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Demonstration of highly efficient orange EL device and warm white OLED

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ABSTRACT

Exciplex is a well-known charge transfer excited-state complex formed between electron-donor and electron-acceptor molecules. The exciplex studied here is resulted from between the donor mCP and the acceptor PO-T2T, which acts as the host of orange emitting OLEDs. The emitter is orange phosphor Ir(pq)2acac. Our device based on the exciplex host and orange phosphor is very simple, but excellent EL performance was observed. That is, peak current efficiency of 35.3 cd/A, power efficiency of 31.0 lm/W and EQE of 18.7% at a 1.5 wt% orange phosphor, respectively. To date this is the highest orange OLED compared with other orange OLEDs with identical color performance. The better results were ascribed to reasonable device design and contribution from both singlet and triplet of exciplex host as well efficient energy transfer from exciplex host to the orange phosphor. The conclusion is based on the fact that exciplex emission comes from TADF process in which there is small $\Delta E_{s\text{-}t}$. This strategy enables to resolve the problem of the lower reverse intersystem crossing (RISC). Besides, warm white OLED was also achieved by lowering the dopant concentration, by which incomplete energy transfer from exciplex host to the orange phosphor could take place.

1. Introduction

Recent years, organic light emitting diodes (OLEDs) have attracted much attention owing to promising application of panel displays and solid-state lighting [1–4]. Thus, white OLED(WOLED) would be required. White OLED could be consisted of either three primary colors (R G B) or two primary colors with orange- and blue sub-emissive colors, which were referred to three or two primary color WOLED respectively [5–8]. The various emitters generally have been fabricated by doping a luminescent dye to host. For two primary color case, the effect of electroluminescent (EL) performance of the orange component on WOLED would be essential. Thus, highly efficient orange diodes have attracted a great deal of attention [9,10]. For a high strong EL orange emissive OLED, host materials are usually necessary in order to prevent triplet-triplet (T-T) annihilation because concentration quenching effect is easily happened for phosphorescence(PH) emitters. Therefore, carefully selecting suitable host should generally be important, i.e., the materials must possess suitable singlet (S_1) and triplet (T_1) state, so as to ensure efficient energy transfer from host to an orange phosphor emitter dopant [11–15]. To realize easily carrier injection from two electrodes and balance recombination of two carriers in the emitting layer (EML), a bipolar host would be required for reducing exciton quenching and injection barrier [10–15]. Recently, it is reported that phosphorescent OLEDs with exciplex as the host have exhibited high EL performance [1–4,9,13]. The exciplex can either emit itself or transfer energy to the dopant. In addition, on account of presence of small singlet-triplet energy difference thermally activated delayed fluorescence (TADF) exciplex, i.e., up-conversion process between singlet/triplet levels could be taken place [10,12,16–20]. The exciplex that derived from hole- and electron-transporting donor (D)/acceptor (A) pair usually presents the bipolar carrier transporting properties at an appropriate D-to-A-ratio, at such a molar ratio exciton recombination region of the exciplex host could be distributed throughout EML range, which can decrease the density of singlet- and triplet-exctions, so that the two kinds of exciton quenching would be lowered [21].

In this work, we demonstrated high effcient orange device in which exciplex and orange phosphor were as host and dopant. In exciplex host is formed between mCP(N,N′-dicarbazolyl-3,5-benzene) and PO-

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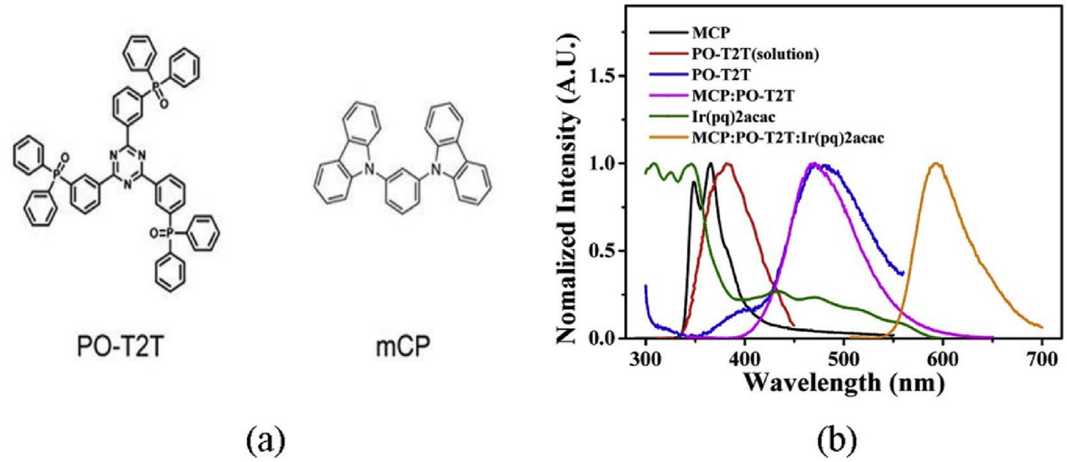


