



# Indolo[3,2,1-*jk*]carbazole-derived planar electron transport materials realizing high efficiency in green phosphorescent organic light-emitting diodes

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**ABSTRACT** Two novel electron transporting materials (ETMs), 2-(4,6-diphenyl-1,3,5-triazin-2-yl)indolo[3,2,1-*jk*]carbazole (DPTrz-ICz) and 5,11-bis(4,6-diphenyl-1,3,5-triazin-2-yl)-2-phenylindolo[3,2,1-*jk*]carbazole (2DPTrz-ICz), were developed based on the indolo[3,2,1-*jk*]carbazole (ICz) core, combined with 2,4-diphenyl-1,3,5-triazine (DPTrz). The introduced ICz core in ETM exhibited enhanced intermolecular charge transport properties owing to its planar and rigid geometry and high triplet energy, demonstrating the potential of designing ETM. Consequently, 2DPTrz-ICz, featuring high triplet energy of 2.84 eV, effectively prevented triplet exciton quenching and showed increased electron mobility owing to the activation of intermolecular hopping charge transfer facilitated by stacking architecture. The two materials were utilized as ETMs in green phosphorescent organic light-emitting diodes. 2DPTrz-ICz exhibited a superior external quantum efficiency of 21.5%, higher than that of the spirobifluorene-derived ETM.

**Keywords:** electron transport layer, indolocarbazole, efficiency, OLED

## INTRODUCTION

Developing organic light-emitting diodes (OLEDs) has opened an era of next-generation displays through unprecedented high-resolution, thinness, and flexibility and has supplemented the limitations of conventional liquid crystal displays, such as narrow viewing angle and slow response speed [1–5]. Although several OLED types have been developed, phosphorescent OLEDs (PHOLED) have been widely studied because they can realize an internal quantum efficiency of 100% *via* harvesting triplet excitons for light emission [6].

Many studies have been conducted on host molecules responsible for transporting charges in the emitting layer (EML) and dopant molecules that directly participate in light emission *via* recombining charges because both are critical to the efficiency and device lifetime of PHOLEDs [7–14]. However, the high efficiency of PHOLEDs cannot be realized without the assistance of hole transport material and electron transport material (ETM) injecting and transporting carriers into EML. Generally, electron mobility is inherently lower than hole

mobility, which encourages the development of high-mobility ETM. There are three major requirements for ETM to increase the efficiency of PHOLED. (1) Good electron injection/hole blocking ability, which can be realized by adjusting the lowest unoccupied molecular orbital (LUMO) of the ETM to have an appropriate value to form a stepwise configuration between the LUMO of the EML and the work function (WF) of the cathode for efficient electron injection. Additionally, the highest occupied molecular orbital (HOMO) of the ETM should be much deeper than that of EML for charges to recombine only in the EML. (2) High electron mobility can be achieved by introducing an electron-deficient moiety with a heteroaromatic ring, such as triazine, pyridine, pyrimidine, and dibenzofuran, or *via* enlarging the  $\pi$ -conjugation of the core. (3) High triplet energy ( $E_T$ ) can be obtained by introducing a high triplet energy moiety with a limited degree of conjugation. Because all excitons relax and emit light from the lowest triplet-excited state and have a relatively long lifetime ( $\mu$ s scale), the nonradiative loss should be minimized by suppressing triplet exciton quenching using high-triplet-energy ETMs. In addition to charge balance, requirements (1) and (2) can reduce the turn-on voltage ( $V_{on}$ ) and driving voltage ( $V_d$ ), respectively.

High-mobility ETMs derived from an anthracene core with a condensed  $\pi$ -conjugated structure have been previously studied [15–18]. Duan's group [15] recently reported an anthracene derivative ETM, DPPyA, taking advantage of high charge mobility *via* inter/intramolecular hydrogen bonds by adding pyridyl units to enable a facile electron hopping mechanism. However, owing to the incompatibility of high charge mobility and the high  $E_T$  of the large  $\pi$ -conjugated structure, DPPyA has a low  $E_T$  of 1.77 eV, which is insufficient to prevent triplet exciton leakage from the EML with green phosphor. ETMs using spiro structures that complement the low  $E_T$  of anthracene-based ETM have also been actively studied [19–24], because they can have a high  $E_T$  of  $\geq 2.3$  eV using the  $sp^3$  carbon connecting the two spiro moieties. 2,7-Bis(4,6-diphenyl-1,3,5-triazin-2-yl)-9,9'-spirobifluorene] (SBFTrz) with a high  $E_T$  of 2.53 eV was developed by adding an electron transporting triazine moiety to a spirobifluorene (SBF) core [23], which has an excellent ability for electron transport and efficient exciton blocking with the green phosphor in EML [25]. As a result, it showed lower  $V_{on}/V_d$  and higher efficiency than 2,8-bis(4,6-diphenyl-1,3,5-triazin-2-

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yl)dibenzo-[*b,d*]furan (DBFTrz), which has dibenzofuran as a core structure and is commonly used for ETL. However, ETMs for PHOLED must be further developed to upgrade the device performances of green PHOLED.

