

Molecular hosts for triplet emitters in organic light-emitting diodes and the corresponding working principle

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This paper summarizes the mechanism and routes for excitation of triplet emitters in dopant emission based phosphorescent organic light-emitting diodes (PhOLEDs), providing a comprehensive overview of recent progress in molecular hosts for triplet emitters in PhOLEDs. Particularly, based on the nature of different hosts, e.g., hole transporting, electron transporting or bipolar materials, in which the dopant emitters can be hosted to generate phosphorescence, the respective device performances are summarized and compared. Highlights are given to the relationships among the molecular structure, thermal stability, triplet energy, carrier mobility, molecular orbital energy level and their corresponding device performances.

phosphorescent organic light-emitting diodes (OLEDs), working principle, electrophosphorescence, hosts, triplet emitter

1 Introduction

Organic light-emitting diodes (OLEDs) are current driven emissive-devices in which organic materials are used as active elements. Compared to their inorganic counterpart, organic materials have the advantages of low-cost synthesis, functional tunability, flexibility and good thin film-forming ability. The devices based on organic materials are generally capable of simple and low-temperature manufacture processes such as spin-coating, ink-printing, and dipping, hence less expensive. Organic materials bear the potentials for large-area fabrication, flexible substrates, and are environmentally friendly. In the past two decades, OLEDs have been intensively studied, due to their potential application for next generation flat panel displays (FPDs) and solid state lighting (SSL).

OLEDs utilizing phosphorescent materials as emitters (PhOLEDs) are more attractive than those based on fluorescent emitters because of their ability to harvest both singlet and triplet excitons, hence reaching 100% internal quantum efficiency [1, 2]. Non-doped PhOLEDs for blue [3], green [4, 5], orange [6], and red [7, 8] were demonstrated. However, those devices usually face the problem of luminescence quenching from the condensed state of emitters. In addition, it is not an economical consideration to apply phosphorescent materials as hosts, because the phosphors which generally consist of transition metals of Ir or Pt are very expensive due to the high cost of starting materials and synthesis. Moreover, doped OLEDs have been demonstrated to possess prolonged operational lifetime without significant concentration quenching [9, 10]. In doped OLEDs, the carrier injection barrier between the emitting layer (EML) and adjacent carrier transport layers as well as the carrier transporting ability of EML can be adjusted feasibly by the host, adding flexibility for the design of device

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structures. This can also simplify the molecular design, because the electronic and photo-physical requirements of EMLs can be considered separately with the former property mainly depending on the host and the latter dominated by the dopant. Thus, most work in the field of PhOLEDs focused on phosphorescent guest-host system.

To disclose the device mechanisms and realize the PhOLED application, a huge number of research works, including many good review articles [11–16], were published. However, despite the fact that the investigation of suitable host materials for triplet emitters has been conducted extensively since the early stage of PhOLEDs research [17], there are rare reviews focusing on this aspect. Here, the mechanism and scenarios for energy transfer from the host to the triplet dopant are summarized and discussed, and the molecular host materials for phosphorescent dopant emitters are reviewed. In particular for host materials, after the discussion of basic characterization methods for the host materials, recent advances directed towards the molecule design as well as their application in devices are addressed. The phosphorescent dopant materials that are discussed in the following parts are shown in Figure 1. We used iridium complexes as representative dopants, because these materials show the best performance among phosphorescent materials, and are the utmost promising ones for applications.

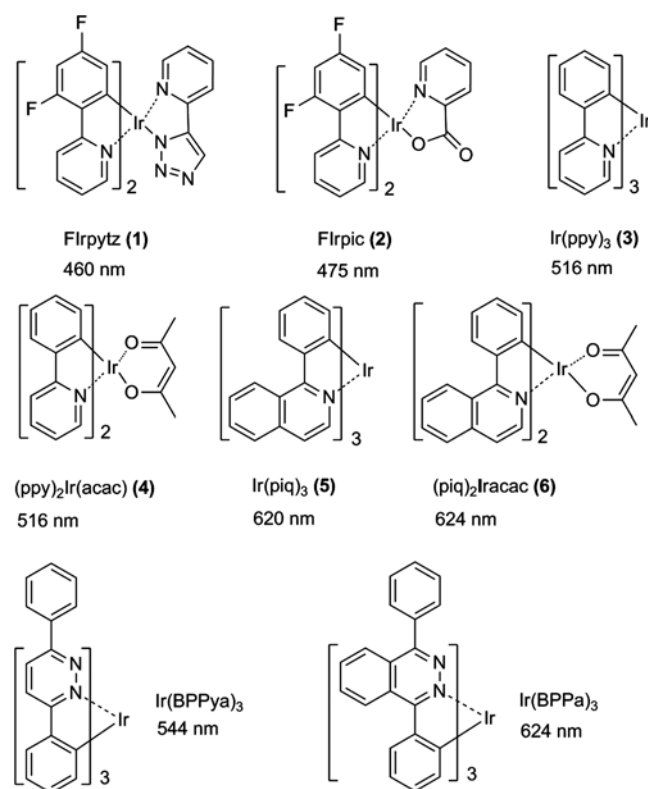


Figure 1 Molecular structures of phosphorescent dopants discussed in this work. The numbers in unit of nm indicate their emission peaks in dispersion (either film or solution).

2 Excitation of triplet emitters dispersed in the host under an electric field

Figure 2(a) shows the basic device architecture for dopant emitter based PhOLEDs, including the hole transport layer (HTL), doped emitting layer (EML) and electron transport layer (ETL) in sequence, sandwiched between two electrodes. Attributed to the long lifetime of the phosphorescent emitter, PhOLEDs usually suffer from large efficiency roll-off at high current density [18], which can be relieved by the selection of the phosphorescent materials with short triplet lifetime, the suitable host material, and the judicious design of device structure such as insertion of the carrier/exciton block layer or buffer layer [19]. Among them, the design of suitable host materials for PhOLEDs is a challenging task, because complex and competing channels exist for host/dopant emitters to be excited as well as for excitons to be confined in the EML, as shown in Figure 2(b). Generally, the ways of electrical excitation of the dopant emitter in PhOLEDs can be categorized into two types: one is direct recombination of carriers on dopants (charge trap) and the other is energy transfer from the excited host.

The favorable charge trap requires the HOMO/LUMO level of the dopant locate above/below that of the host, respectively. In this case, the dopant is likely to act as a hole/electron trap. Sequentially, the trapped holes/electrons on the dopant may provide sites for electron-hole recombination, forming a variety of excited states, such as singlet and triplet excitons. In phosphorescent emitters, it is generally believed that the intersystem crossing (ISC) process from the singlet to the triplet occurs with high probability, thus singlet excitons are expected to transfer to the corresponding triplet state efficiently. Due to the strong spin orbital coupling, strong phosphorescence can be produced via the dominant radiation decay process of triplet excitons. Figure 2(b) illustrates these reaction channels.

Energy transfer is the most significant process in dopant emitter based OLEDs and mainly includes Förster energy transfer and Dexter energy transfer (in Figure 2(b), represented as thin arrow lines and broad arrow lines, respectively). The criteria for efficient Förster energy transfer are (1) good spectra overlap between the emission of the host and the absorption of the dopant, (2) strong to moderate emission of the host, (3) strong absorption of the dopant, and (4) within 100 Å separation between host and dopant molecules. Good Dexter energy transfer must meet (1) spectra overlap between the emission of the host and the absorption of the dopant to provide orbital overlap of the two concerned energy levels, (2) spin conservation, and (3) within 10 Å separation between host and dopant molecules to render spatial vicinity.

