The Strategy to Construct Multifunctional TADF Material for Deep Blue and High Efficiency Yellow Fluorescent Devices

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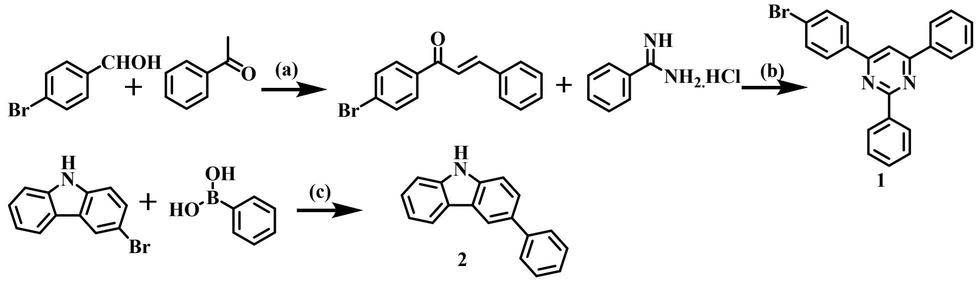
Device fabrication and measurement

The device fabrication and measurement are nearly identical with our previous reports.[1, 2] Indium tin oxide (ITO) with a sheet resistance of 15 Ω square^{-1} was used as the substrate. Prior to use, the ITO glass substrates were pre-cleaned carefully and treated by oxygen plasma for 5 min before device fabrication. Then the sample was transferred to the deposition system. All organic layers and MoO_3 were deposited at a rate of 1 Å s^{-1}, and subsequently LiF was deposited at 0.2 Å s^{-1} and then capped with Al (ca. 4 Å s^{-1}) through a shadow mask in a vacuum of 2×10^6 Torr. For all the OLEDs, the emitting areas were determined by the overlap of two electrodes as 0.09 cm^2. The EL spectra, CIE coordinates and *J–V–L* curves of the devices were measured with a PHOTO RESEARCH SpectraScan PR 655 photometer and a KEITHLEY 2400 SourceMeter constant current source. The EQE values were calculated according to previously reported methods. All measurements were carried out at room temperature under ambient conditions.



Scheme S1. The synthesis route of intermediate **1** and **2**.

Synthesis of 4-(4-bromophenyl)-2,6-diphenylpyrimidine (1):

First, a mixture of NaOH (10.8 g, 270 mmol), 50 mL ethanol, 150 mL water was added into a three-necks round flask, stirring in 0 ^{o}C ice bath. Then, acetophenone (6.48 g, 54 mmol) was added to the reaction bottle. After 30 minutes, bromobenzaldehyde (10 g, 54 mmol) was dissolved with as little ethanol as possible, and dripped into the reaction mother liquor, and stirred at room temperature for 7-9 hours. After that, cooled to 0 ^{o}C and orange solid was obtained by filtration, which directly proceeded the next step. Intermediate in the previous step, the same equivalent of benzoamidine hydrochloride and 5-fold equivalent NaOH were added in a three-necks round flask. The ethanol was added to dissolve the reactant, stirred and refluxed at 78 ^{o}C for 6 hours. The orange-white solid was obtained by thermal filtration. Finally, the white product was washed with ice ethanol for 2-3 times with a yield of 59%. ^{1}H NMR (400 MHz, CDCl$_3$) δ [ppm]: 8.74-8.68 (m, 2H), 8.31-8.26 (m, 2H), 8.19-8.16 (d, J = 8.8 Hz, 2H), 7.98 (s, 1H), 7.70-7.68 (d, J = 8.4 Hz, 2H), 7.59-7.50 (m, 6H).

Synthesis of 3-phenyl-9*H*-carbazole (2):

3-bromocarbazole (6 g, 24.3 mmol), phenylboronic acid (3.1 g, 25.6 mmol), 50 mL of 2 mol/L potassium carbonate solution, 100 mL of toluene, 50 mL of ethanol were added into a three-necks round flask, and the mixture was stirred and nitrogen was bubbled for 10 minutes. Then, Pd(PPh$_3$)$_4$ (0.84 g, 0.72 mmol) was quickly added. The resulting solution was stirred for 12 h at 100 °C under nitrogen. After completion of the reaction, the mixture was extracted with dichloromethane, washed with water, and then evaporated. The white products were obtained by silica gel column chromatography with a yield of 89%. 1H NMR (600 MHz, CDCl$_3$) δ [ppm]: 8.30 (s, 1H), 8.14-8.13 (d, J = 7.8 Hz, 1H), 8.06 (s, 1H), 7.72-7.71 (d, J = 7.8 Hz, 2H), 7.69-7.67 (d, J = 8.4 Hz, 1H), 7.49-7.47 (t, J = 7.8 Hz, 3H), 7.46-7.42 (m, 2H), 7.38-7.34 (t, J = 12.6 Hz, 1H), 7.29-7.26 (m, 1H).

