

# Carbon Nanodots Derived from Natural Products

*by* Mochamad Zakki Fahmi

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# Carbon Nanodots Derived from Natural Products

Yaung Kwee<sup>a</sup>, Alfinda Novi Kristanti<sup>a,\*</sup>, Kaido Siimon<sup>b</sup> , Nanik Siti Aminah<sup>a</sup> and  
 Mochamad Zakki Fahmi<sup>a,c,\*</sup> 


<sup>a</sup>Department of Chemistry, Universitas Airlangga, Surabaya 60115, Indonesia.

<sup>b</sup>Institute of Physics, University of Tartu, Ravila 14c, Tartu, 50411 Estonia.

<sup>c</sup>Supramodification Nano-micro Engineering Research Group, Universitas Airlangga, Surabaya 60115, Indonesia.

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## ABSTRACT

This review highlights recent improvements, and present situations concerning the practical application of, natural products-based carbon nanodots (CNDs) in nanomedicine, sensors, drug delivery, bio-imaging, solar cells, photocatalysis  and nano-encapsulation. CNDs are zero-dimensional carbonaceous nanomaterials that have recently drawn much attention because of their unique physicochemical properties, such as excellent biocompatibility and tunable photoluminescence, easy functionalization, solubility in water, dispersibility, and low toxicity. Additionally, they are environmentally friendly, abundant, easily accessible, and rich in multiple elements. Since CNDs derived from organic products have unique properties, we explored and found that the properties of CNDs may depend on the preparation method and the used precursors. This study also informs on the positioning of natural products in nanotechnology.

## KEYWORDS

Carbon nanodots, natural product, synthesis, applications.

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
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## 1. Introduction

Carbon is one of the most plentiful elements in the earth's crust. Many applicable materials, including various nanomaterials, are carbon-based materials.<sup>1,2</sup> With all the recent

nanotechnology advancements, carbon-based materials have developed from low-cost waste to high-value materials. Adding value is often achieved by using new synthesis methods, which allow controllable and straightforward preparation of nanomaterials.<sup>3,4</sup> Fluorescent carbon-based nanomaterials, like carbon nanodots (CNDs), have received interest due to their

 <sup>39</sup>  
 \* To whom correspondence should be addressed.  
 E-mail: A.N.K., alfinda-n-k@fst.unair.ac.id / M.Z.F., m.zakki.fahmi@fst.unair.ac.id



low cost and low toxicity. Therefore, researchers have put much effort into developing new synthetic approaches to prepare such materials.<sup>5,6</sup>

In 2004, CNs were first explored during the purification of single-walled carbon nanotubes.<sup>7</sup> Subsequently, Sun *et al.*<sup>8</sup> verified the properties of luminescent CNs synthesized by surface passivation and laser ablation in 2006. Since then, research regarding the nature of CNs has been developing fast.<sup>8</sup> In the last few years, substantial progress has been made in synthesizing CNs, characterizing their properties, and developing their potential applications.<sup>9–22</sup> However, the sources of carbon and its importance have been largely neglected.<sup>23</sup>

There are abundant organic products in our environment that are good sources of carbon for the synthesis of organic products-based CNs.<sup>24,25</sup> These CNs are green and eco-friendly, in contrast with other man-made carbon dots. Man-made carbon dots are prepared from precursors containing some toxic material, and oxidation of some chemical agents.<sup>26</sup> Additionally, organic products as sources of CNs are advantageous in many ways, for example, being inexpensive,<sup>27</sup> easy to obtain,<sup>28</sup> eco-friendly, relatively nontoxic<sup>29</sup> and abundant in nature.<sup>30</sup> Furthermore, it is often possible for CNs to produce valuable and useful materials from low-value organic waste.<sup>31</sup> Some of the other useful properties of CNs are: tuneable fluorescence, nontoxicity, sustainability, small size distribution, good biocompatibility, excellent photostability, electrochemiluminescence and multi-photon excitation (up-conversion). These properties make them potentially useful in applications for biomedicines,<sup>32</sup> sensors,<sup>33,34</sup> optronic<sup>35</sup> and photo-catalysis.<sup>36</sup> Furthermore, the as-prepared CNs can be functionalized using biomolecules, enhancing their optical properties, optimizing and making them chemically inert to be used for some intended therapy.<sup>37</sup> These modified CNs are also used for sensors, optronics as catalytic materials, and effective carriers in drug delivery applications and biological imaging.<sup>25,38</sup>

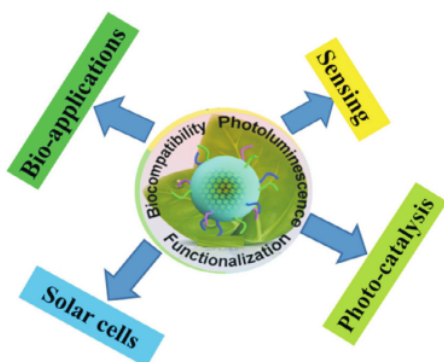
Mostly, in the comparison of conventional semiconductor quantum dots, organic dyes, and up-converting nanoparticles, photoluminescent organic products derived-CNds have several advantages which include nontoxicity, eco-friendliness, the ability to use cost-effective synthetic methods, high solubility in water, durable photostability, robust chemical inertness, and ease of modification.<sup>39</sup> Synthesis of CNs can be carried out either by 'top-down' approaches like laser ablation,<sup>40</sup> electrochemical preparation methods<sup>41</sup> and oxidation,<sup>42</sup> or 'bottom-up' approaches like hydrothermal,<sup>43</sup> or microwave treatment,<sup>44</sup> pyrolysis,<sup>45</sup> chemical oxidation,<sup>42</sup> and solvothermal<sup>46</sup> approaches. CNs have been derived from various organic sources<sup>21</sup> such as organic compounds,<sup>47</sup> bamboo leaves,<sup>48</sup> shrimp shellwaste,<sup>49</sup> plant extracts,<sup>50</sup> fruit shells<sup>51</sup> and peels,<sup>52,53</sup> fruit juice,<sup>54,55</sup> algae<sup>56,57</sup> and eggshells,<sup>58,59</sup> depending on the specific applications. The produced extract is mostly carbonized to become carbon black, turning into CNs after dispersion, separation, and purification steps.<sup>60</sup> Many researchers have reported using CNs derived from natural products for sensing metal ions.<sup>61</sup> Such CNs have been synthesized from quince fruit,<sup>62</sup> mushrooms,<sup>63</sup> cherry blossoms,<sup>64</sup> coffee,<sup>65</sup> sesame<sup>66</sup> and sago starch.<sup>67,68</sup> Additionally, some nanoencapsulation studies of bioactive compounds are also described briefly in this review. Encapsulation can improve the water solubility of bioactive compounds, such as essential oils.<sup>69</sup> Nanoencapsulation also improves bioavailability and health beneficial activities. Meanwhile, working in the nanoscale means an increased surface area, which leads to increased activity.<sup>70</sup> Lee *et al.*<sup>71</sup> have reported

CNs derived from major polysaccharides of *Ocimum basilicum* (basil) seeds. Fahmi *et al.*<sup>72</sup> emphasize the potential application of CNs prepared from waste bamboo leaves and their potential to enhance drug delivery efficacy. In particular, tumour drug doxorubicin was attached to loaded CNs for tumour imaging and therapy.<sup>72</sup> Tang *et al.*<sup>73</sup> also reported water-soluble crystalline CNs derived from glucose, synthesized by a facile microwave-assisted hydrothermal treatment, for potential application in the field of DUV (deep ultraviolet) photonic devices. The as-prepared graphene-like CNs have a diameter of 1.65 nm, and they are less than 5 layers thick. Konwar *et al.*<sup>74</sup> reported CNs derived from tea leaves, including polyphenols such as catechins,<sup>75</sup> tannins,<sup>76</sup> and flavonoids,<sup>77</sup> which showed excellent antioxidant properties. These tea carbon dots were used to prepare a chitosan-tea carbon dots nanocomposite hydrogel film with developed physico-mechanical properties. Recently, Hsu *et al.*<sup>78</sup> prepared CNs from ground coffee using a simple method consisting of four stages: dehydration, polymerization, carbonization, and passivation. Such nanodots had a  $5 \pm 2$  nm average diameter, and their quantum yield was 3.8 % for cell imaging. Dong *et al.*<sup>79</sup> prepared fluorescent CNs and graphene oxide (GO) by changing carbonization capability of an usual organic precursor, namely citric acid. Wang *et al.*<sup>80</sup> also reported using *Enteromorpha prolifera* (EP) as the starting material to probe the radical scavenging of natural phenolic compounds, these novel luminescent CNs were synthesized using a facile microwave-assisted hydrothermal approach. Vandarkuzhali *et al.*<sup>83</sup> prepared the pineapple-peel-derived CNs for use in applications like sensors, molecular keypad lock and memory devices. Fuciños *et al.*<sup>81</sup> have reported caffeine encapsulation into  $\alpha$ -lactalbumin ( $\alpha$ -LA) nanotubes through nanotube synthesis.  $\alpha$ -LA nanotubes were around 100 % successful in caffeine loading.<sup>81</sup>

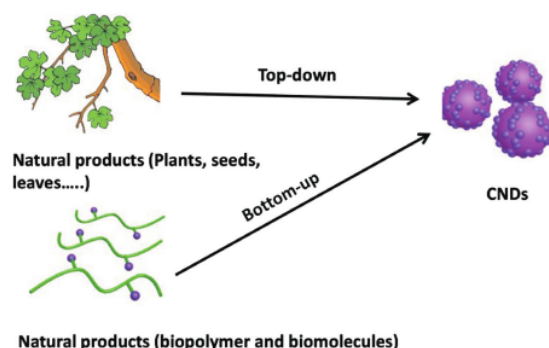
It could be deduced from the respective overviews above that the differently efficient CNs done by many studies have given promising models for various applications. However, information on utilizing organic product-based material for CNs and its application has not been clearly reported as a review. It became an essential factor in promoting other possibilities for further application. It is hoped that the potential findings of the various organic products-based CNs would be profitable because the starting materials are sustainable, eco-friendly, and readily available. This review has collected many examples of current developments of green fluorescent CNs with high potential. CNs are widely prepared by various methods and from various carbon sources<sup>11</sup> (Fig. 1). Hence, this review aims to extensively examine their synthesis, properties and applications in sensing, solar cells, catalysis, bioimaging and drug delivery and nanoencapsulation of bioactive compounds.