Accordingly, in the viewpoint of energy transfer from the host to the triplet state of the dopant, possible mechanisms can be described as follows (refer to Figure 2(b)):

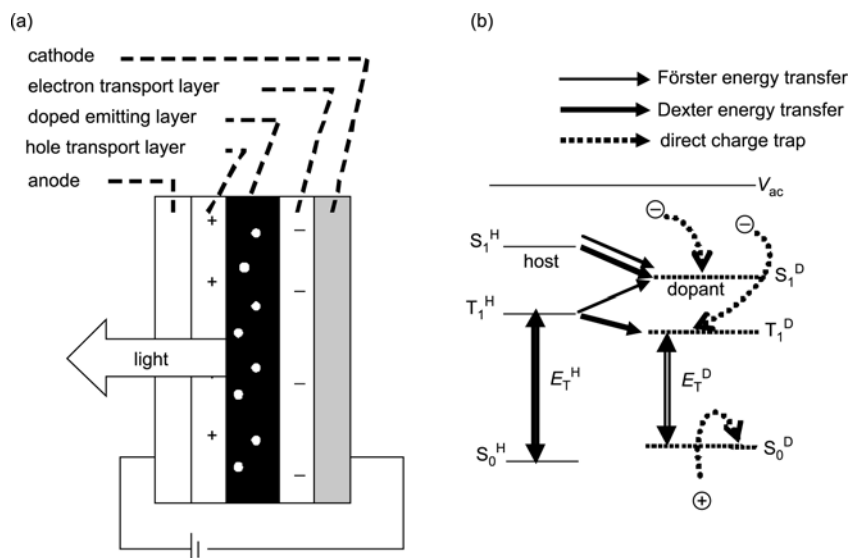
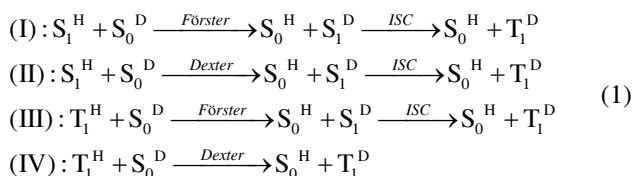


Figure 2 (a) Layered OLEDs structure with a dopant emitter. (b) The main ways for the triplet dopant to be excited under an electric field.



where singlet and triplet states are represented by S and T, respectively; the subscript 1 and 0 indicate an excited state and the ground state, respectively; the superscript H and D refer to the host and the dopant, respectively; ISC means inter-system crossing; Förster and Dexter refer to Förster and Dexter energy transfer, respectively. (I) and (II) are the main routes for energy transfer from the singlet excited state of the host to the triplet of the dopant, while (III) and (IV) represent the dominant channels of those from the triplet excited state of the host to the triplet of the dopant.

It is noteworthy that energy transfer from the singlet of the host (S_1^H) to the triplet of the dopant (T_1^D) is unfavorable, due to the weak absorption of the triplet (not satisfying Förster energy transfer) and the spin difference (S_1^H vs. T_1^D , not meeting the spin conservation rule for Dexter energy transfer). Similarly, Dexter type energy transfer from T_1^H to S_1^D (due to the spin conservation) and Förster energy transfer from T_1^H to T_1^D (due to the weak absorption of the dopant triplet state) are unlikely to happen. In addition, process (III) is only preferential when the host is phosphorescence (meeting the emissive requirement for Förster energy transfer) [17]. In the case of the fluorescent host, the dominant energy transfer process from the host triplet is Dexter energy transfer. Thus, high doping concentration of the dopant is necessary. In addition, the triplet energy of the host (E_T^H , indicated in Figure 2(b)) should be larger than that of the phosphorescent dopant (E_T^D , shown in Figure 2(b)) so as to provide efficient triplet to triplet (T-T) energy transfer from the host to the dopant via exothermic process

and confine the triplet exciton in the dopant.

For evaluation of exciton confinement in the triplet dopant, the difference between the triplet energies of the host and the guest (ΔG ($\Delta G = E_T^D - E_T^H$)) is a critical parameter [17]. Baldo *et al.* have demonstrated the energy transfer process between triplets of the host and phosphorescent dopant under an electric field [17]. By analyzing the transient electroluminescence (EL) and the deduced radiative lifetime, they found that energy transfer from the triplet of host could occur in a system with ΔG slightly greater than, smaller than, or close to zero. However, the most favorite way is $\Delta G < 0$. In 2005, using integrating sphere to measure the photoluminescence (PL) quantum yield (Φ_{PL}) of phosphor-doped films with various hosts under different concentrations, Kawamura *et al.* concluded that the Φ_{PL} of the doped film was strongly dependant on the host material [2]. When the blue phosphor, bis[(4,6-difluorophenyl)pyridinato-N,C^{2'}](picolnato) iridium(III) (FIrpic (2) in Figure 1), was doped into the conventional host material CBP (4,4'-bis(*N*-carbazolyl)-2,2'-biphenyl), the maximum Φ_{PL} of 79% was obtained at a high doping concentration of about 15 mol%. By contrast, the Φ_{PL} of FIrpic-doped mCP (*m*-bis(*N*-carbazolyl)benzene) film reached 100% at the concentration of 1.4 mol%.

As shown in Figure 3, in the former case, because the triplet energy of FIrpic is larger than that of the CBP host, backward energy transfer from FIrpic (2) to CBP occurs exothermically, reducing Φ_{PL} . It is not the case for FIrpic (2) doped mCP film. Due to the smaller triplet energy of FIrpic (2) compared to that of mCP host, backward energy transfer from FIrpic (2) to mCP is greatly suppressed, hence Φ_{PL} is increased and the optimized doping concentration is low.

An example of backward energy transfer from the triplet of the guest to that of the host was demonstrated by the

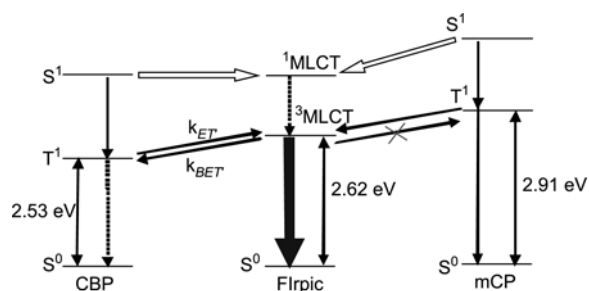


Figure 3 Energy transfer scheme for FIrpic with CBP or mCP.

triplet emission from the host at low temperature in BAiq (bis(2-methyl-8-quinolinato)4-phenylphenolate aluminum(III))-Ir(ppy)₃ (**3**) [tris(2-phenylpyridine)iridium(III)] system [20]. Tsuboi *et al.* also investigated the energy transfer in phosphorescent emitters based host-guest system by analyzing the changes of the PL spectra under different temperatures and obtained similar results [21]. Other techniques employed to study the energy transfer in the phosphorescent host-guest system include photo-induced absorption [22, 23] and

transient PL emission [24].

In PhOLED devices based on the same dopant with different hosts, the dominating device mechanism can be different, e.g., either energy transfer or direct charge trapping, hence the device performances can be varied dramatically. Taking Ir(ppy)₃ (**3**), the most intensively studied phosphorescent material in PhOLEDs, as an example, the variations of performance are demonstrated in Table 1, the features of host materials and different devices characteristics, together with device performance are compared, and the related references are listed for further survey [25–44].

The table shows devices manufactured by vacuum deposition process generally exhibit higher efficiency than those using spin coating process, which could be attributed to the ability of reducing defect formation, avoiding extra impurity, as well as the high possibility of forming pinhole-free thin film under high vacuum. The efficiencies of Ir(ppy)₃ (**3**) based devices are highly dependent on the host and device configuration, with the maximum efficiencies of 105 cd/A (29%, or 133 lm/W) [25], 95 cd/A (27%, or 97 lm/W) [26], 73.0 cd/A (19.2%, or 70.2 lm/W) [27], and 77.9 cd/A

Table 1 Performances of Ir(ppy)₃ based PhOLEDs with different hosts ^{a)}

| Host | HOMO/LUMO (eV) | Triplet energy (eV) | Proc | Max. Eff. | | | Device feature | Ref. |
|-------------------------|-------------------------|---------------------|------|-----------|------|--------|-----------------------------------------------------------------------------------------|------|
| | | | | (cd/A) | (%) | (lm/W) | | |
| CBP | NA/NA | NA | dep | 105 | 29 | 133 | low electron injection barrier and high electron transport | [25] |
| TCTA/ CBP ^{b)} | NA/NA | 2.85/2.60 | dep | 95 | 27 | 97 | chemical doping, charge balance, high reflectivity cathode | [26] |
| TCTA | 5.9/2.7 | NA | dep | 73.0 | 19.2 | 70.2 | efficient hole confinement and favorite electron transfer to eml | [27] |
| <i>o</i> -CzOXD | 5.55/2.56 | 2.68 | dep | 77.9 | 20.2 | 59.3 | low hole injection barrier, both the hole and the electron can be trapped, bipolar host | [28] |
| TFTPA | NA/NA | 2.89 | dep | 44.1 | 12.0 | 21.0 | direct charge injection, and homogeneous dispersion of the dopant at high conc. | [29] |
| ttbCBP | 5.7/2.5 | 2.64 | dep | 28.8 | 8.8 | 16.4 | bulky host with efficient energy transfer | [30] |
| DFC | 5.57/2.20 | 2.53 | dep | NA | 10.5 | 25 | efficient energy transfer | [31] |
| TCTA | 5.9/NA | NA | dep | NA | 13.5 | 53 | chemically doping, efficient energy transfer and direct charge trap | [32] |
| TRZ2 | 6.0/2.6 | 2.81 | dep | NA | 10.0 | 15.9 | balanced carrier in eml | [33] |
| TAZ | NA/NA | NA | dep | NA | 15.6 | 42 | good carriers and triplet exciton confinement, electron transport host | [34] |
| OXD | 5.9/NA | NA | dep | 26 | NA | 10 | efficient energy transfer and direct charge trap | [35] |
| Bepp2 | 5.7/2.6 | NA | dep | 38.3 | NA | 46.6 | good electron transport host, extremely low hole injection barrier | [36] |
| BUPH1 | 5.9/2.8 | 2.4 | dep | 28 | NA | 33 | bipolar and high carrier drifting host | [37] |
| BOBP3 | 6.19/2.81 ^{c)} | 2.53 | dep | 26 | NA | NA | efficient energy transfer | [38] |
| DPCLP | 5.45/2.20 | 2.47 ^{c)} | dep | NA | 11.2 | NA | electron transport host | [39] |
| TCTA /CBP ^{b)} | NA | NA | dep | NA | 14 | NA | exciton confinement in eml | [40] |
| TCTA/TPBI mixed EML | NA | NA | dep | NA | ~16 | NA | broad emitting zone by mixing host and with exciton block layer | [41] |
| Me-TBBI | 5.37/2.19 | 2.76 | spn | 27.4 | NA | 4.5 | bipolar host, balanced carrier injection and transport | [42] |
| DM-TIBN | 5.59/2.33 | 2.77 | spn | 27.3 | NA | 7.3 | bipolar host | [43] |
| TDAPB | 5.1/1.6 | NA | spn | 20 | NA | NA | efficient energy transfer/ homogeneous film | [44] |

