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Molecular design strategy of thermally activated delayed fluorescent emitter using CN-substituted imidazopyrazine as a new electron accepting unit

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Abstract

Thermally activated delayed fluorescence (TADF) based organic light emitting diodes (OLEDs) have attracted enormous attention recently due to their capability to replace conventional phosphorescent organic light emitting diodes for practical applications. In this work, newly designed CN-substituted imidazopyrazine moiety was utilized as an electron accepting unit in TADF emitter. Two TADF emitters, 8-(3-cyano-4-(9,9-dimethylacridin-10(9H)-yl)phenyl)-2-phenylimidazo[1,2-a]pyrazine-3-carbonitrile (Ac-CNImPyr) and 8-(3-cyano-4-(10H-phenoxazin-10-yl)phenyl)-2-phenylimidazo[1,2-a]pyrazine-3-carbonitrile (PXZ-CNImPyr), were developed based on the CN-substituted imidazopyrazine acceptor combined with acridine and phenoxazine donor, respectively. CN-substituted phenyl spacer was introduced between the donor and acceptor for sufficiently small singlet-triplet energy gap (ΔE_{ST}) and molecular orbital management. Small ΔE_{ST} of 0.07 eV was achieved for the phenoxazine donor based PXZ-CNImPyr emitter. As a result, organic light emitting diode based on PXZ-CNImPyr emitter exhibited high external quantum efficiency up to 12.7 %, which surpassed the EQE limit of common fluorescent emitters. Hence, the CN-modified imidazopyrazine unit can be introduced as a new acceptor for further modifications to develop efficient TADF OLEDs.

Key words: TADF, acceptor, imidazopyrazine, efficiency, OLED

Introduction

Thermally activated delayed fluorescence (TADF) has been proved as an efficient triplet harvesting mechanism to realize high external quantum efficiency (EQE) in organic light emitting diodes (OLEDs) and hence can be considered as a replacement for the phosphorescence¹⁻⁹. In general, the TADF molecules are small organic compounds based on the donor-acceptor architecture with small singlet-triplet energy gap (ΔE_{ST}). They can exhibit 100 % internal quantum efficiency like phosphors without use of any precious metals such as iridium or platinum¹⁰⁻¹³. Recently, a lot of TADF materials demonstrated very good performances in the OLEDs. Nevertheless, TADF OLEDs still require further improvement in material and device parameters such as short device lifetimes, long delayed fluorescence lifetime, and large efficiency roll-off for their commercial applications^{14,15}. New molecular design strategies for efficient reverse intersystem crossing (RISC) are indispensable to improve these parameters^{16–20}. According to Tamm–Dancoff approximation theory²¹, both singlet-triplet energy gap (ΔE_{ST}) and transition dipole moment are directly proportional to the highest occupied molecular orbital (HOMO)-lowest unoccupied molecular orbital (LUMO) overlap. As the oscillator strength depends on the transition dipole moment; the oscillator strength and ΔE_{ST} are complementary to each other. In the TADF material design, these two complementary parameters need to be matched in a single molecule for high performance OLEDs. Small ΔE_{ST} is necessary to facilitate the triplet excited state (T_1) to singlet excited state (S_1) reverse intersystem crossing (RISC) process in TADF emitters and is obtained by separating the wave function distributions of the HOMO and LUMO^{9,18}. The HOMO and LUMO can be largely separated by inducing perpendicular orientation between the donor and acceptor. Therefore, distorted backbone structure between the electron donor and electron acceptor for the HOMO-LUMO separation is required for small ΔE_{ST} . At the same time, effective HOMO-LUMO overlap for the large oscillator strength with efficient radiative decay from S_1 to the ground state (S_0) is also necessary for high $EQE^{22,23}$.

In the last several years, many electron donating/accepting moieties were tested for the high performing TADF OLEDs. Electron donating moieties including carbazole, triphenylamine, acridine, phenoxazine, and phenazine^{24–28}, and electron accepting moieties including triazine, pyrimidine, ketone, sulfone, phosphine oxide, dicyanobenzene, pyridine and triarylboron^{29–35} were employed as the donors and acceptors in the TADF emitters. Among the electron acceptors, the imidazole unit has been proved as an electron acceptor to realize blue emission due to its

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shallow LUMO level and high triplet energy³⁶. Until now, only a few TADF emitters have been developed based on the imidazole unit due to its weak electron acceptor strength^{26,37}.

