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Efficient exciplex emission from intramolecular charge transfer material

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ABSTRACT

New exciplexes formed between a typical intramolecular charge transfer (ICT) material (bis[4-(9,9-dimethyl-9,10-dihydroacridine)phenyl]sulfone (DMAC-DPS)) and a series of electron donor and acceptors in donor:acceptor system have been systematically demonstrated. It is found that such ICT materials could form exciplex with both standalone electron donor and acceptor materials with itself as acceptor and donor components, which is based on the presence of both donor and acceptor species in the ICT material. The emission spectra of exciplex OLEDs based on ICT materials could be regularly tuned ranging from blue to yellow color by changing energy level alignment between ICT and standalone donor/acceptor materials. Among these exciplexes, DMAC-DPS:PO-T2T combination offered the highest exciplex EL performance, with its peak external quantum efficiency, luminance and current efficiency of 9.08%, 35,000 cd/m^2 and 30 cd/A, respectively. On the other hand, we also found that the exciplex efficiency was insensitive with the weight ratio between ICT material and acceptor, which means 'doping' of ICT material into the acceptor. Our finding extend the usage and selection scope of the TADF material.

1. Introduction

Organic light emitting diodes (OLEDs) are anticipated as one of the next generation energy-saving and eco-friendly light sources and flat panel displays because of their merits like wide-view-angle, low driving voltages, and high efficiencies [1–3]. The traditional OLEDs are mostly based on fluorescent and phosphorescent materials and still suffer from critical disadvantages like low efficiency of fluorescent device and large efficiency roll-off and high application cost of phosphorescent OLED [4–6]. In view of the disadvantages in fluorescent and phosphorescent OLEDs, great efforts have been devoted to develop highly efficient noble metal free materials. Among these efforts, a new mechanism named thermally activated delayed fluorescence (TADF) was raised by the group of Adachi and has been attracted a lot of research interests [7,8]. OLEDs based on TADF materials are considered to have unit internal quantum efficiency because the randomly formed 75% triplet excited states can up convert into the singlet excited state and contribute to light emission [9,10]. In this mechanism, the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) of an emitting material should be sufficiently separated to minimize the energy difference between singlet and triplet excited states [11]. This small energy difference, designed comparable with the K_BT (K_B the Boltzmann constant, T for temperature) at room temperature, enables efficient thermally activated reverse intersystem crossing of triplet states to the singlet state. Both intra molecular charge transferred (ICT) materials and exciplexes formed between electron donors and acceptors are recognized with TADF character [12–15]. Up till now, a great deal of ICT materials and exciplexes were proved have high emitting efficiencies [11,16–20]. Among which, the most impressive OLEDs based on blue and green ICT emitters performed high EQE of 19.5% and near 30% respectively [11,13,21]. However, the properties of the ICT materials in either Photoluminescence (PL) or electroluminescence (EL) situations are still not sufficiently explored. The ICT materials in EL condition can easily form exciplexes with the host materials if the energy levels are not located in the energy gap of the host materials. These exciplex emissions would damage the color purity of the ICT emission and this effect has not been paid enough attentions. On the other hand, the ICT materials have been proved of huge potential to be used in undoped (layered) devices where the ICT materials could also form exciplexes with transporting materials [22–24]. Therefore, it is of great significance to investigate the exciplex behaviors of ICT materials because it can improve the efficiency and quality of ICT

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emission and give new insight of the ICT materials. What's more, it can also be used as primary active layer in device if the ICT material based exciplex is efficient.