Fig. 1. a) Molecular structure of PO-T2T and mCP, b) Normalized PL spectra of mCP film and PO-T2T film and solution(PO-T2T is an excimer which has two peaks, one is the peak of PO-T2T itself coincides with the peak of PO-T2T solution in the figure, and the other is the peak of excimer), mCP:PO-T2T co-deposited film and mCP:PO-T2T:Ir(pq)2acac co-deposited film, as well the absorption spectrum of Ir(pq)2acac phosphor film green line). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

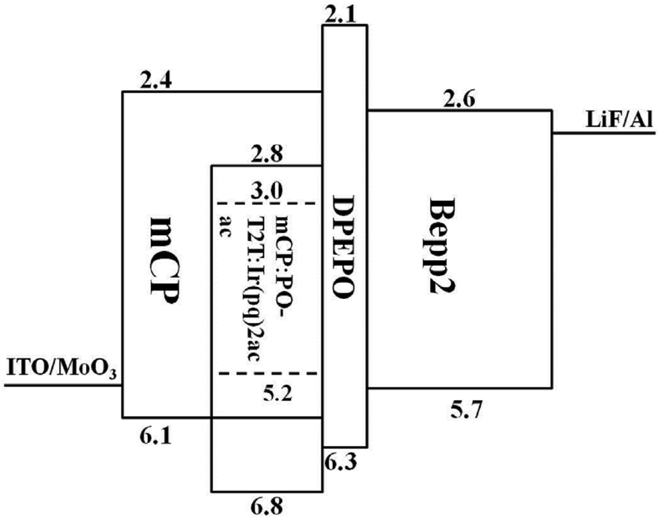


Fig. 2. Schematic device structures in this study, the numbers on LUMO and under HOMO depict the LUMO- and HOMO-levels (eV) of each molecule, respectively.

T2T((1,3,5-triazine-2,4,6-triyl)tris(benzene-3,1-diyl) tris(diphenylphosphine oxide)) which serve respectively as the electron-donor- and acceptor components, as well orange phosphor is (iridium (III)bis(1phenylquinoline)-acetylacetonate Ir(pq)2acac)). By comparing it is observed that the T_1 level is lower for the exciplex (2.64 eV) than for either mCP (2.94 eV) or PO-T2T (2.99eV), and higher than that of phosphor Ir(pq)2acac (2.2 eV) [1,22–25]. Thus, we dope different concentrations of Ir(pq)2acac into the exciplex host in which mCP:PO-T2T ratio also was tuned. As a result, high EL performance based several orange emitting OLEDs were successfully achieved, therein, orange OLED with maximum efficiency was developed, That is, at a concentration of 1.5 wt% maximum current- and power-efficiency are respectively 35.3 cd/A and 31.0 lm/W, as well EQE reaches up to 18.7%. It is also observed that as the quantity of orange phosphor was further lessened in exciplex host, WOLED was successfully constructed, which is attributed to an incomplete energy transfer from the exciplex host to the phosphor dopant and presence of blue emission derived from the exciplex host. For the purpose of hybrid WOLED by overlapping blue exciplex TADF emissive spectrum with orange emitting band of the phosphor, an interlayer would be required, because such an interlayer can prohibit mutual energy transfer and exciton quenching [26,27]. However, if our device structure contains interlayer, the EL efficiency would be lowered. Note that blue phosphor emission has generally a poor stability which restricts its application [5–8]. Thus, in this manuscript we use the blue exciplex with a small singlet-triplet splitting to realize high efficiency OLED by utilizing reverse intersystem crossing mechanism from T_1 to S_1. In addition, broad EL spectrum of the exciplex also facilitates to fabricate WOLED. Optimal WOLED provides a current-efficiency of 15.55cd/A, a power-efficiency of 13.69 lm/W, as well an EQE of 7.49%, as a result, WOLED with highly simple structure was obtained as the phosphor concentration is 0.1 wt% in exciplex host.

