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## REVIEW

# Sensor heavy metal from natural resources for a green environment: A review relation between synthesis method and luminescence properties of carbon dots

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## Abstract

Carbon dots (CDs) are 10-nm nanomaterial classes as excellent candidates in various applications: physics, biology, chemistry, and food science due to high stable biocompatibility and high surface expansive. CDs produced from natural materials have received wide attention due to their unique benefits, easy availabilities, sufficient costs, and harmless to the ecosystem. The various properties of CDs can be obtained from various synthesis methods: hydrothermal, microwave-assisted, and pyrolysis. The CDs have shown enormous potential in metal particle detection, colorimetric sensors, electrochemical sensors, and pesticide sensors. This review provides systematic information on a synthesis method based on natural resources and the application to the environmental sensors for supporting the clean environment. We hope this review will be useful as a reference source in providing the guidance or roadmap for new researchers to develop new strategies in increasing luminescence properties CDs for multi detection of heavy metals in the environment.

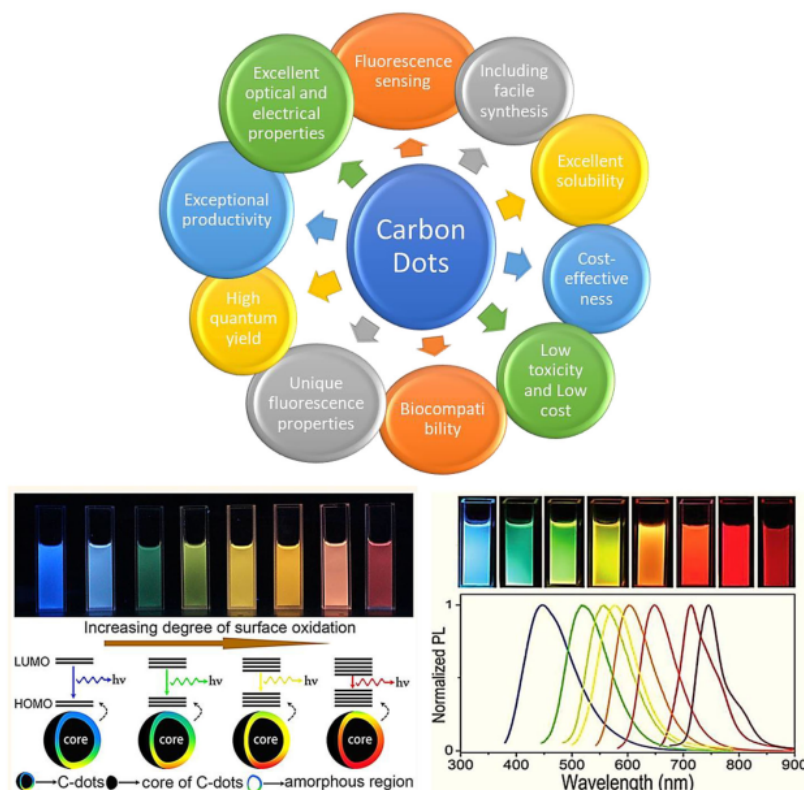
## KEYWORDS

carbon dots, environmental sensor, synthesis method

## 1 | INTRODUCTION

The term of 'carbon dot' is usually used for fluorescent carbogenic materials with an outer shell made of a carboxylate or other useful substances such as graphite, where the carbon at the centre and contains covalent bonds with oxygen and nitrogen.<sup>[1]</sup> The sizes of carbon dots are up to 50 nm but for good fluorescence usually lower than 10 nm.<sup>[2]</sup> In the previous reported references, the various classifications of carbon nanoparticles, for example, green carbon dot (GCD), carbon nanodot (CND), carbon nanocluster, and C-dot or carbon dots (CDs).<sup>[3]</sup> CDs are emerging as environmentally friendly and promising nanomaterials by the use of fluorescence in detection system, easy admixture, excellent solvency, low cost, low toxicity, biocompatibility, attractive fluorescence properties, high quantum yield (QY), phenomenal optical and electrical properties.<sup>[4,5]</sup> The physical, chemical, and electronic properties of nanoscale structures are strongly dependent on the synthesis using nanotechnology.<sup>[6]</sup> CDs as a fluorescent

material, provide wide potential application in various biomedical and optoelectronic<sup>[1]</sup> applications and have been an interesting topic for scientist around the world.<sup>[7]</sup> Some of the properties of CDs have become of interest due to stable photoluminescence (PL), expansive excitation spectrum, iridescence, fluorescence, adjustable surface function and water solvency.<sup>[8]</sup> PL CDs have shown potential in biological, medical, and environmental applications with environmentally friendly concept.<sup>[9]</sup> Some reported references are of chemiluminescence (CL) properties of CDs, where the research is focused to improve the intrinsic properties of CL to gain more applications.<sup>[10]</sup> CL is defined as the production of light through chemical reactions. The CL properties of CDs were first discovered when CDs coexisted with some oxidants, such as potassium permanganate (KMnO<sub>4</sub>) and cerium(IV).<sup>[11,12]</sup> Various properties can be improved by surface modifications of CDs as shown in Figure 1. Incorporating CDs into the framework has been developed with high emission properties, suspended fluorescence, and brightness at room temperature.<sup>[5,13,14]</sup>



**21** **FIGURE 1** Various properties of carbon dots (CDs) and we have included **34** luminescence properties which strongly depend on the surface oxidation state as the effect of the bandgap<sup>[5,13,14]</sup>

Currently, most researchers have focused on how to obtain the excellent luminescence properties of CDs by two ways of synthesizing: bottom-up and top-down methods.<sup>[15]</sup> The reported reference for the top-down methods is laser ablation, electrochemical oxidation, ultrasonication, arc discharge, and chemical ablation.<sup>[16]</sup> For the bottom-up methods they are hydrothermal,<sup>[17]</sup> microwave-assisted<sup>[18]</sup> and pyrolysis.<sup>[19]</sup> For this review, we focus on bottom-up methods for detection of environmental pollution. The properties of CDs: hydrophilicity and luminescence properties have been identified as strongly dependent **1** on the reaction conditions such as carbon source, temperature, and time, and through various planning techniques and combining of CDs with different sources<sup>[20]</sup> as shown in Figure 1.

