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REVIEW

Green Sources Derived Carbon Dots for Multifaceted Applications

Jothi Vinoth Kumar[1] · Ganesan Kavitha[1] · Rajaram Arulmozhi[1] · Velusamy Arul[2] · Subramanian Singaravadivel[2] · Natarajan Abirami[1]

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Abstract

For the past decade, the Carbon dots (CDs) a tiny sized carbon nanomaterial are typically much attentive due to their outstanding properties. Nature is a fortune of exciting starting materials that provides many inexpensive and renewable resources which have received the topmost attention of researchers because of non-hazardous and eco-friendly nature that can be used to prepare green CDs by top-down and bottom-up synthesis including hydrothermal carbonization, microwave synthesis, and pyrolysis due to its simple synthetic process, speedy reactions and clear-cut end steps. Compared to chemically derived CDs, green CDs are varied by their properties such as less toxicity, high water dispersibility, superior biocompatibility, good photostability, bright fluorescence, and ease of modification. These nanomaterials are a promising material for sensor and biological fields, especially in electrochemical sensing of toxic and trace elements in ecosystems, metal sensing, diagnosis of diseases through bio-sensing, and detection of cancerous cells by in-vitro and in-vivo bio-imaging applications. In this review, the various synthetic routes, fluorescent mechanisms, and applications of CDs from discovery to the present are briefly discussed. Herein, the latest developments on the synthesis of CDs derived from green carbon materials and their promising applications in sensing, catalysis and bio-imaging were summarized. Moreover, some challenging problems, as well as upcoming perspectives of this powerful and tremendous material, are also discussed.

Keywords Carbon dots · Microwave synthesis · Electrochemical sensing · Bio-imaging

Introduction

Nanomaterials are colloidal particles in which carbonaceous carbon nanomaterials have been fascinating major research attention as of their special features. Carbon dots (CDs) with amazing properties and applications have become a growing star as a novel nanocarbon member for the scientific community, mainly sp^2 bonded carbon pillar with sizes in the range between 1 to 10 nm [1, 2]. Xu et al. in 2004, during the single-walled carbon nanotubes (SWCNT) refining process carbon dots, were first exposed [3]. These CDs are zero-dimensional (0D) carbon nanomaterials with sizes below 10 nm, which are commonly quasi-spherical particles. Green CDs are raised to fluorescent nature carbon owing to very robust features with exclusive remarkable properties, up-conversion photoluminescence, water-solubility, multicolor wavelength tuned emission, chemical inertness, thermal stability, and biocompatibility defends the extraordinary gratitude grabbed by these interesting nanoparticles [4–7]. One of the main important features of carbon dots is their fluorescence. Various reports have been revealed that carbon dots show color- fluorescence release from the blue to the near-infrared (NIR) region.

A worldwide clarification for the fluorescence source of carbon dots has not yet been planned. As perspicuously shown in (Fig. 1), several scientific literature studies associated with CDs have been gradual increasingly published since 2005.

The improved research interest in such carbon materials may be attributed to many congenital advantages of CDs. Several benefits of CDs, such as simple and carbon source abundance, biocompatibility, low-cost synthesis process, and amazing fluorescent properties. Besides, CDs are inert and chemically stable, which form a colloidal solution and are extremely resistant to photobleaching matched to standard fluorescent organic dyes and semiconductor quantum dots (QD).

✉ Natarajan Abirami
abiramin@srmist.edu.in

[1] Department of Chemistry, College of Engineering and Technology, SRM Institute of Science and Technology, Kattankulathur - 603 203, Kanchipuram, Chennai, Tamil Nadu, India

[2] Department of Chemistry, SSM Institute of Engineering and Technology, Dindigul, Tamil Nadu 624 002, India

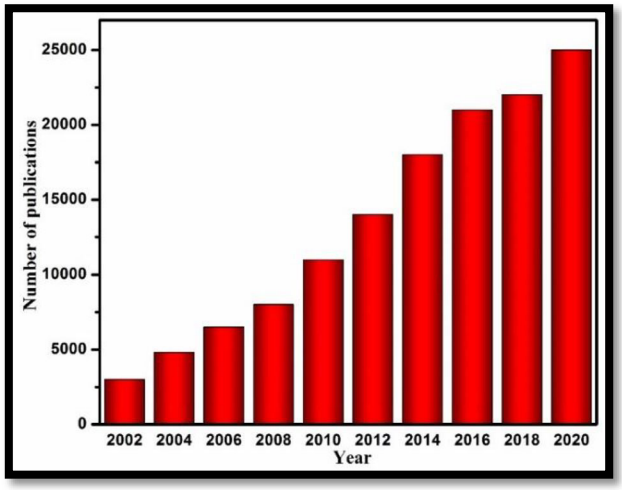

Fig. 1 The number of published literature studies according to the Google scholar search ("carbon dots")

Hence, the fluorescent properties of CDs are broadly used for sensing [8–10], biomedical imaging [11–13], catalysis [14, 15], energy research [16, 17], gene delivery [18, 19], cell differentiation [20, 21]. The carbon dots are usually well-defined as a class of core-shell composites involving a core and surface passivation by numerous chemical groups and frequently achieved through the invention of a highly thin insulating film of oligomeric polyethylene glycol on an acid-treated CD's surface, great fluorescence, high intensities and excellent quantum yield (QY) of CDs can be reached with current surface passivation [22].

Usually, CDs are synthesized by the surface functionalization of CNPs with organic and polymeric molecules. In the past few years, CDs synthesized from many chemical sources have been detected for the synthesis of CDs, such as ammonium citrate [23], citric acid [24], phytic acid [25], carbon nanotube [26], urea [27], boronic acid [28]. Green CDs are produced from green carbon sources. The word green mention materials that are biomass or natural renewable sources such as *Phyllanthus emblica*, sweet potatoes, green tea, egg, lotus root, bitter guard juice, and rice bran [29–35].

Green precursor affords fantastic properties, such as low-cost synthesis process, carbon source abundance, pronounced yield, biocompatibility, and high renewability. Newly, the natural- derived carbon nanomaterials develop a new part in the green synthesis with a financial benefit in widespread applications. Different classes of carbon nanomaterials such as activated carbon, CDs, etc., can be developed from natural wastes depending on the synthetic parameters. Comparatively speaking, the temperature of reaction (100–210 °C) of CDs formed through green synthesis is smaller than that of CDs attained by using some chemical additive which commonly needs a very high temperature of 210 °C. Also, the types of solvents that are very important for the production of CDs typically depends on the dissolving of the starting resources. The natural precursor is essentially reacted in normal solutions. Quantum Yield were optimized following green methods and precursors. The use of the green chemistry approach benefits cheap, renewable, reducing chemical exposure, abundant biomass, reduces environmental waste, and potential to scale up. The revealing of toxic elements, environmental pollutants, pesticides, biological molecules, etc., is much required in biology and chemical sciences.

In this review, we discuss synthetic methods CDs from natural green sources and focus on recent works related to sensors, catalysis, and bioimaging applications are summarized. Most researchers use the green pathway as it has benefits towards the cost, renewable, abundant biomass, less chemical exposure, and finally lesser waste discharge. Lastly, we propose specific upcoming perspectives and some challenges.

Synthetic Methodology

The synthesis methods of CDs can be gathered into the size and surface tuning. The size tuning can be pre-owned to prepare CDs and adjust their fluorescence properties by adjusting their sizes. Surface tuning approaches can be used to surface groups of CDs to prompt different surface states. Fluorescence properties of CDs mainly depend on their size owing to the quantum-confinement effect and the deviation in density and nature of the sp^2 domain. Size-tuning by prolonging or shortening the reaction time can be used to control the energy band gap of CDs.

Top-Down Formation

Normally, CDs can be made-up using top-down and bottom-up approaches as shown in (Fig. 2). The top-down methods in the synthesis of CDs from carbon precursors include synthesis routes. In the top-down process, the break of big carbonaceous materials or distributed into small-sized CDs by chemical or physical methods. Many altered carbon precursors have been used for the making of CDs like graphite [37, 38], candle soot [39], etc. This method of synthesizing CDs include high energy ball milling [40], electrochemical route [41], and laser ablation [42–44].

One of the first methods in the manufacture of CDs was when single-walled carbon nanotubes (SWCNTs) were treated to be purified from arc-discharge soot. The soot oxidized with nitric acid (HNO_3) and then extracted with a NaOH solution. Sun et al. also prepared the CDs over laser ablation, carbon target with argon as carrier gas resulted in the assembly of carbon nanoparticles. Formerly the product was outer passivated with some organic compounds which made the dots [45]. Economical precursors and mass production are also wider benefits of the top-down method [46].