## 2. Synthetic Methods

The synthetic routes of CNs were categorized as 'top-down' and 'bottom-up' methods, as shown in Fig. 2. In the case of a 'top-down' method, the method is generally used to prepare carbon nanotube, graphite nanotubes and nanofibres. Moreover, CNs are also derived from natural products, including fruit shells and peels, plant material and animal waste. All of these sources<sup>12</sup> abundant and relatively macroscopic carbon sources.<sup>24,82,83</sup> On the other hand, in the case of a 'bottom-up' approach, small molecules are expanded to the size of CNs, using different kinds of starting material, such as seeds. This method can be used to prepare CNs from small natural polymers, such as chitin and chitosan.<sup>84,85</sup> However, to prepare a small number of CNs from natural products, the two methods mentioned above have been brought together.<sup>86</sup> Nowadays,



**Figure 1** A schematic diagram of CNDs derived from organic products and their common properties and applications. The image is reprinted with permission from reference [22]. Copyright © 2019 Zhengzhou University.



**Figure 2** A schematic diagram of synthetic routes to CNDs. The image is reprinted with the permission of reference. The image is reproduced with permission from reference [88]. Copyright © 2018 Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

various methods for preparing CNDs from organic products have been developed. Most studies aim to obtain high-quality CNDs using simple, fast, size-controllable methods or large-scale and cost-effective synthetic methods.<sup>87</sup> These two widely used methods are discussed in detail below.

### 2.1. Pyrolysis

Pyrolysis is an irreversible thermolysis reaction that consists of chemical decomposition of organic materials in an inert atmosphere. Both physical and chemical changes occur in organic substances due to pyrolysis, resulting in solid carbonaceous residue. Generally, pyrolysis is carried out under controlled pressure and at a very high temperature above 430 °C (800 °F). Some of the organic raw material sources of carbon can be used to synthesize natural product-based CNDs.<sup>89</sup>

**Table 1** CNDs prepared from natural carbon sources by pyrolysis.

Carbon source materials	Method	QY/%	Size/nm	Applications	References
Rice husk	Pyrolysis	3–6	15	Bio-imaging	92
Lychee seed	Pyrolysis	1.12	10.6	Bio-imaging	93
Coffee grounds	Pyrolysis	5	3.8	Bio-imaging	78
Plant leaves	Pyrolysis	3.7	16.4	Detecting Pb <sup>2+</sup>	94
Peanut shell	Pyrolysis	0.4–2.4	9.91	Multi-colourcell imaging	95
Peanut skin	Pyrolysis	10–40	7	Bio-imaging	96
Bamboo leaves	Pyrolysis	–	2	Imaging and therapy	72

Sun *et al.*<sup>90</sup> have prepared a novel enzyme-free fluorescent sensor to probe catechins in green tea, using bifunctional graphene quantum dots (GQDs) as the starting materials. The GQDs were used as peroxidase-like catalysts and fluorescence probes. The GQDs synthesized by pyrolysis were purified, and then their solution was added into the catechin group compound, which was obtained from tea extract. It was found that an enzyme-free fluorescent nanoplatform was usable to detect catechins, and the detection limit was 5.0 nM (Fig. 3a).

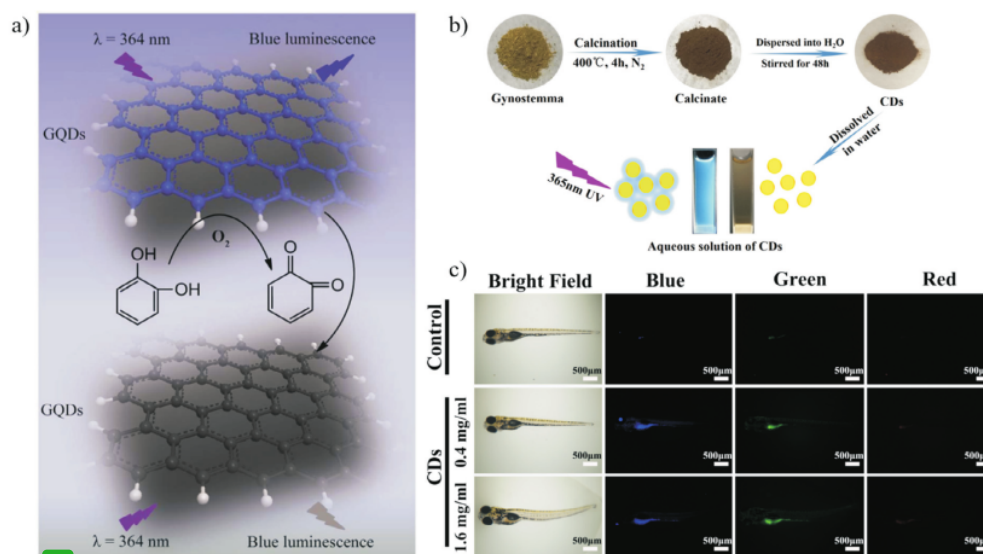
In another study by Wei *et al.*,<sup>91</sup> fluorescent CNDs were synthesized through pyrolysis and direct calcination of *gynostemma* for bioimaging applications. The as-prepared CNDs solution was pale yellow under natural light, whereas it exhibited strong blue fluorescence in 365 nm UV light (Fig. 3b). Their mean size ranged around 2.5 nm. The emitted wavelength has moved to redshift depending on excitation wavelengths (Fig. 3c). Moreover, these CNDs also demonstrated good fluorescence stability even under high ionic strength conditions. Owing to the biocompatibility and excellent fluorescence stability of CNDs, bioimaging observations could be performed in zebrafish. The CNDs can enter zebrafish embryos *via* chorion or mouth. Thus, the fluorescent CNDs could potentially be used in bioimaging in medicinal applications. Moreover, many published articles regarding CNDs prepared from natural carbon sources through pyrolysis for various applications. Their specific information is summarized and presented in Table 1.

### 2.2. Extraction

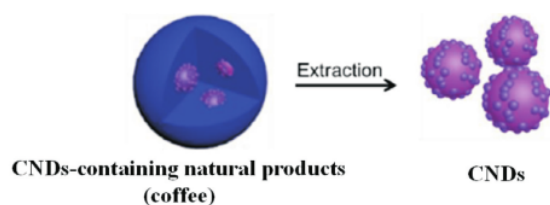
Some organic products-based CNDs can be prepared simply by extraction. Jiang *et al.*<sup>97</sup> prepared the extraction of CNDs directly from instant coffee. In a typically simple process, the prepared coffee powder was mixed with purified water at a heating rate of 90 °C, and the amalgam was subjected to vigorous stirring and then centrifuged. The unsinkable phase was sieved using a membrane to separate sizeable particles. Sephadex G-25 gel filtration chromatography was used to further purify the CNDs, using water as the eluent. Obtaining CNDs using this method is uncomplicated and straightforward. The optimal CNDs demonstrated good biocompatibility and were useful for bioimaging applications in cells and fish (Fig. 4).

In another study, Singh *et al.* 2018<sup>98</sup> prepared fluorescent CNDs from beetroot raw materials using an aqueous extraction method, for *in vivo* live imaging in *C. elegans* and BALB/c mice. The aqueous beetroot extracts displayed blue-emitting CNDs by heating temperature (160 °C) and green-emitting CNDs by oxidation with H<sub>2</sub>PO<sub>4</sub>. Their findings demonstrated that the green-emitting CNDs showed a significant optical response in the intestinal part in live BALB/c mice cleared from the body through faeces, thereby attributing that these green CNDs have potential as diagnostic models in biomedical application.

An additional study by Zhang *et al.*<sup>99</sup> demonstrated that fluorescent CNDs could be fabricated from a polystyrene foam waste through a facile organic solvent extraction method for



**Figure 3** (a) The fluorescence quenching of graphene quantum dots by catechins. The image is reproduced with the permission of the reference [90]. Copyright © 2017 Royal Society of Chemistry. (b) synthesis of CNDs from *Gynostemma* by calcinations, (c) fluorescence photographs of 5 dpf zebrafish incubated in CNDs solution. Images of (b) and (c) are reproduced with permission of reference [91]. Copyright 2019 American Chemical Society.