1.1. *Experiments methods*

All OLED devices were fabricated on indium tin oxide (ITO) coated glass substrates with a sheet resistance of 10 Ω/sq. The ITO processes were carried out, and finished the deposition of organic layers, Al/LiF compound cathode was deposited in the end with a shadow mask, which defined the device an area of $3 \times 3\,\text{mm}^2$. The photoluminescence (PL) spectra were measured with a FluoroMax-4 fluorescence spectrometer (HORIBA Jobin Yvon). The UV–visible absorption spectrum was recorded with a Hitachi U-3900 scanning spectrophotometer. EL spectra were measured through a PR-655 spectra scan spectrometer with computer controlled. The current-voltage-luminance curves were measured with a Keithley 2400 power supply combined with a ST-900M spot photometer. EQE was calculated from the current density-voltage-luminance curve and spectra data. All the organic materials were procured commercially without further purification. All the measurements were carried out at room temperature and under ambient conditions without any protective encapsulation, The substrates were cleaned firstly with acetone, deionized water, acetone and then treated by ultraviolet-ozone for 10 min, after that the ITO substrates were loaded into a high vacuum chamber and a vacuum reaches up to 5.0×10^{-4} Pa and then deposition.

2. Results and discussion

Fig. 1(a) shows the molecular structures of mCP and PO-T2T which are respectively used as electron-donor and -acceptor material in the exciplex system. PL spectrum of mCP:PO-T2T(molar ratio 1:1) is obviously red-shifted compared with that of pure mCP and PO-T2T ones (see Fig. 1(b). Interestingly, the mCP:PO-T2T co-deposited film shows a broad PL spectrum with a peak of 471 nm (2.64 eV), which is close to the energy difference between the $[\text{HOMO}]_{\text{mCP}}$ and $[\text{LUMO}]_{\text{PO-T2T}}$ level, it is referred to PL blue emission of the exciplex [11]. In addition, it is also noted that the PL spectrum of the exciplex overlaps with the

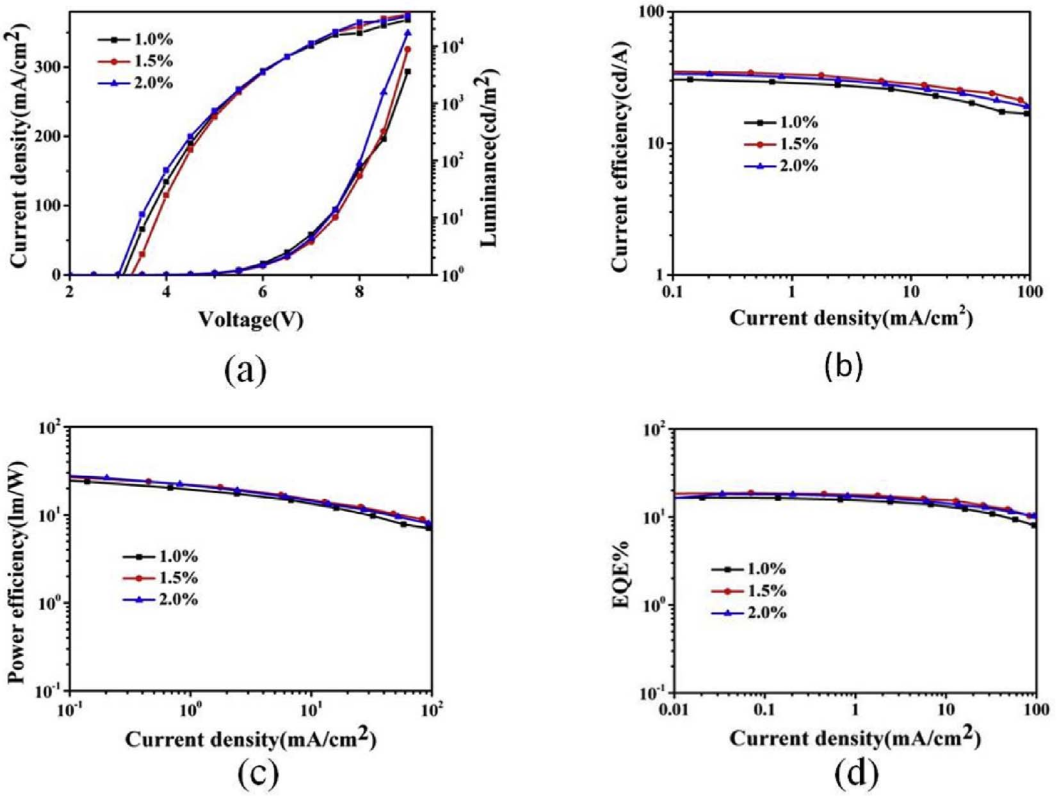


Fig. 3. The EL performance of the OLEDs in this study. (a) Current density-voltage-Luminance, (b) current efficiency-current density, (c) power efficiency-current density (d) EQE-current density (The device is ITO/MoO$_3$(4 nm)/mCP(20 nm)/mCP:PO-T2T:Ir(pq)2acac (xwt% (25 nm))/DPE PO (3 nm)/Bepp2(40 nm)/LiF(1.5 nm)/Al(100 nm).