2. Experimental section

To study the interaction between ICT materials and other electron donors and acceptors, we chose bis[4-(9,9-dimethyl-9,10-dihydroacridine)phenyl] sulfone (DMAC-DPS) as a typical ICT material because DMAC-DPS is one of the most famous blue ICT dopant with high EQE of near 20% [13]. Generally, a highly efficient TADF material have a complete separation between its HOMO and LUMO electron cloud, which are mostly populated at the electron donating and electron accepting fragments respectively [11]. As a result of this separation, the ICT material can form new exciplexes with both electron donor and acceptor materials. The HOMO and LUMO levels of DMAC-DPS are located at 2.9 eV and 5.9 eV respectively [13]. These modest energy levels enable us to design serial devices with consecutive individual emitting spectra. Three electron acceptors, including 2,4,6-tris (biphenyl-3-yl)-1,3,5-triazine (T2T), bis-4,6-(3,5-di-4-pyridyl-phenyl)-2-methylpyrimidine (B4PyMPm) and PO-T2T and one electron donor N,N′-diphenyl-N,N′-bis(3-methylphenyl)-1,1′-biphenyl-4,4′-diamine (TPD) were chosen to form exciplex with DMAC-DPS to modulate the emitting spectra [18,25,26]. The molecular structures of DMAC-DPS, T2T, B4PyMPm, PO-T2T and TPD are given in Fig. 1.

OLED devices are fabricated using pre-cleaned indium tin oxide (ITO) coated glass substrates with a sheet resistance of 15 Ω cm^{-2} and ITO thickness of 150 nm. They were patterned so that the OLED devices had a pixel size of about 12 mm^2. The small molecule and cathode layers were thermally evaporated using the multiple-source organic molecule deposition method. To fabricate the device, the devices were prepared in vacuum at a pressure of $5 * 10^{-4}$ Pa. The MoO_3, hole-transporting materials mCBP and TPD, electron-transporting material T2T, B4PyMPm, and PO-T2T were thermally evaporated at a rate of 0.1 nms^{-1}. Blends of DMAC-DPS:acceptor and TPD:DMAC-DPS were deposited at a rate of 0.1–0.2 nms^{-1}. After the deposition of organic film, a 0.8 nm layer of LiF and a 150 nm layer of aluminum were thermally evaporated onto the organic surface. The four device structures are indicated in Fig. 2.

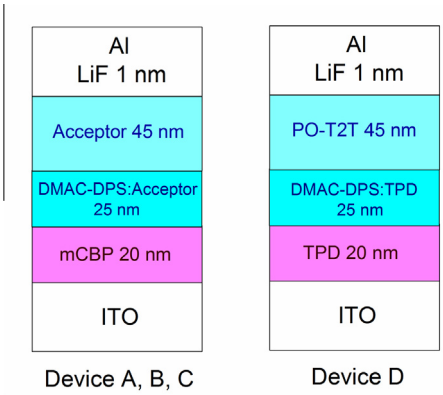

Fig. 2. Device architectures used in this work. The structure of device A, B and C is: ITO/MoO_3 (3 nm)/mCBP (20 nm)/DMAC-DPS:acceptor (25 nm)/acceptor (40 nm)/LiF (0.8 nm)/Al with acceptor as T2T, B4PyMPm and PO-T2T; The structure of device D is: ITO/MoO_3 (3 nm)/TPD (20 nm)/TPD:DMAC-DPS (25 nm)/PO-T2T (45 nm)/LiF (0.8 nm)/Al.

PL spectra of all the films were measured using Hitachi F7000 spectrometer. PL decay times were determined by FLS980 Fluorescence spectrometer. Current–voltage-brightness characteristics were measured by using a Keithley source measurement unit (Keithley2400) with a calibrated silicon photodiode. The EL spectra were measured by a Spectra scan PR650 spectrophotometer. All the EL measurements were carried out at room temperature under ambient condition.