Herein, we designed two ETMs, 2-(4,6-diphenyl-1,3,5-triazin-2-yl)indolo[3,2,1-*jk*]carbazole (DPTrz-ICz) and 5,11-bis(4,6-diphenyl-1,3,5-triazin-2-yl)-2-phenylindolo[3,2,1-*jk*]carbazole (2DPTrz-ICz), *via* introducing indolo[3,2,1-*jk*]carbazole (ICz) core and 2,4-diphenyl-1,3,5-triazine (DPTrz) for high triplet energy and high electron mobility. The DPTrz-ICz and 2DPTrz-ICz showed high  $E_T$  values of 2.98 and 2.84 eV for triplet exciton blocking, respectively. Among the two ETMs, 2DPTrz-ICz particularly achieved a lower  $V_{on}/V_d$  (at 3000 cd m<sup>-2</sup>) of 3.5 V/6.8 V and higher external quantum efficiency (EQE) of 21.5% than SBFTrz in device performances. This is owing to the quenching suppression effect because of the high triplet energy and the improved electron transport property obtained *via* inter/intramolecular interactions.

## RESULTS AND DISCUSSION

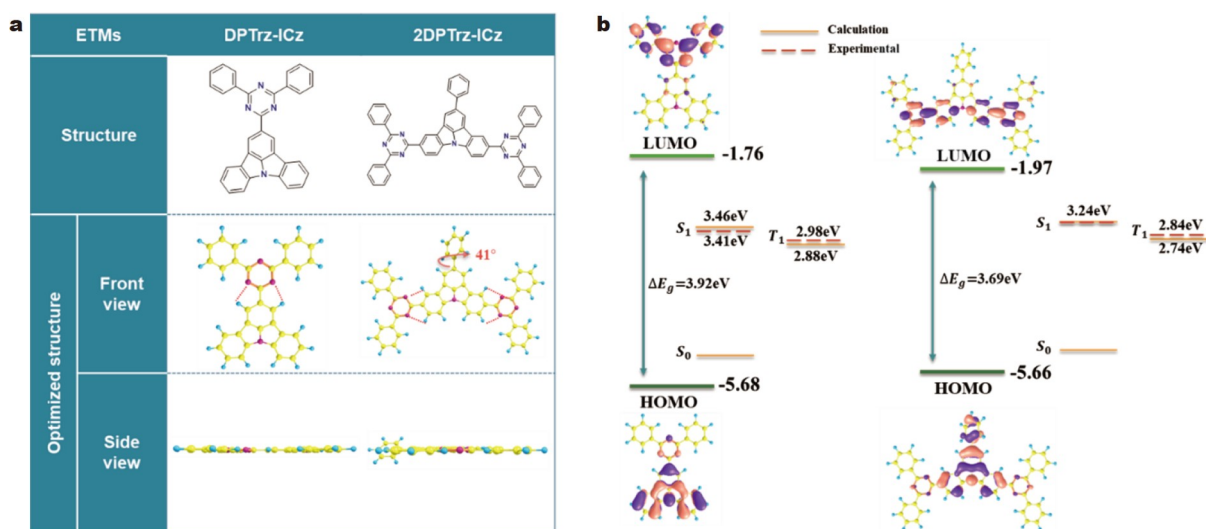
### Material design and synthesis

Conventionally, ICz has been mainly studied as a core structure of green or blue multiresonance emitter [26–28] because it can realize a narrow full width at half maximum and a small Stoke's shift owing to its rigid and planar structure and high singlet/triplet energy [29]. The planarity and rigidity of its structure can also strengthen the intermolecular interaction *via* molecular packing, facilitating the electron hopping mechanism between molecules and increasing electron mobility. Therefore, ICz is suitable as the core structure of the ETM, which requires high electron mobility and high triplet energy. In addition, the ICz has a higher triplet energy by 0.3 eV than SBF and is advantageous to prevent triplet exciton leakage from the emitter. In addition to high electron mobility and triplet energy, combining ICz with prominent DPTrz electron transport units can give ETMs a deep HOMO/LUMO value and low energy barrier for electron injection from the cathode. Furthermore, the nitrogen atoms of the triazine moiety form intramolecular hydrogen bonds with the hydrogen atoms of the ICz core, further

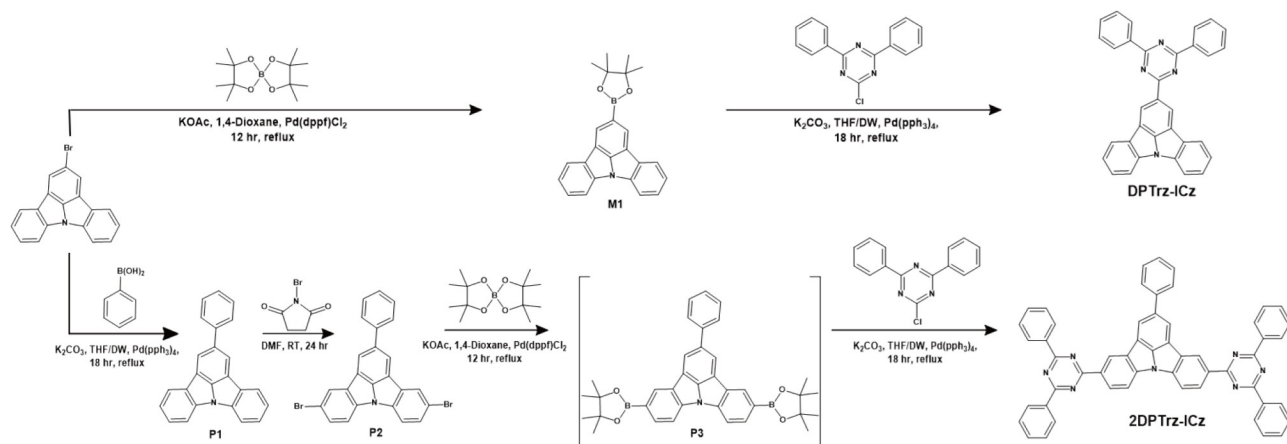
strengthening the rigidity and planarity of the molecule for high electron mobility. Based on this concept, DPTrz-ICz and 2DPTrz-ICz with one DPTrz and two DPTrz units were developed.