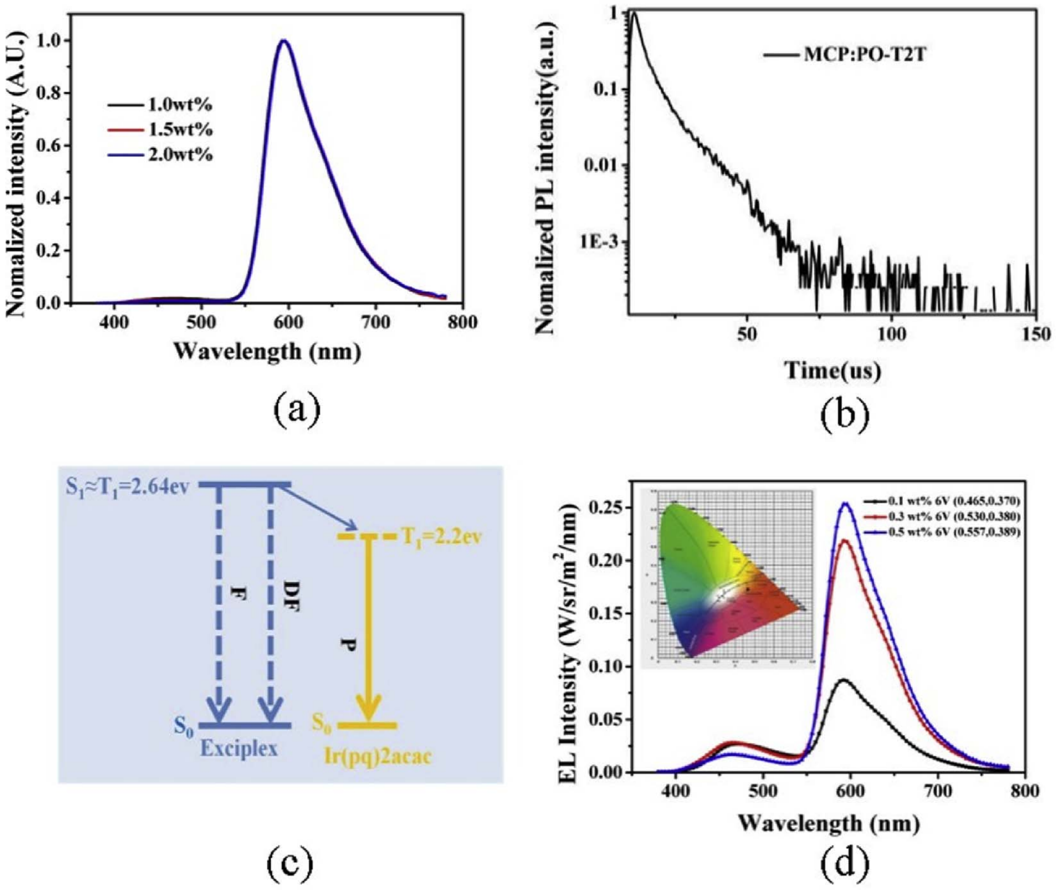
absorption spectrum of Ir(pq)2acac, indicating that effective energy transfer between exciplex and phosphor could be brought about [18,29].

Fig. 2 describes the device structures with ITO/MoO$_3$(4 nm)/mCP (20 nm)/mCP:PO-T2T:Ir(pq) 2acac (xwt%(25 nm))/DPEPO(3 nm)/ Bepp2 (40 nm)/LiF (1.5 nm)/Al (100 nm), here the ratio mCP-to-(PO-T2T) in the EML is a 1:1 and x = 1.0,1.5 and 2.0, respectively. In the device system, ITO and LiF/Al were used as the anode and the compound cathode, respectively, as well MoO$_3$ (molybdenum trioxide) was used as the hole-injection layer, mCP and Bepp2 (bis [2-(2-hydroxyphenyl)-pyridine]beryll-ium) were used as the hole- and electron-transporting layer, respectively. A thin interlayer of DPEPO (oxybis(2,1-phenylene))bis(diphenylpho-sphine oxide) inserted between Bepp2 and the EML acts as the exciton blocking layer, various Ir(pq)2acac concentration were doped into (mCP:PO-T2T) exciplex host, which functions as EMLs. High triplet state level based DPEPO plays an exciton blocking role, which can effectively restrict the singlet- and triplet-excitons within Bepp2 being in favors of energy transfer. So the exciton would be distributed in the entire EML, thereby, the exciton quenching would be reduced, improving the devices efficiency, Such a good result should be attributed to presence of higher triplet of DPEPO compared to Bepp2.