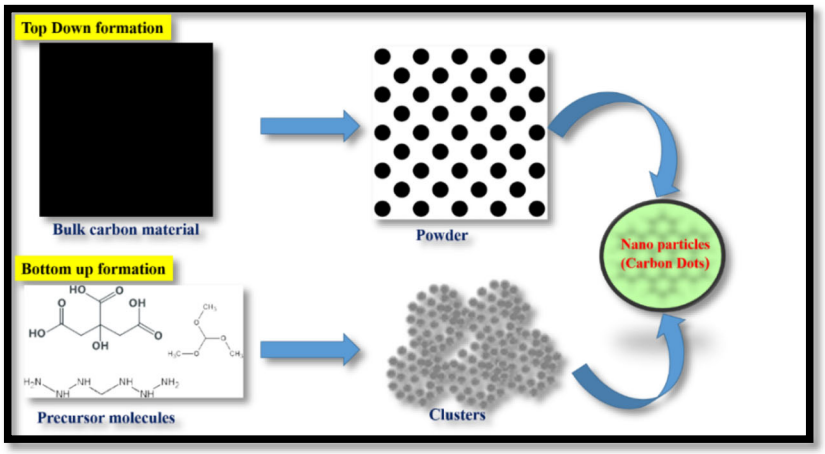

Fig. 2 CDs produced using top-down and bottom-up formation

Bottom-up Formation

The bottom-up method mostly refers to the carbonization and polymerization of small molecules into CDs on a large scale. The techniques such as chemical vapor deposition (CVD) [47], solvothermal [48, 49], combustion [50], microwave-assisted methods [51–53], hydrothermal [54–56], and ultrasonic-assisted methods [57–59], are classified under bottom-up approaches in the production of CDs. The benefit of the bottom-up method is that it has adequate control over the size of the product and the route. The bottom-up methods are simpler, easier, greener, and faster to make a bulky quantity and less cost. Furthermore, maximum renewable sources cover small biomolecules that can be carbonized to form green CDs.

CDs Derived from Green Sources and Methods

The CDs can be well-defined as the carbon quantum dots (CQDs) produced using 'green sources' as a precursor of carbon; the term 'green sources' refers to materials that are renewable natural products. The synthesis of CDs was limited to the use of carbonaceous precursors which caused CDs with less solubility and low quantum yield. To attain cost-effective, simple, environment friendly with unique optical and electronic properties, based on the many synthetic methodologies, several green carbon sources were explored. Since then several reports, green CDs could be prepared from natural sources, as shown in (Fig. 3). The chemical configuration of the resulting CDs is generally dependent on the choice of the raw material. Further, most of these green methods were enhanced with respect to the Quantum yield attained; therefore, this class as well argues the QY attained to altered precursors. The QY and solubility can be increased by surface passivation of the synthesized CDs. These green sources include plants, fruits, vegetables, juices, human derivatives, beverages, and bakery products. For the chosen carbon precursor, we have grouped these CDs in different sessions [60].

Hydrothermal Method

This method was convenient, eco-friendly, cheap. Hsu et al. [61] discovered the coffee grounds to prepared CDs with a diameter of 5 ± 2 nm and a QY of 3.8%. It was suggested that the use of hydrophilic sources results in high QY as a match to a hydrophobic one. Hydrothermal methods are eco-friendly, cheap, and convenient; many researchers have to work synthesize green CDs *Allium fistulosum* was used as a carbon precursor for the synthesized of CDs (with a diameter of 4.22 nm and QY of 10.48%) [62]. The low reaction time and higher temperature are encouraging for combining small particles. Kavitha et al. [63] synthesized green CDs using date palm fronds as the unique carbon source through low-temperature carbonization, with a QY of 33.7%. The achieved CDs had a spherical shape and better catalytic activity. Zhou et al. [64] prepared green CDs using a simple carbonization method of watermelon peels as a starting material. The attained CDs had a (size of 2 nm and QY of 7.1%), in the pH range of 2–11 and thermally stable. An elemental analysis study displayed that the achieved CDs were composed of 64.65% of Carbon, 1.13% of Nitrogen, 7.67% of Hydrogen, and 26.55% of Oxygen.

Microwave Irradiation Method

Microwave synthesis is the furthermost effective and less time method, QY, and quality of the CDs to synthesize green CDs. Hu et al. [68] used albumin (egg white) as the precursor to fabricate CDs in an aqueous medium. The formed CDs had an average diameter of 3.2 ± 1.1 nm and could be switched for semiconductor CDs. Wang et al. [69] prepared CDs from eggshell membranes, the QY was 14% and it has excellent

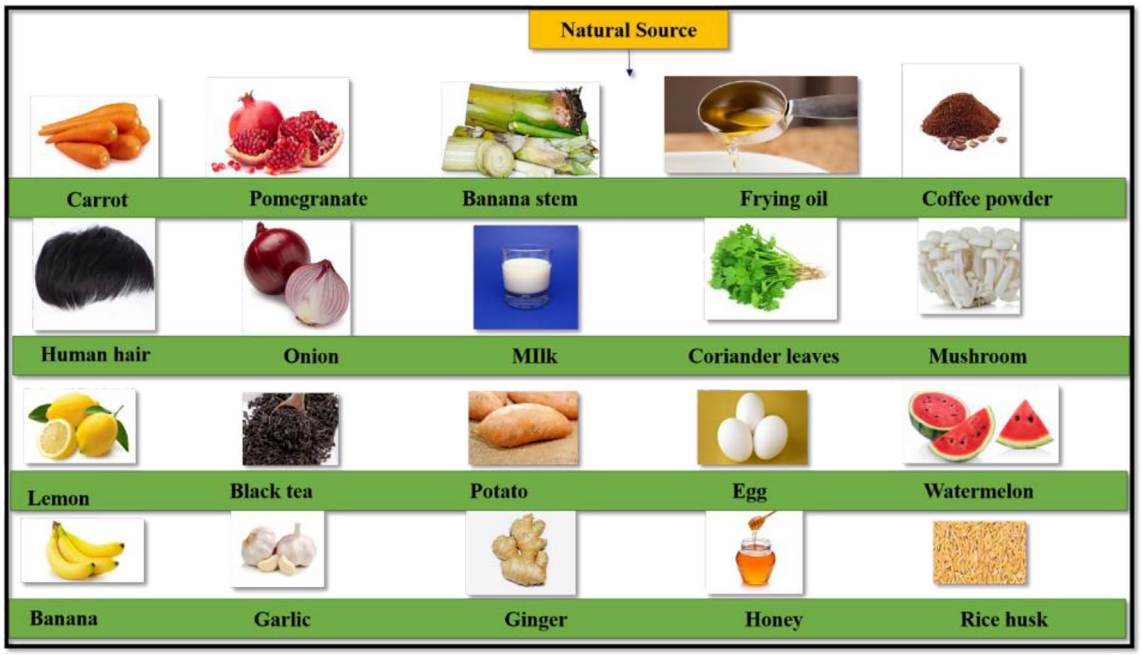

Fig. 3 Green carbon sources

water solubility. Fabrication of CDs using gelatin [70], lignin [71] as the carbon starting material through the microwave method has also been reported.

Pyrolysis Method

The pyrolysis method is a simple technique and well-established for the production of green CDs, followed by separation and purification. Zhang et al. [72] synthesized CDs from (marigold) a flower with a QY of 7.84% and doped the CDs with EDA to form N-CDs. Teng et al. [73] produced N-CDs using konjac flour as the precursor. The synthesized N-CDs with an average diameter of 3.37 nm, and a QY of 22%, were quasi-spherical. Other carbon sources, such as peanut shells [74], crab shells [75], and various plant leaves [76], to synthesize CDs.

Heating Method

The heating method is an economic, green, and simple method used to create CDs. De et al. [77] described the creation of CDs using bananas as the precursor without using any specific reagents for doping and passivation. The attained circular CDs had an atypical size of 3 nm and a Quantum Yield of 8.9%. Va et al. [78] synthesized CDs with diameters of 5 ± 2 nm and a Quantum Yield of 48% by heating method pseudo-stem. Other precursors, such as mangosteen pulp [79] extract, and rice bran [80] have been used to manufacture CDs via the heating method. Scientists have worked on many other unique and simple methods to synthesize CDs. Phadke et al. [81] synthesized CDs using neem gum as a source, with a biogenic method. The size of the CDs was 5–10 nm, and they were steady under several pH and temperature conditions. Zhao et al. [82] synthesized green CDs using corn bract by a cheap, simple, and suitable solvothermal method with ethanol. The Quantum Yield and typical diameter of the synthesized green CDs were 6.9% and 2.6 nm, respectively.

