Double-Network Confined Supramolecular Phosphorescence Light-Harvesting Boosting Photocatalysis

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A double-network phosphorescence light-harvesting (PLH) supramolecular system is successfully fabricated based on synergistic confinement strategy. First, the bicationic vinyl bromophenyl pyridine derivative (G) is encapsulated by cucurbit[8]uril (CB[8]), inducing weak phosphorescence emission at 510 nm. Subsequently, the G/CB[8] is confined by in situ copolymerization with acrylamide, displaying enhanced phosphorescence emission, then coassembled with hyaluronic acid modified hydroxypropyl- β -cyclodextrin (HA-HP- β -CD) to form double-network hydrogel, further improving phosphorescence lifetime from 442 µs upto 5.17 ms. Benefitting from the confinement of double-network of supramolecular system, the doped Eosin Y (EY) as acceptor can be photosensitized by the phosphorescence of G/CB[8] leading to delayed fluorescence emission of EY with long lifetime as 36.4 µs. As compared with EY alone, by the synergistic effect of the confined microenvironment and long lifetime photosensitizer, the PLH system is successfully applied in cross-coupling reaction and exhibits enhanced activity, providing a new strategy for the application of phosphorescence energy transfer boosting photocatalysis.

1. Introduction

In recent years, pure organic room temperature phosphorescent (RTP) materials with unique photophysical properties such as long phosphorescence lifetime, high quantum yield, and large Stokes shift, which exhibited wide application in the fields of biological imaging,^[1,2] information encryption,^[3–5] anti-counterfeiting systems,^[6–8] and organic luminescent devices.^[9–11] Among many strategies for the realization RTP emission, the confinement effect of Cucurbit[*n*]urils (CBs) could simply and conveniently realize the efficient phosphorescence emission in water phase.^[12,13] CBs are pumpkin-shaped, water-soluble macrocyclic compounds formed by the condensation of glycoluril and formaldehyde, which have rigid skeleton and a plurality of carbonyl groups, thus could bind cationic guests by electrostatic interaction to form stable host–guest complexes, and its inner cavity not only provides a hydrophobic

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environment for luminophores, but also restricts the movement of guests, inhibits the nonradiative transition of the excited state of luminophores, reduces the collision of solvent molecules, giving a confined microenvironment for phosphorescence emission.^[14-16] Tian et al.^[17] reported that CB[8] could bind with triazine bridged bromophenyl pyridine derivative to form a stable 2:2 hostguest complex, then could be excited by visible light to give tunable phosphorescence emission in aqueous solution. We reported a red phosphorescence emission "supramolecular pins" with ultrahigh quantum yield up to 99.38% based on CB[8] and alkyl chain bridged phenyl pyridine salt.^[18] Thus, the confinement of CBs can effectively induce RTP emission of organic luminophores. In addition, nanosized supramolecular assemblies not only have assemble confined effect is benefit to improving the luminescent properties

of chromophores, but also could further load dyes as energy acceptors to form efficient energy transfer system. Therefore, taking advantage of the strategy of confinement of macrocyclic compounds and assemble could effectively promote the development of PLH systems.

Artificial light-harvesting system with the characteristics of large donor-acceptor ratio and high energy transfer efficiency, which is convenient to realize large Stokes shift luminescence emission, showing fascinating application prospects in the fields of cell imaging,^[19-21] sensor detection,^[22] catalysis,^[23-25] and luminescent materials.^[26-28] George et al. reported that luminescent supramolecular hybrid could be facilely fabricated by the cationic dibromophthalimide derivative and inorganic clay via electrostatic interaction, exhibiting efficient phosphorescence emission with high quantum yield as 41.8%, which also could be doped with energy acceptor to form artificial lightharvesting system.^[29] We reported that dibromophthalimide derivative could coassemble with CB[7] and amphipathic sulfonatocalix[4]arene to form multivalent phosphorescence assembly, then loading dyes as energy acceptors to construct near-infrared emission PLH system with cascaded energy transfer for cell imaging.^[20] However, the application of pure organic PLH system with long lifetime luminescence in photocatalysis reaction has not been reported as best we know. Herein, a doublenetwork PLH system was successfully fabricated by in situ copolymerization of bicationic vinyl bromophenyl pyridine derivative (G), CB[8], acrylamide, and hyaluronic acid modified