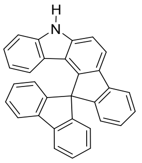
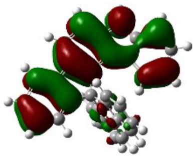
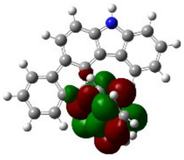
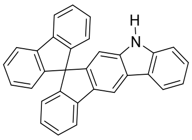
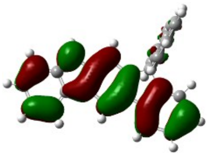
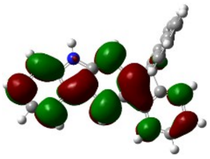
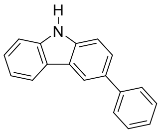
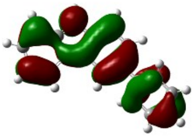
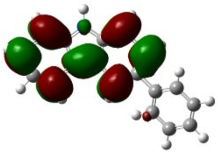
Structure	Orbital distribution		HOMO [eV]	LUMO [eV]
	HOMO	LUMO		
 SFI34			-5.09	-0.77
 SFI23			-5.19	-0.83
 3CzPh			-5.32	-0.72

Fig S1. The HOMO/LUMO orbital distribution of three donors and energy levels.

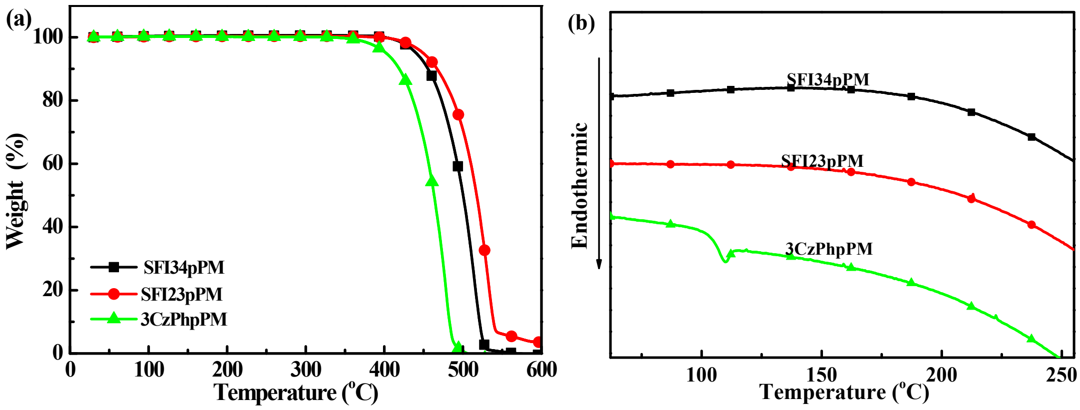


Fig S2. (a) The TGA curves and (b) DSC curves of **SFI34pPM**, **SFI23pPM** and **3CzPhpPM.**

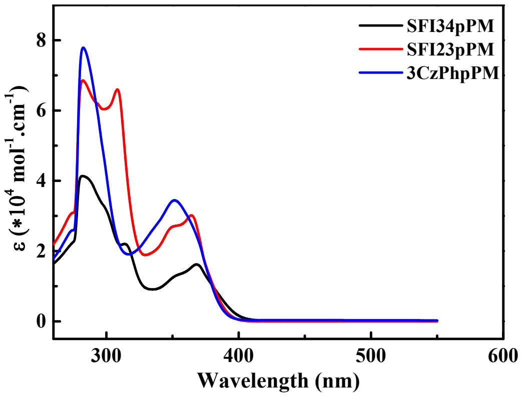


Fig S3. The extinction coefficient (ε) of three compounds in toluene.

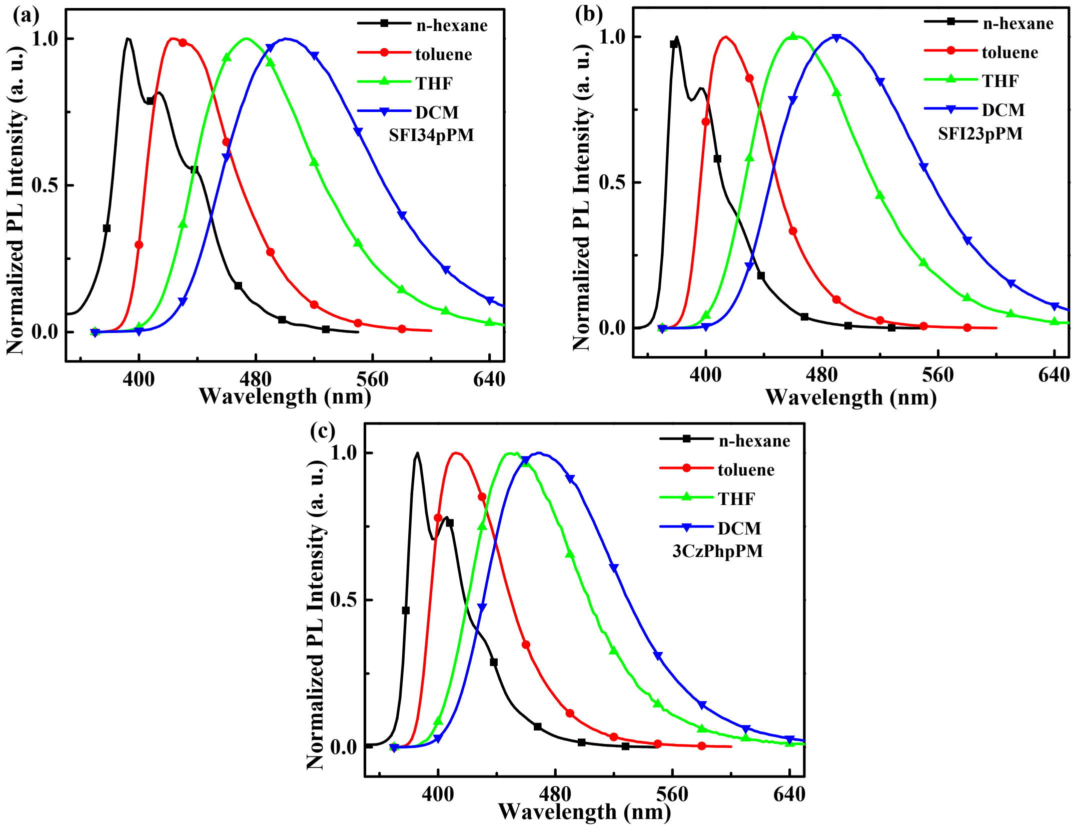


Fig S4. The emission spectra in four different solvents (a) **SFI34pPM**, (b) **SFI23pPM** and (c) **3CzPhpPM.**

