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Dyes and Pigments

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Exciton stabilizing high triplet energy n-type hosts for blue phosphorescent organic light-emitting diodes

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ARTICLE INFO ABSTRACT Keywords: An exciton stabilizing electron transport type (n-type) host material was developed for application in long Blue PHOLEDs lifetime blue phosphorescent organic light-emitting diodes (PhOLEDs). A conjugation breaking diphenylsilyl Device lifetime backbone structure combined with a diphenyltriazine unit was a basic molecular platform to obtain high triplet Triplet energy energy and electron transport property. Two hosts with and without carbazole were synthesized as the bipolar Electron transport type host and unipolar n-type hosts derived from diphenylsilyl backbone and diphenyltriazine unit. The blue PhOLEDs using the two hosts showed high external quantum efficiency above 20% and blue color coordinate of (0.14, 0.17). Particularly, the bipolar n-type host with the carbazole unit extended the device lifetime by more than 1.4 times compared with the unipolar host and by more than three times relative to a conventional diphenyltriazine based host without the diphenylsilyl core because of the exciton stabilizing function of the host. This work proved the potential of the diphenyltriazine and carbazole merged diphenylsilyl skeleton as a chemical platform of the n-type hosts.

1. Introduction

Blue phosphorescent organic light-emitting diodes (PhOLEDs) have been studied because of potential to obtain high external quantum efficiency (EQE) over 20% by full utilization of excitons in the emitting layer [1–9]. Both host and phosphors were critical to the device performances of the blue PhOLEDs and the host materials affected the EQE, driving voltage, and device lifetime. Therefore, the host materials for blue PhOLEDs should have high triplet energy to prevent back energy transfer from phosphors to host, good hole and electron transport properties, and good stability against carriers during device operation.

Single bipolar hosts can satisfy the several host requirements, but the mixed host system is favored to meet the requirements and to achieve high efficiency and long device lifetime, simultaneously. The main causes of short device lifetime of blue PhOLEDs, triplet-triplet annihilation (TTA) and triplet-polaron annihilation (TPA) [10–18], can be suppressed in the mixed host by managing the triplet excitons and polarons. Triplet exciton density triggering the degradation mechanisms of blue PhOLEDs can be controlled by broad recombination zone assisted by bipolar charge transport properties of the mixed host, and polaron leakage and polaron density can be well manipulated by proper

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https://doi.org/10.1016/j.dyepig.2021.109297

Received 4 November 2020; Received in revised form 25 February 2021; Accepted 8 March 2021 Available online 16 March 2021 0143-7208/© 2021 Elsevier Ltd. All rights reserved.

selection of the hole transport type (p-type) and electron transport type (n-type) hosts of the mixed hosts [19,20].

A lot of p-type and n-type hosts have been reported in the literatures and representative n-type hosts have generally been diphenylphosphine oxide, diphenylsulfone, pyridine, pyrimidine and diphenyltriazine derivatives [21–29]. However, the diphenylphosphine oxide and sulfone derivatives are unstable due to small bond dissociation energy, which made them unsuitable for long lifetime blue PhOLEDs [30]. In the case of the diphenyltriazine derivatives, it was difficult to obtain high triplet energy in solid state because of planar backbone structure, which made it difficult to develop the high triplet energy n-type hosts compared to the p-type hosts. Moreover, the design of the n-type hosts which are stable against polarons and excitons formed during the electrical operation of the mixed host device is also challenging. Recently, several diphenylsilyl group based host materials were exemplified as the high triplet energy hosts, but the device lifetime is still short [31,32].

In this work, we developed new n-type host materials with high triplet energy for high EQE and long device lifetime in the blue PhO-LEDs. Diphenylsilane was used as a backbone structure and diphenyl-traizine unit was merged for electron transporting property. Two n-type hosts, bis(3-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl)diphenylsilane







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(2TrzSi) and bis(3-(4-(9H-carbazol-9-yl)-6-phenyl-1,3,5-triazin-2-yl) phenyl)diphenylsilane (2CzTrzSi), were synthesized and evaluated as the n-type hosts in blue PhOLEDs. It was demonstrated that the blue PhOLEDs built on the 2CzTrzSi based mixed host showed high EQE above 20% and extended device lifetime by more than 1.4 times compared with the 2TrzSi device and more than 3.7 times relative to that of the control device with a planar diphenyltriazine derived host. The exciton stabilizing function of the 2CzTrzSi host was responsible for the improved device lifetime.

