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Enabling high efficiency and long lifetime of pure blue phosphorescent organic light emitting diodes by simple cyano modified carbazole-based host

Rajendra Kumar Konidena, Won Jae Chung, Jun Yeob Lee

School of Chemical Engineering, Sungkyunkwan University, 2066 Seobu-ro, Jangan-gu, Suwon, Gyeonggi, 440-746, South Korea

ARTICLE INFO	A B S T R A C T
Keywords: Host High efficiency Phosphor Blue device	Herein, we report two simple and efficient high triplet energy bipolar host materials named 2'-(9 <i>H</i> -carbazol-9- yl)-[1,1'-biphenyl]-2-carbonitrile (CzBPCN) and 9-(2'-cyano-[1,1'-biphenyl]-2-yl)-9 <i>H</i> -carbazole-3-carbonitrile (CNCzBPCN) by connecting carbazole type donor and cyano acceptor on 2 and 2' positions of a biphenyl linker, respectively. The effect of cyano substitution on carbazole building block on photophysical and electrolumi- nescence properties was unveiled. Both the compounds revealed high triplet energy (E_T) of above 3.0 eV attributed to the interrupted interchromophoric electronic interactions between the donor and acceptor units. The computational studies and carrier transport analysis proved that the compounds secured good bipolar charge transporting feature. The applicability of these materials as hosts for blue phosphorescent organic light emitting diodes (PhOLEDs) was tested. Interestingly, the CNCZBPCN hosted device exhibited superior performance with three fold improved external quantum efficiency (EQE_{max}) of 20.2% over the CZBPCN hosted device which showed EOE_{max} of 7.1% Importantly, the CNCZBPCN hosted device demonstrated 11 fold extended operational

1. Introduction

Since the first demonstration, phosphorescent organic light emitting diodes (PhOLEDs) received tremendous impetus from the scientific community as potentially high efficiency devices for next generation solid state lighting applications and displays because of their unitary internal quantum efficiency by harnessing 100% excitons [1-15]. However, phosphors typically possess long exciton lifetime and therefore they need to be embedded in an appropriate host material to nullify the efficiency detrimental factors such as exciton diffusion, concentration quenching and triplet-triplet annulation (TTA) [13,14]. As the major portion of the emitting layer is occupied by the host matrix, undoubtedly the hosts play a crucial role in governing the charge transportation, location of recombination zone and lifetime of the devices [13,14]. Thus, the host materials are equally important as the dopant emitters in achieving the best performances. Presently, potential host materials for green and red PhOLEDs have been developed for achieving high EQE and long lifetime together and successfully applied in commercial devices [13–19]. However, efficient and stable host materials for blue PhOLEDs with superior triplet energy ($E_{\rm T}$) above 3.0 eV, suitable HOMO/LUMO energy levels and bipolar charge transporting feature are scarcely reported because it is challenging to achieve all these credentials simultaneously [20–23]. Importantly, blue PhOLEDs possess far inferior operational lifetime compared to their green and red counterparts [24–26]. Thus, it is urgent to promote the operational lifetime of blue PhOLEDs without scarifying their EQE. Although several factors affect the lifetime of PhOLEDs, the stability of host material plays a vital role [24–26]. Therefore, the development of stable and high triplet energy hosts for blue PhOLEDs is highly desirable. Besides, in the field of organic materials for electronics, synthetic procedures play a crucial role. Especially for industrial applications, the synthesis of host materials for PhOLEDs should be simple, scalable and cost-effective [15].

lifetime of 121 h up to 50% of its intimal luminance ($L_0 = 200 \text{ cd/m}^2$) compared to **CzBPCN**. We believe that this work demonstrates the cost-effective and efficient high triplet energy bipolar host materials for simultaneous

achievement of high EQE over 20% and long operational lifetime of blue PhOLEDs.