The application of CDs that have been widely reported due to luminescence properties for drug transport, optronics, biomedicine, energy, light emitting devices,<sup>[21]</sup> photodynamic therapy, solar cell<sup>[22]</sup> and sensing.<sup>[23]</sup> The focus application in this review is for sensing, which is more specific for fluorescence sensors,<sup>[24]</sup> colorimetric sensors,<sup>[25]</sup> electrochemical sensors,<sup>[26]</sup> and pesticide sensors.<sup>[27]</sup>

Various industries develop every day waste which without any treatment has a negative effect on the environment. A problem for the environment is heavy metal pollution.<sup>[28]</sup> In this review, we provide information on the synthesizing methods and the conditions that affect luminescence properties of CDs and then, we explore the environmental sensor applications. We add the type of CDs for environmental sensors with excellent luminescence performance. Therefore,

it is objective and appropriate to introduce a broad survey and use **5** of CDs for environmental controls and for detection of pollution. This review summarizes the latest improvements of CDs for environmental sensor applications.

The CDs for environmental sensor applications are cheap, and provide reliable means of testing in real time, low toxicity, and biocompatibility.<sup>[4,5,8]</sup> However, there is variability of the fluorescence properties which strongly depend on the synthesized methods and report various sensitivity. **2**

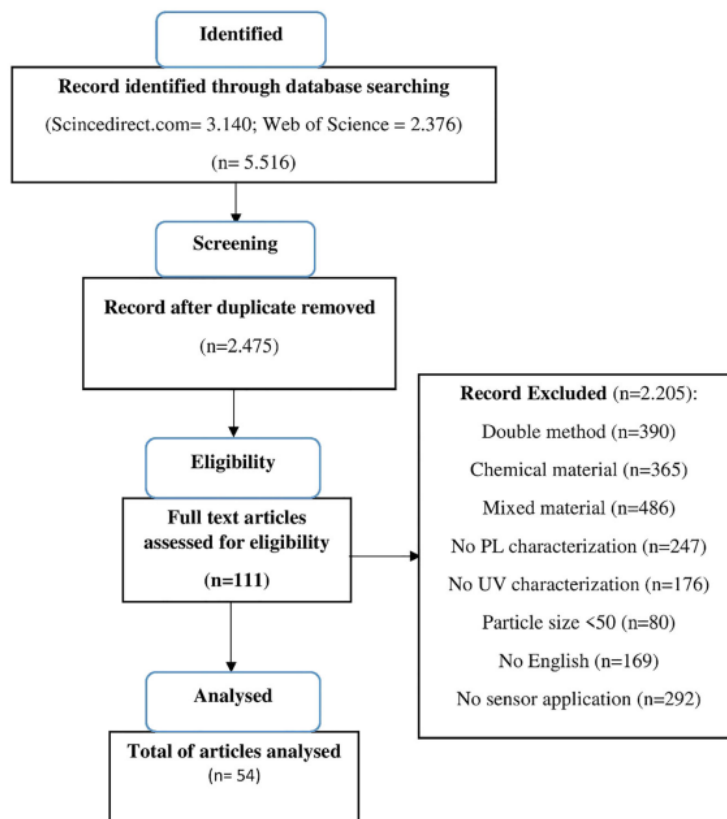
Hence, the purpose of this review was to: (1) record and describe the relation of synthesizing methods with luminescence properties for evaluating the most efficient and effective methods for environmental sensor applications, **2** and (2) collate the data to develop a standardized efficient method of synthesizing CDs for environmental sensor applications.

We believe that this review will provide a reference and guide for scientists to develop carriers in the field of natural sources for sensor environmental pollution based on luminescence performance in the future.

## 2 | METHODS

The systematic review was developed in accordance with the preferred reporting items for systematic review and meta-analysis (PRISMA) statement as shown in Figure 2. **13**

**FIGURE 2** PRISMA diagram demonstrating the full-text article selection process



## 2.1 | Search strategy

A literature search was conducted on the following databases: Scencedirect.com (Elsevier), and the Web of Science electronic database from 2013 to 2021 to identify relevant publications. Reference lists of each eligible article were searched manually to identify potential ones that meet the inclusion criteria.

## 2.2 | Selection of studies

After the removal of duplicates, the title, abstract, synthesis, and characterization of the articles were screened according to the inclusion and exclusion criteria. Inclusions such as having PL characterization, ultraviolet (UV) characterization, having particle size < 50 nm, English writing, natural material, focus on sensor application. Excluded article if dual method, chemical source, mixed material, no PL characterization, no UV characterization, particle size > 50 nm, no application sensor, text not available. Then, the entire text of the article was researched.

## 2.3 | Data extraction

Data was extracted manually by identifying databases through Elsevier and Scholar as many as 5516 articles, the data analysed were

56 articles. The data recorded from each article is divided into three parts, namely hydrothermal synthesis, microwave, and pyrolysis. Each part consists of precursor which is the material used, application which consists of fluorescence sensor, colorimetric sensor, electrochemical sensor, and pesticide sensor. Particle size, QY (%), preparation condition, temperature and time are analysed, linear range, and limit detection in sensor applications.

## 2.4 | Statistical analysis

The data were synthesized qualitatively, using the appropriate totals.

## 3 | CARBON DOTS SYNTHESIS METHOD

CDs are made from different natural materials and synthesis methods: hydrothermal, microwave-assisted, and pyrolysis. There are two ways to produce CDs: top-down and bottom-up methods. The bottom-up methods have an advantage, the synthesis and size are good and easy to control.<sup>[8]</sup> Normal results with heteroatomic can get benefit from obtaining high QYs, fixed properties, and without any extra passive steps.<sup>[20]</sup>

Green sources have widely been used for CDs as a readiness to use environmentally friendly synthesis and raw precursors, e.g. from

parts of plants. The various methods used in synthesizing CDs from several parts of plants have advantages and disadvantages as presented in Table 1.