Fig. 3 (a) shows current density-voltage-luminance. We can see that although the turn-on voltage of all devices is about 3V, higher turn-on voltage may also be arisen from a larger transporting barrier lying between DPEPO and Bepp2. From the J-V curve we also observed that variety of doping concentration almost does not affect on current density. This phenomenon clarifies that the emission process is ascribed to energy transfer mechanism, otherwise, if the current density is dependent on the phosphor doping concentration, dominant emission mechanism would be charge trapping process [1–4,14,20,21,30].

For our EL device the current density does not increase with the Ir (pq)2acac doping concentration. Therefore, there would be energy transfer from the exciplex host to the phosphor. It is better that for the exciplex to ensure the transporting balance between the charge carriers so that the mutual quenching between the polaron and the exciton could be avoided, thus, which could help to improve the EL efficiency of the device. Thus, we adjusted the doping concentration to optimize the EL performance. Fig. 3(b)-(d) specify the current efficiency, power efficiency and EQE curves, respectively. As can be seen from the figures that all devices almost exhibit low efficiency roll-off, which is attributed to the good bipolar property of the exciplex host, wider carrier recombination region due to distributed throughout EML, the appropriate Ir(pq)2acac doping concentration also reduces the exciton quenching. As a 1.0 wt% of Ir(pq)2acac was used the current efficiency, power efficiency and EQE are 30.08cd/A, 31.5 lm/W and 16.52%, respectively. This may be due to the fact that the Ir(pq)2acac dopant concentration is too low to incomplete energy transfer from exciplex host to phosphor was took place [10,16,17,20,31]. When a concentration of 1.5 wt% was used the best device performance has been determined. CE, PE, and EQE are 35.3cd/A,31.0 lm/W and 18.7%, respectively. Besides, we also note that even as the current density reaches up to 10 mA/cm^2, 20 mA/cm^2 and 30 mA/cm^2, the current efficiency, power efficiency and EQE still keep 28.58cd/A,26.56cd/A and 25.18cd/A, 15.22 lm/W,13.07 lm/W and 11.89 lm/W, as well 15.57% and 14.33% and13.25%, respectively. From the data we could conclude that low efficiency roll-off for our orange device was achieved. The results should be ascribed to absence of concentration quenching effect, i.e., full excitons were used for EL emission due to better phosphor doping concentration.

We use the following equation to calculate E.Q.E., here h is Planck's constant, c is the velocity of light, I is operating current and e is electron charge, L is the luminance of the OLED (cd/m^2),K (λ) is the Commission International de l'Eclairage chromaticity (CIE) standard Photopic Luminous Efficiency Function,S(λ) is the luminous intensity of different wavelengths, λ is the wavelength.



(a)

(b)

(c)

(d)

Fig. 4. (a) The normalized EL spectra of OLEDs with 1.0, 1.5 and 2.0 wt% phosphor (b) the blue PL decay curve of mCP, PO-T2T mCP:PO-T2T mixing film (1:1), (c) The energy transfer processes of the EL device with phosphor dopant, here, left: the S_0,S_1 and T_1 represent the ground state, excited singlet state and excited triplet state of exciplex, respectively, blue dotted arrow -F and -DF depict fast and delayed-fluorescence, respectively. Orange line-arrow in right: S_0, T_1 and P denote singlet-, triplet-excited state as well orange phosphorescence emission from the phosphor, which is resulted from exciplex energy transfer. (d) EL spectra of device with different doping concentration and inset are CIE coordinates of the white OLED device with 0.1 wt% phosphor dopant. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Table 1
EL performance combination between our orange device A and other reported orange OLEDs.