**Figure 4** Schematic illustration of CNDs preparation using extraction methods. The image is reprinted with permission from reference [88]. Copyright © 2018 Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

biomedical application. The highly fluorescent CNDs showed very strong fluorescence because of the organic solvent's dispersible ability type. It was deduced from Zhang *et al.*'s findings that the strong photoluminescence, the small size, no toxicity and biocompatibility would contribute to applying these CNDs for bio-imaging and bio-labelling.

### 2.3. Hydrothermal Treatment

Many researchers often use this method because it is a cheap, eco-friendly, and effective technique to synthesize CNDs from diverse organic products. In this method carbon sources, like carbohydrates, were dissolved in water with or without adding some additives (acid or alkali).<sup>100</sup> The prepared solutions were put into an autoclave, heated and kept at a constant heating rate for 7 h. The supernatant, which contains large particles, was separated by centrifugation. Natural products,<sup>101</sup> such as starch, maltose, glucose, sucrose, xylose, amylopectin, saccharose, and cellulose, have often been used as carbon sources to develop efficient organic products-based CNDs for specific applications.<sup>102</sup>

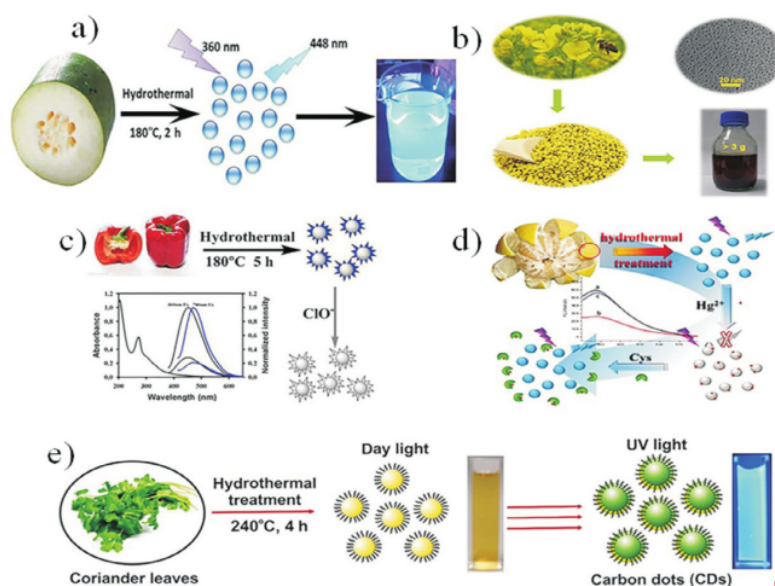
Mewada *et al.*<sup>103</sup> synthesized green CNDs from the Indian water plant, *Trapabispinosa*, peel extract in the absence of any extrinsic oxidizing agents. The size distribution ranged from 5 to 10 nm in the solution, with a leading highly green fluorescence under UV-light ( $\lambda_{\text{ex}} = 365 \text{ nm}$ ). The as-prepared CNDs exhibited

potential fluorescent properties and quantum yield. Thus, the as-synthesized CNDs were promising to be biocompatible in case of Madin–Darby Canine Kidney (MDCK) cells.<sup>103</sup>

Feng *et al.*<sup>103</sup> prepared nitrogen-doped CNDs from edible winter melon as the carbon source, using a facile one-step hydrothermal treatment. It was found that the size distribution diameter of mono-dispersed CNDs was 4.5–5.2 nm. It had 7.51% quantum yield (43). These novel photoluminescent CNDs were successful as bio-imaging agents for hepG2 (liver hepatocellular carcinoma) cells. The graphical abstract of it is presented in Fig. 5a.<sup>104</sup>

Zheng *et al.*<sup>105</sup> published information about synthesizing pollen-CNDs through hydrothermal treatment. The as-synthesized CNDs emitted blue light fluorescence under UV light display excitation, making them potentially useful for tracking the absorption of nitrogen, potassium, and phosphorus and marking the biological carrier platform *Brassica parachinensis* L. They observed that *Brassica parachinensis* L. showed a yield of 42.90 mg during the addition of  $3.5 \text{ mg L}^{-1}$  pollen-CNDs in the nutrient solution. Thus, it would be possible to use the pollen-CNDs at the site of hydroponically grown vegetables. Moreover, those prepared CNDs were biocompatible for imaging of *in vitro/in vivo* studies. The schematic abstract diagram is shown in Fig. 5b.

Yin *et al.*<sup>106</sup> showed that highly efficient CNDs with down- and up-conversion fluorescence could be produced by hydrothermal treatment, using sweet pepper as the carbon source. The quantum yield of CNDs was 19.3%. These CNDs had good photophysical properties, such as narrow and symmetric emission spectra, good photostability against photobleaching and excitation-dependent fluorescence response. The CNDs worked for fluorescent probes to detect hypochlorite ( $\text{ClO}^-$ ) due to the ability to perform both down- and up-conversion fluorescence. For down-conversion measurement,  $0.05 \mu\text{mol L}^{-1}$  of  $\text{ClO}^-$  concentrations were determined as a detection limit, whereas the up-conversion detection of  $\text{ClO}^-$  concentrations had a small observation limit of  $0.06 \mu\text{mol L}^{-1}$  ( $S/N = 3$ ). In this way, it



**Figure 5** Schematic illustration of CNDs prepared by through hydrothermal treatment from organic products, for example, (a) winter melon, (b) bee pollen, (c) sweet pepper pomelo peel, and (e) coriander leaves. Image (a) is reproduced from reference [109]. Copyright © 2015 Royal Society of Chemistry. Image (b) is reproduced with the permission of reference [110]. Copyright © 2015 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. Image (c) is reproduced with the permission of reference [106]. Copyright © 2013 Royal Society of Chemistry. Image (d) is reprinted with the permission of reference [4]. Copyright © 2012 American Chemical Society. Image (e) is reprinted with the permission of reference [108]. Copyright © 2015 Royal Society of Chemistry.

was inferred from the above results that the CNDs prepared by this method are advantageous in anion detection applications due to being simple, sensitive, and dual-signalling models. Additionally, they could also be extensively used in biological and environmental assays (Fig. 5c).

Another report by Lu *et al.*<sup>107</sup> concerning fluorescent organic products-based CNDs, in which fluorescent CNDs were prepared from the cheap waste of pomelo peel as a raw carbon source, using hydrothermal treatment as the preparation method, was published in 2017. The quantum yield was found to be close to 6.9%. The authors further explored that the application of such CNDs was to detect fluorescent  $\text{Hg}^{2+}$ , which is dependent on  $\text{Hg}^{2+}$ -induced luminescent quenching of these CNDs. High level of sensitivity and selectivity toward  $\text{Hg}^{2+}$  was achieved by this sensing method, and the range of detection limit was found to be as low as 0.23 nM. This study also proved that it was promising to use fluorescent CNDs to detect  $\text{Hg}^{2+}$  in lake water samples (Fig. 5d).

Sachdev and Gopinath<sup>108</sup> reported the preparation of CNDs from coriander leaves, through a one-step hydrothermal treatment, for potential use as antioxidants, bioimaging agents, and sensors. According to the antioxidant activity test results, raising the concentration of CNDs from 5 to 70  $\mu\text{g mL}^{-1}$  subsequently increased the scavenging activity from 12 to 94%. Moreover, 2, 2-diphenyl-1-picrylhydrazyl (DPPH) solution, which turned colourless to yellow based on an increase in the concentrations of CNDs, was studied. Additionally, CNDs was assessed for biocompatibility and bioimaging. More than 85% of cell viability was accounted for by L-132 cells at all the tested doses (0.1 to 1  $\text{mg mL}^{-1}$ ) of CNDs.

Meanwhile, the low cytotoxicity of CNDs under *in vitro* conditions strongly attributed to the favourable application of CNDs for bioimaging response. To further describe CNDs synthesized from starting materials of natural products carbon sources, more

examples of published articles regarding CNDs prepared from natural carbon sources using hydrothermal method. Their specific information is summarized and presented in Table 2.

#### 2.4. Chemical Oxidation

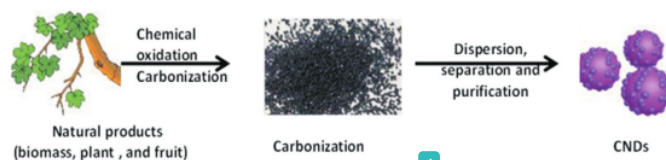
Hydrogen peroxide and oxidizing acids of chemical oxidants are useable as cooperative starting materials to synthesize natural products-based CNDs. Generally, natural products are first carbonized, and then chemical oxidants are added (as shown in Fig. 6). The separation and purification of the obtained CNDs were performed continually.<sup>120</sup>

Chin *et al.*<sup>68</sup> have reported starch nanoparticles prepared from sago starch via chemical oxidation. Raw materials of sago starch are cheap, and starch nanoparticles can be prepared in an eco-friendly way. The obtained CNDs were dehydrated, using concentrated  $\text{H}_2\text{SO}_4$  and simultaneously oxidized with  $\text{HNO}_3$  for 30 min. They discovered that fluorescent CNDs have an excitation-independent, stable wavelength peak emitted at 430 nm when they get excited by distinctly powerful energy photons. Interestingly, they observed that emission wavelength peak did not shift due to particularly homogenous and mono-dispersed physiochemical characteristics. Owing to their simple preparation, shorter reaction time, and effective luminescent emission, these fluorescent CNDs are favourable candidates for diverse biomedical uses in bio-imaging and biosensors.<sup>68</sup>