Other Methods

Researchers have hired various other simple and innovative methods to synthesize CDs, in addition to the above talk about conventional synthetic routes. Ye et al. [83] prepared nitrogen &sulfur co-doped CDs (NS-CDs) through a pyrolysis method using biomass, including pigeon feathers, egg yolk, egg white, and manure, and the QYs of the as-synthesized NS-CDs were 24.87%, 16.34%, 17.48%, and 33.5%, correspondingly. Romero et al. [84] N & S co-doped CDs were prepared photochemically using an aqueous vegetable extract in the front of polyethylene glycol (PEG). The size of the as-prepared CDs reduced with increasing ultraviolet (UV) irradiation time, which led to a rise in the fluorescent intensity. The as-synthesized CDs act as a reducing agent for the reduction of Ag^+ and Au^+ ions into their elemental forms. The aqueous-soluble CDs with a size of 2.5 nm were prepared from *gynostemma* as a source, by the calcination method [85].

From the directly above discussion, we can come to the end that the hydrothermal method is the most extensively used technique for the synthesis of CDs because of its low cost, eco-friendly nature, and ease of synthesis. The other regularly used methods are hydrothermal carbonization, microwave irradiation, and carbonization. However, additional effort

should be made to prepare green CDs from non-conventional (natural) sources, rather than by traditional heating. Then, the microwave irradiation method should be explored further because of its homogenous, efficient delivery of heat, and simultaneous, with very fast reaction rate.

The fluorescence properties and toxicity of green CDs frequently depend on their size distribution. The formation of green CDs with uniform size is very hard challenging, which can avoid their use in biological applications. Finally, a variety of green CDs synthesis methods cause differences in size, fluorescence color, QY, toxicity, etc.,

Fluorescence Mechanisms of CDs

CDs prepared using different ingredients and methods own different fluorescence properties. Fluorescence emission is of enormous importance, as plays a key role in imaging and sensing applications. Presently, the most accepted luminescence mechanisms are the quantum confinement effect [15, 86], molecular fluorescence [24, 87], and surface state emission [88].

Molecular Fluorescence

Current research shows that the development of fluorescent impurities through the bottom-up approach chemical synthesis (i.e., molecular fluorescence) gives to the popularity of emission from CDs. Yang's group [89] established that the QY fluorescence of CDs synthesized by citric acid ($C_6H_8O_7$) and ethylenediamine (EDA)was reasoned to a fluorescent molecule (imidazo [1,2-a] pyridine-7-carboxylic acid, 1,2,3,5-tetrahydro-5-oxo-, IPCA). The CDs have been verified to be a mixture of IPCA, carbon cores, and polymers, in which IPCA donated to the strong blue fluorescence (Fig. 4). Nowadays, there are few key points for the fluorescence foundation of CDs: (i) surface-state emission, which is insistent on by the hybridization of the carbon support and attached functional groups; (ii) the molecular fluorescence, it is made by fluorescent impurities since the by-products created during carbon dots synthesis mainly give to the fluorescence emission; and (iii) carbon core-state emission, which is encouraged by perfect crystals with fewer defects and reformed groups [87].

Surface State Emission and the Quantum Confinement Effect

The surface state emission is broadly accepted mechanism that contains the degree of surface oxidation and functional groups. The red-shifted release of CDs depends on the oxygen content existing on the surface of CDs. As oxygen content rises, the number of surface defects increases. Pang et al. [93] suggested that the CDs outcomes from surface state emission, where the energy gap is presiding over by the size and

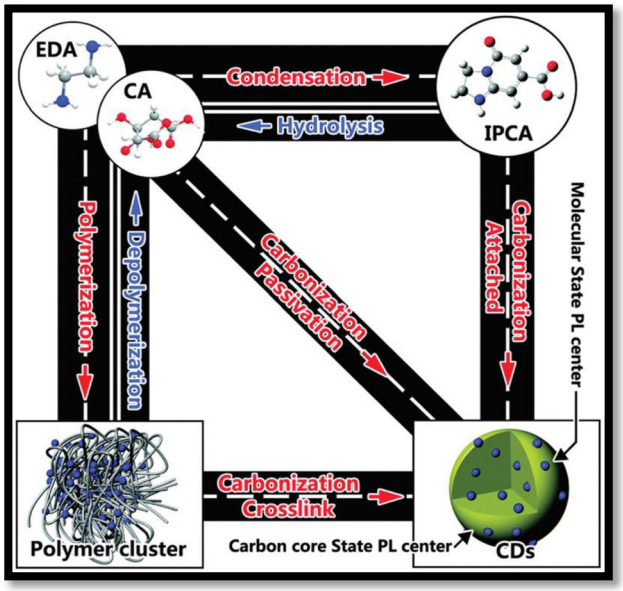

Fig. 4 A relationship between products in hydrothermal method of CA and EDA. Reproduced from ref. [89] with permission. Copyright 2015 Royal Society of Chemistry

surface properties of the CDs. Results revealed that the surface oxidation and π-electron system have a major effect on the energy gaps of the surface states. The upper extent of surface oxidation or the π-electron system has a slighter energy gap of the surface states in the CDs. C.Z. Huang group [94] proposed that the fluorescence of CDs was ascribed to a synergetic effect of the quantum confinement effect and the surface state emission. Sequences of full color emitting carbon quantum dots with the absolute QY greater than 70% were prepared by a hydrothermal method and more purification by column chromatography (Fig. 5). The electron circulation of any semiconductor nanocrystal is partial by the crystal boundary and displays band-gap- and size-dependent energy relaxation manners [95]. Li et al. [15] exhibited a red-shifted emission behavior of prepared CDs with growing sizes from 5 to 35 nm because of the quantum confinement effect.

Physical and Chemical Properties

CDs are mainly containing Carbon (C), Hydrogen(H), Oxygen (O), and Nitrogen (N)components which are existent in the form of several functional groups on the surface and good water solubility. The advantage of natural precursors over chemical objects is that maximum of these methods post-modifications or surface passivation, separate reactant for doping, do not require. A protein and further biomolecules give self - passivation to CDs. Carbon Dots possessing an obvious crystal lattice with sp^2/sp^3carbons. All CDs on the surface various chemical groups presented, such as nitrogen, oxygen, carboxyl, amino groups, and these groups play a vital role. CDs are active in photon-harvesting since of π–π^* transition of C = C bonds.

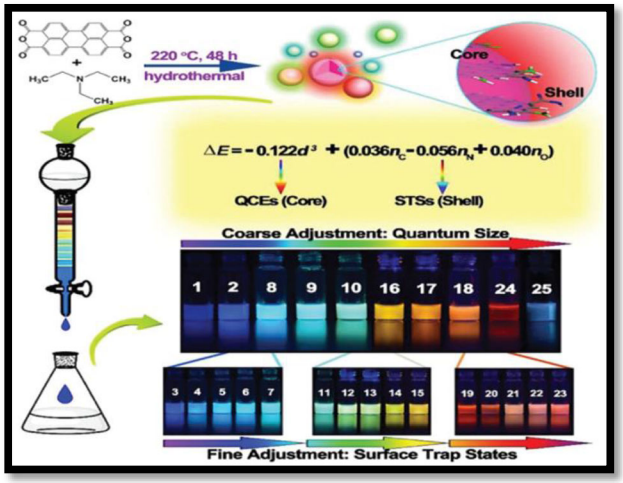

Fig. 5 Synthesis and purification of fluorescent CQDs. The equation and the photos specify the size of the core-shell nanostructured fluorescent CQDs which plays the adjustment of the fluorescence wavelength due to the surface trap states (color online). Reproduced from ref. [95] with permission. Copyright 2019 Royal Society of Chemistry

The absorption peak found in 220–280 nm is normally in the short-wavelength region reason to π–π* transition of C = C bonds, and the shoulder peak located at around 310–380 nm is ascribed to n–π* transition of C = O bonds. The x-ray diffraction (XRD) of CDs typically signifies amorphous nature owing to disordered carbon (C) atoms, broad diffraction peak in the range of 2θ value 20–25°, and a d-spacing between 0.31–0.38 nm [96, 97]. Carbon dots with varied structures have quite parallel optical characteristics in terms of their absorption, phosphorescence, chemiluminescence, fluorescence, upconversion photoluminescence, electro-chemiluminescence, and photo-induced electron transfer property. Photoluminescence is one of the most interesting features of CDs, one constant feature of the PL for CDs is the discrete dependence of the emission wavelength and intensity, the reason for this phenomenon may be the optical variety of nanoparticles of various size or CDs with different emissive traps on the surface. Electrochemiluminescence activities are moderately reactive to surface passivation, wherein the surface-passivated CDs showed weak electrochemiluminescence actions but strong fluorescence. The synthesis of CDs with fluorescence emission is of special importance, as fluorescence plays a key role in imaging and sensing applications. The Quantum confinement effect electron sharing of some of the semiconductor nanocrystal is partial by the crystal boundary and displays size-dependent energy and band-gap relaxation processes [95].

