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## Introduction

Organic light-emitting diodes (OLEDs) have attracted considerable scientific and industrial interest since the pioneering work by Tang et al. in 1987.<sup>1</sup> Generally, phosphorescent OLEDs (PhOLEDs), in which the emitting layers (EML) are normally constructed by doping the phosphors into charge-transport hosts, have much higher electroluminescence (EL) efficiency than the conventional fluorescent OLEDs, due to the radiative harvesting of both electrogenerated singlet and triplet excitons.<sup>2,3</sup> Recently, green and red PhOLEDs with 100% internal quantum efficiency have been reported,4-7 but high performance blue PhOLEDs have not yet been achieved, which are subject to a sky-blue emission with the Commission International de I' Eclarirage (CIE) coordinates of *x* and/or  $y \ge 0.20$  and their instability in the emission color and EL efficiency.8-12 It has become a bottleneck in the development of efficient OLEDs in both flat-panel displays and solid-state lighting. Furthermore, the approximate 0.5-1.0 eV exchange energy losses in power efficiency resulting from the energetic relaxation

E-mail: yuliu@jlu.edu.cn, zuolun\_zhang@yahoo.com.cn

## A novel tetraphenylsilane-phenanthroimidazole hybrid host material for highly efficient blue fluorescent, green and red phosphorescent OLEDs<sup>+</sup>

Dong Liu, Mingxu Du, Dong Chen, Kaiqi Ye, Zuolun Zhang,\* Yu Liu\* and Yue Wang

A novel organosilane compound, bis(4-(1-phenylphenanthro[9,10-*d*]imidazol-2-yl)phenyl)diphenylsilane (Si(PPI)<sub>2</sub>), has been designed and synthesized. It has a high thermal decomposition temperature of 528 °C and is able to form an amorphous glass with a high glass-transition temperature of 178 °C. In addition, it possesses high singlet and triplet energies and displays efficient energy transfer to the selected blue fluorescent and green and red phosphorescent dopants when used as a host material. Electrochemical measurements and single-carrier devices indicate that Si(PPI)<sub>2</sub> is a bipolar transport material, allowing the injection and transport of both electrons and holes. By using (Si(PPI)<sub>2</sub>) as a host, high-performance fluorescent blue (FB) and phosphorescent green (PG) and red (PR) OLEDs with a uniform and simple device configuration have been achieved. These OLEDs exhibit very high peak external quantum efficiency (EQE) and peak power efficiency (PE), *i.e.* 6.1% and 8.0 Im W<sup>-1</sup> for FB, 19.2% and 51.1 Im W<sup>-1</sup> for PG and 12.0% and 15.6 Im W<sup>-1</sup> for PR. Moreover, the high-level EQE of 4.0, 19.1 and 10.6% and PE of 3.0, 41.6 and 7.5 Im W<sup>-1</sup> can be maintained by FB, PG and PR, respectively, at the practical luminance of 100 cd m<sup>-2</sup>. Furthermore, the emission colors of these OLEDs remain almost unchanged within the whole range of driving voltages. Importantly, the blue OLED displays a pure blue emission (CIE: 0.18, 0.17).

following intersystem crossing from the conductive host into the blue emissive triplet state represent another drawback.<sup>13</sup> Considering these issues regarding the blue PhOLEDs, adopting the stable blue fluorescent emitters to realize high-quality blue EL emission is a complementary effective strategy and becomes again an active research area.<sup>14-17</sup> On the other hand, blue, green and red emitters often require different hosts to achieve highly efficient energy transfer from host to emitter.<sup>18-22</sup> Therefore, the full-color and white OLEDs were often established based on a complex material system and, correspondingly, highcost organic syntheses. In this sense, achieving high performance blue, green and red electroluminescence through a simple material system with the aim to reduce the production cost of materials and simplify the manufacturing process is an important issue for OLED applications. Given this, many efficient multi-color and white PhOLEDs based on a universal host have been achieved in the literature.<sup>23–26</sup> However, the design and synthesis of an organic molecule that can be employed as a host to realize the efficient blue fluorescent OLEDs as well as phosphorescent OLEDs with a relatively long-wavelength light, such as green and red, has not been reported previously, and is still a considerable challenge.