The optical imaging frequently utilized on CDs is traditional fluorescence imaging estimations including Stokes utilizing excitation in short frequencies, such as the bright (UV) or blue-green and has some restrictions, for example, (i) signal-to-noise low proportion generated via automatic fluorescence and dense light dispersion from organic tissue; (ii) the low depth UV entry profundity and visible emission in natural tissues; and (iii) the chance of DNA harm in short frequencies particularly UV excitation.<sup>[36]</sup> For fluorescence imaging of cells and tissues, CDs are worthwhile as far as their prepared fluid solvency, physicochemical and photochemical qualities, high optical performance and non-secure flickering, and biocompatibility.<sup>[37]</sup> Lately the fluorescent test is the best technique in the particle-identification, functional effortlessness, and high affectability.<sup>[38]</sup> The various experiments on CDs for fluorescence sensors<sup>[39]</sup> show the same optical properties with UV absorption. Most of the CDs exhibiting fluorescence wavelength emission properties depend on the excitation wavelength, red driven CDs by growing excitation frequency. The understanding of fluorescence CDs is valuable for various applications. As a result, there has been much research dedicated to the study of fluorescence systems.<sup>[31]</sup> UV-visible and PL spectroscopy are known spectroscopic methods commonly used to analyse the optical behaviour of CDs.<sup>[40]</sup>

To measure the QY of CDs, the reference standard quinoline sulphate (QS) with fluorescence were used, the absorbance value (A) in the CDs and QS solutions were measured with the same wavelength excitation. The results of the fluorescence of CDs can be obtained in the following Equation (1):

$$QY_{CQD} = QY_{QS} \times \frac{I_{CQD}}{I_{QS}} \times \frac{A_{QS}}{A_{CQD}} \left( \frac{n_{CQD}}{n_{QS}} \right)^2 \times 100\% \quad (1)$$

QY is the fluorescence quantum yield (%),  $I$  is the fluorescence area of incident light and  $n$  is the refractive index of the solution. The specified QY of the carbon quantum dots (CQDs) is 1.8% and the decay period of fluorescence is determined by the decay curve.<sup>[41]</sup>

### 3.1 | Hydrothermal method of CDs

Among the bottom-up methods, the hydrothermal method is ideally used for CDs because of its advantages such as environmentally friendly, low cost, simplicity and adaptability.<sup>[42]</sup> The hydrothermal synthesis method requires a large amount of heat energy because the temperature response is generally around 200°C<sup>[43]</sup> and easy to achieve high QY for CDs.<sup>[20]</sup> Generally, organic precursor solutions for hydrothermal reactors at high temperatures.<sup>[22]</sup>

The hydrothermal method also provides surface carbon functionalization by presenting carboxylate, amino, and hydroxyl groups with good biocompatibility. This makes it easy to generate CDs with dynamic species for explicit organic applications.<sup>[44]</sup> Hydrothermal

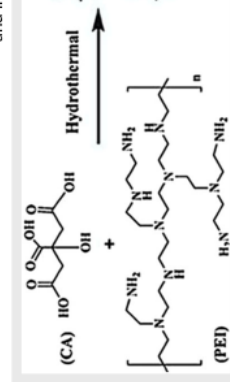
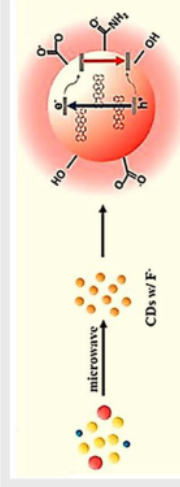
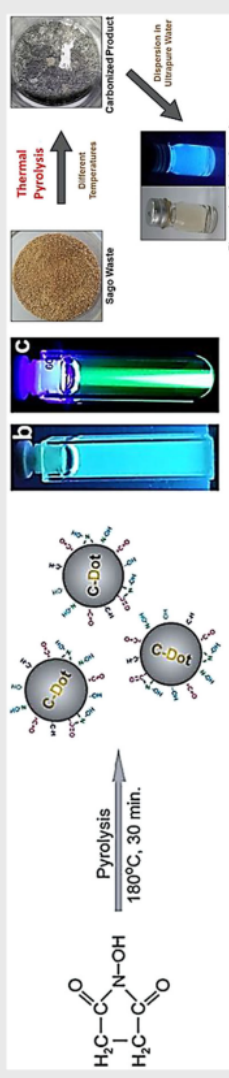
carbonization has provided a remarkable advance directness and the creation of CDs with large QYs<sup>[45]</sup> and *in situ* surface passivation.<sup>[46]</sup> The synthesis has been carried out using natural materials such as: lemon peel,<sup>[47]</sup> pear juice,<sup>[48]</sup> sweet potato,<sup>[49]</sup> banana juice,<sup>[50]</sup> watermelon,<sup>[51]</sup> orange peel,<sup>[52]</sup> coffee bean,<sup>[53]</sup> apple juice,<sup>[54]</sup> bamboo leaves,<sup>[55]</sup> lemon,<sup>[56]</sup> and strawberry.<sup>[57]</sup>

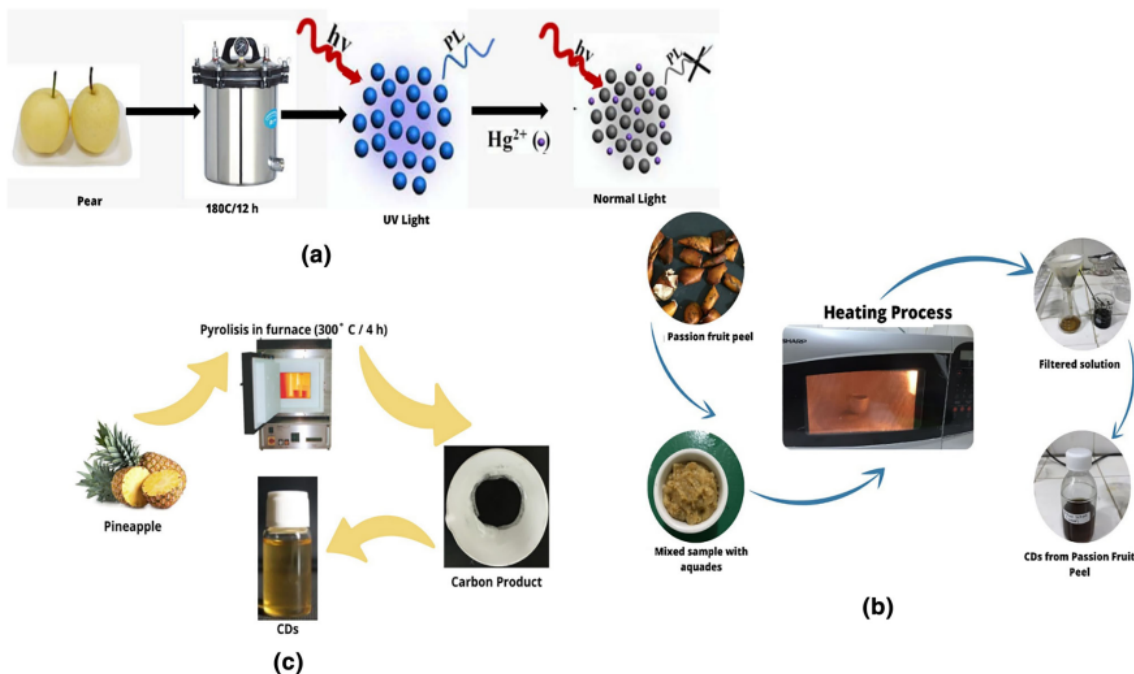
Zulfajri et al has synthesized CDs from cranberry seeds by the pyrolysis method as a carbon precursor at 200 for 18 h and produced size of 3.96 nm with QY of 10.85%, detection limit 9.55 μM and a linear size range of 30 μM to 600 μM.<sup>[58]</sup> The research conducted by Wang et al has also synthesized CDs by the hydrothermal method at 120°C for 4 h with a 34 molecule.<sup>[52]</sup> Raveendran et al has synthesized mint leaves as a precursor for CDs by the hydrothermal method for sensing iron ion (Fe<sup>3+</sup>) with a detection limit of 79 nm.<sup>[59]</sup> Liu et al reported rose-heart radish contains amino acids, sugars, proteins and vitamins for producing a lot of carbon, nitrogen, and oxygen components in CDs with high fluorescence QY 13.6%.<sup>[60]</sup> Figure 3 shows the schematic of the hydrothermal method applied for detection of mercury ion (Hg<sup>2+</sup>). More details can be seen in Table 2 for the synthesis of CDs with various precursors, applications, size of CDs, QY, conditions, linear range, and limit of detection.