	Turn Voltage (V)	Current Efficiency (cd/A)	Power Efficiency (lm/W)	EQE(%)	Emission Peak(nm)	Reference
Device A	3	35.3	31.0	18.7	596	This work
Device B	2.5	23.1	22.4	15.5	604	Ref. [22]
Device C	2.6	25.6	26.6	15.4	596	Ref. [22]
Device D	4.5	14.74	–	8.92	602	Ref. [24]
Device E	2.4	–	62.1	25	524/616	Ref. [9]
Device F	2.4	–	22.7	14.5	616	Ref. [9]
Device G	–	–	73.1	27	556	Ref. [18]

In the table DeviceA:ITO/MoO_3(4 nm)/mCP(20 nm)/mCP:PO-T2T:Ir(pq)2acac (1.5 wt% (25 n m))/DPEPO(3 nm)/Bepp2 (40 nm)/LiF (1.5 nm)/Al(100 nm).

DeviceB: ITO(150 nm)/TAPC(20 nm)/TCTA(10 nm)/TCTA:B3PYMPM:Ir(ppy)2(acac):Ir (mphq)2(acac) (30 nm,8 wt%, × 0.3 wt%)/B3PYMPM (45 nm)/LiF (0.7 nm)/Al (100 nm).

DeviceC:ITO(150 nm)/TAPC(20 nm)/TCTA(10 nm)/TCTA:B3PYMPM:Ir(ppy)2(acac):Ir (mphq)2(acac) (30 nm,8 wt%, × 2.0 wt%)/B3PYMPM (45 nm)/LiF (0.7 nm)/Al (100 nm).

DeviceD:ITO/HATCN (5 nm)/NPB(40 nm)/TCTA (20 nm)/CzTrz (30 nm): 3 wt%PO-01/ BPhen (40 nm)/LiF (0.5 nm)/Al (150 nm.

Device E:ITO/NPB (30 nm)/CC-MP8 with 2% Ir(pq)2(acac)(30 nm)/PBD (50 nm)/LiF (0.5 nm)/Al (130 nm)/LiF (0.5 nm)/Al(130 nm).

Device F:ITO/TPAC (40 nm)/CC-MP4 with 1% Ir(pq)2(acac) (30 nm)/TmPyPb (50 nm)/ LiF(0.5 nm)/Al (130 nm)/LiF (0.5 nm)/Al (130 nm).

Device G:ITO (150 nm)/DNTPD(60 nm)/NPB (30 nm)/SBAF-2C:Ir(pq)2acac (30 nm 3%)/ BCP (5 nm)/Alq3 (20 nm)/LiF (1 nm)/Al (200 nm).

$$\eta_{\mathrm{EQE}} = \frac{\mathrm{N_p}}{\mathrm{N_e}} = \frac{\pi.\ \mathrm{e.L}}{683.\mathrm{h.c.l}} \times \frac{\int_{400}^{760} \mathrm{S}(\lambda)\cdot\lambda d\lambda}{\int_{400}^{760} \mathrm{S}(\lambda)\cdot\mathrm{K}(\lambda) d\lambda}$$

After the phosphor dopant Ir(pq)2acac was rose to 2 wt%, maximum current- and power-efficiency are 33.8 cd/A and 30.4 lm/W as well EQE reaches up to 18.1% respectively. The device efficiency decreases with the increase of the doping concentration, this case is mainly due to the serious TTA of the phosphorescent material. When the doping concentration of Ir (pq) 2acac is increased, the energy transfer from the host to the guest causes. The increase of the exciton concentration makes the triplet excitons to more easily quench each other, which also makes the efficiency roll-off of the 2.0 wt% doping device more severe compared to1.5 wt% doping device [31,32].

Fig. 4 (a) depicts the EL spectra at different Ir(pq)2acac concentrations and EL peak in 596 nm of the phosphor. From the figure we can see that for the device with 1 wt% phosphor a weak EL peak at 471 nm was also observed, which should be ascribed to emission derived from exciplex itself, and as the phosphor concentration of the device is larger 1 wt%, the peak of the exciplex emission was almost quenched by the phosphor emitter, which confirms that as phosphor doping into device is lower than 1 wt%, the incomplete energy transfer from exciplex to phosphor must be generated. As a result, the device offers either blue or orange EL emissions, which derived respectively from exciplex and the phosphor. Furthermore optimal device with 1.5 wt% Ir(pq)2acac exhibits a peak efficiency, i.e., its CE, PE, and EQE