Sensor Application of Green Carbon Dots

Carbon Dots materials were widely working in chemosensors application specifically on the detection of numerous metal ions including compound pollutants and naturally active molecules associated with the biological sciences and environment. The CDs still make use of predominately as a fluorescent probe in the analysis based chemosensors due to their photostability, tremendous water solubility, and tunable fluorescence wavelength with huge quantum yield. The small size, low cytotoxicity, very huge surface functionalization make CDs highly reactive and can easily interact with chemical species; easy functionalization creates CDs an outstanding contestant for fluorescence-based particular sensing applications [98]. CD surface makes a new electron-hole pair through energy transfer, thereby altering the fluorescence intensity of the CDs. The sensing mechanisms of CDs are founded on the subsequent principles: (a) The energy produced from CDs can be re-absorbed by metal ions inner filter effect (IFE); (b) Carbon dots can give photo-excited electrons to the conduction band (CB) of metal ions (Energy Transfer mechanism, electron transfer); (c) CDs can contribute photo-excited electrons to the vacant d-orbital of metal ions; (d) energy can be transferred from the higher state of CDs to metal ions fluorescence resonance energy transfer, (FRET).

Selective detection of metal ion analytes is a very crucial universal task to an observer and reduce the effluent in the environment. Overall, the CDs naturally have oxygen surface functional groups such as hydroxyl and carboxylic groups to form a hydrophilic nature of CDs and high surface active areas. Hence, the metal ions can powerfully work together with CDs along with surface bonding thus resulting in the tuning of CDs properties. In the past few years, many kinds of natural precursors were used to obtain CDs mainly by using synthetic techniques which are listed in Table 1.

Mercury (Hg^{2+}) Sensor

Mercury (Hg^{2+}), ions are measured as very destructive water contaminants even in the drop levels. Traditional orange juice-derived N-CDs were used by Zhili et al. [99] to detect Hg^{2+}in the recovery of samples was ranged from 102.0% to 103.0%. They also confirmed that their approach could be very useful for the environmental water analysis of Hg^{2+}. Radhakrishnan et al. [100] have synthesized CDs through the hydrothermal treatment of *Coccinia indica* and used for high selectivity for the detection of Hg^{2+}, Pb^{2+}, Cu^{2+}, and Fe^{3+}, with a lower limit of detection (LOD) of 3.3 nM as shown in (Fig. 6a). When Hg^{2+} was directly added, the fluorescence intensity of CDs was quenched owing to the interface amid the functional groups on the surface of CDs and Hg^{2+} ions, based on inner filter effects (IFE) and non-radiative electron transfer (ET) processes.

The fluorescence intensity of CDs was quenched is represented by the red bars (Fig. 6b). Fluorescence emission spectra of CDs, fluorescence sensing results for the detection of Hg^{2+},

Table 1 The details for the synthetic techniques and different applications of naturally-derived CDs

Precursor	Method	Reaction condition	QY (%)	Application	[Ref.]
Seville orange	Hydrothermal	130 °C, 12 h	13.3	Fe^{3+}	[36]
Spinach leaves	Hydrothermal	200 °C, 2 h	–	Fe^{3+}	[65]
Allium fistulosum	Hydrothermal	120 °C, 3 h	–	Solar cells	[66]
Cherry Tomatoes	Hydrothermal	180 °C, 6 h	9.7	Trifluralin Herbicide	[67]
Marigold	Pyrolysis	1000 °C,5 h	7.84	Fe^{3+}, bioimaging	[72]
Peanut shells	Pyrolysis	400 °C,4 h	10.58	Cu^{2+}	[74]
Gynostemma	Calcination	400 °C, 4 h	5.7	Bioimaging, antioxidant	[85]
Kelp	Microwave	800 W, 1.5 h	23.5	Co^{2+} and pH sensing	[90]
Shrimp shells	Calcination	230 °C, 2 h	20	Cr^{+6}	[91]
Gardenia fruit	Hydrothermal	180 °C, 5 h	10.7	Hg^{2+} and cysteine	[92]
Orange juice	Hydrothermal	200 °C, 11 h	31.7	Hg^{2+}	[99]
Coccinia indica	Hydrothermal	180 °C, 7 h	30.0	Hg^{2+}, Cu^{2+}, Pb^{2+}, Fe^{3+}	[100]
Lemon peel	Hydrothermal	200 °C, 12 h	14	Cr^{6+}	[109]
Dunaliella salina	Hydrothermal	200 °C, 3 h	8	Hg (II), Cr (VI), cell imaging	[110]
Eleusine coracana	Pyrolysis	300 °C, 3 h	–	Cu^{2+}	[111]
Bamboo leaves	Hydrothermal	200 °C, 6 h	7.1	Cu^{2+}	[112]
Grass	Hydrothermal	180 °C, 3 h	4.2	Cu^{2+}	[113]
Peach gum	Hydrothermal	180 °C, 16 h	28.46	Au^{3+},cell imaging	[115]
Jackfruit seed	Microwave	600 W, 1.5 min	17.9	Au^{3+}, cell imaging	[116]
Gum tragacanth	Hydrothermal	180 °C, 12 h	66.74	Au^{3+}	[117]
Miscanthus grass	Hydrothermal	180 °C, 4 h	11.6	Fe^{3+}	[119]
Mint leaves	Hydrothermal	200 °C, 5 h	–	Fe^{3+}, ascorbic acid	[121]
Snake gourd peels	Hydrothermal	180 °C, 12 h	28.6	Fe^{3+}	[122]
Grape skin	Hydrothermal	190 °C, 3 h	18.67	PA	[125]
Kerosene fuel soot	Oxidative	100 °C, 12 h	–	PA	[126]
Apple seeds	Pyrolysis	300 °C, 1 h	20	4-NP, cell imaging	[127]
Oyster mushroom	Hydrothermal	200 °C, 25 h	12.51	o-NA, p-NA, m-NA, p-NP	[128]
Casein	Microwave	30 min	18.7	Leaf cells	[137]

Note: o-NA: o-nitroaniline, p-NA: p-nitroaniline, m-NA: m-nitroaniline, p-NP: p-nitrophenol, 4-NP: 4-nitrophenol, PA: Picric acid

Pb^{2+}, $Cu^{2+,}$ and Fe^{3+}, selectivity plot, and sensing mechanism (Fig. 6c).

Lu et al. [101] prepared CDs from pomelo peel as a precursor for the highly sensitive detection of Hg^{2+}, with a detection limit of (LOD) 0.23 nM. An original non-radiative electron-hole pair was made when Hg^{2+} ions intermingled with a carboxyl group existing on the surface of the CDs, primary to fluorescent quenching mechanism via FRET. The fluorescence intensity of prepared CDs improved with increasing pH from 2 to 6 and then reduced from pH 7 to 12.

Li et al. [102] prepared N- CDs nitrogen-doped from Chinese yam and some modified them with a carboxy fluorescein (FAM)-labeled DNA, fluorophore, for the particularly sensitive detection of Hg^{2+}, and the limit of detection (LOD) for $Hg^{2+}$was 1.26 nM. Initially introduced 6-mercaptopurine (6-MP) to the FAM-DNA-CD complexes, where it formed stable hydrogen bonds, covalent bonds, and π-π conjugations and slightly increased the fluorescence intensity of the bulky conjugated systems, this system was quenched when $Hg^{2+}$was added to the conjugated system because of the relations between $Hg^{2+}$and the DNA molecule, which affected the FAM-DNA to breakdown away from the CDs. The most hazardous metal ion Hg^{2+} is a heavy eco-friendly threat due to its bio, accumulation and high harmfulness. The same group, has worked flour derived CDs for Hg^{2+} detection with the limit of 0.5 nM.

Guo et al. [103] prepared hair derived CQDs from human products as shown in (Fig. 7). Results revealed highly selectivity and sensitivity towards mercury ion (Hg^{2+}) with a LOD 10 nM and linear reaction in the range of 0 to 75 μM.