In this work, we successfully designed TADF OLEDs based on the 2-phenyl imidazopyrazine (ImPyr) as an acceptor unit. Due to the presence of one extra nitrogen atom in the acceptor unit, the ImPyr unit can be considered as a stronger acceptor than the well-known 2-phenyl imidazopyridine unit. Furthermore, the 3-position of ImPyr was substituted with a CN unit to increase the electron accepting ability and deepen the LUMO level. Finally, 9,9-dimethyl-9,10-dihydroacridine and 10H-phenoxazine electron donors were attached at 8-position of the 3-cyano 2-phenyl imidazopyrazine (CNImPyr) unit through a CN-substituted phenyl linker to construct 8-(3-cyano-4-(9,9-dimethylacridin-10(9H)-yl)phenyl)-2-phenylimidazo[1,2-a]pyrazine-3-

carbonitrile (Ac-CNImPyr) and 8-(3-cyano-4-(10H-phenoxazin-10-yl)phenyl)-2phenylimidazo[1,2-a]pyrazine-3-carbonitrile (PXZ-CNImPyr) as new TADF emitters. The newly developed CNImPyr acceptor unit was successfully employed as a TADF acceptor to reach EQE up to 12.7 %. Due to the feasible synthetic approach, further modification of this acceptor unit is possible for superior OLED device performances.

Results and discussion

Theoretical calculations and synthesis

To deeply understand molecular configuration of **Ac-CNImPyr** and **PXZ-CNImPyr**, and their electronic properties such as HOMO-LUMO energy gap, singlet and triplet energies, oscillator strength, etc; time dependent density functional theory (TD-DFT) calculations were carried out at B3LYP/6-31G (d) level. Figure 1 represents optimized structures, HOMO-LUMO distributions, energy gap, and oscillator strength of the TADF emitters. The HOMO and LUMO energy levels of the CNImPyr acceptor were deepened by 0.47 and 0.50 eV, respectively, as compared to that of ImPyr acceptor without the CN unit. In the case of **Ac-CNImPyr** and **PXZ-CNImPyr**, the HOMO was mainly located on the electron donors of acridine or phenoxazine, while the LUMO was mainly located on the CNImPyr acceptor and the CN-substituted phenyl linker between the donor and acceptor. The phenoxazine substituted **PXZ-CNImPyr** exhibited smaller HOMO-LUMO energy gap (2.16 eV) and higher oscillator strength (0.0245) than the

acridine substituted **Ac-CNImPyr** (2.40 eV and 0.0001). The dihedral angles between the donor unit and phenyl spacer attached to the acceptor of **Ac-CNImPyr** and **PXZ-CNImPyr** were 88.3 ^o and 74.2 ^o, respectively; suggesting twisted molecular structure for HOMO-LUMO separation and TADF characteristic of these compounds.



Fig. 1. Chemical structures, HOMO-LUMO distribution and energy gap (Eg), oscillator strength (f) of the imidazopyrazine, CN-substituted imidazopyrazine acceptor, Ac-CNImPyr and PXZ-CNImPyr.

For sufficient electron accepting character, ImPyr unit was utilized as a new electron accepting unit of the TADF emitters and it was further modified by the incorporating cyano group at 3-position. Scheme 1 represents synthetic route for **Ac-CNImPyr** and **PXZ-CNImPyr**. Commercially available 3-chloro pyrazine 2-amine was reacted with phenacyl bromide to obtain

a cyclized imidazopyrazine intermediate 1, which was further converted into important 3-cyano substituted intermediate 3 through iodination and cyanation reactions. At the same time, 5-bromo 2-fluorobenzonitrile was reacted with acridine or phenoxazine in NaH/dimethylformamide (DMF) reaction condition to obtain intermediates 4 and 5 (scheme S1 in supporting information), which were further converted into intermediate 6 and 7, respectively, by the common boronate ester formation reaction. They were reacted with previously synthesized intermediate 3 by Suzuki coupling reaction to acquire the final products of **Ac-CNImPyr** and **PXZ-CNImPyr**. The TADF materials were confirmed by mass and ¹H/¹³C nuclear magnetic resonance analysis and further purified by sublimation process under a vacuum condition to obtain extra pure materials for the device study.



Scheme 1: synthetic route for Ac-CNImPyr and PXZ-CNImPyr.