a) NA, EML, dep and spn are not available, emitting layer, vacuum deposited, and spin coating, respectively. b) Double EML. c) Deduced from the reference papers.

(20.2%, or 59.3 lm/W) [28], for high performances, 27.4 cd/A (4.5 lm/W) [42] and 20 cd/A [44] for relatively low performances. Utilizing suitable host and the optimizing of device structure according to device mechanism are very significant toward improving PhOLED performance.

Although the confinement of triplet excitons in phosphorescent dopant by choosing a host with relatively high triplet energy is one of the key factors for highly efficient PhOLEDs, particular cases remain. If the host with relatively low triplet energy is employed, as in the case of utilizing polyfluorene (PFO) host for a green triplet emitter, by inserting a buffer layer between the anode and the phosphor-doped PFO layer to block electron drifting out of the EML, electrons will accumulate at the interface between the buffer layer and EML. Meanwhile holes can be trapped directly on the dopant, subsequently recombining with the accumulated electrons at the interface, thus high efficiency can be achieved, with the maximum of 36.8 cd/A and 12.3% [45]. Moreover, it was demonstrated by H. H. Liao et al. that the morphology of the phosphor-doped film was significant for the engenderment of Dexter energy transfer from the host to the dopant, and it could occur in a system without triplet confinement if the triplet exciton lifetime of phosphor was short [22]. In addition, we have studied new green [19] and red [46] Ir complexes in CBP host. By inserting a buffer layer between the HTL and EML, PhOLEDs with high efficiency and low roll-off have been achieved [19].

3 Basic characterization for triplet hosts in PhOLEDs

In OLEDs, a good host material possesses good carrier transport, matched energy level to adjacent layers and high thermal stability. These properties are generally evaluated by carrier mobility (μ), the HOMO and LUMO energy levels, the glass transition temperature (T_g), and the decompose temperature (indicated by 5% thermal decompose temperature, T_d).

3.1 Mobility

To obtain the mobility of an organic semiconductor, time of flight (TOF) [47] and transient electroluminescence (TEL) [48, 49] are usually employed. By measuring the flight time (t_f) by TOF or the transient time (t_T) by TEL of the concerned carrier across a distance (L) under an applied voltage (U), mobility can be calculated as

$$\mu = \frac{L^2}{U \cdot t_T} \quad (2)$$

Figure 4 shows the scheme for these two measurements and the detailed methodology can be found elsewhere [47, 49, 50].

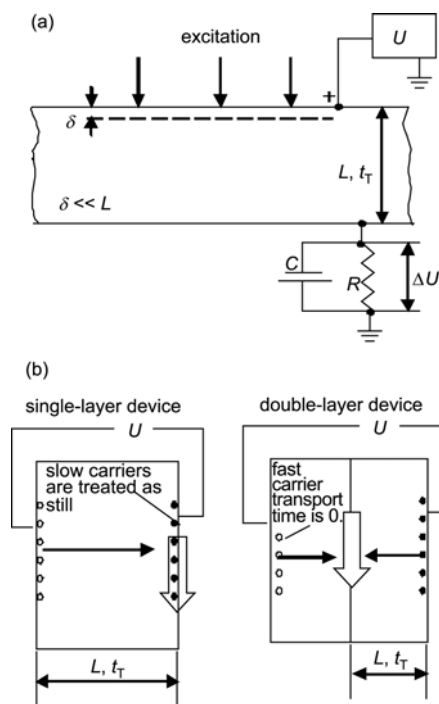


Figure 4 Scheme for mobility measurement. (a) TOF method. δ is the charge generation region, L refers to the thickness of thin film. (b) Transient EL method (the broad arrows represent light emission).

3.2 Energy levels

The HOMO position of organic materials is usually determined either by ultraviolet photoemission spectroscopy (UPS) in the form of thin film, or by cyclic voltammetry (CV) measurement in solution [51–53].

In UPS measurement, HOMO level (E_{HOMO}) which is aligned to vacuum level, can be obtained as

$$E_{\text{HOMO}} = E_{\text{photon}} - \Delta E_{\text{SE-VB}} \text{ (eV)} \quad (3)$$

where E_{photon} is the ultraviolet photo energy for excitation, $\Delta E_{\text{SE-VB}}$ is the energy difference between the low energy secondary electron cutoff and the high energy end of the valence band.

In CV measurement, the values of oxidation potential (E_{ox}) and reduction potential (E_{red}) are related to the HOMO and LUMO level, respectively, and can be corrected to vacuum level alignment:

$$\begin{aligned} E_{\text{HOMO}} &= E_{\text{ref}} - E_{\text{ox}} \text{ (eV)} \\ E_{\text{LUMO}} &= E_{\text{ref}} - E_{\text{red}} \text{ (eV)} \end{aligned} \quad (4)$$

where E_{ref} is the reference, with values of -4.7 eV for Ag/AgCl [54], -4.4 eV for SCE (saturated calomel electrode) [55], -4.8 eV for ferrocene [52], and -4.6 eV for NHE (normal hydrogen electrode) [56].

LUMO can be deduced from the long wavelength cutoff (λ_{cutoff} , nm) of the absorption spectrum and the HOMO:

$$E_{\text{LUMO}} = E_{\text{HOMO}} + \frac{1240}{\lambda_{\text{cutoff}}} (\text{eV}) \quad (5)$$

As a host for triplet emitters, besides the HOMO and LUMO energy levels, the triplet energy of the material (E_T) is also significant to determine the mechanism of T-T energy transfer. This value is usually derived from the phosphorescent emission spectrum, either at room temperature for phosphorescent materials or at low temperature for non-phosphorescent materials. With the high energy peak value (λ_{ph} , nm) from the spectrum, triplet energy can be calculated as

$$E_T = \frac{1240}{\lambda_{\text{ph}}} (\text{eV}) \quad (6)$$

3.3 Thermal analysis

The thermal stability of an organic material is typically evaluated by thermal gravimetric analysis (TGA) with the feature of T_d , and by the differential scanning calorimeter (DSC) with the characteristic value of T_g (sometimes can not be found). Figure 5 shows the typical curves of TGA and DSC. The higher the values of T_d and T_g , the more stable the material.

4 Development of host materials

4.1 Basic requirements for hosts serving for triplet emitters

The basic requirement for a host material in OLEDs is good carrier transport property with the ability of forming pin-hole-free thin film. In addition, enduring thermal and electrochemical stability coupled with stable morphology is desired to ensure a longer device operation time. For organic semiconductors, it is widely accepted that molecules with electron-donating groups usually possess the hole transport feature, while molecules with electron-accepting moieties generally exhibit the electron transport character, and molecules with both electron-donating and electron-

accepting groups may show the bipolar carrier transport property.

Taking into account both the mechanism for exciting the triplet state of the dopant (see Figure 2(b) and discussion thereby) and the device structure in PhOLEDs, we summarized several strategies for host material design: (1) wrapped HOMO/LUMO level of the dopant within those of the host to allow direct charge trap in the dopant material; (2) spectra overlap between the emission of the host and the absorption of the dopant to afford the resonance effect; (3) relatively high triplet energy level of the host to prevent reverse triplet energy transfer from the phosphorescent emitter to the host; (4) balanced hole and electron transport in the host to give chance for better confining of the exciton in the emissive layer, thus offering broad exciton recombination region, and avoiding exciton drift. This point is very significant, because evenly and broadly distributed excitons in a confined region can reduce exciton density for the same current density, thus efficiently preventing T-T annealing; (5) matched HOMO/LUMO level with adjacent hole/electron transport layer to reduce the hole/electron injection barrier, which is desirable for low driving voltage. It is difficult to satisfy all the criteria, however, the trade-off among the carrier injection to EML, the high triplet energy of the host, and the direct charge trap on the dopant need to be considered.