Detection of Pb^{2+}Sensor

Toxic heavy metal lead (Pb^{2+}) has extended growing attention for its strong effects on humans, animals, and the environment. Radhakrishnan et al. [100] described the amine-

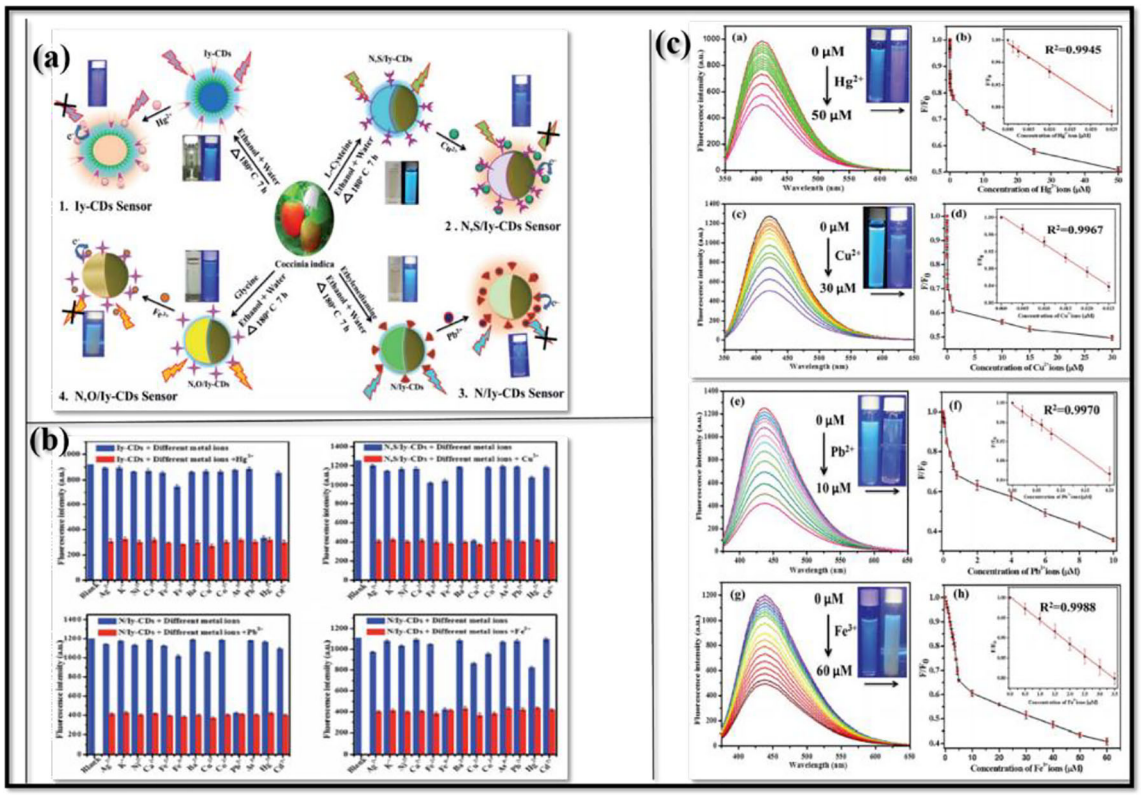

Fig. 6 **a** Schematic representation for the detection process of *Coccinia indica* derived CDs towards Hg^{2+}, Pb^{2+}, Cu^{2+}, and Fe^{3+}ions. **b** The relative quenching of the fluorescence intensities metal ions is represented by the red bars. **c** Fluorescence emission spectra of CDs, inset plot shows the linear of the curve. Reproduced from ref. [100] with permission. Copyright 2019 Royal Society of Chemistry

functionalized CDs-based sensor to selective detect Pb^{2+} with a determined limit of detection of $0.21X10^{-6}$m. The fluorescence intensity of CDs was successfully quenched by the addition of Pb^{2+} ions owing to the electron transfer from the amine group in the excited state of CDs to vacant d-orbitals of Pb^{2+}. Chocolate as source derived soluble CDs were used for the detection of Pb^{2+} from aqueous solution selective chelating effect toward lead ions with the supporting of the surface hydroxyl group through fluorescence quenching mechanism, with a detection LOD of 12.7 nM [104].

Bandi et al. [105] synthesized CDs using berries these CDs possess amine and primary functional groups on the surface. Lead sensors revealed outstanding sensitivity to the detection of Pb^{2+} with a LOD of $9.64X10^{-9}$m. The amine-functionalized CDs act as an electron donor because the nitrogen atom of the amine groups can donate a lone pair of electrons to the empty d-orbital of Pb^{2+} ions through a

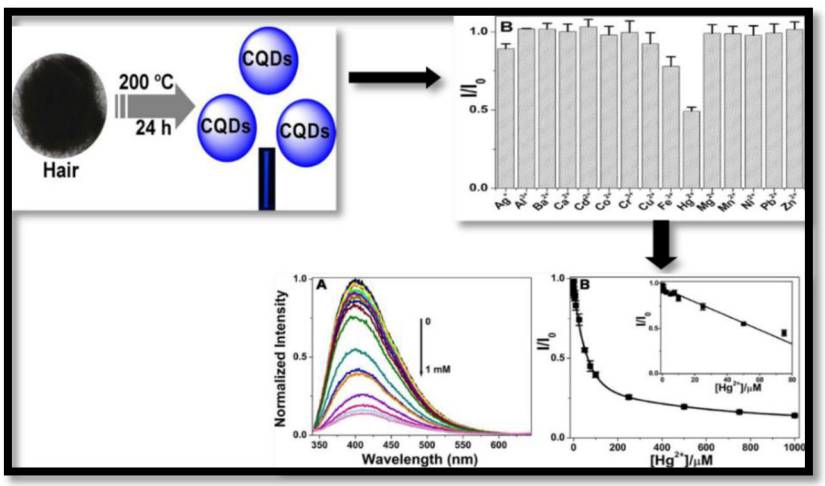

Fig. 7 Hair derived CQD for sensing of Hg^{2+}. Reproduced from ref. [103] with permission. Copyright 2019 Nature

nonradiative electron transfer. Sahu et al. [106] prepared CDs using *Ocimum sanctum*as precursor, a sensitive detection sensor for $Pb^{2+}$with detection limit 0.59 nM and range of 0.01–1.0 μM. This sensing method is due to the effective binding affinity between vacant d-orbital of lead (Pb^{2+}) ions and amine group existing on CDs surface, nitrogen atoms donate a couple of electron to vacant d-orbital of lead ions and origins nonradiative electron transfer.

Chromium (Cr^{6+}) Sensor

One of the most highly toxic metal ions (Cr^{6+}) leads to cancer it damages the immunological systems and hazardous environmental pollutant. Roshni et al. [107] synthesized CDs from groundnut for sensitive detection sensor for Cr(VI) with detection limit 0.1 ppm (1.9 μM). The quantum yield (7.8%) the fluorescence intensity slowly decreased with an increase in the concentration of Cr (VI) based on the Stern-Volmer plot. Chinawooth et al. [108] prepared CDs from shallot extract to detect Cr (VI) ion created on fluorescence quenching process with a limit of 11.7 μM and the high quantum yield of 32.34%. Tyagi et al. [109] used lemon peel-derived CDs for selective sensing of Cr^{6+} with a detection limit of 73 nM. The occurrence of vacant d–orbital in Cr^{6+} and small lying d-d transition state support non-radiative electron-hole pair recombination and leads to fluorescence quenching mechanism of CDs. The selective detection of Cr^{6+} using dunaliella salina biomass-derived CDs has also been reported by FRET or ET mechanisms [110].

Copper (Cu^{2+}) Sensor

Copper ion is one of the most indispensable abundant commonly found in natural water and can affect kidney or liver upon long-term uncovering with very high concentrations and easily accumulate in our body system. Murugan et al. [111] synthesized Eleusine coracana derived CDs used for 'turn-off' sensor probe for detection of Cu^{2+} with a limit of detection (LOD) 10 nM and the linear range 0–100 μM. The bamboo leaves derived CDs [112] were capped with a BPEI capping agent for selective detection of Cu^{2+} ions with a LOD 115 nM. The concentration increasing Cu^{2+} used photoluminescence quenching beside with a blue shift in adsorption of Cu^{2+} and desorption of BPEI from CDs surface.

Liu et al. [113] used N-CDs nitrogen-doped and derived from grass for the selective detection of Cu^{2+} with a detection limit of 1 μM. Cu^{2+} can combine with the nitrogen and oxygen atoms present on the CDs surface because of their highly chelating power, subsequently quenching the fluorescence intensity of the N-CDs via ET or FRET. Aqueous soluble CDs from eggshell membranes are used for the selective detection of Cu^{2+} with a LOD of 0.8 nM, using the turn-off fluorescence mechanism through FRET or ET [69]. Wang et al. [114] synthesized N-CDs from crown daisy leaves as a carbon source, used as a fluorescent probe for Cu^{2+} ion, the linear range from 10.0 to 120.0 nM and a very low detection limit of 1.0 nM of Cu^{2+}. In this probe for real water sample analysis, a series of concentrations of Cu^{2+} was added from the real water samples ranged from 92.1% to 103.8%.