In comparison to unipolar hosts, the bipolar host materials comprised of both the electron donor and acceptor units together in a single molecule are ideal to improve the charge carrier mobility,

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^{*} Corresponding author. *E-mail address:* leej17@skku.edu (J.Y. Lee).

broaden the recombination zone and boost the device performance [13, 14]. However, the development of host materials with bipolar charge transport and superior $E_{\rm T}$ (>3.0 eV) is challenging due to their intrinsic charge transfer interactions [20-26]. To solve this, numerous molecular design approaches have been developed, among which the connectivity of donor and acceptor units in non-conjugative mode of linkage through high triplet energy linker is promising to interrupt effective π -conjugation and strong charge transfer interactions [27–30]. Generally, carbazole has been used as stable hole transporting unit for designing high $E_{\rm T}$ blue hosts [13,14,20-30]. Several acceptor units such as triazine, phosphineoxide, benzimidazole, oxadiazole and cyano were reported as electron transporting units, in which the cyano moiety has drawn significant attention over unstable phosphine oxide, low $E_{\rm T}$ triazine and benzimidazole due to its robustness and less extended conjugation [13, 14,20–37]. As a linker, biphenyl unit is attractive due to its high $E_{\rm T}$, good chemical stability and wide scope for chemical modification [35]. It is anticipated that modification of biphenyl unit at 2 and 2' positions with donor and acceptor units would effectively hamper the strong interchromophoric interactions and limit the conjugation for high triplet energy.

Inspired by the above discussions, in this work we designed and synthesized two simple and easily accessible high triplet energy bipolar host materials named, CzBPCN and CNCzBPCN comprising of carbazole/cyanocarbazole donor and cyano acceptor connected through biphenyl linker at 2 and 2' positions, respectively. The effect of cyano substituent on carbazole building block on photophysical, electrochemical, thermal and electroluminescence properties were investigated in detail. Both the compounds showed high triplet energy over 3.0 eV. Theoretical studies and single carrier analysis witnessed good bipolar charge transporting feature for the materials. The potency of these materials as hosts for blue PhOLEDs was tested by employing Ir(cb)₃ blue dopant. The CNCzBPCN hosted device demonstrated excellent performance with three fold improved EQE_{max} of 20.2%, current efficiency (CE) of 30.8 cd/A compared to its congener CzBPCN, which exhibited EQEmax of 7.1% and CE of 10.3 cd/A. Notably, the simple cyano modification extended the operational lifetime time of the CNCzBPCN device by 11 fold with LT50 of 121 h compared to the **CzBPCN** device with LT_{50} of 11 h.

2. Results and discussion

2.1. Molecular design and synthesis

The molecular design of this work involved stable building blocks such as carbazole/cyanocarbazole as hole transporting units, biphenyl as a π -linker and cyano unit as an electron transporting unit. We presumed that substitution of donor and acceptor units on 2 and 2'-positions of biphenyl linker would hamper the effective conjugation between donor and acceptor units and interrupt the strong electronic communication between them, which would inherit high triplet energy without sacrificing the bipolar charge transporting property for the host materials. The accomplished synthetic procedure for the materials is shown in Scheme 1. The cesium carbonate mediated *N*-arylation of carbazole/cyanocarbazole with 1-bromo-2-fluorobenzene (1) afforded intermediate 1a/1b, respectively. Subsequently, the intermediates 1a and 1b were reacted with (2-cyanophenyl)boronic acid under palladium catalyzed Suzuki-Miyaura reaction conditions to yield the desired host materials, CzBPCN and CNCzBPCN, in reasonable yields after column chromatography and vacuum train sublimation purification methods. The chemical structures of the intermediates and host materials were confirmed by the nuclear magnetic resonance spectroscopy (NMR) and mass spectral methods.