### 3.2 | Microwave-assisted method of CDs

For microwave energy to convert into heat energy medium electromagnetic radiation is required to produce heat.<sup>[78]</sup> Microwave radiation offers short and rapid heating, and it significantly improves productivity and properties of CDs.<sup>[27]</sup> Usually, the sample is dissolved in a solvent and then heated in a microwave chamber, the CDs that are formed are separated and filtered.<sup>[79]</sup> Owing to its effectiveness and simple operation, the microwave-assisted method is a low-cost and robust approach with strategic advantage to provide many fluorescent CDs.<sup>[80]</sup> A schematic illustration for CDs from passion fruit peel using the microwave-assisted technique is shown in Figure 3 with the result of homogeneity size of CDs.<sup>[81]</sup> Furthermore, most ordinary microwave appliances can transmit electromagnetic radiation at 2.45 GHz to keep away from impedance with media transmission devices.<sup>[18]</sup> Microwave irradiation of organic compounds is a fast and inexpensive method for synthesizing CDs using natural resources for green luminescent CDs.<sup>[23]</sup> Apart from the most striking benefits of the microwave-assisted method, the short usage time and temperature control make it a preferred method by researchers.<sup>[82]</sup> Generally, the microwave method can provide heating and considerably reduce the reaction time, so that the sub-atomic size of the CDs is homogeneity.<sup>[30]</sup> The microwave synthesis method of CDs is used for various types of precursors, particle size, preparation condition, limit of detection, and linear range as shown in Table 3. The particle size obtained is one of its fundamental properties<sup>[90]</sup> which affect the physical and optical properties. For the CDs mixture, the polarity interaction between the precursor and the solvent has an influence on the hydrophilic, hydrophobic, or amphiphilic properties and shows the different

TABLE 1 The advantages and disadvantages of hydrothermal, microwave-assisted, and pyrolysis in synthesizing carbon dots

Methods	Advantages	Disadvantages	References
<p>Hydrothermal</p>  <p>(CA) + (PEI) <math>\xrightarrow{\text{Hydrothermal}}</math> passivated C-dots</p>	<p>Cheap, non-harmful, simple, controlled reaction, the plentiful and inexpensive precursors</p>	<p>Poor size control, long synthesis duration, high Temperatures, and pressures</p>	<p>[1,29–35]</p>
<p>Microwave-assisted</p>  <p>microwave <math>\rightarrow</math> CDs w/ Urea • CA • F</p>	<p>Easy, fast, versatile, minimal cost, eco-accommodating and efficient</p>	<p>size that is not easy to control</p>	
<p>Pyrolysis</p>  <p><chem>NC(=O)N</chem> <math>\xrightarrow{\text{Pyrolysis } 180^\circ\text{C, 30 min.}}</math> C-Dot</p> <p>Sago Waste <math>\xrightarrow{\text{Thermal Pyrolysis (Different Temperatures)}}</math> Carbonized Product <math>\xrightarrow{\text{Dispersion in Ultrapure Water}}</math> Fluorescing C-dots Sample</p>	<p>simple, efficient synthesis time, eco-friendly</p>	<p>large size and difficult to increase</p>	



**FIGURE 3** (a) Preparation of CDs and application for  $\text{Hg}^{2+}$  detection<sup>[54]</sup>; (b) schematic illustration of preparation of CDs from passion fruit peel by microwave assisted method<sup>[88]</sup>; (c) schematic of the synthesis of pineapple CDs using the pyrolysis method<sup>[92]</sup>

types of carbon.<sup>[18]</sup> Gu et al studied lotus root by the microwave method with QY of 19.0%<sup>[82]</sup> and Gul et al used banana peels at 80°C for 12 h and applied it as a colorimetric sensor for hydrogen peroxide ( $\text{H}_2\text{O}_2$ ).<sup>[83]</sup> Ramezani et al reported the use of quince fruit for CDs by the microwave method and successfully applied as an arsenic ion ( $\text{As}^{3+}$ ) sensor.<sup>[84]</sup>

### 3.3 | CDs pyrolysis method

Pyrolysis is the simplest method for the synthesis of CDs by heating to produce carbon black which is then separated and purified.<sup>[79]</sup> This is known as a bottom-up method.<sup>[19]</sup> Generally, pyrolysis occurs under pressure and high temperatures that is easily controlled<sup>[77]</sup> to produce uniform sized organic precursors (see Figure 3) and the corresponding results for various precursors are presented in Table 4. There are three stages of the pyrolysis method used as follows: (i) absorbing energy from the heat of natural precursors, (ii) carbonaceous materials, (iii) the release of synthesized CDs.<sup>[23]</sup>

Generally, pyrolysis produces carbonaceous material which can be separated and purified to obtain CDs.<sup>[29]</sup> In 2019, Jiao et al reported CDs by the pyrolysis method using mango peel as carbon precursor at 300°C for 2 h with a QY of 8.5%, detection limit 1.2  $\mu\text{M}$  and linear size range of 4  $\mu\text{M}$  to 16  $\mu\text{M}$ .<sup>[91]</sup> A similar report by Ma et al used peanut shells for sensor of copper ion ( $\text{Cu}^{2+}$ ) with a QY of 10.58%.<sup>[92]</sup> Tan et al reported the effect of pyrolysis conditions on the physical, molecular size, and fluorescence strength prepared from

sago waste with low cytotoxicity for cells and could subsequently be used for cell imaging.<sup>[35]</sup>

## 4 | APPLICATION OF SENSOR

The CDs can easily distinguished impurities attached on the surface due to the ability to fluorescence in various colours. The fluorescence CDs display promising optical properties due to wide excitation spectrum, easy adjustability, and high photostability.<sup>[98]</sup>

CDs has been widely applied as a nano sensor due to their capacity to recognize particles and atoms with very high selectivity as a sensor environment as shown in Figure 4. The fluorescence for sensing depends on the type of CDs: graphene, carbon, and polymer, the size of CDs, and the concentration of metals ion in the environment as clearly shown in Figure 4. Some spectroscopy is also used to detect and to measure concentration of metals ion including atomic absorption and emission and auger electrons, however, complicated sample planning and expensive instrumentation is required. Therefore, CDs is a new solution by use of appropriate precursors<sup>[2]</sup> with detection ranges from many molar to a few nanomolar.<sup>[28]</sup>

### 4.1 | Fluorescence sensor

Recently, different test groups have created the fluorescence-based sensor to detect certain metal particles which is very sensitive to

**TABLE 2** Hydrothermal synthesis method of carbon dots (CDs) for various type of precursor, size, preparation condition, limit of detection, and linear range

Precursor	Application	Size	Quantum yield	Preparation condition	Linear range	Limit of detection	References
Lemon peel	Sensing	1–3	~14%	200°C/12 h	2.5–50 $\mu$ M	~73 nM	[47]
Juice pear	Detection of $\text{Cu}^{2+}$	10	–	150°C/2 h	–	0.1 mg/L	[48]
Sweet potato	$\text{Fe}^{3+}$ sensing	3.49	8.64%	180°C/8 h	1 to 100 $\mu$ M	0.32 $\mu$ M	[49]
Banana juice	Copper(II) detection	1.27	32%	150°C/4 h	1–800 $\mu$ g/ml	0.3 $\mu$ g/ml	[50]
Watermelon juice	Detection ion $\text{Fe}^{3+}$	–	10.6%	180°C/3 h	–	0.16 $\mu$ M	[51]
Orange peel	Sensor chromium(VI)	2.34	14%	120°C for 4 h	92.09% to 104.87%	10 nM	[52]
Coffee bean	$\text{Fe}^{3+}$ detection	4.6	5.56%	180°C/12 h	0–0.10 mM	15.4 and 16.3 nM	[53]
Apple juice	Detection of $\text{Hg}^{2+}$	2.8 nm	6.4%	180°C/12 h	5.0 to 100.0 nM	2.3 nM	[54]
bamboo leaves	Detection $\text{Cu}^{2+}$	3.6 nm	7.1%	200°C/6 h	0.333 to 66.6 M	115 nM	[55]
Lemon	Detection $\text{Mo}^{6+}$	–	1.33%	150 to 280°C/12 h	–	20 ppm	[56]
Strawberry	Fluorescent for $\text{Hg}^{2+}$	5.2 nm	6.3%	180°C/12 h	10 nM to 50 $\mu$ M	3 nM	[57]
Cranberry beans	Sensor for selective detection of $\text{Fe}^{3+}$	3.96	10.85%	200°C/18 h	9.55 $\mu$ M	30–600 $\mu$ M	[58]
Mint leaf	$\text{Fe}^{3+}$ detection	4 nm	7.64%	200°C/5 h	0.9984 $\mu$ M	79 nM	[59]
Rose heart	$\text{Fe}^{3+}$ detection	1.2–6	13.6%	180°C/3 h	0.02 to 40 M	0.13 M	[60]
Onion waste	Detection $\text{Fe}^{3+}$	7	28%	120°C/15 h	0–20 $\mu$ M	0.31 $\mu$ M	[61]
Garlic	Detection $\text{Fe}^{3+}$	–	10.5%	200°C/6 h	1–3 nM	0.2 $\mu$ M	[62]
Lentil	Colorimetric sensor	7 $\pm$ 4 $\mu$ m	–	220°C/7 h	5–150 $\mu$ g/L	3 $\mu$ g/L	[63]
Banana plant	Nanosensor	2.5	48%	–	–	6.4 nm	[64]
Greengage plums	Detection pesticide	–	48%	180°C/7 h	–	10 <sup>-8</sup> ppm	[65]
Leek	Peptidases sensor for dichlorvos	2–3.3	–	180°C/4 h	10 <sup>-9</sup> –10 <sup>-3</sup> M	5 $\times$ 10 <sup>-10</sup> M	[66]
Waste tea	Detection of $\text{CrO}_4^{2-}$ , $\text{Fe}^{3+}$	10	–	150°C/6 h	–	–	[67]
Tulsi leaves	Detection chromium(VI) sensors	5 nm	3.06%	240°C/4 h	1.6 $\mu$ M to 50 $\mu$ M	4.5 ppb	[68]
Lycii Fructus	Colorimetric sensor	2–5 nm	17.2%	200°C/5 h	0–30 $\mu$ M	21 nM	[69]
Cherry tomato	Detection of $\text{Cu}^{2+}$ and pesticide quinalphos	2–3 nm	26%	100°C/5 h	0–16 $\mu$ M	0.3 nm	[70]
Apple juice	Detection of acetylcholinesterase pesticides	3.5 nm	–	180°C/12 h	0–2.60 nM	0.05 nm	[71]
cucumber, orange, and strawberries	Detection of pesticide flumioxazin	4–8 nm	12%	140/150 minute	0–35.4 $\mu$ g/L	0.027 $\mu$ g/L	[72]
Carrot, orange and pepper	Sensor pesticide	0.49–2.24 nm	–	160/1 h, 180/3 h, 200/5 h	–	0.05 $\mu$ g/L	[73]
cauliflower juice	Detection of pesticides	1.54 nm	43%	120/5 h	–	0.25, 0.5, and 2 ng/ml	[74]
Lycii Fructus	Colorimetric detection	–	–	180°C/5 h	0.1–100 $\mu$ M	0.04 $\mu$ M	[75]
Carnation flower	Colorimetric sensing pH	6–9 nm	11.36%	–	–	10.6–5	[76]
Reed	Electrochemical sensor for hydrazine determination	2.7 nm	–	180°C/3 h	0.99 $\mu$ M to 5,903 $\mu$ M	0.024 $\mu$ M	[77]

**TABLE 3** Microwave synthesis method of carbon dots (CDs) for various type of precursor, size, preparation condition, limit of detection, and linear range

Precursor	Application	Size	Quantum yield	Preparation condition	Linear range	Limit of detection	References
Lotus root	Hg <sup>2+</sup> ions detection	9.41	19.0%	–	0.1 to 60.0 μM	18.7 nM	[82]
Banana peel	Colorimetric sensor for H <sub>2</sub> O <sub>2</sub>	1.48	–	80°C/12 h	0.1–100 μM	9 nM and 10 nM	[83]
Quince fruit	As <sup>3+</sup> determination	4.85 ± 0.07	8.55%	220°C/1 min	0.1 to 2 μg/mL	–	[84]
Banana peel	Electrochemical sensor for target DNA determination	5	–	90°C/1 h	5.0 × 10 <sup>-16</sup> to 1.0 × 10 <sup>-10</sup> mol/L	1.82 × 10 <sup>-17</sup> mol/L	[85]
Jackfruit seeds	Sensor for Au <sup>3+</sup> determination	5	17.91%	37°C/48 h	–	239 nM	[86]
Wool	Pesticide sensor for glyphosate detection	2.8	16.3%	200°C/1 h	0.025–2.5 μg/ml	12 μg/ml	[87]
Kelp	Detection of cobalt(II) ions	3.7	23.5%	200°C/10 min	1–200 μmol/L	0.39 μmol/L	[88]
Vegetable	Detection of paraoxon-ethyl pesticide	–	–	80°C/30 min	5.80 mM	0.22 ± 0.02 μM	[89]

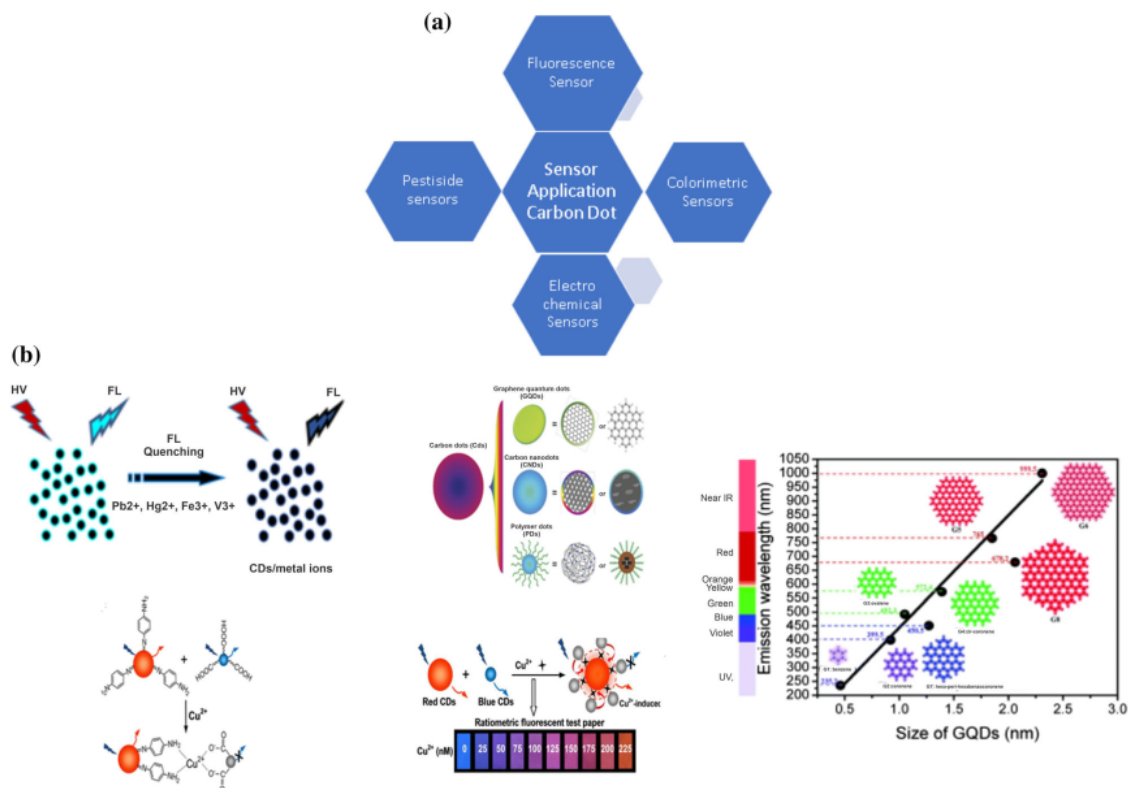
**TABLE 4** Pyrolysis synthesis method of carbon dots (CDs) for various type of precursor, size, preparation condition, limit of detection, and linear range

Precursor	Application	Size	Quantum yield	Preparation condition	Linear range	Limit of detection	References
Mango peel	Fluorescent sensor for Fe <sup>2+</sup>	2–6	8.5%	300°C/2 h	4 μM–16 μM	1.2 μM	[91]
Peanut shell	Fluorescent sensor for Cu <sup>2+</sup> determination	1.8–4.2	10.58%	300°C/4 h	–	4.8 mM	[92]
Sago waste	Metal ions sensing	10 μm to 15 μm	–	250°C to 450°C/1 h	–	7.49 μM and 7.78 μM	[35]
<i>Carica papaya</i> waste	Sensor for chromium	<10	23.7%	60°C/24 h	0.708 ppb and 2.4 ppb	0.708 ppb	[93]
<i>Borassus flabellifer</i> flowers	Sensor for Fe <sup>3+</sup> determination	3–8	13.97%	300°C/2 h	0 to 30 nM	10 nM	[94]
Purple boom	Sensor Br <sub>2</sub> detection	–	18%	300°C/2 h	0.95–13.3 μM	4.90 nM	[95]
Lychee seed	Colorimetric detection	1.12	10.6%	300°C/2 h	0–1000 μg/ml	50 mM	[96]
Plant leaf	Detection Fe <sup>3+</sup>	3.7 nm	–	250°C to 450°C/1 h	0–100 μM	–	[97]

oxygen interaction on the outer layer of the CDs with metal ions and forming complexes as can be seen in Figure 4. CDs can easily be used as a fluorescent test to distinguish metal ions, such as CDs from peanut shells.<sup>[92]</sup> The CDs surface has important parameters to increase luminescence intensity by modification during or after synthesis. The surface state can help in increasing the effect and selectivity of CDs sensor for inorganic metal ions, the optical absorption for optical sensors of heavy metals by colorimetry, absorbance, and PL.<sup>[101]</sup> Heavy metals, which are metallic elements with a much higher density than water, have raised global ecological concerns since they cause

environmental pollution and there has been an increase in the elements in various industries including agriculture. Environmental pollution around mines, foundries, smelters, and others are also a major concern for production plants.<sup>[102]</sup> CDs have been reported by biochemical design for application sensor of Cu<sup>2+</sup>, Fe<sup>3+</sup>, Hg<sup>2+</sup>, Mo<sup>6+</sup>, and Au<sup>3+</sup> ions with highly sensitive and selective reactions.<sup>[56]</sup>

