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Synthesis of Tunable Fluorescent Carbon Dots: A Mini Review

Varsha UshaVipinachandran, Sathish Rajendran, Indhumathi Ashokan, Kabir Hussain Badagoppam Haroon and Susanta Kumar Bhunia*

Department of Chemistry, School of Advanced Sciences, Vellore Institute of Technology, Vellore, 632014, India
Email: susanta.bhunia@vit.ac.in; susanta.chem@gmail.com

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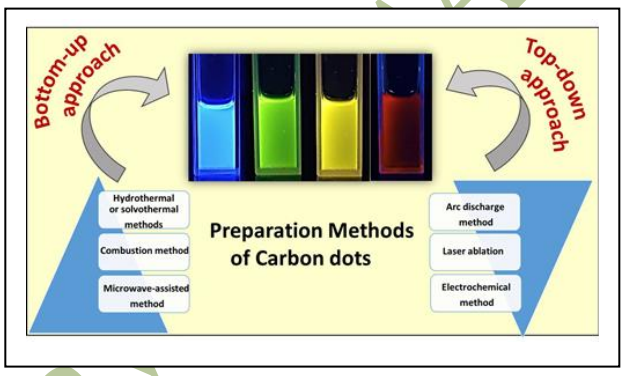
Abstract

Among carbon nanomaterials family, small sized carbon dots (CDs) have received considerable research interest because of their interesting properties including tunable fluorescence, strong emission, easy surface modification, wider availability of carbon precursors, chemical stability, low photobleaching, etc. Moreover these dots are potential competitor compared to conventionally used semiconductor quantum dots. These remarkable properties allow the CDs to use in sensing, bioimaging, drug delivery, catalytic, and light emitting diode applications. Therefore the synthesis of tunable fluorescent CDs remains a crucial factor and challenging issue. In this minireview, we have explained two synthetic methods namely top-down and bottom-up approaches for synthesis of CDs. Different techniques under these two approaches have been well explained.



Keywords: Carbon dots; tunable fluorescence; synthetic approaches; top-down approach; bottom-up approach

1. Introduction

The zero-dimensional carbon dots (CDs) have paved their own milestone in various fields like chemo- and biosensing, cellular imaging, theranostics and photonics due to its unique optical, electrical and surface properties.[1] The CDs are typical luminescent with nano spherical shaped structure and mainly consist of carbon as major element. As these dots exhibit spontaneous luminescence property with excellent biocompatibility, photo stability and non-toxicity, it involves as inevitable participant in cell imaging as well as in sensing.[2, 3, 4, 5] The CDs have the potentiality in biomedical field for cancer cell imaging with active drug delivery. Moreover, the inclusion of different surface functional moieties on the CDs surface plays the selective receptor-acceptor activity to execute the chemo-gene and antibiotic therapy. The green synthetic methods imply their safety and eco-friendly nature. Besides low-photobleaching power and light to heat generation efficiency help CDs for cancer cells treatment.[6,7] Quasi spherical nanodots has facilitated with quantum confinement, as they are size manipulated under 10 nm to mold the properties of CDs.[8,9] CDs consist of sp^2 hybridized graphitic core and these cores are decorated by different functional groups that help to tune the fluorescent property through surface modification and fluorescence.[10,11] Carbonization temperature plays a major role to tune the fluorescent property (temperature-dependent fluorescent).[12] Although band gap tunes fluorescence property based on size manipulation to capture the excited light and resulting in alteration of fluorescence properties. Strong fluorescence emission after the absorption of particular energy made them to use as the sensitizers in solar cell. That means CDs are able to absorb the excited light to impart the electron transference. The property of electronic transition depends on nature of emission property. Blue emissive CDs have higher energy band gap compared to red emissive CDs that results in different electronic transition properties. Thus, the synthesis of fluorescent CDs from carbonaceous precursors is a considerable subject. The synthetic methods are broadly classified under the category of top-down and bottom-up approaches. One of the fascinating peculiarities of CDs is that they can be synthesized through economic as well as environment friendly and green synthetic methods. Surface passivation can play a vital role for fluorescence improvement. Moreover, the emission color can be tuned by doping hetero elements such as oxygen, nitrogen, sulphur, phosphorous, etc. which can control the electronic transition. Emission spectra of different emissive CDs (blue to red) under different excitation wavelengths has been shown in Figure 1a that displays their respective maximum emission wavelengths. Fluorescence digital images of CDs have been captured under hand held UV lamp. Figure 1b represents the