2.2. Photophysical properties

The optical properties such as electronic absorption and emission of the compounds were investigated using ultraviolet-visible (UV-Vis) absorption and photoluminescence (PL) measurements. The absorption and emission spectra of the compounds collected in tetrahydrofuran (THF) solution are displayed in Fig. 1 and the relevant data are presented in Table 1. The compounds showed more than two indistinguishable absorption bands in the range of 274-341 nm. The short wavelength absorption appearing below ca. 300 nm is assigned to the localized π - π^* electronic transitions of the carbazole/cyanocarbazole and biphenyl units. Whereas, the long wavelength absorption band stemming above ca. 300 nm is attributed to the $n-\pi^*$ transitions of the carbazole building block. Both CzBPCN and CNCzBPCN revealed high optical band gaps of 3.59 eV and 3.53 eV, respectively, indicating their wide band gap nature. The solutions of the materials showed deep-blue emission on exposure to UV-Vis light with maximum wavelength fell in the range of 380-420 nm. Indeed, the CzBPCN showed ca. 40 nm redshifted emission compared to CNCzBPCN. This could be attributed to the strong CT character of CzBPCN due to the presence of strong carbazole donor compared to cyanocarbazole in CNCzBPCN. This is also supported by the featureless broad emission profile of CzBPCN. The CNCzBPCN showed vibrational pattern characteristic of weak CT emission [34,38]. To assess the credibility of the materials to function as hosts for PhOLEDs, their lowest triplet excited states were analyzed by recording the phosphorescence spectra of the materials in THF at low temperature of 77 K. Similar to fluorescence features, the phosphorescence spectra of the compounds witnessed vibronic profile, indicating that the phosphorescence of the materials stems from the local triplet excited states [32,33]. The triplet energy (E_T) of the materials was determined from the highest energy vibronic peaks and was found to be 3.04 for CzBPCN and 3.06 eV for CNCzBPCN. The high E_T is attributed to the interrupted conjugation between the chromophores by virtue of non-conjugation mode of linkage on 2 and 2' positions of biphenyl unit [34]. Though, the singlet energy is different for these materials, but triplet energy is similar. This anomaly can be explained as follows. Generally, the singlet energy is strongly affected by the CT property of the organic compounds. The strong CT of CzBPCN lowered the singlet energy, while the weak CT of CNCzBPCN did not. As the CT excited state



Scheme 1. Synthetic route for the desired bipolar host materials.



Fig. 1. Electronic absorption and photoluminescence spectra of the materials recorded in THF solution.



Fig. 2. Phosphorescence spectra of the bipolar hosts collected at 77 K in THF matrix.

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Compound	$\lambda_{ m abs},{ m nm}$ ($arepsilon_{ m max},{ m M}^{-1}$ cm ⁻¹ × 10 ³) ^a	$\lambda_{\rm em} ({\rm nm})^{\rm a}$	E _T (eV) ^b	HOMO/ LUMO (eV) ^c	E _g (eV) ^d	T _{5d} (°C) ^e
CzBPCN	292 (21.4), 323 (sh), 338 (6.3)	409	3.04	-5.86/- 2.70	3.16	287
CNCzBPCN	274 (73.3), 326 (sh), 341 (6.8)	345, 363.4, 377.5, 393 (sh)	3.06	-5.90/- 2.77	3.13	325

^a Collected in THF solution.

^b Determined from phosphorescence spectra.

^c HOMO and LUMO deduced from the CV.

^d Electrochemical band gap.

^e Thermal decomposition temperature (T_{5d}).

energy of the emitter is smaller than the local excited state energy, the singlet energy is dominated by the CT excited state energy. However, the triplet energy is governed by the local triplet excited state energy because it is smaller than the CT triplet excited state energy. Therefore, the CT property has no effect on the triplet energy in the current two hosts, resulting in similar triplet energy in the two compounds. The E_T of the **CzBPCN** and **CNCzBPCN** was high enough to serve as potential host materials for the blue phosphor for efficient forward energy transfer while preventing back energy transfer (see Fig. 2).

2.3. Electrochemical and thermal properties

To ascertain the electrochemical behavior of these materials, cyclic voltammetry (CV) measurements were performed on their dilute solutions in dichloromethane using 0.1 M tetrabutlyammonium perchlorate as a supporting electrolyte and ferrocene as an internal standard to approximate the peak potentials (Fig. S1 and Table 1). The compounds showed an irreversible oxidation process with an anodically shifted oxidation potential compared to the ferrocene reference, suggesting the removal of electron from the appended carbazole/cvanocarbazole units. Indeed, the CzBPCN revealed facile oxidation as witnessed by its low oxidation potential (E_{ox}) of +1.06 V compared to CNCzBPCN (+1.10 V) due to the presence of relatively strong carbazole donor in CzBPCN compared to cyanocarbazole in CzBPCN. The HOMO energy level of the compounds was determined from their onset potential of the first oxidation wave with reference to ferrocene and was -5.86 eV for CzBPCN and -5.90 eV for CNCzBPCN. The LUMO energy level of the compounds was estimated by subtracting the HOMO energy level from the electrochemical band gap and found to be -2.70 eV for CzBPCN and -2.77 for CNCzBPCN.

