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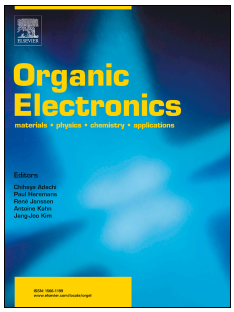

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Highly Efficient Green Phosphorescent Organic Light-Emitting Diodes based on Tetraphenyl Silicon Derivative Host Materials

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Abstract

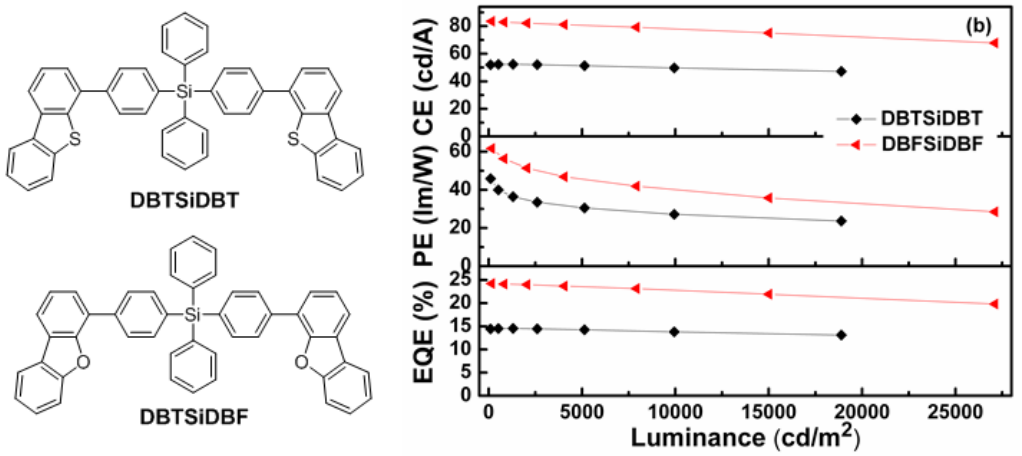
We report herein two novel tetraphenyl silicon derivatives, bis(4-(dibenzo[b,d]thiophen-4-yl)phenyl)diphenylsilane (**DBTSiDBT**) and bis(4-(dibenzo[b,d]furan-4-yl)phenyl)diphenylsilane (**DBFSiDBF**) achieved by coupling dibenzothiophene (DBT) and dibenzofuran (DBF) moieties with bis(4-bromophenyl)diphenylsilane. This linking strategy raises the triplet energy, glass transition temperature (*Tg*) and thermal stability of **DBTSiDBT** and **DBFSiDBF**.

Therefore, they can serve as hosts, producing phosphorescence at high efficiency in organic light-emitting diodes. Consequently, (2-phenylpyridine) iridium(III) ($Ir(ppy)_3$) based phosphorescence organic light-emitting diodes (PHOLEDs) with green emission were designed by applying **DBTSiDBT** and **DBFSiDBF** as host materials. The champion external quantum efficiency (EQE) and maximum luminance of the devices were 24.5% and over 27000 cd/m^2 respectively.

Keywords:

Dibenzothiophene, Dibenzofuran, Organosilicon derivatives, Host material, High triplet energy, Green phosphorescent organic light-emitting diode

Graphical



Highly Efficient Green Phosphorescent Organic Light-Emitting Diodes based on Tetraphenyl Silicon Derivative Host Materials

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Abstract: We report herein two novel tetraphenyl silicon derivatives, bis(4-(dibenzo[b,d]thiophen-4-yl)phenyl)diphenylsilane (**DBTSiDBT**) and bis(4-(dibenzo[b,d]furan-4-yl)phenyl)diphenylsilane (**DBFSiDBF**) achieved by coupling dibenzothiophene (DBT) and dibenzofuran (DBF) moieties with bis(4-bromophenyl)diphenylsilane. These two compounds show high triplet energy states, high glass transition temperature (*Tg*) and high decomposition temperature (*Td*). Therefore, they can serve as hosts in organic light-emitting diodes. Consequently, (2-phenylpyridine) iridium(III) ($Ir(ppy)_3$) based phosphorescence organic light-emitting diodes (PHOLEDs) with green emission were studied with **DBTSiDBT** and **DBFSiDBF** as host materials. The champion external quantum efficiency (EQE) and maximum luminance of the devices were 24.5% and over 27000 cd/m^2

respectively.