To theoretically prove our design concept for the ETMs, density functional theory (DFT) calculation and time-dependent DFT (TD-DFT) calculation of the ETMs were performed using the Gaussian 16 program with B3LYP 6-31G\* basis. As depicted in the side view of ETMs in Fig. 1a, the triazine and ICz moieties of both molecules are completely in the same plane, and only the 2DPTrz-ICz molecule has a phenyl group rotated by 41° at the C2 position of the ICz core. As expected, this suggests that the intramolecular hydrogen bond can be formed between the ICz and the DPTrz (red dash line in Fig. 1a). Since this geometry is advantageous for strengthening intermolecular stacking, it may contribute to expanding the electron transfer channel *via* increasing the  $\pi$ - $\pi$  orbital overlap between molecules. The HOMO and LUMO distributions of the two ETMs are shown in Fig. 1b. The LUMO is distributed on the strongly electron-deficient triazine moiety, while the HOMO is dispersed on the weakly electron-rich ICz core. However, the HOMO and LUMO distributions are completely separated in the DPTrz-ICz, while LUMO is extended through the DPTrz and ICz units. This might positively impact electron mobility for electron hopping owing to extensive orbital overlap. Although the calculated triplet energy (2.84 eV) of 2DPTrz-ICz is slightly lower than that of DPTrz-ICz (2.98 eV), both molecules are expected to have high triplet energy to suppress triplet exciton quenching of green phosphors owing to the high triplet energy of the ICz moiety. In addition, the simulated HOMO–LUMO gap of 2DPTrz-ICz (3.69 eV) is smaller than that of DPTrz-ICz (3.92 eV), and the LUMO level of 2DPTrz-ICz is deeper than that of DPTrz-ICz owing to the extra DPTrz moiety in 2DPTrz-ICz.

The synthesis processes of DPTrz-ICz and 2DPTrz-ICz are schematically shown in Scheme 1. The two ETMs were synthesized as follows. The C2 position of 2-bromoindolo[3,2,1-*jk*]carbazole (SM) was first substituted with a boronic ester group *via* borylation with bis(pinacolato)diboron (B<sub>2</sub>pin<sub>2</sub>), forming M1. DPTrz-ICz was then synthesized *via* Suzuki coupling between the boronic ester of M1 and the chlorine atom of the 2-



**Figure 1** (a) Chemical structures and front/side view of the optimized structures of DPTrz-ICz and 2DPTrz-ICz using the Gaussian 16 program. (b) DFT/TD-DFT calculations of DPTrz-ICz (left) and 2DPTrz-ICz (right).



**Scheme 1** Synthesis processes of DPTrz-ICz and 2DPTrz-ICz.

chloro-4,6-diphenyl-1,3,5-triazine (DPTrz-Cl). Moreover, P1 was obtained *via* Suzuki coupling between phenylboronic acid and SM, and P2 was synthesized following the procedure reported in our previous work [30]. Then, the bromine located at the 5 and 11 positions of the ICz core in P2 was substituted with boronic ester groups *via* borylation with B<sub>2</sub>pin<sub>2</sub>, forming P3. In this reaction step, mono- and di-substituted boronic esters were generated in the ratio of 2:8. The mixture was directly used in the subsequent reaction without separating the compounds. Finally, 2DPTrz-ICz was prepared *via* Suzuki coupling between P3 and DPTrz-Cl. Further details on the reaction conditions and reagents can be found in the Supplementary information.

### Thermal analysis

Thermogravimetric analysis (TGA) and differential scanning calorimeter (DSC) measurements were conducted to determine the thermal stability of the two ETMs. Firstly, the decomposition temperature ( $T_d$ ) at a weight loss of 5% from the initial weight of the organic matter was confirmed. The  $T_d$  of DPTrz-ICz and 2DPTrz-ICz were 402 and 550°C (Fig. 2c), respectively, and 2DPTrz-ICz showed excellent thermal stability, indicating that it can stably evaporate thermally with little decomposition at high temperatures. This results from the synergistic effect induced by the highly rigid ICz core and the hydrogen bonds from the triazine moiety. The DSC measurement was also performed for both molecules, but it was impossible to detect their glass transition temperature owing to planar and rigid geometry that limited the local motion of molecules.