Organic derivatives and metal complexes of phenanthroimidazole (PPI) have been widely utilized as blue fluorescent emitters



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State Key Laboratory of Supramolecular Structure and Materials,

College of Chemistry, Jilin University, Changchun 130012, P. R. China.

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and/or the host materials for highly efficient OLEDs,<sup>27-32</sup> due to their high-energy emission and bipolar transport properties. Additionally, tetraarylsilane compounds have attracted a fair amount of attention as organic EL materials, attributed to their wide bandgap and the high thermal stability resulting from their non-conjugated tetrahedral molecular geometry.<sup>25,33-37</sup> In this contribution, by integrating one tetraphenylsilane and two PPI groups, we have prepared bis(4-(1-phenylphenanthro[9,10-d]imidazol-2-yl)phenyl)diphenylsilane (Si(PPI)<sub>2</sub>) with the expectation that this compound will have good thermal stability, high-energy emission and good carrier-transport ability to work as a high-performance host material. We have found that, through doping of fluorescent blue or phosphorescent green or red emitters into Si(PPI)<sub>2</sub>, a series of host-guest systems with efficient energy transfer can be realized. They showed very high peak external quantum efficiency (EQE) and power efficiency (PE) values of 5.6% and 8.0  $\text{Im W}^{-1}$ for blue emission with the CIE coordinates of (0.18, 0.17), which are among the highest EL efficiencies ever reported for the pure blue fluorescent OLEDs that meet the standard of the  $CIE_{x,y}$ coordinates of  $x \le 0.20$  and  $y \le 0.20$ , as well as 18.8% and 51.1  $\text{Im W}^{-1}$  for green emission and 11.9% and 15.6  $\text{Im W}^{-1}$  for red emission. They maintain high levels of 3.6, 18.7 and 10.5% and 3.0, 41.6 and 7.5  $\text{Im W}^{-1}$  at the practical luminance of 100 cd m<sup>-2</sup> respectively, and their EL emission color remains almost unchanged within the whole range of the corresponding driving voltages.

We will present a comprehensive investigation on  $Si(PPI)_2$ , which encompasses not only the conventional characterization for its thermal, photophysical and electrochemical properties, but also the emphatically studies on its carrier injection/transport abilities and EL characteristics as host material.

### Experimental section

#### General information

Auxiliary materials, such as 1,4-bis[(1-naphthylphenyl) amino]biphenyl (NPB), 4,4',4"-tri(N-carbazolyl) triphenylamine (TCTA), 1,3,5-tris(N-phenylbenzimidazol-2-yl)benzene (TPBi), and the series of emitting fluorescent/phosphorescent materials including An(PPI)<sub>2</sub>,<sup>38</sup> Ir(ppy)<sub>3</sub>,<sup>3</sup> and (bt)<sub>2</sub>Ir(dipba)<sup>39</sup> (Fig. 5) for fabricating the OLEDs were prepared according to literature procedures and purified further by vacuum sublimation prior to use. Starting materials for the synthesis were obtained from commercial suppliers and used without further purification. Anhydrous THF was distilled with sodium benzophenone ketyl under a nitrogen atmosphere and degassed using the freeze-pump-thaw method. All glassware, syringes, magnetic stirring bars and needles were dried in a convection oven for at least 4 hours. Reactions were monitored using thin layer chromatography (TLC). Commercial TLC plates (Silica gel 60 F254, Merck Co.) were developed and the spots were seen under UV light at 254 and 365 nm. Silica column chromatography was done using silica gel 60 G (particle size 5-40 µm, Merck Co.). <sup>1</sup>H NMR spectra were recorded on a Bruker AVANCE 300 MHz spectrometer with tetramethylsilane as the internal standard. Mass spectra were measured on a