2. Results and discussion

Two n-type hosts, 2TrzSi and 2CzTrzSi, were synthesized based on tetraphenylsilane backbone as a conjugation breaking group and diphenyltriazine as a strong electron transport group. The molecular structure of two hosts was targeted to have tetrahedral geometry unlike other aromatic linked diphenyltriazine derived materials with planar structure in order to obtain high triplet energy in solid state by suppressing intermolecular stacking. The diphenyltriazine functional groups were connected to the *meta* position of the phenyl unit to deliver the 2TrzSi host. The 2CzTrzSi host had extra carbazole moieties to control the singlet energy for exciton stabilization.

The synthetic processes of 2TrzSi and 2CzTrzSi are depicted in Scheme 1. A triazine-carbazole intermediate 3 was produced through a lithiation reaction of carbazole followed by reaction with 2,4-dichloro-6-phenyl-1,3,5-triazine. The brominated tetraphenylsilane intermediate 1 prepared according to reported work [33,34] was transformed into a boronic ester form and coupled with 2-chloro-4,6-diphenyl-1,3, 5-triazine and intermediate 3 by Suzuki-Miyaura coupling reaction to provide the 2TrzSi and 2CzTrzSi hosts. All reaction steps were proceeded in good yields over 50% and the chemical structure of the final hosts was confirmed by mass spectrometer and $^{1}H/^{13}C$ nuclear magnetic resonance (NMR) spectrometer (Figure S1 in supporting information).

Photoluminescence (PL) analysis of the two compounds was carried out to investigate the possibility of them as n-type hosts for blue PhO-LEDs. Fluorescent and phosphorescent emission spectra in Fig. 1 were collected to estimate the singlet and triplet energies of the hosts. Room temperature fluorescence spectra of 2TrzSi and 2CzTrzSi in neat film provided singlet energies of 3.51 and 3.14 eV, while the low temperature (77K) phosphorescence spectra in frozen tetrahydrofuran solution delivered triplet energies of 2.97 and 2.98 eV, respectively. The singlet energy of 2CzTrzSi was smaller than that of 2TrzSi due to weak charge transfer (CT) character between carbazole and diphenyltriazine. However, the triplet energy was similar in the two hosts because the additional carbazole unit did not extend the degree of π -conjugation. The extra carbazole in the 2CzTrzSi played a role of stabilizing the singlet energy while preserving the high triplet energy of the 2TrzSi. It also induced broad CT emission spectrum in the fluorescence spectra of



Fig. 1. UV–Vis absorption and PL spectra of (a) 2TrzSi and (b) 2CzTrzSi. UV–Vis absorption and phosphorescence were measured using dilute tetrahydrofuran solution. Fluorescence was obtained from neat film. Phosphorescence was measured at 77 K after delay time of 1 ms.

2CzTrzSi compared to the local emission accompanying vibrational peaks in the 2TrzSi. The ultraviolet–visible (UV–Vis) absorption spectra of 2TrzSi and 2CzTrzSi also reflected the CT properties of the 2CzTrzSi host. The UV–Vis absorption of the two materials below 300 nm was originated from strong π - π * absorption of the aromatic structure separated by the diphenysilane group. In the case of 2CzTrzSi, an extra weak absorption at 300–350 nm was observed by CT absorption of 2CzTrzSi



Scheme 1. Synthetic scheme of 2TrzSi and 2CzTrzSi.

The lowest absorption energies of 2TrzSi and 2CzTrzSi corresponding to HOMO-LUMO gap were 4.12 and 3.65 eV, respectively.

The solid state PL emission of the hosts was further studied to figure out the complex formation with p-type host in the mixed host because the 2TrzSi and 2CzTrzSi hosts were developed as n-type hosts. The fluorescence spectra of the mixed film of p-type and n-type hosts were compared with those of neat films of each host to verify the exciplex formation. Carbazole based conventional host, 3,3-di(9H-carbazol-9-vl) biphenyl (mCBP), was used as the p-type host. The emission spectra of the mixed host films (Fig. 2), mCBP:2TrzSi and mCBP:2CzTrzSi, reflected the PL emission of each host without indistinctive red-shift, indicating no exciplex formation. Two host materials can be justified as exciplex free type mixed host. In the case of 2TrzSi, donor-acceptor interaction between carbazole of mCBP and triazine of 2TrzSi caused the slight red shift of the emission spectrum in the mixed film. Whereas, the slight blue shift of the emission spectrum of 2CzTrzSi is due to suppressed intermolecular interaction between 2CzTrzSi molecules by the mCBP host.