In 2012, Suryawanshi *et al.*<sup>121</sup> synthesized green CNDs by oxidizing dead neem leaves with  $\text{H}_2\text{SO}_4$ - $\text{HNO}_3$ . The prepared CNDs contained various functional groups, for instance, -COOH and epoxy groups, in which these functional groups favour non-radiative integration of electron-hole pairs in the inherent state. The surface of the synthesized CNDs was modified with amines. Meanwhile, the intrinsic fluorescence could be fine-tuned. The -COOH and epoxy groups were substituted by

**Table 2** Promising CNDs prepared from organic resources as a result of hydrothermal treatment.

Carbon source materials	Synthetic method	QY/%	Size/nm	Application	References
Cocoon silk ions	Hydrothermal	38	70	Bio-imaging detecting	25
Kitchen waste	Hydrothermal	50		Drug delivery	111
Lignin biomass	Hydrothermal	21	2–6	Bio-imaging	112
Willow bark	Hydrothermal	6	1–4	Biosensor photocatalyst	113
Peanut shells	Hydrothermal	3.2	5	Multicolour living cell imaging	95
Eggshell membrane	Microwave Hydrothermal	14	5	Biosensor	114
Humic acid	Hydrothermal	5.2	4	Bio-imaging	115
Bamboo leaves	Hydrothermal	7.1	2–6	Detecting Cu <sup>2+</sup>	116
Goose feathers	Microwave hydrothermal	17.1	21.5	Detecting Fe <sup>3+</sup>	117
Coconut water	Microwave hydrothermal	54	1–6	Detecting Cu <sup>2+</sup>	118
Honey	Hydrothermal	19.8	2	Detecting Fe <sup>3+</sup>	119

**Figure 6** Schematic diagram of CNDs prepared by chemical oxidation. The image is reproduced with the permission of reference [88]. Copyright © 2018 Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

the  $-CONH_2$  and  $-C-NH_2$  groups, subsequently suppressing the non-radiative track. Thus, the fluorescence intensity was stronger because the intrinsic state emission increased. This study offered new insights into CNDs from an investigational and mechanistic perspective and provided methods to prepare multicoloured CNDs by oxidation. Yan *et al.*<sup>122</sup> prepared CNDs from starch and oxidized them chemically for the application of detecting the chemotherapy drug, imatinib, by a novel design depended on quenched fluorescence. The modification of surface properties and fluorescence emission of CNDs was easily achieved by chemical oxidation. However, some problems remain. As the primary reason for using CNDs prepared by chemical oxidation, the CNDs solution could increase the biological toxicity, because remaining chemical oxidation agents' can contaminate the as-prepared CNDs, which is a major drawback this method.<sup>122</sup>

Besides, some natural products-based graphene-like nanodots have also been synthesized with a chemical oxidation/reduction method. For example, Dong *et al.*<sup>123</sup> synthesized CNDs with a chemical reduction pathway from humic substances (HS) without further modification and treatment. HS solid materials were dissolved in 0.1 M (pH 7) phosphate buffer solution (PBS), to measure the fluorescence. The as-prepared graphene nanodots could be promising for the clinical platform around a society *in vivo* or *in vitro* testing system. Most of these nano-sheets had the size range in lattice spacings of 0.220–0.240 nm according to HRTEM characterization, fitting well with graphene. HS samples had physical appearances similar to graphene nano-sheets.

These physical properties were modified with oxygen-containing groups. They concluded that those graphene nanoparticles are intrinsically carbon-based dots, showing

excellent optical properties that included UV-vis (ultraviolet-visible), FL (fluorescence), and ECL (effective conjugated length). Natural carbon-based dots in HS could contribute to new insight into HS. These natural products derived carbon-based dots may bring about potential HS utilization in various fields, such as bio-imaging, bio-medicine, sensing and optoelectronics because of their unique properties.<sup>123</sup> To further investigate, many CNDs examples from natural products-based carbon sources were synthesized by using chemical oxidation for biomedical applications provided by Table 3.

## 2.5. Microwave-assisted Synthesis Method

The microwave-assisted process is regarded as one of the most effective and feasible methods to synthesize promising CNDs. Advantageously, this method allows CNDs to be synthesized very fast by using a proper precursor solution. Time-saving is the leading benefit of using the microwave-assisted method.<sup>127</sup>

Simsek *et al.*<sup>50</sup> prepared carbon nanomaterials from *Nerium oleander* (oleander) leaves through the microwave-assisted method. *Nerium oleander* contains a large number of chemical components like cardiac glycosides, triterpenes, polyphenols, flavonoid, and glycosides. The chemical composition of the natural extracts plays a significant role in synthesizing carbon-based nanodots. The *Nerium oleander* extracts were divided into aqueous extract and ethanol extract. The aqueous extract contains sterols, flavonoids, and terpenoids, whereas these compounds are not contained in ethanol extract. To evaluate the significant result of a surface-passivating agent in the process of CNDs formation, 1 g polyethylene glycol (PEG) 10000N was added into some of the preparations. It was found that the highest emission wavelength peaks of these CNDs<sub>E@PEG</sub> (ethanol extract-passivated PEG-CNDs) and CNDs<sub>W@PEG</sub> (water extract-

**Table 3** CNDs reported from organic resources by chemical oxidation.

Carbon source materials	Synthetic method	QY/%	Size/nm	Application	References
Activated carbon	Chemical oxidation	12.6	4.5	Bio-imaging	124
Cow manure	Chemical oxidation	65	4	Bio-imaging	125
Candle soot	Chemical oxidation	–	1	PL properties	126

passivated PEG-CNDs) were red-shifted. This red-shift of CNDs' photoluminescence (PL) emission could be corresponded to growing surface deformity with an oxygenated surface exhibiting smaller energy levels.<sup>50</sup>

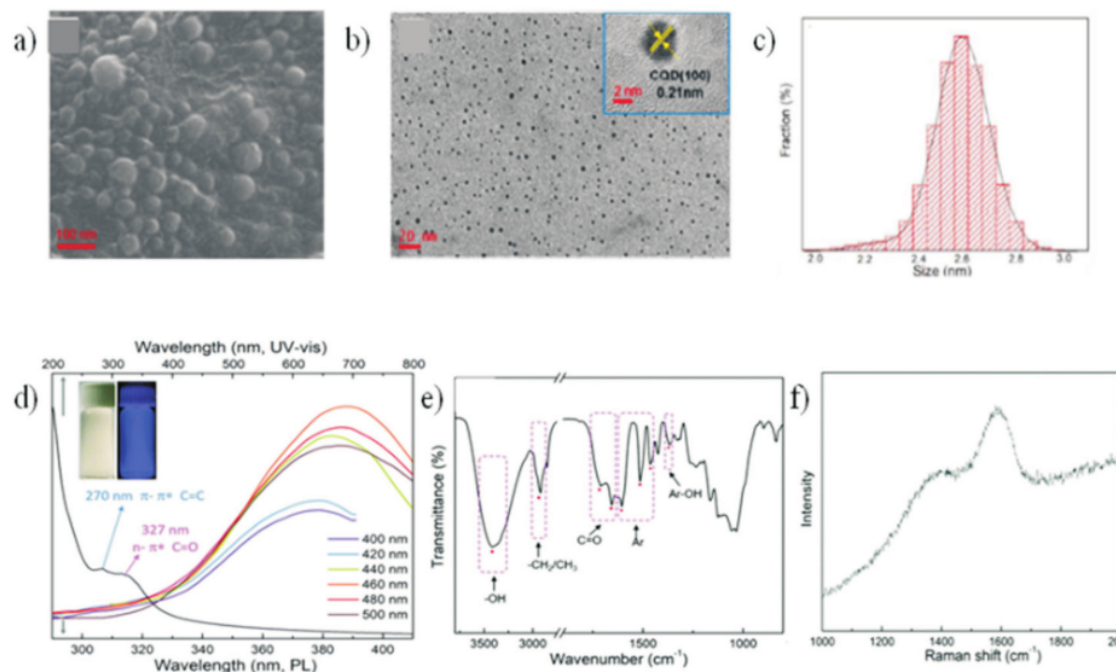
Si *et al.*<sup>128</sup> synthesized CNDs from rice straw as a raw carbon source by treating it with 5% in acid catalyst in a co-solvent preparation of ethanol-water, using a fast microwave-assisted one-pot method. The morphology of the as-prepared lignin-based nanoparticles observed in the sophisticated heterogeneity and exhibited crystallization defects. After lignin-based nanoparticles (LNPs) were dissolved in the selected organic solvent phase, LNPs produced solution-based precipitates of CNDs, facilitating the lignin diffusion into the solution in contrast with depositing on the surface area.

The reliable zeta potential values verified the excellent stability of the lignin-based nanoparticles dispersion (Fig. 7a), which is beneficial to heated deterioration completion and good thermal durability. The sizes ranged from 2 to 3 nm of prepared lignin-based CNDs. The lignin-based CNDs were dominated by the grafting space of 0.21 nm of a crystalline structure according to the HRTEM analysis (Fig. 7b). Interestingly, the solution-based CNDs were influenced by more graphitic crystallization than the only LNPs. The wavelength peak of the UV-vis absorption of the CNDs solution displayed two shoulder peaks at 270 and 327 nm as indicated in Fig. 7d, which corresponded to the  $\pi-\pi^*$  transitions of aromatic  $sp^2$  domains and the  $n-\pi^*$  shifting of the C=O bond, respectively.