Keywords: Green phosphorescent organic light-emitting diode, High triplet energy, Host material, Organosilicon derivatives, Dibenzofuran, Dibenzothiophene.

1. Intruduction

Organic light emitting diodes (OLEDs) emerged about three decades ago as a new luminescent technology [1], and much progress has been achieved since then. OLEDs could be applied in display and lightening. Compared with traditional technologies, OLEDs have many advantages such as thin and flexible structure, wide view angle, fast response and decent transparency. Therefore, OLEDs can be used in various applications [2-8]. However, OLEDs are not stable for a prolonged period and this remains one of the problems that needs to be addressed to achieve a long device lifetime [9-11]. Within a typical fluorescence OLED, the ratio of singlet (*S*) and triplet (*T*) excitons is 1:3. Therefore, the internal quantum efficiency (IQE) does not exceed 25% and the maximum EQE is less than 5% despite its long-term stability. The discovery of phosphorescent emitters has facilitated the achievement of highly efficient OLEDs with the utilization of both singlet and triplet excitons [12]. To date, low energy PHOLEDs (e.g. orange and red PHOLEDs) have exhibited remarkable performances [13-16]. However, achieving high energy PHOLEDs (e.g. blue PHOLEDs) is still a challenge. The reasons are given as follows [9-11]: i) the availability of high efficiency and long lifetime phosphors is limited. The available conventional phosphors lack long-term stability. In addition, PHOLEDs may change their EL spectra with diffenrent driving voltage. ii) In order to match the energy levels of the blue and green phosphors, the host material needs to exhibit wide energy gap. This makes the preparation of such a material a big challenge. In addition, these kinds of host materials often possess deep highest occupied molecular orbital (HOMO) as well as shallow lowest unoccupied molecular orbital (LUMO). This increases injection barriers for electrons as well as holes. The device therefore needs much higher driving voltage which will further limit the application of PHOLEDs. Therefore, a long-lasting and high-efficiency OLED is still on the wish list with no satisfying solution in sight. Since host materials serve as the dispersion matrix for phosphor emitters, it is imperative to develop highly effecient host materials for practical application of PHOLEDs. To meet this requirement, some semi-empirical

principles have to be established. These include good thermal and morphological stability, appropriate HOMO/LUMO levels, optimal charge transport and high triplet energy [9-11]. The design protocol to raise the triplet energy of a host material includes: i) to incorporate the high triplet energy moieties e.g. arylsilane [17-21], dibenzofuran [22-29], dibenzothiophene [22,30-36], fluorene [37-41], and carbazole [42-44] into host material; ii) to restrain the conjugation length in the host material. Silicon [17-21], phosphine oxide [40] and sp^3 carbon [45] effectively interrupt intramolecular π-conjugation and are commonly adopted for this purpose. Also, sterically hindered structures as well as *ortho* or *meta* linking strategies can be used to minimize the conjugation length. This design principle has been applied over the years to develop many highly efficient host materials [22,39-44].

In this work, by linking a tetraphenyl silicon building block with dibenzothiophene (DBT) and dibenzofuran (DBF) units, two new host materials exhibiting elevated glass-transition temperature and triplet energy were prepared. The new host materials are bis(4-(dibenzo[*b*,*d*]thiophen-4-yl)phenyl)diphenylsilane (**DBTSiDBT**) and bis(4-(dibenzo[*b*,*d*]furan-4-yl)phenyl) diphenylsilane (**DBFSiDBF**). The silicon-bridge is capable of facilitating transfer of energy from host to guest for smaller steric hindrance. As result, highly efficient green PHOLEDs with (2-phenylpyridine) iridium(III) ($Ir(ppy)_3$) as emitter were realized using the novel host materials with maximum EQE of 24.5% and luminance over 27000 cd/m^2.

