot effectively reduce the hole current density at low doping level of **Ir-POB**[39]. In addition, the nearly identical LUMO levels of both **Ir-POB** and **TPBi** can make the electrons from **TPBi** layer flow easily to the LUMO of **Ir-POB**, which can enhance electron current density. Hence, higher current density can be furnished at higher doping level of **Ir-POB** in emission layer (Fig. 4b). Based on the excitation mechanisms for the phosphorescent emitters, the variation of white EL spectra of devices **W1**, **W2** and **W3** with increasing driving voltage can be explained as follows. Owing to the direct charge-trapping excitation associated with **Ir-POB**, the charge-trapping process of **Ir-POB** will definitely compete with charge-transporting process in the device. With increasing driving voltage, internal electric field of the device will be enhanced to favor charge-transporting process and disfavor the charge trapping process of **Ir-POB**. In addition, the enhanced charge-transporting process will promote the formation of excitons on the host to facilitate energy-transfer to **FIrpic**. So, with increasing driving voltage in devices **W1**, **W2** and **W3**, the blue content in the white EL spectra is effectively enhanced and the orange content is relatively weakened, inducing undesired variation of white EL spectra (Fig. 3).

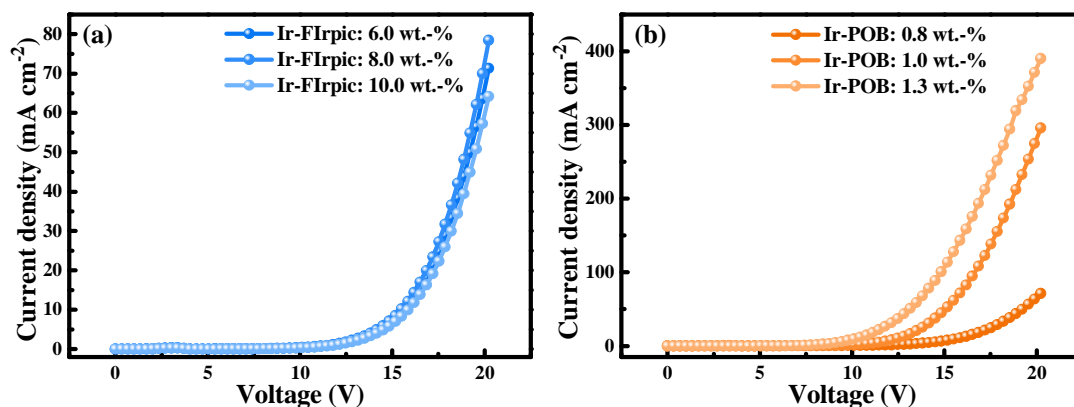
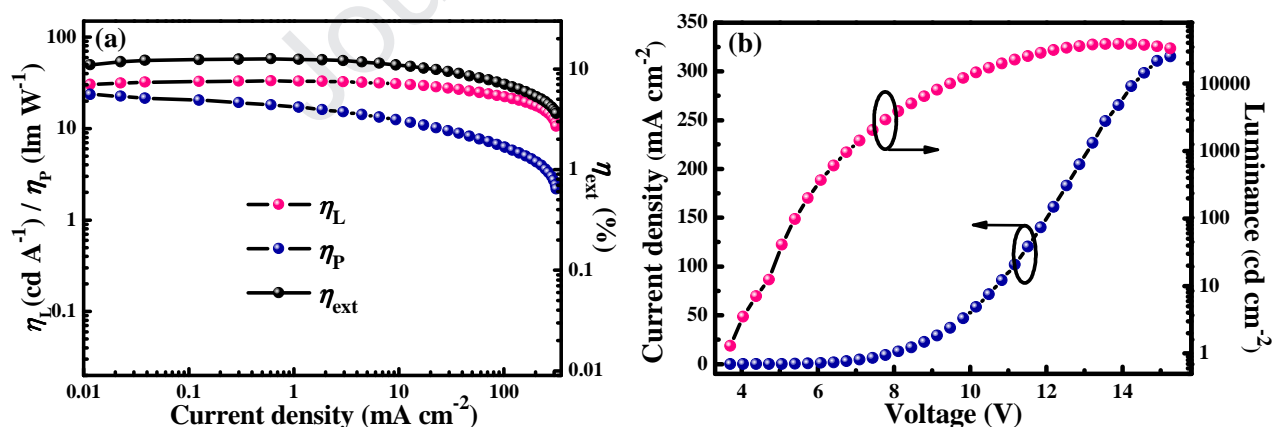