3. Results and discussion

To achieve high emitting efficiency, the ICT materials with TADF character are mostly doped into a host material to reduce its self-quenching events [27]. Usually, the HOMO and LUMO of an ICT dopant should lower and higher than or equal to that of the host. If this requirement is not satisfied, exciplex formed between the host and the ICT dopant would appear [28]. This phenomenon can also observed in the traditional fluorescent emitter case, like AlQ_3 with m-MTDATA [29]. This unexpected exciplex in the traditional fluorophore case were firstly regarded as a negative effect that would reduce the quality of emitting color [30]. But

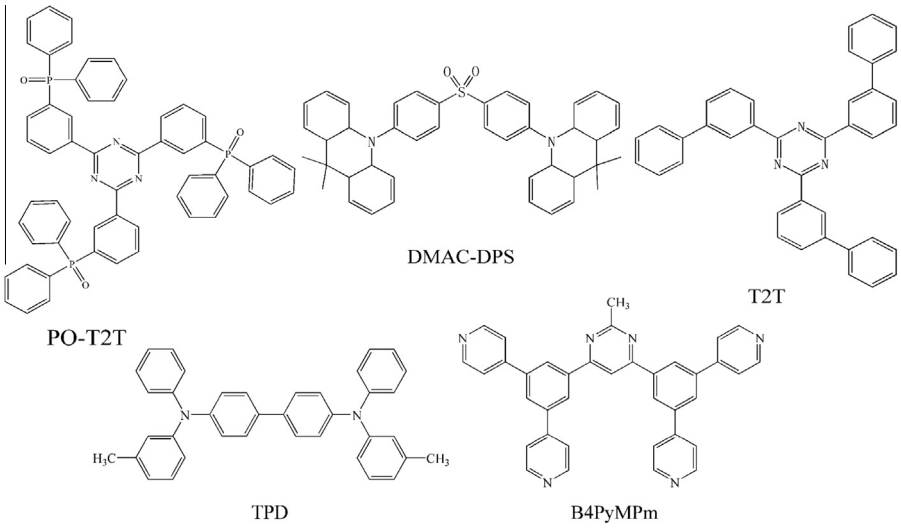

Fig. 1. Molecular structures used in this work. PO-T2T, T2T and B4PyMPm were used as electron acceptor and electron transporting layer. TPD was used as electron donor and hole transporting layer.

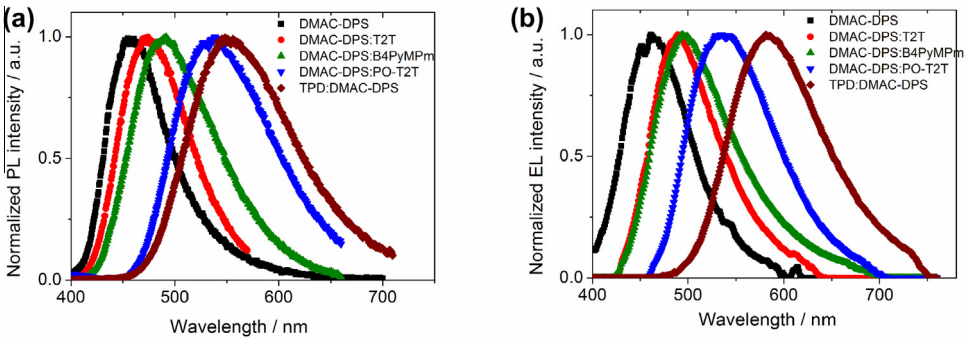

Fig. 3. (a) Normalized PL of mCP:(6 ± 1 wt.%) DMAC-DPS doped film, DMAC-DPS:T2T, DMAC-DPS:B4PyMPm, DMAC-DPS:PO-T2T and TPD:DMAC-DPS mixed films. mCP: N,N′-dicarbazolyl-3,5-benzene. The PL excitation wavelength is 355 nm. (b) The EL spectra of device A, B, C and D under a bias of 6 V. The EL spectra of DMAC-DPS based device with device structure of: ITO/MoO3 (3 nm)/mCP (20 nm)/mCP: 6 ± 1 wt.% DMAC-DPS (25 nm)/TPBi (40 nm)/LiF (0.8 nm)/Al.

