Exciplex hosts for blue phosphorescent organic light-emitting diodes

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ABSTRACT
The host material of organic light-emitting diodes (OLEDs) has been advanced from a single host to a mixed host for high efficiency and long lifetime. Several types of mixed host have been reported in the literature, but the exciplex host has been popular as the mixed host of OLEDs. The exciplex host has been developed mostly for red and green phosphorescent OLEDs, and has upgraded device performances, but it is difficult to develop the exciplex host for blue phosphorescent OLEDs. Recently, several works demonstrated the potential of the exciplex host for blue phosphorescent OLEDs. In this paper, the exciplex host for blue OLEDs is reviewed, and its prospects are presented.

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1. Introduction
Exciplexes are known as the intermolecular charge transfer complex of two or more organic materials driven by the electron-donor/acceptor-type material characteristics [1–7]. They are very popular as the host of phosphorescent organic light-emitting diodes (OLEDs) because they can provide a low driving voltage [8–10], high external quantum efficiency (EQE) [11–14], and a long device lifetime [15–17].

In general, exciplexes have at least one p-type (hole transport type) host and one n-type (electron transport type) host, which generate an intermolecular charge transfer complex. The p-type host plays the role of an electron donor, and the n-type host plays the role of an electron acceptor in the exciplex. When carriers are injected from the charge transport layers to the emitting layer, the holes are mostly carried by the p-type host, and the electrons are mainly carried by the n-type host. Hole and electron carrier transport independently happens in the p- and n-type hosts, and then the electrons in the n-type host recombine with the holes in the p-type host, forming intermolecular charge transfer excitons. The excitons can be converted to photons, and the exciton energy can be transferred to dopant molecules when the dopant is dispersed in the exciplex host. Therefore, the exciplex can emit by itself as the emitter [18–20], and can also work as the host of the phosphorescent [21–23] or thermally activated delayed fluorescent emitters [24]. The exciplex, however, is not as efficient as the high-efficiency emitters because of the non-radiative loss process, which encourages the use of the exciplex as the host rather than the emitter. The working process of the exciplex as the emitter and host is schematized in Figure 1.

The exciplex has several advantages as the host of the phosphorescent emitter. First, the driving voltage is low due to the efficient carrier injection and transport through the exciplex host materials [25]. As the exciplex has at least one p-type host and one n-type host, the carrier injection and transport in the emitting layer are relatively easy compared to that in a single host. The p-type host is commonly a strong hole-transport-type host with a shallow highest occupied molecular orbital (HOMO) and good hole transport properties. The shallow HOMO level of the p-type host facilitates the hole injection from the hole transport layer, and the improved hole transport properties prevent hole accumulation at the interface between the hole transport layer and an emitting layer. Likewise, the n-type host with a strong electron transport character and a deep lowest unoccupied molecular orbital (LUMO) assists the electron injection from the electron transport layer to the emitting layer, and suppresses electron accumulation at the interface between these two layers. Therefore, the driving voltage of OLEDs governed by the energy barrier for carrier injection and carrier mobility is lowered in the exciplex-based phosphorescent OLEDs [26–28].
Second, the efficiency is high and the efficiency roll-off is quite small in the exciplex-hosted OLEDs [29,30]. The efficiency is dominated by the hole-electron balance in the emitting layer [31–33], and can be maximized in the exciplex-based devices because both holes and electrons can be efficiently injected and transported in the emitting layer by the p- and n-type hosts. Simultaneously, the efficiency roll-off is also improved by the exciplex host because exciton quenching mechanisms at high luminance are suppressed. The main mechanisms of the efficiency roll-off at high luminance are triplet-triplet annihilation (TTA) and triplet-polaron annihilation (TPA) [34–38]. The TTA process is closely related to the exciton density in the emitting layer, and a low exciton density is preferred for the reduced TTA process. The exciplex is advantageous because the emission zone in the emitting layer is broadened by an efficient hole and electron injection and transport [39,40]. The holes and electrons recombine over the whole emitting layer, and the excitons are uniformly distributed in the emitting layer. Therefore, the local triplet exciton density is relatively low, which minimizes the TTA process. This is also helpful for reducing the TPA process in the emitting layer. The TPA process occurs through a collision between triplet excitons and polarons, indicating that polaron formation in the phosphorescent emitter needs to be avoided to suppress the TPA mechanism. When the exciplex host is used in the emitting layer, direct hole and electron trapping by the phosphor is difficult because of the small HOMO and LUMO gaps between the host and the phosphor. Moreover, the dominant energy transfer from the exciplex host to the phosphor also hinders the TPA process in the emitting layer. Therefore, the reduced TTA and TPA processes improve the efficiency roll-off of exciplex-based devices.

