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High-Triplet-Energy Bipolar Host Materials Based on Phosphine Oxide Derivatives for Efficient Sky-Blue Thermally Activated Delayed Fluorescence Organic Light-Emitting Diodes with Reduced Roll-off

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1 We designed and synthesized two new ambipolar host
2 materials, namely **CzPO** and **Cz3PO**, which contain
3 electron-donating carbazole and electron-accepting
4 triphenylphosphine oxide moieties. Thermally activated
5 delayed fluorescence (TADF)-based OLEDs employing
6 **CzPO** and **Cz3PO** as host materials and the 9-(4-(4,6-
7 diphenyl-1,3,5-triazin-2-yl)phenyl)-1,3,6,8-tetramethyl-
8 9H-carbazole (**CzTRZ2**) as the emitter resulted in
9 improved maximum external quantum efficiencies, EQE_{max},
10 of 13.1% and 13.2%, respectively, together with small
11 efficiency roll-offs, while the device based on bis[2-
12 (diphenylphosphino)phenyl]ether oxide (**DPEPO**) showed a
13 much more pronounced efficiency roll-off. The reduced
14 efficiency roll-off in the devices based on **CzPO** and **Cz3PO**
15 can be ascribed to their improved ambipolar charge transport
16 capacity compared to that of **DPEPO**, which results in a
17 broader recombination zone in the emitting layer using these
18 ambipolar hosts.

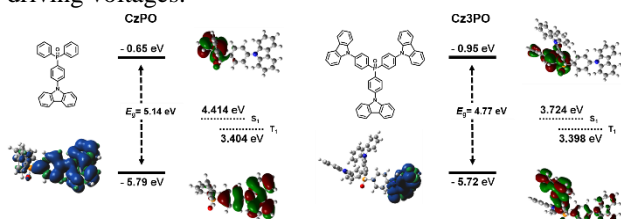
19
20 **Keywords:** Thermally activated delayed fluorescence (TADF) |
21 Organic light-emitting diode (OLED) | Ambipolar host

22 After the invention of fluorescence-based organic
23 light-emitting diodes (OLEDs) in 1987¹, the flat panel
24 display market based on OLEDs is emerging to become
25 the dominant technology. Both organic-based
26 fluorescence and organometallic-based phosphorescence
27 emitters are currently used in OLEDs^{2,3}. In 2012, a series
28 of highly efficient OLEDs based on thermally activated
29 delayed fluorescence (TADF) were demonstrated, in
30 which an efficient process of reverse intersystem crossing
31 (RISC) from a triplet excited-state to a singlet excited-
32 state was realized in purely organic molecules⁴. OLEDs
33 based on TADF emitters achieved comparable an internal
34 quantum efficiency (IQE) of 100%. Since then, a large
35 number of TADF emitters have been reported⁵. In
36 particular, TADF emitters have shown tremendous
37 potential to replace the state-of-the-art fluorophores in
38 high performance deep-blue OLEDs. During the survey of
39 blue-emitting devices, significant efficiency roll-off at

40 high current density was identified in these devices, and
41 the triplet energies (E_T) of the host materials must be
42 sufficiently high compared to the emitters in order to
43 confine the excitons on the emitters and thereby realize
44 efficient blue OLEDs^{5c}. Further, the balance of charge
45 transport carriers must also be considered to mitigate the
46 efficiency roll-off.

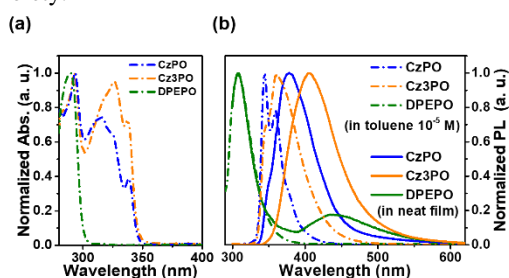
47 For blue TADF-based OLEDs, there are some general
48 requirements for the host material: (1) the E_T of host
49 materials must be higher than that of the emitter to prevent
50 back energy transfer⁶; (2) to facilitate effective charge
51 injection, both the highest occupied molecular orbital
52 (HOMO) and lowest unoccupied molecular orbital
53 (LUMO) energy levels of the host materials should be well
54 matched with both those of the emitter and the other layers
55 of the device⁷; (3) a good host material should possess
56 ambipolar character to best manage charge transport
57 through the emissive layer^{8,9}. In general, the
58 recombination region tends to occur closer to the emissive
59 layer/electron-transporting layer (EML/ETL) interface
60 when using hole-transporting (HT) hosts. Likewise, the
61 recombination region is likely to be located in the
62 EML/HTL interface when using an electron-transporting
63 (ET) host such as the frequently used **DPEPO**. It has been
64 found that poor and unbalanced carrier mobility within the
65 EML is responsible for inferior efficiencies, low stability
66 and efficiency roll-off¹⁰. OLEDs with narrow charge
67 recombination zones lead to devices presenting severe
68 efficiency roll-off due to the local accumulation of high-
69 density triplet excitons, especially at high current
70 densities¹¹⁻¹⁵. As a result, an ambipolar host is more
71 suitable for improving the device performance in blue
72 OLEDs. The simple incorporation of both HT and ET
73 moieties within host materials should give rise to
74 ambipolar host materials¹⁶⁻¹⁹. **DPEPO** is the most popular
75 host material for blue-emitting OLEDs²⁰. However, the
76 use of such ET-type host materials is usually coupled with
77 reduced device stability and efficiency roll-off²¹⁻²⁴. Thus,
78 in this study, we designed two host materials, **CzPO** and

1 **Cz3PO**, by linking carbazole donor units with an electron-
 2 accepting triphenylphosphine oxide group (Fig. 1). Both
 3 hosts show high E_T s and ambipolar charge transport
 4 properties. Sky-blue OLEDs based on these two bipolar
 5 host materials not only showed smaller efficiency roll-off
 6 characteristics but also operated under relatively low
 7 driving voltages.



8
 9 **Figure 1.** Molecular structures, HOMO, LUMO, S_1 and T_1
 10 energy levels of **CzPO** and **Cz3PO** characterized by DFT
 11 calculations at the PBE0/6-31G(d) level of theory, and the
 12 electron density distributions of the frontier molecular
 13 orbitals and spin density distributions of T_1 energy levels.

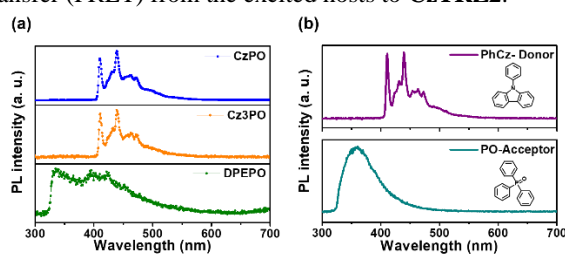
14 The energy levels of **CzPO** and **Cz3PO** were first
 15 investigated computationally using the Gaussian 16 program
 16 package²⁵. Ground-state geometries in the gas phase were
 17 optimized at the PBE0/6-31G(d) level, and the lowest singlet
 18 and triplet excited states were calculated using time-
 19 dependent density functional theory (TD-DFT) based on the
 20 optimized ground-state geometries. The electron density
 21 distribution of the HOMOs and LUMOs of **CzPO** and
 22 **Cz3PO** are depicted in Fig. 1 along with their energies and
 23 the energies of the singlet and triplet excited states and the
 24 spin density distribution of the T_1 states. The HOMOs of both
 25 host materials are delocalized over the electron-donating
 26 carbazole units and bridging phenylene while the LUMOs are
 27 mainly localized over the electron-accepting phosphine oxide
 28 moiety.



29
 30 **Figure 2.** UV-Vis absorption spectra of **CzPO**, **Cz3PO**
 31 and **DPEPO** in toluene (10^{-5} M) and (b) PL emission spectra
 32 of **CzPO**, **Cz3PO** and **DPEPO** in toluene (10^{-5} M) and neat
 33 films.

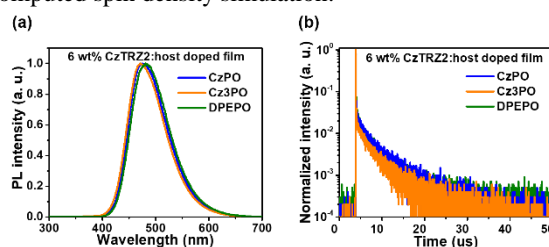
34 The UV-Vis absorption and photoluminescence (PL)
 35 spectra of **CzPO**, **Cz3PO** and **DPEPO** are shown in Fig. 2
 36 respectively. The photophysical data are summarized in
 37 Table S1. In toluene, all host materials show strong
 38 absorption bands below 300 nm, which are assigned as $\pi-\pi^*$
 39 transitions of the phosphine oxide moiety. The absorption
 40 band beyond 300 nm is absent in **DPEPO**, and the lowest-

41 energy absorption bands ranging from 310 to 350 nm are
 42 attributed to the $\pi-\pi^*$ transition of the 9-phenylcarbazole unit
 43 in **CzPO** and **Cz3PO**. In toluene, **CzPO** shows emission
 44 maxima (λ_{PL}) at 344 and 359 nm, while **Cz3PO** shows λ_{PL} at
 45 361 nm. **DPEPO** shows the most hypsochromically shifted
 46 emission at 290 nm. The bathochromically shifted emission
 47 in **Cz3PO** is due to the presence of three donor groups within
 48 the molecule. In neat films, only **CzPO** and **Cz3PO** display
 49 red-shifted emission at 377 and 406 nm, respectively,
 50 whereas **DPEPO** show an additional emission band at 437
 51 nm that may be ascribed to excimer emission.²⁸ In the present
 52 study, **CzTRZ2** was used as the TADF emitter in the emitting
 53 layer as the absorption spectrum of **CzTRZ2** overlaps
 54 strongly with the neat film emission spectra of **CzPO** and
 55 **Cz3PO**, thereby assuring efficient Förster resonance energy
 56 transfer (FRET) from the excited hosts to **CzTRZ2**.



57
 58 **Figure 3.** (a) Phosphorescence spectra of **CzPO**, **Cz3PO**
 59 and **DPEPO** in toluene matrices at 77 K. (b)
 60 Phosphorescence spectra of donor and acceptor moieties in
 61 toluene matrices at 77 K.

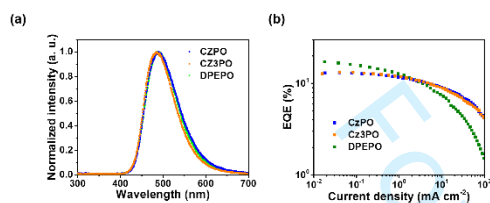
62 The phosphorescence spectra of **CzPO**, **Cz3PO** and
 63 **DPEPO** are depicted in Fig. 3a and these are cross-compared
 64 with the phosphorescence spectra of *N*-phenylcarbazole and
 65 triphenylphosphine oxide (Fig. 3b), which act as reference
 66 electron-donor and electron-acceptor moieties. From the
 67 phosphorescence spectra, the E_T values were determined to
 68 be 3.06 and 3.05 eV for **CzPO** and **Cz3PO**, respectively. The
 69 E_T of **CzPO** and **Cz3PO** are higher than that of **CzTRZ2**
 70 (2.85 eV)^{5d}, ensuring a suppression of back energy transfer.
 71 Both **CzPO** and **Cz3PO** show similar vibronic-structured
 72 emission profiles and E_T to that of *N*-phenylcarbazole.
 73 Therefore, the lowest T_1 excited state of each of **CzPO** and
 74 **Cz3PO** is localized on the donor moieties, consistent with the
 75 computed spin density simulation.



76
 77 **Figure 4.** (a) Steady-state PL spectra and (b) time-resolved
 78 PL decay profiles of 6 wt.%-**CzTRZ2**:host doped thin films
 79 (host = **CzPO**, **Cz3PO**, and **DPEPO**) measured at 300 K
 80 under N_2 (excitation wavelength: 280 nm).

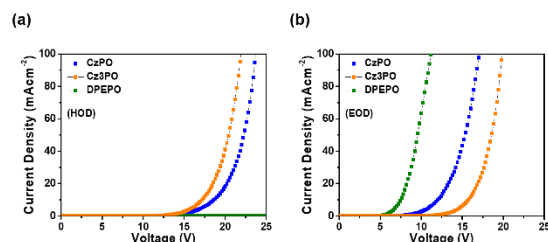
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1 The PL spectra of 6 wt % -**CzTRZ2** doped films in **CzPO**,
 2 **Cz3PO** and **DPEPO** are nearly identical, indicating efficient
 3 FRET (Fig. 4a). The **CzPO**-, **Cz3PO**- and **DPEPO**-doped
 4 films of **CzTRZ2** show broad and structureless sky-blue
 5 emission with high Φ_{PL} of 81%, 75%, and 92%, respectively.
 6 The transient decay profiles of the doped films clearly show
 7 biexponential decay respectively, at 300 K (Figs. S1 and 4b).
 8 Such behavior is characteristic of the TADF emission
 9 previously reported for this compound.^{5d} Furthermore, both
 10 hosts materials possess a high decomposition temperature, T_d ,
 11 of 365 and 454 °C, for **CzPO** and **Cz3PO**, respectively,
 12 which is indicative of high thermal and morphological
 13 stabilities (Fig. S2). The T_d values are substantially higher
 14 than that of **DPEPO** ($T_d = 322$ °C)²⁶.



15
 16 **Figure 5.** (a) The EL spectra measured at 8 V, and (b)
 17 external quantum efficiency (η_{ext})–current density curves of
 18 the TADF-based OLEDs.

19 We next evaluated the EL performance in OLEDs using
 20 **CzTRZ2** as the TADF emitter and **CzPO**- and **Cz3PO** and
 21 **DPEPO** as host matrices in the EML. The devices were
 22 fabricated with the following architecture : ITO (100
 23 nm)/HATCN (10 nm)/TAPC (40 nm)/mCP (10 nm)/6 wt.-%
 24 **CzTRZ2**:host (20 nm)/PPT (10 nm)/TPBi (50 nm)/LiF (0.8
 25 nm)/Al (100 nm), in which 2,3,6,7,10,11-hexacyano-
 26 1,4,5,8,9,12-hexaazatriphenylene (HATCN) is the hole-
 27 injection layer, 1,1-bis(4-ditolylaminophenyl) cyclohexane
 28 (TAPC) and 1,3-bis(*N*-carbazolyl)benzene (mCP) are the
 29 hole-transporting layer and electron-blocking layer,
 30 respectively. The EML layer consists of 6 wt.-% of **CzTRZ2**
 31 doped in **CzPO**, **Cz3PO** or **DPEPO**. 2,8-
 32 Bis(diphenylphosphoryl)dibenzothiophene (PPT) is the hole-
 33 blocking layer and 1,3,5-tris(*N*-phenylbenzimidazol-2-
 34 yl)benzene (TPBi), LiF and Al serve as the electron-
 35 transporting layer, the electron-injection layer and cathode,
 36 respectively. The ET of electron-blocking and hole-blocking
 37 layers, i. e., mCP (2.9 eV) and PPT (2.9 eV), respectively, are
 38 higher than that of **CzTRZ2**, which ensures confinement of
 39 the triplet excitons within the EML²⁷. The EL spectra and
 40 EQE characteristics of the corresponding OLEDs are shown
 41 in Fig. 5. Devices based on **CzPO** and **Cz3PO** as host
 42 materials resulted in high EQE_{max} of 13.1 and 13.2%
 43 respectively. By contrast, the device based on **DPEPO**
 44 showed a higher EQE_{max} of 16.7%. Despite the higher EQE_{max} ,
 45 the **DPEPO**-based devices showed a significantly serious
 46 efficiency roll-off with an EQE of only 1% at 100 mA cm⁻².
 47 Meanwhile, the **CzPO**- and **Cz3PO**-based devices exhibited
 48 reduced efficiency roll-off with EQE_{100} of 4.2% and 4.2%,
 49 respectively, at a display-relevant current density of 100 mA
 50 cm⁻². All the device data are shown in Table S2.