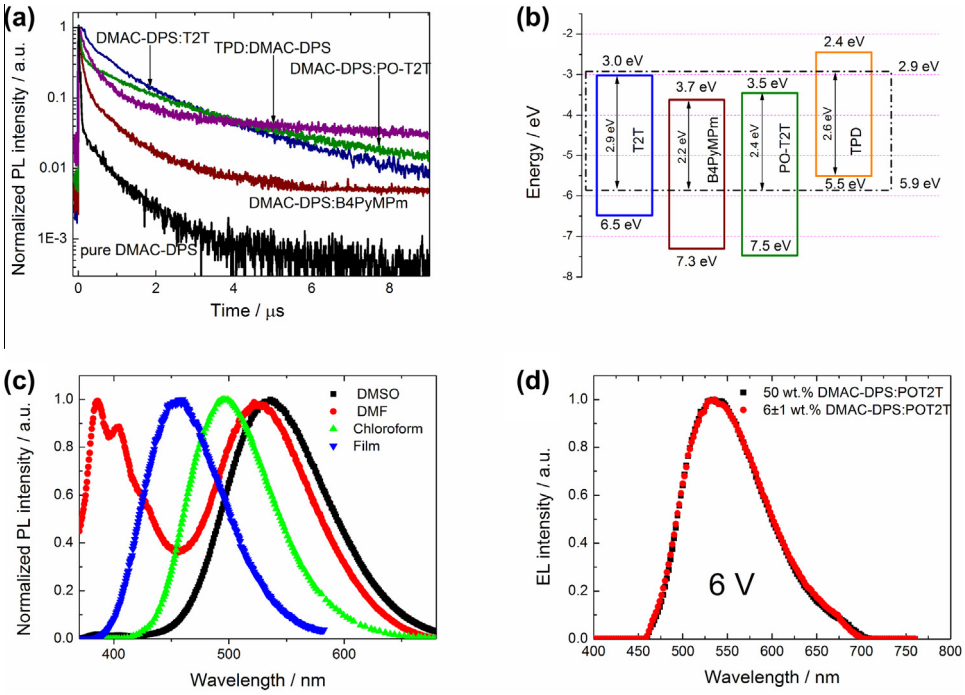

Fig. 4. (a) PL decay characters of DMAC-DPS:T2T, DMAC-DPS:B4PyMPm, DMAC-DPS:PO-T2T and TPD:DMAC-DPS mixed films. The excitation wavelength was 355 nm. (b) Energy diagram of DMAC-DPS, T2T, mB4PyMPm, PO-T2T and TPD. Double arrowed lines in rectangles indicate the theoretically transition energies of exciplex emissions. (c) PL spectra of DMAC-DPS in solvents with different polarities and as pure film. The structured peak in the PL of DMAC-DPS in DMF was from DMF solvent and machinery scattering. (d) EL spectra of device C with 50 wt.% and 6 ± 1 wt.% DMAC-DPS loading under 6 V bias.

Table 1
PL decay components of mixed films and DMAC-DPS doped mCP film.

Films	Component-1		Component-2		CIE (*x*,*y*)[b]
	Time/μs	Percent/%	Time/μs	Percent/%	
DMAC-DPS:T2T	0.27229	60.885	1.62158	39.115	(0.20, 0.37)
DMAC-DPS:B4PyMPm	0.05868	89.771	0.7106	10.229	(0.24, 0.42)
DMAC-DPS:PO-T2T	0.05846	77.134	1.79839	22.866	(0.36, 0.56)
TPD:DMAC-DPS	0.18177	76.871	1.03879	23.129	(0.51, 0.49)
Pure DMAC-DPS	0.03974	51.387	0.73526	42.613	-
DMAC-DPS in mCP[a]	0.021[a]	28.750[a]	3.1[a]	71.250[a]	-

[a] Data from Ref. [13].
[b] Under 6 V bias.

eventually, researchers found that the maximum theoretical efficiency of pure exciplex is as high as unit [31]. Accordingly, several exciplexes indeed give high EQE of above 5% due to its TADF character [9,31]. Similarly, the ICT material can also form exciplex with pure electron donating/accepting materials and this exciplex was treated as negative effects [28].