Scheme 1. The triple confinement activated phosphorescence light-harvesting system boosting photocatalysis (DF = delayed fluorescence).

hydroxypropyl- β -cyclodextrin (HA-HP- β -CD) as well as energy acceptor Eosin Y (EY) (Scheme 1). In this system, the bicationic luminophore was activated by CB[8] to give weak phosphorescence emission at 510 nm and microsecond lifetime (442 µs). Subsequently, the G/CB[8] as supramolecular cross-linking reagent could in situ copolymerize with acrylamide, then coassembled with HA-HP- β -CD to form double-network hydrogel with multiple hydrogen bonds and rigid structures, which further provides confined microenvironment that is beneficial to the luminescence of chromophore, leading to the double-network system gives strong phosphorescence with long lifetime as 5.17 ms. Moreover, energy acceptor EY was doped into the phosphorescent hydrogel to form PLH system, giving delayed fluorescence signal with long lifetime as 36.4 µs and red-shifted emission at 560 nm. The obtained PLH system not only has a long lifetime photocatalyst sensitized by the phosphorescence of G/CB[8], but also possesses confined microenvironment for photocatalytic reaction, thus showing higher activity (1.9 times) than EY alone.

2. Results and Discussion

Firstly, a cationic bromophenyl pyridine derivative with two positive charges and alkenyl group (G) was synthesized as the luminophore of the PLH system (Scheme S1, Supporting Information), and confirmed its structure by nuclear magnetic resonance (NMR) spectrometer and high-resolution mass spectrometer (HR-MS) (Figures S1–S3, Supporting Information). Next, the binding behavior of CBs and G was comprehensively studied by NMR, HR-MS, and UV-visible spectrometer. As shown in **Figure 1**a, With the addition of CB[8], the absorption

peak of the chromophore was shifted from 305 to 310 nm, and its absorbance intensity decreased, which may be ascribed to the G could interact with CB[8] to form host-guest complex in aqueous solution. As can be seen from Figure S5 (Supporting Information), the Job plot experiment gave a maximum value as 0.67, which implied that the guest was encapsulated by CB[8] to form a 2:1 type complex. After determining the stoichiometric ratio of host and guest, the binding constant between G and CB[8] was measured to be $2.71\times 10^{11}~\text{m}^{-1}$ based on the absorbance intensity changes of G upon addition of CB[8] (0-1.0 eq.) at 305 nm (Figure 1b). Additionally, we also investigated the binding behavior between CB[7] and guest. Although the absorption intensity of G at 305 nm was also decreased with the addition of CB[7], but its absorption peak did not shift (Figures S6, Supporting Information). The binding ratio of CB[7] to G was determined to be 1:1, and the binding constant was 3.42×10^6 m⁻¹ (Figures S7,S8, Supporting Information). These results jointly indicated that G and CB[7] adopted a different binding mode compared with G/CB[8].

Subsequently, the host–guest binding mode of G/CBs was also investigated by NMR experiments. As can be seen from **Figure 2** and Figure S9 (Supporting Information), after addition 0.5 eq. of CB[8], the protons $H_{a,b,c,d}$ belong to the aromatic ring of the guest were shifted from 8.86 (H_a), 8.35 (H_b), and 7.84 (H_{c,d}) to 8.77 (H_{a'}), 7.44 (H_{b'}), 6.87 (H_{b'}) and 6.62 (H_{d'}) ppm, respectively. The upfield shift value of the proton $H_{c,d}$ was significantly larger than $H_{a,b}$, which may be due to bromophenyl was inserted into the cavity of CB[8] and loaded in the shielding area, while the cationic pyridine group was in the carbonyl port of CB[8].^[17] The ¹H NMR titration experiment of G/CB[7] showed that the aromatic group of the guest was also included by host (Figure S10, Supporting Information). In addition, the