Photophysical properties

To estimate their charge transfer features and singlet/triplet energies, photophysical properties of **Ac-CNImPyr** and **PXZ-CNImPyr** such as ultraviolet-visible (UV-vis) absorption, low temperature fluorescence (FL) and phosphorescence (Ph) were measured in toluene solution as depicted in Figure 2. Their detailed photophysical parameters are summarized in Table 1. In the absorption spectra of **Ac-CNImPyr** and **PXZ-CNImPyr**, high energy absorption peaks observed

between 300 and 350 nm can be assigned to the π - π * transitions of the CNImPyr acceptor derived backbone structure and the low energy absorption peaks at 430 and 458 nm are attributed to the charge transfer (CT) transitions from acridine or phenoxazine donor to the CNImPyr acceptor. Due to its strong electron donating ability, phenoxazine donor based **PXZ-CNImPyr** exhibited red shifted absorption and PL peaks compared to the acridine donor based Ac-**CNImPyr.** Fluorescence and phosphorescence spectra were measured at 77 K in toluene solvent under a nitrogen atmosphere. Low temperature phosphorescence was discriminated from the fluorescence by measuring it after 2 ms delay time because the TADF is not observed at low temperature due to deactivation of the TADF by little thermal energy. Singlet/triplet energies of Ac-CNImPyr and PXZ-CNimPyr were calculated from the onset energy of the fluorescence and phosphorescence spectra and were measured to be 2.69/2.40 eV for Ac-CNImPyr, and 2.47/2.38 eV for PXZ-CNImPyr. The ΔE_{ST} values of Ac-CNImPy and PXZ-CNImPy were 0.29 and 0.09 eV, respectively. Photoluminescence quantum yields (PLQYs) of Ac-CNImPyr and **PXZ-CNImPyr** emitters were 40 and 52 %, respectively. Comparatively high PLQY and small ΔE_{ST} were noted in the **PXZ-CNImPyr** emitter, suggesting relatively good TADF characteristic to show improved device performances for this material. To confirm their CT properties, PL spectra of Ac-CNImPyr and PXZ-CNImPyr were measured in different organic solvents as represented in Figure 3. As expected, the fluorescence emission spectra of them were red shifted by increasing solvent polarity from non-polar to polar due to CT excited state of these compounds.



Fig. 2. Normalized UV–vis absorption spectra (measured in toluene solution at 300 K), fluorescence and phosphorescence spectra (measured in toluene solution at 77 K) of Ac-CNImPyr and PXZ-CNImPyr emitter.

Emitter	λabs	λ_{PL}	$\Phi_{PL}^{b)}$	$\tau_{p}^{c)}$	$\tau_{d}^{c)}$	номо	LUMO	E _S / E _T ^{e)}	ΔE _{ST} ^{f)}
	[nm]	[nm]	[%]	[ns]	[µs]	^{d)} [eV]	^{d)} [eV]	[eV]	[eV]
	$sol^{\ a)}$	sol ^{b)}							
Ac-CNImPyr	430	524	40	10.1	729.1	-5.82	-3.57	2.69/2.40	0.29
PXZ-CNImPyr	458	568	52	15.1	86.3	-5.56	-3.59	2.47/2.38	0.09

 Table 1. Photophysical parameters of Ac-CNImPyr and PXZ-CNImPyr

a) Measured in toluene solution; b) PLQY measured in 1 wt % doped polystyrene film under oxygen-free conditions at room temperature; c) PL lifetimes of prompt (τ_p) and delayed (τ_d) decay components; d) Estimated from cyclic voltammetry measurements through onset oxidation and reduction potentials; e) Singlet (Es) and triplet (E_T)

energies were obtained from the onset of fluorescence and phosphorescence emission spectra, respectively; f) $\Delta E_{ST} = E_S - E_T$.



Fig. 3. PL spectra of Ac-CNImPyr (a) and PXZ-CNImPyr (b) in different organic solvents.