Fig. 4. Current density (J)-voltage (V) curves for the devices with different doping concentration for (a) blue (ITO/PEDOT:PSS (40 nm)/TCTA:(**FIrpic** = x wt.-%)(**Ir-POB** = 0.8wt.-%) (40 nm)/LiF (1 nm)/Al (100 nm)) and (b) orange (ITO/PEDOT:PSS (40 nm)/TCTA:(**FIrpic** = 10wt.-%)(**Ir-POB** = x wt.-%) (40 nm)/LiF (1 nm)/Al (100 nm)) phosphorescent emitters.

Clearly, the variation in the white EL spectra from devices **W1**, **W2** and **W3** is induced by the different excitation mechanisms associated with **FIrpic** and **Ir-POB**, respectively. Hence, exciting both **FIrpic** and **Ir-POB** with the same mechanism should be a feasible way to cope with this problem. Based on the energy-level layout of the devices with structure **A** (Fig. 2), owing to the higher LUMO level of **TCTA** than that of **TPBi**, electrons can be easily confined at the interface between **TCTA** and **TPBi**. At the same time, the much deeper HOMO level of **TPBi** can effectively block the holes transported from **TCTA** at the interface between **TCTA** and **TPBi**. It can be expected that inserting a layer of **TCTA** between doped **TCTA** emission layer and **TPBi** layer can form a recombination zone or exciton formation zone. By this way, both **FIrpic** and **Ir-POB** can be excited by energy-transfer mechanism to relieve the problem of white EL variation aforementioned. So, the WOLEDs with **TCTA** layer possessing different thickness have been made by adopting structure **B** (Fig. 2) and the EL results have been summarized in Table S1. Among them, device **W4** in which a thin **TCTA** layer with optimized thickness of *ca.* 4 nm acts as exciton formation zone can show the best EL performances (Table S1). The excitons formed in this zone transfer energy to both **FIrpic** and **Ir-POB** (Fig. 2). From the EL results in Fig. 5, device **W4** can show comparable EL efficiencies

to devices **W1**, **W2** and **W3** (Fig. 5a and b) with maximum luminance (L_{\max}) of 38844 cd cm^{-2} , maximum external quantum efficiency (η_{ext}) of 12.6%, maximum current efficiency (η_L) of 33.2 cd A^{-1} and a maximum power efficiency (η_P) of 24.4 lm W^{-1} . Importantly, device **W4** can maintain stable white EL spectra in wide driving voltage range from 5 to 11 V (Fig. 5c), indicated by much smaller CIE coordinate variation with $\Delta x = 0.0044$ and $\Delta y = 0.0197$ than that of **W1**, **W2** and **W3** (Fig. 3d). The weak EL band at *ca.* 420 nm should be assigned to **TCTA** (Fig. 5c), since the recombination zone locates in the thin **TCTA** layer and the formed excitons cannot be consumed by the phosphorescent emitters. The existence of weak EL band from **TCTA** makes EL spectra of **W4** different from those of **W1**, **W2** and **W3**. Obviously, difference in the white EL spectra is due to the different excitation mechanism in **W4** from those in **W1**, **W2** and **W3**. However, with increasing driving voltage $> 11 \text{ V}$, obvious variation in white EL spectra can be detected (Fig. 5c and d), due to the drifting of recombination zone into the doped **TCTA** layer. So, more advanced strategy should be developed to cope with the problem of white EL variation.



