Carbon Dots: Zero-Dimensional Carbon Allotrope with Unique Photoinduced Redox Characteristics

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ABSTRACT: Carbon dots (CDots) exploit and enhance the intrinsic properties of small carbon nanoparticles. Their optical absorptions and photoinduced redox characteristics are competitive with those of conventional semiconductor quantum dots at one end and fullerenes and other carbon nanomaterials at the other. Highlighted in this mini review are the effective photon harvesting over a broad spectral range by CDots and their subsequent excited-state charge transfer processes and interactions, which have enabled their use as sensors, for photodynamic effects, and in various energy conversion technologies.

INTRODUCTION

Carbon dots (CDots), 1 generally defined as small carbon nanoparticles with various surface passivation schemes (Figure 1), exploit and enhance the intrinsic properties of the carbon nanoparticles. Among various nanoscale carbon allotropes, fullerenes have naturally taken the title of carbon nanomaterials at the zero dimension (Figure 1). However, one may argue that fullerenes are, in fact, stoichiometrically defined molecules of not only unique molecular architectures but also distinct electronic structures and properties, fundamentally different from carbon nanomaterials in other dimensions including carbon nanotubes and graphene nanosheets, which obviously cannot be considered as molecules in any stretch of the imagination. On the other hand, small carbon nanoparticles share a key structural feature with carbon nanotubes and graphene nanosheets, namely, the substantially present structural and edge defects (Figure 1),‡ which are clearly not possible in fullerenes. Thus, the nanoparticles should really deserve the title of nanoscale carbon at the zero dimension (Figure 1).

Defects in small carbon nanoparticles and their one- and two-dimensional counterparts play a major role in the optical and redox properties of these carbon nanomaterials. Upon the effective passivation of the defects in small carbon nanoparticles for CDots, these properties are greatly enhanced, as found and established experimentally.¹,³ Among the more pronounced are the observed bright and colorful fluorescence emissions of CDots, which have been investigated extensively for both fundamental understanding and technological applications. Equally interesting and valuable are the unique photoinduced redox characteristics of CDots as both excellent electron donors and acceptors versus fullerenes as potent electron acceptors only. Highlighted in this mini review are the effective photon harvesting over a broad spectral range by CDots and their subsequent excited-state charge transfer processes and interactions, which have enabled their use as sensors, for photodynamic effects, and in various energy conversion technologies.

CDot Syntheses. CDots exploit and enhance the unique properties of small carbon nanoparticles, which as discussed above represent a distinct nanoscale carbon allotrope at the zero dimension (Figure 1). Therefore, in both the structure and composition of CDots, there must be the dominance of nanoscale carbon particles/domains. To ensure such dominance, the classical approach is to use pre-existing carbon nanoparticles, specifically processed and selected, for the preparation of CDots, as in the original finding.¹ The carbon nanoparticles can be sourced from various carbon soot samples produced by laser ablation, arc discharge, and other methods, including those available commercially that are marketed as “carbon nanopowders”. For the required effective surface passivation in CDots, the carbon nanoparticles are functionalized by organic molecules in chemical and thermally induced reactions, including mostly the amidation of carboxylic acid moieties on the nanoparticle surface in the former¹,³ and likely radical additions in the latter.⁴

Among other syntheses in which the dominance of nanoscale carbon particles/domains is evident are those based on the electrochemical etching of graphite,⁷ the pyrolysis of organic materials,⁶ and other high-temperature carbon-
Absorption and Emission Properties. CDots were originally found and developed for their bright and colorful fluorescence emissions that resemble those of conventional semiconductor quantum dots (QDs). For photoexcitation, the optical absorptions of CDots are dictated by electronic transitions in the core carbon nanoparticles (Figure 1), which are known to be associated with π-plasmons. The core carbon nanoparticles may be largely amorphous (for those harvested from some commercially supplied “nanopowder” samples, for example) or more graphitic (from samples produced by laser ablation under some conditions), including those small few-layer graphene pieces often referred to as “graphene quantum dots” in the literature. The absorptions are strong, covering the broad UV to visible spectral region and extending into the near-IR, capable of harvesting up to 65% or more of the solar radiation over 300–800 nm. More quantitatively, optical absorptivities of CDots in terms of per molar concentration of carbon atoms ($M_{C\text{ atom}}$) in the core carbon nanoparticles are $50–100 \times M_{C\text{ atom}}^{-1}$ cm$^{-1}$ at 400–450 nm, compared with 16 $M_{C\text{ atom}}^{-1}$ cm$^{-1}$ for C$_{60}$ at its first absorption band maximum. CDots are also known for being among the very best two-photon absorbers in the near-IR.