In addition, TADF characteristics of these emitters were further confirmed by transient PL measurements. Figure 4 represents transient PL data of **Ac-CNImPyr** and **PXZ-CNImPyr** at room temperature, which show typical TADF decay with nanosecond-order prompt decay and a microsecond order delayed decay under a degassed condition. The short lifetime prompt components (τ_p) can be assigned to $S_1 \rightarrow S_0$ transition, while the long lifetime delayed components (τ_d) are associated with TADF based intersystem crossing (ISC) and RISC processes. The prompt /delayed lifetimes were measured to be 10.1 ns/729 µs and 15.1 ns/86.1 µs for **Ac-CNImPyr** and **PXZ-CNImPyr**, respectively. In consistent with their experimental ΔE_{ST} values, strong phenoxazine donor based **PXZ-CNImPyr** emitter demonstrated shorter delayed fluorescence lifetime than the acridine donor based **Ac-CNImPyr** emitter.



Fig. 4. Delayed transient PL decay curves of 1 % **Ac-CNImPyr** and **PXZ-CNImPyr** doped films in polystyrene at room temperature.(Inset: Prompt PL decay curves of 1 % **Ac-CNImPyr** and **PXZ-CNImPyr** doped films in polystyrene at room temperature).

Thermal and electrochemical properties

Thermogravimetric analysis (TGA) study was performed to study thermal stability of Ac-CNImPyr and PXZ-CNImPyr at high temperature. As represented in Figure 5, thermal decomposition temperatures of Ac-CNImPyr and PXZ-CNImPyr were found to be 423 and 410 °C, respectively, at 5 % weight loss. Both compounds were stable at high temperature and suitable for vacuum evaporation process. Additionally, cyclic-voltammetry study of Ac-CNImPyr and PXZ-CNImPyr was carried out using conventional three electrode system for the calculation of their HOMO and LUMO energy levels (Figure 6). Onset oxidation potentials of Ac-CNImPyr and PXZ-CNImPyr were found to be 1.01 and 0.85 eV respectively, while their onset reduction potentials were -1.24 and -1.22 eV respectively. The HOMO/LUMO energy levels of Ac-CNImPyr and PXZ-CNImPyr were calculated from the respective onset potentials of oxidation and reduction, and were found to be -5.82/-3.57 eV for Ac-CNImPyr and -5.56/ - 3.59 eV for **PXZ-CNImPyr**. The shallow HOMO level of **PXZ-CNImPyr** can be attributed to the comparatively strong electron donating ability of the phenoxazine donor. The LUMO levels were similarly deep by the strongly electron accepting CNImPyr acceptor shared in the two TADF emitters.



Fig. 5. Thermogravimetric analysis (TGA) curves of Ac-CNImPyr and PXZ-CNImPyr.



Fig. 6. Cyclic voltammograms of TADF emitters Ac-CNImPyr and PXZ-CNImPyr.

Electroluminescence Performance

The electroluminescence (EL) performances of **Ac-CNImPyr** and **PXZ-CNImPyr** were evaluated by doping them into 2-phenyl-4,6-bis(12-phenylindolo[2,3-a]carbazole-11-yl) -1,3,5-triazine (PBICT) host material. Figure 7 shows the energy level diagram and chemical structures of the materials used in different layers of the OLED devices. Figure 8, 9 and 10 represent current density–voltage–luminance (J–V–L) characteristics, EQE-L curves and EL spectra of the 10 wt % emitter doped devices, respectively. The turn-on voltage of the two devices was quite similar, but the driving voltage at 1,000 cd/m² was dissimilar. The **PXZ-CNImPyr** emitter based device (5.8 V) possibly due to good hole transport through the **Ac-CNImPyr** emitter. EL spectrum of the **PXZ-CNImPyr** device was red-shifted compared to that of the **Ac-CNImPyr** device, which followed the same trend of the PL spectra. EL peaks of the **PXZ-CNImPyr** and **Ac-CNImPyr** (541 and 570 nm) devices were red-shifted relative to their respective PL peaks (524 and 565 nm) due to the use of comparatively polar PBICT host material. Figure S1-S6 in supporting information represents J–V–L characteristics, EQE-L curves and EL spectra of the **Ac-CNImPyr**

