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Carbazole/oligofluorene End-capped Hexanes: Solutionprocessable Host Materials for Phosphorescent Organic Lightemitting Diodes

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Alkyl substituents are promising in improving the solubility and film morphology of aromatic compounds for the solution processing of organic optoelectronic devices. However, the introduction of electronically inert alkyls will generally hinder hole and electron injection and transport into/between the aromatic units, leading to lower electronic properties of the material and thus inferior device performance. Here, we propose an alternative method in using alkyl groups *via* aromatic double end-capping alkylization, where aromatic carbazole and oligofluorenes were linked at both ends of a hexyl chain to obtain a series of carbazole/oligofluorene end-capped hexanes. These newly designed compounds show high solubility, good film morphology, high thermal stability, blue emission, high triplet energy, and suitable frontier orbital energy levels with good charge transport properties; when used as host materials in solution-processed phosphorescent organic light-emitting diodes (PhOLEDs), these molecules exhibit high device performance with maximum current efficiency of 28.0 cd/A and external quantum efficiency up to 7.8%. Our strategy in integrating both advantages of alkyl and aromatic units offers an effective approach to design high-performance solution-processable organic optoelectronic materials.

Introduction

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As an inherent current-driven device, organic light-emitting diode (OLED) may revolutionize current information displays and solid-state lighting industries, especially after the developments of phosphorescent OLEDs (PhOLEDs)^{1,2} and thermally activated delayed fluorescent (TADF)^{3,4} OLEDs capable of harvesting both singlet and triplet excitons for luminescence with 100% internal quantum efficiency^{5,6}. Besides the efforts in improving device efficiency and lifetime, solution processing of OLEDs through spin coating, die coating, or inkjet printing have attracted increasing attentions recently to benefit from the low cost and large-area of the solution-based fabrication techniques for commercial manufacturing⁷⁻¹⁰. For both PhOLEDs and TADF OLEDs, host materials are essentially needed to disperse the emitters to suppress their concentration quenching and triplet-triplet annihilation, meanwhile maintain

the charge transport and energy transfer processes to activate the guest luminophor electronically^{11,12}. To this end, a series of requirements of host materials have to be satisfied simultaneously to support the efficient emission of the guest for high-performance OLEDs¹³⁻¹⁶: (1) high compatibility to well disperse the guest molecules, (2) efficient and balanced hole and electron injection and transportation during the operation of devices, (3) high triplet energy to confine triplet excitons on guest molecules for high luminescent efficiency, (4) high thermal/chemical stability in long-term device application, (5) easy and efficient in synthesis, and so on. And, it becomes even more challenging in developing applicable host materials when the devices were fabricated through solution processes^{17,18}. To date, high-performance host materials are generally bipolar



Scheme 1. Molecular structures of **SFCz**, **DFCz** and **TFCz** designed by aromatic end-capping alkylization strategy.

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small molecules, which are highly aromatic with low solubility and high tendency in crystallization and phase separation when processed *via* solution techniques^{19,20,21}; polymeric and dendrimeric host materials²²⁻²⁴ usually exhibit inferior device performance, although they can be easily solution-processed.

Introducing alkyl substituents onto the aromatic compounds is a widely used method to improve the solubility and film morphology of the material^{25,26}. However, the introduction of the electronically inert alkyl groups will hinder the hole and electron injection and transport into/between the aromatic units, leading to low electronic properties^{27,28}; this should be a main reason for the lower device performance of optoelectronic oligomers, polymers and dendrimers. To overcome the dilemma in increasing the solubility meanwhile reducing the conductivity when using alkyl groups, we propose an aromatic double end-capping alkylization strategy to arrange the inert alkyl groups inside the molecule and the optoelectronic active aromatic units outside, so as to take both advantages of high solubility and film-forming ability from alkyl groups and high optical and electronic properties from aromatic units.