There are several functionalities employed in tailoring the chemical structure of the host materials, such as carbazolyl, phenylsilanyl, starburst amine, triazinyl, oxadiazolyl, arylphosphine oxide, and emissive metal complex, and these materials will be described in the following parts.

4.2 Hole transport hosts

4.2.1 Carbazole based materials

Materials containing carbazolyl moiety predominately exhibit hole transport ability, and are intensively employed as hosts for phosphorescent emitters in PhOLEDs. It has been demonstrated both by theoretical studies [57, 58] and experimental work [59] that due to the lone-pair electrons from the N atom in carbazole, the exchange energy between

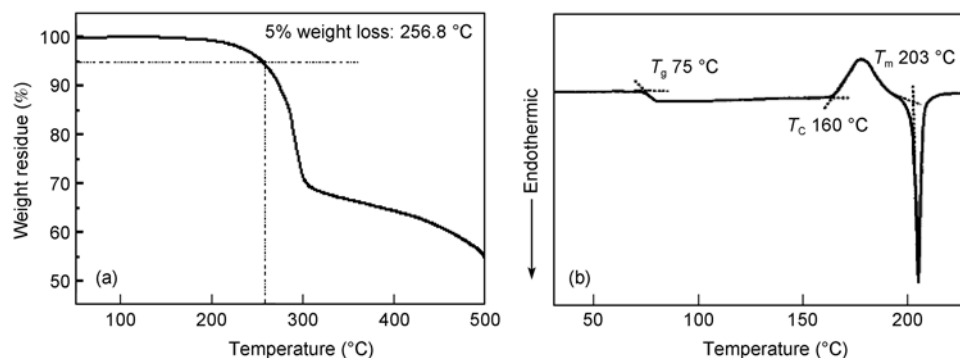


Figure 5 Thermal analysis of (a) TGA curve and (b) DSC curve.

the singlet and triplet of carbazole based materials is generally small, leading to relatively small singlet-triplet splitting hence large triplet energy, which is preferential for triplet dopant emitters. As shown in Figures 6 to 8, we classified carbazole-containing materials into three types: dicarbazole-containing hosts, monocarbazole-based hosts, and carbazole oligomers.

Dicarbazole-containing CBP (**7**) is the most widely used host material for triplet emitters, which can efficiently host green to red phosphorescent emitters with triplet energies smaller than that of CBP (**7**) ($E_T = 2.6$ eV [17]) [46, 60–62]. Using CBP as the host, and employing a high electron transport material as ETL, Ir(ppy)₃ based device achieved efficiency of 133 lm/W (29%) [25], which is the

highest efficiency for Ir(ppy)₃ to date. By attaching sterically hindered t-butyl units to the C-3 and C-6 positions of both carbazole moieties in CBP (**7**), while preserving the characteristics of the wide band-gap and high triplet energy, a more morphologically stable material ($T_g = 175$ °C), ttbCBP (**8**) (Figure 6), was obtained [30]. Devices based on ttbCBP (**8**) show efficiencies of 8.3% (28.8 cd/A, 16.4 lm/W) for green from Ir(ppy)₃ (**3**), and 9.8% (6.7 cd/A, 3.0 lm/W) for red from Ir(piq)₃ (**5**). Hybridizing the 9,9'-spirobifluorene function group with two carbazole units can also afford the thermally stable material ($T_g = 151$ °C) CFL (**9**) (Figure 6) [63]. Based on CFL (**9**) host, EQEs of 13% were obtained from both Ir(piq)₃ and Ir(ppy)₃ (**3**) devices. The lifetime of Ir(piq)₃ (**5**) device based on CFL is nearly 5 times longer

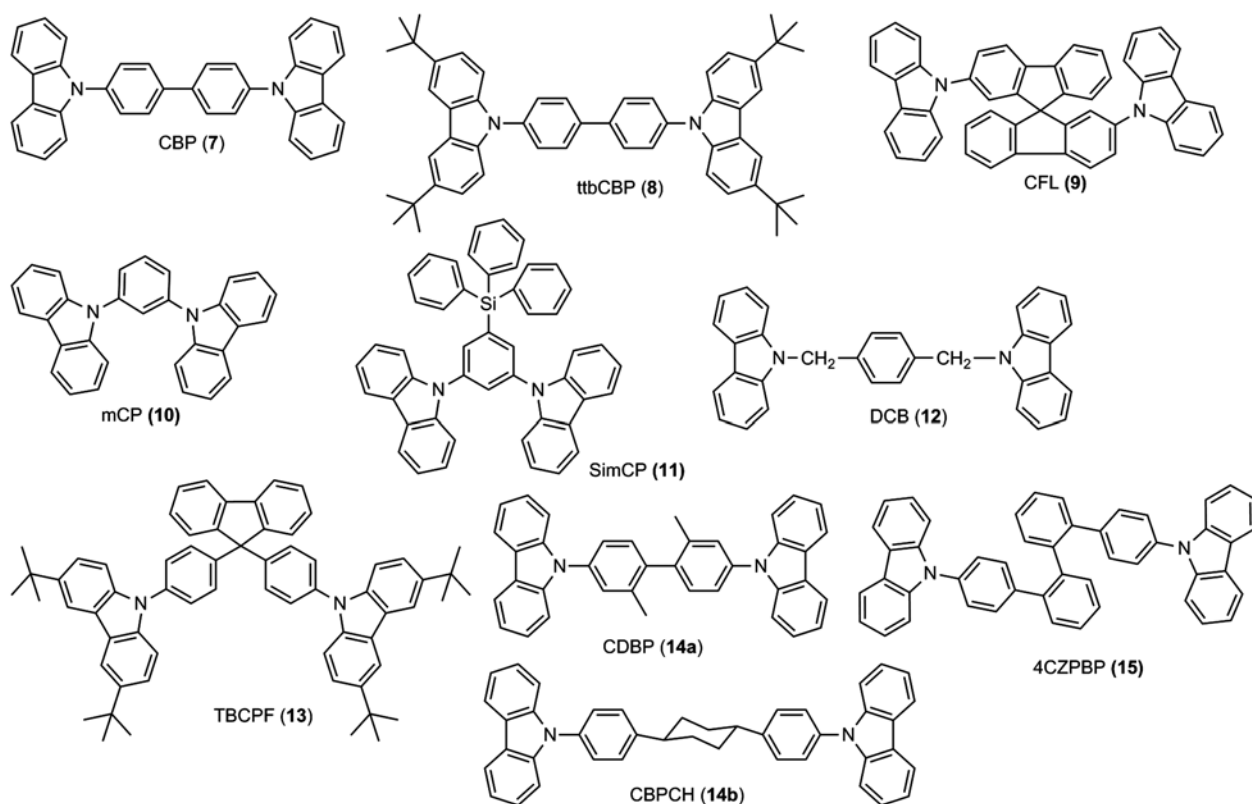


Figure 6 Dicarbazole-containing host materials.

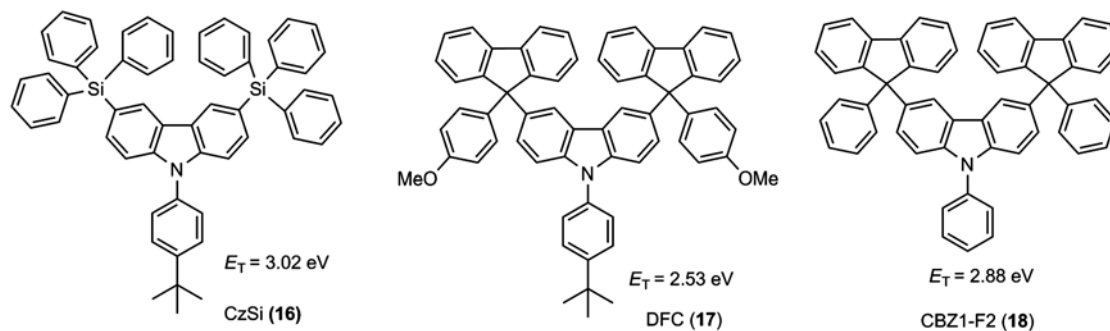


Figure 7 Mono-carbazole based hosts with their triplet energies labeled.