and **PXZ-CNImPyr** devices at different doping concentrations. The EL spectra of the devices were slightly red-shifted by increasing doping concentration from 1 to 10 % mainly due to the interaction between dopant molecules at high doping concentration. The color coordinates of the **Ac-CNImPyr** and **PXZ-CNImPyr** devices were (0.37, 0.55) and (0.50, 0.49), respectively. Relatively high EQE of 12.7 % was exhibited by **PXZ-CNImPyr**. The device result was consistent with comparatively high PLQY (52 %), short delayed fluorescence lifetime (86.3 μ s) and small ΔE_{ST} (0.07 eV) of the **PXZ-CNImPyr** emitter. The EQE values of the devices were decreased at high doping concentration mainly due to concentration quenching effect. Low EQE (5.6 %) of the Ac-CNImPyr device is due to low PLQY (40 %). The 12.7 % EQE value of **PXZ-CNImPyr** emitter is higher than the theoretically estimated value of ~5 % for the conventional fluorescent emitter³⁸⁻⁴⁰ (this is not an absolute limit, but rather based on estimates of the refractive index difference). Therefore, it was confirmed that triplet excitons as well as singlet excitons were utilized through RISC process for the EL emission of the **PXZ-CNImPyr** emitter based TADF emitters as the efficient emitters in the future.



Fig. 7. Energy-level diagram and material chemical structures of the Ac-CNImPyr and PXZ-CNImPyr emitter doped devices.



Fig. 8. Current density–voltage–luminance curves of **Ac-CNImPyr** and **PXZ-CNImPyr** based OLED devices.



Fig.9. Quantum efficiency–luminance curves of the **Ac-CNImPyr** and **PXZ-CNImPyr** based OLED devices.



Fig.10. EL spectra of the Ac-CNImPyr and PXZ-CNImPyr devices.

Emitter	λ_{EL}	Voltage ^a	CE _{max}	PE _{max}	EQE _{max}	CIE (x , y)
	[nm]	[V]	[cd A ⁻¹]	[lm W ⁻¹]	[%]	
Ac-CNImPy	541	5.8	36.9	38.6	5.6	(0.37, 0.55)
PXZ-CNImPy	570	4.9	32.3	29.1	12.7	(0.50, 0.49)

Table 3. Electroluminescence performances of the Ac-CNImPyr and PXZ-CNImPyr devices.

^a At 1000 cd m⁻²

Conclusions

In conclusion, we developed two TADF emitters based on the newly introduced CN-substituted 2-phenyl imidazopyrazine acceptor named CNImPyr. The CNImPyr acceptor was combined with either acridine or phenoxazine donor. Among the two emitters, the strong phenoxazine donor based **PXZ-CNImPyr** emitter showed comparatively high PLQY, short delayed fluorescence lifetime and small ΔE_{ST} , resulting in the device EQE of 12.7 %. This result confirmed efficient TADF process in the **PXZ-CNImPyr** emitter and capability of CNImPyr unit as an efficient electron acceptor for TADF emitters.

Experimental

Intermediate 1 and 2 were synthesized by following the reported procedure⁴¹. General information about measurement and calculation is in supporting information.

Synthesis of 8-chloro-2-phenylimidazo[1,2-a]pyrazine-3-carbonitrile (3)

8-Chloro-3-iodo-2-phenylimidazo[1,2-a]pyrazine (2) (1.3 g, 3.65 mmol) was dissolved in dimethylformamide (50 mL) and copper cyanide (1.63 g, 18.28 mmol) was added in the reaction mixture. The resulting mixture was stirred at 150 °C for 12 h. Reaction mixture was cooled to room temperature and water was added. A solid precipitate was filtered, dried well and purified by column chromatography (60 % DCM in hexane) to afford a pure product (1.05 g, 42.68 %). MS (FAB) m/z 255 [(M + H)+]. ¹H NMR (500 MHz, CDCl₃) δ 8.21-8.27 (m, 3H), 7.96-7.98 (d, J = 7 Hz, 1H), 7.53-7.56 (m, 3H).

Synthesis of 8-(3-cyano-4-(9,9-dimethylacridin-10(9H)-yl)phenyl)-2-phenylimidazo[1,2-a]pyrazine-3-carbonitrile (Ac-CNImPyr)