Transient PL (TRPL) of 2CzTrzSi in Fig. 3 was measured to assess the thermally activated delayed fluorescence (TADF) characteristic of 2CzTrzSi because the singlet-triplet energy gap was small. A clear delayed fluorescence with an excited state lifetime of 15.5 μ s was observed in the TRPL decay data, suggesting that the 2CzTrzSi is a TADF type host for the mixed host.

The lowest unoccupied molecular orbital (LUMO) was calculated from onset value of reduction potential by cyclic voltammetry (CV) in Fig. 4. The HOMO of 2TrzSi was determined from the difference



Fig. 2. Film PL spectra of (a) mCBP:2TrzSi and (b) mCBP:2CzTrzSi in comparison with those of mCBP, 2TrzSi and 2CzTrzSi.

between the LUMO and optical HOMO-LUMO gap from UV-Vis absorption spectra, while that of 2CzTrzSi was obtained from onset value of oxidation potential (Figure S2) as the 2CzTrzSi is a CT type material. The HOMO/LUMO of 2TrzSi and 2CzTrzSi were -7.36/-3.24 and -6.40/-3.32 eV, respectively. The LUMO levels were similar because the same triazine unit was employed as the electron deficient unit, but the HOMO level of 2CzTrzSi was shallower than 2TrzSi due to the carbazole moiety. The experimental energy level analysis results were well correlated with a molecular orbital simulation result by B3LYP 6-31G (Becke, 3-parameter, Lee-Yang-Parr 6-31G) basis set in Fig. 5. The HOMO and LUMO frontier molecular orbitals of 2TrzSi were spread over the diphenyltriazine unit through the tetraphenylsilane backbone structure. However, the HOMO of 2CzTrzSi was predominantly localized on the carbazole. The HOMO/LUMO energy levels from the molecular orbital calculation were -6.60/-1.83 and -5.76/-1.95 eV in the 2TrzSi and 2CzTrzSi, respectively, which followed the trend of the experimentally validated energy levels. The singlet and triplet energies were also kept on the experimentally determined emission energies. All calculated data are summarized in Table 1.

Thermal properties like glass transition temperature (T_g) and thermal decomposition temperature (T_d) are depicted in Figure S3. The T_g s were 118 and 153 °C, the T_d s were 442 and 526 °C in 2TrzSi and 2CzTrzSi, respectively. The T_g and T_d of 2CzTrzSi were higher than those of 2TrzSi due to the large molecular weight and intermolecular interaction induced CT character. The two host materials showed high T_d for good thermal stability in device fabrication process and high T_g for stable device operation.

Based on the material characteristics, the 2TrzSi and 2CzTrzSi were used as the n-type hosts in blue PhOLEDs with mCBP as a p-type host. The mixed host was composed of n-type: p-type host (50:50) and Ir(cb)₃ [35] was utilized as a blue triplet emitter at 20% doping concentration. The device energy diagram and molecular structure of fac-tris(5-(tert-butyl)-1,3-diphenyl-2,3-dihydro-1H-imidazo[4,5-b]pyrazine)iridium (Ir (cb)₃) are represented at Fig. 6. A planar type triazine based DBFTrz [28] reported in our previous paper was a reference n-type host. The current density (J) and luminance (L) plots against voltage (V) are presented in Fig. 7(a). The J of the mCBP:2TrzSi and mCBP:2CzTrzSi was similar, but high J was observed in the mCBP:DBFTrz device. This was well correlated with electron transport properties of the hosts. The single carrier device data in Figure S4 confirmed the electron transport property and the electron current density of hosts were in the order of DBFTrz > $2TrzSi \approx 2CzTrzSi$. The high electron current density of DBFTrz is related with the planar molecular structure by hydrogen bonding between diphenyltriaizne and dibenzofuran. The 2TrzSi and 2CzTrzSi showed low electron current density because the diphenylsilyl backbone structure favors tetrahedral geometry. The tetrahedral structure hinders molecular orbital overlap between molecules, retarding the electron transport of the hosts. The optimized geometry of 2TrzSi and 2CzTrzSi is given in the Figure S5. The turn-on voltage of three mixed host devices were under 2.7 V and driving voltage at 10/100/1000 cdm^{-2} of the mCBP:2TrzSi and mCBP:2CzTrzSi devices were 3.1/3.6/4.5 and 3.1/3.6/4.7 V. The turn-on and driving voltages of blue PhOLED devices were low by transporting carriers separately without energy barrier by mixing the mCBP and n-type hosts.