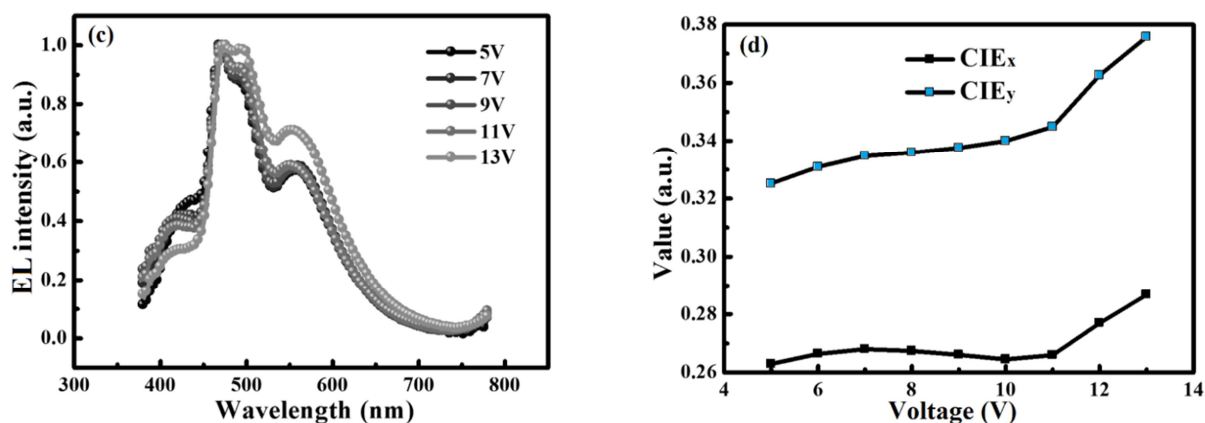


Fig. 5. EL performances of Device **W4**. (a) EL efficiencies vs. current density. (b) Current density (J)-voltage (V)-Luminance (L) curves. (c) EL spectra at different voltages. (d) CIE(x,y) coordinates at different voltages.

3.2. Achieving stable white EL through charge carrier-trapping mechanism for phosphorescent emitters

Generally, white EL spectra can be obtained through exciting phosphorescent emitters by energy-transfer mechanism in complimentary color WOLEDs. However, the energy-transfer excitation mechanism will definitely induce energy loss, disfavoring EL efficiencies. In addition, drifting of recombination zone should be addressed as well. Hence, new strategy should be developed to maintain both white EL stability and high EL efficiencies. In this section, taking advantage of energy-level layout of **FIrpic** and **Ir-POB**, this purpose has been successfully fulfilled by excitation of the phosphorescent emitters through direct charge-trapping mechanism in solution-processed WOLEDs. The prepared WOLEDs adopt structure **C** in Fig. 6. Both **PVK** with high LUMO level and **OXD-7** with deep HOMO level have been employed as co-host materials. Clearly, this strategy makes the energy-levels of both **FIrpic** and **Ir-POB** lie in between those of co-host materials at the same time furnishing ambipolar features.