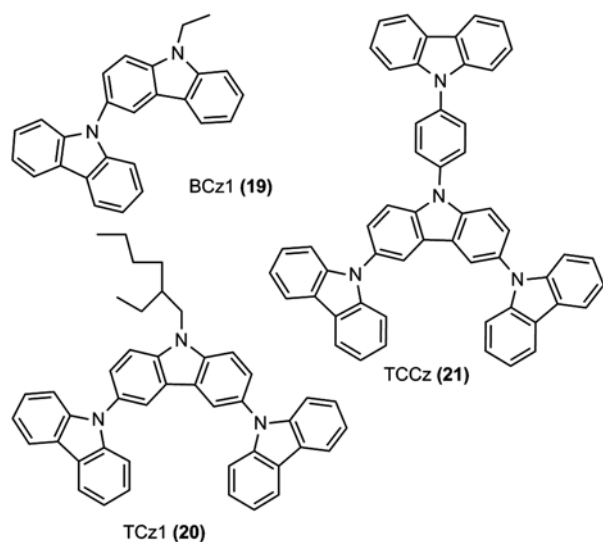


Figure 8 Carbazole oligomer hosts.

compared to the corresponding CBP (**7**) based device.

For blue triplet emitters, CBP (**7**) simply does not have sufficient triplet energy for T-T energy transfer from the host to the dopant, thus the inefficient endothermic energy transfer has to take effect [64], reducing the efficiency of triplet exciton formation in the dopant. To solve this problem, several dicarbazole derivatives with relatively short conjugation have been developed, such as *N,N'*-dicarbazolyl-3,5-benzene (mCP (**10**)) [65], 3,5-di(*N*-carbazolyl)tetraphenylsilane (SimCP (**11**)) [18], *N,N*-dicarbazolyl-1,4-dimethene-benzene (DCB (**12**)), $E_T = 2.95$ eV [66], and 9,9-bis[4-(3',6'-di-*tert*-butylcarbazol-9-yl)phenyl]fluorene (TBCPT (**13**)), $E_T = 2.88$ eV [67]. mCP (**10**) with the triplet energy of 3.0 eV was introduced as a blue phosphorescence host by Forrest's group [65]. They indicated that due to the exothermic T-T energy transfer by replacing host CBP (**7**) with mCP (**10**), the performance of FIrpic (**2**) based blue PhOLEDs was improved from 6.1% (7.7 lm/W) to 7.5% (8.9 lm/W). Modification of mCP (**9**) to SimCP (**11**) was carried out by Yeh *et al.* [18], yielding further improved device performance. The superiority of SimCP (**11**) as a phosphorescent host was further disclosed by the work from the same research group [68]. As demonstrated, mCP (**10**) and SimCP (**11**) have similar hole mobilities of 5×10^{-4} and 4×10^{-4} cm²/Vs, similar HOMO levels of 6.15 and 6.10 eV, respectively, as well as similar triplet energies. However, because of the inert and bulky substitution of triphenylsilyl, SimCP has higher T_g of 101 °C, compared to 55 °C of mCP (**10**).

Tokito *et al.* showed that the triplet energy of CBP (**7**) (2.6 eV) could be improved to 3.0 eV in 4,4'-bis(9-carbazolyl)-2,2'-dimethyl-biphenyl (CDBP, **14a**) by simple attachment of methyl groups to the 2,2' positions of the biphenyl core of CBP (**7**) to produce steric effect which twists the molecule and breaks the conjugation [69]. FIrpic (**2**) and

CDBP (**14a**) based blue PhOLED exhibited an efficiency of 10.4 % (20.5 cd/A, 10.5 lm/W). He *et al.* demonstrated that inserting non-conjugation groups between the biphenyl core of CBP (**7**) also resulted in breaking of the conjugation within the molecule, yielding material CBPCH (**14b**) [1,4-bis(4-(9*H*-carbazol-9-yl)phenyl)cyclohexane] with high triplet energy of 3.01 eV and high glass transition temperature of 115 °C [70].

4CZPBP (**15**) (2,2'-bis(4-carbazolylphenyl)-1,1'-biphenyl) is another example of good host with high triplet state for blue phosphor, which was proposed by the research group of Kido [71, 72]. Based on the emitting system of 4CZPBP (**15**) doped with FIrpic (**2**), high efficiencies can be obtained by two different device architectures. In one structure, besides utilizing high triplet host of 4CZPBP (**15**), a good ETL, bearing high triplet energy coupled with high electron mobility, was employed, which could suppress energy transfer from emitter FIrpic (**2**) to adjacent ETL, as well as lower the driving voltage, hence resulting in high power efficiency of 39 lm/W and EQE of 21% [71], which is one of the best performances for blue PhOLEDs. In another device, even higher efficiencies were obtained, with maximum values of 47.0 lm/W, 22.0%, and 50.5 cd/A [72]. In contrast to the first device, a weak electron transport ETL with a high energy gap was employed, which appears to conflict with the conventional notion that a weaker electron transport material as ETL results in lower efficiency [72]. This improvement is believed to be partially attributed to the more efficient block of the hole by the ETL material with a wide gap, and reflects that confinement of carriers within the triplet emission layer or at EML interface is more crucial than selecting good electron transport material as ETL in blue PhOLEDs.

Tsai *et al.* reported two host materials of 9-(4-*tert*-butylphenyl)3,6-bis(triphenylsilyl)-9*H*-carbazole (CzSi (**16**)) [73] and 9-(4-*tert*-butylphenyl)-3,6-bis(9-(4-methoxyphenyl)-9*H*-fluoren-9-yl)-9*H*-carbazole (DFC (**17**)) [31] containing mono-carbazole moiety (Figure 7), which are substituted at 3,6,9-position of carbazole. The designing strategies are as follows. First, the bulky triphenylsilyl or 9-fluorenyl moiety provides good thermal stability, and serves as an effective spacer to break the conjugation between the peripheral substitution and the π -conjugation of the carbazole core. Second, substitution on the 3,6-position of carbazole suppresses the electrochemical activity of these positions. As a result, these two materials are electrochemically stable with reversible oxidation potential as well as thermally stable with T_g of 131 °C for CzSi (**16**) and 180 °C for DFC (**17**). Concomitantly, they possess large triplet energy of 3.02 eV for CzSi (**16**) and 2.53 eV for DFC (**17**). FIrpic (**2**) and CzSi (**16**) based blue PhOLEDs achieved efficiencies of 16%, 30.6 cd/A, and 26.7 lm/W. Ir(ppy)₃ and DFC (**17**) based green PhOLEDs achieved an efficiency of 10%.

Compared to DFC (**17**), a very structurally similar material 9-phenyl-3,6-bis(9-phenyl-9*H*-fluoren-9-yl)-9*H*-carbazole

(CBZ1-F2 (**18**)) has been developed by another research group (see Figure 7) [74]. Flrpic (**2**) and CBZ1-F2 (**18**) based device yielded efficiencies of 10.2% and 19.2 cd/A. However, the differences in triplet energies between CBZ1-F2 (**18**) and DFC (**17**) are rather large, i.e., 2.53 eV for DFC (**17**), and 2.88 eV for CBZ1-F2 (**18**), considering that the molecular structures of CBZ1-F2 (**18**) and DFC (**17**) are similar, both having two fluorene units attached via the C-9 position to a carbazole moiety at the 3,6-position. The only difference between them is that DFC (**17**) contains optically inert groups of tert-butyl and methoxyl, which solely have very little effect on the photophysical property of materials [34]. Thus we suggest that this difference in triplet energy may be attributed to experimental diversity and needs to be studied in detail to get a clear insight into the underlying molecular design. It is noteworthy that a pure hydrocarbon aromatic compound belonging to fluorene derivatives was found to possess high triplet energy and could serve as a host for the blue triplet emitter, due to its rigid twisted tetrahedral molecular structure [75].

In the design of carbazole oligomers serving as triplet hosts, Brunner *et al.* analyzed the experimental data and concluded that the HOMO level of carbazole oligomers could be tuned by substitution at the 3,6- and/or 9-position while their triplet levels remained sufficiently high [59]. Tsai *et al.* studied carbazole oligomers of 9-ethyl-9*H*-3,9'-bicarbazole (BCz1 (**19**)) and 3,6-9'-dicarbazolyl-9-2''-ethylhexyl-carbazole (TCz1 (**20**)) (Figure 8) with linked topology of 3(6)-9' position. In consistent with the above conclusion, they found that 3(6),9'-linked carbazole oligomers possessed high triplet energy of 2.9–3.0 eV due to the suppressed delocalization of triplet excitons between carbazole units [76]. Blue PhOLEDs based on TCz1 (**20**) and Flrpic (**2**) gave maximum efficiencies of 14.7% (31.1 cd/A) and 28.4 lm/W. Similarly, carbazole oligomer *N*-(4-[9,3',6',9']tercarbazol-9'-yl)phenyl)carbazole (TCCz (**21**), Figure 8) has been employed as a host for green iridium dendrimers in PhOLEDs, achieving a maximum efficiency of 16.6% (57.9 cd/A) [77].