With this strategy in molecular design (Scheme 1), carbazole and oligofluorenes were linked together at their 9 positions by a hexyl chain, leading to a series of carbazole/oligofluorene endof 9,9'-((2-bromo-9H-fluorene-9,9capped hexanes diyl)bis(hexane-6,1-diyl))bis(9H-carbazole) (SFCz), 9,9'-((2,7dibromo-9H-fluorene-9,9-diyl)bis(hexane-6,1-diyl))bis(9Hcarbazole) (DFCz) and 9,9'-((2-(4,4,5,5-tetramethyl-1,3,2dioxaborolan-2-yl)-9H-fluorene-9,9-diyl)bis(hexane-6,1-diyl)) bis(9H-carbazole) (TFCz). The indirect and flexible nonconjugated hexyl linkage offers good solubility of the compound and still remains the high optoelectronic activity of the aromatic carbazole and oligofluorenes units. More importantly, the conjugation length can be also facilely controlled, leading to blue emission and high triplet energy of the materials. When applied as host materials for solution-processed PhOLEDs with 6 wt% green phosphor guest of tris[2-(p-tolyl)pyridine]iridium (III) (Ir(mppy)₃), these carbazole/oligofluorene end-capped hexanes show high device performance with a maximum current efficiency (CE) up to 28 cd/A, a power efficiency (PE) of 11.6 lm/W, and an external quantum efficiency (EQE) of 7.8%, which are comparable to the best results of solution-processed PhOLEDs based on Ir(mppy)₃²⁹⁻³¹.

Results and discussions

Synthesis and physical properties

The alkyl-linked carbazole/oligofluorene compounds of SFCz, DFCz, and TFCz designed by aromatic end-capping alkylization strategy were synthesized in high yields through facile reactions including the alkylation of carbazole and the subsequent alkylation, bromination, boronation and Suzuki coupling of fluorene (see synthesis and characterization in the supporting information, Scheme S1). The facilely prepared target compounds show excellent solubility in common solvents (Table S1), which is favourable for fabricating optoelectronic



Figure 1. TGA curves (a) and AFM height images (5×5 μ m² solution-processed film on glass substrate) of SFCz (b), DFCz (c), and TFCz (d).

devices through solution processes³². Good thermal stability was also observed by thermal gravimetric analysis (TGA) and differential scanning calorimetry (DSC) measurements, showing the increasing decomposition temperature (T_d) from 358°C, 388°C to 458°C and glass transition temperature (T_g) from 56°C, 69°C to 74°C, due to a combined result of increased molecular weight and enhanced molecular rigidity with enlarged fluorene oligomers from **SFCz**, **DFCz** to **TFCz** (**Figure 1a, S1** and **Table 1**).

The morphology of the organic layer, a significant factor in influencing the efficiency of solution-processed PhOLEDs, was then explored. Revealed by atom force microscopy (AFM), the surface of spin-coated **SFCz**, **DFCz** and **TFCz** films is very smooth with a small root-mean-square roughness (RMS) of 0.294, 0.297, and 0.277 nm, respectively (**Figures 1b-1d**). More interestingly, even after annealed at 50°C for 20 min, these films remain amorphous with almost identical RMS values of 0.306, 0.288, and 0.281 nm, respectively (**Figures S2**). The stable, smooth and amorphous films of these carbazole/oligofluorene end-capped hexanes without significant crystallization tendency through spin-coating are highly attractive for solution-processed OLEDs³³.

Optical property

The absorption spectra of **SFCz**, **DFCz** and **TFCz** in CH₂Cl₂ solution show three absorption peaks at 346, 330 and 294 nm (**Figure 2**). The 346 nm low-energy band, which is similar to that observed in fluorene-based molecules, is attributed to π - π * electronic transitions of the fluorene backbone³⁴. Hence, its strength enhancement from **SFCz** to **TFCz** is due to the increased content of fluorene units. The 294 and 330 nm energy bands are attributed to the absorption of peripheral carbazole units, since carbazole shows the characteristic absorption peaks at 293 and 334 nm in dilute solution³⁵. This assignment was further confirmed by the absorption spectra in solid film state, which are very similar to that in solution with only a very small redshift (< 5 nm). Further, from the absorption edges, optical

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bandgaps (E_g) were figured out to be 3.47, 3.34, and 3.13 eV for SFCz, DFCz and TFCz, respectively. In contrast to the absorption spectra that determined by both carbazole and fluorene units, the photoluminescence (PL) spectra are dominated by fluorene units with characteristic 0-0 and 0-1 electric transitions, because fluorenes have higher luminescent efficiency than carbazoles^{36,37}. In CH₂Cl₂ solution, the two PL peaks gradually red-shifted from 356 and 372 nm of SFCz to 399 and 419 nm of TFCz when the number of fluorene unit increases from 1 to 3. At the solid state, the emission peaks of fluorene units show very small red-shifts, showing only decreased 0-0 emission peaks (Table 1); this should be related to the small intermolecular interactions between the emissive fluorene groups in solid state due to the protection of the alkyl-linked peripheral carbazoles, which interrupt the conjugation of fluorene with the alkyl chain and prevent their interactions by the bulky carbazoles. Nevertheless, due to the π - π stacking of the peripheral carbazoles, the emission of carbazole excimer was observable around 520 nm³⁸. The excimer emission is the strongest in SFCz, probably because of its high molecular freedom in rendering the strong interactions between carbazoles. And, this effect also leads to the lowest carbazole excimer emission in DFCz, since the intermolecular interactions of DFCz in partially asymmetric configuration can be remarkably restrained.

Electrochemical Properties

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The electrochemical properties of SFCz, DFCz and TFCz were measured by cyclic voltammetry (Figure S3) to identify their frontier orbital energy levels³⁹. Reversible oxidative electrochemical processes were observed in SFCz, DFCz and TFCz thin films deposited on the surface of glass carbon electrodes, showing oxidation peaks at 0.73, 0.61, and 0.49 V respectively. Based on the oxidative curve and reference energy level of ferrocene (4.8 eV below vacuum)⁴⁰ under the identical electrochemical conditions, the highest occupied molecular orbital (HOMO) energy levels were estimated to be -5.49, -5.37 and -5.21 eV for the corresponding three compounds (Figure 3). However, the reduction curves of the carbazole/oligofluorene end-capped hexanes cannot be observed. Therefore, the lowest unoccupied molecular orbital (LUMO) energy levels are estimated by combining the results of optical bandgap (E_g) measured via the onset of absorption in solid state, according to the relation of LUMO=HOMO+ E_g^{41} . The LUMO energy levels of SFCz, DFCz and TFCz were found to be close, which are at -2.02, -2.03 and -2.08 eV, respectively (Figure 3).

Theoretical investigations

Density functional theory (DFT) calculation can provide in-depth theoretical insights into the nature of optical and electronic properties of organic optoelectronic molecules⁴². From the frontier orbital energy levels and isosurfaces illustrated in **Figure 3**, carbazole units dominate HOMO with similar energy levels, while oligofluorenes determine LUMO with decreased energy levels from momofluorene in **SFCz** to trifluorene in **TFCz**. However, this theoretical prediction is contrary to the experimental observations, which exhibit nearly unchanged



Figure 2. Absorption (closed symbols) and photoluminescence (open symbols, excited at 300 nm) spectra of **SFCz**, **DFCz** and **TFCz** (a) in dilute $(1 \times 10^{-5} \text{ mol } \text{L}^{-1})$ CH₂Cl₂ solution and (b) in thin film.

LUMO and slightly increased HOMO energy levels in SFCz, DFCz and TFCz (Table 1). Such difference may be due to the fact that there are two independent optoelectronic units of carbazole and oligofluorenes linked by an inert alkyl chain. DFT calculations can find the global minima in frontier orbital energies; while CV measurements involve electron/hole injection and extraction processes in/from the molecular surface, where the peripheral electronically active carbazoles may kinetically determine the electrochemical properties, when the inner oliofluorenes are encapsulated by the electronically insert alkyl chains. Consequently, the peripheral carbazoles actually determines both HOMO and LUMO with small energy level variations (<0.3 eV) in the three compounds, regardless the change of the inner oligofluorenes. Therefore, it is not strange that their HOMO and LUMO energy levels are close to the reported frontier orbital energies of carbazole (HOMO = -5.8 eV^{43} and LUMO = -2.1 eV^{44}), indicating clearly the dominating role of peripheral carbazoles on the electrochemical properties of these aromatically end-capped alkyl compounds.

The protection effects of peripheral carbazole substituents on the emissive core of fluorene oligomers were further evidenced in their absorption and emission spectra in different solvents (**Figure S6** and **Table S2**). From the separated electron distributions of HOMO and LUMO, the three compounds seem to be typical donor (D)- acceptor (A) materials, which should show varied PL spectra in solvents with different polarity⁴⁵. In contrast, the features of **SFCz**, **DFCz** and **TFCz** in both absorption and emission spectra are nearly independent of the solvent

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Figure 3. Theoretical (black) and experimental (red) HOMO and LUMO energy levels, triplet energies, frontier orbital isosurfaces and T₁ state spin density distributions of **SFCz**, **DFCz** and **TFCz** (in eV).

polarity with the peak shift less than 5 nm, suggesting that both the ground (S₀) and the lowest singlet excited state (S₁) of the designed compounds are free of solvation. Theoretical (DFT) analysis further reveals that the direct transition from HOMO to LUMO (HOMO \rightarrow LUMO) is almost forbidden in simulated absorption spectra of these compounds, since HOMO and LUMO are separately located at the peripheral carbazole and the oligofluorene core that can hardly overlap for direct photoexcitation (**Table S3**)⁴⁶.

As to the lowest triplet excited state (T_1) energy of the designed molecules, the computed spin density distributions show that oligofluorenes begin to dominate T_1 in **DFCz** and **TFCz** with low theoretical T_1 energies of 2.38 and 2.32 eV respectively. However, the experimental T_1 energies according to the 0-0 transitions of the phosphorescence spectrum measured in 2-

methyltetrahydrofuran at 77 K with a delay time of 5 ms. (Eigure S5) were estimated to be 3.02, 2.69 and 2.68¹eV for SFC2, OFC2 and TFC2, respectively⁴⁷. These T₁ energy values are significantly higher than that of oligofluorenes, which should be attributed to the stronger phosphorescent emission of carbazole units. Therefore, besides the charge transport properties, the peripheral carbazoles actually dominate further the triplet excited states of the carbazole/oligofluorene end-capped alkyls.

Device Performance as host materials

In light of the excellent solubility, high thermal stability, good amorphous film formation property, and apparently high T₁ energy of the designed compounds, the device applications of SFCz, DFCz and TFCz as host materials in solution-processed PhOLEDs were investigated. A simple device configuration was adopted as ITO/PEDOT:PSS (30 nm)/PVK(30 nm)/SFCz (DFCz or TFCz): Ir(mppy)₃ (94:6 wt%, 20 nm)/TPBi (55 nm)/Ca (25 nm)/AI (100 nm) (Figure 4a), in which PEDOT:PSS is poly(3,4ethylenedioxythiophene): poly(styrenesulfonate) as holeinjection layer, PVK is poly(9-vinylcarbazole) as the holetransporting layer, Ir(mppy)₃ is tris[2-(p-tolyl)pyridine]iridium (III) as the phosphorescent emitter, TPBi is 1,3,5-tri(1-phenyl-1H-benzo[d]imidazol-2-yl)phenyl, and the Ca/Al composite cathode is used to enhance the electron injection⁴⁸. The green phosphor of Ir(mppy)₃ has a T_1 of 2.44 eV⁴⁹, which is higher than that of difluorene and trifluorene but lower than that of carbazole^{50,51}. The electroluminescent spectra of the PhOLED devices show exactly the characteristic emission of Ir(mppy)₃ without any emission from the hosts (Figure 4b), indicating that the hosts of SFCz, DFCz and TFCz do have higher T₁ energy to support the complete and efficient energy transfer from host (SFCz, DFCz and TFCz) to dopant (Ir(mppy)₃).

From the current density-voltage-luminance (*J-V-L*) values of these green PhOLEDs based on these new host materials at a low Ir(mppy)₃ doping concentration of 6% (**Figure 4c**), the **DFCz**-hosted device show the best device performance with the lowest driving voltage and the highest electroluminance, reaching a maximum current efficiency (CE) up to 28 cd/A, a power efficiency (PE) of 11.6 Im/W, and an external quantum efficiency (EQE) of 7.8% (**Figure 4d**). These device efficiencies are among the best of the reported solution-processed green PhOLEDs based on the guest of Ir(mppy)₃ (**Table S4**)²⁹⁻³¹. The better performance of the **DFCz**-based PhOLEDs than those of **SFCz** and **TFCz**-hosted devices (**Table S5**) is mainly due to the more balanced hole and electron transportation in **DFCz**-based

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Comp.	T_{d^a}	λ _{abs} (nm)		^{opt} Eg ^b	λ _{em} (nm)		^{exp} ET	CV (eV)		DF	DFT calculation (eV)			
	(°C)	CH ₂ Cl ₂	Film	(eV)	CH ₂ Cl ₂	Film	(eV)	НОМО	LUMO	НОМО	LUMO	^{cal} Eg	^{cal} E _T	
SFCz	358	294,333,347	297,305,333,349	3.47	356,372	362,372,389,515	3.02	-5.49	-2.02	-5.31	-0.85	4.46	2.97	
DFCz	388	294,331,346	297,334,349	3.34	369,387	375,390,411	2.69	-5.37	-2.03	-5.31	-1.36	3.95	2.38	
TFCz	458	294,333,347	297,334,349	3.13	399,419	424,448,526	2.68	-5.21	-2.08	-5.32	-1.55	3.77	2.32	

Table 1. The optical and electrochemical properties of SFCz, DFCz and TFCz.

^aAt 5% weight loss; ^bEstimated from the absorption edge in solid film state.

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Figure 4. (a) Device configuration and energy diagram of the green PhOLEDs hosted by **SFCz**, **DFCz** and **TFCz**; (b) Normalized electroluminescent spectra of the PhOLEDs at 1 mA/cm²; (c) Current density (open)-luminance (solid) voltage curves and (d) efficiency-current density curves of the green PhOLEDs.

devices⁵², as evidenced in the current density-voltage curves of hole-only and electron-only devices in configurations of ITO/PEDOT:PSS (30 nm)/host (20 nm)/Al (100 nm) and ITO/host (20 nm)/TPBi (55 nm)/Ca (25 nm)/Al (100 nm) respectively (Figure S6). By fitting the experimental J-V curves with the fielddependent space charge limited current (SCLC) model (Equation S5), the hole mobilities of SFCz, DFCz and TFCz were found to be 8.25×10⁻⁶, 9.12×10⁻⁶ and 7.69×10⁻⁶ cm² V⁻¹ s⁻¹ respectively, while their electron mobilities are 4.69×10⁻⁷, 7.04×10⁻⁷, 1.38×10⁻⁷ cm² V⁻¹ s⁻¹. The lowest performance of SFCz-hosted device may be also related to its heavy excimer formation in solid film as revealed by PL spectrum (Figure 2b); the excimer with red-shifted emission and lower T₁ energy will significantly reduce the PhOLED's efficiencies. It seems that both the conjugation length and spatial positions of the aromatic units linked together by an alkyl chain into a molecule are important to balance the solubility and electronic properties of the designed materials to an optimal extent for highperformance device applications.

Conclusions

We have demonstrated an aromatic ring end-capped alkyl chain molecular design strategy for solution-processable host materials of OLEDs. The prepared carbazole/oligofluorene endcapped hexanes show high solubility, good film morphology, high thermal stability, blue emission, high triplet energy, and suitable frontier orbital energy levels with good charge transport properties, leading to high device performance with a maximum CE of 28.0 cd/A and an EQE of 7.8%, when used as host materials for solution-processed PhOLEDs of Ir(mppy)₃. To take optimal advantages of this molecular design strategy, both the conjugation length and spatial position of the aromatic units should be carefully controlled. Our results suggest that it is possible to benefit from both the high solubility and good filmforming properties of the alkyl groups and the high optical and electronic activities of the aromatic units, if aromatic units were linked on both ends of alkyl chains in a proper manner. This work would provide important insights into the molecular design of solution-processable organic materials for optoelectronic devices.

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Carbazole/oligofluorene End-capped Hexanes: Solution-processable

Host Materials for Phosphorescent Organic Light-emitting Diodes

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High-performance solution-processable host materials for electrophosphorescent devices have been developed *via* a new method of aromatic double end-capping alkylization.