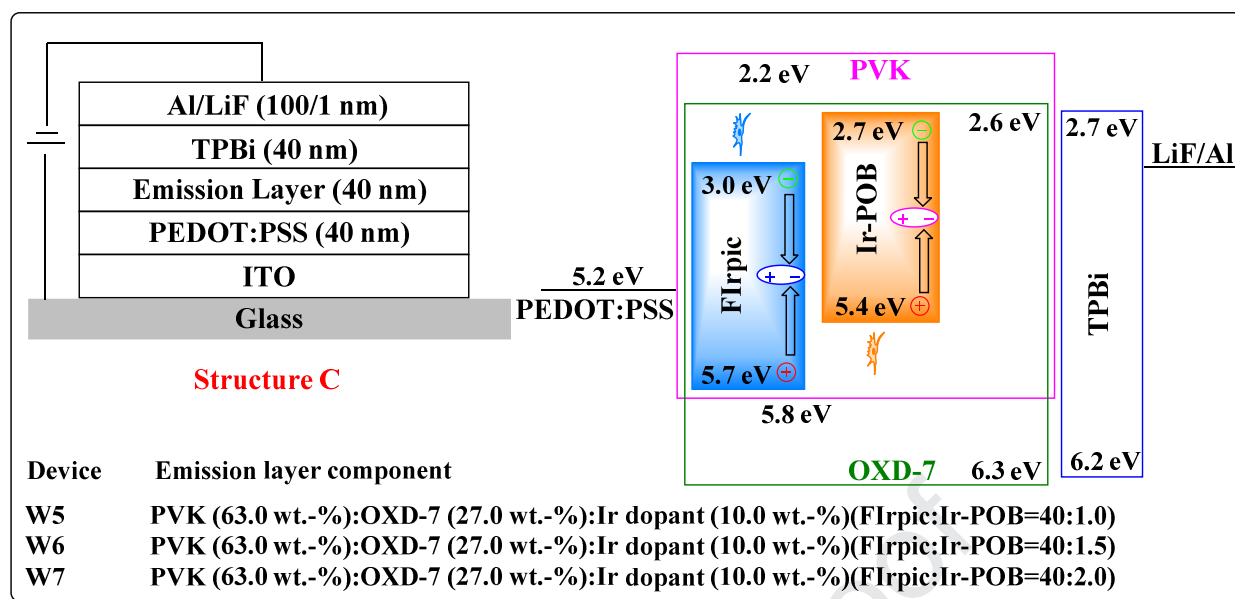


Fig. 6. Structure energy-level layout for the WOLEDs with phosphorescent emitters excited by charge carrier trapping.

The energy-level pattern between hosts and phosphorescent guests will favor direct charge-trapping excitation mechanism for both **FIrpic** and **Ir-POB**. In order to confirm both **FIrpic** and **Ir-POB** are dominantly excited by direct charge-trapping mechanism, PL and EL spectra have been compared (Fig. S3). The EL spectra of device **W6** taking as an example are quite different from the PL spectrum of the film with the same content to that of the emission layer of device **W6**. For the PL process, there should be just energy-transfer process without direct charge-trapping. Obviously, if there were obvious energy-transfer in the EL process, great resemblance between EL and PL spectra would be observed. So, it can be safely concluded that **FIrpic** and **Ir-POB** are mainly excited through direct charge-trapping mechanism. In addition, the obvious dependence of *J-V* curves on the content of **FIrpic** and **Ir-POB** in the emission layer have clearly indicated their charge-trapping excitation mechanism in the WOLEDs (Fig. S4).

Furthermore, in order to clarify the excitation behavior of **FIrpic** and **Ir-POB**, device **T1** (ITO/PEDOT:PSS(40 nm)/EML(40 nm)/Al(100 nm), without electron injection layer) and device **T2**