different emissive CDs which correlate with their emission spectra. In this mini review, we have explained different synthetic strategies for the CDs preparation.

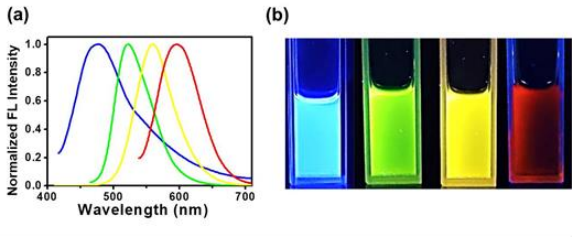


Figure 1. (a) Normalized emission spectra and (b) digital fluorescence images of different CDs.

2. Synthesis of CDs

Synthesis of CDs are classified into two categories namely bottom-up and top-down approach. The bottom-up approach concerns the synthesis of CDs from small molecules via hydrothermal/ solvothermal method, combustion method and microwave-assisted method. Whereas, top-down approach comprises mainly arc-discharge, laser ablation, and electrochemical oxidation processes that involve breaking down of bulk materials into small sized CDs. A schematic presentation has been shown in Figure 2.

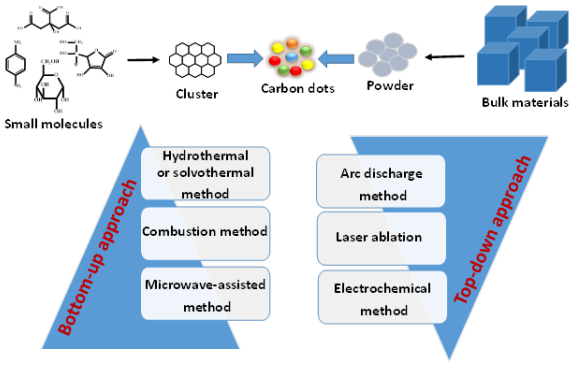


Figure 2. General synthetic approaches of CDs.

2.1. Bottom-up approach

As the name suggests, the synthesis procedure involves sizing up the molecular level precursors to nanometer level particles to exhibit exceptional optical, electrical and magnetic properties. The piling up of the molecules to nanometer size is not that much easy. It must requires a large amount of external energy sources like thermal, sound or microwave.[13] As the chemicals used are at the molecular level, it is more flexible to the endless tuning of CDs with the aforementioned properties. There are mainly four steps involved in the CDs formation during the synthetic time namely condensation, polymerization, carbonization, and passivation. The small molecules come closer and condense to form chain compounds as intermediate in the condensation process. The compounds become polymerized with the progression in time to form polymeric CDs in the second step. The functional groups attach on the polymeric CDs surface in this step to impart fluorescence property. In the carbonization technique, crystalline graphitic core with sp^2 hybridization are formed from polymeric CDs. The remaining precursor molecules act as surface passivation to enhance fluorescence property of the CDs.