Thermal stability of the compounds was ascertained by thermogravimetric (TGA) analysis under an inert atmosphere (Fig. 3). The compounds showed decent thermal stabilities evident from their high thermal decomposition temperature (T_{5d}) above 287 °C corresponding to 5% weight loss. Interestingly, the **CNC2BPCN** displayed marked thermal stability with T_{5d} of 325 °C compared to **C2BPCN** ($T_{5d} =$ 287 °C). This highlights the importance of cyano substituent in improving the thermal robustness of the compounds. We also performed differential scanning colorimetric measurements, but no definite glass transition temperature (T_g) was observed for the compounds due to the



Fig. 3. TGA traces of the bipolar host materials.

small molecular weight of the two compounds for local motion of the molecular structure. The marked thermal stabilities suggest these materials can tolerate heat during vacuum processed OLED device fabrication.

2.4. Theoretical studies

To understand the electronic structure of these materials, density functional theoretical computations were executed on the compounds by employing B3LYP/6-31G* functional [40]. The optimized geometries and frontier molecular orbital diagrams are shown in Fig. 4. The low energy conformation of the compounds displayed a non-planar three dimensional geometry with a large twisting angle between appended donor and biphenyl linker of >70° and between two phenyl units of biphenyl of >58°. This can be due to the severe steric repulsions between the chromophores at the 2-position of biphenyl linker. These results suggest that the effective conjugation between the donor and biphenyl would interrupt and hamper the strong charge transfer interactions between donor and acceptor, which is beneficial for securing high triplet energy with bipolar charge transporting feature for the host

materials. In addition, the highly twisted geometry is advantageous to improve the thermal and morphological stability of the materials. As shown in Fig. 4, the HOMO orbitals are localized on the carbazole/cyanocarbazole building blocks, while the LUMO orbital was delocalized on the biphenyl linker and cyano acceptor. The well separated HOMO and LUMO orbitals on the donor and acceptor, respectively, further supports the bipolar charge transporting property for the materials. The calculated HOMO/LUMO/band gap of the compounds **CzBPCN** and **CNCzBPCN** are of -5.45 eV/-1.49 eV/3.96 eV and -5.91 eV/-1.71 eV/4.20 eV, respectively, and the trend is in good agreement with the experimentally deduced values.

2.5. Carrier transport properties

Bipolar charge transport of the host materials is one of the most important parameters for balanced charge injection and transportation, broad recombination zone and superior device performances of PhO-LEDs. Thus, to understand the relationship between the molecular structure and charge transporting properties of these materials, single carrier devices were constructed by fabricating hole only device (HOD) and electron only device (EOD). The current density (J)-Voltage (V) plots of the compounds are shown in Fig. 5. Both the compounds exhibited decent HOD and EOD current densities compared to conventional mCBP host material, indicating that the materials possess good hole and electron transporting ability and secured bipolar charge transporting feature. Indeed, the compound CzBPCN showed high hole current density compared to CNCzBPCN due to the presence of relatively strong carbazole donor compared to cyanocarbazole, while the CNCzBPCN witnessed high electron current density compared to CzBPCN due to the presence of two cyano acceptors. Nonetheless, the single carrier devices of both the compounds showed sufficiently high current densities for efficient hole and electron injection and transportation and proved that the materials possess bipolar charge transporting ability. In addition, the hole and electron motilities of the materials quantified by the space charge limited current (SCLC) method and were found to be 4.50×10^{-6} cm² V⁻¹ s⁻¹ and 3.45×10^{-7} cm² V⁻¹ s⁻¹ for **CzBPCN** and 2.33×10^{-6} cm² V⁻¹ s⁻¹ and 8.98×10^{-7} cm² V⁻¹ s^{-1} for CNCzBPCN, respectively. This proves that the CNCzBPCN has better bipolar charge transport properties for balanced carrier density and efficient exciton formation.



Fig. 4. a) Optimized geometries b) FMO distribution of the bipolar hosts calculated using B3LYP/6-31G* basis set on Gaussian 09 program.



Fig. 5. I-V plots of a) hole-only and b) electron-only device.