Third, the device lifetime is extended by the exciplex host. Several factors dominate the device lifetime of the phosphorescent OLEDs. Among these, the TTA and TPA degradation mechanisms have been known to be critical for the phosphorescent device lifetime [41–43]. As described above, both the TTA and TPA mechanisms can be controlled by the exciplex host due to the wide recombination zone and energy-transfer-dominated emission mechanism with little charge trapping, which elongates the operation lifetime of the phosphorescent OLEDs. Other than the TTA and TPA mechanisms, improved polaron stability is also partially responsible for the elongated device lifetime of the exciplex-based devices [44,45]. Compared to the single host, which carries holes and electrons at the same time, the p- and n-type hosts carry only holes or only electrons. Generally, the p-type host is designed to be stable under holes, and the n-type host is designed to be stable under electrons. This indicates that the p- and n-type hosts in the exciplex host can be stable during electrical operation. The single host, however, is exposed to both holes and electrons, and cannot survive under two different conditions. Therefore, the device lifetime is improved when the exciplex is used as the host of phosphors.

2. Exciplex host for sky blue phosphorescent OLEDs

As earlier described, the exciplex is favored as the host of phosphorescent OLEDs and has been widely used as the host of red and green phosphorescent OLEDs. It is difficult to develop the exciplex host for blue phosphorescent OLEDs, however, because of the high triplet energy requirement. The common blue phosphorescent
dopants have 2.6–2.7 eV triplet energy, indicating that the exciplex host should have over 2.7 eV triplet energy for energy transfer. In general, the emission energy of the exciplexes is governed by the HOMO of the p-type host and the LUMO of the n-type host. As the exciplex is formed between a strong p-type host and a strong n-type host by the HOMO-LUMO offset, the singlet and triplet energies of the exciplexes are limited. The large HOMO-LUMO offset between the two hosts is desired for the large singlet and triplet energies, and this requirement can be met when weak p- and n-type hosts are used to form the exciplex [46]. Therefore, proper management of the p- and n-type hosts is critical for the development of the exciplex hosts for blue phosphorescent OLEDs.

In the development of the exciplex host for blue phosphors, either a weak p-type host or a weak n-type host needs to be employed to increase the triplet energy of the exciplex. The first exciplex host for blue phosphorescent OLEDs was the N,N′-dicarbazolyl-3,5-benzene (mCP):bis-(3,5-di-3-pyridylphenyl)-2-methylpyrimidine (B3PYMPM) host [47]. mCP was a weak p-type host, and B3PYMPM was a strong n-type host. The 0.67 eV HOMO level gap and 0.7 eV LUMO level gap between the two hosts drove the exciplex formation. The mCP:B3PYMPM showed exciplex emission in the blue wavelength range, and harvested triplet excitons of sky-blue-emitting bis[2-(4,6-difluorophenyl)pyridinato]iridium(III) (FIrpic) dopant. Full harvesting of the triplet excitons of FIrpic and assistance from the horizontal dipole orientation realized a high EQE of 29.5% in the sky blue phosphorescent OLEDs.

A similar approach adopting the weak p-type host and the strong n-type host was reported by the same group. The two hosts for the exciplex formation were mCP and (1,3,5-triazine-2,4,6-triyl)-tris(phenylphosphineoxide) (PO-PH) compounds were also classified as a weak p-type and strong n-type hosts combined exciplex [49]. Instead of B3PYMPM, the strong n-type PO-T2T host was employed to develop the exciplex host. The large LUMO level difference by the deep LUMO level (−3.5 eV) of the PO-T2T enabled exciplex formation between mCP and PO-T2T. FIrpic phosphor was doped in the exciplex, which enabled a high EQE of 30.3% in the sky blue phosphorescent OLEDs. A low driving voltage of 2.75 V at 100 cd/m² was also achieved by energy-barrier-free carrier injection.

The exciplex host of mCP:1,1′,1″′-(pyridine-2,4,6-triyl)tris(2-phenyl-1H-benzo[d]imidazole (iTPBIPy) can also be classified as a weak p-type and strong n-type hosts combined exciplex [49]. 2,2,2′-((3,5-phenylene)tris(1-phenyl-1H-benzimidazole (TPBI) is well known as a weak n-type host, and it was modified with a pyridine unit to strengthen the electron transport character of TPBI. Three TPBI-derived n-type hosts were applied as the n-type host to generate exciplex hosts, among which the mCP:iTPBIPy exciplex host provided the best EQE of 19.3% in the sky blue phosphorescent OLEDs. The application of the exciplex host enhanced the EQE and reduced the driving voltage of the phosphorescent OLEDs. The relatively low triplet energy (~2.65 eV) of the TPBI derivatives, however, limited their application in sky blue phosphorescent devices.