4.2.2 Tertiary amine based materials

Triphenylamine end-capped with fluorenes (tris[4-9-phenylfluoren-9-yl]phenyl)amine) (TFTPA (**22**)) [29] or carbazoles (4,4',4''-tri(*N*-carbazolyl) triphenylamine) (TCTA (**23**)) [27] was demonstrated to be efficient hosts for triplet emitters. Their molecular structures are shown in Figure 9. TFTPA (**22**) possesses high T_g of 186 °C, and is electrochemically as well as morphologically stable. PhOLED devices with TFTPA (**22**) host reached the maximum efficiencies of 13.1% (29.4 cd/A) and 18.1 lm/W for blue from Flrpic (**2**), 12.0% (44.1 cd/A) and 21.0 lm/W for green from Ir(ppy)₃ (**3**) and 9.6% (10.2 cd/A) and 9.0 lm/W for red from (piq)₂Ir(acac) (**6**). Although the triplet energies of TFTPA (**22**) (2.89 eV [29]) and TCTA (**23**) (2.85 eV [26]) are similar, there are rare previous reports with TCTA (**23**)

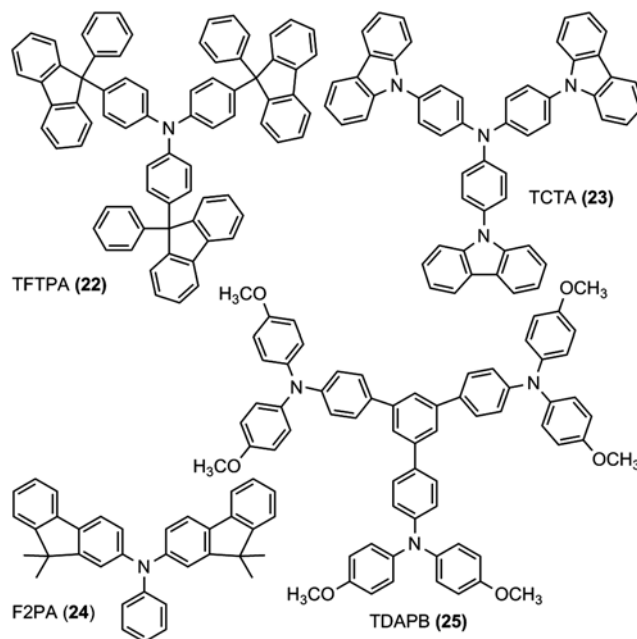


Figure 9 Tertiary amine based hosts.

as a host for triplet emitters other than Ir(ppy)₃ (**3**) [27, 32]. Nevertheless, when double EMLs consisting of TCTA (**23**) and CBP (**7**) as hosts were used, the Ir(ppy)₃ (**3**) based PhOLEDs could obtain extremely high efficiencies of 27% (95 cd/A) and 97 lm/W [26]. Another finding with the double EMLs employing CBP (**7**) and TCTA (**23**) as hosts is that Ir(ppy)₃ based device exhibits low efficiency roll-off at high current density [40]. Ir(ppy)₃ (**3**) based devices with the feature of small efficiency roll-off can also be obtained by utilizing the mixture of TCTA (**23**) and TPBI [1,3,5-tr(*N*-phenyl(benzimidazole-2-yl)benzene) as the host [41].

Other tertiary amines tested as hosts for triplet emitters include F2PA (**24**) [78] and TDAPB (**25**) [44] (Figure 9). However, the device performances based on them are not as promising as other hosts.

4.3 Electron-transport hosts

Toward searching hosts for triplet emitters, as shown in Figure 10, typical electron transport materials with functionalities of triazole (TAZ1 (**26**)) [34, 79], phenanthroline (BCP (**27**)) [34], oxadiazole (BOBP-3 (**28**)) [35, 38], and silole (si(bph)₂ (**29**)) [80]) were studied. Mainly due to their relatively low triplet energy, these electron transport materials are scarcely used for the blue triplet emitter.

Adachi *et al.* studied several electron transport materials as hosts for Ir(ppy)₃ (**3**), and found that TAZ1 (**26**) is better than BCP (**27**). The green device based on TAZ1 (**26**) and Ir(ppy)₃ (**3**) achieved an efficiency of 15.4% (42 lm/W) [34]. Later, they reported that with TAZ1 (**26**) and (ppy)₂Ir(acac) (**4**), the green device with the efficiency of 19.0% (65 lm/W) was obtained [79]. Oxadiazole derivatives (BOBP-3 (**28**)) as

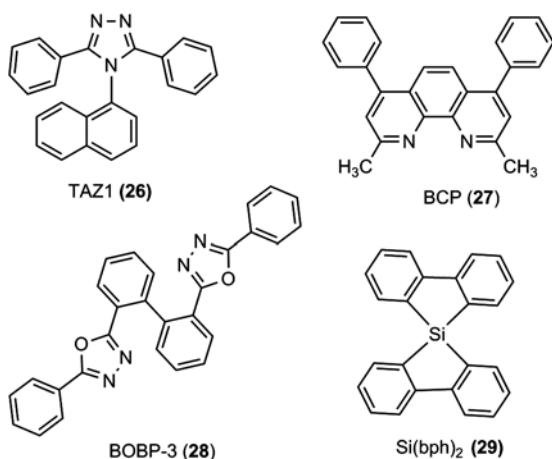


Figure 10 Typical electron transport materials serving as hosts.

triplet hosts were introduced by Lee and Leung *et al.* [35, 38]. BOBP-3 (**28**) and Ir(ppy)₃ (**3**) based device exhibits an efficiency of 24 cd/A (10 lm/W). This device was claimed of longer lifetime compared to the corresponding CBP (**7**) host based device [38]. Lee *et al.* regarded that EML consisting of BOBP-3 (**28**) (they termed as OXD) doped with Ir(ppy)₃ (**3**) was an ambipolar layer, due to the electron transport property of BOBP-3 (**28**) and the hole transport property of Ir(ppy)₃ (**3**) [35]. They obtained similar device performance as that in ref. [38]. A silole derivative with the triplet energy of 2.72 eV, 5,5'-spirobi (dibenzosilole) (Si(bph)₂ (**29**)), has been tried as the host for Ir(ppy)₃ (**3**) [80]. The device gave an efficiency of 4.5% at a luminance of 10000 cd/m², apparently not as efficient as other host materials based devices.

The introduction of carbazole into the electron transport functionalities of quinoline or triazine unit was claimed to yield electron transport materials of CQs (**30–33**) [81] or TRZ2 (**34**) [33] with favorable hole injection/transport features (Figure 11). When used as the host for Ir(ppy)₃ (**3**),

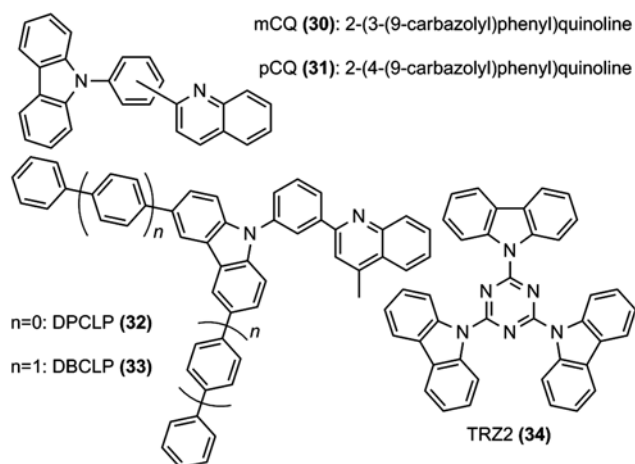


Figure 11 Carbazole containing electron transport materials as hosts.

TRZ2 (**34**) based device gave an efficiency of 11.2% (16.0 lm/W), and CQs (**30–33**) based device achieved an efficiency of 11.2%.

Electron transport phosphine oxides (PO1 (**35**), PO6 (**36**), and PO15 (**37**)), with chemical structures shown in Figure 12, were found to possess high triplet energies that are able to host blue phosphorescent emitters [82–86]. The best performance based on this kind of electron transport host materials is 8.1% (25.1 lm/W), using 10% FIrpic (**2**) doped into PO6 (**36**) host as EML [86]. As discussed in the following part, when these phosphine oxides are bipolar, the performance of FIrpic (**2**) based devices can be improved further.

4.4 Bipolar hosts

Theoretically, bipolar hosts are ideal. Besides simple device structure, bipolar hosts can offer balanced charge carriers in EMLs, making evenly and broadly distributed excitons possible. Thus, the exciton density can be reduced, leading to lower T-T annihilation and low efficiency roll-off. The most widely used strategy to tailor bipolar molecules is the combination of electron-rich (donor) and electron-deficient (acceptor) moieties to form a donor-acceptor type molecule. However, due to the possibility of charge delocalization between donor and acceptor moieties, this type of materials usually present a low energy band gap, and concomitantly, the triplet energy will decrease, making them less suitable for triplet emitters that require high triplet energy in order to prevent reverse energy transfer.