### Photophysical properties

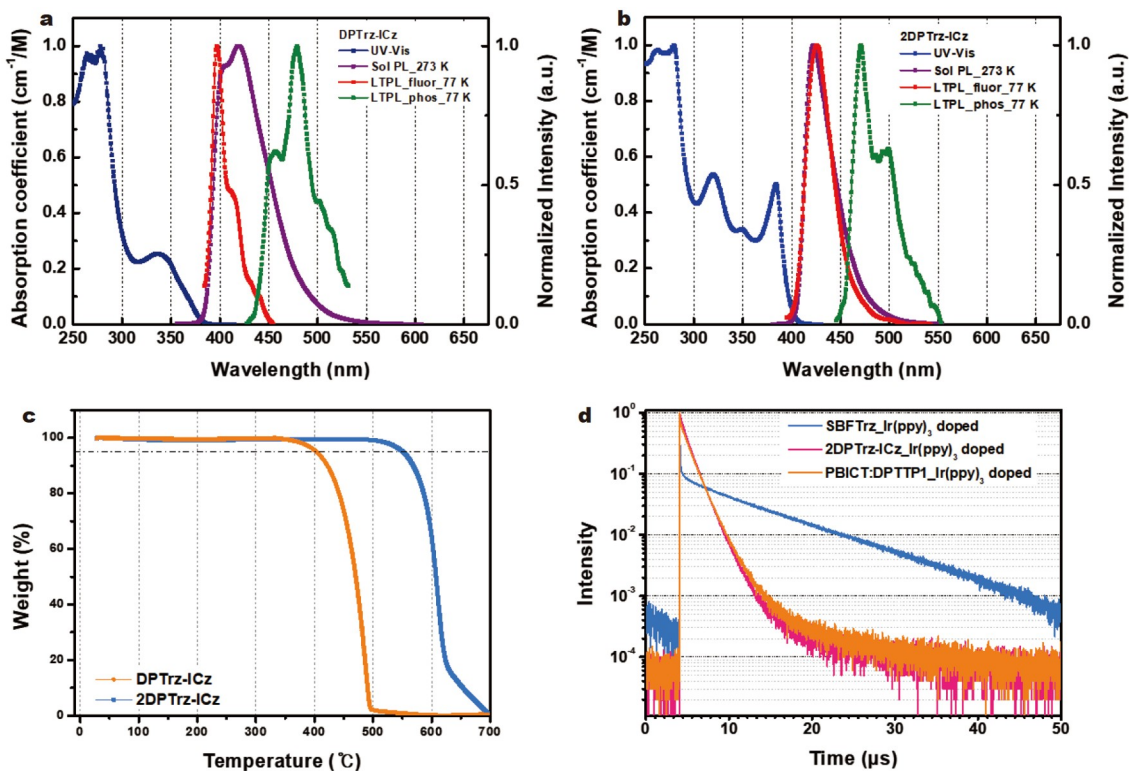
To identify the photophysical properties, the ultraviolet-visible (UV-vis) absorption and photoluminescence (PL) spectra were measured at a concentration of  $10^{-5}$  mol L<sup>-1</sup> in tetrahydrofuran (THF) solvent, as depicted in Fig. 2a, b and summarized in Table 1. The UV-vis absorption spectra of both molecules showed  $\pi$ - $\pi^*$  transitions at wavelengths below 300 nm and  $n$ - $\pi^*$  transitions at 300–400 nm. The 2DPTrz-ICz tended to absorb light at longer wavelengths owing to its extended conjugation within the backbone structure. The HOMO-LUMO gap estimated from the absorption onset was smaller in 2DPTrz-ICz (3.11 eV) than that in DPTrz-ICz (3.23 eV), consistent with the theoretical calculation. PL spectra were also analyzed to investigate the fluorescence and phosphorescence properties.

DPTrz-ICz and 2DPTrz-ICz exhibited similar peak wavelengths of 398 and 401 nm in THF at 273 K. Low temperature (77 K) PL measurements of DPTrz-ICz and 2DPTrz-ICz offered high triplet energy of 2.98 and 2.84 eV, respectively, owing to the high triplet energy of the ICz, which was higher than that of previously reported SBFTrz. This proves both compounds can be excellent ETMs to prevent triplet exciton quenching from green phosphorescent dopant.

The triplet exciton blocking effect of the ETMs was analyzed using transient PL (TRPL) measurement. The TRPL measurement was conducted using 2DPTrz-ICz film doped with 5% tris-(2-phenylpyridine)iridium (Ir(ppy)<sub>3</sub>), SBFTrz doped with Ir(ppy)<sub>3</sub>, and 2-phenyl-4,6-bis(12-phenylindolo[2,3-*a*]carbazole-11-yl)-1,3,5-triazine (PBICT):4-(3-(triphenylene-2-yl)phenyl)-dibenzo-[*b,d*]thiophene (DBTTP1) (75:25) film doped with Ir(ppy)<sub>3</sub> (Fig. 2d and Table S1). The PBICT:DBTTP1 film was used as a control sample because it was used as a host in the fabrication of the devices. The fluorescence decay curve of the 2DPTrz-ICz film was similar to that of the PBICT:DBTTP1 film, suggesting that the triplet excitons of Ir(ppy)<sub>3</sub> were not quenched by the 2DPTrz-ICz of ETM owing to its high triplet energy. However, the SBFTrz film displayed a completely distinct decay pattern from the other two curves, suggesting a triplet exciton quenching of the emitter by SBFTrz. Furthermore, PL quantum yield (PLQY) was measured to determine the luminous efficiency of the three Ir(ppy)<sub>3</sub>-doped films. Consistent with the TRPL trend, the 2DPTrz-ICz film showed a high PLQY of 92.3% compared with that of the control sample (96.0%), while the SBFTrz film demonstrated a considerably low efficiency of 24.8% owing to its triplet exciton quenching. Therefore, the TRPL and PLQY measurements infer that 2DPTrz-ICz is a more suitable ETM for enhancing EQE *via* effectively confining the triplet exciton within the EML in green PHOLED.