3. Results and Discussion

3.1 Syntheses and Characterization

Scheme 1 outlines the preparation steps for bis(4-(dibenzo[*b*,*d*]thiophen-4-yl) phenyl)diphenylsilane (**DBTSiDBT**) and bis(4-(dibenzo[*b*,*d*]furan-4-yl)phenyl) diphenylsilane (**DBFSiDBF**). 1,4-Dibromobenzene and dichlorodiphenylsilane were used as precursors for the synthesis of the intermediate, bis(4-bromophenyl)diphenylsilane. Finally, the target products were obtained in good yield by one step Suzuki−Miyaura coupling reaction with appropriate boronic acids of dibenzothiophene (DBT) and dibenzofuran (DBF).

3.2 Thermal Analyses

Thermogravimetric analysis (TGA) as well as differential scanning calorimetry (DSC) were conducted to determine the thermal characteristics of **DBTSiDBT** and

DBFSiDBF. **Figure 1** shows that the two compounds possess high thermal stability. The T_g was measured as 112 ^oC for **DBTSiDBT**, while **DBFSiDBF** shows slightly lower T_g of 102 ^oC. The T_g data for these materials show a slight difference which may be ascribed to the similarities in their molecular weight and molecular structure. **DBTSiDBT** and **DBFSiDBF** also showed T_d of 474 and 429 ^oC respectively at 5% weight loss. The T_ds exhibit a similar trend to their T_g data.

3.3 Optoelectronic Properties

Figure 3 presents the room temperature UV-vis absorption and PL spectra of **DBTSiDBT** and **DBFSiDBF** in dichloromethane as well as their phosphorescence (Phos) spectra at 77 K in 2-methyltetrahydrofuran (2-MeTHF). The detailed parameters are presented in **Table 1**. **DBTSiDBT** and **DBFSiDBF** exhibit similar absorption feature. Nonetheless, the maximum absorption wavelengths (Abs λ_{max}) as well as the absorption onset of DBT-based compounds are red-shifted by 16 nm with respect to those of the DBF-based analogue. Therefore, the band gap (E_g) of **DBTSiDBT** (3.59 eV) is narrower than that of **DBFSiDBF** (3.77 eV). On the other hand, **DBTSiDBT** and **DBFSiDBF** exhibited very similar PL characteristics. The similar and structureless PL curves might be due to their similar molecular configuration [22]. As shown in their Phos spectra, **DBTSiDBT** and **DBFSiDBF** exhibit well defined vibronic bands with the highest values at 464 nm and 460 nm and triplet energies (E_T) of 2.67 and 2.70 eV respectively. Due to their suitable and high levels of E_T, these compounds could serve as host materials in phosphorescent OLEDs.

3.4 DFT Simulation.

The electronic distributions of **DBTSiDBT** and **DBFSiDBF** were further elucidated by simulating their FMO spatial distributions through DFT calculations at a B3LYP/6-31G(d) level. **Figure 3** presents the spatial distributions of the highest and lowest frontier molecular orbitals of these host materials. These two materials exhibit similar distributions of LUMO and HOMO. The HOMOs of **DBTSiDBT** and **DBFSiDBF** are localized on the DBF and DBT unit, respectively, and the LUMOs spread across the molecular backbone.

3.5 FMO Energy Levels

To obtain energy levels of these materials, their HOMO levels were evaluated via

UPS analyses. **Figure 4** presents the UPS spectra of **DBTSiDBT** and **DBFSiDBF**. The HOMOs can be extracted using the equation; $E_{HOMO} = hv - E_k$, where, hv is photon energy (He I, hv = 21.22 eV) and E_k is kinetic energy of photo-electrons. LUMOs can be evaluated using equation; $E_{LUMO} = E_g + E_{HOMO}$, E_g is the optical energy gap. Therefore, levels of HOMO/LUMO of **DBTSiDBT** and **DBFSiDBF** are calculated to be -6.24/-2.65 and -6.45/-2.68 eV respectively.