The research of bipolar hosts for triplet emitters is limited, most of which is within the last three years. Carbazole is the most popular functionality. As shown in Figure 13, bipolar materials taking carbazole as hole transport moieties include 4,7-di-carbazol-9-yl-[1,10]-phenanthroline [BUPH1 (**38**)] [37], 2,5-bis(2-(9H-carbazol-9-yl)phenyl)-1,3,4-oxadiazole [*o*-CzOXD (**39**)] [28], and 2,6-bis-(3-(carbazol-9-yl)phenyl)pyridine [26DCzPPy (**41**)] [87], which employ electron transport functional groups of phenanthroline, oxadiazole, and pyridine, respectively. The similar strategy for the design of the above molecules is the suppression of electron delocalization between the donor and acceptor moieties by

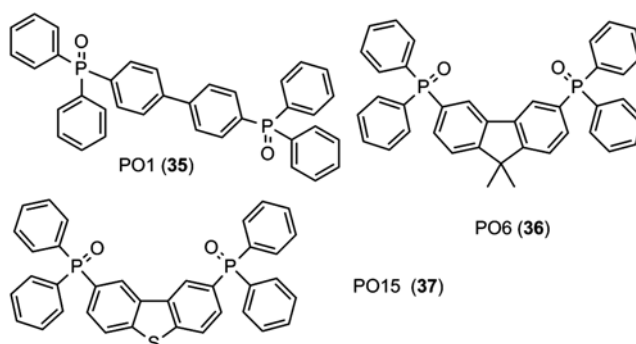


Figure 12 Phosphine oxides as hosts for triplet emitters.

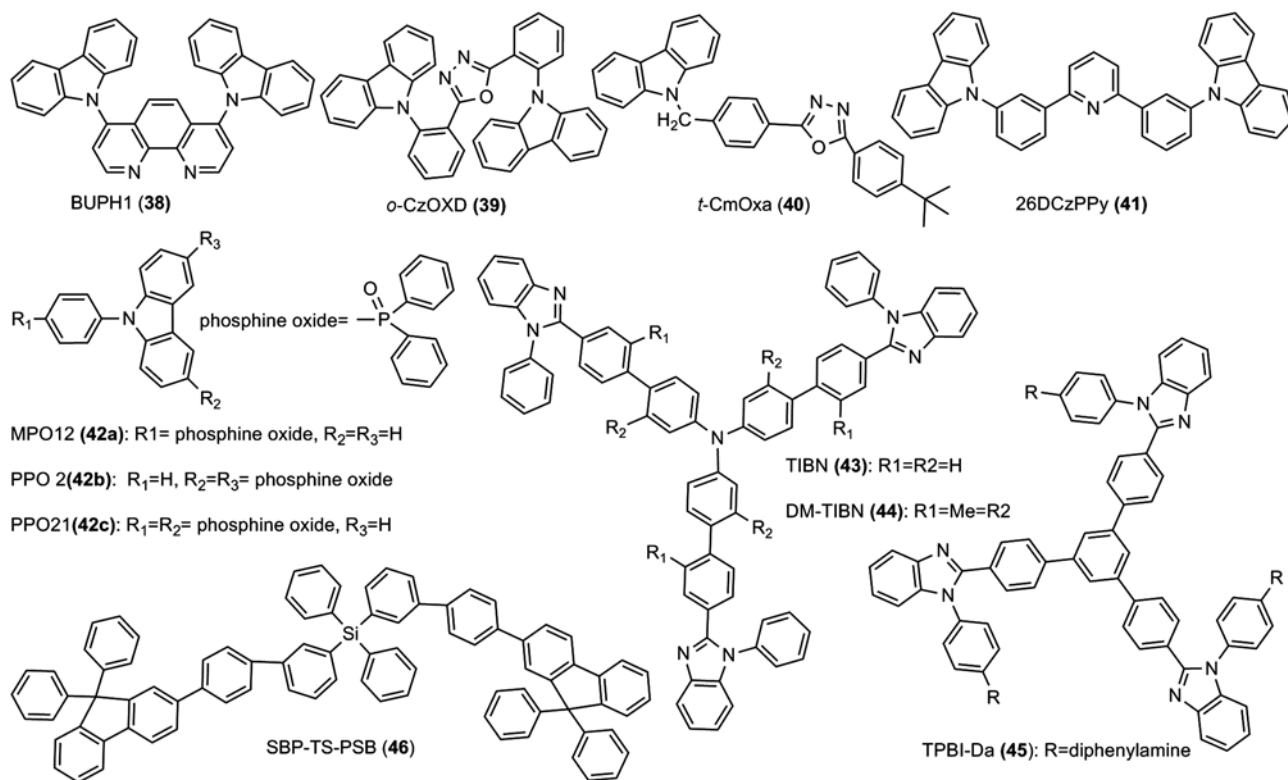


Figure 13 Bipolar host materials.

breaking the conjugation between them in order to keep a high energy gap and relatively large triplet energy.

We introduced bipolar host BUPH1 (38) as the triplet host and demonstrated its promising potential in PhOLEDs [37]. By peripherally capping two hole transport carbazole moieties to the electron transport phenanthroline core at the 4,7-position, BUPH1(38) molecules were demonstrated to be a bipolar material with high thermal stability ($T_g=155\text{ }^\circ\text{C}$ and $T_d=449\text{ }^\circ\text{C}$). Because the carbazole attachment via its 9-position of the non-conjugated sp^3 N atom effectively suppresses the conjugation between carbazole and phenanthroline, the band gap of BUPH1(38) (3.1 eV) as well as its triplet energy (2.4 eV) are reasonably high to accommodate triplet dopants of green to red triplet emitters. Mainly due to the good bipolar-transport property of the host, Ir(ppy)₃ (3) doped BUPH1(38) PhOLEDs achieved very low driving voltages of 2.6, 3.5 and 5.3 V for current density of 1, 10, and 100 mA/cm², and exhibited power efficiencies of 33 and 20 lm/W at 100 and 1000 cd/m², respectively.

o-CzOXD (39), another promising bipolar triplet host with two carbazole units and one oxadiazole hybridizing, was introduced by Tao *et al.* [28]. They held that due to the twist structure between the donor and the acceptor, the intramolecular electron delocalization from carbazole to oxadiazole was reduced, thus high triplet energy level of 2.68 eV was obtained. *o*-CzOXD (39) is thermally stable with T_g of 97 °C and T_d of 428 °C. Devices containing *o*-CzOXD (38) as the

host show maximum efficiencies of 20.2% (77.9 cd/A) for green from Ir(ppy)₃ (3) and 18.5% (13.6 cd/A) for deep red based on (piq)₂Ir(acac) (6). Another carbazole and oxadiazole hybrid material 9-[4-[5-(4-*tert*-butylphenyl)-[1,3,4]oxadiazol-2-yl]-benzyl]-9*H*-carbazole (*t*-CmOxa (40)) with a large singlet energy gap of about 3.5 eV was synthesized as the host for red phosphor [88]. The device based on *t*-CmOxa (42) and bis (dibenzo [f,h] quinoxalino) (acetylacetonato) Ir(III) exhibits efficiencies of 9.5% and 9.9 lm/W.

A star-shaped molecule, tris(2,2'-dimethyl-4'-(1-phenyl-1*H*-benzimidazol-2-yl)biphenyl-4-yl)amine (DM-TIBN (44)), with hole-transport triphenylamine attached to electron-transport benzimidazole moieties, was reported to be a bipolar host with relatively large triplet energy of 2.77 eV, attributed to the localization of HOMO and LUMO at its respective hole- and electron-transporting moieties (triphenylamine and benzimidazole, respectively) [43]. The solution processed DM-TIBN (44) and Ir(ppy)₃ (3) based device exhibits maximum efficiencies of 27.3 cd/A and 7.3 lm/W. However, the driving voltages are rather high, e.g., the luminance turns on at 6.9. Takizawa *et al.* demonstrated that the hybridization of starburst benzimidazole derivative TPBI (1,3,5-tris(*N*-phenylbenzimidazol-2-yl)-benzene) core with diphenylamine could result in bipolar material TPBI-Da (45). The bilayer device based on TPBI-Da (45) and Ir(ppy)₂acac (4) achieved an efficiency of 15% (59.2 cd/A) [89].

When two spirobifluorene units with the feature of ambipolarity are attached to non-conjugated tetraphenylsilane inert core, a bipolar and thermally stable molecule, bis(4'-(9,9-di-phenyl-9H-fluoren-2-yl)biphenyl-3-yl)diphenylsilane (SBP-TS-PSB (**46**), $T_g = 194$ °C, Figure 13), with high triplet energy of 3.04 eV is achieved [90]. (piq)₂Ir(acac) (**6**) and SBP-TS-PSB (**46**) based red PhOLED shows a maximum efficiency of 14.6% (10.3 lm/W).