### Device characteristics

To investigate the electron transport characteristics of each molecule when incorporated into a device, the electron-only device was fabricated with the following configuration for the developed ETMs: indium tin oxide (ITO, 50 nm)/diphenyl[4-(triphenylsilyl)phenyl]phosphine oxide (TSPO1, 10 nm)/ETMs (60 nm)/LiF (1.5 nm)/Al (200 nm). TSPO1 was used as a hole-blocking layer. The ETMs were DPTrz-ICz, 2DPTrz-ICz, and



**Figure 2** UV-vis absorption spectra, fluorescence spectra (273 K/77 K), and phosphorescence spectra at 77 K of  $10^{-5}$  mol L $^{-1}$  in THF solutions of (a) DPTrz-ICz and (b) 2DPTrz-ICz. (c) TGA curves of DPTrz-ICz and 2DPTrz-ICz. (d) TRPL of 5 wt% Ir(ppy) $_3$ -doped 2DPTrz-ICz, SBFTrz, and PBICT:DBTTP1 (75:25).

**Table 1** Photophysical and electrochemical properties and thermal stability of 2DPTrz-ICz and DPTrz-ICz

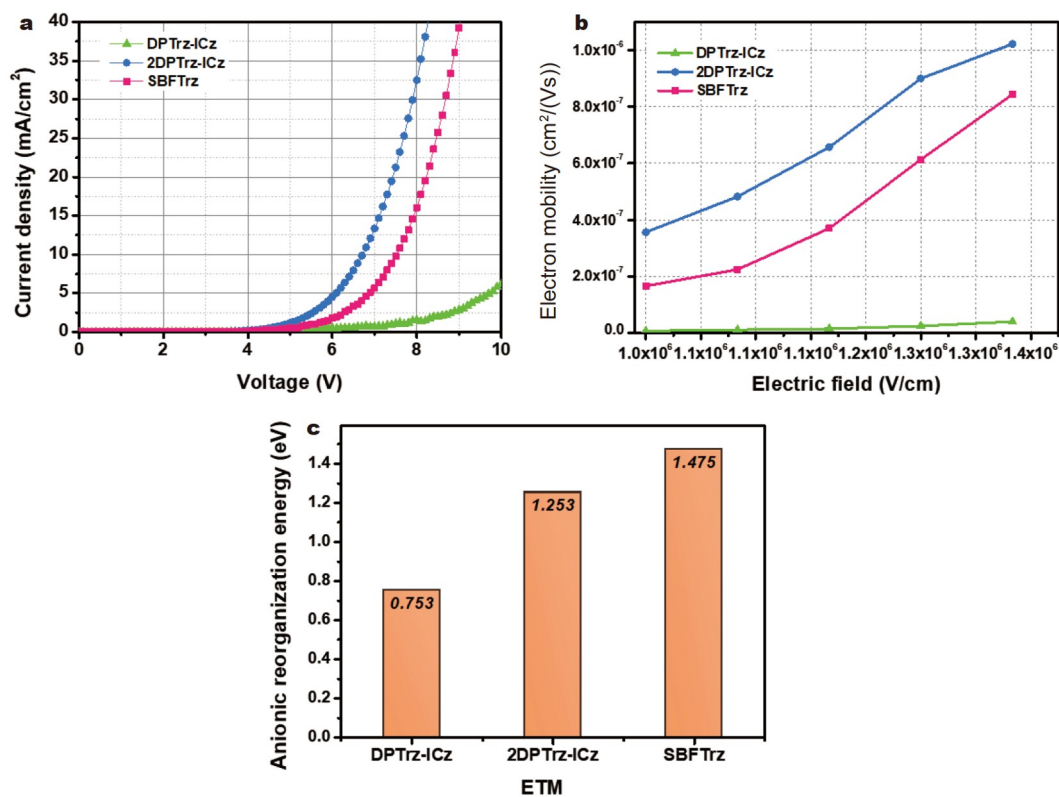
	$\lambda_{ab}^a$ (nm)	$\lambda_{em,max}^b$ (nm)	$E_S^{c/d}$ (eV)	$E_T^{e/f}$ (eV)	$E_g$ (eV)	HOMO/LUMO $^g$ (eV)	$T_d^h$ (°C)
DPTrz-ICz	381	398	3.41/3.31	2.98/2.68	3.23	-5.71/-2.48	402
2DPTrz-ICz	398	401	3.24/3.06	2.84/2.73	3.11	-6.13/-3.02	550

a) Onset wavelength of UV-vis absorption spectra in THF solution ( $10^{-5}$  mol L $^{-1}$ , 273 K). b) The peak wavelength of the PL spectra in THF solution ( $10^{-5}$  mol L $^{-1}$ , 273 K). c) The onset and d) peak wavelength of low-temperature fluorescence in frozen THF ( $10^{-5}$  mol L $^{-1}$ , 77 K). e) The onset and f) peak wavelength of low-temperature phosphorescence in frozen THF ( $10^{-5}$  mol L $^{-1}$ , 77 K). g) Ionization energy (HOMO) derived from UV photoelectron spectroscopy,  $E_{LUMO} = E_{HOMO} + E_g$ . h) Decomposition temperature at which the weight loss of 5% from the initial weight occurs.

