Generating long-wavelength absorption bands with enhanced deep red fluorescence and photothermal performance in fused carbon dots aggregates

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Abstract
Carbon dots (CDs) with long-wavelength absorptions and emissions are highly desired for biological applications. Herein, we report a new supra-CDs strategy to construct long-wavelength absorption bands based on fused CDs aggregates (f-CDAs) through a concentration-induced interparticle dehydration process among green emissive CDs (r-CDs) under solvothermal treatment. The obtained fused f-CDAs exhibit an obvious absorption band in 550–700 nm and significantly enhanced deep red fluorescence in N,N-dimethylformamide with photoluminescence quantum yields of 15.6% and high photothermal conversion efficiency up to 26.1% in water. Benefiting from the high photothermal performance, in vivo tumor photothermal therapy has been realized via intratumoral injection of f-CDAs under 655 nm laser irradiated at 0.5 W/cm².

KEYWORDS aggregates, bandgap controlling, carbon dots, deep red emission, photothermal therapy

INTRODUCTION
As an emerging carbon-based nanomaterial, carbon dots (CDs) have received widespread attention due to their distinct advantages such as excellent biocompatibility, low preparation cost, high photoluminescence (PL), and tunable optical properties.1–8 These merits make them potential candidates for various biological applications, including bioimaging,9,10 biosensing,11 and photothermal therapy (PTT).12,13 To satisfy biological applications, in vivo long-wavelength absorption and emissions, peculiar in red to near-infrared (NIR) regions, are highly desired due to the high penetration and low absorption of biological tissues in this wavelength region.14,15

Up to now, a lot of blue and green emissive CDs (r-CDs) with main absorption bands in the ultraviolet (UV) and blue light regions have been reported.16 Great efforts have been paid to tune the optical bandgap of CDs to longer wavelength regions by various strategies, such as size screening,16 heteroatom doping,17 and surface oxidation.18,19 For example, Xiong and co-workers synthesized red emissive CDs by increasing the size of sp²-conjugated domains and incorporation of oxygen species into their surface structures following separation via silica column chromatography.20 Rogach and co-workers developed a seed growth strategy to prepare CDs with the maximum emission centered at 580 nm through increasing the particle sizes.21 Fan and co-workers reported a solvothermal route to obtain N,S-codoped CDs with tunable PL from green to red emissions through increasing S doping concentration.22 In our previous work, we reported the NIR absorption band with enhanced NIR fluorescence from CDs, which can be realized through surface modification with molecules or polymers rich in sulfioxide/carbonyl groups.23 It is worth mentioning that these reported methods of engineering the bandgaps of CDs are mainly in dispersed systems, which usually require precise separation and purification, and are not easy to reproduce. Assembling is an effective approach to construct novel optical and...
photophysical properties in supra nanomaterial systems. In our previous work, we reported supra-CDs with a strong visible-NIR absorption band centered at 700 nm via assembly of UV absorbing CDs through interparticle hydrogen bonds and electrostatic interactions. This assembling induced a long-wavelength absorption band in the supra-CDs did not feature red or NIR fluorescence but resulted in high photothermal conversion efficiency of 59.2%. Recently, we constructed NIR emissive carbon nanorolls by a solvothermal-induced fusing process from r-CDs. Under solvothermal treatment, the concentration-induced interparticle dehydration process happened among r-CDs, resulting in the f-CDAs with obvious absorption bands from 550 to 700 nm wavelength region. The obtained f-CDAs not only inherit the green fluorescence from the r-CDs but also exhibit significantly enhanced deep red fluorescence in N,N-dimethylformamide (DMF) and high photothermal conversion efficiency up to 26.1% in water. Via intratumor injection f-CDAs aqueous solution, satisfied in vivo tumor PTT performance was demonstrated in mice model under 655 nm excitation with a low irradiation density at 0.5 W/cm².

RESULTS AND DISCUSSION

r-CDs were synthesized from urea and citric acid by microwave heating method according to our previous work. f-CDAs were prepared by post solvothermal treatment of high concentrated r-CDs in DMF. Note that, 70 mg/ml r-CDs DMF solution was solvothermal heated at 220°C for 2 h. The obtained dark solution was collected, mixed with twice its volume of ethanol, and centrifuged at 8000 rpm for 10 min to remove residual byproducts. The precipitate was collected and freeze-dried into a dark powder of f-CDAs.