2.6. Electroluminescence properties

The high triplet energy and bipolar charge transporting feature of the CzBPCN and CNCzBPCN hosts encouraged us to employ them as bipolar host materials for deep-blue phosphorescent emitter, [[5-(1,1dimethylethyl)-3-phenyl-1H-imidazo[4,5-b]pyrazin-1-yl-2(3H)-ylidene]-1,2-phenylene]bis[[6-(1,1-dimethylethyl)-3-phenyl-1H-imidazo [4,5-b]pyrazin-1-yl-2(3H)-ylidene]-1,2-phenylene]iridium (Ir(cb)₃). The Ir(cb)₃ was chosen as the phosphor because of pure emission color, high efficiency and moderate lifetime [24,25]. The optimized device structure of the multi-layered deep-blue PhOLED device was ITO/BPBPA:HATCN/BPBPA/mCBP/CzBPCN or CNCzBPCN:Ir (cb)₃/DBFTrz/ZADN/LiF/Al. Here, BPBPA:HATCN and BPBPA were used as a hole injection and hole transporting layers, respectively. The mCBP and DBFTrz served as hole transporting type and electron transporting type exciton blocking layers and ZADN was employed as an electron transporting layer. The emitting layer was developed by doping 30 wt% of Ir(cb)3 either in CzBPCN or CNCzBPCN host. The energy level alignment plot of the device and chemical structures of each layer used in the device are shown in Fig. 6. The current density (J)-voltage (V)-luminance (L) plots, EQE vs L, EL plot and relative L vs operational time of the blue devices are portrayed in Fig. 7 and data are summarized in Table 2.

As shown in Fig. 7, EL spectra of both the compounds displayed single peak blue emission with a peak maximum of 469 nm, which corresponded to the emission of the blue $Ir(cb)_3$ dopant. The EL spectra were in agreement with the PL spectra of the emitting layer film (Fig. S2). The absence of any other emissions from the host or any layers in the device supports efficient exothermic energy transfer from the hosts to Ir(cb)₃ dopant. The Commission Internationale de l'Elcairage (CIE) coordinates of the devices lie in the range of pure blue coordinates of x \sim 0.14 and 0.18 < y < 0.19. The **CzBPCN** hosted device exhibited maximum EQE (EQE_{max}) of 7.1%, current efficiency (CE) of 10.3 cd/A and power efficiency (PE) of 6.5 lm/W. Whereas, the CNCzBPCN hosted device showed EQE_{max} of 20.2%, CE of 30.8 cd/A and PE of 26.9 lm/W. Certainly, the CNCzBPCN hosted device demonstrated superior performance over CzBPCN based device with three-fold improved EQE and CE. This can be attributed to the good charge transporting ability. Besides, the operational lifetime is one of the important issues of blue PhOLEDs, thus we have measured the lifetime of the devices and presented the L versus time plot at an initial luminance (L_0) of 200 cd/m² in Fig. 7. The CNCzBPCN hosted device exhibited 11 times extended



Fig. 6. Energy level diagram and the chemical structures of materials used in the device.



Fig. 7. a) I-V-L plots, b) EQE vs L, c) EL plot and d) Relative luminance vs time plots of the blue devices.

Table 2	
EL parameters of the blue devices.	

Host	Voltage $(V_{on})^a$	EQE (%)		CE (cd/A)		PE (lm/W)		CIE (x, y)	LT_{50} (h) ^b
		Max.	@1000 cd/m ²	Max.	@1000 cd/m ²	Max.	@1000 cd/m ²		
CzBPCN CNCzBPCN	3.2 3.1	7.1 20.2	5.8 14.8	10.3 30.8	8.1 21.2	6.5 26.9	3.9 11.4	0.14, 0.18 0.14, 0.19	11 121

^a turn-on voltage (V) at 1 cd/m² ${}^{b}L_{0} = 200$ cd/m.².