3.6 Electroluminescent Properties

The electroluminescent (EL) characteristics of **DBTSiDBT** and **DBFSiDBF** were investigated by fabricating green phosphorescent OLEDs based on $Ir(ppy)_3$ with a structure of ITO/HAT-CN (10 nm)/TAPC (55 nm)/Host: 9 vol% $Ir(ppy)_3$ (20 nm)/TmPyPB (35 nm)/Liq (2 nm)/Al (120 nm). 9 wt% $Ir(ppy)_3$ were doped into **DBTSiDBT** and **DBFSiDBF** to achieve the emitting layer (EML). The corresponding molecular structures and energy diagrams are shown in **Figure 5**. The current density–voltage–luminance (*J*–V–L) characteristics; current efficiency (CE), power efficiency (PE) and external quantum efficiency (EQE) as a function of luminance and the EL spectra are shown in **Figure 6(a)**, **6(b)** and **6(c)**, respectively. **Table 2** presents the EL parameters.

As revealed in **Figure 6(a)**, the devices hosted by **DBTSiDBT** and **DBFSiDBF** exhibit relatively low turn-on voltages of 4.4 and 4.7 V at 1000 cd/m^2, respectively. The **DBTSiDBT** and **DBFSiDBF** based devices can achieve the maximum luminance of 19000 and 27000 cd/m^2, respectively. **Figure 6(b)** presents the efficiencies of green devices based on $Ir(ppy)_3$. For **DBFSiDBF** hosted device, high efficiency of 83.5 cd/A, 61.7 lm/W and 24.5% were realized for CE, PE and EQE respectively. **DBTSiDBT** based devices show relatively inferior maximum CE, PE and EQE of 52.0 cd/A, 45.8 lm/W and 14.4% respectively. Moreover, it is noteworthy that both devices exhibited flat efficiency roll-offs. At 1000 cd/m^2, devices based on **DBTSiDBT** showed an increase in EQE from 14.4% to a maximum value of 14.5%. At 10000 cd/m^2, the EQE retained a high value of 13.7%. At 1000 cd/m^2, the device based on **DBFSiDBF** demonstrated a reduction in EQE from the highest value 24.5% to 24.4%, and was able to retain 23.6% at 5000 cd/m^2. Notably, at 10000 cd/m^2, the EQE was only slightly reduced to 23.3%, which was still higher than the maximum EQE of 14.4% for devices based on **DBTSiDBT**. It is worth noting that the relatively good hole and electron transport properties of DBF-based host material (Figure 7) and

the higher E_T will favor the balanced charge transport and efficient energy transfer from host to dopant, which may account in part for its better EL performance. The green devices exhibit typical CIE coordinates of $Ir(ppy)_3$ based devices are (0.28, 0.64) at 5 mA/cm^2. It means that emission is originated from $Ir(ppy)_3$ and not from **DBTSiDBT** or **DBFSiDBF**. This could be attributed to the high E_Ts exhibited by these hosts which effectively confines emission within the dopant molecule.

4. Conclusion

In conclusion, two novel organosilicon derivatives, **DBTSiDBT** and **DBFSiDBF**, achieved by the attachment of DBT or DBF to bis(4-bromophenyl)diphenylsilane were designed, synthesized and characterized. Both compounds exhibited high triplet energy and good thermal stability due to the suitable linking strategy. The materials were applied as hosts for $Ir(ppy)_3$ emitter in green phosphorescence OLEDs with simple device structures. Green PHOLEDs based on **DBFSiDBF** achieved a maximum EQE of 24.5% with low efficiency roll-off. These results reveal the great potentials of tetraphenyl silicon derivatives and the possibility of subjecting them to extended study for high efficiency OLEDs.

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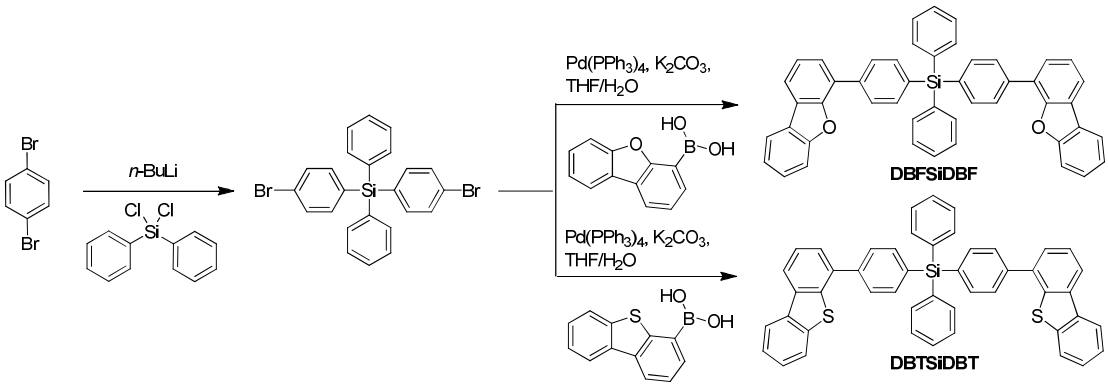