The maximum EQEs of the mCBP:2TrzSi and mCBP:2CzTrzSi devices were 21.4 and 20.7%, respectively. The EQEs at 100/500/1,000 cd/m⁻² were 21.3/21.2/20.5% in mCBP:2TrzSi and 20.7/20.4/19.8% in mCBP:2CzTrzSi devices. The mCBP:2TrzSi device maintained the high EQE over 20% even at high luminance and the EQE of the mCBP:2CzTrzSi device was still high though it was slightly declined under 20%, indicating the efficiency drop at high luminance was improved. The mCBP:DBFTrz device showed poor maximum EQE of 9.7% because the energy transfer from host to guest was inefficient due to the low triplet energy of DBFTrz (2.94 eV) in film state. The EQE plots are described in Fig. 7(b) and device performances are summarized in Table 2.



Fig. 3. Transient photoluminescence data of (a) prompt decay and (b) delayed decay of 2CzTrzSi.



Fig. 4. Reduction CV curves of 2TrzSi and 2CzTrzSi in dichloromethane solution.

The electroluminescence (EL) spectra in Fig. 7(c) represented $Ir(cb)_3$ phosphor emission without host emission. The EL peak wavelength of the mCBP:2TrzSi and mCBP:2CzTrzSi devices was 466 nm, which agreed with pure $Ir(cb)_3$ EL emission with color coordinate of (0.14, 0.17) and PL spectrum with peak wavelength of 463 nm (Figure S6). In the case of the mCBP:DBFTrz device, the EL wavelength was slightly red-shifted to 468 nm and y color coordinate was also shifted with color coordinate of (0.14, 0.19) owing to the endothermic energy transfer.

The device operational lifetime confirmed at a constant current density operating condition is in Fig. 7(d). The device lifetime at 50% of initial luminance (LT_{50}) was 923 h in the mCBP:2TrzSi device and 1270 h in the mCBP:2CzTrzSi device, whereas that of mCBP:DBFTrz device was only 347 h. The device lifetime of the mCBP:2CzTrzSi device was extended by more than 1.4 times and 3.7 times compared to that of the mCBP:2TrzSi device and mCBP:DBFTrz device, respectively. The longer device lifetime of the mCBP:2CzTrzSi devices than that of the mCBP:DBFTrz device is due to the prevention of the reverse energy transfer by high solid state triplet energy of the 2CzTrzSi device



Fig. 5. Molecular simulation results of 2TrzSi and 2CzTrzSi using B3LYP 6-31G (Becke, 3-parameter, Lee–Yang–Parr 6-31G) basis set.

Table 1			
Measured material	properties of 2Trzsi	hne	2C7Tr7Si

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	Es	E _T	Eg	HOMO	LUMO	T _g ∕T _d		
	(eV)	(eV)	(eV) ^a	(eV)	(eV)	(°C)		
2TrzSi	3.51	2.97	4.12	-7.36^{b}	$-3.24 \\ -3.32$	118/442		
2CzTrzSi	3.14 ^c	2.98	3.65	-6.40^{d}		153/527		

 $^{\rm a}~{\rm E_g}$ was absorption energy calculated from onset value of UV–Vis absorption spectrum.

^b HOMO of 2TrzSi was calculated from difference between LUMO and Eg.

^c E_s of 2CzTrzSi was obtained from onset value of fluorescence spectrum.

^d HOMO of 2CzTrzSi was calculated from oxidation potential of CV.

is due to the exciton stabilization effect of the 2CzTrzSi host through low singlet energy by TADF character. Moreover, the short triplet exciton lifetime of 15.5 μ s by reverse intersystem crossing mechanism also contributed to the improved device lifetime of the 2CzTrzSi device.