Fig. 3a displays the PL spectra of various exciplexes formed between DMAC-DPS and different donor or acceptor materials.

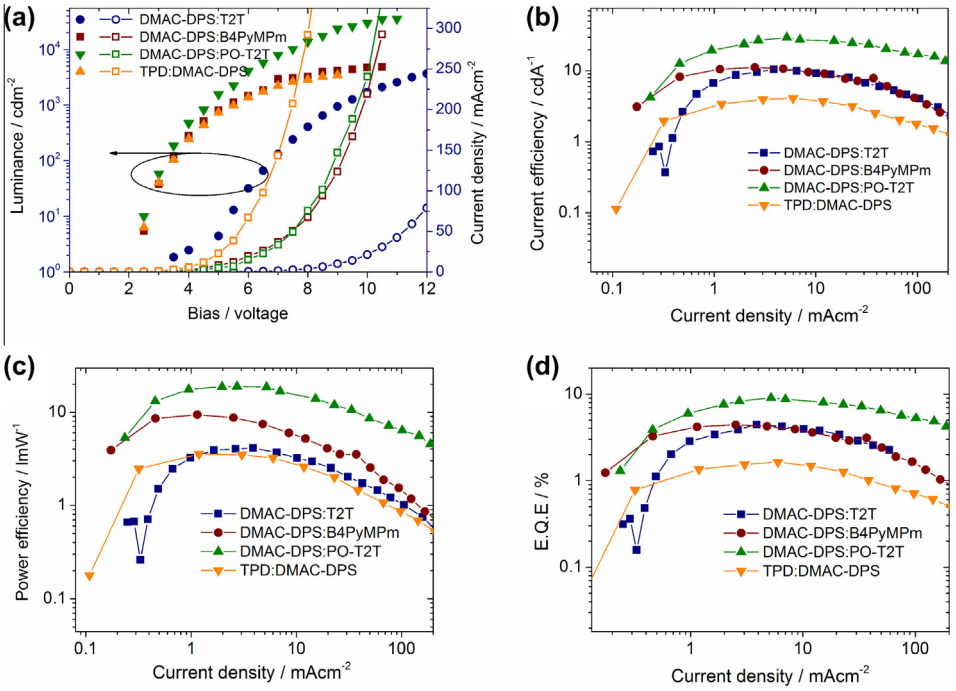

Fig. 5. (a) Luminance and current density vs operating bias of device A, B, C and D. (b) Current efficiency, (c) power efficiency and (d) EQE vs current density of device A, B, C and D.