2.1.1. Hydrothermal/ solvothermal method

The most common and cost-effective method for the synthesis of CDs is hydrothermal method. It involves the polymerization followed by carbonization of carbon precursors on assistance with appropriate solvent, in which the carbon precursors completely dissolves. Generally, the precursors and solvent mixture is fetched into a teflon-lined autoclave and heated in an oven at desired temperature for particular time duration. The temperature and carbonization duration been chose based on optimum fluorescent CDs generation. High amount of thermal energy needed for the carbonization as well as for passivation processes. Various types of fluorescent CDs can be synthesized from wide range of carbon precursors including glucose, sucrose, amino acids, citric acid, biowastes, etc. Hu et al. synthesized blue fluorescent CDs having fluorescence quantum yield (QY) of 22% from a mixture of citric acid and L-histidine using hydrothermal method.[14] Han et al. prepared blue fluorescent nitrogen doped CDs from biomass using highland barley as carbon precursor and ethylenediamine as nitrogen source for surface passivation.[15] Mercury ion (Hg^{2+}) has been selectively detected with this type of CDs. Nitrogen doped blue emissive CDs have been also prepared from cellulose and ethylenediamine mixture.[16] Ferric ion (Fe^{3+}) has been detected through fluorescence quenching technique. Hydrothermal technique has been used to prepare bright blue fluorescent CDs from mandelic acid and ethylenediamine mixture by Zhang et al.[17] The CDs showed QY of 41.4% and have been used for picric acid detection. A mixture of citric and glycine was used for blue fluorescent CDs preparation.[18] Khan et al. reported temperature assisted one-step synthesis of water soluble nitrogen and sulphur co-doped CDs with blue fluorescence from a mixture of L-lysine and thiourea.[19] Tan et al. synthesized blue emissive CDs with a fluorescence QY of 48.3% using citric acid and poly (ethylenimine) mixture.[20] Phenylenediamine isomers were separately heated in ethanol solution to prepare blue, green, and red emissive CDs with respective QY of 4.8%, 10.4%, and 20.6%.[21] The authors applied these CDs in bioimaging and flexible full-color emissive PVA films. One step solvothermal technique has been reported for red fluorescent CDs synthesis from 1,2,4,5-benzenetetramine tetrahydrochloride dissolved in ethanol solvent.[22] The QY value was reported as 30.2% and methylene blue has been detected via fluorescence quenching strategy. Yellow fluorescent CDs with QY of 78.6% were solvothermally synthesized from sodium citrate, carbamide, and anhydrous calcium chloride in toluene.[23]

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Blue-green-yellow fluorescent CDs were prepared in reverse micelle system by Prikhozhdenko et al.[24] Hexane was used as non-polar oil phase, nonionic polyoxyethylene lauryl ether as a surfactant, and either water or dextran sulfate aqueous solution was used as polar phase. The major advantages of hydrothermal method over others are the production of stable, well composed CDs with minimal loss of precursors. This method encourages the synthesis of hydrophilic CDs other than hydrophobic CDs. That is the CDs produced through hydrothermal method usually dissolve in polar solvents and impart good biocompatibity.

2.1.2. Combustion method

Simple heating under benign conditions also favours the synthesis of CDs. It involves the combustion of candle soot under favourable condition to get illuminating CDs.[25] Blue emissive CDs were synthesized from candle soot in the presence of HNO_3 and the CDs has been purified with PAGE (polyacrylamide gel electrophoresis).[26] Chen et al. successfully synthesized photobleaching resistive and green fluorescent B, N-doped CDs through normal combustion of aminophenyl boronic acid (APBA) as the starting material.[27] The resulting CDs have been used for the detection of Cu^{2+} ions via static fluorescence quenching. Soots from candle, paraffin oil, corn stalk, natural gas were oxidized by HNO_3 to synthesize CDs by scientists.[28,29,30,31] Although, this method is cost-effective, but low fluorescent QY CDs synthesis limits their wide usage.