Fig. 6. (a) Device energy diagram and (b) molecular structure of Ir(cb)_{3.}



Fig. 7. (a) Current density-voltage-luminance plots, (b) EQE-luminance plots, (c) EL spectra and (d) device operation lifetime of the Ir(cb)₃ doped blue PhOLEDs.

Additionally, the mixed host devices worked better than the single host devices of mCBP, 2TrzSi, 2CzTrzSi as presented in Figure S7. In the electrical operation process of the mixed host devices, the hole carriers are transported through the p-type host dominantly, but they can be leaked to the n-type host under high electric field and the excitons are generated in the n-type host. The low singlet energy of 2CzTrzSi decreases the generated exciton energy and can stabilize the host against

excitons. This can be proven by tracing the degree of PL intensity decline under UV exposure in Fig. 8. The reduction of PL intensity according to UV irradiation time was observed, which demonstrated that the degree of PL degradation was small in the 2CzTrzSi host compared to the 2TrzSi host. The result indicated that 2CzTrzSi was stable against UV light induced PL degradation and the improved exciton stability of 2CzTrzSi assisted achieving longer device lifetime in the 2CzTrzSi device than the

Table 2

Device data of blue PhOLEDs.

	V _{on} (V) ^a	EQE (%)			CIE	Lifetime (h,
		100 cdm ⁻²	1000 cdm ⁻²	Max		LT ₅₀)
mCBP:2TrzSi	2.61	21.3	20.5	21.4	(0.14, 0.17)	923
mCBP:2CzTrzSi	2.65	20.7	19.8	20.7	(0.14, 0.17)	1270
mCBP:DBFTrz	4.14	9.31	8.06	9.66	(0.14, 0.19)	347

^a V_{on} is turn-on voltage at 1 cdm⁻²



Fig. 8. Relative change of PL intensity of 2TrzSi and 2CzTrzSi according to UV exposure time.

2TrzSi device. The obtained results were compared with those of the common diphenylphosphin oxide host in Figure S8, which demonstrated poor device lifetime because of unstable diphenylphosphin oxide unit.

3. Conclusions

Two materials with high triplet energy, 2TrzSi and 2CzTrzSi, were synthesized as the n-type hosts for blue PhOLEDs. The tetrahedral structure of diphenylsilane conjugation breaking group enabled high triplet energy close to 3.0 eV by disrupting the planar structure of diphenyltriazine. Additionally, the carbazole exciton stabilizing unit in the 2CzTrzSi largely elongated the device lifetime of the blue PhOLEDs. The 2CzTrzSi based mixed host showed high EQE above 20% and extended device lifetime by more than 1.4 times compared with the 2TrzSi device and more than 3.7 times relative to that of the control device with a planar diphenyltriazine derived host. The results substantiated that the exciton stabilizing n-type host design can enhance the device lifetime of the blue PhOLEDs.

4. Experimental

4.1. Device fabrication and measurements

Blue PHOLED devices were fabricated using the device stack of ITO/ BPBPA:HATCN (40 nm, 30% doping)/BPBPA (10 nm)/mCBP (10 nm)/ EML (30 nm)/DBFTrz (5 nm)/ZADN (20 nm)/LiF (1.5 nm)/Al (200 nm). BPBPA is N,N,N'N'-tetra[(1,1'-biphenyl)-4-yl]-(1,1'-biphenyl)-4,4'diamine, and HATCN is dipyrazino[2,3-f:2',3'-h]quinoxaline-2,3,6,7,10,11-hexacarbonitrile. HATCN was doped in BPBPA at a doping concentration of 30%. DBFTrz is 2,8-bis(4,6-diphenyl-1,3,5-triazin-2-yl) dibenzo[b,d]furan and ZADN is 2-[4-(9,10-di-naphthalen-2-ylanthracen-2-yl)-phenyl]-1-phenyl-1H-benzoimidazole. The emitting layer (EML) was a mixed host of mCBP and electron transport type host materials doped with 20% of $Ir(cb)_3$ triplet emitter. The host materials were mixed at a ratio of 50:50.

CRediT authorship contribution statement

Ju Hui Yun: Investigation, Writing – original draft. Junseop Lim: Investigation, Writing – original draft. Won Jae Chung: Investigation. Jun Yeob Lee: Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

This work was supported by National Research Foundation of Korea (2016M3A7B4909243).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.dyepig.2021.109297.

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