

Conquering Aggregation-Induced Solid-State Luminescence Quenching of Carbon Dots through a Carbon Dots-Triggered Silica Gelation Process

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Supporting Information

ABSTRACT: Aggregation-induced luminescence quenching of carbon dots (CDots) is the main obstacle for their applications in the solid state. Herein, we report a method to produce strongly luminescent CDots@silica composite gels with highly concentrated emitting centers, where the gelation of tetraethyl orthosilicate is initiated by the surface hydroxyl groups of CDots. The key feature of this approach is to prevent both the collision between high concentrated CDots in gelating solution and their aggregation upon drying. The resultant CDots@silica composite xerogel exhibits both high CDots loading fraction (19.2 wt %) and photoluminescence quantum yields (>40%).



INTRODUCTION

Luminescent carbon dots (CDots) possess distinctive merits,¹ such as excitation-dependent luminescence,^{2–6} low cost,⁷ chemical inertness,⁸ low cytotoxicity,⁹ high photostability,^{10–14} and excellent biocompatibility.^{15–18} These advantages inspired extensive studies on CDots, which have already resulted in several applications in various fields,^{19–27} such as CDots-based bioimaging²⁸⁻³⁴ and their use as fluorescent inks.³⁵ Recently, CDots-based solid-state luminescent composites have received increased attention, as they are promising fluorophores for carbon-based light-emitting diodes (LEDs).³⁶⁻⁴³ Conventional white LEDs are based on rare-earth-based phosphors, which are currently experiencing a serious shortage in supply.⁴⁴ Semiconductor quantum dots (QDs) are considered as a potential alternative for LEDs, but the high-performance QDs cause toxicity concerns, because of the presence of Cd. Consequently, CDots, as an emerging and environmentally friendly class of carbon-based luminescent nanomaterials, are coming in focus for developing LEDs. One of the obstacles in realizing efficient CDots-based phosphors is their aggregation-induced luminescence quenching, which can seriously deteriorate their photoluminescence quantum yields (PLQYs) in the solid state.³⁶ The common method of avoiding this undesirable effect is an incorporation of CDots into solid-state matrices, such as poly(vinyl alcohol) (PVA),³⁶ polyhedral oligomeric silsesquiox-⁸ inorganic salts,⁴³ and silica.⁴⁵⁻⁵⁴ High PLQYs of such ane,³¹ CDots-based solid-state luminescent composites are rather

difficult to realize for high CDots loading fractions.^{45,55} For instance, CDots embedded into PVA matrix achieved high PLQYs up to 84% when the CDots loading fraction is only 0.6 wt %, while PLQYs decreased to 16% for the CDots loading fraction of 7.4 wt %.⁵⁵ This happens because CDots have a tendency to collide in solution and aggregate in the solid state if their concentration is high during the fabrication of composites, resulting in decreased PLQYs.⁴⁵ However, in practical applications, high-performance luminescent devices require not only high PLQYs of the constituting fluorophores, but also an overall strong PL emission output, which is guaranteed by high concentration of the luminescent centers in the materials. Hence, a combination of the high PLQYs and the high loading fraction of luminescent CDot centers in their solid-state luminescent composites is strongly desirable.

To decrease the chances of CDots to collide and aggregate in solid state, the movement of CDots should be restricted in the compositing process.⁵⁶ In particular, gel has served as a host structure to obtain a broad variety of nanocomposites with tailored functionalities and superior PL properties.⁵⁷ However, simply loading CDots into the gel would not provide efficient restriction of their movement and still suffer the aggregation-induced luminescence quenching during the drying process.

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Instead, they should be able to contribute into the formation of cross-linked gel architecture serving as gelation centers. To the best of our knowledge, there have been no reports on such a function of CDots, and this is what has been the focus of the present study. In this work, we have employed CDots prepared using citric acid and urea with plenty of hydroxyl groups on their surfaces, being able to form links with gelating silica. Gelation reaction between tetraethyl orthosilicate (TEOS) and CDots at relatively high concentration (0.8 mg/mL) efficiently restricts collisions between CDots, resulting in well-separated emitting centers distributed in formed xerogel, with both high CDots loading fraction (19.2 wt %) and high PLQY (41%).

EXPERIMENTAL SECTION

Materials. Citric acid (99.5%), ammonia-water (25%), tetraethyl orthosilicate (TEOS), ethanol (99.7%), and urea (99%) were purchased from Aladdin. Polydimethylsiloxane (PDMS) elastomer kits (Sylgard 184) were purchased from Dow Corning (Midland, MI, USA). All chemicals were used without further purification. The water used in all experiments was purified through a Millipore system.

Synthesis of CDots with Green Emission. CDots with green emission were synthesized according to our previously published procedure.⁵⁸ Three grams (3 g) of citric acid and 6 g of urea were dissolved in 20 mL of water, and the mixture was heated for 5 min in a domestic 750 W microwave oven, resulting in a dark-brown clustered precipitate. The latter was separated by directly dissolved in ethanol, followed by three cycles of centrifugation at 8000 rpm to remove aggregated particles. The resulting transparent, yellow-brownish CDots ethanol solutions with variable concentration were used for further experiments.

Synthesis of CDots with Blue Emission. CDots with blue emission were synthesized by adding 3 g of citric acid into 20 mL of ammonia–water.⁵⁵ The mixture then was heated in a domestic 750 W microwave oven for \sim 5 min, during which the color of the solution changed from a colorless liquid to dark-brown liquid, indicating the formation of CDots. The products were added into ethanol and centrifuged to remove aggregated particles at a speed of 8000 rpm for 10 min three times.

Fabrication of CDots@Silica Composites. TEOS (0.6 mL) and 0.3 mL of ammonia–water were sequentially added into 8 mL of a CDots ethanol solution. For CDots concentrations in the range of 0.05–0.4 mg/mL, CDots@silica composite nanoparticles were obtained after 10 h at room temperature, while for the CDots concentration of 0.8 mg/mL, it took only 6 h. Xerogel was obtained by freeze-drying of CDots@silica composite gel. CDots@silica composite gel and xerogel with blue emission were synthesized via a similar method, except using a 1.2 mg/mL CDots ethanol solution with blue emission.

Fabrication of LEDs. Commercially available blue-emitting InGaN LED chips (Shen Zhen Hongcai Elecronics Co., Ltd.) were used as an excitation source emitting at 450 nm. For the preparation of the color conversion layer, the CDots@silica composite xerogel with green emission was mixed with PDMS precursor in a mass ratio of 1:1, and filled into the cup-shaped voids of LED chips, followed by curing at 80 $^{\circ}$ C for 1 h.

Characterization. Ultraviolet-visible (UV-Vis) absorption spectra were collected using a Shimadzu Model UV-3101PC spectrophotometer. Photoluminescent (PL) spectroscopy was performed on a Hitachi Model F-7000 spectrophotometer. The absolute PL quantum yields (PLQYs) of CDots ethanol solutions were measured in a calibrated integrating sphere in a Edinburgh Instruments Model FLS 920 spectrofluorimeter. Before characterizing the PL properties of CDots@silica composites, the composites are separated from the solutions through centrifugation. The obtained precipitates then are freeze-dried. The final powders are characterized to obtain PLQYs with a calibrated integrating sphere in a Edinburgh Instruments Model FLS 920 spectrofluorimeter, while their PL intensities are examined *in situ* by the fluorescence microscope equipped with a spectrometer.

Time-resolved luminescence decay curves were measured by a LifeSpec-II dedicated lifetime spectrometer (Edinburgh Instruments). In the photostability studies, the CDots@silica composite xerogel is placed on a quartz plate, and its PL intensity is examined in situ using a fluorescence microscope that has been equipped with a spectrometer for different irradiating times. The commercial fluorescein sodium dye solution is deposited on a quartz plate by drop-casting, allowed to dry, and its bleaching under irradiation is studied using the same equipment. The CDots are deposited on paper to prevent their aggregation-induced luminescence quenching,³⁶ and their photostability is evaluated by the mentioned equipment. Transmission electron microscopy (TEM) images were recorded on a FEI Tecnai-G2-F20 TEM at 200 kV. Scanning electron microscope (SEM) images were obtained on a JEOL Model FESEM 6700F electron microscope with primary electron energy of 3 kV. Energy-dispersive spectra (EDS) and elemental mapping were conducted with the help of Inca X-Max instrument (Oxford Instruments). Powder X-ray diffraction (XRD) measurements were carried out on a Siemens Model D5005 diffractometer. The fluorescence microscopy images were collected on a C2+ confocal microscope system (Nikon). The spectra of the LEDs were measured by a Spectrascan Model PR-650 spectrophotometer with an integral sphere under ambient conditions at room temperature.

RESULTS AND DISCUSSION

CDots were prepared by microwave-assisted synthesis, according to our previous report (see the Experimental Section for details).58 The as-prepared CDots form stable colloidal dispersions both in water and ethanol, with PLQYs of 15% and 36% under 405 nm excitation, respectively. Conventional silicabased nanocomposites are commonly prepared via the Stöber approach, where nanoparticles, TEOS, ammonia, water, and ethanol are directly mixed together.⁵⁹ In the previously reported application of the classical Stöber method to fabrication of CDots@silica composties, CDots were subjected to the surface treatment with silane or other stabilizers, to avoid phase separation, 45-49 while this procedure is complicated. To simplify the process, the as-prepared CDots without premodification are directly used. Nevertheless, our attempts to fabricate CDots@silica composites within the classical Stöber approach failed: Centrifugation of the reacting mixture of TEOS and ammonia-water in CDots water/ethanol (volume ratio of 1:2) separated white silica particles without obvious PL emission as a precipitate, while green emissive CDots hold in the supernatant solution (see Figure S1 in the Supporting Information). This shows that TEOS reacts with water molecules rather than with functional groups on the surface of CDots, and the hydrolyzed TEOS form silica particles that do not physically encapsulate CDots. To further prove this conclusion, the EDS characterizations are performed (Figure S2 in the Supporting Information), indicating the absence of CDots.

We then exploited chemical reactions between TEOS and CDots utilizing the abundant functional groups on the surface of CDots to develop luminescent composites. The gelation process of silica can be described by two types of reactions, generally referred to as the hydrolysis (first step) and the subsequent condensation reaction.⁶⁰ As shown in Scheme 1, TEOS can be hydrolyzed by water to form Intermediate Product 1 (IP1), where k_w is the rate constant of this reaction. Likewise, it has been demonstrated that there are plenty of hydroxyl groups on the surface of CDots (Figures S3 and S4 in the Supporting Information),⁵⁸ and hydroxyl groups can react with TEOS according to reaction 2 in Scheme 1, leading to Intermediate Product 2 (IP2), with k_c as the rate constant.

Scheme 1. Reactions of TEOS with Water (Reaction 1) and CDots (Reaction 2)^a



^{*a*}Reactions 1 and 2 are the corresponding rate expressions, and k_w and k_c are the respective rate constants of the two reactions.

Therefore, the water content in the reaction mixture should play a key role in determining the degrees of reaction of reactions 1 and 2. In the presence of an excess of water, the formation of IP1 is much faster than for IP2, leading to the conversion of TEOS into pure silica. Reducing the water content in the reaction mixture may help to suppress the hydrolysis of TEOS, and favor the reaction between TEOS and CDots.

To realize this scenario, water was not added into the reaction system but CDots were directly dissolved in ethanol, with the maximum possible concentration measured as 0.8 mg/ mL. The synthesis of CDots@silica composites has been conducted by the stepwise addition of TEOS and ammonia solution into the CDots ethanol solution. We found out that increasing the concentration of CDots in ethanol led to the formation of more products (see Figure 1), where the weight ratio of these products is 1/10/22/45/78, corresponding to the increase of CDots concentration in the precursor solution from 0 to 0.05 mg/mL, to 0.1 mg/mL, to 0.2 mg/mL, to 0.4 mg/mL. This result also indicates that simply mixing TEOS and ammonia-water in ethanol only resulted in a small amount of pure silica (Figure 1), since the amount of water in the ammonia solution is small, leading to decreases in the rate of reaction 1 in Scheme 1. In contrast, the reaction between the hydroxyl groups of CDots and TEOS (reaction 2 in Scheme 1) became dominant, resulting in the formation of CDots@silica composites. It is worth noting that the hydrolysis did not happen without adding the ammonia solution for any longer stirring times, which demonstrates that water in the ammonia solution molecules is crucial. Meanwhile, the reality that more CDots could result in more CDots@silica composites further demonstrates that the reactions shown in Scheme 1 are reliable.⁶¹ The above observation is a strong hint that CDots do not simply act as a dopant, but rather as the gelating agent for the formation of silica in form of the CDots@silica composites.

Scanning electron microscopy (SEM) images of CDots@ silica composites reveal quasi-spherical nanoparticles with narrow size distributions (see Figures 2a and 2b). As the CDots concentration in the precursor solution increased from 0.05 mg/mL to 0.1 mg/mL, to 0.2 mg/mL, to 0.4 mg/mL, the diameters of the nanoparticles increased from 68 ± 8 nm to 78 ± 8 nm, to 90 ± 10 nm, to 128 ± 20 nm, respectively. More



Figure 1. Change in the appearance of ethanol solutions containing CDots, TEOS, and ammonia–water during time periods up to 10 h (0 h (top), 5 h (middle), and 10 h (bottom)). The concentration of CDots in the solution varies from 0 mg/mL, to 0.05 mg/mL, to 0.1 mg/mL, to 0.2 mg/mL, and to 0.4 mg/mL, as indicated in the top photograph.

CDots present as gelating centers in the precursor solution provide more hydroxyl groups for the hydrolysis of TEOS, thereby leading to faster growth and larger particle sizes. Energy-dispersive spectroscopy (EDS) confirmed the existence of C and N in the composite nanoparticles, both increasing with increased CDots concentrations (see Figure 2c, as well as Table S1 in the Supporting Information). We further note that mixing TEOS and ammonia-water in absolute ethanol without the presence of CDots results in a formation of nonuniform silica nanoparticles (see Figures S5a and S5b in the Supporting Information), which highlights the role of CDots as nuclei to form CDots@silica composites. Transmission electron microscopy (TEM) imaging further demonstrates the spherical morphology of CDots@silica composite nanoparticles (see Figure 3a); high-resolution transmission electron microscopy (HRTEM) image allows us to visualize a single CDot with a lattice spacing of 0.20 nm (Figure 3b), consisting of the (100) crystallographic facet of graphitic carbon.58 The HRTEM imaging also shows that the silica matrix surrounding CDots is amorphous, which is further confirmed by the powder XRD pattern (Figure 3c).

As a proof of the successful inclusion of CDots, CDots@ silica composite nanoparticles possess PL emission properties of the encapsulated CDots, namely their distinct excitationdependent PL behavior. The fluorescent colors change from turquoise to green to red, when changing the excitation from UV to blue to green (Figures 3d-f). Under 375 nm excitation, the PL peak of the CDots@silica composite nanoparticles obtained from 0.4 mg/mL CDots ethanol solution is located at 520 nm (Figure S6 in the Supporting Information). Its PLQY is measured to be 31% at 405 nm excitation, while the CDots@ silica composite nanoparticles synthesized from 0.05 mg/mL CDots ethanol solution show a higher PLQY of 52% (Table S1



Figure 2. Representative scanning electron microscopy (SEM) images ((a1) CDots concentration = 0.05 mg/mL, (a2) CDots concentration = 0.1 mg/mL, (a3) CDots concentration = 0.2 mg/mL, and (a4) CDots concentration = 0.4 mg/mL; scale bar = 500 nm for each graph), corresponding size histograms (b1–b4; obtained from more than 200 particles for each graph), and corresponding EDS patterns (c1–c4) of CDots@silica composite nanoparticles obtained when varying the concentrations of CDots in the reaction system from 0.05 mg/mL, to 0.1 mg/mL, to 0.2 mg/mL, to 0.4 mg/mL.

in the Supporting Information). At the same time, Table S1 shows that the PLQY still decreases from 52% to 31% when the CDots loading fraction increases from 2.1 wt % to 8.4 wt %, synthesized from CDots concentrations from 0.05 mg/mL to 0.4 mg/mL, respectively. As discussed above, the reduction in PLQY is mainly due to the collision and, thereby, aggregation of CDots during the fabrication process. We note that upon increasing the CDots concentration in a precursor solution to 0.4 mg/mL, an appearance of the samples in SEM images changed from isolated nanoparticles (panels (a1)-(a3) in Figure 2) to the cross-linked structures (panel (a4) in Figure 2), the effect of which has become even more pronounced when the CDots concentration further increased to 0.5 mg/mL (see Figure S7 in the Supporting Information). This observation inspired us to further increase the CDots concentration to eventually achieve a more perfect cross-linked architecture, where the movement of CDots during the

fabrication process could become sufficiently restrained, thus avoiding their aggregation. Indeed, upon an increase of CDots concentration to 0.8 mg/mL (which is the maximum possible concentration of CDots in ethanol), the solution rapidly became viscous, and after an aging for 6 h, a CDots@silica composite gel with a strong green luminescence was formed (Figure 4a). After freeze-drying the gel, a green-emitting CDots@silica xerogel was obtained (see Figures 4b and 4c), and SEM and TEM images revealed its cross-linked, intercalated 3D architecture (Figures 4d and 4e). The diffuse reflectance spectrum of CDots@silica xerogel has a similar absorption band, compared with that of CDots, and the high absorbance of the xerogel ranging from 380 nm to 460 nm suggests the possibility of obtaining a strong PL emission (Figure S8 in the Supporting Information).

The PLQY of the CDots@silica composite xerogel has been measured to be 41% under 405 nm excitation, with a high



Figure 3. (a) TEM image, (b) HRTEM image, and (c) XRD pattern of CDots@silica composite nanoparticles synthesized in 0.2 mg/mL CDots ethanol solution. (d-f) The fluorescence images were observed via fluorescence microscopy under UV excitation (panel (d)), blue excitation (panel (e)), and green excitation (panel (f)), respectively. The exposure time was 100 ms (panel (d)), 150 ms (panel (e)), and 2000 ms (panel (f)). Scale bars in panels (d)–(f) are equal to 10 μ m.



Figure 4. Photographs of the CDots@silica composite gel taken under sunlight (panels (a1) and (a3)) and UV light (panels (a2) and (a4)). After freeze-drying the gel, CDots@silica composite xerogel is obtained, with the PL emission spectrum (panel (b)) taken at 375 nm excitation and a photograph taken under UV light in the inset (panel (c)). An SEM image (panel (d)) and a TEM image (panel (e)) are also shown.

CDots loading fraction of 19.2 wt % (Table S1). We ascribe simultaneous realization of both high PLQYs and high CDots loading fraction in the xerogel to the restrained movement of CDots, which effectively avoid their aggregation in the resulting composites. Figures 5a and 5b show variations of PLQYs and PL intensity of the CDots@silica composites versus the concentration of CDots in the precursor ethanol solution. The PL intensity data have been collected on the fluorescence microscope equipped with a spectrometer (see Figure 5b, as well as Figure S9 in the Supporting Information). For the CDots concentration lower than 0.4 mg/mL, the PLQYs of the



Figure 5. Variations of (a) PLQY (open black squares), (b) PL intensity (open red circles), and (c) PL lifetime (open blue triangles) of the CDots@silica composites versus the concentration of CDots ethanol solution. Solid lines are solely provided as a guide to the eye.

resulting composites gradually decrease upon increasing the CDots concentration, which is mainly due to the collision and aggregation of CDots. When the CDots concentration is further enhanced above 0.4 mg/mL, cross-linked architectures start to develop, which restrains the aggregation of CDots and is favorable for increasing their PLQYs.

We further conducted time-correlated single-photon counting measurements (405 nm excitation) on the composite samples (see Figure 5c, as well as Figures S10 and S11 in the Supporting Information). The fluorescence decay curves of CDots ethanol solutions with different concentrations (recorded at 520 nm), which were used to prepare composite nanoparticle and xerogel, are almost identical (Figure S10), indicating that no aggregation of CDots in solutions occurs in this concentration range. At the same time, average PL lifetimes of the CDots@silica nanoparticles are gradually shortened from 11.6 ns to 10.4 ns, to 9.8 ns, to 8.8 ns, when the CDots loading fraction increases from 2.1 wt % to 3.8 wt %, to 5.4 wt %, to 8.4 wt %, respectively (see Table S1 in the Supporting Information, Figure 5c, and Figure S11 in the Supporting Information). It is worth mentioning that two lifetime components, which are related to different recombination processes in CDots@silica composites,¹⁰ exhibit similar trends with variations of the CDots loading fraction (Table S1). These results are in accord with the increased energy losses in the composites due to the aggregation of CDots when the CDots loading fraction increases, leading to the reduction of PLQYs from 52% to 40%, to 35%, to 31%. However, the average PL lifetime of CDots@silica composite xerogel is extended to 10.3 ns (Table S1 and Figure 5b), although the CDots loading fraction is up to 19.2 wt %, which demonstrates that the aggregation of CDots could be prevented in the gelation process.

According to the above analysis, Scheme 2 is proposed to illustrate the differences in the formation of CDots@silica composite nanoparticle and xerogel. At low CDots concentration, the amount of IP2 is rather low, while water from

Scheme 2. Schematics of the Formation of CDots@Silica Composite Nanoparticles/Gel at (a) Low, (b) Medium, and (c) High Concentration of CDots Ethanol Solutions



ammonia aqueous solution continues to react (albeit slower) with TEOS to form quasi-spherical silica nanoparticles, which gradually encapsulate IP2. As the CDots concentration increases, the amount of IP2 is increased, and for the concentration of CDots reaching 0.8 mg/mL, this happens fast, resulting in an increase of viscosity of the gelating solution. In this case, the formation of silica and IP2 occurs in parallel, leading to an intercalated three-dimensional (3D) architecture of the resulting composites.

The CDots@silica composite xerogel possesses a superior resistance toward UV light irradiation, which is a useful prerequisite for lighting applications. Its photostability was investigated by comparing the PL intensity of CDots, the CDots@silica composite xerogel, and commercial fluorescein sodium dye solution under the same conditions of the continuous irradiation by UV light (1.6 W/cm²). As seen from Figure S12 in the Supporting Information, PL of the dye is almost completely quenched after 65 min of irradiation, while the PL intensity of CDots is preserved by more than 80%, indicating CDots' high resistance to UV light irradiation, which is consistent with the previously reported results.⁵⁶ The CDots@silica composite xerogel exhibit improved photostability, because of the protection by silica. The PL intensity of the xerogel is preserved by more than 90% after 65 min of irradiation.

Besides green emissive CDots, blue emissive CDots synthesized from citric acid and ammonia–water are also utilized to prepare the CDots@silica composites, which is equally facilitated by the presence of abundant hydroxyl groups on the surface.⁵⁵ Based on the above strategy, blue emissive CDots@silica composite gel could be fabricated as well, resulting in a strongly blue luminescent composite xerogel (Figure S13 in the Supporting Information). Mixing the green or the blue emissive CDots@silica xerogels with polydimethylsiloxane (PDMS) and solidifying the mixtures at 80 °C for 1 h allows us to obtain strongly luminescent solid-state materials that are easy to cast in different shapes (Figure 6a),



Figure 6. (a) Photograph of the xerogel-PDMS composites excited by an ultraviolet lamp. (b) Emission spectrum of a down-conversion white LED prototype entirely based on a powdered CDots@silica composite xerogel with a photograph of the working device (inset, panel (c)).

which can be employed as color converters in down-conversion LEDs. Because of the concern about the UV radiation of UV LED chips always needing to be considered, blue-emitting InGaN LED chips are usually used as an excitation source with emission at 450 nm. Because of the distinct excitationdependent PL behavior of CDots, the PL emission of the CDots@silica composite xerogel shows an obvious red-shift when changing the excitation wavelength from 375 nm to 532 nm (Figure S14 in the Supporting Information). Under 450 nm excitation, the CDots@silica composite xerogel emits a greenyellowish light, with a PL peak centered at 543 nm and a PLQY of 36% (see Figure S14). Apart from the excitation-dependent PL emission of CDots, increasing the contents of the CDots@ silica composite xerogel in the color conversion layer results in a further red-shift of the emission peak, because of the progressive reabsorbance and re-emission effect, which has been considered in detail in previous publications $^{41,42,62}\ A$ mixture of powdered CDots@silica composite xerogel (as shown in Figure S14) and PDMS has been deposited on an InGaN LED chip, and cured at 80 °C for 1 h to produce an

LED prototype with emission maxima centered at 450 and 558 nm (Figure 6b), and an emission tail extending to longer wavelength over the entire visible spectrum. As shown in Figure 6c, such LEDs emit a white light, with the following parameter values: Commission Internationale de L'Eclairage (CIE) coordinate, (0.33, 0.34); color temperature, 5603 K; color rendering index, 79; and luminous efficacy of radiation, 28 lm/W.

CONCLUSIONS

In summary, we fabricated CDots@silica composite gels (where CDots represents carbon dots) via a reaction between tetraethyl orthosilicate (TEOS) and the surface hydroxyl groups of CDots, instead of the classic Stöber approach. During the gelation process, collision and aggregation among CDots is prevented. The resulting CDots@silica composite xerogel not only possesses high photoluminescence quantum yields (PLQYs) exceeding 40%, but also high CDots loading fraction (19.2 wt %). The as-prepared solid-state xerogel exhibits excellent photostability, which permits us to fabricate white LED prototypes. This work demonstrates the design and fabrication of strongly emitting CDots-based phosphors by conquering the aggregation-induced solid-state luminescence quenching of CDots, and further highlights their applicability in solid-state lighting.

ASSOCIATED CONTENT

S Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.chemma-ter.6b05375.

An attempt to prepare CDots@silica composites using the classical Stöber approach, PL spectrum of the CDots@silica composite nanoparticles synthesized in 0.2 mg/mL CDots ethanol solution, time-resolved PL decay curves, and blue-emitting CDots@silica composite gel (PDF)

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Notes

The authors declare no competing financial interest.

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