Scheme 1. Synthetic routes to **DBTSiDBT** and **DBFSiDBF**.

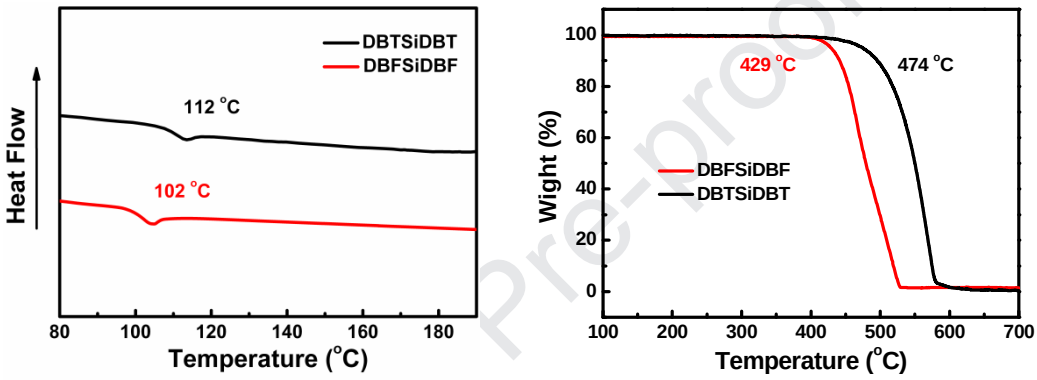

Figure 1. Differential scanning calorimetry (DSC, left) and thermogravimetric analyses (TGA, right) of **DBTSiDBT** and **DBFSiDBF**.

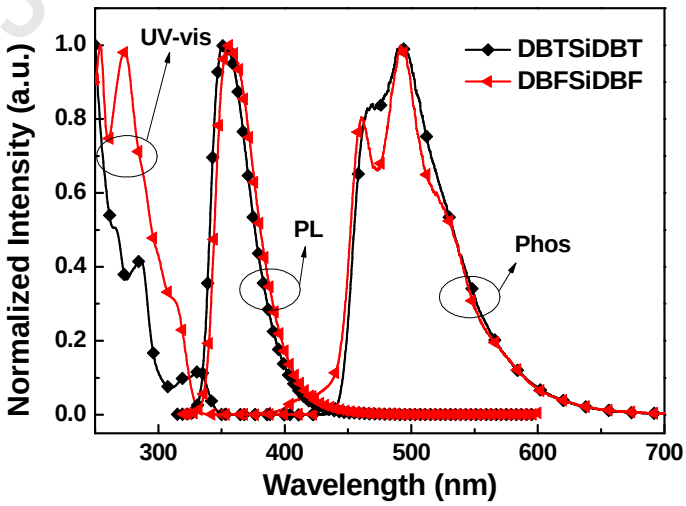

Figure 2. UV-Vis absorption, PL and Phos spectra of **DBTSiDBT** and **DBFSiDBF**.

Table 1. Physical Properties of **DBTSiDBT** and **DBFSiDBF**.

Host	Abs λ_{max}[a] /nm	PL λ_{max}[a] /nm	T_g[b]/ °C	T_d[c]/ °C	E_g[d]/ eV	E_T[e]/ eV	HOMO[f]/eV	LUMO[g]/eV
DBTSiDBT	285	355	112	474	3.61	2.67	-6.24	-2.65
DBFSiDBF	254	351	102	429	3.50	2.70	-6.45	-2.68

[a] Measured in toluene solution at room temperature. [b]T_g: Glass transition temperature. [c]T_d: Decomposition temperature. [d]E_g: Band gaps, calculated from the corresponding absorption onset. [e]E_T: Measured in 2-MeTHF glass matrix at 77 K. [f] HOMO levels, calculated from UPS data. [g] LUMO levels, calculated from the HOMO and E_g.