The iron metal is used in daily life because of its fundamental capacity in oxygen transport, oxygen digestion, and electronic exchange.<sup>[83]</sup> However, iron has a great impact on the structure of life showing detrimental consequences for cells and tissues and



**FIGURE 4** (A) Application of carbon dots as an environmental sensor. (B) The dependence of size to the emission wavelength, type of CDs from graphene, carbon, and polymer are included.<sup>[99]</sup> The surface state is important for sensor application by attaching the molecule and view of fluorescence depends on the concentration of metals ions<sup>[100]</sup>

forming open oxidative intermediates.<sup>[38]</sup> Iron is the fourth most plentiful component in the world and is a micronutrient in the human body that conveys oxygen and structures of the haemoglobin, myoglobin, and enzymes. Natural waters contain variable measures of iron depending upon the topographical region and other synthetic parts of the stream. The Fe<sup>2+</sup> and Fe<sup>3+</sup> particles are considered, and consent of scientist related to the oceanic climate. In groundwater iron is ordinarily present as a solvent Fe<sup>2+</sup>. It seems, high Fe<sup>2+</sup> contents lead to more potential persistent infections.<sup>[10]</sup> The research on Fe<sup>3+</sup> detection has been carried out by Murugan and Sundramoorthy, they reported that the CDs from *Borassus flabellifer* flowers by the pyrolysis method with a linear range of 0 to 30 nm and detection limit of 10 nm.<sup>[94]</sup> Green synthesis research of CDs for sensor of Fe<sup>3+</sup> was reported from watermelon,<sup>[51]</sup> coffee beans,<sup>[53]</sup> cranberry beans,<sup>[58]</sup> rose heart,<sup>[59]</sup> and tea waste.<sup>[67]</sup> It is important to improve the efficient detection of Fe<sup>3+</sup> as this is very important for the environment.<sup>[61]</sup> Sun et al have used garlic in producing CDs which contain nitrogen in the formation of C-N and C=N which can be used as a sensor for Fe<sup>3+</sup> with a detection limit of 0.2 M.<sup>[62]</sup> Shen et al found that CDs by the hydrothermal method using sweet potato could be fluorescent sensors to detect Fe<sup>3+</sup> in biological systems,

The CDs preparation was distinguished by Fe<sup>3+</sup> with a detection limit of 0.3 M and a linear range of 1–100 M. The scientists recommend that the fluorescence quenching CDs in detection of Fe<sup>3+</sup> are ascribed the electronic construction when hydroxyl clusters on the surface interact with iron particles.<sup>[49]</sup>

Among heavy metals, copper is the third most abundant heavy metal and has an important position in physiological, dioxygen transport, initiation, signal transduction, and energy.<sup>[49]</sup> Chromium contamination in water sources starts from industrial waste during operations, such as electroplating dyes and leather tanning.<sup>[93]</sup> Purbia and Paria showed that the Cu<sup>2+</sup> used in the research acts as a fluorescence indicator, interacting with the C-point (forming a charge transfer complex) and dampening the emission intensity, because of the non-radiative exchange of electrons from CDs to the orbitals of Cu<sup>2+</sup>.<sup>[90]</sup> Das et al studied sensitivity and showed that there should be a relationship between fluorescence quenching and Cu<sup>2+</sup> fixation that can be selected for quantitative detection of Cu<sup>2+</sup> (0–300 M).<sup>[21]</sup> Other research on metal ion sensing has been carried out, research by Chaudhary et al in 2020 showed a multipurpose green synthesis from banana juice and when applied for the detection of copper(II) the limit of detection was 0.3 g/ml.<sup>[50]</sup> Detection of Cu<sup>2+</sup> from CDs has been

widely studied from various plants such as: pear juice,<sup>[48]</sup> bamboo leaves,<sup>[54]</sup> and peanut shell.<sup>[92]</sup>

For fast and sensitive detection of molybdenum ion ( $\text{Mo}^{6+}$ ) with a simple and natural technique at a low cost is crucial.<sup>[55]</sup> In 2017, Hoan et al reported CDs from lemon to identify  $\text{Mo}^{6+}$  QY of 1.33% with high sensitivity and detection limit of 20 ppm.<sup>[56]</sup> Another heavy metal is  $\text{Hg}^{2+}$  which is toxic to marine biological systems and contamination through drinking water and food can cause chronic diseases in the human body. Several researchers are conducting research to distinguish mercury particles by using CDs which will remove mercury metal particles in the environment.<sup>[103]</sup> In 2017, Gu et al reported nitrogen-doped CDs (N-CDs) from lotus roots as carbon precursors for the identification of  $\text{Hg}^{2+}$  particles by a detecting component.<sup>[82]</sup> The squeezed apple as a carbon source of CDs was reported for  $\text{Hg}^{2+}$  sensors with a detection limit of 2.3 nm from Ref. [52] and similar research from highland barley in Ref. [102].

The gold ion ( $\text{Au}^{3+}$ ), which in recent years has become a topic of concern for research in both the biological sciences and chemical sciences. Generally, it has been used in gold plating, environmental testing, anticancer, nanomaterials, catalysts, organic sensors, and drug transport systems depending on the nature of the compound, unique properties and high biocompatibility that can be used as a medium to maintain environmental ecosystems from natural materials such as CDs.<sup>[38]</sup> Raji et al has reported natural green advances for the synthesis of fluorescent CDs from jackfruit seeds by the microwave-assisted method at 37°C for 48 h. In addition, the method used offers an opportunity for the large-scale synthesis of fluorescent CDs. The obtained CDs have a spherical morphology with a normal molecular size of about 5 nm, excellent water solubility, high quantum yield of 17.91% as well as fantastic stability, and large capacity to withstand pH, ionic strength, and photobleaching, which are natural applications being widely used showing low cytotoxicity and phenomenal biocompatibility of pre-regulated CDs. CDs were successfully applied for  $\text{Au}^{3+}$  sensing with detection limit of 239 nm and multicolour live cell imaging.<sup>[86]</sup>