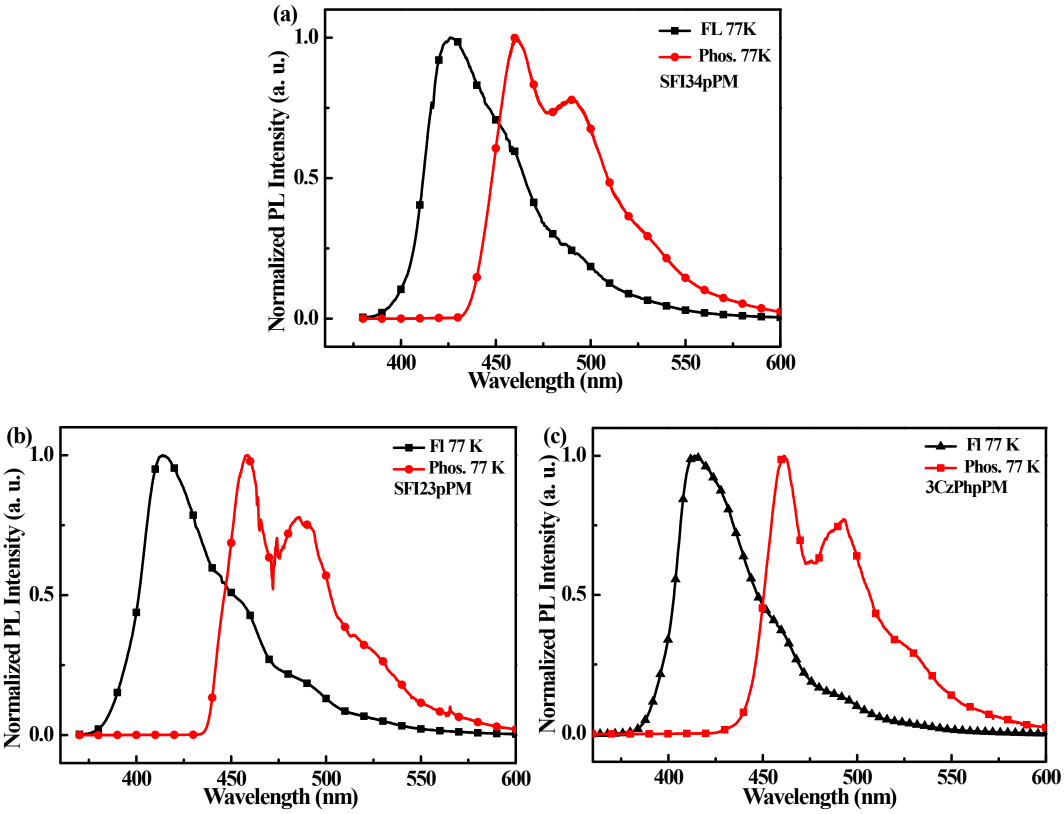


Fig S5. The fluorescence and phosphorescence spectra at 77 K in dimethyl tetrahydrofuran, (a) **SFI34pPM**, (b) **SFI23pPM** and (c) **3CzPhpPM.**

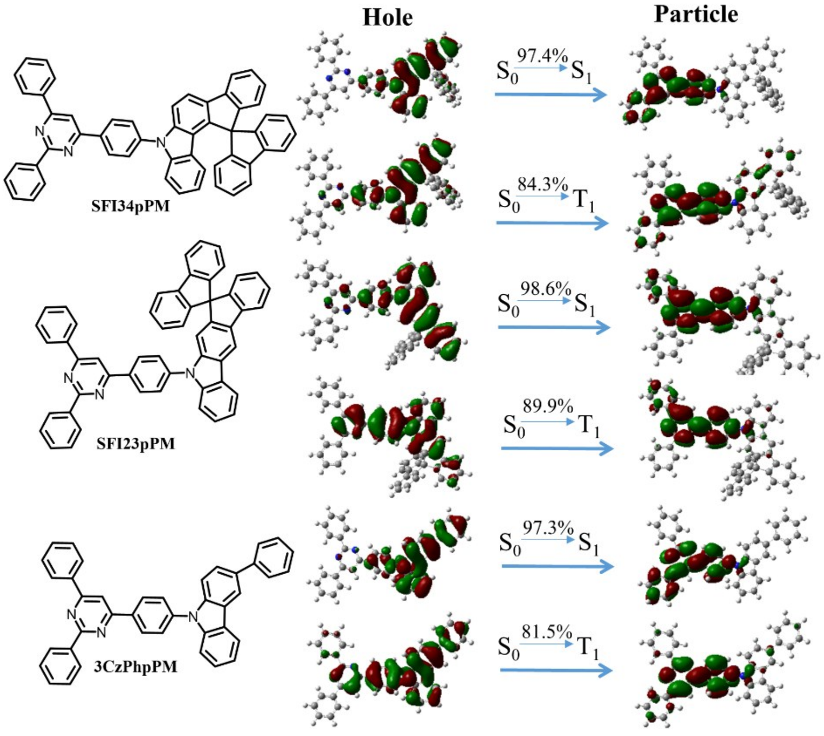


Fig S6. The NTO calculations of three PM compounds.