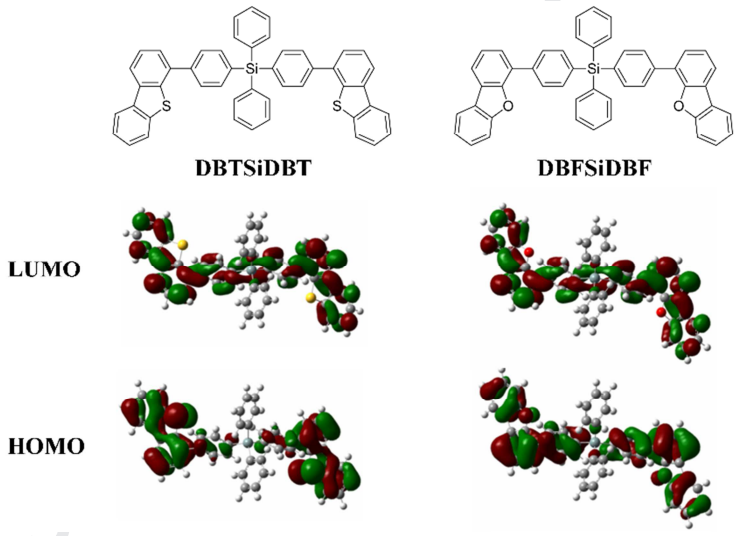

Figure 3. HOMO/LUMO distributions and molecular structure of **DBTSiDBT** and **DBFSiDBF**.

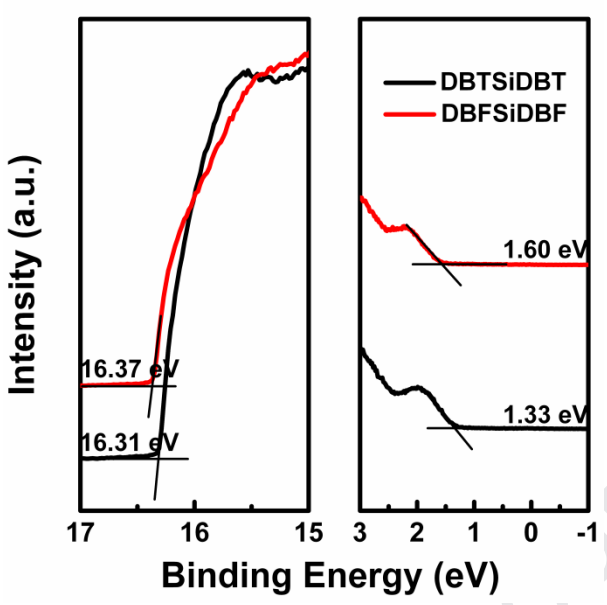

Figure 4. UPS spectra of **DBTSiDBT** and **DBFSiDBF**.