## 4.2 | Colorimetric sensors

Recently, chemical and colorimetric sensors have received attention due to the capacity to distinguish various bacteria present in the environment with high accuracy<sup>[35]</sup> and low cost.<sup>[63]</sup> The colorimetric sensor exhibits potential to identify large numbers of microorganisms in the environment with easily to construct paper-based colorimetric sensors for evidence of microorganism identification.<sup>[25]</sup> Colorimetric sensors are simple devices to qualify and to evaluate by observing colour chromaticity. They have drawn a large amount of interest due to low costs, quick investigation, and great results.<sup>[17]</sup>

Amjadi et al has reported CDs from lentils by the hydrothermal method for a thioridazine hydrochloride colorimetric sensor and shows shadow red because of the limits of thioridazine hydrochloride to the N-CD/silver nanoparticle. They showed that the colorimetric is a simple and sensitive sensor for the detection of thioridazine hydrochloride in

human serum samples with a detection limit of 3 g/L.<sup>[63]</sup> Sun et al synthesized CDs from Lycii Fructus by hydrothermal method at 200°C for 5 h and showed selectivity fluorescent assay to detect  $\text{Fe}^{3+}$  successfully.<sup>[69]</sup> The CDs have low cytotoxicity and high biocompatibility, effectively applied for multicolour cell imaging and detection of  $\text{Fe}^{3+}$  intracellular.<sup>[63]</sup> Zhang et al has conducted research on CDs from seaweed as a sensor colorimetry with fluorimetry for detection of cobalt ion ( $\text{Co}^{2+}$ ), the solution undergoes a colour change that can be seen clearly with the naked eye under visible light and UV light.<sup>[88]</sup>

## 4.3 | Electrochemical sensors

The electrochemical sensor works based on electrodes which are modified by the development of a catalyst for purpose to reduce nitrite in the reaction.<sup>[26]</sup> Li et al reported CDs from kiwi seeds, white sesame seeds, and black sesame seeds prepared by the hydrothermal method for nitrite sensors and shows good sensitivity. For nitrite detection, the CDs have been shown to have an increase in electrocatalytic properties of 0.23 M, high correlation with increasing electrical conductivity on the CDs surface. Moreover, the CDs-based nitrite sensor exhibits a low detection limit but may in the future use many biomass materials that can be used for the synthesis of CDs as a new electrochemical sensor for food material detection.<sup>[105]</sup> Huang et al reported CDs from banana peels in the form of Pd-Au@CDs/GCE for comparison sensing of DNA via mounted on the outer layer by the carboxyl group at the surface of CDs. The largest linearity of the target DNA was achieved by clustering  $5 \times 10^{-16}$  to  $1 \times 10^{-10}$  mol/L.<sup>[85]</sup>

## 4.4 | Pesticide sensors

Pesticides are usually used for expanding crop yields and to improve the quality of food sources. However, misuse of pesticides causes potential hazardous environmental damage and food safety problems.<sup>[106]</sup> In most cases, pesticides are used as chemical control agents or to eradicate pests and diseases of plants or animals. Pesticides can be herbicides, insect sprays, fungicides, or other types depending on their respective targets, they include special chemical compounds, and the most well-known are arsenic, carbamates, organophosphates, pyrethroids, and nitrophenol derivatives. Pesticides can be grouped based on their biological structure and chemical structure.<sup>[107]</sup> Organophosphate pesticides have been widely used in horticulture due to their ability to prevent, to control or to kill the dreaded crawling insects, increasing crop yields and vegetable production.<sup>[27]</sup> The detected organophosphate method is a combination of gas and liquid chromatography with various detectors.<sup>[108]</sup> Currently, an environment with even low pesticide has effect on human health. Therefore, it is important to detect pesticides, several methods have been reported including gas chromatography (GC), electron capture-site gas chromatography (ECD-GC), gas chromatography-mass spectrometry (GC-MS), high-pressure liquid chromatography (HPLC), and high-

pressure liquid chromatography–mass spectrometry (HPLC-MS). However, these methods are expensive, and the detection time is slow and difficult when distinguishing for different types of pesticides. A new material is needed with high efficiency,<sup>[64]</sup> high photostability, low hazard level, and good biocompatibility such as CDs working with optical sensor systems for the detection of pesticides.<sup>[17]</sup>


The development and sensitivity for OP identification is becoming urgent to protect the environment, screen food quality, measure pesticide hazards, and maintain the general welfare. Typically, OPs have been identified by gas and liquid chromatography, mass spectrometry, measurement of immunosorbent-associated compounds (ELISA) and electrochemical investigations.<sup>[27]</sup> Wang et al reported CDs from eggs for sensors organophosphate pesticides with detection limit of 4.48  $\mu\text{M}$ .<sup>[109]</sup> Korram et al reported a sensor for acetylcholinesterase peptide from apple juice with a detection limit of 0.05  $\text{nm}$ .<sup>[71]</sup> In recent years, abundant nanomaterials have been created and applied in agribusiness to screen for plant health, to promote crop development, to work on the capabilities of fertilizers and pesticides and to control infections, insects, and environmental impacts.<sup>[110]</sup>

## 5 | CONCLUSION AND PROSPECTS

In this review, we reported synthesize methods of CDs, modification strategies, fluorescent properties, and the ability for sensors of heavy metals in the environment. Several methods for CDs preparation are discussed including hydrothermal, microwave, and the pyrolysis methods. Natural resources from various plants can be used as precursors for synthesizing CDs. The hydrothermal method was the most reported used owing to the simple operation, environmentally friendly, and low cost. Meanwhile, we also introduced CDs applications for metal ion sensors, colorimetric sensors, electrochemical sensors, and pesticide sensors. In the future, better synthesis methods, and better CDs luminescence properties, easy to produce, and high sensitivity need to be developed for environmental sensors. In addition, a more prominent understanding of the optical properties of CDs is needed and requires more possibility to find better materials. For example, green plums are synthesized as precursors of CDs and used to drive the development of pesticide sensors. There are several challenges for applying CDs as sensors that need to be addressed for further research as follows:

1. Colour fluorescence systems and CDs electrochemical detection components should be investigated further.
2. Need high sensitivity of CDs for various kinds of application. With continuous research of CDs with new synthesis methods for easy tuning luminescence properties and lower detection limit will continue to be developed. In addition, a mixture of different green synthesis methods is also expected to contribute to increasing sensitivity of CDs as an environmental sensor.

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