2.1.3. Microwave-assisted method

Usage of higher energy for the synthesis of CDs is highly essential to bring up all the small molecules to accumulate together. Microwave technique can meet the criteria in the synthesis procedure of CDs because of its higher energy and used to prepare hydrophilic, hydrophobic or even amphiphilic CDs.[32] Yang et al. have synthesized blue fluorescent CDs through microwave pyrolysis from green natural material “kelp” as the main carbon source as well as ethylenediamine as the nitrogen dopant.[33] The resulting CDs have been applied for colorimetric detection of Co^{2+} ions. High fluorescent CDs with QY of 49.9% have been reported from succinic acid and tris(2-aminoethyl)amine as an “A_2 /B_3” monomer and the CDs displayed its high efficiency in cellular imaging. In et al. focused on the harsh synthesis method (pyrolysis using microwave) self-assembly followed carbonization to produce CDs from succinic acid and tris (2-aminoethyl)amine as an “A2 + B3” monomer set.[34] Green emissive CDs were synthesized from phthalic acid and triethylenediamine hexahydrate mixture in a very short time (60 s) by Wang et al.[35] Roasted chickpea was used for microwave-assisted synthesis of blue fluorescent CDs which was used for Fe^{3+} detection.[36] Fluorescent N-doped CDs were synthesized from microwave heating of L-ascorbic acid and β-alanine mixture.[37] The CDs were used in MDCK and HeLa cells bio-imaging application. So that it is very clear that microwave-assisted CDs synthesis is possible for both inorganic and green precursors.

2.2. Top-down approach

This approach involves the break down or cleavage of large carbon materials into small-sized CDs. The formation mechanism builds upon the disintegration of the bond between carbon atoms.[38] The most commonly used method for this approach includes mainly arc discharge, laser ablation and electrochemical oxidation.[39] This method generally possesses sp^2 hybridized carbon structures and they do not exhibit bright fluorescence properties. Surface modification is used to increase the luminescent property.[40, 41] Since this method involves the breaking down of the substances to the nanometer level, the process needs high energy to impart the breaking/ exfoliation. Generally carbon nanotube, graphene sheet, carbon fiber, etc. are used as starting materials. Some of the synthetic methods are explained in the following.

2.2.1. Arc discharge method

This method involves cutting down of bulk carbon materials to small carbon particles under gas plasma enclosed in the sealed container. The plasma generates high electric current which helps to maintain a high temperature inside the reactor. Decomposition of bulk carbon materials takes place at anode and the deposition of small carbon particles takes place in cathode. Scrivens et al. prepared single walled carbon nanotube by arc discharge method.[42] During purification by gel electrophoresis, fluorescent CDs were obtained as side product. Substitution of hetero atoms in graphene alters its band gap which has well explained by Rao et al. on their study of synthesis of blue fluorescent CDs under two different atmosphere.[43] Where the boron doped graphene quantum dot (B-GQDs) prepared under the atmosphere of H_2 + He + B_2H_6 as the gas phase of arc-discharge. Whereas the nitrogen doped graphene carbon quantum dot (N-GQDs) prepared with the mixture containing H_2 + He + NH_3. During this synthesis, most of the carbon atoms replaced with the heteroatoms provided like boron and nitrogen. The N- carbon quantum dot had slightly blue shift compared to B-CQDs because of the changes in the band gap. Su et al has performed an interesting study on the impurities formed during the arc discharge of single walled carbon nanotube and they successively did the chemical oxidation of such carbon by products to obtain fluorescent graphitic CDs with higher fluorescent upconversion under long wavelength irradiation.[44] They synthesized narrow distribution of graphitic CDs by controlling particle size through centrifugation followed by reflux with strong acid. Although, the fluorescence depends on the different energy level of the surface states which has been achieved by surface passivation. The obtained graphitic CDs contained significant amount of -OH groups on its surface to give upconversion fluorescence on irradiation at 550 nm to 900 nm. Downconversion spectra were also observed from these CDs. Mustelin et al. synthesized blue to yellow-green emissive CDs during arc oxidation of single walled carbon nanotube were oxidized by nitric acid.[45] Poursalehi et al. synthesized CDs through arc discharge among two high-

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purity graphite electrodes in distilled water for a duration of 2-4 sec.[46] The authors coupled TiO_2 with CDs which exhibit lower band gap of about 2.41 eV which helps the visible active photo-catalytic applications.