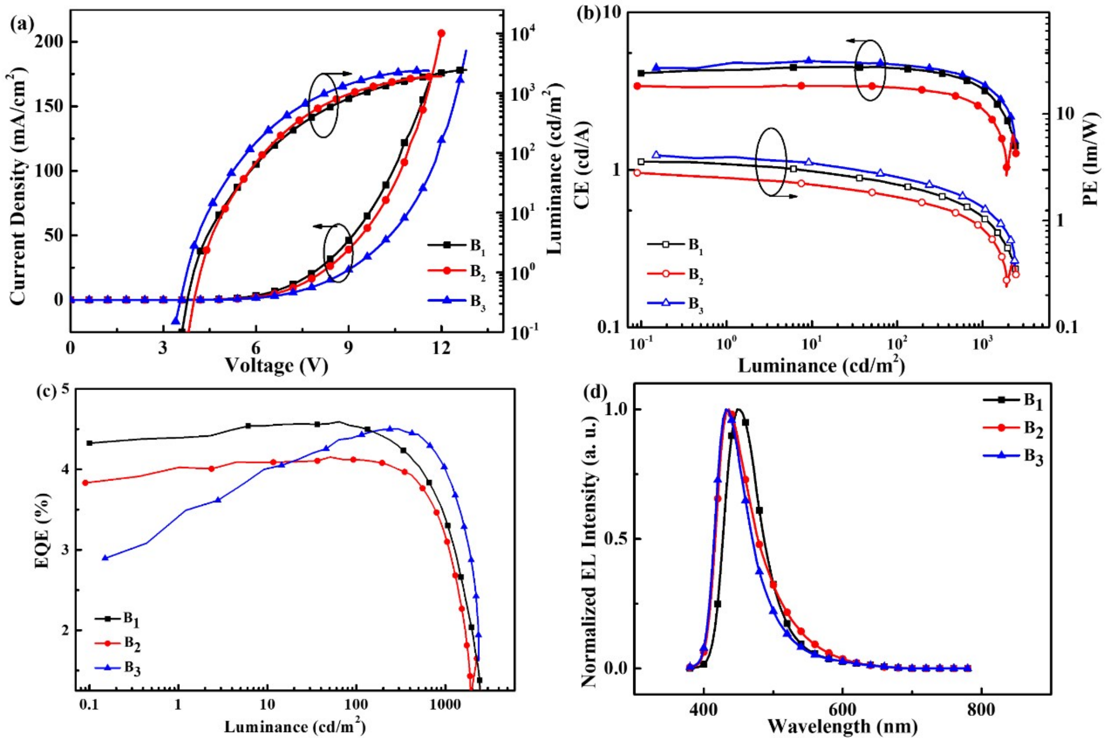
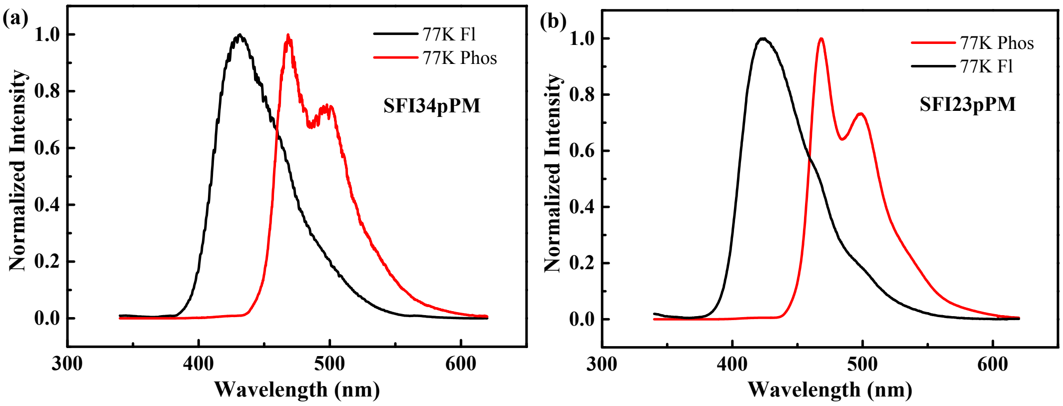


Fig S7. (a) Current density–voltage–luminance (C–V–L) curves; (b) CE-Luminance-PE; (c) EQE-Luminance; (d) Normalized EL spectra for device B$_1$-B$_3$.

Device structure B$_1$-B$_3$: ITO/MoO$_3$ (10 nm)/TAPC (40-50 nm)/*m*CP (5-10 nm)/**SFI34pPM** or **SFI23pPM** or **3CzPhpPM** (20 nm)/DPEPO (5 nm)/TmPyPB (20-40 nm)/LiF (1 nm)/Al.