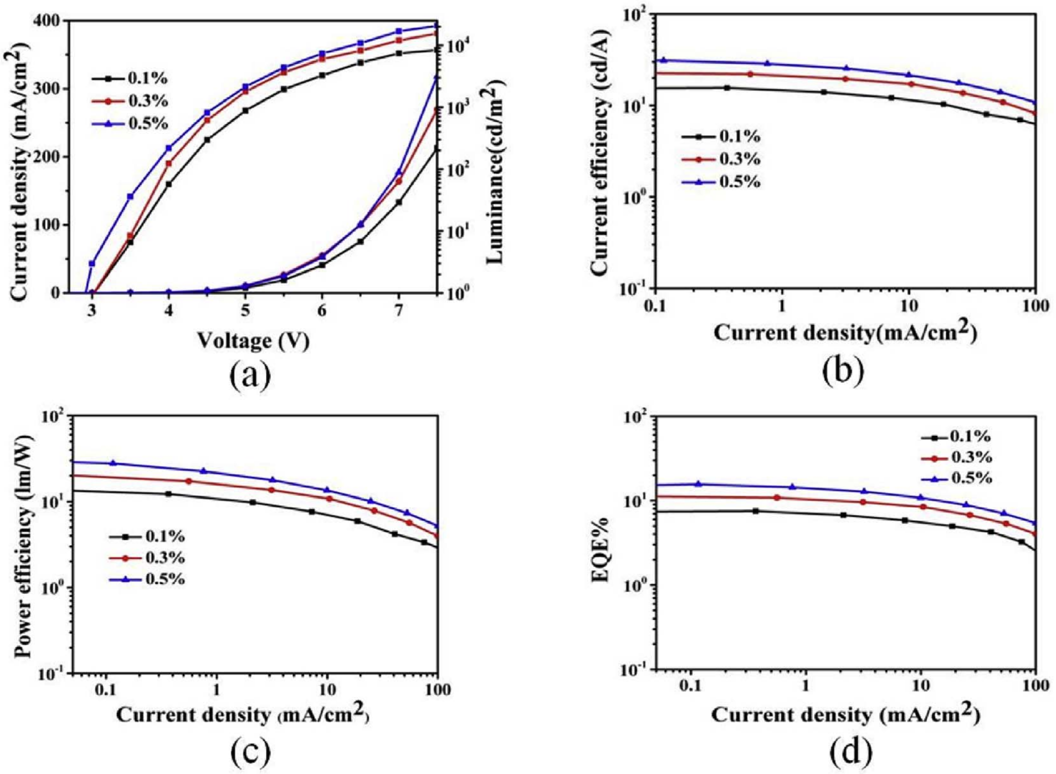


Fig. 5. The EL performance of three OLEDs in this study. (a) Current density-voltage-luminance (b) Current efficiency-current density (c) power efficiency-Current density (d) EQE-current density.

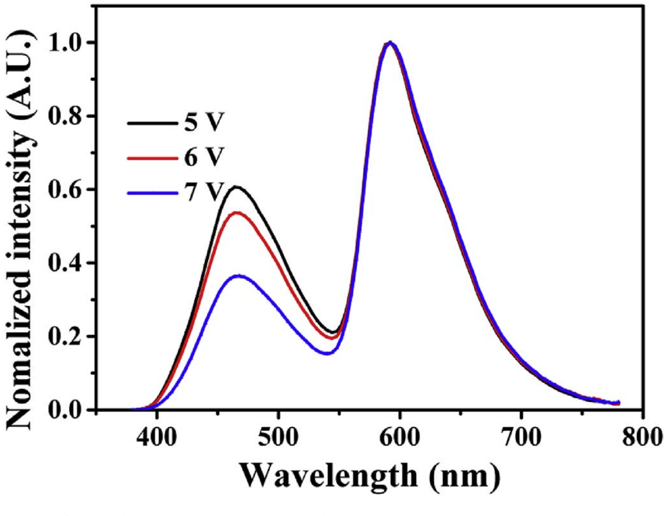


Fig. 6. The EL spectra of 0.1 wt% doping device under different voltages.

are 35.3cd/A, 31.0 lm/W and 18.7%, respectively.

In order to validate TADF property of mCP:PO-T2T blue exciplex PL, its transient decay curve was determined, as described in Fig. 4(b). According to the blue transient PL decay curve we used formula (1) to calculate τ_1 and τ_2 which are 1.74μs and10.83 μs, respectively. Which behaves excited state lifetimes of the exciplex emission, total decay time is about 5.77 μs, which prove that $\Delta E_{s\text{-}t}$ is extremely small. Thus the fact that the blue exciplex emission should be ascribed to TADF processes has been adequately affirmed [13,19,33,34].

$$\tau = \frac{B_1\tau_1^2 + B_2\tau_2^2}{B_1\tau_1 + B_2\tau_2} \qquad (1)$$

here, B1 and B2 are preexponential factor.