Additionally, the optical features of the CNDs solution exhibited bright purple fluorescence when irradiated. Since the increased excitation wavelength occurred from 400 nm to 500 nm, the emission wavelength shifted to irreversible lower

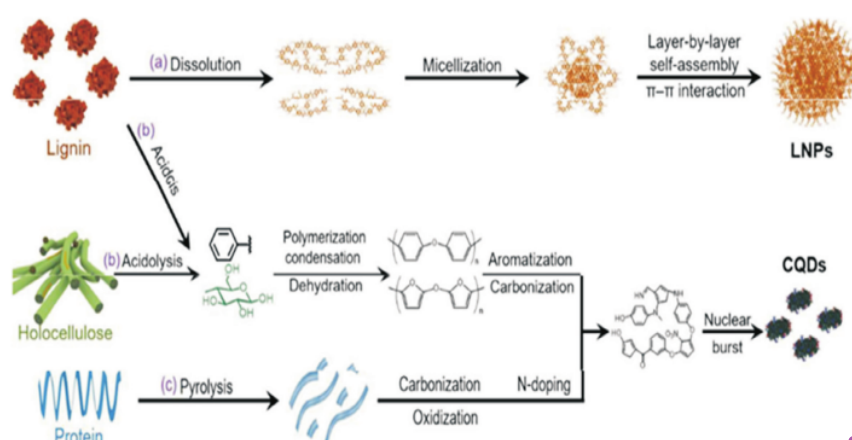
peaks of  $\sim 380$  nm. The infra-red spectroscopy results of LNPs demonstrated that the stretching absorption bands of -OH (hydroxyl groups) appeared at  $3420\text{ cm}^{-1}$  and, methyl and methylene groups absorb stretching at  $2927\text{ cm}^{-1}$ . Subsequently, these functional groups occurred abundantly in the LNPs. Moreover, the bending vibration of the phenolic hydroxyl group appeared at  $1636\text{ cm}^{-1}$ . The two bands, namely the C=O stretching at  $1710\text{ cm}^{-1}$  and  $1655\text{ cm}^{-1}$ , attributed to the presence of not only conjugated but also unconjugated C=O in the LNPs (Fig. 7e) appeared. The typical peaks in the Raman analysis centred at  $1360$  and  $1590\text{ cm}^{-1}$  for the CNDs indicated as the D and G bands of prepared carbon nanomaterials, respectively, confirmed the intrinsic nature of nanocrystalline graphitic  $sp^2$  and  $sp^3$  carbon deformations (Fig. 7f). In this novel one-pot microwave-assisted approach, the acidolysis released the monosaccharide, and the organic acid formed and solvated into the lignin fraction. Furthermore, Fig. 8 illustrates the synthesis of CNDs from natural lignin, holocellulose and protein through dehydration, polymerization and condensation, as well as from aromatization or carbonization.<sup>128</sup> Therefore, it was conferred from the physicochemical and optical properties of selected solution-based CNDs that these CNDs are more favourable than lignin-based nanoparticles alone.

Liu *et al.*<sup>129</sup> conducted a rapid microwave-assisted synthesis of amphiphilic CNDs from glucose as starting material combined with distilled water and oxidized by concentrated  $\text{H}_3\text{PO}_4$ . The optimal CNDs solution could be dissolved and readily taken out by chloroform and other organic solvents (ethanol, THF, toluene), exhibiting their amphipathic form. When it was excited at 360 nm, the CNDs displayed two well-defined fluorescent wavelengths in the middle parts at both 447 and 560 nm. More-



**Figure 7** (a) SEM photograph of lignin-based nanoparticles. (b) Solution-based CNDs images captured by TEM and HRTEM. (c) Histogram of the corresponding dot size distributions of CNDs. (d) UV-vis absorption and PL spectra of the CNDs at different excitation wavelengths; inset: photograph of the CNDs aqueous solution captured under daylight and 480 nm UV light lamp on left and right, respectively. (e) FTIR spectra of the LNPs. (f) Raman spectrum images of CNDs. Images and diagrams are reprinted with permission from reference [128]. Copyright © 2018 Royal Society of Chemistry.





**Figure 8** General mechanism for co-preparation of lignin-based nanoparticles and solution-based CNDs. The image is reprinted with permission of reference [128]. Copyright © 2018 Royal Society of Chemistry.

over, the CNDs exhibited excitation-dependent emission feature connected with higher potency observed at 442 nm and had a quantum yield (QY) of 7.6 % in water. To further study CNDs produced from natural products-based carbon sources using chemical oxidation for biomedical applications, their specific information was summarized and presented in Table 4.

## 2.6. Molecular Aggregation

The molecular aggregation method is not well-known for the preparation of CNDs. Only a few reports, including that of the Niu group,<sup>133</sup> is available. This method also offered easy preparation from organic sources of natural products derived CNDs (Fig. 9) without heating or other kinds of energy input. Owing to the presence of lignin, the formation of CNDs played an important role in forming J-type molecular clusters in the prepared solution. These aggregates optimized photoluminescence, and the newly created CNDs showed excellent results for bioimaging applications. The CNDs emitted both up-converting and down-converting emission following excitation with ultraviolet-visible and near-infrared (NIR) radiation. It is worth noting the following two limitations of this method. Firstly, organic raw materials must have aromatic structures (as shown in Fig. 10). Secondly, the CNDs formed by aggregation do not exhibit high chemical and photostability, since the J-type bulks are easily affected by extrinsic interference.<sup>133</sup>

In 2018 Zhang *et al.*<sup>134</sup> prepared through a facile molecular aggregation approach luminescent CNDs from bean-shell phenolic extracts for use in the antioxidation and bioimaging. The as-prepared CNDs exhibited a favourable antioxidation efficiency with photo-bleaching resistance. Their size distribution

ranged from 1 to 5 nm with pH/excitation dependent fluorescence. Their novel findings demonstrated that the bio-compatible CNDs are helpful for banana preservation and bioimaging tumour cells *in vivo*.

Reckmeier *et al.*<sup>135</sup> prepared carbon dots using citric acid and ammonia reactants through the molecular aggregation of ammonothermal synthesis. The resulting nanoparticles showed amorphous aggregates of molecular fluorophores. Those aggregates displayed the blue-green double emission spectrum of prepared CNDs' concentration-dependent optical features and also exhibited graphitic domains to the emission behaviour.

Liu *et al.*<sup>136</sup> reported fluorescent CNDs from tannic acid (TA), synthesized by hydrothermal treatment, that exhibited aggregation-induced emission enhancement to visualize permissive detection. They explored organic solvents induced aggregation emission improvement of CNDs for permissive detection and demonstrated promising applications in new sensing devices.

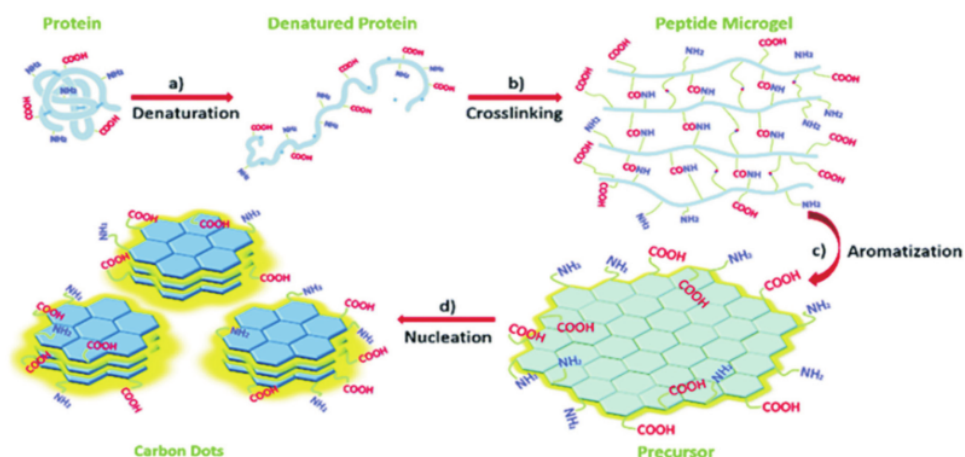
## 3. Properties

### 3.1. Physical Properties

CNDs mostly comprise carbon, hydrogen, nitrogen and oxygen, depending on the precursor and synthetic route. Among them, carbon and oxygen are the most plentiful components with many carboxylic acids. The potential of CNDs is highly dependent on the precursor and respective synthetic methods. These moieties could be helpful for both well-dissolved water and functionalized surface with other functional groups.<sup>87</sup> Generally, all of the organic-based CNDs are in the range of 2–100 nm-sized nanoparticles made of an sp<sup>2</sup>-hybridized

**Table 4** CNDs prepared from different precursors, using microwave-assisted reactions.

Precursors	Solvents	Additive	Reaction conditions	Size/nm	Application	References
Sucrose	Water	Phosphoric acids	100 W for 3 min 40 s	3–10	Drug delivery and biosensing	130
Chitosan	Water	Polyethylene glycol	450 W for 5 min	2–10	Drug delivery	131
Lotus root	Water	–	800 W for 6 min	2–5	Detection of Hg <sup>2+</sup>	132
BSA and urea	Water	–	700 for 5 min	2.3–5.4	Intracellular imaging and temperature sensing	129
Natural lignocellulose	Ethanol–water co-solvent system	–	10 min	2–3	Bioimaging	128