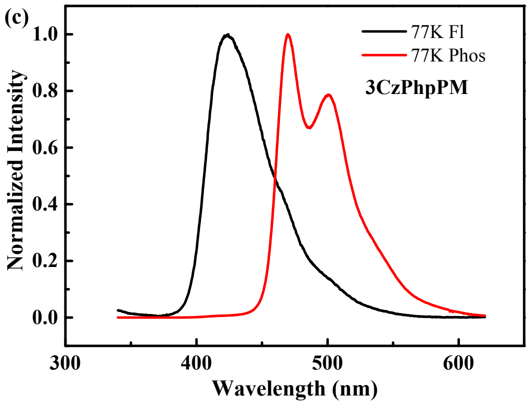


Fig S8. The low temperature fluorescence and phosphorescence of doped films (a) **SFI34pPM**; (b) **SFI23pPM**; (c) **3CzPhpPM**.

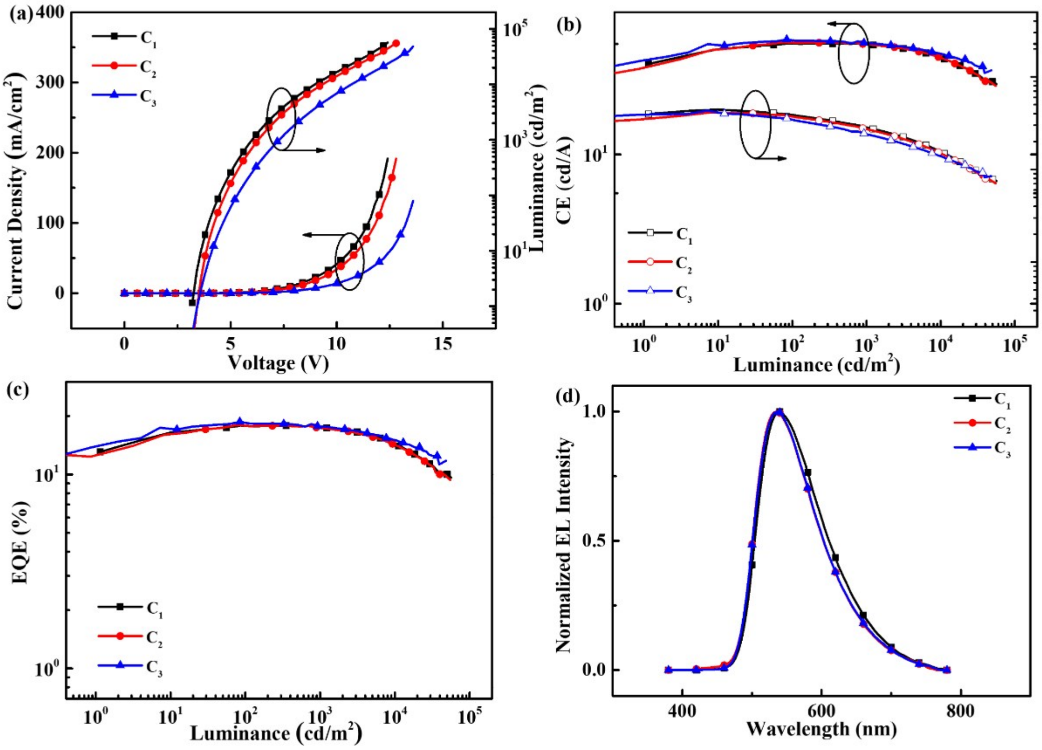


Fig S9. (a) Current density–voltage–luminance (C–V–L) curves; (b) CE-Luminance-PE; (c) EQE-Luminance; (d) Normalized EL spectra for device C_1-C_3.

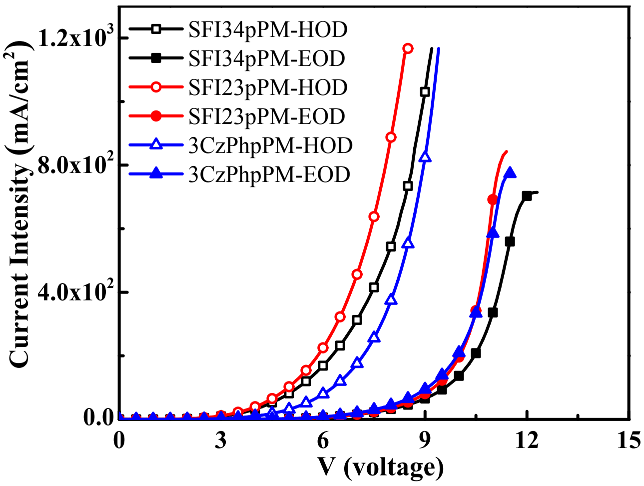


Fig S10. Current density–voltage curves of the hole and electron only devices based on the three compounds.