In addition to the weak p-type host and strong n-type host combined exciplex host, strong-p-type-host- and weak-n-type-host-based exciplex hosts were reported for blue phosphors. Several weak n-type hosts were designed, and they were mixed with common p-type hosts like 4,4,4-tris(N-carbazolyl)triphenylamine (TCTA) and di-[4-(N,N-ditolyl-amino)-phenyl)cyclohexane (TAPC). TAPC:5′′′-sulfonyle-di-1,1′:3′,1″-terphenyl (BTPS) is an early exciplex host for blue phosphorescent OLEDs [50]. It was built on the basic platform of a strong p-type host and a weak n-type host. The BTPS n-type host worked as the weak n-type host because the weak electron-withdrawing diphenylsulfone core was used in the backbone structure. The singlet and triplet energies of the exciplex hosts were 2.97 and 2.82 eV, respectively, which could induce energy transfer from the exciplex host to the FIrpic dopant. The high power efficiency of 50 lm/W, the low driving voltage of 2.9 V, and a high EQE of 21.7% were claimed using the TAPC:BTPS exciplex host doped with a FIrpic dopant.

Another weak n-type host for the exciplex hosts was (5-Terphenyl-1,3-Phenylene)bis(diphenylphosphine oxide) (POPH), which has an aromatic-based backbone structure modified with two diphenylphosphine oxide units [51]. Diphenylphosphine oxide usually works as a weak electron-withdrawing unit that does not degrade the triplet energy of the main skeleton. Therefore, the POPH host was designed as the weak n-type host to allow exciplex formation with a p-type host. The p-type hosts in the exciplexes were TAPC, TCTA, and mCP. All the p-type hosts exhibited exciplex emission by mixing with the POPH host. Among the three exciplexes, the TCTA:POPH host showed the best device performances as the host of solution-processed FIrpic devices. A low turn-on voltage of 2.7 V, 25.8 cd/A current efficiency, and 22.5 lm/W power efficiency were demonstrated using the TCTA:POPH exciplex host and the FIrpic dopant.

1,4-phenylenebis(diphenylphosphine oxide) (2PO) and ((phenylphosphoryl) bis(4,1-phenylene))bis(diphenylphosphine oxide) (3PO) compounds were also weak n-type hosts for solution-processed phosphorescent OLEDs [52]. The 2PO and 3PO hosts were designed to have two and three diphenylphosphine oxide units, respectively, for an n-type character. The two molecules were synthesized as the weak n-type host with a wide

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Figure 2. Chemical structures of the p- and n-type hosts for the exciplexes of sky blue phosphorescent OLEDs.

HOMO–LUMO gap and high triplet energy. The 2PO and 3PO hosts were mixed with TCTA, which afforded exciplex hosts with 2.73 and 2.76 eV triplet energies, respectively. A 14.6% EQE and 29.2 cd/A current efficiency were obtained in the FIrpic-doped blue phosphorescent OLEDs. Moreover, the efficiency roll-offs of the TCTA:2PO and TCTA:3PO devices were only 3.5% and 0.4% at 1,000 cd/m², respectively.

The 1,3,5-tris(diphenylphosphoryl)benzene (TPO) host is also a diphenylphosphine-oxide-modified n-type host [53]. TPO is quite similar to the 3PO host in that they share phenyl and diphenylphosphine oxide units. TPO and 3PO are isomers with the same building blocks. TPO was used as the n-type host of the exciplex host for the solution process due to its good solubility. The triplet energy of the TCTA:TPO exciplex was 2.77 eV for blue phosphorescent device application. The solution-processed devices doped with FIrpic achieved high current efficiency (23.8 cd/A) and power efficiency (15.8 lm/W). The TPO and 3PO hosts were comparable as the n-type host of the exciplex host.