Gold (Au^{3+}) Sensor

Liao et al. [115] prepared highly water-soluble N-CDs using peach gum polysaccharides and EDA for fast, selective, and sensitive exposure of Au^{3+} ions, with a LOD of 64 nm. Fluorescence quenching could happen via the synergetic effect of non-radiative electron transfer and FRET from the N-CDs to Au^{3+} and gold particles act as an acceptor to absorb light emitted by N-CDs. The CDs derived from jackfruit seed are used as a tremendous fluorescence probe in the detection of Au^{3+} with great sensitivity of and 239×10^{-9}m. CDs as a fluorescence probe were established to sense Au^{3+} ions based on the fluorescence turn-off mechanism [116]. Zeinab et al. [117] synthesized N-CDs from gum tragacanth by hydrothermal treatment with a high quantum yield (at 436 nm) was 66.74%. The LOD reached 2.69 μM using the Stern-Volmer plot and a linear range of 0–100 μM.

Iron (Fe^{3+}) Sensor

The Fe^{3+} metal ions play a crucial role in genetic functions can cause critical diseases like Alzheimer, Parkinson, heart attack and some other diseases. Fe^{3+}sensing via fluorescence quenching mechanism of CDs was reported by Zhu et al. [76].This group prepared CDs from numerous plant leaves by pyrolysis method, fluorescence quenching of CDs to happen via the interaction between the functional groups on the CD surface and Fe^{3+} ions and following electron transfer between them. Dou et al. [118] honey-derived CDs for Fe^{3+}selective and sensitive interaction of Fe^{3+} ions with several functional groups–NH_2, -OH, and–COOH existent on the surface of CDs, with a detection limit of 1.7 X 10–9 mol/L. Some of the other green precursor CDs have been active for the sensing of Fe^{3+}, Miscanthus grass with the QY 11.6% [119], Milk via fluorescent sensing [120]. Mint leaves derived CDs promising on-off-on fluorescent sensor with a limit of detection of 374 nM [121], Snake gourd peels derived CDs exhibited a quantum yield of 28.6%, detection of Fe^{3+} ions by fluorescence quenching mechanism with (LOD) of 0.398 μM, linearity range of 10–100 μM [122].

pH Sensor

The pH homeostasis has an essential physiological impact and hereafter this sensor has revealed important attention. The waste frying oil derived S-CDs acts as a pH sensor range from

3 to 9 as shown (Fig. 8). The fluorescence intensity displays a linear increase in pH range 2 to10 and also displays a reversible behavior [123]. The pH responsive behavior was increasing de-protonation with accumulative pH that leads to the upper concentration of carboxyl group on the CDs surface.

Biomolecules Sensing

In recent years' researchers have used green synthesized CDs for the selective sensing of biomolecules, such as carbohydrates, amino acids, glutathione, and hydrogen peroxides, the biosensing mechanisms are parallel to chemical sensing. Date kernel derived CDs were used to determination of zoledronic acid in human serum [124]. N-CDs were synthesized from Konjac flour through a pyrolysis treatment and were used for off-on sensing of L-lysine [73]. The major key components of Konjac flour are glucomannan, proteins, heteropolysaccharides, inorganic salts, and starch. In this N-CDs pH-dependent photoluminescence (PL) manners, the intensity could be improved by the addition of NaOH and amino acids, such as L-arginine and L-lysine. Wang et al. [69] developed water-soluble CDs using egg shell membranes via a microwave method and used the sensor for glutathione and fluorescence off and on mechanism. Initially, CDs were added to Cu^{2+} ions, the fluorescence intensity decreased by the ET mechanism, glutathione is added to recover the fluorescence, the CDs surface removed the Cu^{2+} ions via coordination with Cu^{2+}. The CDs/Cu^{2+} sensor exhibited a linear range of detection of 0.5–80 μM, with a LOD of 0.48 μM.

Nitro Compounds Sensing

A nitro compound is highly dangerous and explosive by nature that can strictly affect environment. The water solubility nature of compound surgeon environmental pollution over soil and water thereby creating health issues to living organisims. Nitro compounds including nitrobenzene (NB), nitrotoluene (NT), nitromethane (NM), picric acid (PA) nitrophenol (NP), nitroethane (NE), 2,4,6-trinitrotoluene (TNT), etc.,

The improvement of a chemosensor selective to nitro compounds is of more importance to prevent environmental contamination. CDs derived from grape skin used to sensor system displayed an even greater sensitivity of 10×10^{-9} m toward the sense of picric acid (PA) [125]. The electron transfer involved in the sensing mechanism that electron transfer (ET) from a donor of the amino group on CDs to acceptor of the nitro group on picric acid resulted in the fluorescence quenching of CDs (Fig. 9a). Srinivasan et al. [126] prepared CDs using kerosene fuel soot and applied for the PA sensor have suggested hydrogen bonding interaction in the detection of picric acid along with inner filter effect (IFE). The hydrogen bonding among the carboxyl group on the surface of CDs and the nitro group of picric acid was verified by the FTIR analysis of CDs with PA. The CDs synthesized from apple seeds emit blue fluorescence that was successfully quenched by 4-nitrophenol (4-NP) via the fluorescence resonance energy transfer [127]. The acceptor (4-NP) and donor (CDs) in distance between were calculated to be 2.66 nm and with a detection limit of 13×10^{-9}m (Fig. 9b). Muhammad et al. [128] have developed oyster mushroom-derived CDs for a rapid, selective sensor and emitted cyan color with a quantum yield of 12.51%. The sensor sense nitroarenes (NAs) based on FRET, IFE, and PET which endowed the sensing mechanisms (Fig. 9c). The detection limits were 0.78 μM for o-NA, 2.29 μM for m- NA, 0.50 μM for p-NA, and 1.56 μM for p-NP.

Detection of Pesticides

Pesticides are toxic to humans, chemical-based materials that are used to safeguard crops alongside rodents, weeds, fungi, insects, and some pests. The pesticides can be categorized as herbicide, fungicide, insecticide. Among pesticides, carbamate pesticides and organo-phosphorus were broadly used in the agricultural field owing to their wide spectrum and very

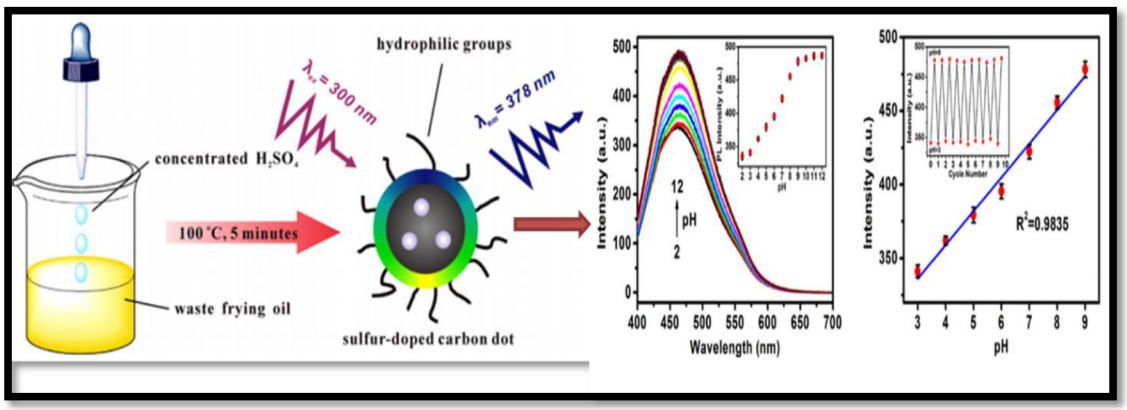

Fig. 8 Frying oil derived S-CDs for pH sensor. Reproduced from ref. [123] with permission. Copyright 2014 Elsevier

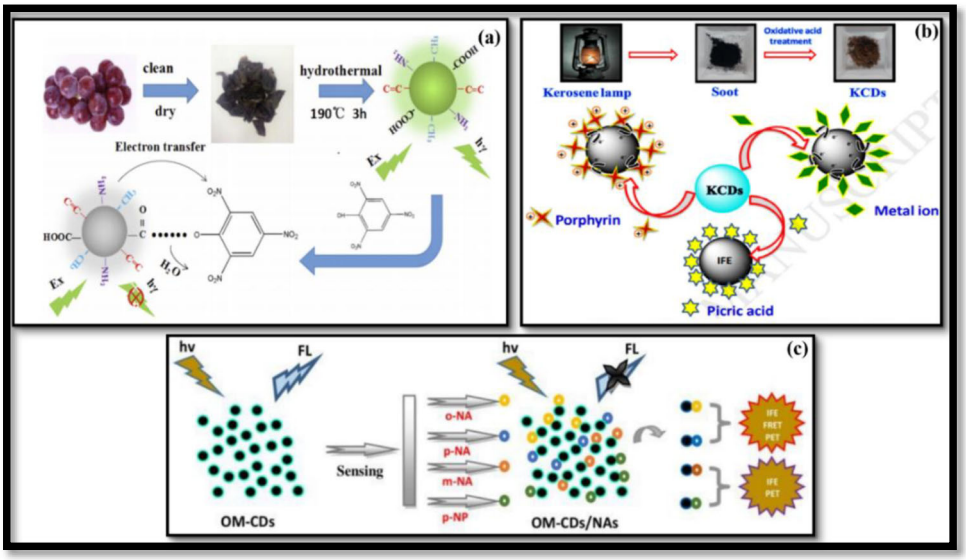

Fig. 9 Schematic illustration for synthesis method and detection mechanism of CDs toward nitro compounds (**a**) ET-based picric acid detection. Reproduced from ref. [125] with permission. Copyright 2018 Elsevier. (**b**) IFE-based picric acid detection. Reproduced from ref. [126] Copyright 2019 Elsevier. (**c**) FRET, IFE, PET-based o-NA, p-NA, m-NA, p-NP detection. Reproduced from ref. [128] with permission. Copyright 2020 Royal Society of Chemistry

great efficiency. Deka et al. [129] water hyacinth derived CDs are applied as a sensor for detecting the (pretilachlor) herbicide. The quantum yield is 17.02% with the LOD is found to be 2.9 μM and shows green fluorescence. This sensor also shows admirable selectivity and specificity for natural soil samples.