Bipolar hosts for blue triplet emitters are hard to obtain, because the requirement of even larger triplet energy than that of the blue triplet phosphor puts further strain on donor-acceptor type molecules. It has been found that the inductive withdrawing of electrons by phosphine oxide moiety was able to tune the HOMO and LUMO of carbazole compounds while preserving high triplet energy [91]. For instance, when diphenylphosphoryl group connects with carbazole at the *N*-phenyl position, *N*-(4-diphenylphosphorylphenyl)carbazole (MPO12 (**42a**), Figure 13) and PPO2 (**42b**) [3,6-bis(di-phenylphosphoryl)-9-phenyl-9H-carbazole] are ambipolar due to the spatial separation of the HOMO and LUMO on different parts of the molecule. The blue device based on MPO12 (**42a**) and FIrpic (**2**) gave an efficiency of 9.1% with a low driving voltage of 4.8 V at brightness of 800 cd/m² [83]. The blue device based on PPO2 (**42b**) and FCNIr [tris((3,5-difluoro-4-cyano-phenyl)pyridine) iridium] gave an efficiency of 18.4% [84]. In 2010, it was reported in *Adv. Mater.* that a similar material (PPO21, **42c**) with carbazole and phosphine oxide moieties, [3-(diphenylphosphoryl)-9-(4-(diphenyl-phosphoryl)phenyl)-9-carbazole], exhibited bipolar carrier transport property and the device based on it and FCNIr exhibited an efficiency of 18%. However, the stability of the device was not satisfactory [85].

Another case of bipolar host for blue phosphor was the hybrid of high triplet electron donor of carbazole with high electron affinity acceptor of pyridine [87]. The resulted high triplet compound (26DCzPPy (**41**), $E_T = 2.71$ eV) was demonstrated to be able to transport both holes and electrons. FIrpic (**2**) and 26DCzPPy (**41**) based blue PhOLEDs achieved efficiencies of 24% and 46 lm/W with low roll-off at high current density.

4.5 Inert hosts

A series of tetraarylsilanes were investigated as hosts for triplet emitters [80, 92–94], including UGH1 (**47**), UGH3 (**48**), UGH2 (**49**), BSB (**50**) and BST (**51**), as shown in Figure 14. These materials exhibit large energy gaps (3.8 to 4.4 eV) and deep HOMO levels (6.3 to 7.2 eV). Due to their deep HOMO levels and inertness in conductivity, the devices employing them as phosphor-hosts need high driving voltage and the excitation mechanism is direct carrier trapping by the dopant. For instance, BSB (**50**) and FIrpytz (**1**) based blue PhOLED gave high efficiencies of 19.3%, 37.8 cd/A and 16.8 lm/W, but its turn-on voltage was as high as

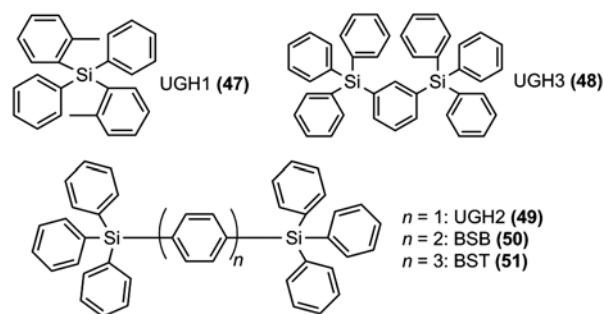


Figure 14 Inert host materials.

5.1 V [93].

4.6 Fluorescent metal-complex hosts

Fluorescent metal complexes of Zn(BTP)₂ (**52**) [95, 96], Be(bq)₂ (**53**) [97] and Be(pp)₂ (**54**) [36], shown in Figure 15, have been found useful as hosts for green to red iridium complexes in PhOLEDs, and the similarity among these materials based devices is low driving voltage as well as relatively simple device structure. Zn(BTP)₂ (**50**) with triplet energy of 2.5 eV can serve as an efficient host for Ir(piq)₃ (**5**), leading to an efficiency of 8.6 cd/A (8.8 lm/W and 10.3%). Be(bq)₂ (**53**) was claimed to transport electrons and have very small exchange energy between the singlet and triplet state. When it was employed as a host, Ir(piq)₃ (**5**) based device with a simple bilayered structure achieved a red emission with an efficiency of 9.7 cd/A (6.9 lm/W). The device using electron transporting Be(pp)₂ (**54**) as the host for Ir(ppy)₃ (**3**) was reported from the same group. Employing a simple double-layer device structure, Be(pp)₂ (**54**) and Ir(ppy)₃ (**3**) based device produced a green emission with an efficiency of 38.3 cd/A (46.6 lm/W).

4.7 Phosphorescent metal-complex hosts

As discussed in the introduction section, if the host is a phosphorescent material, by proper selection, the long-range and rapid Förster energy transfer can be the dominant mechanism in PhOLEDs, hence the typical requirement of high concentration in the system of fluorescent hosts can be ignored. Furthermore, another expected advantage of using phosphorescent hosts is the low driving voltage, because in

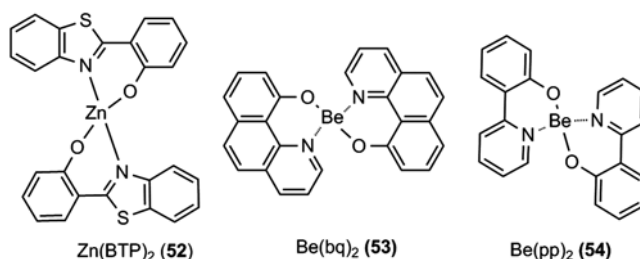


Figure 15 Fluorescent metal complex hosts.

principle, the phosphorescent materials generally have small band gaps compared to fluorescent materials having similar triplet energy, thus the resulted shallow level of HOMO or deep level of LUMO will facilitate hole or electron injection, reducing the driving voltage. This aspect of research is rare, partially due to the expensiveness of phosphorescent materials, making them unfavorable to be consumed in a large scale such as hosts. The device based on a platinum complex emitter and a phosphorescent host of Ir(ppy)₃ (**3**) is one of the early work [98].

The structures of some phosphorescent hosts are shown in Figure 16. Green phosphorescent (ppy)₂Ir(acac) (**4**) was studied as the host for the red emitter Ir(piq)₃ (**5**) by Tsuzuki *et al.* [99]. It was found that (ppy)₂Ir(acac) (**4**) was a good hole transport material with the energy gap of 2.4 eV and triplet energy of 2.3 eV. The optimized concentration of Ir(piq)₃ (**5**) in (ppy)₂Ir(acac) (**4**) host based device is only 0.3%. This device shows a maximum efficiency of 11 lm/W, reflecting low driving voltage. However, the efficiency roll-off at high current density is large. Another study using phosphorescent host in PhOLED was reported by the same group, where the solution process was employed after modifying the host (ppy)₂Ir(acac) (**4**) to (ppy)₂Ir(C9-acac) (**55**) or (ppy)₂Ir(Cy6-acac) (**56**) [100]. The maximum efficiency of 10% for red PhOLED was achieved.

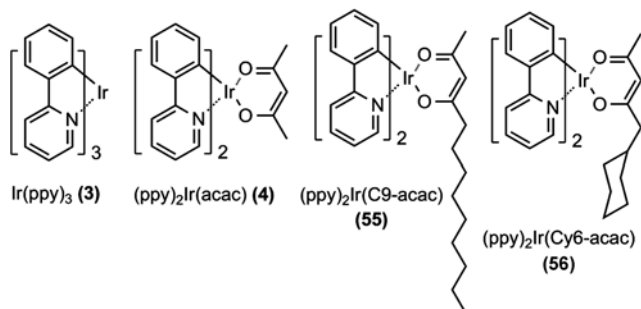


Figure 16 Phosphorescent host materials used in PhOLEDs.

5 Perspective

Research has demonstrated that better choice of suitable hosts for triplet emitters in PhOLED can not only improve device efficiency, but also reduce the efficiency roll-off at high current density. The improvement in device stability has been demonstrated as well [101]. There are apparent achievements in the pursuit of good hosts for the blue triplet emitter. For solid state lighting, the efficiencies of white PhOLEDs are high enough for applications. Besides small molecular hosts discussed in this review, polymer hosts for triplet emitters also have been intensively studied [102]. Nevertheless, unknown factors and problematic issues remain in PhOLEDs. For instance, due to the tough requirements of both large bandgap and high triplet energy, there is

a lack in satisfactory hosts for blue phosphors. Another limitation in PhOLED research is that most of the work on host materials in PhOLEDs emphasizes the efficiency improvement. Studies of the impact on device lifetime by the introduction of the hosts are rare, especially for blue emission. Further research for satisfying different applications and reaching the intrinsic limitation of PhOLED technology may be directed in simultaneous improvement of the efficiency and lifetime, reduction of the driving voltage and achievement of small efficiency roll-off at high current density.

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