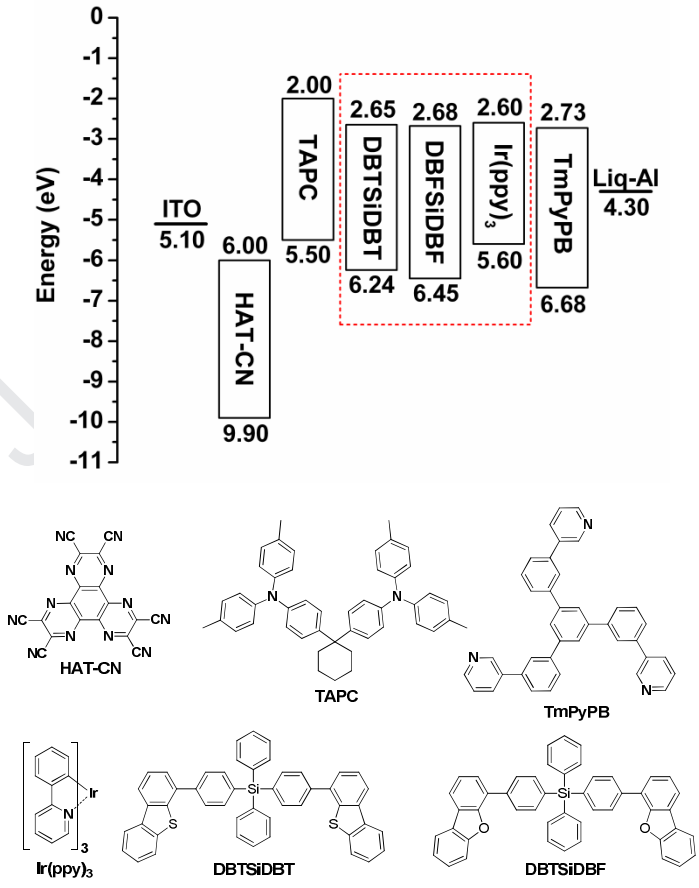

Figure 5. Molecular structures and representative energy level diagram of the materials.