(ITO/EML(40 nm)/TPBi(40 nm)/LiF(1 nm)/Al(100 nm), without hole injection layer) had been fabricated. The component of devices **T1** and **T2** is similar to that of device **W6** with **PVK** (63.0 wt.-%):**OXD-7** (27.0 wt.-%):Ir dopant (10.0 wt.-%)(**FIrpic**:**Ir-POB** = 40:1.5). It have been found that the turn-on voltage for both **T1** and **T2** was *ca.* 10 V. EL spectra for both **T1** and **T2** can show obvious difference (Fig. S5). For device **T1** without electron injection layer, there are mainly holes in the emission layer with driving voltage less than 10 V. When driving voltage is higher than 10 V, electrons are injected into the emission layer of **T1**. At this time, holes and electrons can combine to form excitons and hence induce EL to turn on the device. Form the EL spectrum of device **T1** (Fig. S5), it can be clearly seen that the blue EL band from **FIrpic** is much higher than that from **Ir-POB**. It indicates that **FIrpic** should possess much higher ability to trap electrons than **Ir-POB**. Based on the similar analysis and the EL pattern of device **T2**, it indicates that **Ir-POB** should possess much higher ability to trap holes than **FIrpic**. In addition, the *J-V* curves for the hole-only devices with different doping levels of **FIrpic** for hole-only devices (Fig. S6a) and **Ir-POB** for electron-only devices (Fig. S6b) have been obtained. Generally, the *J-V* curves are quite similar under different doping levels of the phosphorescent dopants. This result indicates that **FIrpic** and **Ir-POB** cannot act as effective hole and electron trap, respectively. Together with the energy-levels of **FIrpic** and **Ir-POB**, it can be concluded that electrons can be easily injected into **FIrpic**, serving as electron trap, while **Ir-POB** can capture injected holes and serve as hole trap (Fig. 6). Importantly, owing to the fact that **FIrpic** and **Ir-POB** act as trap for electron and hole, respectively, there is no obvious competition in their excitation process, guaranteeing relatively stable content of blue and organic light in the final white EL spectra. It should be noted that the mixed ambipolar host can guarantee both holes and electrons distribute more evenly in the emission layer of the WOLEDs. It will reduce

the competition between **Flrpic** and **Ir-POB** in the excitation process as well. Definitely, it will benefit the stable white EL as well.

Encouragingly, the EL performances of WOLEDs **W5**, **W6** and **W7** have realized this idea (Fig. 7, 8 and S7). All the WOLEDs can show very stable white EL spectra in wide range of driving voltage (Fig. 7). From 6 to 13 V, CIE coordinate variation for **W5** is $\Delta x = 0.010$ and $\Delta y = 0.015$, while $\Delta x = 0.0075$ and $\Delta y = 0.0078$ for **W6** together with $\Delta x = 0.0078$ and $\Delta y = 0.0053$ for **W7** (Fig. 7d). In addition, device **W7** can exhibit the highest EL efficiencies of 22.4%, 58.4 cd A⁻¹ and 41.3 lm W⁻¹ among the three WOLEDs.

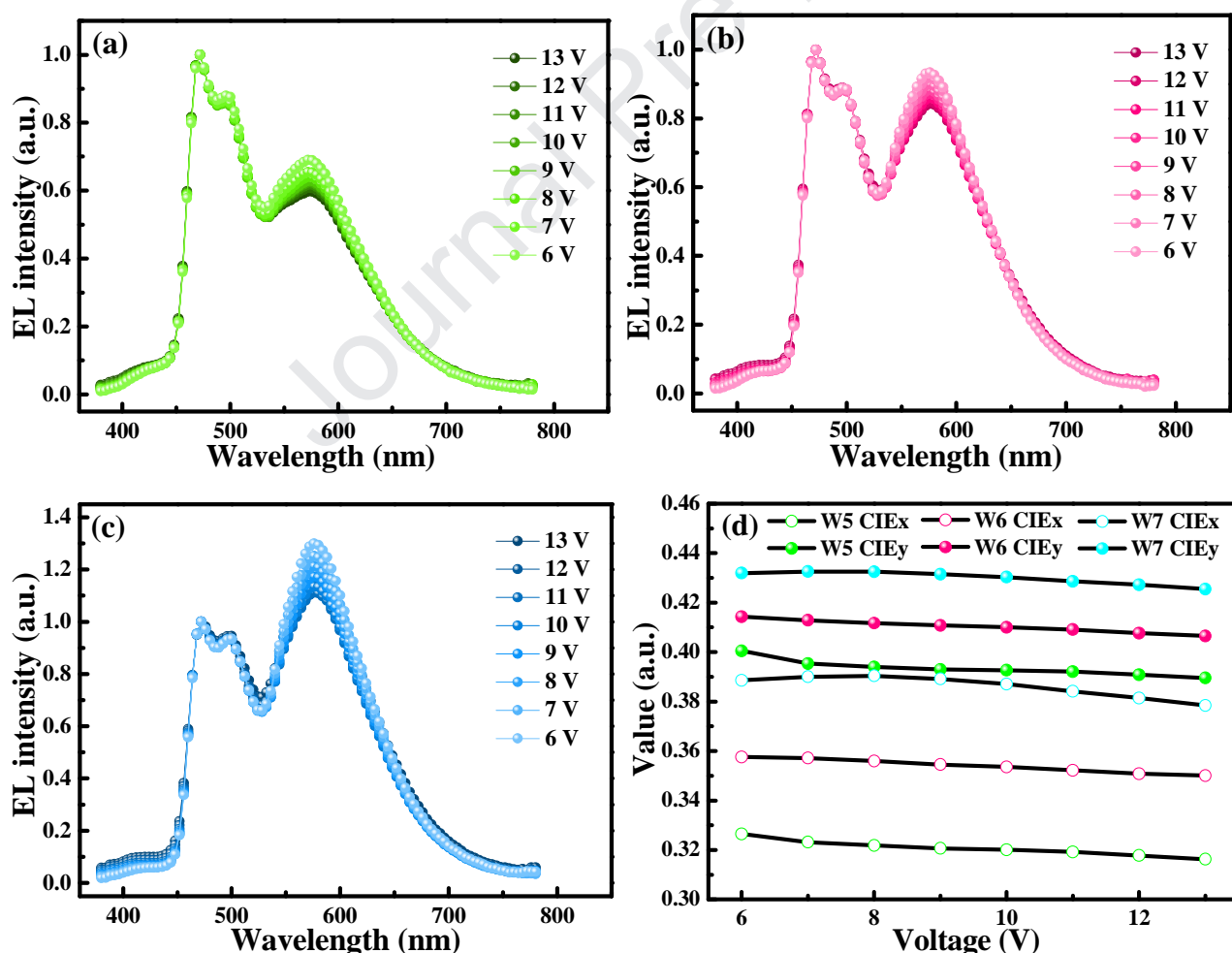
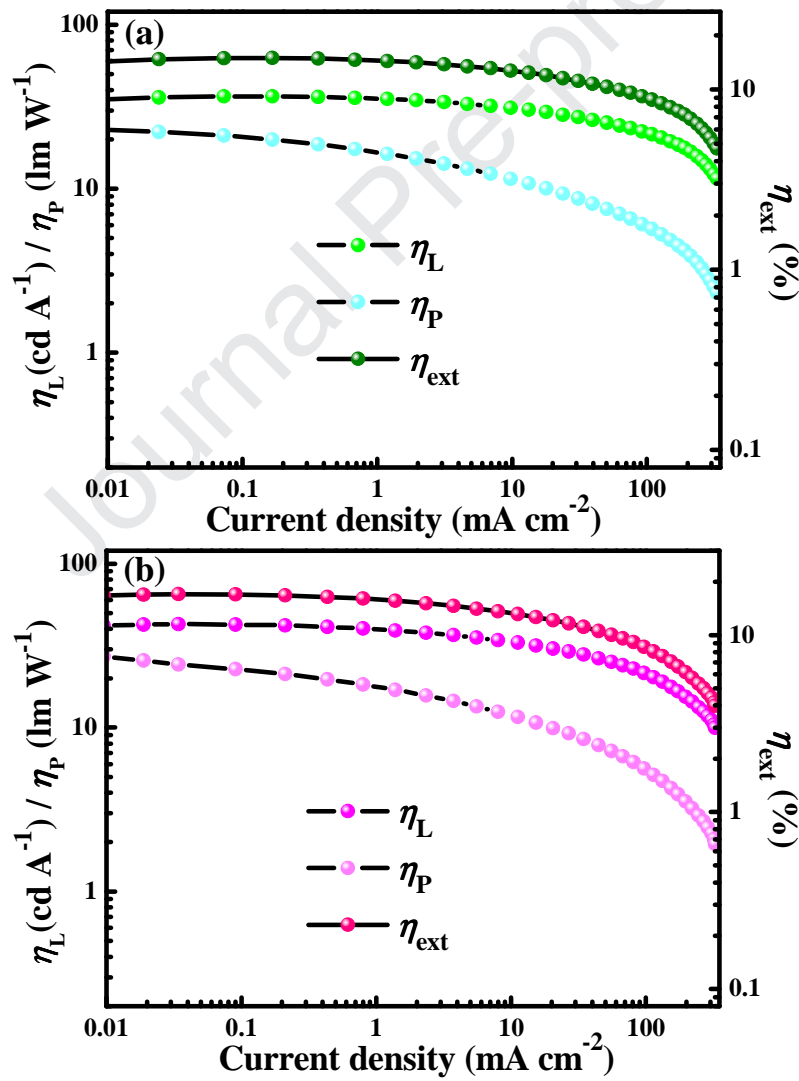