Figure 1. a) UV-Vis spectrum of G upon addition of different mount of CB[8]. b) Nonlinear least-squares analysis of the absorbance intensity changes of G upon addition of CB[8] at 305 nm to calculate the binding constant value (Ks) between G and CB[8] (G = 30×10^{-6} m).

mass spectrometry experiments further confirmed that G/CB[8] and G/CB[7] formed a stable 2:1 and 1:1 complex, respectively (Figures S11,S12, Supporting Information). Therefore, through the above research on the binding behavior of CBs and guest, we can reasonably speculate that the bicationic G could be encapsulated by CB[8] and CB[7] to form a 2:1 head-to-tail type and a 1:1 pattern complexes, respectively.

We further studied the influence of confinement of CBs on the luminescence behavior of the guest. The photoluminescence titration experiment showed that the luminophore gave a fluorescence peak at 380 nm when excited at 310 nm (Figure S13, Supporting Information), its fluorescence intensity was gradually decreased with the addition of CB[8], and a new emission peak at 510 nm was observed at the same time. Furthermore, the phosphorescence spectroscopy (delayed 0.1 ms) showed that the G was induced by CB[8] to give a new long

lifetime signal at 510 nm (Figure 3a). The color of G solution was turned from blue to cyan after CB[8] was added under 365 nm UV lamp (Figure 3a inset). It is preliminarily speculated that the observed novel signal may belong to the longlived phosphorescence emission activated by the confinement of CB[8]. The excitation spectrum of G/CB[8] ($\lambda_{em} = 510$ nm) was consistent with the absorption band (Figure 3b), indicating the emission signal at 510 nm was derived from the excited G/CB[8] complex. The luminescence lifetime of this new signal was measured to be 442 µs (Figure 3c), excluding it was a short lifetime fluorescence peak. In addition, the luminescence intensity of G/CB[8] was obviously increased after nitrogen gas was bubbled into the system (Figure 3d) to reduce the influence of dissolved oxygen on luminophores.^[17,18] Thus, this novel emission peak at 510 nm should be ascribed to the phosphorescence signal from G/CB[8]. In sharp contrast to G/CB[8]



Figure 2. ¹H nuclear magnetic resonance (NMR) spectrum of a) G and b) G/CB[8] ([G] = 1×10^{-3} M, [CB[8]] = 0.5×10^{-3} M). The peaks marked with asterisk (*) were assigned to the protons of CB[8].







Figure 3. a) The phosphorescence spectral of G upon addition of different mount of CB[8] (delayed 0.1 ms) ([G] = 30×10^{-6} M). Inset: A: G, inset B: G/CB[8] under 365 nm. b) Normalized absorption and phosphorescence excitation spectrum of G/CB[8]. c) Photoluminescence lifetime of G/CB[8] ($\lambda_{em} = 510$ nm). d) The phosphorescence spectral of G/CB[8] and G/CB[8] under N₂.

system, the fluorescence intensity of the guest (380 nm) was increased upon addition of CB[7], and no new emission peak was observed in G/CB[7] solution (Figure S14, Supporting Information). Based on the above experiment results, we can reasonably infer that the phosphorescence emission of G/CB[8] may be ascribed to the CB[8] provide a hydrophobic environment for guest and restricts the movement of molecules, inhibiting the nonradiative transition of excited state of luminphores and also reduce the collision of solvents, providing a microenvironment benefit to the phosphorescence emission of G.^[14–16] Thus, by the confinement effect of CB[8], the phosphorescent supramolecular complex of G/CB[8] may be used as a donor in artificial PLH system. The obtained G/CB[8] not only could emit phosphorescence, but also possesses alkenyl groups, which may be acted as supramolecular cross-linking reagent of the phosphorescence polymer. Under UV light irradiation, the double-network supramolecular hydrogel (G/CB[8]-Gel) was successfully obtained by in situ copolymerization of G/CB[8], acrylamide, and HA-HP- β -CD (Figure S4, Supporting Information), then characterized by scanning electron microscopy (SEM) and rheological testing instrument. As shown in **Figure** 4a, the freeze-dried G/CB[8]-Gel exhibited porous structures, indicating crosslinked networks were existed in this polymer. The result of force–strain sweep experiment (Figure 4b) manifested that the hydrogel structure of G/CB[8]-Gel was maintained in the



Figure 4. a) Scanning electron microscopy (SEM) image of G/CB[8]-Gel. b) Strain sweep test of G/CB[8]-Gel ($\omega = 1.00$ Hz, G': storage modulus; G'': loss modulus).



shear strain range from 0.1% to 100%. As shown in Figure S15 (Supporting Information), the loss modulus (*G*") value was less than the storage modulus (*G*') at 1% strain, indicating *G*/ CB[8]-Gel possessed stable hydrogel structure. Although *G*/ CB[8] and acrylamide also could be copolymerized to form hydrogel (Figures S16,S17, Supporting Information), the tested value of *G*' (6.70×10^3 Pa) was smaller than the *G*/CB[8]-Gel (1.25×10^4 Pa) at the 1% shear strain, implying the presence of HA-HP- β -CD could improve the mechanical properties of *G*/CB[8]-Gel. Therefore, the supramolecular hydrogel *G*/CB[8]-Gel with double-network was successfully fabricated by in situ copolymerization of multibuilding blocks.

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Possessing hydroxyl, carbonyl, and carboxyl, the HA-HP- β -CD was selected as the building block, which could coassembly with phosphorescent polymer by multivalent interactions to form rigid microenvironment confining the movement of chromophores, inhibiting the nonradiative transition of the excited state of luminophores, enhancing phosphorescence emission. As shown in Figure 5a, G/CB[8]-Gel gave a phosphorescence emission signal at 510 nm in delayed photoluminescence spectroscopy, and bright cyan color of which was observed by the naked eyes under 365 nm lamp. Time-resolved luminescence experiment showed that the luminescence lifetime of G/CB[8]-Gel was measured to be 5.17 ms (Figure 5b), which was enhanced 11.7 times than G/CB[8] in aqueous solution. In order to further study the influence of double-network structure on the luminescence behavior of supramolecular system, we also investigated the luminescence lifetime of the polymer without HA-HP- β -CD (G/CB[8]/polyacrylamide). Although the luminescence lifetime of G/CB[8]/polyacrylamide reached 1.31 ms (Figure S18, Supporting Information), which was enhanced 3.0 times than that of G/CB[8], this value was obviously less than G/CB[8]-Gel (5.17 ms) with double-network structure. Therefore, by the confined in situ copolymerization with acrylamide, the photoluminescence properties of G/CB[8] were efficiently improved, then the obtained supramolecular polymer coassembled with the second polymer network (HA-HP- β -CD) to form more rigid double-network structure with multi-hydrogen bonds could further enhance phosphorescence emission of luminophores. Furthermore, Figure S19 (Supporting Information) showed that the chemical shift of the protons H_{ad} in G/CB[8] were not changed in the present of 1.0 eq. HP- β -CD, thus the G would not release from CB[8] due to the strong host-guest interaction between G and CB[8]. On the other hand, the protons of G were also without change in the present of HP- β -CD (Figure S20, Supporting Information). The UV-visible titration spectrum (Figure S21, Supporting Information) showed that the absorption band of G did not change with the addition of 5.0 eq. HP- β -CD, indicating the bicationic G could not be included by HP- β -CD. On the basis of these experiments results we reasonably inferred that the functional group (G) was not released from CB[8] and could not encapsulated by HP- β -CD in the coassembly.

Considering the fabricated double-network phosphorescent system not only has good luminescent properties, but also contains HP- β -CD, thus which could further bind with dyes as acceptors to form functional energy transfer systems. As shown in Figure 5a, the absorption band of EY has a good overlap with the emission spectrum of phosphorescence hydrogel, indicating EY may be used as the acceptor in energy transfer system. Subsequently, different mount of EY was added into the phosphorescent hydrogel to investigate the possibility of



Figure 5. a) Normalized UV–Vis spectrum of Eosin Y (EY) and phosphorescence spectral of G/CB[8]-Gel. b) Photoluminescence lifetime of G/CB[8]-Gel ($\lambda_{em} = 510 \text{ nm}$). c) The phosphorescence spectra of G/CB[8]-Gel upon addition of different amount of EY. Inset: A) G/CB[8]-Gel, B) G/CB[8]-Gel/EY. d) Photoluminescence lifetime of G/CB[8]-Gel/EY ($\lambda_{em} = 560 \text{ nm}$).



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the luminescent polymer as light harvesting platform. With the addition of EY (Figure 5c), the phosphorescence signal of the polymer was gradually decreased, and the emission peak at 560 nm belong to EY was observed at the same time. Even trace mount of EY (one percent molar ratio of donor) was added, the emission peak from acceptor still could be clearly observed. When the ratio of donor to acceptor reached 10:1, the energy transfer efficiency was calculated as 62% according to the change of the phosphorescence intensity of the G/CB[8]-Gel at 510 nm. The color of luminescent hydrogel was turned from cyan to yellow under 365 nm lamp (Figure 5c and Figure S22, Supporting Information). Thus, we preliminarily infer that the efficient energy transferred from excited luminophores to the acceptor EY in this double-network phosphorescence system. Additionally, the luminescence lifetime experiments of the energy transfer system showed that the lifetime of the luminophore at 510 nm was decreased from 5.17 to 2.15 ms in the presence of 0.1 eq. acceptor (Figure S23, Supporting Information). Interestingly, the luminescence lifetime of EY at 560 nm in the PLH system was cal. as 36.4 µs, which should be assigned to the delayed fluorescence emission (Figure 5d). Furthermore, the excitation spectrum intensity of EY ($\lambda_{em} = 560$ nm) in the PLH hydrogel system (Figure S24, Supporting Information) was obviously more intensive than the absorption band in the range of 300-400 nm, indicating the long lifetime fluorescence emission at 560 nm was mainly derived from the excited donor. Therefore, high efficiency phosphorescence emission supramolecular system could be achieved by employing the synergistic confinement strategy of macrocyclic compounds and double-network polymer assembly, and then bind with acceptor via host-guest interaction of HP- β -CD, leading to the efficient energy transfer from the triplet G/CB[8] to the singlet acceptor

(TS-FRET path), then giving long-lived fluorescence emission of EY (**Figure 6**c), realizing the construction of PLH system.

We also try to apply this PLH system in cross-coupling reaction to investigate the influence of long lifetime supramolecular photosensitizer originated from phosphorescence resonance energy transfer on photocatalysis. Although EY exhibited reaction activity for the photochemical synthesis under the irradiation of green light, which was also limited by the weak absorption in the wavelength range of ultraviolet and inefficient to use short-wavelength UV light. It is well known that the light harvesting system of plants can efficiently use the captured energy of multiwavelength sunlight for photocatalytic reaction.^[23-25] In our PLH system, G/CB[8]-Gel could be excited by UV light and acted as donor, then benefiting to efficient phosphorescence energy transfer from exited antenna molecules (G) to acceptors, thus the EY in PLH system could be photosensitized by the ultraviolet and visible light (Figure S24, Supporting Information) to give long-lived supramolecular photocatalyst, implying the PLH hydrogel may be used as supramolecular reactor with wide-spectrum light absorption for photocatalytic reaction. Subsequently, xenon lamp was selected as the light source of PLH system to mimic the solar light source, and then the cross-coupling reaction between benzothiazole (I) and diphenyl phosphine oxide (II) (Figure 6a) was used to evaluate the catalytic activity of PLH system. As shown in Figure 6b, the conversion of compound I was up to 82% and the yield of compound III was 62% for photocatalytic reaction in the PLH system. In sharp contrast, the control experiments used free EY as photocatalyst showed that the conversion of compound I and the yield of III were 44% and 32%, respectively. Furthermore, the ratio of phosphorescence light-harvesting system (PLH) G/CB[8]-Gel/EY in the reaction system was fixed at 2 mol%



Figure 6. a) Photoreaction of compound I and II under the catalysis of phosphorescence light-harvesting (PLH) system. b) The conversion of compound I under the catalysis of EY (A) and PLH system (C); the yield of compound III under the catalysis of EY (B) and PLH system (D). c) The simplified Jablonski diagram for the illustration of PLH system and the diagram of photocatalytic mechanism (Abs. = absorption, Fluo. = fluorescence, ISC = intersystem crossing, Non.rad. = non-radiation, Phos. = phosphorescence, TS-FRET = triplet to singlet Förster resonance energy transfer).



(Figure S34, Supporting Information), 4 mol% (Figure S32, Supporting Information), and 6 mol% (Figure S35, Supporting Information), respectively. It was found that 4 mol% amount of photocatalyst gave the highest yield (62%) under the same condition. To understand the possible mechanism of the reaction, the radical scavenger 2,2,6,6-tetramethyl-1-piperidinyloxy (TEMPO) was added into the reaction system, and found that no product was found under the same condition (Figure S36, Supporting Information), which indicated that the photocatalytic reaction may go through a radical mechanism. Based on the above experiment results and relevant literature,^[23] we proposed the possible mechanism of photocatalysis (Figure 6c). This photocatalytic reaction was started with reductive quenching of the excited [EY]^{+•} with compound II to give radical II' and hydrogen radical. Next, compound I reacted with radical II' by nucleophilic addition reaction to form radical III'. Finally, the radical III' was deprotonated to give III, and the hydrogen radicals in the system were combined to form hydrogen. The following advantages of PLH system may be account for the good activities of supramolecular photocatalysis. Firstly, the photocatalyst EY in PLH system can efficiently use UV and visible light to form more active EY species with long lifetime (Figure 6c), which is beneficial to the catalytic reaction. On the other hand, PLH system has hydrophobic environment such as the cavity of HP- β -CD and the hydrophobic region formed by double-network polymer that may provide a confinement microenvironment for the reaction substrates, which is also favorable for the photoreaction. In addition, the photocatalyst EY can be bind by HP- β -CD in the confined microenvironment of macrocyclic and polymer, which can effectively reduce the influence of long-time light irradiation on dye and maintain the photocatalytic activity. Therefore, taking advantage of the confined microenvironment and long lifetime photosensitizer, the PLH system was successfully applied in cross-coupling reaction and exhibited enhanced activity, showing a new insight for the application of phosphorescence energy transfer boosting photocatalysis.

3. Conclusion

In conclusion, a double-network PLH supramolecular system was successfully fabricated by in situ copolymerization of CB[8]/vinyl bromophenyl pyridine salt, acrylamide, and HA-HP- β -CD as well as energy acceptor EY. In this system, the mechanical and phosphorescence properties of the double-network hydrogel could be efficiently enhanced by the triple confinement of CB[8], HA-HP- β -CD, and polyacrylamide, leading to phosphorescence emission at 510 nm and long luminescence lifetime (5.17 ms). Furthermore, acceptor EY could be efficiently loaded into the phosphorescent hydrogel to form PLH system, resulting in efficient energy transfer from triplet phosphors to singlet acceptors, giving long lifetime (36.4 µs) fluorescence emission at 560 nm. Finally, by the synergistic effect of the confined microenvironment and long lifetime photosensitizer, the PLH system exhibited enhanced activity (1.9 times) than EY alone in cross-coupling reaction. Therefore, the double-network confined supramolecular assembly was fabricated by in situ copolymerization not only could induce RTP, but also induced

efficient phosphorescence energy transfer, achieving delayed fluorescence and boosting photocatalysis.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

cucurbituril, cyclodextrin, light-harvesting, phosphorescence, supramolecular assembly

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