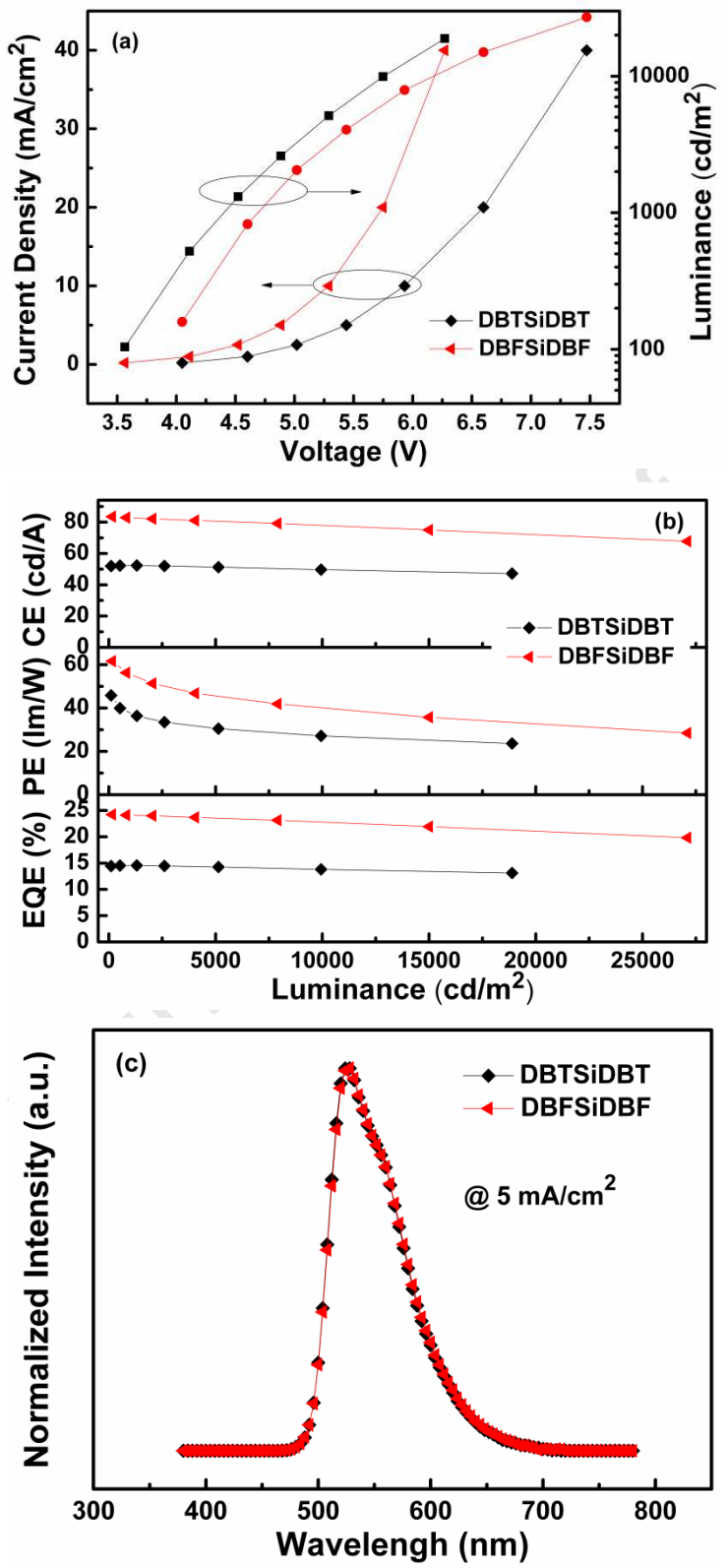

Figure 6. Current density–voltage–luminance (*J*–V–L) characteristics (a); current efficiency (CE), Current density–voltage–luminance (*J*–V–L) characteristics (a); current efficiency (CE), power efficiency (PE) and external quantum efficiency (EQE) versus luminance curves for $Ir(ppy)_3$-based devices (b) and EL spectra at 5 mA/cm^2 (c).

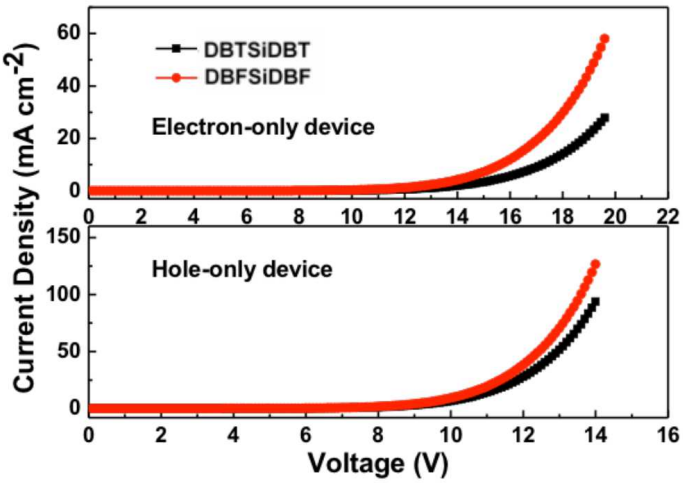

Figure 7. *J*–V characteristics of hole- and electron-only devices. Hole-only devices: ITO/ MoO_3 (10 nm)/ **Host** (100 nm)/ MoO_3 (10 nm)/ Al (100 nm); electron-only devices: ITO/ TmPyPB (20 nm)/ **Host** (100 nm)/ TmPyPB (20 nm)/ Liq (2 nm)/ Al (100 nm).

Table 2. Electroluminescence characteristics of the devices

Device[a]	Host	V[b]	η_{CE}[c]	η_{PE}[c]	EQE[c]	CIE[d]
		[V]	[cd A^{-1}]	[lm W^{-1}]	[%]	[x, y]
1	DBTSiDBT	4.4	53.4, 53.4, 50.0	45.8, 37.7, 28.4	14.5, 14.5, 13.7	0.28, 0.64
2	DBFSiDBF	4.7	83.5, 82.9, 78.0	61.7, 56.1, 40.9	24.5, 24.4, 23.2	0.28, 0.64

[a] The notations 1 and 2 in devices indicate the devices fabricated with DBTSiDBT and DBFSiDBF as the hosts respectively. Device configuration: ITO/HAT-CN (10 nm)/TAPC (55 nm)/Host: 9% $Ir(ppy)_3$ (20 nm)/TmPyPB (35 nm)/Liq (2 nm)/Al (120 nm). [b] Voltages at 1000 cd/m^2, [c] Efficiencies in the order of the maxima at 1000 cd/m^2 and at 10000 cd/m^2, [d] Commission International de Ieelaiage coordinates measured at 5 mA/cm^2.

- Two novel organosilicon derivatives, DBTSiDBT and DBFSiDBF, were designed, synthesized, and applied as host materials in green PHOLEDs.
- Both materials showed good thermal stability and high triplet energy.
- The $Ir(ppy)_3$ based PHOLED hosted by DBFSiDBF exhibited relatively high EQE of 24.5% with lower efficiency roll-off.

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: