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Efficient manipulation of Förster resonance energy transfer through host–guest interaction enables tunable white-light emission and devices in heterotopic bisnanohoops†

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In this study, we synthesized and reported the heterotopic bisnanohoops **P5-[8,10]CPPs** containing cycloparaphenylenes (CPPs) and a pillar[5]arene unit, which act not only as energy donors but also as a host for binding energy acceptors. We demonstrated that a series of elegant FRET systems could be constructed successfully through self-assembly between donors **P5-[8,10]CPPs** and acceptors with different emissions *via* host–guest interaction. These FRET systems further allow us to finely adjust the donors **P5-[8,10]CPPs** and acceptors (BODIPY-Br and Rh-Br) for achieving multiple color-tunable emissions, particularly white-light emission. More importantly, these host–guest complexes were successfully utilized in the fabrication of white-light fluorescent films and further integrated with a 365 nm LED lamp to create white LED devices. The findings highlight a new application of carbon nanorings in white-light emission materials, beyond the common recognition of π-conjugated molecules.

Introduction

Förster resonance energy transfer (FRET)[1] is a distance-dependent photophysical process, in which the energy is transferred from an excited donor to proximal ground-state acceptor fluorophores by means of nonradiative dipole–dipole coupling.[1] Efficient FRET requires a substantial spectral overlap between the emission spectrum of the donor and the excitation spectrum of the acceptor, as well as an optimal distance between them.[2] Upon excitation of the donor, there is a partial transfer of energy between the donor and acceptor fluorophores, resulting in emission from both interactive partners[3] and enabling color-tunable emissions and even white-light emission.[4] On the other hand, white-light emission can also be developed through appropriate integrations of fluorophores that emit the three primary colors (red, green and blue) or two complementary colors within a single molecular backbone *via* covalent linkages.[5] However, in addition to the intricate synthesis process, a significant obstacle for achieving white-light emission is the tendency for molecular arrangements to exhibit undesired non-panchromatic emission due to intra-molecular FRET or through-bond energy transfer.[6] Therefore, an efficient strategy for fabricating FRET systems based on non-

covalent bonds has been developed through host–guest inter-actions, which not only circumvents complex synthesis processes but also minimizes potential emission changes caused by covalent functionalization.[7] During the past few decades, various FRET systems based on host–guest interac-tions[8] have been designed and successfully applied in the construction of white-light emitting materials.[5d,9]

Host–guest systems characterized by non-covalent interac-tions[10] have garnered significant attention in recent years due to their facile syntheses, complementary host and guest properties, optical tenability, various supramolecular structures and unique complexation behaviors.[11] Such host–guest interaction plays a crucial role in the development of tunable white-light emission because it provides an effective approach to generate white-light emission by self-assembly of multiple donor–acceptor mole-cules.[12] In recent decades, there have been various macrocyclic molecules,[13] such as crown ethers,[14] cyclodextrins,[15] calixarenes,[16] cucurbiturils,[17] cycloparaphenylenes[18] and pillararenes,[19] which have been applied to investigate host–guest interaction. Among them, pillar[5]arene is a very attractive host due to its C5-symmetric structure and an overall cylindrical or pillar-like cavity,[20] which enable pillar[5]arene and its derivatives to demonstrate excellent host–guest complexing abilities towards electron-poor or neutral guest molecules.[21] Therefore, they provide an effective pathway for constructing FRET systems and further fabricating tunable fluo-rescent materials.[10a,22] However, few examples have been investi-gated regarding the white-light emission achieved through host–guest interactions based on macrocycles.

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Strained cycloparaphenylenes (CPPs) consisting of *para*-linked phenylenes[23] not only exhibit a common absorbance maximum while their emissions are red-shifted as their sizes decrease,[24] but also can bind fullerenes in their curved cavity.[25] We recently reported a [12]CPP nanoring able to bind non-fullerene guests thus forming ternary complexes with a narcissistic chiral self-recognition.[26] In the present work, by taking advantage of the exceptional size-dependent photophysical properties of CPPs and the host–guest interaction of pillar[5]arene, we incorporated a pillar[5]arene unit into the CPP framework to generate heterotopic bisnanohoops **P5-[8,10]CPP**s with broken symmetry, in which the CPP moieties would serve as energy donors while the pillar[5]arene would act as a host for binding various energy acceptors with different emissions for achieving color-tunable emissions including white light by manipulating FRET systems through host–guest interactions between energy donors and acceptors. It is worth noting that the size-dependent photophysical properties of CPPs suggest that their fluorescent colors can be tuned by simply altering their size without changing their excitation wavelengths. In specific, the **P5-[8]CPP** and **P5-[10]CPP** not only display green and blue emissions upon excitation at the same wavelength, respectively, but also can host energy acceptors emitting red emission in an organized assembly for achieving FRET systems that emit tunable colors in the visible spectral range (400–700 nm), in particular, white-light emission without cutting off the margin region through a filter. It is very important for developing white-light emitting materials through simultaneously using energy donors with different emissions because a slight deviation in excitation wavelength would destroy the balance of colors comprising white-light emission.

In this article, we synthesized heterotopic bisnanohoops (**P5-[8]CPP** and **P5-[10]CPP**) through the incorporation of a pillar[5]arene unit into the CPP framework (Fig. 1) and demonstrated that these elegant FRET systems could be efficiently constructed through self-assembly between **P5-[8,10]CPP** donors and BODIPY- or rhodamine-based acceptors (BODIPY-R or Rh-R) *via* host–guest interaction with a high energy transfer efficiency from **P5-[8,10]CPP**s to BODIPY-R/Rh-R. The combinations of the donors emitting blue or green colors with the acceptors emitting green or red colors through host–guest interactions allow us to finely regulate FRET systems for obtaining multiple color-tunable emissions, particularly white-light emission (Fig. 1). More importantly, these host–guest complexes were not only utilized in the fabrication of white-light fluorescent films but also integrated with a 365 nm LED lamp to create white LED devices. To the best of our knowledge, it is unprecedented that CPPs have been employed for regulating white-light emission. Overall, this work presents a straightforward and efficient strategy for achieving regulation of multiple color emissions, including white light.

Results and discussion

Synthesis and photophysical properties of P5-[8,10]CPPs

In order to construct heterotopic bisnanohoops **P5-[8,10]CPP**s, we incorporated a pillar[5]arene unit into the CPP framework.
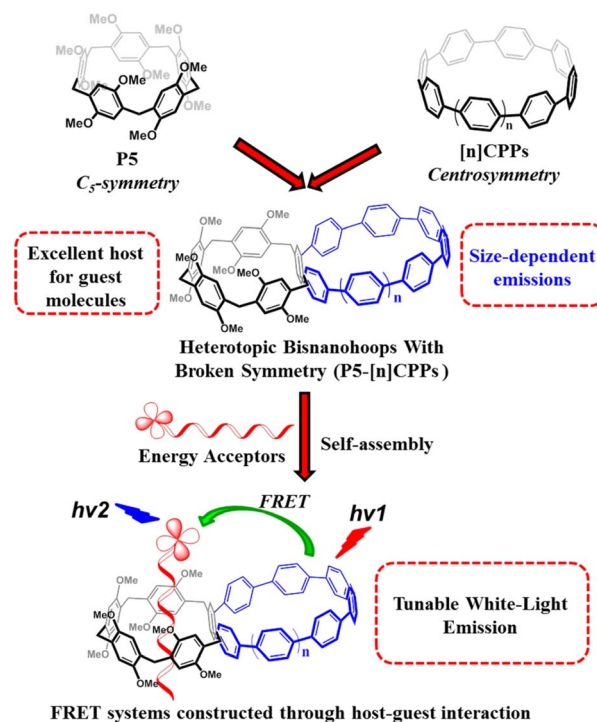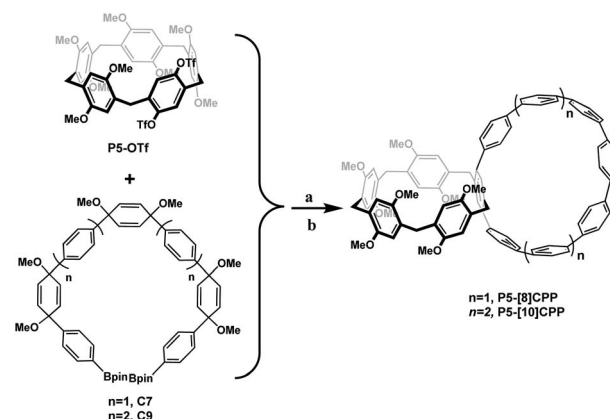

Fig. 1 The chemical structure of heterotopic bisnanohoops and illustration of Förster resonance energy transfers in the energy donors (heterotopic bisnanohoops) and acceptors (BODIPY-R/Rh-R) for achieving tunable white-light emission.

Thus, **P5-[8]CPP** and **P5-[10]CPP** were designed; their synthesis is outlined in Scheme 1 and the detailed procedures are included in the ESI.† The key precursor P5-[*n*]CPP-OMe was obtained from the Suzuki cross-coupling reaction between P5-OTf and compound C7 (or C9). A mixture of equimolar amounts of P5-OTf and compound C7 (or C9) was refluxed with $Pd(PPh_3)_4$ as a catalyst in THF/H_2O (5 : 1) to provide crude P5-[*n*]CPP-OMe, which was directly subjected to a reductive aromatization reaction without further purification in the presence of

Scheme 1 Reaction conditions: (a) $Pd(PPh_3)_4$, K_2CO_3, THF/H_2O (5 : 1), N_2, 70 °C, 36 h; (b) $SnCl_2 \cdot H_2O$, HCl, THF, room temperature, 24 h, yield (two steps): **P5-[8]CPP**, 26%; **P5-[10]CPP**, 25%.

H_2SnCl_4 to generate the desired products **P5-[8]CPP** and **P5-[10]CPP** (ref. 27) with a two-step yield of 26% and 25%, respectively. The structures of **P5-[n]CPPs** were confirmed by ^{1}H NMR, ^{13}C NMR and MALDI-TOF MS.

The photophysical properties of **P5-[8]CPP** and **P5-[10]CPP** were examined by UV-vis and fluorescence spectroscopies, and the photophysical parameters are summarized in Table 1. Similar to the [n]CPPs, the **P5-[n]CPPs** have a common absorption maximum at around 330 nm (Fig. 2) from HOMO → LUMO+1 and HOMO → LUMO+2 transitions (Fig. S2 and S3†), which correspond to the calculated absorption maxima (341–366 nm) with large oscillator strength (f) values (Fig. S4 and S5†). A shoulder peak appearing at 380 nm is assigned to the HOMO → LUMO transition for **P5-[8]CPP**, as well as the HOMO → LUMO forbidden transition for **P5-[10]CPP**. In addition, in this series, there is a red-shifting fluorescence emission as the size of the hoop decreases (visible as a peak at 460 and 510 nm for **P5-[10]CPP** and **P5-[8]CPP**, respectively). Impressively, the fluorescence quantum yields of **P5-[8,10]CPPs** displayed a significant enhancement reaching 43% for **P5-[8]CPP** and 74% for **P5-[10]CPP**, respectively, compared to those of pristine [8]CPP (10%) and [10]CPP (65%). The phenomena were mainly ascribed to symmetry breaking caused by introduction of the pillar[5]arene unit into the CPP backbone. Additionally, fluorescence lifetimes (τ) of **P5-[n]CPPs** (6.3 and 2.9 ns for **P5-[8]CPP** and **P5-[10]CPP**, respectively) decrease in comparison to those of [n]CPPs (17.6 and 6.6 ns for [8]CPP and [10]CPP, respectively) (Tables 1 and S7†). In order to explain enhancement of fluorescence quantum yields, we thus optimized the S_1 state of **P5-[n]CPPs** (Fig. S6†) and calculated the theoretical k_r, which was related to two key parameters (oscillator strength and transition energy). The theoretical k_r values were calculated to

be 1.27×10^8 s^{-1} for **P5-[8]CPP** and 3.64×10^8 s^{-1} for **P5-[10]CPP**, which were consistent with the experimental ones (Table S5†). It is well known that the oscillator strength originates from the overlap of the frontier molecular orbitals.[28] As shown in Fig. S6,† the HOMOs and LUMOs of **P5-[n]CPPs** at the S_1 state all tend to concentrate on cycloparaphenylene portions, so the frontier molecular orbitals exhibit maximum overlap, which results in large oscillator strengths ($f_{max} = 0.5$ and 1.23). In addition, the transition energies of **P5-[n]CPPs** were also calculated to be 5.0 eV, and the synergistic effect between oscillator strength and transition energy led to higher k_r values for **P5-[n]CPPs**. Furthermore, the competition between k_r and k_{nr} contributed to their high fluorescence quantum yield.

Investigation of the FRET process

It is widely acknowledged that pillar[5]arene exhibits a notable capacity to accommodate numerous alkyl derivatives with moderate to high affinity.[29] Accordingly, BODIPY and rhodamine[30] were carefully selected and functionalized with different alkyl chains (Fig. 3A). The functionalized fluorescent dyes were utilized as energy acceptors for constructing FRET systems with **P5-[n]CPPs** through host–guest interactions between the functionalized alkyl chains and pillar[5]arene (Fig. 3B). First, the host–guest interaction between **P5-[10]CPP** and BODIPY-Br was investigated by ^{1}H NMR titration experiments. As indicated in Fig. S10,† the related ^{1}H NMR signals on BODIPY-Br displayed obvious upfield shifts, while the protons on part of pillar[5] arene in **P5-[10]CPP** showed an obvious downfield shift upon addition of BODIPY-Br into a solution of **P5-[10]CPP** in CDCl$_3$ (Fig. S11 and S12†). These observed changes in chemical shifts indicated that the electron-rich cavity of the pillar[5]arene unit within **P5-[10]CPP** is capable of binding BODIPY-Br to form a host–guest complex with a binding constant 3.20×10^5 M^{-1} (K_a was calculated from the fluorescence titration experiment), which is higher than those of most previously reported pillar[5] arene derivatives for similar guest molecules.[31,32] The enhancements in K_a values may be attributed to the alteration of electron density within it's electron-rich cavity caused by functionalization of pillar[5]arene, which subsequently affects the binding affinity towards guests. These results provided an opportunity for the development of FRET-based **P5-[n]CPPs** *via* host–guest interactions.

Generally, one of the most important criteria that should be satisfied for FRET to take place is a significant overlap between the absorption spectrum of the acceptor fluorophore and the emission spectrum of donors. Hence, the investigations on the absorption and emission spectra of **P5-[10]CPP** and BODIPY-Br revealed that the emission spectrum of **P5-[10]CPP** (λ = 460 nm) highly overlap with the absorption spectrum of BODIPY-Br (λ = 500 nm) (Fig. S13†). Therefore, it shows great potential to construct a fluorescence energy transfer system through the self-assembly of **P5-[10]CPP** and BODIPY-Br (Fig. 3B). In order to explore the fluorescence energy transfer from **P5-[10]CPP** to BODIPY-Br, the fluorescence titration experiment was carried out by gradually adding BODIPY-Br into a solution of **P5-[10] CPP** in DCM (Fig. 3C(IV)). The results indicate that the gradual

Table 1 Summary of photophysical properties of **P5-[n]CPPs**

Compounds	λ_ex/nm	λ_em/nm	Φ	Lifetime (ns)
P5-[8]CPP	330	510	43%	6.3
P5-[10]CPP	332	460	74%	2.9

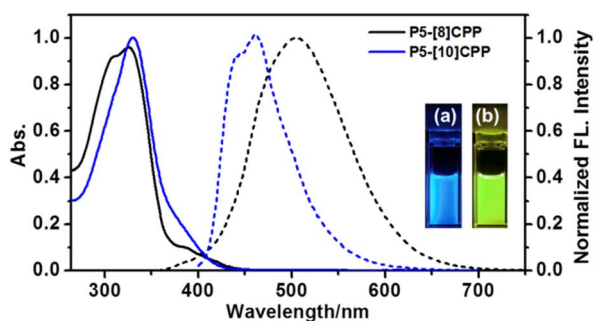**Fig. 2** UV-vis absorption and fluorescence emission spectra of **P5-[8] CPP** and **P5-[10]CPP** in DCM (C = 1 × 10^{-5} M, λ_ex = 330 nm) (inset: fluorescence photographs of (a) **P5-[10]CPP** and (b) **P5-[8]CPP** under UV irradiation at λ = 365 nm).
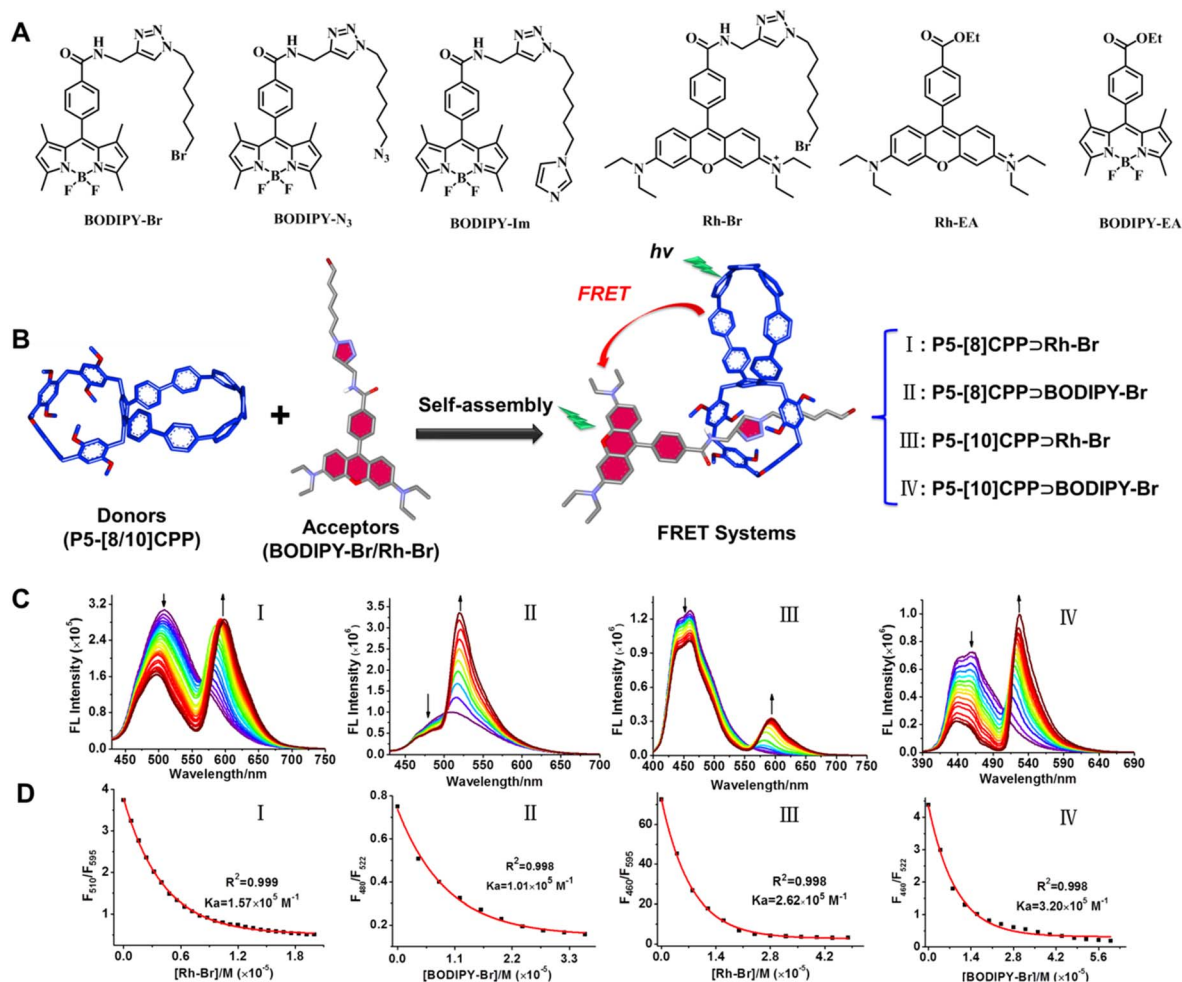
Fig. 3 (A) Structures of acceptors BODIPY-R and Rh-R (R = -Br, -N$_3$, -Im, -EA); (B) the fabrication of the representative FRET systems between donors (P5-[n]CPPs) and acceptors (Rh-Br and BODIPY-Br); (C) fluorescence titration spectra of donors (1 × 10^{-6} M) with addition of receptors in DCM (λ_{ex} = 330 nm); (D) fluorescence titration plot for determination of the binding constant of donors in the presence of various amounts of acceptors ((I) P5-[8]CPP⊃Rh-Br; (II) P5-[8]CPP⊃BODIPY-Br; (III) P5-[10]CPP⊃Rh-Br; (IV) P5-[10]CPP⊃BODIPY-Br).

addition of BODIPY-Br to a solution of **P5-[10]CPP** led to a decrease in fluorescence emission of the donor at 460 nm and a simultaneous increase in fluorescence emission of the acceptor at 522 nm, confirming successful energy transfer from the **P5-[10]CPP** moiety to the BODIPY one, which is consistent with expectations for FRET. The energy transfer efficiency was calculated to be 78% (Table 2). The K_a value was extracted to be 3.20×10^5 M^{-1} (Fig. 3D(IV)). Additionally, the FRET from **P5-**[10]CPP to BODIPY-Br was further confirmed by fluorescence decay profiles (Fig. S29†), and the results showed that the lifetime of **P5-[10]CPP** slightly decreased from 2.90 ns to 2.80 ns upon addition of BODIPY-Br under identical measurement conditions. Furthermore, in order to confirm that FRET resulted from the self-assembly *via* host–guest interaction between **P5-[10]CPP** and BODIPY-Br, a control experiment was carried out by gradually adding ester group-decorated BODIPY (BODIPY-EA) into a solution of **P5-[10]CPP** (Fig. S25†), and the findings showed that the gradual addition of BODIPY-EA to a solution of **P5-[10]CPP** does not induce a decrease in fluorescence emission at 460 nm, but rather results in an increase in fluorescence emission at 522 nm, which can be attributed to the amplification of BODIPY-EA fluorescence resulting from an increase in its concentration. These observations have revealed that the FRET phenomenon arises from self-assembly through host–guest interactions between the donor (**P5-[10]CPP**) and acceptor molecules (BODIPY-Br) (Fig. 3B).

A series of similar FRET systems were also successfully constructed by self-assembly of donors (**P5-[8]CPP** or **P5-[10]**

Table 2 Summary of K_a values and energy transfer efficiency for fluorescence titration of donors (P5-[n]CPPs) and acceptors

Donors	Acceptors	K_a/M^{-1} (× 10^5)	Energy transfer efficiency
P5-[8]CPP	Rh-Br	1.57 ± 0.05	53%
	BODIPY-Br	1.01 ± 0.16	26%
P5-[10]CPP	BODIPY-Br	3.20 ± 0.09	78%
	BODIPY-Im	1.04 ± 0.07	57%
	BODIPY-N$_3$	1.06 ± 0.07	60%
	Rh-Br	2.62 ± 0.10	21%

CPP) with other acceptors (BODIPY-R or Rh-R) (Fig. 3B). The fluorescence titration experiments (Fig. 3C, D and S19–S24†) demonstrated that energies were successfully transferred from the P5-[n]CPPs moiety to the BODIPY (or rhodamine) moiety. The energy transfer efficiency and comparable binding constants were calculated and are listed in Table 2. Furthermore, the alterations in their lifetimes provide additional evidence of energy transfer from donors to acceptors (Fig. S27–S32 and Table S7†). These observations further confirmed the successful fabrication of FRET systems from self-assembly of P5-[n]CPPs and BODIPY-R (or Rh-R).

Moreover, it is very apparent from the fluorescent features of these fluorophores (P5-[10]CPP, P5-[8]CPP, BODIPY-Br and Rh-Br) that these compounds emit the three primary colors (red, green and blue, Fig. 4a). The suitable combination of these fluorophores in an organized assembly would cover the visible

spectral window (400–700 nm), leading to white-light emission without cutting off the margin region through a filter. By fine adjusting self-assembly between different energy donors and acceptors, various FRET systems with tunable multiple color emissions from purple to blue, green, yellow, orange, then to near-white, and even to pure white were readily prepared without complex synthesis and structural modification (Fig. S33–S44 and Table S8†). In particular, upon mixing P5-[10]CPP, P5-[8]CPP and Rh-Br in a molar ratio of 1 : 1 : 1 or P5-[10]CPP, P5-[8]CPP, BODIPY-Br and Rh-Br in a molar ratio of 1 : 1 : 1 : 1, pure white-light emission could be readily obtained (Fig. 4a). In addition, the 1931 Commission Internationale de l'Eclairage (CIE) chromaticity coordinates for the corresponding fluorescence spectra were calculated using "GOCIE" software.[33] The observed CIE coordinates for the 1 : 1 : 1 mixture of P5-[10]CPP, P5-[8]CPP and Rh-Br and for the 1 : 1 : 1 : 1 mixture of P5-[10]CPP, P5-[8]CPP, Rh-Br and BODIPY-Br are (0.33, 0.31) and (0.32, 0.33), respectively, which all match well with the CIE coordinates of the natural white light (0.33, 0.33) (Fig. 4b).

We next conducted a meticulous investigation of the tunable white-light emission process through fluorescence titration experiments, which was carried out by gradually introducing Rh-Br into a 1 : 1 molar ratio mixture of P5-[10]CPP and P5-[8]CPP in DCM. The results suggest that the gradual addition of Rh-Br leads to a progressive decrease in the intensity of the fluorescence emission band within the range of 400 nm to 550 nm, accompanied by a simultaneous increase in fluorescence intensity at 580 nm (Fig. 5b). Throughout the process, the addition of varying ratios of the acceptor (Rh-Br) into the donor (a 1 : 1 mixture of P5-[10]CPP and P5-[8]CPP) resulted in a diverse range of emissions spanning from blue to white, and even to red hues (Fig. 5a). In particular, when the ratio of Rh-Br, P5-[10]CPP and P5-[8]CPP is 1 : 1 : 1, the FRET system exhibits white-light emission. These findings in the emission of the FRET system are consistent with the CIE chromaticity diagram, where the emission spectrum shifts from blue to white and even pink hues (Fig. 5c).

Moreover, we conducted fluorescence titration experiments to investigate the impact of Rh-Br in mixtures of varying ratios of P5-[10]CPP, P5-[8]CPP and BODIPY-Br on the regulation of white-light emission in the FRET process. First, the fluorescence titration experiments were carried out by dropwise adding Rh-Br into the 1 : 1 : 1 ratio mixture of P5-[8]CPP, P5-[10]CPP and BODIPY-Br in DCM. The results indicate that the gradual addition of Rh-Br led to a gradual decrease in the intensity of the fluorescence emission band ranging from 400 nm to 550 nm, while the fluorescence intensity at 593 nm gradually increased (Fig. 5e). Simultaneously, the mixture of Rh-Br and P5-[10]CPP/P5-[8]CPP/BODIPY (1 : 1 : 1) in different ratios yields multiple color emissions from blue-green to white, even orange (Fig. 5d). When Rh-Br, P5-[10]CPP, P5-[8]CPP, and BODIPY-Br reached a ratio of 1 : 1 : 1 : 1, the FRET system displayed white-light emission. In addition, the CIE chromaticity diagram demonstrated a well-matched correlation with these changes in multi-color emission, transitioning from the blue-green region to white and even orange (Fig. 5f). Similarly, upon gradual addition of Rh-Br into the 1 : 1 : 0.5 or 1 : 1 : 2 ratio mixture of P5-[10]CPP, P5-[8]CPP and BODIPY-Br, respectively, a similar change in fluorescence emission could
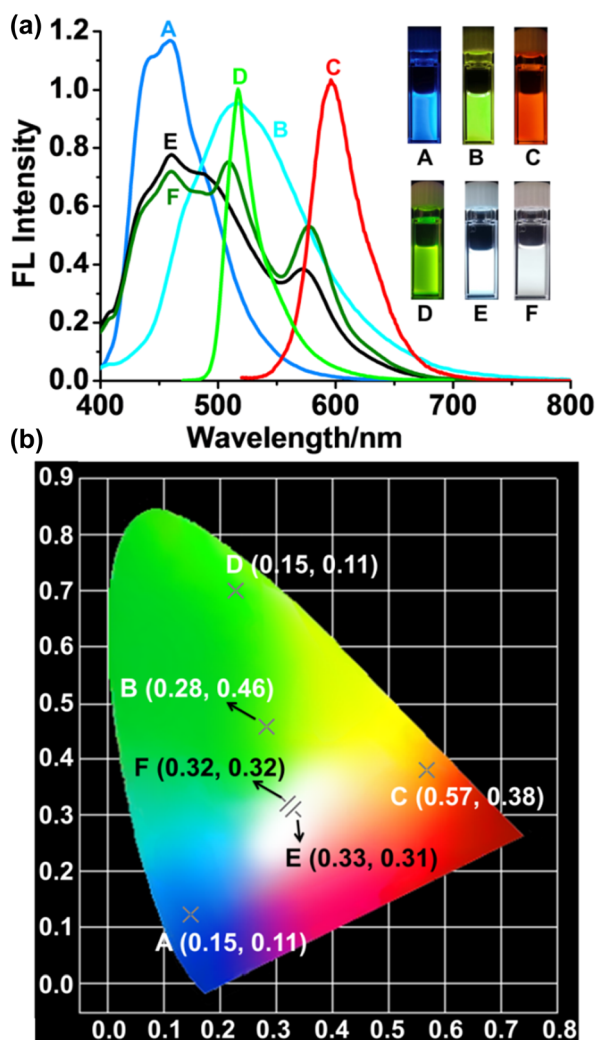

Fig. 4 (a) Fluorescence emission spectra ($C = 1 \times 10^{-5}$ M, $\lambda_{ex} = 330$ nm, DCM); (b) CIE spectrum ((A) P5-[10]CPP; (B) P5-[8]CPP; (C) Rh-Br; (D) BODIPY-Br; (E) the 1 : 1 : 1 ratio mixture of P5-[10]CPP, P5-[8]CPP and Rh-Br; (F) the 1 : 1 : 1 : 1 ratio mixture of P5-[10]CPP, P5-[8]CPP, BODIPY-Br and Rh-Br. Inset: fluorescence photographs under UV irradiation at $\lambda = 365$ nm).
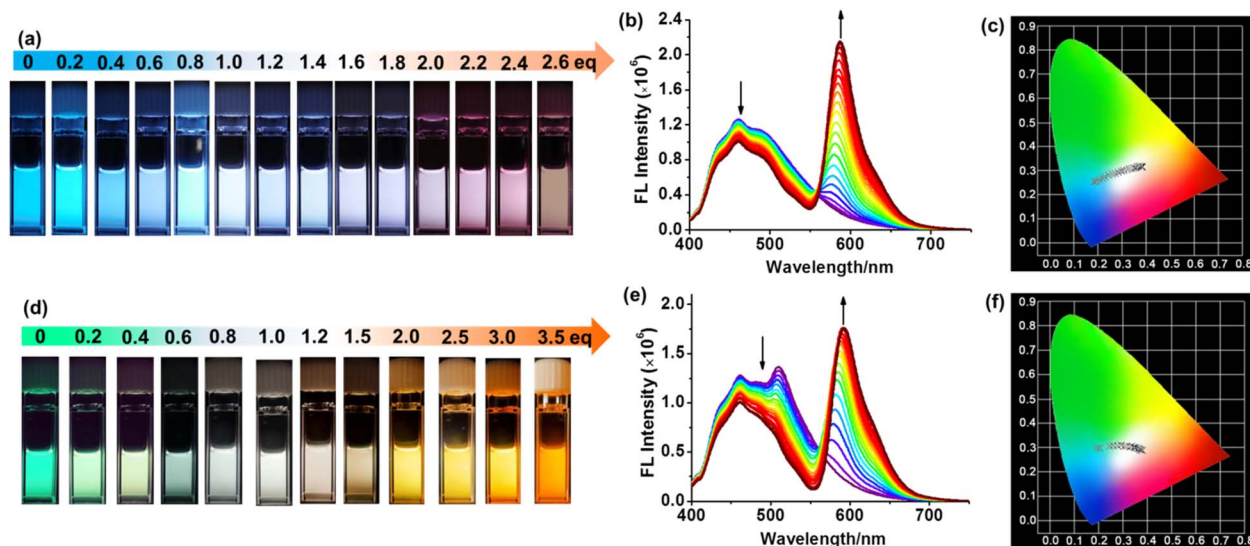
Fig. 5 (a) Fluorescence photographs of the 1 : 1 ratio mixture of **P5-[10]CPP** and **P5-[8]CPP** in the presence of Rh-Br under UV irradiation at $\lambda = 365$ nm (the ratio of Rh-Br to the mixture of **P5-[10]CPP** and **P5-[8]CPP** (1 : 1) ranges from 0 to 2.6); (b) fluorescence spectra of **P5-[8]CPP/P5-[10]CPP** (1 : 1, 1×10^{-5} M, $\lambda_{ex} = 330$ nm) in the presence of Rh-Br; (c) CIE spectrum of **P5-[8]CPP/P5-[10]CPP** (1 : 1, 1×10^{-5} M, $\lambda_{ex} = 330$ nm) in the presence of Rh-Br; (d) fluorescence photographs of the 1 : 1 : 1 ratio mixture of **P5-[10]CPP**, **P5-[8]CPP** and BODIPY-Br in the presence of Rh-Br under UV irradiation at $\lambda = 365$ nm (the ratio of Rh-Br to the mixture of **P5-[10]CPP**, **P5-[8]CPP** and BODIPY-Br (1 : 1 : 1) ranges from 0 to 3.5); (e) fluorescence spectra of **P5-[8]CPP/P5-[10]CPP**/BODIPY-Br (1 : 1 : 1, 1×10^{-5} M) in the presence of Rh-Br; (f) CIE spectrum of **P5-[8]CPP/P5-[10]CPP**/BODIPY-Br (1 : 1 : 1, 1×10^{-5} M) in the presence of Rh-Br.

also be observed (Fig. S47 and S49†), which are consistent with the CIE chromaticity diagram, changing from a blue-green region to a white, even an orange one (Fig. S47 and S49†). The evidence obtained from these experiments suggests that different donor ratios demonstrate a consistent FRET process for Rh-Br, resulting in the emission of fluorescence shifting from blue-green to white or even orange, as well as enabling the creation of a white-light system.

To further investigate the impact of different ratios of acceptors Rh-Br and BODIPY-Br on the regulation of white-light emission in FRET processes, we prepared various ratios of acceptors by increasing the ratio of Rh-Br to BODIPY-Br from 0.0 to 2.0 equivalents. As shown in Fig. 6b, upon the addition of the aforementioned acceptors to a solution containing a 1 : 1 ratio mixture of **P5-[10]CPP** and **P5-[8]CPP** in DCM, respectively, the fluorescence emission in the range of 400–550 nm exhibited a decrease, while the fluorescence intensity at 593 nm showed an increase. The fluorescence exhibited a color shift from green to white and even pink hues, which was in great agreement with the CIE chromaticity diagram (Fig. 6a, S51 and S52†). More importantly, when Rh-Br, BODIPY-Br, **P5-[10]CPP** and **P5-[8]CPP** reached a ratio of 1 : 1 : 1 : 1, that system displayed white-light emission. These findings further suggest that different acceptor-to-donor ratios can facilitate efficient FRET processes with multi-color emission and result in white-light emission.

Finally, we prepared the PMMA films of **P5-[10]CPP**, **P5-[8] CPP**, the mixture of **P5-[10]CPP**, **P5-[8]CPP** and Rh-Br (1 : 1 : 1), and the mixture of **P5-[10]CPP**, **P5-[8]CPP**, BODIPY-Br and Rh-Br (1 : 1 : 1 : 1), respectively. Under a UV lamp excited at $\lambda = 365$ nm, the PMMA films of **P5-[8]CPP** and **P5-[10]CPP** exhibit green and blue emissions, respectively. Significantly, the PMMA

film exhibits white-light emission upon doping with 1 equivalent of Rh-Br or a 1 : 1 mixture of Rh-Br and BODIPY-Br in the 1 : 1 ratio mixture of **P5-[10]CPP** and **P5-[8]CPP**, thus enabling its potential use in solid-state lighting applications (Fig. 7A). Inspired by this, a white-light-emitting diode was fabricated by coating a 365 nm light-emitting diode (LED) bulb with
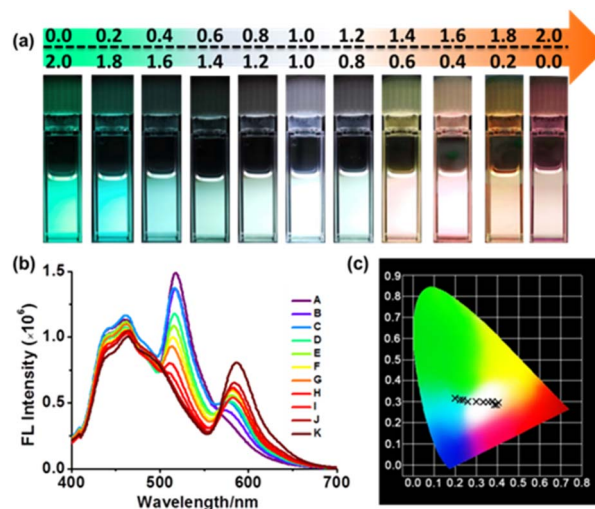

Fig. 6 (a) Fluorescence photographs of the 1 : 1 ratio mixture of **P5-[10]CPP** and **P5-[8]CPP** in the presence of BODIPY-Br and Rh-Br under UV irradiation at $\lambda = 365$ nm; (b) fluorescence spectra of **P5-[8] CPP/P5-[10]CPP** (1 : 1, 1×10^{-5} M, $\lambda_{ex} = 330$ nm) in the presence of BODIPY-Br and Rh-Br; (c) CIE spectrum (the ratio of BODIPY-Br to Rh-Br: (A–K) 0/2.0, 0.2/1.8, 0.4/1.6, 0.6/1.4, 0.8/1.2, 1.0/1.0, 1.2/0.8, 1.4/0.6, 1.6/0.4, 1.8/0.2, and 2.0/0).
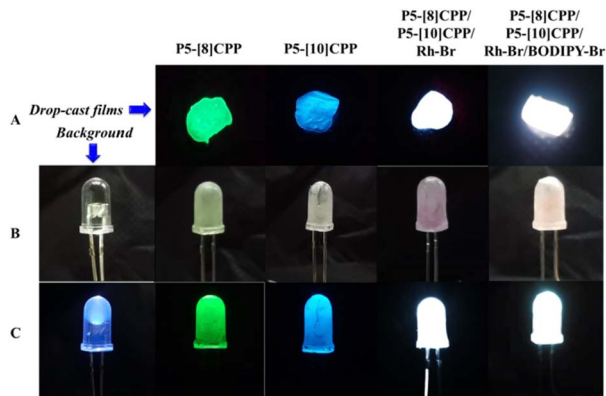
Fig. 7 (A) Drop-cast films; (B) photographs of uncoated LED bulb and coated LED bulb before illumination; (C) photographs of uncoated LED bulb and coated LED bulb after illumination.

a solution containing PMMA and the aforementioned mixture (1 : 1 : 1 ratio mixture of **P5-[10]CPP**, **P5-[8]CPP** and Rh-Br; and 1 : 1 : 1 : 1 ratio mixture of **P5-[10]CPP**, **P5-[8]CPP**, BODIPY-Br and Rh-Br) with white-light emission (Fig. 7B). With the application of a 3 V voltage, these LEDs emit bright white light (Fig. 7C), suggesting that **P5-[n]CPPs**-based drop-cast films with white-light emission may hold potential for future applications in OLED light-emitting devices.

Conclusions

In conclusion, we synthesized heterotopic bisnanohoops (**P5-[8,10]CPPs**) by imbedding a pillar[5]arene unit into the CPP skeleton. The **P5-[8,10]CPPs** showed excellent photophysical properties. Importantly, a series of elegant FRET systems containing donors (**P5-[8,10]CPPs**) and acceptors (BODIPY-R or Rh-Br) were constructed by host–guest interactions. Within these FRET systems, the fluorescence energy was transferred from the CPP unit to the BODIPY (or rhodamine) part with the energy transfer efficiency reaching 78%. Significantly, by fine regulating the self-assemblies between donors (**P5-[8,10]CPP**) and acceptors (BODIPY-Br and Rh-Br) in these FRET systems, multiple color-tunable emissions, particularly white-light emission, were readily achieved without complex synthesis and structural modification. More importantly, these host–guest interactions could not only be utilized in the fabrication of white-light fluorescent films but also integrated with a 365 nm LED lamp to create white LED devices. The findings highlight a new application of carbon nanorings in white-light emitting materials, beyond the common recognition of π-conjugated molecules. The present studies suggest that carbon nanorings can offer a highly efficient and versatile platform for developing white-light emitting devices with potential applications in lighting, displays and sensing technologies.

Data availability

The data that support the findings of this study are available in the ESI† or on request from the corresponding author.

Author contributions

Yanqing Fan designed the experiments, synthesized the compounds, carried out the characterization, and collected and analyzed the data. Hua Jiang and Yanqing Fan wrote the manuscript. Shimin Fan, Jing He, Shengzhu Guo, Zhe Lian, Xiaonan Li and Weijie Guo synthesized partial compounds. Ying Wang analyzed the data; Lin Liu and Xuebo Chen carried out theoretical calculations. Hua Jiang supervised the project. All authors participated in the discussion and revised the manuscript.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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