8-Chloro-2-phenylimidazo[1,2-a]pyrazine-3-carbonitrile (0.3 g, 1.17 mmol) and 2-(9,9-dimethylacridin-10(9H)-yl)-5-(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)benzonitrile (0.61 g, 1.41 mmol) were dissolved in 1, 4 dioxane (20 mL). The reaction mixture was degassed with argon for half an hour. tetrakis(triphenylphosphine)palladium (0.14 g, 0.12 mmol) was added followed by K₂CO₃ (6 mL, 0.2 M solution in water) and the reaction mixture was stirred at 100 ° C for 12 h. After cooling to room temperature, water was added to the reaction mixture followed by extraction with ethyl acetate three times (50 mL). The organic layer was dried over MgSO₄, concentrated on rotary evaporator and purified by column chromatography (40 % dichloromethane in hexane) to afford a pure product (0.55 g, 88.42 %). MS (FAB) MS (FAB) m/z 529 [(M + H)⁺]. ¹H NMR (500 MHz, CDCl₃): δ 9.58 (d, *J* = 2 Hz, 1H), 9.41-9.43 (dd, *J* = 8.5 and 2 Hz, 1H), 8.35-8.38 (m, 2H), 8.31-8.32 (d, *J* = 7 Hz, 2H), 7.75-7.76 (d, *J* = 8.5 Hz, 1H), 7.52-7.59 (m, 5H), 7.00-7.02 (m, 4 H), 6.24-6.26 (m, 2H), 1.76 (S, 6H). ¹³C NMR (125 MHz, CDCl₃): δ 31.3, 36.4, 111.5, 113.6, 116.2, 116.8, 118.3, 121.9, 125.8, 126.8, 127.9, 129.5, 130.3, 130.9, 131.3, 131.9, 133.9, 135.8, 135.9, 136.9, 139.9, 146.5, 146.9, 153.8.

Synthesis of 8-(3-cyano-4-(10H-phenoxazin-10-yl)phenyl)-2-phenylimidazo[1,2-a]pyrazine-3-carbonitrile (PXZ-CNImPyr)

Similar procedure as described for the synthesis of Ac-CNImPyr was used except that 10Hphenoxazine was used as the reactant instead of 9,9-dimethyl-9,10-dihydroacridine, to afford a yellow solid (0.28 g, 71.06 %). MS (FAB) MS (FAB) m/z 503 [(M + H)⁺]. ¹H NMR (500 MHz, CDCl₃): δ 9.55 (d, *J* = 2 Hz, 1H), 9.37-9.40 (dd, *J* = 8.5 and 2 Hz, 1H), 8.35-8.39 (m, 2H), 8.30-8.32 (d, *J* = 7 Hz, 2H), 7.76-7.78 (d, *J* = 8.5 Hz, 1H), 7.54-7.61 (m, 3H), 6.73-6.79 (m, 4H), 6.64-6.68 (m, 2H), 5.97-5.99 (d, *J* = 8 Hz, 2H). ¹³C NMR (125 MHz, CDCl₃): δ 111.3, 113.2, 115.7, 116.2, 116.6, 118.1, 122.6, 123.4, 127.6, 129.3, 130.1, 131.1, 131.6, 132.6, 132.8, 135.8, 136.3, 136.6, 139.6, 143.9, 144.0, 146.4, 153.6.

Device fabrication and measurements

The device configuration was ITO (50 nm)/PEDOT:PSS (60 nm)/TAPC (20 nm)/ mCP (10 nm)/ PBICT:Ac-CNImPyr or PXZ-CNImPyr (25 nm, X wt%)/TSPO1(5 nm)/TPBi (40 nm)/LiF(1.5 nm)/Al(200 nm); where poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) was used as a hole injection layer; 4,4'-cyclohexylidenebis[N,N-bis(4-methylphenyl)aniline] (TAPC) as a hole transporting layer and 2,2',2''-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi) as an electron transporting layer. N,N-Dicarbazolyl-3,5-benzene (mCP) and diphenyl-4-triphenylsilylphenyl phosphine oxide (TSPO1) were employed as hole- and electron-transport type exciton blocking layer, respectively. Vacuum evaporation process was used to produce the devices and the fabricated devices were encapsulated with a glass cover to protect the devices at ambient condition. The devices were characterized using CS2000 spectroradiometer and Keithley 2400 source measurement unit.

Conflicts of interest

There are no conflicts of interest to declare.

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Table of Contents

Molecular design strategy utilizing imidazopyrazine as new electron accepting unit for two thermally activated delayed fluorescence (TADF) emitters (Ac-CNImPyr and PXZ-CNImPyr) was proposed. Especially TADF emitter with strong phenoxazine donor (PXZ- CNImPyr) exhibited sufficiently small ΔE_{ST} to initiate RISC process and realize high external quantum efficiency up to 12.7 %. This new CN-modified imidazopyrazine (CNImPyr) unit was successfully tested as proficient electron acceptor for the future TADF emitter design.