The phosphine oxide modification approach was also applied to design the 1,3,5-tris(1-(4-(diphenylphosphoryl)enyl)-1H-benzo[d]imidazol-2-yl)benzene (TPOB) host derived from the TPBI host [54]. The weak n-type character of TPBI was intensified by additional phosphine oxide units. The phosphine oxide units additionally improved the solubility of the host in organic solvents. The mixing of TPOB with TCTA formed an exciplex with 2.67 eV triplet energy. The introduction of the diphenylphosphine oxide unit twisted the TPBI backbone structure, which increased the solubility and triplet energy. It also deepened the LUMO level of the host for facile electron injection. The solution-processed FIrpic devices exhibited a low turn-on voltage of 2.8 V and improved efficiency roll-off in addition to a maximum EQE of 13.8%.

The chemical structures of the p- and n-type hosts that were used for the exciplexes of sky blue phosphorescent OLEDs are shown in Figure 2, and the device performances are shown in Table 1.

3. Exciplex host for deep-blue phosphorescent OLEDs

Although the appropriateness of several exciplexes as the host of blue phosphors was verified, they were suitable only for sky blue phosphors and could not be used for deep-blue phosphors because their triplet energy was not sufficiently high to harvest triplet excitons of deep-blue phosphors. The triplet energy of the exciplex host required for deep-blue phosphors is above 2.8 eV. To satisfy the requirement of the exciplex host for deep-blue phosphors, design strategies to increase the triplet energy of the exciplex have been reported. One such design strategy is to increase the HOMO–LUMO offset between the p- and n-type hosts. In this case, the two hosts should have higher triplet energy than the phosphor, and the HOMO and LUMO gaps should be large enough to drive exciplex formation. One example is the exciplex between mCP and 2,4-bis(4-(diphenylphosphoryl)phenyl)pyridine (BM-A10) [55]. The mCP was a weak p-type host with a deep HOMO level, and the BM-A10 was a weak n-type host with a shallow LUMO level. The matching of the HOMO and LUMO levels in the two hosts resulted in the exciplex with 3.0 eV
Table 1. Device performance of sky blue OLEDs that used the exciplex host system.

| EML structure (host/dopant) | $V_{on}$ [V] | EQE [%] | LE [lm W$^{-1}$] | J [Cd A$^{-1}$] | CIE (x, y) | Ref. |
|-----------------------------|-------------|---------|-----------------|-------------|-------------|------|
| mCP:B3PYMPM/FIrpic          | 3           | 29.5    | 55.4            | 62.2        | –           | [48] |
| mCP:BOD-T2T/FIrpic          | 2.75        | 30.3    | 66              | –           | –           | [48] |
| mCP:TBPB/FIrpic             | 3.2         | 19.3    | 36.9            | 38.5        | (0.15, 0.29)| [49] |
| TAPC:BTPS/FIrpic            | 2.9         | 21.7    | 50              | 46.1        | –           | [50] |
| TCTA:PO-PH/FIrpic           | 2.7         | –       | 22.5            | 25.8        | (0.15, 0.32)| [51] |
| TCTA:2P0/FIrpic             | –           | 14.6    | –               | 29.2        | –           | [52] |
| TCTA:3P0/FIrpic             | –           | 12.2    | –               | 26.8        | –           | [52] |
| TCTA:TPO/FIrpic             | 3.0         | –       | 15.8            | 23.8        | (0.16, 0.33)| [53] |
| TCTA:TPOB/FIrpic            | 2.8         | 13.8    | 22.0            | 28.2        | (0.15, 0.33)| [54] |

The high triplet energy of the exciplex host confined the triplet excitons of the tris((3,5-difluoro-4-cyanophenyl)-pyridine)iridium (FCNIr) dopant with little loss, and afforded a high EQE of 24% in the deep-blue phosphorescent OLEDs with deep-blue color coordinates of (0.15,0.21).

The other design strategy to increase the triplet energy of the exciplex is to weaken the exciton binding between the p- and n-type hosts because the strong binding of the two hosts decreases the emission energy. Two p-type hosts, 3,5-di(9H-carbazol-9-yl)-N,N-diphenylaniline (DCDPA) and N,N-bis(4-(tert-butyl)phenyl)-3,5-bis(3,6-di-tert-butyl-9H-carbazol-9-yl)amine (t-DCDPA) [56], were compared as the p-type hosts in combination with the 2,8-bis(4,6-diphenyl-1,3,5-triazin-2-yl) dibenzofuran (DBFTrz) n-type host. The t-DCDPA was designed to have t-butyl blocking groups around the molecule to control the exciton binding between the p- and n-type hosts. In general, the electron and hole are captured by the Coulomb attraction between two molecules, and they are bound together. Therefore, the exciton binding energy might be decreased by the t-butyl blocking group due to the weak Coulombic force [57,58]. Although the HOMO–LUMO offset was reduced by the shallow HOMO level of the t-DCDPA, the singlet and triplet energies of the t-DCDPA:DBFTrz exciplex were quite similar to those of the DCDPA:DBFTrz, proving the validity of the exciton binding energy control through the sterically hindered molecular structure. The DCDPA: DBFTrz and t-DCDPA:DBFTrz exciplexes well performed as the exciplex host for deep-blue phosphorescent OLEDs doped with FCNIr by providing high EQEs of 16.4% and 15.3%, respectively, along with deep-blue color coordinates of (0.14,0.19) and (0.14,0.20).