Carbon Dots in Catalysis

The functional CDs are displaying their greater capability as a catalyst in dye degradation of water contaminants application. CDs can act as an electron mediator (donor and acceptor) this specific characteristic nature makes them a potential catalyst. The synthesized green CDs display satisfactory photocatalytic activity related to other photocatalysts, such as TiO_2, ZnO, and CdS because of their higher chemical stability, better water solubility, and lower toxicity. The green CDs has absorption of light in the near-infrared region. The light of CDs in the (NIR) allows them to act as an effective catalyst since organic compounds can be broken by high-energy radiation [130].

Photocatalysis objects to accelerate reactions, which is a novel type of "green" technology for the degradation of pollutants and gaining ideal hydrogen energy from water and sunlight [131]. The photocatalysis mechanisms by CDs are created on the following major principles: (a) CDs with upconversion PL absorb light in the visible wavelength and emit UV light, which is absorbed by metal/metal oxides to form photo-excited electrons; (b) π -conjugated CDs can act as a sensitizer by providing their photo-excited electrons to the conduction band (CB) under visible light irradiation; (c) CDs can also support to bind organic dyes via π-π* interaction, increasing the probability of degradation. (d) CDs act as electron reservoir to inhibit e^-/ h^+ recombination by accepting photo-excited electrons from metal/metal oxides; Willow bark derived CDs used as a catalyst for the instantaneous reduction of the Au(III) complex and graphene oxide(GO) for the development of reduced graphene oxide (rGO) -Au- CD nanocomposites under UV light irradiation, there was no reduction of the Au(III) complex and GO in the absenteeism of CDs under same situations, thereby representing the photo-induced electron transfer capacity of the CDs [132].

Arul et al. [133] prepared nitrogen-doped CDs (NCDs) using *Hylocereus undatus* extract its carbon source, these N-CDs were used as a catalyst for the methylene blue (MB) dye reduction. The absorbance of methylene blue solution containing $NaBH_4$ reducing agent slowly reduced with time upon the addition of N-CDs confirming the role of CDs. The MB and $NaBH_4$ might be adsorbed on the surface of N-CDs owing to the occurrence of surface functional groups in which N-CDs act as electron transfer (ET) mediator to permit for electron transfer from BH_4 - ions to MB over CDs. Prasannan et al. [134] synthesized CQDs from orange peels and coupled them with ZnO to formation a ZnO-CQDs, this composite was used as a catalyst for the degradation of dye naphthol black-blue azo (NBBA) under UV light radiation. A graphic illustration of the dye-degradation mechanism of CQDs/ZnO is shown in (Fig. 10a). An electron/hole pair was produced on the surface of ZnO upon light irradiation when the electrons were excited from the VB to the CB and transported to the CB of the conjugated CDs, the photo-generated electrons and holes more combined with water and oxygen made hydroxyl ($OH^·$) and superoxide ($O_2^·$) radicals, which degraded the dye.

Extremely fluorescent red-emitting Magnesium-nitrogen-doped carbon dots (Mg-N/CDs) using bougainvillea plant leaves extract was established for the sunlight-instigate catalytic degradation of MB dye [135]. The Mg-N/CDs shows emission peaking at 678 nm with a QY of ≈ 40%. In the

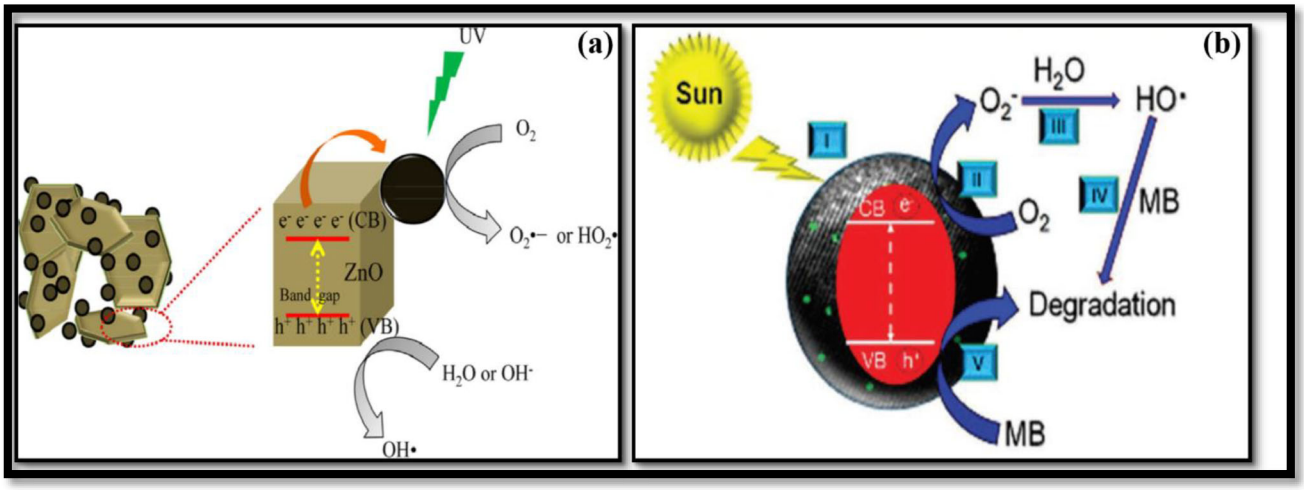

Fig. 10 (**a**) Schematic mechanism for the photocatalytic degradation of dyes on CQDs/ZnO. Reproduced from ref. [134] with permission. Copyright 2013 American Chemical Society. (**b**) sunlight irradiation the reactive oxygen radicals were formed. Reproduced from ref. [135] with permission. Copyright 2018 American Chemical Society

reduction of MB, Mg-N/CDs attained 99.1% photocatalytic activity under light irradiation for 2 h. Upon sunlight irradiation, the reactive oxygen radicals were formed by the collaboration between photoinduced electrons and oxygen molecules (Fig. 10b). Formerly, the reactive oxygen radicals (O_2^-) act together with water molecules to form the hydroxyl radicals ($^{\cdot}OH$) which were then reacted with adsorbed methylene blue molecules on the surface of CDs important to the decomposition of MB. The usage of CDs in the field of catalysis is limited compared with chemical precursor derived CDs. Hence, more research in the area of green derived CDs-based photocatalysis should be done.

Carbon Dots in Bioimaging

As we mentioned previously, the properties of CDs are extraordinary, which facilitate their in vitro and in vivo applications. Their amazing properties such as biological, photostability, aqueous solubility, and photobleaching resistivity have made CDs widely studied material for bioimaging. In the past few years, numerous researchers have used a variety of sources to synthesize a variety of CDs for bioimaging. Barbosa et al. [136] used cow manure for the preparation of CDs and used it for the fluorescent imaging of MCF-7 cells. The obtained CDs showed more localization within the cytoplasm, whereas alteration of the CDs through acid activation better localization inside the nucleoli.

Nowadays, CDs prepared from microalgae (*Dunaliella salina*) have been used for the bioimaging of human embryonic kidney (HEK-293) cells [110]. The as-synthesized CDs exhibited high biocompatibility and excitation-dependent emission colors (red and green after treatment of 50 μg/mL of CDs for 24 h) within HEK-293 cells (Fig. 11a). Feng et al. [96] prepared CDs under microwave method using silkworm chrysalises as a carbon source. The CDs show low cytotoxicity at the huge concentration of 15 mg/mL^{-1}and display dark blue, red, and green colors under a fluorescence microscope in cellular imaging experiments of HeLa cells obtained under a bright-field (Ai, Bi); excited at 340 nm, 495 nm, and 550 nm (Fig. 11b). Bajpai et al. [137] developed N-doped (N-CDs) using a milk protein, casein, and used them for the bioimaging of spinach leaf cells. They also studied the uptake dynamics of N-CDs by plant guard cells and experimental that the percentage of uptake of NCDs increased with increasing incubation time, then attained a maximum value, and then decreased.