GC/MS system. Elemental analyses were performed on a flash EA 1112 elemental analyzer. IR spectra were recorded on a Bruker VERTEX 80v spectrometer. UV-vis absorption spectra were obtained using a Shimadzu UV-2550 UV-vis spectrometer. Photoluminescence (PL) spectra were recorded on a Perkin-Elmer LS-55 fluorescence spectrometer using a Xe arc lamp excitation source. Solid state PL efficiencies were measured using an integrating sphere (C-701, Labsphere Inc.), using a 365 nm Ocean Optics LLS-LED as the excitation source, and the light was introduced into the integrating sphere through an optical fiber. Time-resolved fluorescence spectra were collected on an Edinburgh mini- $\tau$  fluorescence lifetime spectrometer, using an Edinburgh EPL-405 picosecond pulsed diode laser as the excitation source. Electrochemical measurements were performed using a BAS 100 W Bioanalytical electrochemical workstation, using a Pt disk as the working electrode, platinum wire as the auxiliary electrode, and a porous glass wick Ag/Ag<sup>+</sup> as the pseudo-reference electrode, standardized against ferrocene/ ferrocenium. A 0.1 M solution of n-Bu<sub>4</sub>NPF<sub>6</sub> in CH<sub>2</sub>Cl<sub>2</sub> (for oxidation) or DMF (for reduction) was used as the supporting electrolyte. A scan rate of 100 mV s<sup>-1</sup> was adopted. All the redox potentials are given against ferrocene/ferrocenium.

#### Fabrication of the OLEDs and EL measurements

The general architecture of the complex multilayer diodes used in this study is as follows: the ITO (indium-tin oxide) coated glass substrates (20  $\Omega$  square<sup>-1</sup>) were pre-cleaned carefully in ethanol, followed by washing in acetone in a soap ultrasonic bath and treated by UV/O3 for 2 min. The devices were prepared in a vacuum at a base pressure of around 5 imes 10<sup>-6</sup> Torr. All organics were thermally evaporated at a rate of 1.0 Å  $s^{-1}$  at a base pressure of around  $3.5 \times 10^{-4}$  Pa. A LiF layer (0.5 nm) was deposited at a rate of 0.2 Å  $s^{-1}$ . The finishing Al electrode (cathode) was deposited at a rate of 10 Å  $s^{-1}$  in another chamber. The thicknesses of the organic materials and the cathode layers were controlled using a quartz crystal thickness monitor. The electrical characteristics of the devices were measured using a Keithley 2400 source meter. The EL spectra and luminance of the devices were obtained on a PR650 spectrometer. All the device fabrication and device characterization steps were carried out at room temperature under ambient laboratory conditions. Current-voltage characteristics of single-carrier devices were measured using the same semiconductor parameter analyzer as for OLED devices.

#### Synthesis

The synthetic route to  $Si(PPI)_2$  is shown in Scheme 1.

Di(4-formylphenyl)-diphenylsilane<sup>40</sup>. *n*-BuLi (1.6 M in hexane, 20 mL, 32 mmol) was added slowly to a THF (120 mL) solution of 4-bromobenzaldehyde dimethyl acetal (7.40 g, 32 mmol) at -78 °C, and then the mixture was stirred at this temperature for 1 h. After dichlorodiphenylsilane (1.52 mL, 12.5 mmol) was added, the reaction mixture was stirred at -78 °C for 1 h, slowly warmed to r.t., stirred for 12 h at r.t., quenched with 2 N HCl and extracted with ether. The organic phase was washed with brine and dried over MgSO<sub>4</sub>, and then the solvent was evaporated under reduced



pressure. To the obtained residue, acetic acid (10 mL) and H<sub>2</sub>O (3 mL) were added, and the obtained mixture was stirred for 3 h at r.t., poured into saturated aqueous NaHCO<sub>3</sub> and extracted with ether. The extracts were washed with brine, dried over MgSO<sub>4</sub>, filtered and concentrated under vacuum. The obtained residue was purified using column chromatography (silica gel, hexanes/EtOAc 4:1) to give the product as a white solid (2.85 g, 58%). <sup>1</sup>H NMR (300 MHz; CDCl<sub>3</sub>; Me<sub>4</sub>Si):  $\delta$  10.01 (s, 2H), 7.89 (d, *J* = 8.0 Hz, 4H), 7.74 (d, *J* = 8.0 Hz, 4H), 7.56–7.40 (m, 10H).

Bis(4-(1-phenylphenanthro[9,10-d]imidazol-2-yl)phenyl) diphenylsilane (Si(PPI)<sub>2</sub>). A mixture of 9,10-phenanthrenequinone (312 mg, 1.5 mmol), di(4-formylphenyl)-diphenylsilane (294 mg, 0.75 mmol), aniline (0.55 mL), ammonium acetate (1.16 g, 15 mmol) and glacial acetic acid (30 mL) was refluxed for 24 h. After being cooled to room temperature, the reaction mixture was poured into stirred methanol. The precipitate was filtered, washed with methanol and dried to give the crude product, which was further purified by vacuum sublimation to provide the desired compound as a white powder (414 mg, 60%). <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>, ppm):  $\delta$  8.89 (d, J = 7.2 Hz, 2H), 8.77 (d, J = 8.4 Hz, 2H), 8.71 (d, J = 8.1 Hz, 2H), 7.75 (t, J = 7.5 Hz, 2H), 7.69-7.61 (m, 12H), 7.56-7.43 (m, 16H), 7.37 (t, J = 7.2 Hz, 4H), 7.29–7.24 (m, 2H), 7.17–7.14 (d, J = 8.1 Hz, 2H). <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>, ppm):  $\delta$  120.87, 122.84, 123.07, 124.09, 124.93, 125.66, 126.25, 127.15, 127.27, 127.91, 128.15, 128.33, 128.38, 128.48, 128.60, 128.67, 128.88, 129.07, 129.11, 129.38, 129.42, 129.53, 129.74, 129.95, 130.09, 130.20, 133.03, 133.53, 136.19, 136.38, 137.96, 138.71, 138.77. Ms m/z: 919.9 [M]<sup>+</sup> (calcd: 920.3). Anal. calcd (%) for C<sub>66</sub>H<sub>44</sub>N<sub>4</sub>Si: C, 86.05; H, 4.81; N, 6.08. Found: C, 86.38; H, 4.73; N, 6.05. IR (KBr,  $\nu$ , cm<sup>-1</sup>):  $\nu$  = 3092, 3062, 3010, 1599, 1486, 1455, 1425, 1378, 1296, 1234, 1183, 1152, 1106, 1019, 957, 839, 804, 752, 696, 568, 506.

## Results and discussion

#### Thermal properties

The thermal properties of  $Si(PPI)_2$  were investigated using thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) under a nitrogen atmosphere at a heating rate of 10 °C min<sup>-1</sup>. The compound exhibits very good thermal stability, as evidenced by its high decomposition temperature (corresponding to 5% weight loss) of 528 °C in the TGA thermogram (Fig. 1). This will prevent the compound from decomposing during the processes of vacuum deposition and





device operation. According to the DSC analysis, Si(PPI)<sub>2</sub> has a high melting point of 385  $^{\circ}$ C (Fig. 1, inset). More importantly, it has the ability to form an amorphous glass with a high glass-transition temperature of 178  $^{\circ}$ C during the second-heating scan, which is beneficial for the formation of homogeneous and amorphous films upon thermal evaporation, as well as to keep the stability of the film and to decrease the phase separation of the host–guest system when Si(PPI)<sub>2</sub> is used as a host material.

#### Photophysical properties

Fig. 2a displays the UV-vis absorption and photoluminescence (PL) spectra of Si(PPI)<sub>2</sub> in dichloromethane solution  $(5 \times 10^{-5} \text{ M})$ at room temperature (298 K) and its phosphorescence spectrum in frozen 2-MeTHF (5  $\times$  10<sup>-5</sup> M) at 77 K. Si(PPI)<sub>2</sub> exhibits intense deep-blue emissions in dichloromethane solution as well as in the solid film.  $An(PPI)_2$  is considered to be an appropriate blue dopant for our system because of its preferred pure-blue fluorescence and, importantly, a large overlap between its absorption spectrum and the solid-state emission spectrum of Si(PPI)<sub>2</sub>, which may lead to efficient energy transfer from Si(PPI)<sub>2</sub> to  $An(PPI)_2$ . In the low-temperature (77 K) emission spectrum of Si(PPI)<sub>2</sub> (Fig. 2a), a phosphorescence band showing vibrational structures can be observed. By using the highest-energy vibronic sub-band, the triplet energy  $(E_{\rm T})$  of Si(PPI)<sub>2</sub> is estimated to be 2.55 eV, which is clearly lower than that of the classical blue phosphor bis[2-(4,6-difluorophenyl)pyridinato-N,C2'](picolinato)iridium(m) (FIrpic) (~2.65 eV)<sup>12</sup> and is not high enough to sensitize this blue phosphor, leading to incomplete host-dopant energy transfer accompanied by the poor EL performance (see the ESI<sup> $\dagger$ </sup>). However, this  $E_{T}$  level of Si(PPI)<sub>2</sub> is sufficiently higher than those of most green and red phosphorescence emitters. This result suggests that, besides the singlet-singlet (Förster and/or Dexter) energy transfer, a triplet-triplet Dexter energy transfer can be expected when doping appropriate green and red emitters into Si(PPI)2. The classical green phosphor Ir(ppy)<sub>3</sub> and a high-performance red phosphor (bt)<sub>2</sub>Ir(dipba) were selected as the phosphorescent dopants in this study. The PL spectra of the thin films prepared by doping respectively



**Fig. 2** (a) UV-vis absorption and PL spectra of Si(PPI)<sub>2</sub> (H) and An(PPI)<sub>2</sub> (B) measured in dichloromethane solution (5 × 10<sup>-5</sup> M) at room temperature and the phosphorescence spectrum of Si(PPI)<sub>2</sub> in 2-MeTHF solution (5 × 10<sup>-5</sup> M) at 77 K. (b) PL spectra of the neat films of Si(PPI)<sub>2</sub> (H), blue fluorophore (B), green (G) and red (R) phosphors and these dopant emitters doped in Si(PPI)<sub>2</sub> (H) thin films at 10 wt% doping concentration.

10 wt% of the selected blue, green and red dopants into Si(PPI)<sub>2</sub> are measured to confirm the efficient energy transfer. As can be seen from Fig. 2b, all these doped films show an emission very close to those of the neat-films of the respective dopants, while the emission of Si(PPI)<sub>2</sub> cannot be observed. They exhibit the PL quantum efficiency values of  $8 \pm 1\%$ ,  $96 \pm 3\%$  and  $24 \pm 2\%$  respectively, and their corresponding transient PL decay curves show almost single-exponential decay with the lifetimes of 3 ns, 0.97 µs and 0.71 µs at room temperature (see ESI† file), indicating the complete energy transfer from the host (Si(PPI)<sub>2</sub>) to the three dopants. Therefore, these host–dopant systems are expected to have the potential to realize high-performance OLED devices. The slightly red-shifted emission from the doped films to the neat films of corresponding dopants should be related to the tighter packing between the dopant molecules in the neat films.

#### Electrochemical properties and theoretical calculations

Cyclic voltammetry (CV) was performed to investigate the electrochemical properties of  $Si(PPI)_2$  (Fig. 3a). The HOMO

and LUMO energy levels were estimated to be *ca.* -5.6 eV and -2.2 eV respectively from the onsets of the oxidation and reduction potentials with regard to the energy level of ferrocene ( $-4.8 \text{ eV}^{41}$  below vacuum). This HOMO level is much higher than those ( $\sim -7.0 \text{ eV}$ ) of previously reported tetraarylsilane-based series of UGH host materials,<sup>33</sup> reflecting a better hole-injection property of Si(PPI)<sub>2</sub> due to the derivatization by the PPI group. Moreover, the HOMO level of Si(PPI)<sub>2</sub> is even higher than that of the classic fluorescent host material 4,4'-N,N'-dicarbazolylbiphenyl (CBP) ( $\sim -6.0 \text{ eV}$ ) and matches well with the most widely used hole-transporting material NPB ( $\sim -5.4 \text{ eV}$ ) and electron-blocking material TCTA (-5.7 eV), implying little or no hole-injection barrier from NPB or TCTA to this compound.

To gain more insight into the electronic structure of  $Si(PPI)_2$ , DFT calculations (B3LYP/6-31G(d)) were carried out. The HOMO



Fig. 3 (a) Cyclic voltammogram of  $Si(PPI)_2$  in  $CH_2Cl_2$  for oxidation and DMF for reduction. (b) Contour plots of HOMO and LUMO in  $Si(PPI)_2$  using DFT calculations.

is mainly distributed on one of the two phenanthroimidazole groups, with a small contribution from the  $C_6H_4$  group attached to it while the LUMO is localized partly on the other phenanthroimidazole group and mainly on the C<sub>6</sub>H<sub>4</sub> group between this phenanthroimidazole group and the Si atom (Fig. 3b). The almost complete spatial separation of HOMO and LUMO energy levels suggests that the HOMO-LUMO excitation would shift the electron density distribution from one side of the  $Si(PPI)_2$  molecule as the donor to the other side as the acceptor, leading to a polarized excited state. It is more important that such separation of HOMO and LUMO can provide hole- and electron-transporting channels respectively, where holes and electrons can realize intermolecular hopping smoothly along their respective conducting pathways. Thus, this is likely to result in this compound possessing bipolar charge transporting ability.42-46

#### Charge carrier injection and transport properties

To further understand both hole and electron injection/ transport properties of Si(PPI)<sub>2</sub>, two single-carrier devices were fabricated using the following configurations: ITO/NPB (10 nm)/ Si(PPI)<sub>2</sub> (30 nm)/NPB (10 nm)/Al (100 nm) (hole-only device) and ITO/TPBi (10 nm)/Si(PPI)2 (30 nm)/TPBi (10 nm)/LiF (1 nm)/Al (100 nm) (electron-only device). NPB and TPBi layers are used to prevent electron and hole injection from the cathode and anode, respectively.<sup>47-49</sup> Fig. 4 shows the current density versus voltage curves of both devices, respectively. It is obvious that both devices can conduct significant current, indicating that  $Si(PPI)_2$  is capable of transporting both electrons and holes, and exhibits its bipolar transporting nature, which is effective for balancing holes and electrons in the emitting layer (EML) based on Si(PPI)<sub>2</sub>. The substantially lower electron current density than the hole curve at the same voltage level should be attributed to the high-lying LUMO energy level of Si(PPI)<sub>2</sub>, which means a certain injection barrier ( $\sim 0.5$  eV) from ETL.32

#### Characterization of fluorescent/phosphorescent OLEDs

According to the above studies, Si(PPI)<sub>2</sub> could be a good host material for both fluorescent and phosphorescent OLEDs. To evaluate its practical utility, a series of OLEDs with a uniform and simple configuration of [ITO/NPB (30 nm)/TCTA (5 nm)/ EML (25 nm)/TPBi (20 nm)/LiF (1 nm)/Al (100 nm)] were fabricated, and the energy diagram of the materials used in the EL devices is shown in Fig. 5. NPB was used as the holetransporting material (HTL), TCTA was used as the triplet exciton-blocking layer in the PhOLEDs, and TPBi was used as the electron transport/hole-blocking layer (ETL/HBL). The films of An(PPI)<sub>2</sub>,  $Ir(ppy)_3$  and  $(bt)_2Ir(dipba)$  doped in Si(PPI)<sub>2</sub> at the concentration of 10 wt% were adopted as the EMLs to fabricate the fluorescent blue device FB, the phosphorescent green and red devices PG and PR respectively. The device FB exhibits pure blue emissions (CIE: 0.18, 0.17) with the emission maximum at 452 nm at the luminance of 1000 cd  $m^{-2}$  (Fig. 6), which remains almost unchanged over a wide range of driving voltages from 3.5 V to 12 V. The EL spectrum is consistent with the PL spectrum of  $An(PPI)_2$ , suggesting that the blue EL emission results from the intrinsic emission of An(PPI)<sub>2</sub>. On the other hand, the devices PG and PR present bright green and saturated red emissions with the maximum peaks at 516 and 612 nm and  $CIE_{x,y}$  coordinates of (0.32, 0.61) and (0.64, 0.36) respectively at the luminance of 1000 cd  $m^{-2}$ , and no additional emission from the host was observed.

The current density–voltage–luminance (*J–V–L*) characteristics of these OLEDs are shown in Fig. 7a. All these devices displayed low turn-on voltages of 3.1–3.4 V (Table 1), and the corresponding current density and luminance exhibited sustained increase with increasing the driving voltage. The driving voltages at the practical luminance of 100 cd m<sup>-2</sup> are 5.6 V, 4.7 and 6.2 V, respectively. At driving voltages of 12.6, 9.1 and 13.5 V for FB, PG and PR respectively, very high luminance values of >10000 cd m<sup>-2</sup> were obtained. The quickly increasing luminance and current density indicate a good carrier



Fig. 4 Current density *versus* voltage characteristics of the hole-only and electron-only devices for Si(PPI)<sub>2</sub>.



Fig. 5 Energy level diagram of the materials used and the chemical structures of the emitting materials.



Fig. 6 EL spectra of devices FB, PG and PR at the luminance of 1000 cd  $m^{-2}$ 



**Fig. 7** Current density–voltage–luminance (J-V-L) curves (a) and power efficiency (PE)–luminance–external quantum efficiency (EQE) curves (b) of devices FB, PG and PR.

injection and transport property of the host  $(Si(PPI)_2)$ , which is consistent with our results obtained from the single-carrier devices given above. The external quantum efficiency (EQE) and

Table 1 Electroluminescence properties of the devices<sup>a</sup>

Device	V <sub>on</sub> /V	$L_{ m max}/ m cd\ m^{-2}$ (V at $L_{ m max}$ )	EQE <sup>b</sup> /%	PE <sup>b</sup> / lm W <sup>-1</sup>	EL emission peak, CIE $(x,y)^c$
FB	3.1	15000	6.1, 4.0, 1.9	8.0, 3.0, 0 9	452, (0.18, 0.17)
PG	3.4	60 470 (12 5)	19.2, 19.1,	51.1, 41.6,	516, (0.22, 0.61)
PR	3.3	(12.5) 12 330 (14.0)	17.8 12.0, 10.6, 7.3	26.5 15.6, 7.5, 3.3	(0.32, 0.61) 612, (0.64, 0.36)

<sup>*a*</sup> Abbreviation:  $V_{on}$ : turn-on voltage (recorded at 1 cd m<sup>-2</sup>).  $L_{max}$ : maximum luminance. EQE: external quantum efficiency. PE: power efficiency. <sup>*b*</sup> In the order of maximum, then values at 100 and 1000 cd m<sup>-2</sup>. <sup>*c*</sup> Measured at 1000 cd m<sup>-2</sup>.

power efficiency (PE) of these devices plotted with respect to the luminance are shown in Fig. 7b, and the EL performance data are summarized in Table 1. The blue device FB exhibited the maximum EQE and PE of 6.1% and 8.0  $\text{lm W}^{-1}$  respectively. To the best of our knowledge, these values are among the highest EL efficiencies ever reported for the pure blue fluorescent OLEDs that meet the standard of the  $CIE_{x,y}$  coordinates of  $x \le 0.20$  and  $y \le 0.20$ .<sup>50–56</sup> Notably, the EQE and PE values of FB at the brightness of 100 cd  $m^{-2}$ , which is the practical level used in the fields of display or lighting, remain as high as 4.0% and 3.0  $\text{Im W}^{-1}$  respectively. They are much higher than those of the reference devices adopting the conventional host materials CBP or N,N'-dicarbazolyl-3,5-benzene (mCP) with the low peak EQE and PE < 1% and 2 lm W<sup>-1</sup> (see the ESI<sup>†</sup> file). Furthermore, the green and red PhOLEDs PG and PR exhibited rather high peak EL efficiency values of 19.2, 12.0% for EQE and 51.1, 15.6  $\text{lm W}^{-1}$  for PE. Although these EL efficiencies showed a certain roll-off, the EQE values maintained the levels of 17.8% for PG and 7.3% for PR at a luminance of 1000 cd  $m^{-2}$ , and were as high as 14.2, 4.6% even when the luminance reached as high as 10000 cd m<sup>-2</sup>. Obviously, in terms of the performance, these two devices are comparable with the highly efficient green and red PhOLEDs reported to date.<sup>4-7</sup> Such high and stable EL performance should be ascribed to the balanced carrier injection/transportability of Si(PPI)2 as implied in single-carrier devices, which result in a broad distribution of recombination regions within the corresponding EMLs,<sup>57,58</sup> and correspondingly, a low probability of triplet-triplet annihilation that usually causes an efficiency roll-off at high current densities for the PhOLEDs. Charge confinement in the EML is another key factor for the high EL efficiency. There is a large energy barrier of 0.6 eV for hole leakage from Si(PPI)2 to TPBi together with the unipolar property of NPB for electron leakage from Si(PPI)2 to NPB, respectively. Therefore, holes and electrons can be effectively confined inside the EML, being crucial in achieving high efficiency and low roll-off OLEDs. Besides, on the premise that the  $E_{\rm T}$  (~2.55 eV) of (Si(PPI)<sub>2</sub>) is high enough for working as a host, but not much higher than those of the green (Ir(ppy)<sub>3</sub>,  $E_{T}$ : ~2.4 eV) and red ((bt)<sub>2</sub>Ir(dipba),  $E_{T}$ : ~2.1 eV) phosphorescent dopants, the energy loss during the host-todopant energy transfer process can be reduced as far as possible. It is notable that the excellent performances of all our blue, green and red OLEDs were obtained from the same device configuration

through adopting the same host material. The simple material system and the easily controlled fabrication process are of significance and importance for reducing the cost and enhancing the process stability in commercial mass production.

## Conclusions

In summary, we have developed a novel tetraphenylsilanephenanthroimidazole hybrid compound, Si(PPI)<sub>2</sub>, which can act as a host that realizes the highly efficient blue fluorescent OLEDs as well as green and red phosphorescent OLEDs by adopting a uniform and simple device configuration. The pureblue device with the CIE coordinates of (0.18, 0.17) exhibited the maximum EQE and PE values of 6.1% and 8.0  $\text{Im W}^{-1}$ , respectively, which are among the highest EL efficiencies ever reported for pure-blue devices. Furthermore, the EL performance of our green and red phosphorescent devices is also comparable with that of the most efficient green and red phosphorescent OLEDs reported to date. The high device performance is due to the high thermal stability, wide bandgap, efficient energy transfer and the balanced carrier-transport abilities of Si(PPI)<sub>2</sub>. Our results indicate that Si(PPI)<sub>2</sub> is a rare and excellent common host, which may have abilities to simplify the fabrication process and to reduce the material cost in commercial mass production of high-performance multi-color OLEDs. Moreover,  $Si(PPI)_2$  will also enable us to design the structurally simple three-primary-color fluorescent/phosphorescent hybrid white OLEDs, which are important and attractive for full-color display and white lighting. Further application of Si(PPI)<sub>2</sub> in white OLEDs is ongoing in our lab.

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