51
 52 **Figure 6.** Current density–voltage (J – V) characteristics of
 53 (a) hole-only devices (HODs) and (b) electron-only devices
 54 (EODs) based on the **CzPO**, **Cz3PO** and **DPEPO** hosts.

55 To investigate the reason for the reduced roll-off
 56 characteristics, hole-only devices (HODs) and electron-only
 57 devices (EODs) with different host materials were fabricated
 58 to evaluate the carrier transporting properties of the
 59 corresponding OLEDs. These device configurations were:
 60 ITO/HAT-CN (10 nm)/TAPC (20 nm)/ host (60 nm)/TAPC
 61 (20 nm)/Al (100 nm) for the HODs and ITO/TPBi (20
 62 nm)/host (60 nm)/TPBi (20 nm)/LiF (0.8 nm)/Al (100 nm)
 63 for the EODs. Figure 6 showed the J – V curves of the
 64 fabricated HODs and EODs. For the HODs (Fig. 6a), the
 65 **Cz3PO** and **CzPO**-based devices showed hole
 66 transporting/injection properties due to the presence of more
 67 electron-donating carbazole units, while no significant
 68 electron current was observed in **DPEPO**-based device. In
 69 contrast, for EODs, the order of the electron
 70 transporting/injection properties of devices was **DPEPO** >
 71 **CzPO** > **Cz3PO** (Fig. 6b). From both the HODs and EODs
 72 characteristics, **CzPO** and **Cz3PO** were confirmed to be
 73 ambipolar in nature, in which **CzPO**- and **Cz3PO**-based
 74 devices exhibited balanced hole and electron
 75 transporting/injection properties. Thus, the well-balanced
 76 charge flux and broad distribution of the charge-
 77 recombination zones within the EML resulted in low
 78 efficiency roll-off characteristics for **CzPO**- and **Cz3PO**-
 79 based OLEDs.

80 In summary, reduced efficiency roll-offs were
 81 demonstrated in sky blue TADF-based OLEDs using **CzPO**
 82 and **Cz3PO** as ambipolar hosts compared to the device with
 83 the reference **DPEPO** host. Sky blue TADF-OLEDs based
 84 employing **CzPO** and **Cz3PO** host materials showed
 85 maximum EQEs of 13.1 and 13.2%, respectively, and
 86 reduced roll-offs with EQEs of 4.2% and 4.2%, respectively,
 87 at current densities of 100 mA cm⁻².

88
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Graphical Abstract	
Textual Information	
A brief abstract (required)	<p>We designed and synthesized two new ambipolar host materials, namely CzPO and Cz3PO, which contain electron-donating carbazole and electron-accepting triphenylphosphine oxide moieties. Thermally activated delayed fluorescence (TADF)-based OLEDs employing CzPO and Cz3PO as host materials and the 9-(4-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl)-1,3,6,8-tetramethyl-9H-carbazole (CzTRZ2) as the emitter resulted in improved maximum external quantum efficiencies, EQE_{max}, of 13.1% and 13.2%, respectively, together with small efficiency roll-offs, while the device based on bis[2-(diphenylphosphino)phenyl]ether oxide (DPEPO) showed a much more pronounced efficiency roll-off. The reduced efficiency roll-off in the devices based on CzPO and Cz3PO can be ascribed to their improved ambipolar charge transport capacity compared to that of DPEPO, which results in a broader recombination zone in the emitting layer using these ambipolar hosts.</p>
Title(required)	<p>High-Triplet-Energy Bipolar Host Materials Based on Phosphine Oxide Derivatives for Efficient Sky-Blue Thermally Activated Delayed Fluorescence Organic Light-Emitting Diodes with Reduced Roll-off</p>
Authors' Names(required)	<p><i>Jong Uk Kim,^{1,2} Michael Y. Wong,³ Shiv Kumar,³ Oliver G. Hayes,³ Finlay Duncan,³ Chin-Yiu Chan,¹ Ben Yiu-Wing Wong,¹ Hao Ye,¹ Lin-Song Cui,¹ Hajime Nakanotani,^{1,2,4} Eli Zysman-Colman^{*3} and Chihaya Adachi,^{*1,2,4}</i></p>
Graphical Information	
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