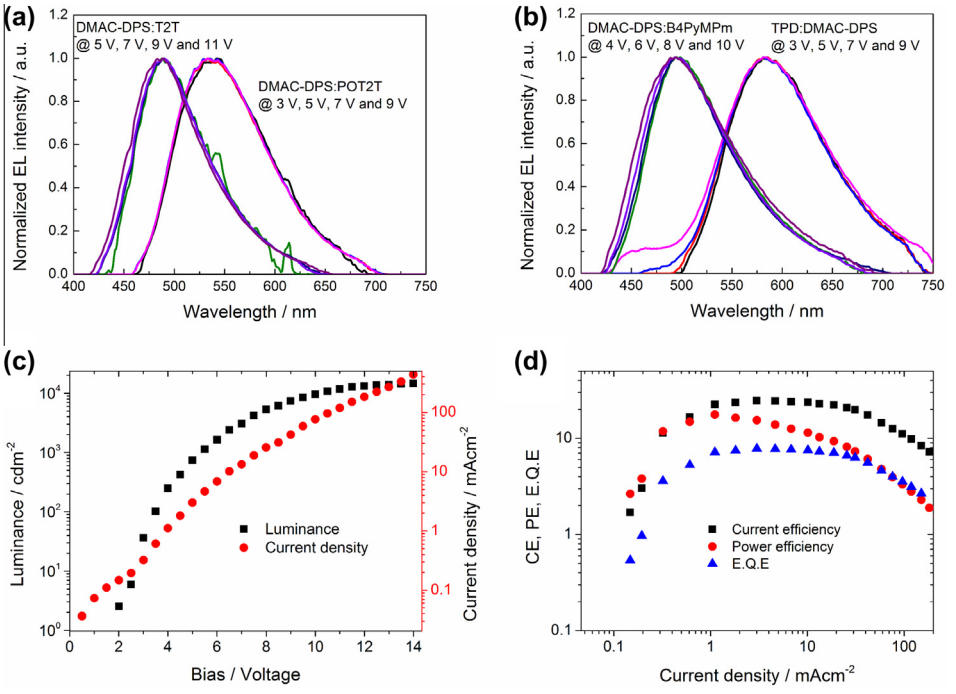

Fig. 6. (a) and (b) EL spectra of device A, B, C and D under different biases. (c) Luminance and current density vs operating bias of device C with 6 ± 1 wt.% DMAC-DPS. (d) Current efficiency, power efficiency and EQE vs current density of device C with 6 ± 1 wt.% DMAC-DPS.

The pristine PL spectra of DMAC-DPS as a blue dopant in the host of mCP are also presented in Fig. 3a for comparison (with spectra peak of 463 nm). When combined with T2T, B4PyMPm and PO-T2T in 50 wt.%:50 wt.% ratio, the mixed films performed consecutive PL spectra with peaks located at 480 nm, 493 nm and 535 nm. When combined with TPD as a representative donor material in 50 wt.%:50 wt.% ratio, the mixed film performed PL spectra with a peak of 550 nm. Forming exciplex with both donor and acceptor materials is a unique property of the ICT materials, which has never been reported. This dual property of ICT materials is origin from its combination of both donating and accepting components in one molecule. In this aspect, the ICT materials are different from traditional bipolar host materials like mCP, mCBP, TCTA and CBP [32]. A traditional bipolar host usually has both donating and accepting properties in one molecule but the HOMO and LUMO are mostly have large overlap so that it cannot form exciplex with

both donor and acceptor. To evaluate the EL performance of these exciplexes, we have also fabricated exciplex based OLEDs with mixed films of DMAC-DPS:Acceptor (50:50 wt.%) and Donor:DMAC-DPS (50:50 wt.%). The EL spectra under bias of 6 V are shown in Fig. 3b with pristine EL spectra of DMAC-DPS (doped in mCP) based OLED under 6 V for comparison. As general exciplexes, the EL spectra of the ICT material based exciplex universally red shifted slightly relative to the PL spectra [16,31].

Usually, an efficient exciplex always behaves double exponential PL decay character due to its TADF from its completely separated HOMO and LUMO population. The PL decay character of our four DMAC-DPS based exciplexes were also examined, as depicted in Fig. 4a. The PL decay times of all four exciplexes contain two components including a prompt and a delayed one. The double exponential fitted decay times of four exciplexes are listed in Table 1. Comparing of the delayed components in the four exciplexes, it is found that the scale and percent of the delayed component in the PL decay have less influence on the quantum efficiency than the radiative recombination rate of singlet exciplex.

Generally, the peak of the exciplex PL emission spectra is determined by the energy difference between the acceptor's LUMO and the donor's LUMO level:

$$h\nu_{(\text{Exciplex})\text{Max}} \cong I_D - A_A - E_C \quad (1)$$

where I_D (HOMO) and A_A (LUMO) denote the ionization potential of the donor and electron affinity of acceptor, respectively. E_C expresses the coulombic attraction energy between D- and A-species [33,34]. In the case of DMAC-DPS:PO-T2T, DMAC-DPS:T2T and TPD:DMAC-DPS, the $I_D - A_A$ value are 2.4 eV, 2.9 eV and 2.6 eV which are roughly consist with Eq. (1) with peak spectra positions of 535 nm (2.32 eV), 480 nm (2.58 eV) and 580 nm (2.14 eV) respectively. However, in the case of DMAC-DPS:B4PyMPm, the $I_D - A_A$ value are 2.2 eV which is far deviate from the energy of peak spectra positions of 493 nm (2.52 eV), as depicted in Fig. 4b. The same anomalous energy behaviors have been previous reported in the polymer based exciplexes, where the nonbonding *n* electron orbitals played a critical effect [35]. On the other hand, most recently, Vygintas Jankus et al. systematically studied the PL (in solvent and film states) and EL spectra of an ICT material [36]. In that work, they found that the ICT material can perform both ICT emission (as PL in polar solvent) and exciplex emission in the ICT:TAPC mixed film and devices. In the case of DMAC-DPS based mixed films, there are also probability of ICT emission because the DMAC-DPS exhibit solvatochromism phenomenon too. To ascertain the origin of emissions of the DMAC-DPS based mixed films, PL spectra of DMAC-DPS emission in solvent with different polarity and in pure film were determined, as given in Fig. 4c. Since the PL spectra of pure DMAC-DPS film peaks at ca. 474 nm, it is impossible that the emission of the DMAC-DPS based mixed films were from aggregation of DMAC-DPS. The PL decay character of pure DMAC-DPS film are given in Fig. 4a and Table 1. It is obviously that the mixed films have different decay trend from that of the pure DMAC-DPS film. If the new emission of the DMAC-DPS based mixed films were from ICT emission in the acceptors with large permanent dipole moments, the spectra should vary with the mixed ratio. Therefore, representative device based on DMAC-DPS:PO-T2T with small mixed ratio of 6 ± 1 wt.% were fabricated. The EL spectrum of device C with small DMAC-DPS loading of 6 ± 1 wt.% almost coincide with the spectrum of the 50 wt.% loading based device, as displayed in Fig. 4d. This result suggests that the emissions of the mixed films and the relative devices were from exciplexes.

The device performances of four exciplexes are illustrated in Fig. 5a–d respectively. The blue, sky blue, green and yellow OLED had a maximum EQE of 4.44%, 4.40%, 9.08% and 1.63% which are among the highest exciplex based OLEDs. Impressively, the green exciplex gave a maximum luminance above 35,000 cd/m^2 and a maximum current efficiency of 30 cd/A with small efficiency roll off. The relative low efficiency of the TPD:DMAC-DPS based device is attributed to the relative planar spatial molecular structure of TPD which results in a low radiative decay efficiency of the singlet exciplex [37]. The EL spectra of four exciplex based devices are stable with only slight changes in a wide range of operating bias, as given in Fig. 6a and b. None the less, we have successfully manipulated the emitting spectra of an ICT material based exciplex system. Once efficient D:A system is found, one would cast efficient white OLED using only one dopant. And it is very interesting when the exciplex based device consists a 25 nm 6 ± 1 wt.% DMAC-DPS:PO-T2T film as emitting layer, the green device still performed a high EQE of 8.0% with a modest maximum luminance of 14,500 cd/m^2 (with other layers the same as in device C), as shown in Fig. 6a and b. This result was a little lower than that of device C but still among the highest green exciplex based OLEDs. The small loading of DMAC-DPS simultaneous means a host–guest manner of the exciplex based device.

4. Conclusion

In summary, through a systematic study, we investigated the interactions of a typical ICT material with a series of electron donor/acceptors. We found that the ICT materials could form new exciplexes with both an electron donor and an electron acceptor because the ICT molecules incorporate electron donor and acceptor moieties at the same time. The spectra of the ICT material based exciplex could be modulated gradually from blue to yellow color. In addition, the exciplex based OLED behaved excellent even when the donor and acceptor were mixed with a host guest weight ratio. These results open up a new developing route and new usage of the prevailing ICT materials.

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