**Figure 9** Formation mechanism of CNDs prepared from protein carbon source by the microwave-assisted method. The image is reprinted with the permission of reference [129]. Copyright © 2016 Royal Society of Chemistry.

graphite nucleus or core, which is modified and functionalized by a polar hydroxy group.<sup>137,138</sup> CNDs also possess good physical and optical properties to develop <sup>46</sup> their potential applications effectively. Mechanical and optical properties of the CNDs play an important role in the application of electronic devices.<sup>139,140,141</sup> Moreover, the fluorescent features also play an important role in bio-medicinal use.<sup>142</sup>

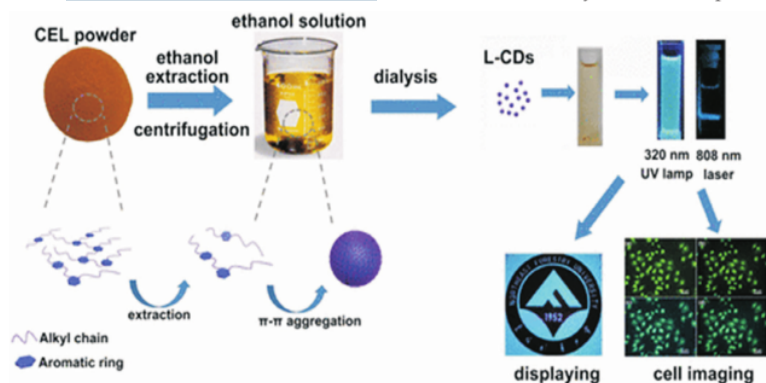
The organic products-derived CNDs typically involve carbon, oxygen and other heteroatoms.<sup>100,143</sup> These doped-heteroatom CNDs <sup>41</sup>ve been synthesized from hair and protein, respectively. Heteroatom-rich natural products also contain nitrogen and sulphur. Since lone pair electrons occupied by heteroatoms were conjugated with top orbitals in the carbon materials, the nitrogen/sulphur-doped CNDs exhibits excellent optical and electronic manners. Generally, natural products-derived CNDs showed both amorphous and crystalline features due to surface moieties on the CNDs' surface area. Generally, most of these heteroatom-doped CNDs from organic products have low crystallinity, even though many studies have reported the presence of crystalline  $sp^2$ -carbon. The surface groups of the prepared organic products-based CNDs are complex, particularly oxygen-related functional groups.<sup>144</sup>

### 3.2. Optical Properties <sup>8</sup>

In the past decade, CNDs <sup>8</sup>have received much attention due to

the material's properties. According to a fundamentally applicable point of view, photoluminescence (PL) has drawn many researchers' attention, and it is also one of the most promising characteristics of CNDs. PL means fluorescence that does not result from heating.<sup>145</sup> PL emission peaks of CNDs are dependent on the excitation wavelength and are based on particle size, appearance, conformation, and intrinsic structure of CNDs. Thus, tuning of the emission <sup>11</sup> peaks could be done for further real-life application. Mostly, the corresponding absorption wavelength is shorter than the wavelength of emission. That is why the energy of the absorption is higher than that of emission. Tuning the wavelength emission spectrum is an extensively considered fact of CNDs.<sup>146</sup> CNDs can not only donate electrons, but they can also receive or accept them.<sup>147</sup>

CNDs normally <sup>15</sup>ow photoluminescent emission, but that fluorescence can be quenched by adding electron acceptors due to photo-induced electron transfer between the CNDs and electron acceptors. Equally, CNDs can also serve as powerful electron acceptors because the luminescence can be quenched by electron donors, for example, N, N-diethyl aniline.<sup>147</sup> This characteristic formation is solvent-dependent and more effective in polar solvents. Photo-induced electron transfer is significant in CNDs-sensitized matters and CNDs-based photocatalyst structuring. The photo-induced electron transfer of CNDs is influenced by electron-hole pairs, surface moieties, and



**Figure 10** Schematic diagram <sup>9</sup> of CNDs preparation from lignin followed by the molecular aggregation method to be applied in displaying and bioimaging. The image is reproduced with permission of reference [133]. Copyright © 2017 American Chemical Society.

adherent processes.<sup>148</sup> Moreover, CNDs have unique optical properties. Then fluorescence absorption and emission of organic products derived-CNDs did not come initially from a bandgap but were affected by radiative recombination and p-plasmon of the surface-restricted electrons holes.<sup>139</sup> This differs from man-made semiconductor carbon nanomaterials.

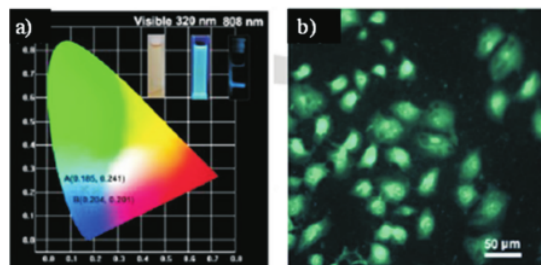
In relation to absorbance, the green CNDs typically demonstrated that optical emission is absorbed in the spectrum's UV region, shifting to the visible region to a small degree.<sup>10</sup> However, the organic products derived-CNDs prepared by standard and simple methods usually showed absorption peaks in the UV range. Each UV range position of CNDs from various carbon sources (man-made carbon sources and carbon sources from organic products) is different.<sup>19</sup> Some absorption peaks corresponded to the  $\pi-\pi^*$  shifting signal of the C-C bonds, the  $n-\pi^*$  relocating behaviour of C=O bonds and/or others. The degree of emission depends on the particle's radius, so as the particle gets smaller, both the excitation and emission spectrum shift to shorter wavelengths.<sup>149</sup> Yoshinaga *et al.*<sup>150</sup> prepared CNDs from glucose solution through microwave-assisted pyrolysis. The average size distribution is of diameter 1.65 nm, and the fluorescence QY is 7–10%.<sup>150</sup> The clear UV absorption peaks showed two prominent bands occupied in the middle part of 228 and 282 nm. Since the length of microwave treatment increased, the intensity of both UV absorption bands changed without tuning band areas, which attributed no size-related areas of the prepared nanomaterials.<sup>150</sup>

CNDs with various sizes, which emitted different fluorescence have been synthesized by diverse methods to describe the range of the fluorescence properties. These emission spectra varied from the visible region to the near-infrared region showing blue, green, yellow, and red. Pan *et al.*<sup>151</sup> reported the preparation of CNDs via hydrothermal method; the prepared CNDs emitted high fluorescence in alkaline solution situation, whereas this photoluminescence was entirely dim in acidic solution matter. When the experiment was repeated in an alkaline solution, the photoluminescence intensity of CNDs recovered. It confirmed that the pH of the solution does not affect the emission wavelength range, but it had a significant impact on the intensity of photoluminescence. The facile CNDs demonstrated excitation-dependent emission related to that of other CNDs derived from conventional semi-quantum dots. Liang *et al.*<sup>147</sup> prepared chitosan-based CNDs as a novel design for a specific study of the quenching fluorescence process. The behaviours of quenching CNDs were studied using a functional group of nitro-aromatics with various ring components, and the practical evidence showed that the preliminary procedure for CNDs-quenching is Förster resonance energy transfer (FRET) not electron transfer. However, the fluorescence emission mechanism draws little attention or interest. Owing to the fluorescence phenomenon of CNDs with plausible mechanisms regarding surface passivation/deformations,<sup>152</sup> quantum size effect,<sup>153</sup> surface state effect,<sup>144</sup> and edge state and carbon-core state composition might be significant to explaining the CNDs' fluorescence emission in detail.<sup>144</sup>

Some strategies were also summarized for increasing quantum yield and red-shifting absorption wavelength of CNDs. In particular, surface functional moieties,<sup>154</sup> degree of surface functionalization,<sup>155,156</sup> quantum size effect/conjugated  $sp^2$ -domain effect<sup>155,157</sup> and element doping<sup>141,158</sup> are usually used to explain the characteristics of the long-wavelength and tunable emission.

In the up-conversion fluorescence phenomenon, CNDs have a significant feature of intense and tunable PL (photolumines-

cence). PL emission spectra are usually shoulder peaks, positioning in the transition from the visible to near-infrared (NIR) position, based on the wavelength excitation.<sup>22</sup> The distinctive emission peaks can be obtained by evaluating excitation wavelengths or particle sizes to provide fluorescence imaging and optical labelling. More interestingly, besides down-converted or normal PL, certain CNDs possessed incredible up-converted PL, and this up-conversion fluorescence emission is significantly agreeable for *in vivo* bioimaging application. A two-photon system might be performed in the up-conversion. For example, Wang *et al.*<sup>159</sup> prepared nitrogen-doped CNDs of 1–3 nm through a facile one-pot route from the carbon source of melamine and glycerol reaction. These nitrogen-doped CNDs showed multi-photon up-conversion fluorescence and had a strong two-photon absorption cross-section. That up-converting potency of CNDs is applicable for multi-photon single-molecule imaging. Similarly, the CNDs synthesized from some organic sources could offer large two-photon absorption by cross-sections. At the same time, they could absorb two long-wavelength photons upon excitation, and one short-wavelength photon is released. Besides, the CNDs with electron donors and electron acceptors could trigger triplet-triplet annihilation up-converting emission. The applications for up-converting emission have already been published in several papers, but the detailed observation of the mechanism is still challenging.<sup>106</sup> Niu *et al.*<sup>133</sup> tried to use up-converted emission from lignin-derived CNDs as cellular imaging agents, as shown in Fig. 11a,b. Up-converting emission can also be applied in potential catalysts, biomedicine, and energy storage application technology.