Mingyue et al. [138] lychee exocarp-derived NBCDs were used as a near-infrared (NIR) fluorescence imaging for cancer cell's targetable and photodynamic therapy. The cytotoxicity was evaluated using (Bel-7404 and HL-7702) by MTT assays, a 0.5 mL of NBCDs solution was injected into the tumor areas of mice, the NIR fluorescence signal was immediately detected from the tumor, was detectable for up to 120 h (Fig. 11c). Most bioimaging applications of natural derived CDs are very limited to in vitro studies. Therefore, researchers should also use CDs to discover some other biological systems, such as in vivo and ex vivo models, which are also very limited. In the future, deep-tissue bioimaging is expected to be done.

Carbon Dots in Other Fields

In addition to the above-indicated and varied applications, there are some additional applications, such as anti-counterfeiting materials, light display materials, and confidential materials. Zhang et al. [139] investigation of CDs developed from wastewater used for the production of tofu. This work is related to the application of fluorescent CDs as light

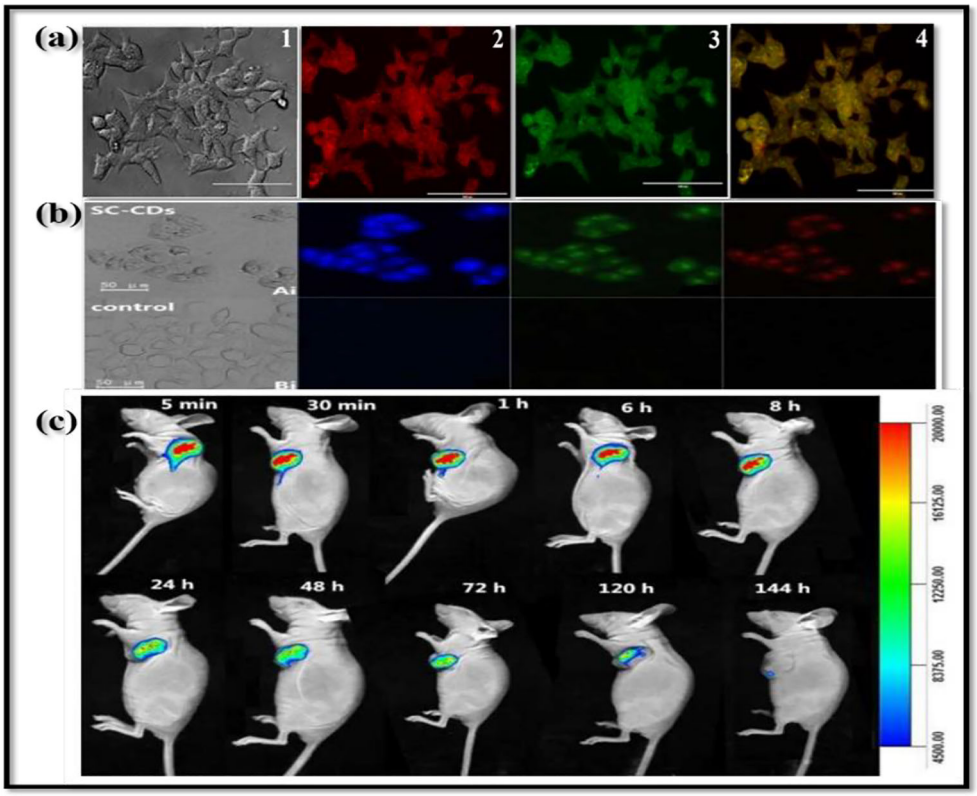

Fig. 11 (**a**) Cell imaging of HEK-293 cell lines. Reproduced from ref. [110] with permission. Copyright 2019 Elsevier. (**b**) Fluorescent microscope images of HeLa cells (Ai - CDs, Bi - control). Reproduced from ref. [96] with permission. Copyright 2016 Royal Society of Chemistry. (**c**) Real-time NIR fluorescence images of the mice tumor with NBCDs at different time points. Reproduced from ref. [138] with permission. Copyright 2018 Royal Society of Chemistry

display materials. Liu et al. [140] synthesized fluorescent CDs from hair as a carbon precursor by a pyrolysis treatment. These CDs are valuable in fluorescent patterns, anti-counterfeiting labeling, and flat panel displays. The application of CDs is very different, and we can research varied applications that are helpful to human development.

Conclusions and Upcoming Perspectives

In this review, we discussed common synthetic routes, properties, and practical applications of CDs. Green CDs prepared from natural, green and biomass sources are more suitable for many applications including sensing, catalysis, bioimaging, solar cells, and drug/gene delivery. Significantly, as related to some former nanomaterials, the normal wasted materials could be sustainably and powerfully changed into CDs. For most of the natural/biomass-derived CDs, the surface is self-passivated owing to the occurrence of several functional moieties.

In the upcoming, green CDs has to be prepared using underutilized, low cost and undervalued sources. The broad mechanism of the formation of CDs is still not fully understood. The great consideration on selectivity and sensitivity of green source derived CDs has significant sensing on hazardous metals like Cr^{6+}, Pb^{2+}, Hg^{2+} and Fe^{3+} and also on biomolecules, nitro compounds, pesticides etc,. Green source derived CDs display outstanding environmental friendliness, simplicity, greenness, cost-effectiveness, non-toxicity, and innovation. CDs were coupled with a conservative photocatalyst, increased the catalytic effectiveness of CDs by the low-energy gap between the HOMO and LUMO, photo-induced electron transfer mechanisms as well as a bridged charge recombination. CDs-based materials had shown outstanding catalytic performance to the reduction of environmental contaminants. CDs have also been explored as bioimaging for cell lines, both in vitro, in vivo, and ex vivo. So, natural carbon precursor is likely to become the greatest popular carbon sources in the future period. While greatly research has concentrated on developing new natural resources, synthetic methods, innovative applications still some challenges remain on the following.

1. Resulting in an effective and bulk production method to prepare better-quality CDs with less size and great QY.
2. CD based sensors would also be used in main sensing areas such as radiation prevention, early detection of diseases, global security, and safety of public utilities.
3. CDs have lesser cytotoxicity than of ordinary chemical-derived CDs. Hence, the use of green derived CDs for catalyst and bioimaging could be better in the future.
4. CDs can be used as a catalyst for the low-energy gap among the HOMO and LUMO. This area has not been studied by researchers using green CDs.

5. Researchers have commonly performed in vitro (cell lines) bioimaging; so, further biological systems, such as in vivo and ex vivo imaging applications, should be traverse using green derived CDs.
6. There are few reports on photodynamic and photothermal therapy with green CDs. Hence, improved attention is essential in this field.

Abbreviations *CDs,* Carbon dots; *SWCNT,* Single-walled carbon nanotubes; *0D,* Zero dimensional; *NIR,* Near-infrared region; *QD,* Quantum dots; *QY,* Quantum yield; *NaOH,* Sodium hydroxide; *CVD,* Chemical vapor deposition; *CQDs,* Carbon quantum dots; *EDA,* Ethylenediamine; *PEG,* Polyethylene glycol; *IFE,* Inner filter effect; *FRET,* Fluorescence resonance energy transfer; *ET,* Electron transfer; *PET,* Photo-induced electron transfer; *LOD,* Limit of detection; *rGO,* Reduced graphene oxide; H_2O_2, Hydrogen peroxide; *GCE,* Glassy carbon electrode; *CV,* Cyclic voltammetry; *NB,* Nitrobenzene; *NT,* Nitrotoluene; *NM,* Nitromethane; *PA,* Picric acid; *4-NP,* 4-Nitrophenol; *NE,* Nitroethane; *TNT,* 2,4,6-trinitrotoluene; TiO_2, Titanium dioxide; *ZnO,* Zinc oxide; *CdS,* Cadmium sulfide; *MB,* Methylene blue; $NaBH_4$, Sodium borohydride; *MTT,* 3-(4,5-Dimethylthiazol-2-yl)-2,5-Diphenyltetrazolium Bromide; *HOMO,* Highest occupied molecular orbital; *LUMO,* lowest unoccupied molecular orbital.

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CRediT Authorship Contribution Statement **Jothi Vinoth Kumar**: Methodology, Writing – original draft. **Ganesan Kavitha**: Visualization. **Rajaram Arulmozhi**: Formal analysis. **Velusamy Arul**: Conceptualization, **Subramanian Singaravadivel**: Editing. **Natarajan Abirami**: Writing – Review & Editing, Supervision.

Data Availability All data generated or analysed during this study are included in this published article.

Declarations

Conflict of Interest All authors declare no conflicts of interest to disclose.

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