SBFTrz. Fig. 3a shows that the electron current density of 2DPTrz-ICz is considerably higher than that of DPTrz-ICz and even superior to that of SBFTrz. This can be attributed to the planar ICz core compared with the sp $^3$  carbon-based SBF core. The almost-planar molecular backbone structure and wide-spread LUMO promoted electron hopping between 2DPTrz-ICz molecules. However, the electron current density of DPTrz-ICz was considerably lower than that of the other two molecules, possibly owing to localized LUMO distribution. The electron mobility values measured using impedance spectroscopy at  $1.25 \times 10^6$  V cm $^{-1}$  for DPTrz-ICz, 2DPTrz-ICz, and SBFTrz were  $2.35 \times 10^{-8}$ ,  $9.00 \times 10^{-7}$ , and  $6.13 \times 10^{-7}$  cm $^2$  V $^{-1}$  s $^{-1}$ , respectively (Fig. 3b). The high electron mobility of 2DPTrz-ICz, which is ~1.5 times higher than that of SBFTrz, is derived from its flat and robust molecular structure. The planar geometry of 2DPTrz-ICz induces a densely packed architecture in the film, enhancing the strong overlap of p-orbitals between adjacent molecules. Consequently, the electrons in the p-orbital can easily move to neighbor molecules *via* phonon and electronic vibronic

coupling caused by the vibration of the organic molecules, activating hopping transport [31].

The molecular packing of 2DPTrz-ICz was indirectly analyzed by monitoring the peak wavelength changes in solution and solid films. The PL spectra of 2DPTrz-ICz and SBFTrz were measured by changing the solvent composition of the THF/water mixed solvent. As shown in Fig. S1, the change in the PL peak wavelength owing to intermolecular interaction was observed by inducing the precipitation of the molecules in a water-rich solvent. After aggregation, 2DPTrz-ICz showed a bathochromic shift of 43 nm, while SBFTrz showed a maximum shift of 6 nm. This result presumes that the intermolecular interaction of 2DPTrz-ICz is stronger than that of SBFTrz, facilitating electron hopping. In addition, an anionic reorganization was calculated because it is one of the critical factors affecting the electron mobility of organic materials [32–35]. The calculation results are shown in Fig. 3c. 2DPTrz-ICz had a lower reorganization energy of 1.253 eV compared with 1.475 eV for SBFTrz, indicating that 2DPTrz-ICz can enhance the electron



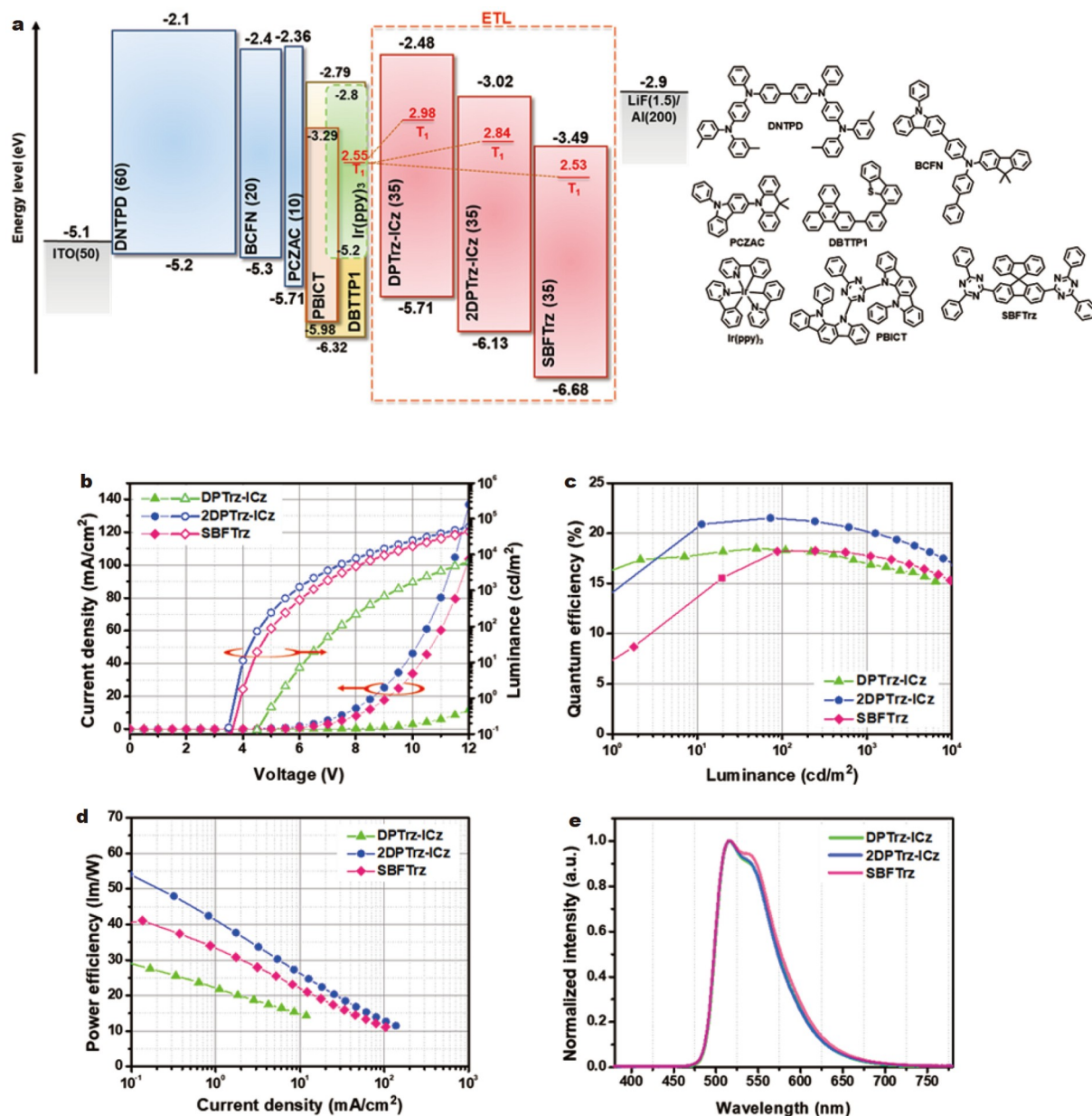
**Figure 3** (a) Current density-voltage curves of 2DPTrz-ICz, DPTrz-ICz, and SBFTz electron-only devices. (b) Electric field *versus* electron mobility curves of DPTrz-ICz, 2DPTrz-ICz, and SBFTz using impedance measurement. (c) Comparison of the anionic reorganization energies of 2DPTrz-ICz, DPTrz-ICz, and SBFTz.