In terms of the decay curve of mCP, PO-T2T and mCP: PO-T2T mixing film (1:1) and above the EL energy transfer processes, we can deduce that phosphor emission actually originates from the energy transfer from exciplex host, as displayed in Fig. 4(c). Right orange narrow points phosphor orange EL emission, which grows out of energy transfer from the exciplex host. The triplet exciplex energy was either up-converted to exciplex singlet by RISC (reverse intersystem crossing) processes, or was transferred partly to triplet state of phosphor by energy transfer mechanism [8,10,26,35,36]. So in theory triplet excitons and singlet excitons were fully used for harvesting EL efficiency and improvement in efficiency roll-off. On the other hand, three EL spectra and CIE coordinates of OLEDs are described in Fig. 4(d), we see that 0.1 wt% phosphor doping based device offers warm white, as indicated in the inset i.e., CIE coordinates(0.465,0.370). The warm white emissive EL efficiency would also be further optimized, but dependences of EL efficiency on current density make clear that all efficiency roll-off are diminutive. The achievement of hybrid WOLED should be ascribed to incomplete energy transfer from exciplex host to the phosphor.

Table 1 summaries EL performance between our orange device (deviceA) and other reported orange OLED. From this table we note that the orange device with Ir(pq)2acac phosphor as emitter furnishes an optimal EL performance. Recently other groups have also reported orange OLED device. Such as, Kim group's device B also content green emission component with 524 nm, which leads to high EL intensity.

Besides, Duan group has reported orange device, but the EL spectral peak of their orange device lies at shorter waveband than our orange EL peak, So short orange wavelength of the OLED would offer higher EL intensity than our pure orange emission. In other words, we clearly observed their EL spectra do not lie orange spectral range, i.e, the spectra almost locate at more shorter waveband so that such spectra based EL intensity would be higher than our pure orange ones. Besides, WOLED made by combining orange light with shorter wavelength with the blue one could only yield narrower bandwidth, i.e, poor color rendering index.

To achieve hybrid WOLED we further reduced phosphor doping content to fabricate a new OLED. The device has structures of ITO/ MoO_3(4 nm)/mCP(20 nm)/mCP:PO-T2T:Ir(pq)2acac (xwt%(25 nm))/ DPEPO(3 nm)/Bepp2(40 nm)/LIF (1.5 nm)/Al (100 nm), in exciplex EML donor-to-acceptor ratio is 1:1 and x = 0.1, 0.3 and 0.5, respectively.

From Fig. 5 we can see that the device efficiency increases with the doping concentration, the fact is mainly due to more effective energy transfer from exciplex to the dopant as concentration of Ir(pq)2acac increases, reducing the non-radiative decaying from the triplet excitons themselves, thereby, improving the efficiency of the device [8,16,17,26,37]. At the same time we also observed that their efficiency roll-off also are weaker from relation of EL properties vs current density.

Fig. 6 shows the EL spectra of 0.1 wt% doping concentration device under different voltage. As can see from the figure that a good white light spectrum was observed, but the spectral stability of the device under different voltages is not particularly good, which is mainly attributed to difficult of managing excitons in the a single emissive layer. This also provides a reference for the next preparation of multi-emissive white light devices.

3. Conclusions

We have used mCP:PO-T2T exciplex as host and orange phosphor as dopant a good performance orange phosphorescent device was achieved. As 1.5 wt % phosphor (Ir(pq)2acac) was doped into exciplex host maximum current efficiency, power efficiency and EQE are 35.3 cd/A,31.0 lm/W, 18.7%, respectively, which are highest to date. Furthermore, as 0.1 wt% phosphor was doped into exciplex host, current efficiency, power efficiency and EQE of the WOLED are 15.55 cd/A 13.69 lm/W 7.49%, respectively. Thus, warm white OLED was achieved by incompletely energy transfer from exciplex host to orange dopant. Both orange and warm white OLED offer very low efficiency roll-off, which would be significant for their future application. The architectonics design of our pure orange- and warm white—OLEDs are highly simple, which would be in favour of industrial applications.

Author contributions statement

X.Z.Y. wrote the main manuscript and carried out most of the experiments and data analysis. C.L.Q.,W.H., L.W.L., L.X.G., X.B.S. and Z.B. guided the progress of experiments, manuscript writing and participated in mechanism discussions. All authors reviewed the manuscript.

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