operational lifetime up to 50% of its initial luminance (LT_{50}) of 121 h compared to CzBPCN hosted device with LT₅₀ of 11 h. The long lifetime of the CNCzBPCN device may be attributed to the formation of broad recombination zone in emitting layer due to the balanced carrier mobility by addition of CN unit. The similar hole and electron mobilities assist enhancing the device lifetime by reducing TTA and triplet-polaron annihilation through the wide emission zone. Moreover, the large LUMO gap between the mCBP electron blocking layer and CNCzBPCN host suppresses electron leakage damaging the mCBP layer, elongating the device lifetime. The device lifetime of the CNCzBPCN device was much longer than that of the conventional mCBP device. Furthermore, it is even longer than that of the CNCzCN1 device reported in our previous work considering the color coordinate of the blue PhOLEDs [41]. Although the absolute device lifetime was about 200 h in previous work, it was obtained from the sky-blue PhOLEDs and the hosts could work only in the sky-blue devices. Therefore, the CNCzBPCN can be regarded as the deep blue host providing long lifetime. These results would provide insightful information to design simple and efficient high triplet energy bipolar host materials for simultaneous achievement of high EQE over 20% and long operational lifetime of blue PhOLEDs.

3. Conclusions

In conclusion, we have designed and synthesized two simple and

efficient high triplet energy bipolar host materials *viz.*, **CzBPCN** and **CNCzBPCN** for blue PhOLEDs by modifying biphenyl with carbazole type donor and cyano acceptor at 2 and 2' positions, respectively. Photophysical properties revealed that the donor and acceptor interactions were effectively minimized due to large dihedral angles, which resulted in high triplet energy above 3.0 eV for the host materials. The **CNCzBPCN** hosted device demonstrated three-fold improved performance over **CzBPCN** with EQE_{max} of 20.2% and CE of 30.8 cd/A. Notably, the **CNCzBPCN** revealed 11 times extended lifetime with LT_{50} of 121 to **CzBPCN** ($LT_{50} = 11$ h). These results are either comparable or superior to the reported deep-blue PhOLEDs (Table S1) [25,41–45]. We believe that these results would provide valuable guidelines to design cost-effective and efficient high triplet energy host materials for blue PhOLEDs to reach high EQE and long lifetime together.

4. Experimental section

4.1. General methods

The commercially purchased chemicals and solvents were used as received. All the reactions were performed under an inert atmosphere. The intermediates and desired host materials were purified using column chromatography methods using silica as a stationary phase. The nuclear magnetic resonance spectroscopy (NMR) data were recorded on 500 MHz JEOL spectrometer and tetramethylsilane was used as an internal standard to approximate the chemical shift values. The solutions used for analytical studies were prepared at room temperature. The absorption and photoluminescence measurements were carried out on UV–vis absorption and photoluminescence spectrophotometer. Thermogravimetric analysis data were collected under a N₂ atmosphere at a heating rate of 10 °C/min. Cyclic voltammetry measurements were carried out on CHI electrochemical analyzer using three electrode system consisting of carbon working electrode, platinum counter electrode and Ag/AgNO₃ reference electrode. Density functional computations were performed on Gaussian 09 program using B3LYP/6-31G* basis set.

4.2. Synthesis

The starting materials, 1-bromo-2-fluorobenzene (1) and (2-cyanophenyl)boronic acid, were purchased from TCI chemicals and intermediate 9-(2-bromophenyl)-9*H*-carbazole (1a) and 9-(2-Bromophenyl)-9*H*-carbazole-3-carbonitrile (1b) was prepared as described in the literature [39,40].

2'-(9H-carbazol-9-vl)-[1,1'-biphenvl]-2-carbonitrile (CzBPCN). The 1a (1.0 g, 3.1 mmol), (2-cyanophenyl)boronic acid (5.4 g, 3.72 mmol), potassium carbonate (1.2 g, 9.3 mmol), and tetrakis (triphenylphosphine)-palladium(0) (0.2 g, 0.15 mmol) were dissolved in 20 mL of THF:H₂O (3:1) mixed solvent. The reaction was allowed to reflux at 80 °C for 6 h. The reaction was monitored with TLC examination. After completion of the reaction, extraction in dichloromethane was performed and the organic layer was dried on anhydrous MgSO₄. The solvent was removed using rotary evaporation. The resulted black crude mixture was purified on column chromatography using 1:2 ratio of DCM:hexanes, which resulted in CzBPCN as a white solid, Yield (0.85 g and 85%): ¹H NMR (500 MHz, CDCl₃, δ ppm): 8.0 (d, J = 10.0 Hz, 2H), 7.75-7.73 (m, 1H), 7.68-7.66 (m, 2H), 7.58-7.57 (m, 1H), 7.53-7.51 (m, 1H), 732-7.31 (m, 2H), 7.19-7.13 (m, 4H), 7.06-7.03 (m, 1H), 6.94–6.91 (m, 1H), 6.78 (d, J = 8.0 Hz, 1H). ¹³C NMR (125 MHz, CDCl₃, δ ppm): 142.22, 137.72, 135.55, 133.35, 132.31, 132.06, 130.78, 130.37, 129.23, 129.15, 128.05, 126.15, 123.14, 120.35, 120.00, 118.74, 112.13, 109.98. MS (APCI) *m*/*z* 345.41 [(M + H)⁺]. Elemental analysis: Found: C, 88.33; N, 7.98; H, 4.84%; molecular formula C₂₅H₁₆N₂ requires C, 88.18; N, 8.13; H, 4.68%