Fig. 7. EL spectra at different voltages for (a) Device **W5**, (b) Device **W6** and (c) Device **W7** together with (d) corresponding CIE(x,y) coordinates.

Table 2 Maximum EL performances of devices **W5**, **W6** and **W7**.

Device	W5	W6	W7
$V_{\text{turn-on}}$ (V)	4.4	4.2	4.1
Luminance L (cd m ⁻²)	38788	33903	37008
η_{ext} (%)	14.9%	17.1%	22.4%
η_L (cd A ⁻¹)	36.5	42.7	58.4
η_P (lm W ⁻¹)	23.1	27.9	41.3



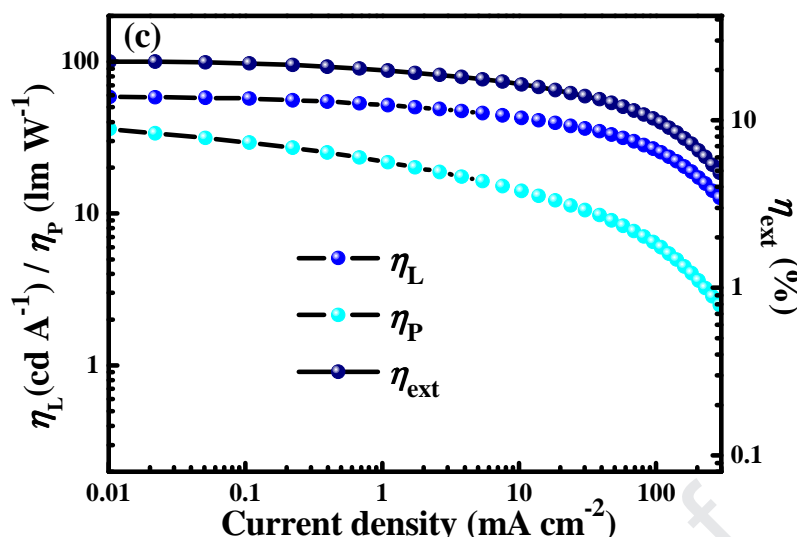


Fig. 8. EL efficiencies vs. current density curves for (a) Device W5, (b) Device W6 and (c) Device W7.

In brief, both of the two strategies can fulfill stable white EL spectra in wide driving voltage range. Importantly, exciting both the phosphorescent emitters *via* charge carrier-trapping mechanism can achieve optimized trade-off between EL efficiency and EL spectral stability in WOLEDs. Clearly, good match of the energy-level layout of **Ir-POB** with that of other functional materials in the WOLEDs has played a critical role in fulfilling this purpose. Obviously, the two strategies have been realized by control the energy-level layout of the materials in the WOLEDs. With the emitters possessing energy-level layout similar to that of **FIrpic** and **Ir-POB**, WOLEDs with stable white EL spectra can be obtained. Hence, the strategies employed in this research should be universal.