The bright fluorescence emissions of CDots are excitation-wavelength-dependent in a rather characteristic pattern for both observed spectra and quantum yields (Figure 2), and the pattern is consistent among CDots of different surface functionalities and from different preparations as long as the dominance of nanoscale carbon particles/domains in the dot structures is ensured. Mechanistically, the photoexcited CDots undergo rapid charge transfers for separated electrons and holes, which are trapped at various surface sites stabilized by the surface passivation (Figure 3). The radiative recombinations of the electrons and holes are responsible for the fluorescence emissions, with the yields of the recombinations for the formation of emissive excited states ($\Phi_2$) reflected in the observed fluorescence quantum yields, $\Phi_1 = \Phi_2 \Phi_3$, where $\Phi_2$ represents emission yields from the emissive excited states ($\Phi_3$).

Photoinduced Redox Characteristics. Carbon powdery materials are typically used in reductive processes, such as the popular Pd/C catalyst. However, photoexcited CDots are both excellent electron donors and acceptors, as clearly demonstrated in the results of fluorescence quenching by electron donor and acceptor molecules as quenchers. For both fluorescence intensities ($\Phi_3$) and lifetimes ($\tau_p$, representing the averages in multieponential decays), the quenching is highly efficient, in general, which has been attributed in part to
the quenching radius beyond the edge of an individual dot for reduced diffusion distance. Also commonly observed has been the decoupling of the $\Phi_F$ quenching from the $\langle \tau_F \rangle$ quenching, with the former being much more efficient and seemingly beyond the diffusion-controlled limit. In the mechanistic framework for CDots (Figure 3), there are roughly and effectively two processes on different time scales, with yields of $\Phi_1$ and $\Phi_2$.13 The process of $\Phi_1$ is orders of magnitude faster than that of $\Phi_2$, as reflected experimentally by the fluorescence rising (picoseconds) and decay (nanoseconds) times. The $\langle \tau_F \rangle$ quenching is mostly dynamic in nature, affecting only the $\Phi_2$ process, namely, decays of the emissive excited states (Figure 3). On the other hand, the quenching of $\Phi_F$ ($\Phi_1 \Phi_2$) has a significant static component due to higher than bulk near-neighbor concentrations of quencher molecules, capable of affecting the $\Phi_1$ process via partially interrupting the radiative recombinations (Figure 3).13 Similar decoupling between $\Phi_F$ and $\langle \tau_F \rangle$ has also been found in other responses of CDots to external effects beyond quenching, such as the enhancement in fluorescence emissions when CDots are confined in a more restrictive environment like in polymer films. The mechanistic account of the decoupling is important to the understanding of many reported results on the sensing of metal ions with CDots based on fluorescence quenching (see below).

A more vivid display of such decoupling is in the case of fluorescence quenching due to the surface doping of CDots with a small amount of noble metal like gold.14 The metal has a high electron affinity and also right on the dot surface, capable of soaking up the trapped electrons to interrupt the radiative recombinations (Figure 3) in a purely static fashion. Such static quenching, also referred to as “dark quenching process” in photophysics textbooks for its not visible in fluorescence decays, has no effects on $\langle \tau_F \rangle$, as observed experimentally. Consistent with such a mechanistic picture is that the noble-metal-doped CDots are excellent photocatalysts for some of the most difficult energy conversion reactions, such as the photocatalytic reduction of CO$_2$ into small organic molecules.14,15

Relevant Applications. For technological applications, CDots have been explored for their properties that are either uniquely advantageous or competitive to those of fullerenes at one end and conventional semiconductor QDs at the other. Highlighted below are some representative relevant applications that take advantage of the photoinduced redox characteristics of CDots.