Although the exciplex hosts assisted in reaching the high EQE in the blue phosphorescent OLEDs, the lifetime of the exciplex-based blue phosphorescent devices was not reported because of the instability of the Irpic and FCNIr dopant. The lifetime study of the exciplex-based blue phosphorescent OLEDs was carried out using fac-tris(3-(1-(2,6-diisopropylphenyl)-1H-imidazol-2-yl)benzonitrile)iridium (Ir(CNpi))-doped blue phosphorescent devices. Ir(CNpi) is a deep-blue-emitting phosphor that can show a high EQE of over 20%. Therefore, exciplex hosts for Ir(CNpi) were developed using the molecular structure with the CN unit and the CN-modified carbazole unit. An n-type host named 9-(3′-(9H-carbazol-9-yl)-5-cyano-[1,1′-biphenyl]-3-yl)-9Hcarbazole-3-carbonitrile (CNmCBPCN) [59] was designed as the n-type host, and it generated an exciplex with a p-type 2,2′-di(9H-carbazol-9-yl)-1,1′-biphenyl (oCBP) host. It did not generate an exciplex with a weak p-type 3,3′-di(9H-carbazol-9-yl)-1,1′-biphenyl (mCBP), but it generated an exciplex with oCBP with a relatively shallow HOMO level for a small HOMO-LUMO offset with the CNmCBPCN. The oCBP:CNmCBPCN exciplex showed 2.94 eV triplet energy and confined the triplet excitons of Ir(CNpi). The EQE of the Ir(CNpi)-doped oCBP:CNmCBPCN device was 18.8% after the triplet harvesting of Ir(CNpi). In particular, the exciplex host extended the lifetime of the blue phosphorescent OLED 1.8 times compared to the single host, confirming the role of the exciplex host as a lifetime-elongating host.

The chemical structures of the p- and n-type hosts used for the exciplexes of deep-blue phosphorescent OLEDs are shown in Figure 3, and the device performances are shown in Table 2. Additionally, the EQE of the sky and deep-blue phosphorescent OLEDs are plotted against the emission wavelength in Figure 4.

4. Summary

The exciplex host for blue phosphors effectively improved the EQE of the blue phosphorescent organic light-emitting diodes (OLEDs) and reduced the driving voltage. It proved to have a potential as the host of blue phosphors. Several challenges remain, however, in the development of the exciplex host for blue phosphors. First, the number of exciplex hosts for deep-blue phosphors is limited, and further exploration of a high-triplet-energy exciplex host with higher than 2.90 eV triplet energy (onset energy of phosphorescence spectra) is needed. The current approaches to increasing...
the triplet energy of the exciplex host are managing the HOMO-LUMO offset and exciton-binding energy between the p- and n-type hosts. To control the HOMO-LUMO offset, the database of high-triplet-energy p- and n-type hosts has to be expanded by developing numerous hosts. The exciton binding energy can be manipulated by designing a molecular structure that can regulate the geometrical structure for large separation and weak binding between molecules. Geometrical control of the backbone structure and bulky blocking groups can relieve the tight binding of the molecules and increase the triplet energy. Second, the lifetime of the exciplex-based blue phosphorescent OLEDs needs to be extended. Only one lifetime result of the blue phosphorescent OLEDs has
been reported using the exciplex host. Although several exciplex hosts for blue phosphors have been developed, their lifetime was not validated possibly due to the poor stability of the blue phosphors like Flrpic and FCNIr. Further study of the device lifetime of the exciplex-based blue phosphorescent OLEDs should be carried out to derive an exciplex host for practical application. Both p- and n-type hosts that can survive during the electrical operation process of the phosphorescent OLEDs should be designed to guarantee a long lifetime.

Although the current device performances of the exciplex-based blue phosphorescent OLEDs are not good enough for practical application, intensive study of the high-triplet-energy exciplex host for blue phosphors will upgrade the device performances of the blue phosphorescent OLEDs.

Disclosure statement
No potential conflict of interest was reported by the author(s).

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