2.2.2. Laser ablation

In order to provide high energy to decompose large matter, high energy laser has imparted for the breaking down. Here, the surface of the matter gets etched with the high-energy beam laser, which can successively produce high temperature and pressure to melt the surface of the matter undergoes recrystallization to produce nanocarbon particles. Laser treatment of large carbon material produces small sized CDs with very less fluorescence properties Surface modification with polymers like polyethylene glycol (PEG1500N) and poly propionyl ethylenimine-co ethylenimine imparts fluorescence on the CDs surface.[47] The fluorescence enhancement can be facilitated by passivating with inorganic salts like NaOH or Na_2S. Moreover, the surface of the CDs can be modified by choosing the appropriate solvent on accordance with the property that require. The emission wavelength of the CDs can be tuned either by controlling the ablation duration or by altering the ablation and excitation laser wavelength. The laser ablation can be carried in vacuum, liquid or gaseous medium. In liquid medium, a part of the energy is absorbed by the liquid before the energy reaches to the matter been ablated. The CDs produces through the laser ablation is strong enough to tune the photoluminescence (PL) with different emission wavelength. Agglomeration helped to bring up to the particular PL that depends on the laser ablation duration.[48] It is an effective method to synthesize CDs with high water solubility, good fluorescence character, and different morphologies. Nevertheless, the only default demerit of this method is requirement of large amount of carbon source material to ablate. As the laser ablation method is very adventitious in short period as well as ease of handling made the method been used by Sun et al. for the synthesis of CQDs from carbon cloth.[49] Here they used homemade double beam pulse laser for effective synthesis. In order to increase the efficiency and time shortening, they divided the single laser beam into two parts. An intense thermionic electron emission after the coulomb explosion produces huge amount of temperature and pressure with electromagnetic fields exploit the carbon cloth on irradiation with laser. Within this high temperature and pressure, the carbon cloth split up to produce sp^2 carbon domains. Which further group up with pyrolyzed substituent having oxygen and sulphur as the active elements on its surface. While, the second laser beam

Table 1

S. No.	Synthetic approach	Preparation method	Advantages	Disadvantages	Ref.
1.	Bottom-up	Hydrothermal / Solvothermal	High energy and unique pressure with tunable particle size as well as fluorescence	Hard to synthesize CDs with hydrophobic nature	[39]
2.	Bottom-up	Combustion	facile procedure, ease of scale-up production, precise controllable design of initial molecules, low cost, and environmental benign operation	Need very high temperature	[56]
3.	Bottom-up	Microwave-assisted	Short reaction time, uniform heating, lower instances of side reactions and fewer by-product formation, more uniform physical, chemical and optical properties and need very less purification steps	Restricted to small volumes, limited in large scale synthesis, reducing the usability of solvents with lower boiling points	[32]
4.	Top-down	Arc discharge	Simple and inexpensive, possible to synthesize hydrophobic CDs	Hard to purify the fragments obtained. Normally used for CNT synthesis, low yield, limited distribution	[47]
5.	Top-down	Laser ablation	Easy passivation of the synthesized CDs	High cost equipment and laser, produced a heterogeneous mixture of CDs with very less fluorescence	[57]
6.	Top-down	Electrochemical	Ease of operation, abundance of raw materials, potential for mass production, low cost and not involving any harsh or toxic chemicals	The purification of CDs is tedious. No formation of CDs with electrolytes as H_2SO_4/EtOH	[58]

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accelerated the probability of domain formation and promote 35.4% photo luminescent CQDs. Silva et al. synthesized CDs with dimension of about 100 nm by UV pulsed laser ablation of carbon precursors dissolves in water.[50] Thus synthesized CDs showed fluorescence after the functionalization with NH_2-polyethylene-glycol (PEG_{200}) and N-acetyl-L-cysteine (NAC). The prepared CDs displayed two major average size dispersions of about 63 and 373 nm and the smaller size was obtained from the laser ablation of bigger particles.