**Figure 11** (a) CIE chromaticity description and the photograph of lignin-derived CNDs showing emission colours. (b) Inverted fluorescence microscopic photographs of HeLa cells incubated by lignin-derived CNDs upon excitation at 800 nm. Images are reproduced with the permission from reference [133]. Copyright © 2012 American Chemical Society.

#### 4. Applications

In contrast with green CNDs prepared from organic products, there are some drawbacks in many conventional ways of synthesizing quantum dots for semiconductor materials. For example, using toxic chemical reagents as the starting materials, requiring high temperature, tedious running, and cumbersome synthetic steps.<sup>160</sup> Consequently, these man-made carbon quantum dots (CQDs) have more drawbacks than organic products-based CNDs. Therefore, those man-made CQDs are more limited in term of synthesis and applications.

In earlier years using simple diverse synthetic methods, green CNDs have been developed from many natural products, for instance, soy milk,<sup>161</sup> eggshells,<sup>162</sup> garlic,<sup>163</sup> honey,<sup>164</sup> ground coffee<sup>65</sup> and prawn shells.<sup>165</sup> These carbon source starting materials from plants, leaves, fruits, animals, microorganisms, and their excreta offered organic products derived-CNDs.<sup>21</sup> These

CNDs have several advantages, such as readily and abundantly available raw materials, low cost, convenient synthesis, acceptable renewability, multicoloured photoluminescence, and excellent biocompatibility. Researchers have been keen to synthesize these new profitable and environmental friendly nanomaterials with novel applications. In this article, the applications of organic product based-CNDs in medicinal applications, for example, bioimaging and therapy, drug delivery, sensing and detection, solar cells, and photo-catalysis, will be described.

#### 4.1. Bioimaging and Therapy

There have been many approaches to preparing organic products-based CNDs to stain cancer cells or tumour cells to address the core for fast and accurate targeted cell detection.

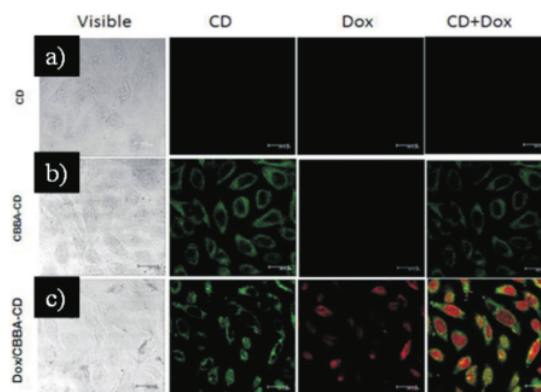
Barbosa *et al.*<sup>125</sup> synthesized CNDs from animal excreta, like cow manure, and from glucose. The new modified CNDs showed strong fluorescence. The fluorescence means that they could be used effectively to probe living cells with magnificant sub-cellular selectivity. Also, the as-prepared CNDs can stain nucleoli in breast MCF-7 selectively. The CNDs were sensitive to four other cellular patterns and displayed the corresponding cellular selection in live-cell imaging observations.<sup>125</sup> Wang and Zhou<sup>166</sup> synthesized non-toxic CNDs from the milk carbon source through microwave radiation without any chemical reagents. The CNDs exhibited low-toxic behaviour in the concentrated range of 15 mg mL<sup>-1</sup> and bright blue, green, and red colours of emission were showed under an inversely photoluminescent microscope during cellular imaging investigation.<sup>166</sup>

Song *et al.*<sup>167</sup> prepared CNDs from linseed. The prepared CNDs can potentially be applied in both biosensors and bioimaging. Feng *et al.*<sup>104</sup> synthesized green CNDs from edible winter melon *via* hydrothermal treatment and the resulting CNDs are successful bioimaging agents for HepG<sub>2</sub> cells. Wei *et al.*<sup>91</sup> synthesized heteroatom-doped CNDs like nitrogen/sulphur-doped CNDs from *Allium fistulosum* through a one-pot hydrothermal approach. The as-prepared CNDs showed 85 % of cell viability against Hela cells, even when incubated at high concentration of 20 mg mL<sup>-1</sup>. Because of their non-toxic behaviour, the as-synthesized CNDs could be selectively applied for multicolour cellular imaging in K562 and MCF-7 as excellent fluorescent probes.

Fahmi *et al.*<sup>72</sup> have found that CNDs can be prepared from bamboo leaves by pyrolysis. The as-prepared CNDs were modified with CBBA (4-carboxy-benzylboronic acid) to obtain specific targeting of HeLa tumour cells. More interestingly, an anticancer agent of doxorubicin (Dox) was loaded onto the modified CBBA-CNDs to use them for drug delivery. Potential internalization of CND, CBBA-CNDs, and Dox/CBBA-CNDs into HeLa tumour cells is depicted in Fig. 12. It is worth noting that significant green fluorescence was investigated from CBBA-CNDs on the cytoplasm of HeLa tumour cells after 1 h of incubation.

Meanwhile, green fluorescence's significant presence confirmed that CNDs were favourably absorbed by the cells *via* endocytosis (Fig. 9b). Moreover, Dox delivery was successfully performed using CBBA-CNDs with red fluorescence; the typical emission of Dox was explored in the nuclei of cells treated with Dox/CBBA-CNDs. Dox plays a role as a tumour drug by interfering with the DNA helix structure in the cell nucleus. The formation of red fluorescence in the cell nucleus, after incubation with Dox, confirmed that the drug had been well delivered to the target.

Moreover, Wu *et al.*<sup>168</sup> prepared surface passivated CNDs based on a solvent-free synthesis from commercial food-grade



**Figure 12** (a) CLSM photographs of CNDs. (b) CBBA-CNDs images. (c) Dox/CBBA-CND images after incubation for 1 h with HeLa cells at 37 °C. Images are reprinted with permission from reference [72] Copyright © 2018 Royal Society of Chemistry.

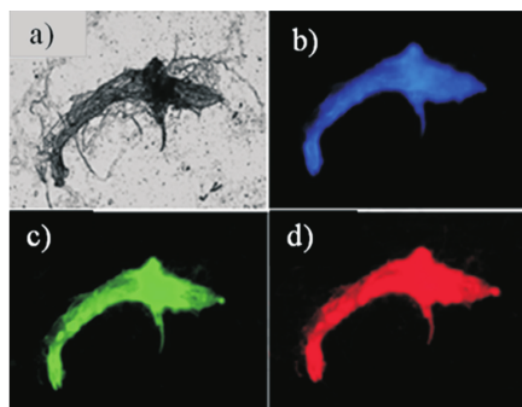
honey as starting material. Since intense near-infrared absorption was detected, it can be concluded that microscopic size and rapid lymphatic carrier of CNDs have dislocated from the paw to the axillary lymph node after loading. Therefore, the evidence of that lymph node is significantly improved. They were also discarded from the vessels relatively fast. This exploration can play a vital role in observing sentinel lymph nodes (SLN), where cancer cells can propagate in case of breast cancer.

Another report by Bhamore *et al.*<sup>169</sup> described the preparation of green CNDs from *Acacia concinna* seeds (a carbon source) for fluorescence imaging of fungal cells by using the microwave hydrothermal approach. To assess imaging efficiency by the as-prepared CNDs, the ultra-small CNDs-incubated *Penicillium* sp. cells showed the optical performance of different kinds of fluorescence such as luminous blue, green and red colours by following laser excitation wavelengths of 405, 488 and 561 nm. This optical performance means that the prepared ultra-small CNDs effectively entered (successful internalization) the cells *via* endocytosis. Hence, these CNDs localized in the cytoplasm and were specially wrapped in the nucleus of cells. The novel exploration clearly explained that the ultra-small CNDs synthesized from *Acacia concinna* seeds exhibited excellent biocompatibility and internalization into the cells with multicoloured imaging, implying their promising use in cell imaging, as shown in Fig. 13.<sup>169</sup>

Amin *et al.*<sup>170</sup> also reported water-dispersible nitrogen-doped CNDs (N-CNDs) from the date kernel as a starting matter through a one-pot hydrothermal synthesis without any preceding treatment. The resulting N-CNDs significantly showed 12.5 % of a quantum yield, which was relatively higher than most N-CNDs synthesized from different carbon sources. Interestingly, the as-prepared N-CNDs intrinsically have a great potential regarding optical properties and the higher QY without the addition of high cost or toxic chemical reagents. More importantly, they observed the blue excitation photographs of MG-63 cells after N-CNDs solution (20.0 mg mL<sup>-1</sup>) incubation. They showed bright green fluorescence (Fig. 14b). Thus, several amounts of N-CNDs easily entered the cell membranes. It was suggested that they were able to keep their fluorescent behaviour in the cellular environment.<sup>170</sup>

#### 4.2. Drug Delivery

Nanomaterial-based drug delivery system is one of the biomedical treatments, which is not used much yet. The organic



**Figure 13** Multicolour cell imaging potential of *Penicillium* sp. Cells with respective (a) bright field, (b) blue emission, (c) green emission and (d) red emission when treated with oxidized CNDs under different laser excitation wavelengths. Images are reproduced with permission from reference [169]. Copyright © 2018 Elsevier.