Sensing. The highly efficient charge transfer quenching of fluorescence emissions has made CDots excellent sensors for both electron-deficient and electron-rich substances. However, more popular have been their uses in the sensing of electron-deficient analytes, such as nitrotoluenes, as signature compounds for TNT and related explosives. For example, EDA-CDots [EDA = 2,2′-(ethylenedioxy)bis(ethylamine)] were explored for the detection of 2,4-dinitrotoluene (DNT) with the highly efficient fluorescence quenching both in intensities ($\Phi_F$) and lifetimes ($\langle \tau_F \rangle$).13 The quenching of fluorescence intensities was apparently beyond the diffusion control, which was attributed to static quenching contributions associated with higher local DNT concentrations around the

Figure 2. Absorption spectrum (ABS) and fluorescence (FLSC) spectra (inset, from the left corresponding to excitation wavelengths of 400 nm in 20 nm increments) and quantum yields at different excitation wavelengths for EDA-CDots.

Figure 3. Cartoon illustration (top) and state diagram (bottom) on the mechanistic framework for CDots.
CDots and the quenching radius larger than the average radius of individual dots. The \(r_{\text{eff}}\) quenching was dynamic and still very efficient at the upper limit of diffusion control. Thus, the former is more useful to the need for high sensitivity, pushing the detection limit lower, whereas the latter is valuable to the quantification of the analyte concentrations above the detection limit.

The sensing of various metal cations in many reported studies might be more affected or dominated by the substantial or extreme static quenching of fluorescence intensities, with the apparent Stern–Volmer quenching constants as large as \(3.0 \times 10^{7}\). The overwhelming majority of these studies were based on dot samples prepared in one-pot carbonization syntheses under relatively mild processing conditions, which were nanoscale carbon domains mixed with abundant organic species. As a result, such samples must be porous or even “spindle-like”, capable of soaking up and/or adsorbing the metal cations to have much higher local concentrations for the substantial or extreme static quenching, responsible for the observed low detection limits. However, a downside of the dominating static quenching would be the vulnerability to minor changes in the sample structure and/or morphology in standard versus analyte solutions, which could have major effects on the analyte quantification based on standard curves.

Among some investigations in which the dot samples were similar or close to those obtained from pre-existing carbon nanoparticles, Chang and co-workers used the dot sample from carbonizing l-cysteine hydrothermally at 300 \(^\circ\)C for 2 h to detect various metal cations based on the quenching of fluorescence intensities. For \(\text{Cu}^{2+}\) in water and vitamin B\(_12\) samples, as an example, the detection limit was reported to be as low as 10 nM. Huang and co-workers used microwave-assisted carbonization of solid-state organic mixture to prepare dot samples for the detection of \(\text{Cr}(VI)\) along with various other metal cations in water. It was found that \(\text{Zn}^{2+}\) could actually enhance the fluorescence emissions somewhat, in contrast to the quenching by \(\text{Ag}^{+}\), \(\text{Cu}^{2+}\), \(\text{Fe}^{3+}\), and especially \(\text{Cr}(VI)\). For \(\text{Cr}(VI)\), the reported detection limit was as low as 120 nM. According to the comparison between the effects of \(\text{Cr}(VI)\) on fluorescence intensities and decays, the quenching was apparently all static. Interestingly, therefore, even for these dot samples of likely more significant nanoscale carbon contents, there was still the dominance of static quenching associated with high local concentrations around the emissive entities in the samples.

**Photocatalysis.** The effective photon harvesting by CDots in the visible spectrum and beyond and the photoexcited CDots serving as both excellent electron donors and acceptors have prompted their many uses in photocatalytic oxidation and reduction reactions. Mechanistically, the photoexcitation of CDots results in the rapid charge transfers and separation for the trapped electrons and holes (Figure 3), which could be exploited for catalytic activities directly, such as the photocatalytic reduction of \(\text{CO}_2\) and degradation of organics. The former represents one of the most challenging yet extremely valuable photocatalytic conversions. It was shown that the electrons could be concentrated by doping the surface of CDots with noble metals to enhance substantially the photocatalytic performance for much higher \(\text{CO}_2\) conversion quantum yields. A surprising finding was the dependence of the conversion quantum yields on \(\text{CO}_2\) concentrations. In fact, for the use of gold-doped CDots as catalysts in liquid \(\text{CO}_2\) with visible light irradiation (405−720 nm), the quantum yields estimated on the production of formic acid alone could reach 3−5%, thus among the best ever achieved with conventional semiconductor and/or other photocatalysts for the \(\text{CO}_2\) reduction. As known in the literature, the best-performing semiconductor photocatalysts for \(\text{CO}_2\) conversion were mixtures of several semiconductors of different band gaps, with the implied need mechanistically for some kind of coordinated actions of multiple catalysts. In this regard, the observed high performance of CDots may be rationalized as being due to their broadly distributed electronic transitions and charge transferred states in individual dots, which are analogous to the catalyst configuration of combining multiple semiconductors in a single nanostructure.

CDots and derived photocatalysts have been popular in water splitting for hydrogen molecules. In fact, the photocatalytic reduction of \(\text{CO}_2\) in aqueous media could mechanistically go through the route of hydrogen generation first, which then reduces \(\text{CO}_2\) into small organic molecules. Interestingly, among the reported studies on water splitting with the use of CDots as photocatalysts, it seems that the outcomes are significantly dependent on the dot samples used. For the CDot-based photocatalysts found to be highly effective in the \(\text{CO}_2\) conversion, as discussed above, they were also capable of the photocatalytic hydrogen generation from water under visible light but with a performance significantly poorer than that in the \(\text{CO}_2\) reduction.

The dot structures of carbon composites or combined with conventional semiconductors have been explored for enhanced photocatalytic functions. In these catalyst configurations, the nanoscale carbon domains serve as photosensizers for the semiconductors, in addition to their own catalytic activities in some cases. For example, various carbon−\(\text{TiO}_2\) composite dots have been prepared as photocatalysts for the \(\text{CO}_2\) conversion, but there have been no major breakthroughs in terms of enhanced catalyst performance for much improved photo-reduction outcomes. Similarly, the composite dot structures of carbon with \(\text{TiO}_2\) were evaluated in photocatalytic oxidation reactions, such as the popular uses in photocatalytic degradation of organic dyes.

For example, Yu et al. prepared the carbon−\(\text{TiO}_2\) nanosheet composites as photocatalysts to degrade rhodamine B under visible light. The degradation experiments were relatively straightforward, in which the photocatalysts in an aqueous rhodamine B water solution were irradiated by using a 500 W xenon lamp. The decreasing concentration of rhodamine B with the photoirradiation was determined every 20 min in optical absorption measurements. The results led to the conclusion that the photocatalysts were effective for the dye degradation purpose.

**Optoelectronics.** Conventional semiconductor QDs have been widely used in optoelectronic devices, with famous examples including “QLED” television sets and “quantum dot displays” in general. They are also popular in various photovoltaic devices, so are fullerene, despite the generally poor photon-harvesting capabilities of the widely used \(\text{C}_60\) derivatives like PCBM (([6,6]-phenyl \(\text{C}_61\) butyric acid methyl ester). CDots resemble QDs in optical properties and photoinduced redox characteristics, including the sharing of some mechanistic features, and they also compete favorably with fullerene. Thus, CDots have been explored for their uses in various components of optoelectronic devices.
Rogach and co-workers prepared two kinds of dot samples for light-emitting diodes, one emitting white light and the other emitting blue light with different applied voltages. The dot syntheses were based on the thermal carbonization of the mixture containing octadecene, 1-hexadecylamine, and citric acid with somewhat more aggressive processing conditions of 300 °C for 3 h. The fluorescent dots were used in the emissive layer of the devices (Figure 4). The maximum brightness was 24 cd/m² for the blue LED and 96 cd/m² for the white LED. The devices had a low turn-on voltage of 5 V. The white LED could emit light in different colors under different voltages (Figure 4).

Yan et al. used a CVD setup (acetylene as the feed at 1000 °C) to prepare dot samples for the electron transporting layer (ETL) in polymer solar cells (PSCs). The purpose was to overcome the limitations of the commonly used ETL materials such as LiF and Ca for the higher thermal stability, enhanced electron injection, and lower interface resistance in the solar cells, as achieved in the study. Similarly, Cui et al. used the CVD-produced dot samples to prepare composite films with poly(3-hexylthiophene) (P3HT). As made evident by the quenching of fluorescence emissions, there were electron transfer interactions in the composite films with the dots as electron acceptors. The electron acceptor characteristics of the dots were exploited for their combination with or replacing the C_60 derivative PCBM in the solar cells, and both were found to be able to enhance the cell performance. It was concluded that CDots could be used as an inexpensive alternative to C_60 derivatives for the electron acceptor function in polymer solar cells.

Antimicrobial Function. CDots have been shown for their visible-light-activated microbicidal functions, including the observed inhibition of bacteria like *Escherichia coli* under ambient room light, which serves as an example for effectiveness. The antimicrobial actions of CDots are similar to those of the widely used TiO₂ nanoparticles (except for the required UV excitation of TiO₂), involving the photoinduced generation of reactive oxygen species (ROS). Mechanistically, the ROS generation could be associated with the initial charge-separated species following the photoexcitation of CDots and/or the emissive excited states from radiative recombinations (Figure 3). The available experimental results seem to suggest contributions by both.

Similar to the photocatalysts based on dot structures in which nanoscale carbon domains are composited or combined with conventional semiconductors, as discussed above, the carbon-based/derived hybrid nanostructures could be explored for enhanced antimicrobial activities. In such nanostructures, the carbon domains again serve the function of effectively harvesting visible photons to sensitize the nanoscale semiconductors like TiO₂ in the hybrids, in addition to their own ROS generation and related antimicrobial actions. CDots are intrinsically nontoxic, and they can be produced from abundant and often renewable precursor materials, amenable to microbial control in food/water safety and related applications that require the microbicidal agents to be benign and also low or ultralow costs.

Relationships to Carbon Nanomaterials in Other Dimensions. The optical properties and photoinduced redox characteristics of CDots are associated with the passivated defects in the core carbon nanoparticles or nanoscale carbon domains in the dots. Similar structural and edge defects are present in carbon nanotubes and graphene nanosheets (Figure 1), and their passivation via chemical functionalization or in other schemes could result in largely the same bright and colorful fluorescence emissions and excited-state charge transfers. In fact, the observation of bright green fluorescence from functionalized carbon nanotubes preceded the finding of CDots. In all of the carbon nanomaterials in different dimensions (except for fullerene molecules, Figure 1), the unavoidable defects apparently play a critical role or even “it is all about defects” in terms of the photoexcited state properties and processes in these nanomaterials. In this regard, each passivated defect area in the carbon nanostructures may be considered phenomenologically equivalent to a CDot (Figure 5). Such a structural view may help the fundamental understanding and technological exploitation of the nanoscale carbon allotropes.

**SUMMARY AND PERSPECTIVES**

CDots have emerged to represent a rapidly advancing and expanding research field, with their properties and potential applications competing broadly and in most cases effectively with those of conventional semiconductor QDs and other carbon nanomaterials. In the further development of this new and transformative carbon nanomaterial platform, the relevant
research community must have the courage and determination to address the systemic issue of the incorrectly or poorly prepared dot samples that are contaminated substantially or nearly completely by dye-like species/mixtures. In this regard, the dominant contents of carbon nanoparticles or nanoscale carbon domains must be ensured in all dot samples, as required in the definition of CDots. Nevertheless, with properly prepared high-quality dot samples, their highly competitive and/or novel uses across a number of important technological areas may be envisaged.

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