2.2.3. Electrochemical method

All the discussed methods need a high amount of thermal condition for effective working. While the electrochemical methods operate in normal temperature and pressure. Even though, the electrochemical method is known for its facile tuning the particle size as well as the photoluminescence.[51] The CDs are formed through four steps like electro-oxidation, electro-polymerization, carbonization, and passivation. As the major types of CDs are more sensitive towards the pH of the solution, the CDs from the electrochemical method are also very much sensitive to pH by changing their fluorescence brightness with change in a pH.[51] Zhang et al. synthesized blue fluorescent CDs through electrochemical carbonization of sodium citrate and urea for the effective sensing of Hg(II) in water.[52] Usually, graphite or carbon rods are taken as the electrode for exfoliation. After the exfoliation, the crystal defect may happen by the removal of particles from the cathode and collectively dissolves in the corresponding electrolyte. The nanometer-sized carbon particle imparts good fluorescence. The size of the CDs majorly depends on the intensity of current applied in between the electrodes.[53] Zhang et al. successfully synthesized the carbon nano dots from alcohols through electrochemical method.[54] The uniformly formed CDs under electrolysis of small alcohol molecule in between two platinum rods act as both anode and cathode. As if the CDs preparation involves several combinations with ethanol, the alkaline combination with ethanol find good result on the production of efficient CDs. Strong blue luminescent nanocrystals developed by Ding et al. through electrochemical degassing of multiwalled carbon nanotube (MWCNT) of acrylonitrile solution with tetrabutylammonium perchlorate (TBAP) to synthesize CDs from MWCNTs.[55] As-obtained small nanocrystal on evaporation of acrylonitrile had a uniform spherical shape and a narrow size distribution of 2.8 to 0.5 nm in diameter. The MWCNT getting structure modulated through entangling or curled on electrochemical treatment, emphasising the surface area enhancement and there by the crystal deficiency. As if the electrochemical method used for graphite or carbon rod, though the chances of synthesis of CDs from MWCNTs are inevitable. There are many studies have been going on with it.

3. Conclusions

In this review, the preparation of fluorescent CDs has studied well through various top-down and bottom-up synthetic approaches. The CDs contains high amount of carbon along with other elements such as oxygen, nitrogen, etc. Different type of functional groups including hydroxyl, carboxylic, amine, amide attach on the CDs surface resulting in surface passivation that result in enhancement of fluorescence property. Although there are many synthetic methods available for bright fluorescent CDs synthesis, still there is a large difference of QY between semiconductor QDs and CDs. Therefore, there is still a remaining challenge to further improvement of QY of CDs. It can be anticipated that increasing effort and detailed study will create a progressive room for high quality CDs synthesis for various application.

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About the authors

Varsha UshaVipinachandran received her MS degree (2018) from University of Kerala, India. Currently she is pursuing Ph.D. at Vellore Institute of Technology, Vellore Campus. Her research focuses on smart nanostructured materials for environmental application.

Sathish Rajendran received his MS degree (2017) from Periyar University, India. Currently he is doing Ph.D. at Vellore Institute of Technology, Vellore Campus. His research focuses on design of fluorescent nanomaterials for sensing application.

Indhumathi Ashokan received her MS degree (2019) from Bharathidhasan University, India. Currently she is pursuing her Ph.D. at Vellore Institute of Technology, Vellore, India. She is working on fluorescent nanostructures as sensor and environmental application.

Kabir Hussain Badagoppam Haroon received his MS degree (2019) from Thiruvalluvar University, India. Currently he is a Ph.D. student at Vellore Institute of Technology, Vellore, India. His research interest focuses on plasmonic-based nanostructures for energy and environmental application.

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Dr. Susanta Kumar Bhunia received his Ph.D. (2015) from Indian Association for the Cultivation of Science, India. He is currently employed as Assistant Professor at Vellore Institute of Technology, Vellore Campus. His research focuses on the development of advanced nanostructured materials for various technological applications, especially nano-biotechnology, sensing and environmental application.

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