4. Conclusion

Two strategies have been developed in solution-processed complimentary color WOLEDs to furnish stable white EL in wide range of driving voltage through controlling energy-level layout of the involved materials. It has been found that unifying excitation mechanism of the phosphorescent emitters with either energy-transfer or direct charge-trapping, is very crucial for fulfilling stable

white EL spectra. The energy-transfer excitation has been realized by constructing an exciton formation zone in the WOLEDs, while laying the energy-levels of phosphorescent emitters between those of co-host materials can fulfill direct charge-trapping excitation. By these advanced strategy, stable white EL spectra with CIE coordinate variation of $\Delta x = 0.0078$ and $\Delta y = 0.0053$ have been achieved at the same time maintaining high EL efficiencies of 22.4%, 58.4 cd A⁻¹ and 41.3 lm W⁻¹. This research can be an effective outlet to obtain high performance WOLEDs.

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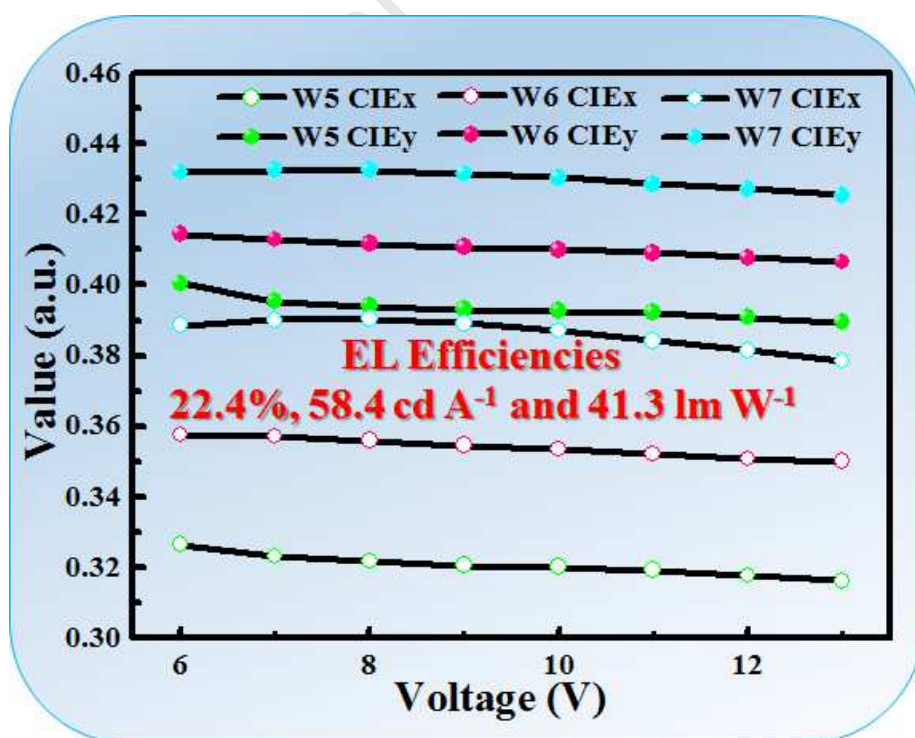
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Highlights

1. Stable white EL has been achieved by unifying excitation mechanism of the phosphorescent emitters.
2. Functional orange phosphorescent emitter with 9-phenyl-9-phosphafluorene oxide (PhFIPO) moiety has been employed for WOLEDs
3. Optimized trade-off between stable white EL spectra and efficiencies has been fulfilled in solution-process WOLEDs.

Graphical Abstract



Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: