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# Highly-Emissive Organic Single-Molecule White Emitters by Engineering o-Carborane-based Luminophores

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**Abstract:** The development of organic single-molecule solid-state white emitters holds a great promise for advanced lighting and display applications. Here we report a novel molecular engineering for highly-emissive single-molecule white emitters by design and synthesis of a series of o-carborane-based luminophores. These luminophores are able to induce multiple emissions to directly emit high-purity white light in solid state. By tuning both molecular and aggregate structures, a significantly improved white-light efficiency has been realized (absolute quantum yield 67%), which is the highest value among the known organic single-molecule white emitters in solid state. The fine-tuning of the packing modes from H-to J- and Cross-stacking aggregate as well as intermolecular hydrogen bonds are successful in one molecular skeleton. These are crucial for highly-emissive white-light emission in solid state.

Upon growing need in the advanced lighting and display applications, organic white-light devices have received increasing attentions.<sup>[1]</sup> As white light covers the whole visible region ranging from 400 to 700 nm, white-light devices are usually fabricated by the incorporation of multicomponent emitters which exhibit three primary or two complementary colors of light.<sup>[2]</sup> This approach, however, encouters many thorny problems, such as spectral instability, bad color reproducibility and fabrication complexity.<sup>[3]</sup> The best strategy to solve these problems is to develop single-molecule white emitters. To date, however, few strategies enable single-molecule luminophores to emit white light efficiently in solid state.<sup>[4]</sup> Two crucial issues have determined such slow research progress. On one hand, to achieve white-light emission requires precise manipulation of excited state of emitters through a rational molecular design, which is very difficult to realize. On the other hand, restricted by both the broad band gap<sup>[5]</sup> and the aggregation-caused quenching (ACQ) effect,<sup>[6]</sup> the luminous efficiency in solid state is usually low. Therefore, the design of a highly-emissive solidstate white emitter still remains a big challenge.

To realize a single-molecule white emitter, herein, a new-type molecular design strategy of multiple emissions from tailorable *o*-carborane-based luminophores was proposed (Scheme 1). The unique *o*-carborane bears multiple properties including three-dimentional *pseudo* cage structure and dual electronic effects,<sup>[7]</sup> and has proven to be a promising building block for photofunctional materials.<sup>[8,9]</sup> However, highly efficient white-light emission in solid state by one single carborane-based organic molecule has not been reported. As an electronic acceptor of the

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o-carboranyl unit, multiple emissions from both charge transfer (CT) and locally-excited (LE) state, might be generated for ocarborane-based luminophores. Moreover, the o-carborane cage offers functionalizable positions with which both molecular structures and crystal packing structures might be tuned. All the characteristics of o-carborane motivates us to initiate a novel molecular engineering for highly emissive single-molecule white emitters.

Based on above strategy, in this work, a new class of ocarborane-based luminophores with a D- $\pi$ -A structure was designed and synthesized (Scheme 1). Through the optimization of the  $\pi$ -conjugated donors, o-carborane-based phenanthrene, 1-ph (Scheme 1), is capable of inducing dual emissions from both LE and CT state in solid state to give rise to complementary-color white light with a CIE (Commission Internationale de l' Eclairage) coordinate of (0.33, 0.36) and an absolute quantum efficiency of 46%. Through further finetailoring the molecular structure of 1-ph by suitable substituents (Scheme 2), both the crystal packing structures and intermolecular hydrogen bonds can be tuned to lead to significantly improved white-light luminous efficiency of 67% (3f in Scheme 2). To the best of our knowledge, this is the highest value among the single-molecule white-light emitters in solid state.[3,4]



Scheme 1. The design strategy for single-molecule white emitters.

The *o*-carborane-based luminophores were synthesized through a Suzuki coupling reaction in yields of 41–61% (Scheme S1). Their structures were firmly identified by <sup>1</sup>H, <sup>13</sup>C, <sup>11</sup>B NMR and MS (see Supporting Information). Moreover, the crystal structures of all the *o*-carborane-based compounds were solved (Figure S1). Owing to the bulkiness of the *o*-carboranyl unit, in

general, no intermolecular  $\pi \cdots \pi$  interaction was observed in the packing diagrams (Figure S2). As a result, ACQ effect could be suppressed.

To examine whether the o-carboranyl unit can induce multiple emissions for the designed molecules shown in Scheme 1, the photophysical properties of all compounds were investigated. The absorption spectra display bathochromic shifts in contrast to the corresponding o-carborane-free model compounds (Figure S4), demonstrating the participation of the o-carboranyl unit in an extended  $\pi$ -conjugation.<sup>[10]</sup> In addition, both the o-carboranebased luminophores and the corresponding model compounds feature similar photoluminescence (PL) spectra in CH<sub>2</sub>Cl<sub>2</sub> (Figure S5), but the former are nearly non-emissive ( $\varphi_f < 0.06$ , Table S3). This is ascribed to the variable C-C bond in the ocarboranyl unit which is involved in the excited state to dissipate energy.<sup>[11]</sup> In order to gain in-depth insight into this phenomenon, the time-dependent density functional theory (TD-DFT) calculations were performed. The highest occupied molecular orbitals (HOMO) of these compounds locate in the donor units (e.g. triphenylamine in 1-tpa), whereas the lowest unoccupied molecular orbitals (LUMO) are distributed on the o-carboranyl unit (Figure S14). Particularly, both the charge change (such as 41.6% for 1-tpa, Table S6) and the bond length variation (e.g.  $\Delta$ (C-C)<sub>S1-S0</sub> = 0.79 Å in **1-tpa**, Table S7) for the C-C bond of the o-carboranyl unit can be observed during emission process. This adequately indicates that the C-C bond variation leads to a nonradiative decay from the excited state.



**Figure 1.** a) The PL spectra of *o*-carborane-based luminophores in toluene, inset: luminescence photographs; b) The calculated HOMO and LUMO distributions of **1-ph** and **1-na** in the excited state; c) PL spectra of **1-ph** in THF at 293 K and 77 K; d) PL spectrum of **1-ph** in crystalline state, inset: CIE coordinate, luminescence photographs and the packing structures of **1-ph**.

Next, the PL spectra of all compounds in *n*-hexane and toluene were investigated (Figures S6 and S7). Only **1-tpa**, **1-an**, **1-py** and **1-cz** show the similar multiple emissions (Figure 1a). The emissions around 380-500 nm arise from the LE states of the donor units, and the broad emissions around 500-700 nm are ascribed to the CT emissions from the donor units to the *o*-carboranyl unit.<sup>[9b]</sup> These assignments were further substantiated by TD-DFT calculations (Figure S14). In sharp contrast, **1-ph** and **1-na** exhibit only LE state emission (Figures

S6 and S7) in spite of the presence of good electronic communication between o-carboranyl and the donor unit (Figure 1b). It might be the easy intramolecular rotation of the donor units that disrupts the radiative transition from CT state.<sup>[12]</sup> The DFT calculations (Figure S15) show the rotation barrier (E<sub>rb</sub>) of 6.29 eV for 1-an, 2.26 eV for 1-ph and 1.75 eV for 1-na. The lower Erb for the later two indicates more active intramolecular rotation, thus inhibiting CT emission.<sup>[6]</sup> To further confirm, the low temperature (77 K) PL spectra of 1-na, 1-ph and their corresponding o-carborane-free control compounds in THF were measured (Figures 1c and S8). As a result, a weak broad emission band around 500-700 nm could be observed only for 1na and 1-ph. This demonstrates that the intramolecular CT state can be generated by the introduction of o-carborane, but inhibited in solution for 1-na and 1-ph owing to the ready rotation of the donor unit.

Furthermore, the PL spectra in the crystalline state were studied (Figures 1d and S9). To our delight, 1-ph was the sole compound to display white-light emission with a CIE coordinate (0.33, 0.36) because of the balanced blue and red emissions (Figure 1d). To explore the origin of the white-light emission, other aggregation-state photophysical properties were measured For example, 1-ph exhibits evident aggregation-induced emission (AIE) behavior (Figure S10) in contrast to 9phenylphenanthrene (2-ph) owing to restricted intramolecular motion in aggregation state.<sup>[6b]</sup> With water fraction over 80%, dual emissions were observed. The blue emission, similar to that of 2-ph, arises from the LE state of phenanthrenyl unit. The broad emission around 580 nm, corresponding to the low temperature (77 K) PL spectrum in solution, is attributed to CT emission. Moreover, if doped into polyvinylpyrrolidone (K90), 1ph also exhibits dual emissions (Figure S11). However, the absolute quantum efficiency ( $\phi_{abs}$ ) of **1-ph** in crystals (46%, Table 1) is superior to those of powder (22%) and doped samples (18-22%, Table S4). Thus 1-ph possesses CIEE (crystallization-induced emission enhancement) property.[13] The wide-angle X-ray diffraction measurement for a powder sample presents considerably decreased peak intensity and increased peak widths (Figure S12), implying that ordered molecular packing has been destroyed. In sharp contrast, the crystal structure of 1-ph shows a H-aggregate via intermolecular  $C_{cage}$ -H··· $\pi$  interaction (2.814 Å) between the o-carboranyl and the phenanthrenyl units (Figure 1d). This effectively avoids the  $\pi \cdots \pi$  stacking of phenanthrenyl units, thus giving bright whitelight emission in solid state.

Even having an acceptable white-light emitter **1-ph** in hand, both the H-aggregate and the rotatable spherical shape of the ocarborane cage (Figure 1d) are not beneficial for  $\varphi_{abs}$ .<sup>[14]</sup> We assumed that such a packing structure might be changed *via* regulating the molecular structure of **1-ph** by substitution at the o-carboranyl unit or the phenyl linker. Thus a series of derivatives of **1-ph** were synthesized (Scheme 2, Figure S1). Indeed, the molecular packing modes can be tuned from H-aggregate (**1-ph**, **3-me**, **3-ph**, **3-cl** and **3-f**) to J-aggregate (**3-pr**) and Cross-stacking aggregate (**3-ph-me**) (Scheme 2). Compounds **3-f**, **3-ph-me** and **3-cl** still exhibit white-light emission in crystalline state (Table 1, Figures 2a and S13). Unexpectedly, the bluish-green emission was observed for **3-ph**, **3-me** and **3-pr** (Figure 2b, Table 1). The TD-DFT calculations

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reveal that **3-f**, **3-ph-me** and **3-cl** display the similar distributions of HOMO and LUMO as in **1-ph** (Figure S16a). The introduction of different substituents at the phenyl linker does not alter the orbital distributions. Hence, both the CT state and LE state are present, showing white-light emission. However, the HOMO of **3-me**, **3-pr** and **3-ph** locates in the phenanthrenyl unit, whereas the LUMO centers on the *o*-carboranyl unit, reflecting different electron-density populations in comparison to **1-ph** (Figure S16b). In this case, hence, only CT state is formed, giving rise to a broad bluish-green emission.



Scheme 2. The molecular and packing structures of 1-ph derivatives.

 Table 1. Summary of photophysical parameters of o-carborane-based phenylphenanthrenes.

	1-ph	3-f	3-ph- me	3-cl	3-pr	3-me	3-ph
Colour	white	white	white	white	bluish- green	bluish- green	bluish- green
CIE	(0.33, 0.36)	(0.29, 0.26)	(0.26, 0.26)	(0.28, 0.36)	(0.17, 0.39)	(0.15, 0.28)	(0.15, 0.30)
$\phi_{\text{abs}}$	46%	67%	50%	17%	98%	70%	10%

Surprisingly, the  $\varphi_{abs}$  of these compounds in crystalline state are of significant difference (Table 1), dependent on aggregate structures. In the case of the bluish-green emitters, **3-pr** shows excellent  $\varphi_{abs}$  (98%), much higher than those of **3-me** (70%) and **3-ph** (10%). This proves that J-aggregate facilitates  $\varphi_{abs}$ , whereas the dipole-forbidden nature of H-aggregate determines a lower  $\varphi_{abs}$  (Figure 2c).<sup>[14]</sup> The uniform H-aggregate determines a lower  $\varphi_{abs}$  (Figure 2c).<sup>[14]</sup> The uniform H-aggregate alignment in **3-ph** even leads to the lowest luminous efficiency (Figure S2j). For the white emitters, the  $\varphi_{abs}$  of **3-f** has reached 67%, higher than the known organic single-molecule white emitters (i.e., the reported largest  $\varphi_{abs} = 47\%$ ).<sup>[3,4]</sup> **3-ph-me** and **3-cl** afford 50% and 17%, respectively. **3-ph-me** adopts a Cross-stacking aggregate which also promotes  $\varphi_{abs}$  owing to a progressive transition from the lowest excited state (Figure 2c).<sup>[15]</sup>

We note that the similar H-aggregate of 3-cl, 1-ph and 3-f leads to distinct  $\varphi_{abs}$  values (Table 1). To better understand this,

the single crystal structures were carefully investigated (Figures 1d, S2k, S2l and Scheme 2). In 3-f, both stronger C<sub>cace</sub>-H···F (2.126 Å) and  $B_{cage}-H\cdots\pi^{[16]}$  (2.683 Å) interactions are present, whereas in **3-cl** the  $C_{cage}-H\cdots Cl$  and  $B_{cage}-H\cdots\pi$  hydrogen bonds are no longer formed, and the sole  $C_{cage}$ -H··· $\pi$  interaction (3.291 Å) is extremely weak. In **1-ph**, the only  $C_{cage}$ -H··· $\pi$  (2.814 Å) exsits. We suggest that these intermolecular hydrogen bonds can restrict the rotation of the o-carboranyl cage which is closely related to  $\varphi_{abs}$ . To confirm, the TD-DFT calculations were used to optimize the molecular structures at different rotation angles of the o-carboranyl unit (20°) (Figure S17). The compounds also show a large C-C bond variation (up to 0.73 Å) in the ocarboranyl unit during LUMO→HOMO transition, which implies the deactivation pathway from the excited state.<sup>[11]</sup> The calculations reveal the stronger intermolecular noncovalent interactions around the o-carboranyl unit in 3-f (Figures 3a and S18) which lead to a more compact packing structure (voids 0.2%) than **1-ph** (voids 0.8%) and **3-cl** (voids 1.2%) (Figures 3b and S3). Moreover, by calculating the energy levels of both the single molecule and its dimer, the most stable aggregate is also attributed to 3-f, followed by 1-ph and 3-cl (Figure S19). Hence, the rotation of the o-carboranyl unit can be efficiently restricted by the multiple and stronger intermolecular hydrogen bonds in 3f to form a more stable and rigid aggregate structure, [17] thus emitting strongest white light among the compounds studied.



**Figure 2.** a,b) The solid-state PL spectra of **1-ph** derivatives shown in Scheme 2, inset: CIE coordinates,  $\phi_{abs}$  and luminescence photographs; c) Splitting of the optically allowed transitions of H-, J- and Cross-stacking aggregate.

We also explored the potential applications of these white-light emitters by taking **1-ph** as an example. The thermogravimetric analysis shows high thermal stability with decomposition at 359 °C, over 100 °C higher than the carborane-free control compound (**2-ph**) (Figure S20). Obviously, it is the introduction of *o*-carborane that improves the thermal stability of the organic luminophores. To utilize the efficient white-emissive property of

**1-ph**, a white-emitting device was fabricated as shown in Supporting Information. The device presents a pure and bright white light with CIE coordinates (0.33, 0.36) (Figure 3c). The white-light brightness can reach  $1.4 \times 10^4$  cd m<sup>-2</sup> under a low operating voltage (4.0 V) (Figure S21), far beyond practical use (i.e.  $1.0 \times 10^3$  cd m<sup>-2</sup>). This may provide promising applications for future lighting.



**Figure 3.** a) The calculated intermolecular interactions for **3-cl** and **3-f**, green indicates noncovalent interactions; b) The voids of the unit cells for **3-f** and **3-cl**; c) CIE chromaticity coordinates and PL spectrum of the white LED device from **1-ph**, inset: luminescence photographs.

In summary, an efficient strategy for highly emissive organic single-molecule white emitters has been developed in the ocarborane-based luminophores. Our results demonstrate that incorporation of o-carborane to phenanthrene can induce CT emission and lead to high-purity complementary-color white light in solid state. For the first time, the new-type intermolecular hydrogen bonds (B<sub>cage</sub>-H… $\pi$ , C<sub>cage</sub>-H… $\pi$  and C<sub>cage</sub>-H…F) from the o-carborane cage were designed to suppress nonradiative decay to have realized high-efficiency solid-state white-light emission ( $\phi_{abs}$  up to 67%) for a simple carborane-phenanthrene conjugate. Such a  $\phi_{abs}$  is the highest among so far reported organic single-molecule white-emitters in sold state. In view of the advantages such as easy preparation, pure white light, high  $\varphi_{abs}$  and thermal stability, the functionalization of organic luminophores with o-carborane may open a new gateway for high-performance solid-state light-emitting materials and devices.

#### **Experimental Section**

The synthesis of all compounds was shown in Scheme S1. All experimental details can be found in the Supporting Information.

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**Key words:** carborane • white light • organic luminophore • aggregation induced emission • hydrogen bonds

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### Entry for the Table of Contents (Please choose one layout)

Layout 1:

## COMMUNICATION

A novel molecular engineering for highly-emissive organic singlemolecule white emitters was developed. Through tailoring the molecular structures of o-carboranebased luminophores, crystal packing modes can be tuned, leading to a satisfactory white-light luminous efficiency up to 67%. To the best of our knowledge, this is the largest value among the organic singlemolecule white emitters in solid state.



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