The morphologies of r-CDs and f-CDAs were characterized by transmission electron microscopy (TEM) and atomic force microscopy (AFM). TEM and AFM images illustrated r-CDs were in typical spherical shapes with sizes of 3–6 nm (Figure 1A,E). The inserted high-resolution TEM images of r-CDs showed well-resolved uniform lattice fringes of 0.26 nm, which was attributed to the (0 2 0) lattice plane of graphite. In contrast, f-CDAs exhibited a much larger diameter of 12–22 nm (Figure 1B,D) and higher heights of 7–13 nm (Figure 1E,G) in TEM and AFM images, respectively. Further, a high-resolution TEM image revealed that the f-CDAs were composed of several graphene-like domains with lattice fringes of 0.26 nm in random orientations (Figure 1C and Figure S3). Therefore, f-CDAs were composed of r-CDs aggregates. Considering the heights of f-CDAs were much smaller than their diameters, it can be inferred that the f-CDAs were derived from r-CDs through two-dimensional growth by fusing the edges of the graphite-like cores. Dynamic light scattering analysis further verified the much larger sizes of f-CDAs than r-CDs (Figure 1H). We also post-treated r-CDs in the same solvothermal condition at a low concentration of 1 mg/ml for comparison, which was named with h-CDs. h-CDs represents CDs that fails to fuse together under high temperature solvothermal condition. h-CDs exhibited similar particle sizes with r-CDs under TEM and AFM observations (Figures S1 and S2), indicating the much larger sized f-CDAs were formed through concentration-induced fusing growth of r-CDs under solvothermal treatment.

The X-ray powder diffraction patterns of r-CDs and f-CDAs were shown in Figure 1I. In comparison, the diffraction peak of f-CDAs at 26.4° was much sharper than that of r-CDs, indicating larger particle sizes with higher content of crystalline graphitic cores, which agrees well with their TEM observations.

The chemical compositional changes between r-CDs and f-CDAs were further investigated by X-ray photoelectron spectroscopy (XPS). Full XPS spectra and the calculated C, N, and O contents of r-CDs and f-CDAs are shown in Figure 1J and Table 1. Comparing with r-CDs, f-CDAs exhibited decreased O content from 25.7% to 20.2% and increased C content from 54.3% to 60.8% (Table 1). The O 1s spectra of r-CDs and f-CDAs (Figure 1K) showed three peaks at 530.9, 531.9, and 533.1 eV, which were corresponded to C=O, C-O/C=OH, and O=C=O bonds. Obviously, the relative contents of C=O/C=OH and O=C=O bonds in f-CDAs was much lower than that in r-CDs, indicating that the carbonyl groups are dehydrated with hydroxyl or amino functional groups among the r-CDs during the post solvothermal treatment. The Fourier transform infrared spectrum of r-CDs and f-CDAs (Figure 1L) exhibited the characteristic absorption bands of ν(C=O) (1704/cm) and ν(C=C) (1624/cm). By comparison, the obviously decreased C=O bonds and enhanced C=C bonds in f-CDAs indicate the extended conjugated domains. Combining with the TEM observations, it can be concluded that f-CDAs were composed of fused r-CDs aggregates through the interparticle dehydration process among the carboxyl and hydroxyl groups on the surface of r-CDs.

The optical properties of CDs were further investigated. Figure 2A and S4 showed the UV-visible (UV-vis) spectra of r-CDs, h-CDs, and f-CDAs in DMF and water dilute solutions. It can be seen that both of the main absorption bands of h-CDs and f-CDAs slightly red-shifted to 425 nm, indicating extended π-π conjugation caused by increased carbonization of the carbon cores through post solvothermal treatment. It should be noted h-CDs only exhibited enhanced absorption tails in visible regions, while f-CDAs exhibited clearly new absorption bands in 550 to 700 nm regions. Under 405 nm excitation, the green fluorescence from h-CDs and f-CDAs was weakened compared to that from r-CDs (Figure 2B). Both of the h-CDs and r-CDs exhibited very weak emissions under red light excitation (Figures S5 and S6), while significantly enhanced deep red emissions were observed in f-CDAs under 635 nm excitation with photoluminescence quantum yield up to 15.6% (Figure 2C and Table 2). Thus, it can be inferred that the enhanced long-wavelength absorption and emission are generated from the interface of the fused r-CDs aggregates.

The PL lifetimes of the green and deep red emissions from r-CDs, h-CDs, and f-CDAs were further investigated.
At 405 nm excitation, the green emission band from r-CDs exhibited a single exponential decay with a lifetime of about 11 ns (Figure 2D). In contrast, the green emission bands from h-CDs and f-CDAs exhibited multi-exponential decays with decreased average lifetimes, indicating energy dissipation caused by post solvothermal treated surface or fused particle interfaces. At 640 nm excitation, the emissions from r-CDs and h-CDs exhibited different multi-exponential decays with gradually decreased average lifetimes in longer wavelength regions (Figure 2E,F), indicating different and non-uniformed long-wavelength emission centers with continuous energy dissipation by defect states.[33] In contrast, f-CDAs exhibited a single exponential decay with an unchanged lifetime of 2.3 ns monitored from 675 to 745 nm, indicating a certain deep red emission center. Considering the clear absorption band of f-CDAs with efficient deep red emission, it can be inferred the enhanced absorption tails in the longer visible region for h-CDs were attributed to increased surface defect states generated after post solvothermal treatment in the dispersed system.

Based on the results above, it can be concluded that the fused interfaces in f-CDAs accounted for the emerged deep red bandgap emissions. A possible mechanism for the formation process from r-CDs to f-CDAs and energy structures is illustrated in Figure 3. Under the solvothermal condition, high concentrated r-CDs dynamically collided and
dehydration processes happened among the abundant carboxyl and hydroxyl groups on the edge sites of the graphitic cores, leading to the formation of fused r-CDs aggregates in three-dimensional growth, which are the as-prepared f-CDAs. In our previous work, we have demonstrated supra-CDs with long-wavelength absorption can be generated through the self-assembling of CDs with absorption bands in the UV region.\textsuperscript{[24]} The long-wavelength absorption band of supra-CDs was proposed to be induced from the neighbored electron-withdrawing groups and electron-donating groups in the supra-CDs. Thus, we proposed the long-wavelength absorption and emission of f-CDAs are generated from the fused interface structure, in which the electron-withdrawing groups are close to the electron-donating groups on the
TABLE 2 Photoluminescence quantum yields (PLQYs) of green emissive CDs (r-CDs), h-CDs, and fused CDs aggregates (f-CDAs) in N,N-dimethylformamide (DMF) and water solutions under 420 and 640 nm excitations.

|       | r-CDs | h-CDs | f-CDAs |
|-------|-------|-------|--------|
| 420 nm Water | 23.8% | 3.13% | 11.2% |
| DMF   | 33.5% | 9.48% | 26.3% |
| 640 nm Water | 0.011% | 0.02% | 1.11% |
| DMF   | 0.37% | 0.43% | 15.6% |

surface of the fused r-CDs aggregates. As the original green emissive carbon cores were trapped by the fused particle interfaces, the f-CDAs inherited the green bandgap emissions. It is interesting to find the deep red fluorescence from f-CDAs can be significantly quenched in an aqueous solution with a much shorter fluorescence lifetime, as shown in Figure 4A, indicating a serious nonradiative transition in the form of heat caused by water molecules. Thus, the photothermal property of f-CDAs in water was further tested. The photothermal performance of f-CDAs aqueous solution was visualized using an infrared thermal camera (Figure S7). It was clearly seen that after irradiation for 10 min, the temperature of the f-CDAs aqueous solution quickly increased by 48.2°C from room temperature, while the temperature of r-CDs and h-CDs aqueous solutions (200 μg/ml) and pure water only increased by 15.1, 16.4, and 0.5°C, respectively, under the same laser irradiating condition.

Figure 4C showed the temperature increases of 800 μg/ml f-CDAs aqueous solutions at various power densities. The temperature of f-CDAs aqueous solutions rapidly increased by 34.8, 58.1, and 64.5°C from room temperature under 655 nm laser irradiation for 10 min at 0.5, 1, and 1.4 W/cm², respectively. The photothermal conversion efficiency of f-CDAs in aqueous solution at 200 μg/ml was calculated to be approximately 26.1% under 655 nm laser irradiation at 1.4 W/cm².[12,34] Furthermore, the f-CDAs exhibited good photothermal stability in an aqueous solution (Figure 4D). After five cycles of laser heating and natural cooling, no substantial deterioration of the photothermal performance was observed in f-CDAs aqueous solution, indicating f-CDAs can be used as a good photothermal agent for PTT application.[15,35,36] The cytotoxicity of the r-CDs and f-CDAs was examined first before the investigation of in vitro tumor treatment. 4T1, HCT116, and Bel7402 cells were incubated with the two CDs. According to Figures S8 and S9, r-CDs and f-CDAs did not inhibit cell viability at concentrations up to 800 μg/ml, suggesting high biosafety and very low cytotoxicity. Then, we investigated the feasibility of using the f-CDAs for PTT in vivo via intratumor (i.t.) injection. Twenty 4T1-tumor-bearing mice were randomly divided into four groups. For the PTT treatment group (G1), the mice were intratumorally injected with f-CDAs (800 μg/ml, 100 μl) and then irradiated by the 655 nm laser for 5 min at 0.5 W/cm² power density. The other three control groups included mice intratumorally injected with r-CDs (800 μg/ml, 100 μl) with laser irradiation (G2), mice intratumorally injected with 100 μl phosphate-buffered saline (PBS) with laser irradiation (G3), and mice intratumorally injected with f-CDAs (800 μg/ml, 100 μl) without laser irradiation (G4).

Under 655 nm laser irradiation, temperature changes at the tumor sites were recorded by an IR thermal mapping apparatus. According to the IR thermographic images (Figure 5A), the temperature of the tumor region injected with f-CDAs rapidly rose to 70°C within 5 min laser irradiation. However, under the same irradiation conditions, the maximum temperature of the tumor areas injected with r-CDs and PBS were only approximately 42–43°C (Figure S10), which had a slight
AGGREGATE

FIGURE 5 (A) Infrared (IR) thermal images of mice with intratumor (i.t.) injection of fused CDs aggregates (f-CDAs) and green emissive CDs (r-CDs) aqueous solutions (800 µg/ml, 100 µl) at 0, 1, 2, 2.5, 3.5 and 5 min under irradiation at the tumor sites by 655 nm laser at 0.5 W/cm². (B–E) Brightfield photos of mice in G1–G4 in the corresponding days after treatments. (F) Tumor growth curves of 4T1 tumors in mice of G1–G4 after treatments (n = 5 per each group). The curves of each group were processed based on the tumor sizes (mean ± SD) of mice. (G) Survival rates of G1–G4 after different treatment.

The fused interfaces in f-CDAs accounted for the generated long-wavelength absorption bands from 550 to 700 nm wavelength region. The obtained f-CDAs not only inherit the green fluorescence from the r-CDs but also exhibit significantly enhanced red and deep red fluorescence in DMF and high photothermal conversion efficiency up to 26.1% in water. In vivo tumor PTT was realized via i.t. injection of f-CDAs (800 µg/ml, 100 µl) under 655 nm laser irradiated (0.5 W/cm², 5 min). After therapy, the life span of mice was prolonged to over 90 days due to the eliminating of tumors. We prospect the fusing supra-CDs strategy could broaden the means of optical bandgap modifications of CDs and generate more attractive properties for carbon nanomaterials.

CONCLUSION

In summary, we developed a new kind of luminescent f-CDAs through a concentration-induced interparticle dehydration process from r-CDs under solvothermal treatment.

EXPERIMENTAL SECTION

Materials and characterization

Citic acid, urea, and DMF were purchased from Sigma Aldrich, and all were used as received without further purification.

A transmission electron microscope (FEI Tecnai–G2–F30 operated at 200 kV) was used for morphological studies. UV-vis absorption spectra were collected on a UV-vis Spectrophotometer (Jasco V-770), and PL spectra–on an Ocean Optics QE pro spectrophotometer. XPS was performed on an ESCALAB 250Xi photoelectron spectrometer (Thermo Fisher Scientific) using Mo as the excitation source. X-ray diffraction analysis was conducted on an X-ray Diffractometer (Rigaku SmartLab 9 kW). The PL quantum yield and PL decay of all nanomaterials were collected at room temperature on an Edinburgh FS5 spectrophotometer. The laser (655 nm) was generated from CNI laser HD-655NM-HS-3W-18061393. Photothermal images were captured by a FLIR E50 (FLIR Systems Inc.) thermal imaging camera.

Synthesis of r-CDs

r-CDs were prepared according to our previous works. Typically, 3 g of citric acid and 6 g of urea were dissolved into 20 ml of deionized water. Then, the mixture was heated in a domestic 750 W microwave oven for about 5 min, during which the color of the solution changed from a colorless liquid to a light brown and finally dark brown clustered solid, indicating the formation of CDs. The solid was then dissolved in water and centrifuged to remove aggregated particles at a speed of 8000 rpm for 20 min three times. The solutions were further purified by dialysis in deionized water using a membrane (MWCO = 0.1–0.5 KD, Spectrum Laboratories). Finally, the clear, yellow-brown r-CDs solutions were obtained and further freeze-dried for use.

Synthesis of f-CDAs

f-CDAs were prepared by post solvothermal treatment of r-CDs in DMF. Note that, 70 mg/ml r-CDs DMF solution was solvothermally heated at 220°C for 2 h. The obtained dark solution was collected, mixed with twice its volume of ethanol, and centrifuged at 8000 rpm for 10 min to remove residual
byproducts. The precipitate was collected and freeze-dried into a dark powder of f-CDAs.

**Photothermal effect measurement**

UNT-T323 digital thermometers with a K-type thermocouple were used for measuring the photothermal effect data. A 1 ml volume of r-CDs and f-CDAs aqueous solutions was introduced into quartz cuvettes, then the cuvettes were irradiated with 655 nm laser power at the power density of 1.4 W/cm² for 10 min. Pure water was used as a control. The thermocouple probe linked the digital thermometer inserted into the tested solutions, which was perpendicular to the light path. The temperature was recorded by the inserted thermocouple of the thermometer.

**In vitro cytotoxicity study**

4T1, HCT116, and Bel7402 cells were used for evaluating the r-CDs and f-CDAs cytotoxicity in the dark through the Alamar Blue assay. Typically, cells were seeded in 96-well plates at a density of 5000 cells per well and cultured overnight. Then the medium was replaced with 100 μl fresh medium containing r-CDs or f-CDAs with various concentrations. After incubation for 24 h, the medium was replaced by a 100 μl fresh medium containing 10% Alamar Blue, and an additional 2–3 h incubation was conducted for the cells. The relative cell viability was calculated according to the absorbance of the correlated cells at 560 nm by a microplate spectrophotometer with the cells only cultured with the medium as a control. Each trial was performed with three wells in parallel.

**In vivo PTT**

The 4T1 tumor mouse model of the Balb/c mice was generated by subcutaneously injected 4T1 cells (5 * 10⁵ per mouse) into the dorsal area of each female Balb/c mouse. When the tumor size reached 100–200 mm³, these Balb/c mice with 4T1 allograft tumors were randomly divided into four groups (n = 5 in each group). The mice in the PTT treatment group were named G1, which was i.t. injected with an f-CDAs aqueous solution (800 μg/ml, 100 μl) and irradiated with 655 nm laser at a power density of 0.5 W/cm² for 5 min. The three control groups included mice i.t. injected with r-CDs (800 μg/ml, 100 μl) with laser irradiation (G2), mice i.t. injected with PBS (100 μl) with laser irradiation (G3), and mice i.t. injected with f-CDAs (800 μg/ml, 100 μl) without laser irradiation (G4). The temperature change of the tumor site was monitored and recorded with a FLIR E50 (FLIR Systems Inc.) thermal imaging camera. Sizes of tumors were measured by using a digital caliper. These mice were anesthetized by tribromoethanol/avertin anesthesia.

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**CONFLICT OF INTEREST**

The authors declare that they have no conflict of interest.

**ETHICAL APPROVAL**

All animal experiments were approved by the University of Macau Animal Ethics Committee under Protocol No. UMAEC-037-2015.

**DATA AVAILABILITY STATEMENT**

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

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