# Distinct electroluminescent properties of triphenylamine derivatives in blue organic light-emitting devices

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Two highly fluorescent triphenylamines (TPA) end-capped respectively with one and three pyrene arms, namely *N*,*N*-diphenyl-4-(pyren-1-yl)aniline (PyTPA) and 4,4',4"-trispyrenylphenylamine (TPyPA), have been designed, synthesized and applied as hole-transporting emitters in organic light-emitting devices (OLEDs). While the two compounds have similar chemical structures (the only difference being the number of pyrene arms), devices based on them show distinct electroluminescent characteristics. PyTPA-based devices exhibit intense deep-blue emission with Commission Internationale de L'Eclairage coordinates (CIE) of (0.14, 0.11); whilst TPyPA-based devices emit white light with CIE of (0.31, 0.35). It is considered that the three electron-donating pyrene arms in TPyPA increase its electron-donating ability, which facilitates exciplex formation with the electron-transporter. This gives rise to a yellow exciplex emission and thus shifts the electroluminescence from deep-blue to white. More importantly, both devices can operate at record-low driving voltages (<4V at 20 mA cm<sup>-2</sup>), indicating the TPA-cored emitters offer not only high luminance efficiencies, but also good hole-injection and transporting features.

## Introduction

Since the first report on high efficiency multilayered organic light-emitting devices (OLEDs),<sup>1</sup> this research topic has drawn considerable attention owing to the huge market potential in full-color flat panel displays as well as solid-state lighting. Development of efficient blue electroluminescence (EL) materials is of particular interest due to their applications in both full-color displays and lighting. Compared with the other two primary colors (red and green), the performance of pure blue emitters is still lacking behind. In particular, deep-blue host emitters which can be used in a non-dopant configuration are highly desirable, not only for a more simple fabrication process, but also to avoid possible degradation due to segregation in a host-dopant system.

To address the above needs, extensive studies have been carried out to design and synthesize blue fluorescent host emitters,<sup>2-21</sup> including derivatives with anthracene,<sup>3-7</sup> fluorene,<sup>8-12</sup> pyrene,<sup>13,14</sup> or carbazole<sup>15-17</sup> cores. Among all blue emitting materials, anthracene cored compounds are widely investigated because of their superior performance. However, high efficiency devices that can match the National Television System Committee (NTSC) standard for blue emission, *i.e.* Commission Internationale de L'Eclairage coordinates (CIE) of (0.14, 0.08), are still rather rare.

Apart from anthracene derivatives, we have recently synthesized an efficient light-emitting material with a hole-transporting triphenylamine (TPA) core, namely 4,4',4''-tris[8-(7,10-diphenylfluoranthenyl)]phenylamine (TDPFPA).<sup>22</sup> EL efficiencies of 9.9 cd A<sup>-1</sup> and 10.6 lm W<sup>-1</sup> and green emission with CIE of (0.24, 0.54) were obtained. It is worth noting that while TPA derivatives (*e.g.* TPD, TCTA, m-MTDATA *etc.*) have been extensively used as hole-transporting or -injecting materials, they are seldom used as light-emitting materials in OLEDs.

In this work, we report two deep-blue emitting TPA derivatives developed in-house. The two TPA derivatives are respectively end-capped with one and three pyrene arms, namely N,N-diphenyl-4-(pyren-1-yl)aniline (PyTPA) and 4,4',4"-trispyrenylphenylamine (TPyPA). It was observed that PyTPA has good hole-transporting properties and strong deep-blue emission. A non-doped PyTPA device shows a turn-on voltage ( $V_{turn-}$ on at 1 cd A<sup>-1</sup>) of 2.8 V and a driving voltage (at 20 mA cm<sup>-2</sup>) of 3.6 V. To the best of our knowledge, these are the lowest values reported for non-doped deep-blue fluorescent OLEDs. The deepblue (CIE of (0.14, 0.11)) device has a maximum current efficiency of 2.0 cd A<sup>-1</sup> (corresponding to 2.0 lm W<sup>-1</sup>) and mild roll-off at high brightness (1.9 and 1.7 cd A<sup>-1</sup> at 1000 and 5000 cd  $m^{-2}$  respectively). These parameters suggest that PyTPA has comparable performance among the state-of-the-art deep-blue fluorescent emitters. On the other hand, while TPyPA has a similar chemical structure to that of PyTPA as well as a blue photoluminescence (PL) peak; its devices show a white emission due to an additional exciplex emission. With these results, relationships between the chemical structures and the exciplex formation are also discussed.

## Experimental

<sup>1</sup>H NMR spectra were recorded with a Bruker DPX-400 spectrometer (400 MHz) and mass spectrometry was performed on

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a PE SCIEX LC/MS spectrometer. Elemental analyses were carried out on a Vario EL Elementar by the Flash EA 1112 method. Modulated differential scanning calorimetry (MDSC) and thermogravimetric analysis (TGA) were performed under a nitrogen atmosphere at a heating rate of 10 °C min<sup>-1</sup> on a TA Instruments MDSC 2910 and TGA Q50, respectively. Solution PL quantum yields ( $\Phi_{PL}$ ) were determined by using the method of Demas and Crosby<sup>23</sup> with a degassed aqueous solution of quinine sulfate as a reference. Absorption and PL spectra were measured with a Perkin Elmer Lambda 2S UV/VIS spectrometer and a Perkin Elmer LS 50B luminescence spectrometer, respectively. Cyclic voltammetric measurements (CV) were carried out on a CH Instruments CHI 600A electrochemical analyzer.

OLEDs were fabricated on indium-tin oxide coated glass substrates with sheet resistance of 30  $\Omega$  square<sup>-1</sup>. Prior to successive film deposition, they were cleaned with Decon 90, rinsed in de-ionized water, then dried in an oven, and finally treated in an ultraviolet-ozone cleaner. The organic materials used have acronyms as follows: NPB: N,N'-diphenyl-1,1'biphenyl-4,4'-diamine; BPhen: 4,7-diphenyl-1,10-phenanthroline; and TPBI: 1,3,5-tris(N-phenylbenzimidazol-2-yl) benzene. Devices with a base structure of either ITO/NPB (70 nm)/PyTPA or TPyPA (30 nm)/TPBI (20 nm)/LiF (0.5 nm)/Al (100 nm) or ITO/PyTPA or TPyPA (100 nm)/TPBI (20 nm)/LiF (0.5 nm)/A1 (100 nm) were fabricated by thermal evaporation in a high vacuum chamber (base pressure  $\sim 10^{-6}$  Torr). In the three-layer devices (i.e. NPB/TPA derivatives/TPBI), the TPA derivatives were used as host emitters and NPB and TPBI were used as the hole-transporting layer (HTL) and the electron-transporting layer (ETL), respectively. The TPA derivatives function as both the host emitter and the HTL in the two-layer devices (i.e. TPA derivatives/TPBI) such that their hole-transporting abilities can be accessed. All films were sequentially deposited at a rate of 0.1-0.2 nm s<sup>-1</sup> without vacuum break. A shadow mask was used to define the cathode and to make four 0.1 cm<sup>2</sup> devices on each substrate. Current density-voltage-luminance (J-V-L) characteristics and EL spectra were measured simultaneously with a programmable Keithley model 237 power source and a Photoresearch PR650 spectrometer. All measurements were carried out at room temperature under ambient atmosphere without any encapsulation.

#### **Results and discussion**

Scheme 1 shows the chemical structures of PyTPA and TPyPA, the identities of which have been confirmed by <sup>1</sup>H-nuclear magnetic resonance, elemental analysis, and mass spectrometry. Incorporation of the TPA with a pyrene group is expected to lead to efficient hole-injection and transportation in these compounds, and to form non-coplanar structures which bear bulky substituents in order to disrupt the intermolecular interaction and inhibit the problematic re-crystallization, and thus improve the thermal and morphological stability of these materials. From the TGA data, both PyTPA and TPyPA are highly stable in nitrogen with high decomposition temperatures ( $T_d$ ) of 416 and 565 °C respectively. On the other hand, these compounds can be obtained in high yields *via* simple Suzuki coupling reactions between commercial starting materials.<sup>24</sup> endowing them with an important attribute of low production cost required for large-scale commercial applications.

Apart from the  $T_d$ , melting and glass transition temperatures  $(T_m \text{ and } T_g)$  of TPyPA and PyTPA were also measured. As determined by MDSC,  $T_m$  and  $T_g$  of TPyPA are 316 and 174 °C; while PyTPA melts at 160 °C (PyTPA has no obvious glass transition). The high  $T_m$  for TPyPA may be attributed to the two extra pyrene groups at the C4 position of the TPA backbone, which effectively enhance the degree of symmetry and increase the molecular weight. In addition, both PyTPA and TPyPA show high  $\Phi_{PL}$ ; whereas solution  $\Phi_{PL}$  were estimated to be 74 and 80% for PyTPA and TPyPA, respectively.

Fig. 1 depicts the absorption and PL spectra of PyTPA and TPyPA neat films deposited on quartz substrates. It is well established that the increase in conjugation length may result in a red-shift of onset absorption edge ( $\lambda_{abs.edge}$ ) and PL peak ( $\lambda_{PL}$ ). As expected, the TPyPA film shows slight red-shifts of 10 and 16 nm for  $\lambda_{abs.edge}$  and  $\lambda_{PL}$  compared to the PyTPA film. In particular, strong blue emission was observed with a single peak maximum at 452 nm (full spectral width at half maximum, FWHM = 54 nm) for PyTPA, relative to that for TPyPA, *i.e.* 468 nm (FWHM = 52 nm). Intense PL peaks with small FWHM of the two materials suggest that they have good potential to offer saturated blue emission in OLEDs. All photophysical data of PyTPA and TPyPA are summarized in Table 1.

Fig. 2 shows normalized EL spectra of the PyTPA- and the TPyPA-based OLEDs with either TPBI or BPhen as ETL at L =100 cd m<sup>-2</sup>. While the EL spectra for PyTPA-based devices with different ETL are nearly identical, EL spectra of TPyPA-based devices with TPBI and BPhen ETL show obvious differences. We also observed that the PyTPA/TPBI device performs considerably better than the PyTPA/BPhen device; our efforts are thus concentrated on the PyTPA/TPBI device hereafter. For the TPyPA devices, in addition to the main peak at 472 nm which corresponds well with the PL peak, an additional peak at 570 nm was observed. The broad and featureless shape of the 570 nm peak indicates that it might be related to exciplex emission. This is further confirmed by the same peak appearing in a PL spectrum of a blended film of TPyPA and BPhen (Fig. 2). With the addition of the strong exciplex emission, the TPyPA/BPhen device gives an efficient white emission. The TPyPA/BPhen device was thus further explored for its efficient white emission.

It is worth noting that exciplex emission is found to originate from the delocalization of electronic excitation over the two molecules and from partial electron transfer between the electron-donating and electron-accepting molecules. As reported by Offermans *et al.*,<sup>25</sup> the charge transfer character of the exciplex is strongly dependent on the strength of the electron-donating properties of the donor materials and with the electron affinity of the acceptor. Thus, it is reasonably inferred that the formation of exciplex emission in TPyPA is due to the increase in electrondonating ability of TPyPA resulting from an increase in the number of pyrene arms. This is not the case for the PyTPA-based device, in which no additional exciplex peak is observed when it is in contact with the same electron-accepting material (TPBI or BPhen).

Fig. 3 shows J–V–L characteristics of the PyTPA/TPBI and TPyPA/BPhen devices with and without the NPB layer. It can be seen that at the same voltage, the bi-layer PyTPA/TPBI device



Scheme 1 Synthetic routes and molecular structures of TPyPA (top) and PyTPA (bottom).



**Fig. 1** Absorption (Abs) and photoluminescence (PL) spectra of PyTPA and TPyPA films on quartz substrates.

shows higher brightness and current than the tri-layer NPB/ PyTPA/TPBI device. For example, at a current density of 20 mA cm<sup>-2</sup>, the driving voltages for bi-layer and tri-layer PyTPA-based devices are 3.57 and 3.68 V, respectively. On the other hand, for TPyPA-based devices, higher current and brightness were observed in the tri-layer device. This can be attributed to the difference between the highest occupied molecular orbital energies ( $E_{\rm HOMO}$ ) of PyTPA and TPyPA. Based on their oxidation onset potentials,  $E_{\rm HOMO}$  values of PyTPA and TPyPA were estimated to be 5.19 and 5.62 eV, respectively. Their lowest unoccupied molecular orbital energies ( $E_{\rm LUMO}$ ) were then estimated from the differences between the HOMO energies and the optical bandgaps ( $E_{opt}$ ) determined from their absorption spectra, *i.e.*  $E_{LUMO} = 2.37$  and 2.86 eV for PyTPA and TPyPA, respectively. For the TPyPA-based devices, NPB ( $E_{HOMO} =$ 5.3 eV) can act as a stepping-stone to facilitate hole-injection from ITO ( $E_{Fermi} = 4.8 \text{ eV}$ ) to the TPyPA ( $E_{HOMO} = 5.62 \text{ eV}$ ) layer. However, in the case of PyTPA, the hole-injection barrier at ITO/PyTPA is only 0.39 eV. This small barrier allows efficient hole-injection directly from ITO to PyTPA. Inserting an extra NPB layer would instead increase the hole-injection barrier to 0.5 eV and thus increase the driving voltage. Nevertheless, it should be pointed out that both devices can work at record-low driving voltages (<4V at 20 mA cm<sup>-2</sup>) and that, to the best of our

**Table 1** Photophysical properties of PyTPA and TPyPA. Thermal properties were characterized with thermogravimetric analysis (TGA) and modulated differential scanning calorimetry (MDSC) measurements at the same heating rate of 10 °C min<sup>-1</sup>.  $\Phi_{PL}$  was measured in solution.  $\lambda_{abs.max}$  and  $\lambda_{PL}$  were measured in thin-film.  $E_{opt}$  was estimated from the long wavelength onset of the absorption band.  $E_{HOMO}$  was defined by cyclic voltammetry measurement while  $E_{LUMO}$  was estimated by  $E_{LUMO}$  =  $E_{HOMO} - E_{opt}$ 

Material	$T_{ m g}/T_{ m m}/$ $T_{ m d}$ (°C)	$\Phi_{ m PL}$ (%)	$\lambda_{abs.max} (nm)$	$\lambda_{\rm PL}$ (nm)	$\lambda_{abs.edge}$ (nm)/ $E_{opt}$ (eV)	$E_{\rm HOMO}/E_{\rm LUMO}$ (eV)
РуТРА	—/160/416	74	374	452	440/2.82	5.19/
ТРуРА	174/316/ 565	80	381	468	450/2.76	5.62/ 2.86



Fig. 2 Electroluminescence spectra of various OLEDs viewed in the normal direction at a luminance of 100 cd  $m^{-2}$ . All devices have the configuration of ITO/NPB (70 nm)/PyTPA or TPyPA (30 nm)/TPBI or BPhen (20 nm)/LiF (0.5 nm)/Al (100 nm). Photoluminescence spectrum of a 10% BPhen : TPyPA blended film is also shown for reference.



Fig. 3 Current density-voltage-luminance characteristics of various devices. Device configurations: ITO/PyTPA (100 nm)/TPBI (20 nm)/LiF (0.5 nm)/Al (100 nm); ITO/NPB (70 nm)/PyTPA (30 nm)/TPBI (20 nm)/LiF (0.5 nm)/Al (100 nm); ITO/TPyPA (100 nm)/BPhen (20 nm)/LiF (0.5 nm)/Al (100 nm); ITO/NPB (70 nm)/TPyPA (30 nm)/BPhen (20 nm)/LiF (0.5 nm)/Al (100 nm).

knowledge, the driving voltage of the bi-layer PyTPA-based device is the lowest reported among non-doped small-molecular fluorescent blue OLEDs.

Fig. 4 and 5 show the efficiencies as a function of current density for the PyTPA- and the TPyPA-based devices. As depicted in Fig. 4, the maximum current, power and external quantum efficiencies ( $\eta_C$ ,  $\eta_P$ ,  $\eta_E$ ) for the bi-layer PyTPA-based device are 1.92 cd A<sup>-1</sup>, 2.01 lm W<sup>-1</sup>, and 1.97%, respectively, whereas those for the tri-layer device are only 1.72 cd A<sup>-1</sup>, 1.85 lm W<sup>-1</sup>, and 1.62%, respectively. Meanwhile, the maximum  $\eta_C$ ,  $\eta_P$ ,  $\eta_E$  for tri-layer TPyPA-based devices are 9.36 cd A<sup>-1</sup>, 8.98 lm W<sup>-1</sup>, 3.93%, respectively, whereas those for bi-layer devices are only 7.37 cd A<sup>-1</sup>, 7.30 lm W<sup>-1</sup>, and 3.25%, respectively (as shown in Fig. 5). It is worth pointing out that the common problem of efficiency roll-off is not observed in the present devices, which show only a mild decrease (~3–9% drop from the maximum values) in current efficiencies. At a current density of 200 mA



Fig. 4 Efficiencies as a function of current density. Device configurations: ITO/PyTPA (100 nm)/TPBI (20 nm)/LiF (0.5 nm)/Al (100 nm); ITO/NPB (70 nm)/PyTPA (30 nm)/TPBI (20 nm)/LiF (0.5 nm)/Al (100 nm).



Fig. 5 Efficiencies as a function of current density. Device configurations: ITO/TPyPA (100 nm)/BPhen (20 nm)/LiF (0.5 nm)/Al (100 nm); ITO/NPB (70 nm)/TPyPA (30 nm)/BPhen (20 nm)/LiF (0.5 nm)/Al (100 nm).



**Fig. 6** Electroluminescence spectra of the PyTPA-based device viewed in the normal direction at a driving voltage of 3–6 V. Device configuration: ITO/PyTPA (100 nm)/TPBI (20 nm)/LiF (0.5 nm)/Al (100 nm).

cm<sup>-2</sup>, the current efficiencies of bi-layer and tri-layer PyTPAbased devices only drop by 9% (corresponding to 1.74 cd  $A^{-1}$ ) and 3% (corresponding to 1.67 cd  $A^{-1}$ ), respectively; while those for bi-layer and tri-layer TPyPA-based devices are only 9% (6.72 cd  $A^{-1}$ ) and 3% (9.08 cd  $A^{-1}$ ), respectively. The absence of current induced luminance quenching may be attributed to the highly balanced recombination of holes and electrons within the present devices.

For practical display applications, the color in terms of CIE and EL should not vary to a large extent when the device is operating at different driving voltages. Fig. 6 and 7 depict the EL spectra for bi-layer PyTPA- and tri-layer TPyPA-based devices viewed in the normal direction at a driving voltage of 3-5 V. Apparently, the EL spectra and CIE for both devices are almost



Fig. 7 Electroluminescence spectra of the TPyPA-based device viewed in the normal direction at a driving voltage of 3-5 V. Device configuration: ITO/NPB (70 nm)/TPyPA (30 nm)/BPhen (20 nm)/LiF (0.5 nm)/A1 (100 nm).

Table 2 Electroluminescent performance of PyTPA and TPyPA and other blue host materials with anthracene,<sup>3-7</sup> fluorene,<sup>8-12</sup> pyrene,<sup>13,14</sup> and carbazole15-17 cores

Material <sup>Ref</sup>	V <sub>turn-on</sub> (V)	V at 20 mA cm <sup>-2</sup> (V)	$\eta_{\mathrm{C}}$ (cd A <sup>-1</sup> )	$\eta_{ m P} \ ( m lm \ W^{-1})$	η <sub>E</sub> (%)	CIE (x,y)
РуТРА	2.8	3.6	1.92	2.01	1.97	0.14,0.11
TPyPA	2.6	3.3	9.36	8.98	3.93	0.31,0.35
T3 <sup>2</sup>	5.0		0.47		1.40	0.16,0.07
ADN⁴		8.8	0.69			0.16,0.14
ADF⁴	_	9.5	0.85			0.15,0.08
NSA <sup>5</sup>		7.7	7.85	3.29		0.15,0.18
MADN <sup>6</sup>		6.2	1.40	0.70		0.15,0.10
BDMA <sup>7</sup>	4.4	8.4	2.20	1.57		0.16,0.12
TCPC-6 <sup>8</sup>	4.0		1.35		3.72	0.16,0.05
DFDF <sup>9</sup>	3.7	5.2	3.78	2.56	2.30	0.16,0.23
DPF <sup>10</sup>	3.3	6.4	6.00	3.60		0.15,0.19
TPHF <sup>11</sup>	4.0		1.52		2.70	0.17,0.07
MDP3FL <sup>12</sup>			_	1.82	1.90	0.15,0.12
DPB <sup>13</sup>		5.2	5.18	10.6		0.19,0.25
P214	3.5	5.9	3.08	1.17		0.17,0.19
Cz2F5 <sup>15</sup>	5.2	7.0	0.52		0.44	_
Cz-NPh <sup>16</sup>	3.0	5.3	1.96	1.51	2.34	0.14,0.09
DDPFTBC <sup>17</sup>	2.8	5.2	8.70	9.10	3.93	0.18,0.36
In2Bt <sup>18</sup>	3.9		0.86	0.76	1.30	0.16,0.08
cis-TSDTB <sup>19</sup>	5.0	5.6	3.40	1.90		0.15,0.10
BDPQ <sup>20</sup>	4.0	9.5	3.60	1.57	4.50	0.15,0.12

independent of the driving voltage; CIE of (0.141  $\pm$  0.001,  $0.112 \pm 0.000$ ) and  $(0.311 \pm 0.005, 0.355 \pm 0.005)$  were obtained for PyTPA- and TPyPA-based devices. These small color shifts offer convenience for commercial display applications. In particular, due to the difference in the charge carrier mobility in the employed organic layers, it is a common phenomenon that the EL spectra of blue and white OLEDs change under different electric fields as a result of the shift of the light-emitting zone. However, it is not the case for the present devices. This suggests that the hole and electron recombination is well confined within the emissive layer as well as at the emissive layer/ETL interface. Table 2 compares the device performance of the present PyTPAand TPyPA-based devices with those of other recently reported blue OLEDs.<sup>2,4-20</sup> Remarkably, the PyTPA-based device shows comparable or even better performance than other deep-blue OLEDs, in terms of efficiencies, driving voltage (also  $V_{turn-on}$ ), and color purity. More importantly, it exhibited a saturated blue color with CIE of (0.14, 0.11), that can significantly reduce the power consumption of full-color displays. These excellent device performances illustrate that these TPA-cored materials, PyTPA and TPyPA, are promising materials for OLED applications.

#### Conclusions

Two fluorophores consisting of a TPA core and peripheral pyrene groups, namely PyTPA and TPyPA, have been designed, synthesized and characterized. The introduction of a TPA core and a rigid pyrene side group gave high thermal stability and high  $\Phi_{\rm PL}$  in solution. Non-doped OLEDs fabricated with these materials demonstrate distinct EL characteristics. PyTPA-based devices exhibit intense deep-blue emission with CIE of (0.14, 0.11), while TPyPA-based devices emit white light (0.31, 0.35). The discrepancy can be rationalized in terms of greater electrondonating ability with increasing number of pyrene arms. The introduction of three pyrene arms in TPyPA is expected to increase the electron-donating ability, leading to the formation of exciplex emission in the TPyPA-based devices. This results in a color shift from deep-blue to white. More importantly, both devices can work at record-low driving voltages (<4V at  $20 \text{ mA cm}^{-2}$ ).

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