transport *via* the hopping process. Therefore, the low reorganization energy and extensive orbital overlap *via* the planar molecular geometry are responsible for the high electron mobility of 2DPTrz-ICz. In the case of DPTrz-ICz, despite the lowest reorganization energy of 0.753 eV, its electron mobility was smaller than that of 2DPTrz-ICz. This can be attributed to the influence of poor orbital overlap by the localized distribution of the LUMO solely on the triazine moiety as supported by DFT calculations. Additionally, the lack of an electron-deficient group sufficient to offset the hole transport property of ICz appears to be a contributing factor to the reduced electron mobility. To demonstrate this, we fabricated hole-only devices using the following structure to investigate the hole transporting characteristics of three ETMs: ITO (50 nm)/poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS, 40 nm)/N-([1,1'-biphenyl]-4-yl)-9,9-dimethyl-N-(4-(9-phenyl-9H-carbazol-3-yl)phenyl)-9H-fluoren-2-amine (BCFN, 10 nm)/ETMs (50 nm)/BCFN (20 nm)/Al (200 nm). As shown in Fig. S2, the current density with increasing voltage reveals prominent characteristics of hole transport in DPTrz-ICz. These bipolar properties of DPTrz-ICz compromise the electron mobility compared with other ETMs. On the other hand, for 2DPTrz-ICz, the hole transport characteristics of ICz are almost offset, confirming the unipolar behavior for electrons.

The green PHOLEDs with the following layer configuration (Fig. 4a) were fabricated to evaluate the performance of novel ETMs on a device level: ITO (50 nm)/N,N'-diphenyl-N,N'-bis-[4-(phenyl-*m*-tolyl-amino)-phenyl]-biphenyl-4,4'-diamine (DNTPD, 60 nm)/BCFN (20 nm)/9,9-dimethyl-10-(9-phenyl-9H-carbazol-2-yl)-9,10-dihydro-acridine (PCzAc, 10 nm)/

DBTTP1:PBICT:Ir(ppy)<sub>3</sub> (30 nm, dope concentration = 25:75:5)/ETM (35 nm)/LiF (1.5 nm)/Al (200 nm). ITO was utilized as the anode, and LiF/Al was used as the cathode. DNTPD was used as a hole injection layer. BCFN and PCzAc acted as the hole transport and electron blocking layers, respectively. Then, DBTTP1:PBICT (25:75) was used as the cohost in EML, and DPTrz-ICz, 2DPTrz-ICz, and SBFTz were employed as the ETL for each device.