The single-carrier devices were fabricated with the configuration of ITO/MoO_3 (10 nm)/TCTA (20 nm)/**SFI34pPM /SFI23pPM /3CzPhpPM** (30 nm)/TCTA (20 nm)/MoO3 (10 nm)/Al for hole only device (HOD) and ITO/LiF (1 nm)/TmPyPB (20 nm)/ **SFI34pPM /SFI23pPM /3CzPhpPM** (30 nm)/TmPyPB (40 nm)/LiF (1 nm)/Al for electron only device (EOD), in which MoO_3 and LiF respectively served as the hole- and electron-injecting layer.

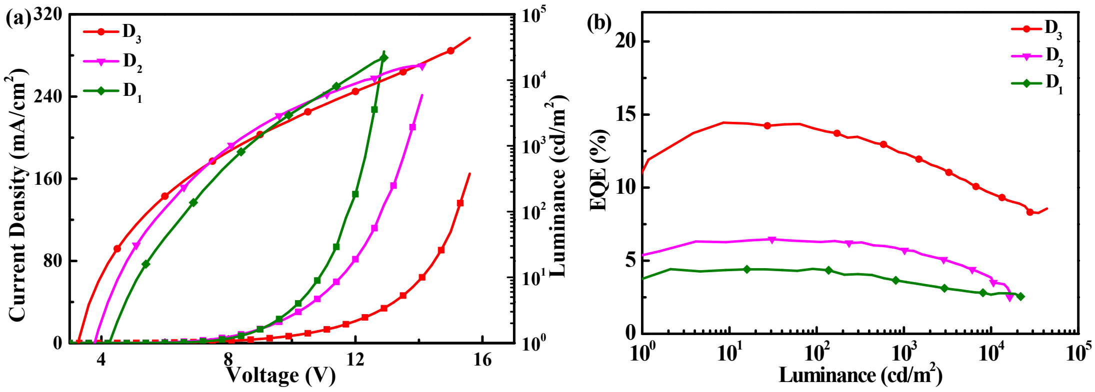


Fig S11. (a) Current density–voltage–luminance (C–V–L) curves; (b) EQE-Luminance of device D_1-D_3.

Device D_1: ITO/MoO_3 (10 nm)/TAPC (60 nm)/TCTA (5 nm)/ CBP: 1 wt%TBRb (30 nm)/ TmPyPB (40 nm)/LiF (1 nm)/Al (EQE_{max}=4.4%)

Device D_2: ITO/MoO_3 (10 nm)/TAPC (80 nm)/TCTA (5 nm)/ 34AcCzTrz: 1 wt%TBRb (30 nm)/ TmPyPB (40 nm)/LiF (1 nm)/Al (EQE_{max}=6.5%)

Device D_3: ITO/MoO_3 (10 nm)/TAPC (80 nm)/TCTA (5 nm)/ CBP:4 wt% 34AzCzTrz:1 wt%TBRb (30 nm)/TmPyPB (40 nm)/LiF (1 nm)/Al (EQE_{max}=14.2%)