9-(2'-Cyano-[1,1'-biphenyl]-2-yl)-9H-carbazole-3-carbonitrile (CNCzBPCN). The CNCzBPCN was synthesized as per the procedure used for CzBPCN using 1b (1.0 g, 2.88 mmol), (2-cyanophenyl)boronic acid (0.5 g, 3.45 mmol), potassium carbonate (1.2 g, 8.64 mmol), and tetrakis(triphenylphosphine)-palladium(0) (0.16 g, 0.14 mmol). White solid, Yield 0.87 g and 87%: ¹H NMR (500 MHz, CDCl₃, δ ppm): 8.29 (s, 1H), 8.02 (d, J = 10.0 Hz, 1H), 7.75–7.72 (m, 3H), 7.59–7.58 (m, 3H), 7.44 (s, 1H), 7.30–7.26 (m, 3H), 7.23–7.11 (m, 1H), 7.09–6.98 (m, 1H), 6.97–6.93 (m, 1H). ¹³C NMR (125 MHz, CDCl₃, δ ppm): 141.58, 137.79, 134.30, 133.37, 132.54, 132.19, 131.04, 129.88, 129.52, 128.69, 128.45, 127.70, 125.57, 123.35, 121.29, 120.71, 120.30, 118.29, 112.80, 110.76, 110.63. MS (APCI) *m/z* 370.42 [(M + H)⁺]. Elemental analysis: Found: C, 85.80; N, 11.34; H, 4.58%; Molecular formula C₂₆H₁₅N₃ requires, C, 85.63; N, 11.37; H, 4.49%

4.3. Device fabrication details

The blue PhOLED devices were fabricated by employing the following device configuration; indium tin oxide (ITO, 50 nm)/PEDOT: PSS (60 nm)/N4, N4,N4',N4' -tetra([1,1'-biphenyl]-4-yl)-[1,1'biphenyl]-4,4'diamine (BPBPA):1,4,5,8,9,11-Hexaazatriphenylenehexacarbonitrile (HATCN) (20 nm)/N4, N4,N4',N4' -tetra ([1,1'-biphenyl]-4-yl)-[1,1'-biphenyl]-4,4'-diamine (BPBPA) (10 nm)/ 3,3'-di(9H-carbazol-9-yl)-1,1'-biphenyl (mCBP) (10 nm)/emitting layer (EML) (25 nm)/2,8- 5 bis(4,6-diphenyl-1,3,5-triazin-2-yl)dibenzo[b,d] furan (DBFTrz) (5 nm)/2-(4-(9,10-di(naphthalen-2-yl)anthracen-2-yl) phenyl)-1-phenyl-1H-benzo[d]imidazole(ZADN) (20 nm)/LiF(2 nm)/Al

(200 nm). The emitting layer was made by doping 30 wt% Ir(cb)₃ dopant either in **CzBPCN** or **CNCzBPCN** hosts. The vacuum evaporation process was used under 3.0×10^{-7} Torr for fabrication of the devices. The devices was equipped with a glass lid filled with nitrogen and stored in glove box to protect them from oxygen, and all device performances were measured outside the glove box. The characterization of the devices was performed using a Keithley 2400 source meter, and optical characterization was carried out using a CS 2000 spectroradiometer.

Author statement

Rajendra Kumar Konidena: 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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.dyepig.2020.109118.

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