As shown in Fig. 4b–e and Table 2, 2DPTrz-ICz showed a low  $V_{on}$  of 3.5 V and low  $V_d$  of 6.8 V at a brightness of 3000 cd m<sup>-2</sup> owing to its previously demonstrated high electron mobility and the designed step-like configuration of LUMO level for the charge to overcome the energy barrier between the WF of the cathode and the LUMO of EML. Notably, it exhibited an enhanced EQE<sub>max</sub> of 21.5%, higher than that of the previously reported SBFTz, owing to the higher triplet energy of 0.3 eV in 2DPTrz-ICz than that of Ir(ppy)<sub>3</sub> [25], as compared in the diagram in Fig. 4a. This suggests that 2DPTrz-ICz can effectively prevent triplet exciton leakage from EML, increasing its recombination efficiency. Furthermore, the power efficiency was also improved using 2DPTrz-ICz ETM. In contrast, DPTrz-ICz showed considerably high  $V_{on}$  (5.1 V) and  $V_d$  (10.8 V), which can be attributed to the considerably shallow LUMO of the ETM, resulting in a high electron injection barrier from the electrode. And also, despite the small reorganization energy and high triplet energy (2.98 eV), it exhibited underperformed EQE and  $\eta_{PE}$  due to the poor charge balance caused by low electron mobility and shallow HOMO/LUMO levels. Meanwhile, the electroluminescence (EL) wavelength analysis revealed that DPTrz-ICz and 2DPTrz-ICz exhibited a low-intense secondary



**Figure 4** (a) Device configuration of green PHOLED. (b) Current density ( $J$ )-voltage ( $V$ )-luminance ( $L$ ) curves; (c) EQE curves based on luminance; (d) the curves of current density versus power efficiency; (e) the EL wavelength curves of the devices using different ETMs.

peak at 540 nm compared with SBFTrz, indicating the advantages of our ETMs in achieving high-color purity green emission. Furthermore, the device lifetime was measured at 3000 cd m<sup>-2</sup>, and 2DPTrz-ICz showed similar degradation to SBFTrz, demonstrating good stability of 2DPTrz-ICz (Fig. S3). This potential of 2DPTrz-ICz as an ETM in green PHOLED is attributed not only to the inherent property of ICz with a high triplet energy than SBF but also to the enhanced intermolecular charge transfer *via* the fused core and the vigorous activation of intramolecular charge transfer *via* the flat and robust molecular geometry. Therefore, this study confirms the potential of ICz as an ETM beyond the conventional application as an emitter, providing a new paradigm for designing future ETMs.

In addition, in order to confirm the versatility of ETMs in yellow PhOLED (Fig. S4) as well as green PhOLED, yellow

PhOLEDs were fabricated using the following device configuration: ITO (50 nm)/DNTPD (60 nm)/BCFN (20 nm)/PCzAc (10 nm)/CzTrz:PO-01 (30 nm, 5 wt% doped)/ETM (35 nm)/LiF (1.5 nm)/Al (200 nm). As a yellow phosphorescent dopant, iridium(III) bis(4-phenylthieno[3,2-*c*]pyridinato-*N,C2'*)acetylacetonate (PO-01), which showed high PLQY and intersystem crossing rate (ISC rate) [36], was employed and 9-(3'-(4,6-diphenyl-1,3,5-triazin-2-yl)-[1,1'-biphenyl]-3-yl)-9*H*-carbazole (CzTrz) was applied as the host material. The yellow PhOLED employing 2DPTrz-ICz ETM achieved a maximum EQE of 24.1%, demonstrating the universality of 2DPTrz-ICz as an ETL in yellow PhOLED (Table S2).

## CONCLUSION

Herein, two ETMs, DPTrz-ICz and 2DPTrz-ICz, were developed

**Table 2** EL performance data for each device using DPTrz-ICz, 2DPTrz-ICz, and SBFTrz

	$V_{on}^a$ (V)	$V_d^b$ (V)	EQE (%) <sup>c</sup> (3000 cd m <sup>-2</sup> )/Max	$\eta_{PE}$ (lm W <sup>-1</sup> ) <sup>d</sup> (3000 cd m <sup>-2</sup> )/Max	$\eta_{CE}$ (cd A <sup>-1</sup> ) <sup>e</sup> (3000 cd m <sup>-2</sup> )/Max	Color coord. (x,y)
DPTrz-ICz	5.1	10.8	16.2/18.5	16.9/35.6	57.9/66.4	(0.32,0.62)
2DPTrz-ICz	3.5	6.8	19.0/21.5	31.8/59.1	68.5/77.6	(0.32,0.62)
SBFTrz	3.8	7.4	17.0/18.2	25.7/41.0	60.8/65.5	(0.32,0.62)

a) Turn-on voltage at 1 cd m<sup>-2</sup>. b) Driving voltage at 3000 cd m<sup>-2</sup>. c) EQE at 3000 cd m<sup>-2</sup>, maximum. d) Power efficiency at 3000 cd m<sup>-2</sup>, maximum. e) Current efficiency at 3000 cd m<sup>-2</sup>, maximum, of each device.

using ICz as a core. The ICz core, which possesses both planarity and robustness, was coupled with triazine to induce molecular stacking, thereby contributing to enhanced intermolecular charge transfer and electron mobility *via* reinforcing p-orbital overlap. Moreover, the high triplet energy of ICz effectively prevented triplet exciton leakage from EML. Furthermore, incorporating two DPTrz moieties into 2DPTrz-ICz effectively reduced the energy barrier to electron injection between the cathode and the emissive layer. As a result, compared with the previously reported SBFTrz core, the developed ETM showed excellent device performance, including a high EQE of 21.5% and a low  $V_{on}$  of 3.5 V, and it had a positive impact on achieving high-purity green emission.

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**Conflict of interest** The authors declare that they have no conflict of interest.

**Supplementary information** Experimental details and supporting data are available in the online version of the paper.



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