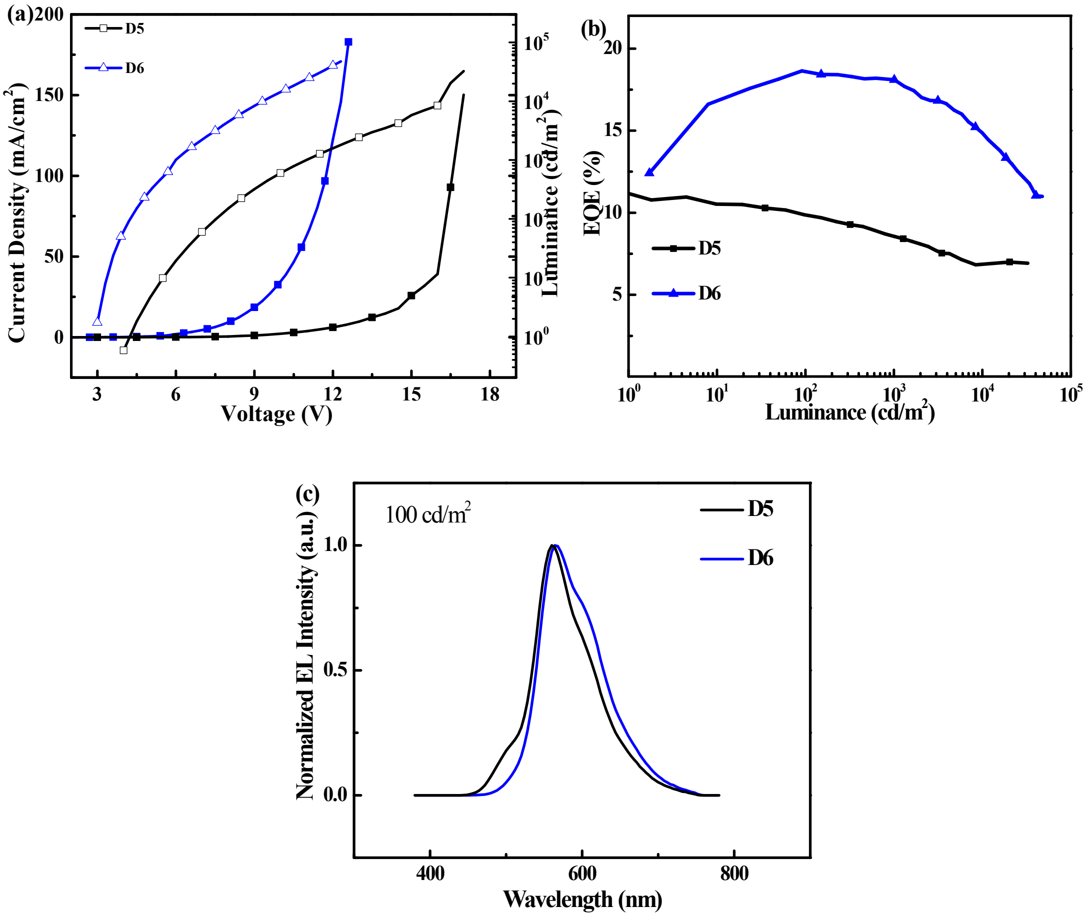


Fig S12. The EL performances of doping concentration dependent TADF sensitized fluorescent devices, (a) Current density–voltage–luminance (C–V–L) curves; (b) EQE-Luminance; (c) EL spectra of D_5 and D_6.

Device D_5: ITO/MoO_3 (10 nm)/TAPC (80 nm)/TCTA (5 nm)/ 3CzPhpPM:4 wt% 34AzCzTrz:0.5 wt%TBRb (30 nm)/TmPyPB (40 nm)/LiF (1 nm)/Al (EQE_{max}=11.5%)

Device D_6: ITO/MoO_3 (10 nm)/TAPC (80 nm)/TCTA (5 nm)/ 3CzPhpPM:4 wt% 34AzCzTrz:2 wt%TBRb (30 nm)/TmPyPB (40 nm)/LiF (1 nm)/Al (EQE_{max}=18.6%)

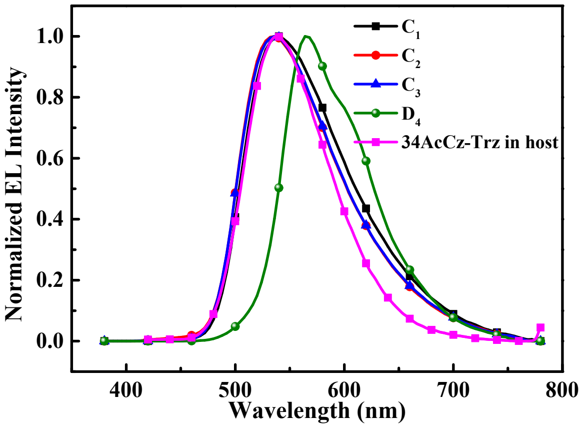


Fig S13. The comparison of the emission spectra with the EL spectra.

References

1 X. Lv, W. Zhang, D Ding, C Han, Z. Huang, S. Xiang, Q. Zhang, H. Xu and L. Wang, *Adv. Optical Mater.*, 2018, **6**, 1800165.

2 Q. Zhang, S. Sun, w. liu, P. Leng, X. Lv, Y. Wang, H. Chen, S. Ye, S. Zhuang and L. Wang, *J. Mater. Chem. C*, 2019, **7**, 9487-9495.