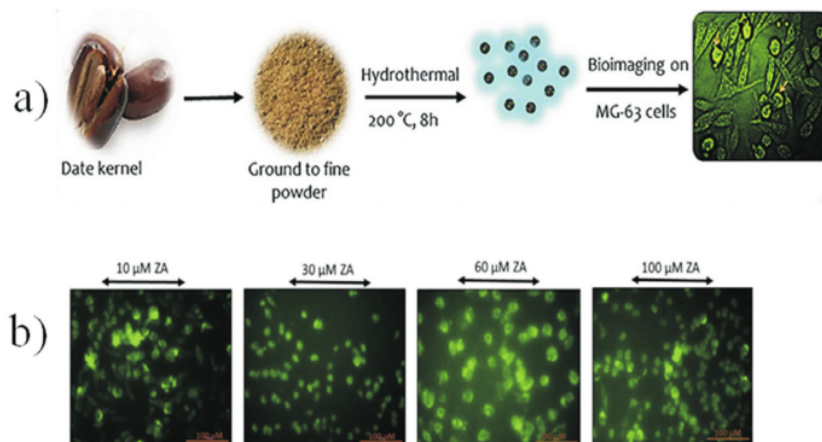
products-based CNDs have been one of the most attractive nanomedicine models. They have drawn researchers' interest and attention because of their selective targeting response, low toxic behaviour, and agreeable drug efficacy, in contrast with conventionally delivered systems.<sup>171,172</sup> Furthermore, they can serve as multifunctional vehicles for loading and releasing drugs. That is why the most promising qualities of organic products-based CNDs and graphene anodots are strong fluorescence, excellent biocompatibility, rapid cellular uptake, and high stability.<sup>22,173</sup>

Mehta *et al.*<sup>174</sup> reported the facile synthesis of doped heteroatoms CNDs from pasteurized milk, using hydrothermal treatment. The synthesized CNDs have many functional groups (amino or hydroxyl groups) in their site and surface, which can subsequently be functionalized and modified. The synthesized green CNDs were loaded with the lisinopril drug. The optimal lisinopril-loaded CNDs (Lis-CNDs) showed high cell uptake in the HeLa cells. Additionally, they also have excellent physicochemical properties, such as ultrafine size, multifunctional groups, and excellent photoluminescence. Thus, the fluorescent

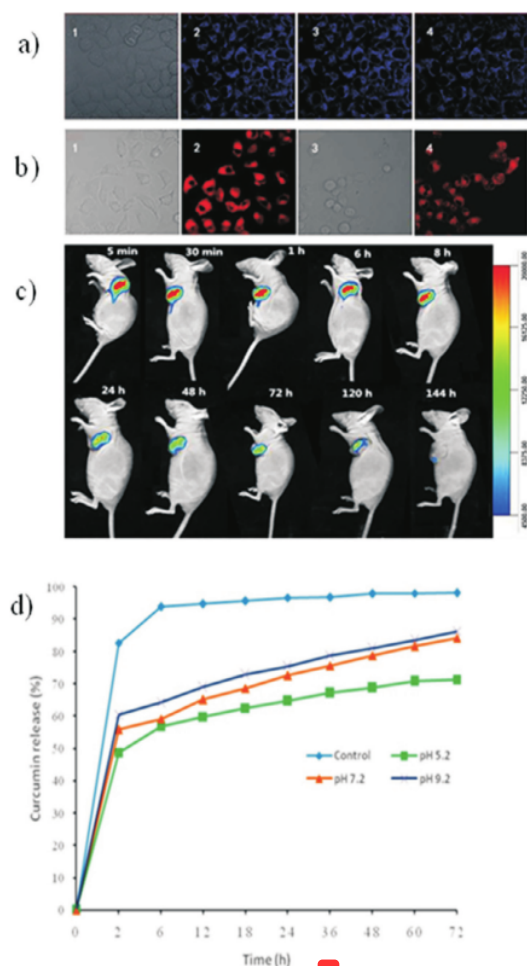
loaded Lis-CNDs could be used as drug carriers for drug loading and release. In addition to assessing the powerful ability of CNDs-based drug delivery methods, these lisinopril-loaded CNDs were found to have good stability when these CNDs circulated in the blood. Moreover, they released encapsulated drugs immediately after penetrating the targeted tumour cells. It was found that the release of the lisinopril drug was still stable up to 24 h. Subsequently, it was found that the release was constant over 48 h in the PBS conditions of pH 5.2, 6.2, and 7.4, respectively, as shown in Fig. 15a,b,c.<sup>174</sup>

Rai *et al.*<sup>175</sup> also used a microwave-assisted green and simple procedure to synthesize sulphur-doped CNDs from sulphur-containing lignosulfonate lignin as the starting raw materials. The synthesized CNDs were further modified by adding NaBH<sub>4</sub> into the solution according to a simple chemical reduction method. Owing to the surface moieties of lignin CNDs, the reduced CNDs (r-CNDs) can offer a potential tool for drug loading and release. Moreover, curcumin was loaded on Lignin r-CNDs to reach the specifically targeted cells. It was found that the delivery of the optimal percentage of curcumin from the r-CNDs was complete, followed by supporting curcumin, as indicated by the standard calibration curve (Fig. 15d). At biological pH calibrated conditions, a maximum value of curcumin release percentage was slowly rising to 82.7% when calibrated at pH 7.2 after 72h, but curcumin loaded r-CNDs solution at pH 5.2 offered a burst release within early 2 h. The cumulative maximum release of 69.1% was achieved after 72 h. Significantly, at pH 9.2 calibration, 86.37% of maximum curcumin release was observed due to the hydrophilicity development, when the pH calibration was increased specifically from 5.2 to 9.2. In conclusion, the curcumin loaded r-CNDs of fast-acting dosage can benefit the novelty of drugs for initial chemotherapy treatment.<sup>175</sup>

D'souza *et al.*<sup>176</sup> reported the preparation of nitrogen-doped fluorescent CNDs (N-CNDs) from dried shrimp as carbon precursors through a one-step hydrothermal route. They were intended to be used for potential drug delivery, transport agents and its impact on cancer cells was tested. Subsequently, boldine was loaded on the surface of the as-prepared CNDs and intracellular distribution, internalization process, and release rate were studied. Interestingly, the fabricated CNDs show diverse surface moieties (-NH<sub>2</sub>, -COOH, and -OH), which facili-



**Figure 14** (a) Schematic illustration of prepared nitrogen CNDs for bioimaging MG-63 cells from date kernel as a raw material. (b) Fluorescence microscopic photographs of MG-63 cells incubated with 20.0 mg mL<sup>-1</sup> N-CNDs at 37 °C for 25 min. Images are reproduced with the permission of reference [170]. Copyright © 2018 Elsevier.



**Figure 15** (a) Diagrammatic photographs of HeLa cells incubated with r-CNDs, NAC, and  $\text{Fe}^{2+}$ . (b) CLSM photographs of Bel-7404 cells treated with r-CNDs for 6 h at 37 °C. (c) The mice tumour treated with r-CNDs under NIR fluorescence at different time intervals. Images are reprinted with the permission from reference [22]. Copyright © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (d) Curcumin percentage release (%) from curcumin loaded r-CNDs concerning time in PBS versus pH of 5.2, 7.4, and 9.2 with control. The image is reproduced with the permission of reference [175]. Copyright © 2017 Elsevier.

tates loading boldine on the surface of r-CNDs through hydrogen bonding. The fluorescent N-doped r-CNDs served as novel fluorescent probes to give simultaneous imaging/mapping of MCF-7 and SH-SY5Y (human neuroblastoma) cells and act as drug carrier agents for delivering boldine drug to specific target cells. The internalized vesicles will trigger the ability to release boldine from the r-CNDs, thereby applying anticancer drug delivery and cell imaging to MCF7 cells. The fluorescence of r-CNDs will help to monitor drug release progress in the cells. As a result, the fluorescent N-doped r-CNDs could sensitively probe the targeted imaging and can serve as drug carriers for selective target delivery system.<sup>176</sup>

#### 4.3. Sensing and Detection

One of the interesting applications of r-CNDs lies in the field of sensing. Many organic products-based r-CNDs have been respon-

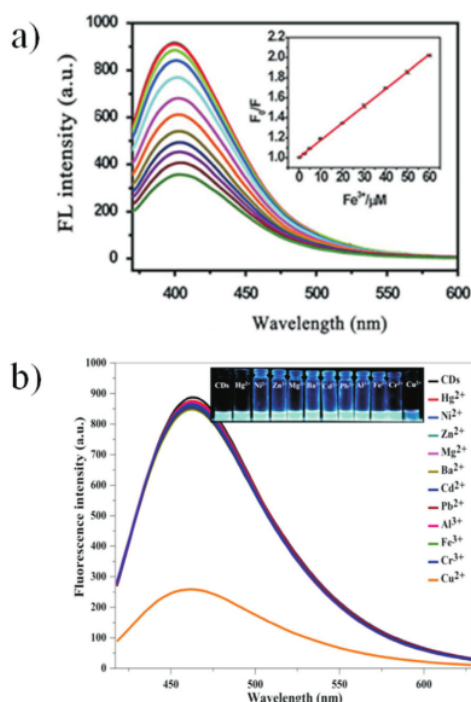
sible for fluorescent directed-sensors to detect metal ions, anions, and molecules because they have unique optical and electrical properties. Several mechanisms play a significant role in the fluorescence changes of quenching or tuning of r-CNDs, including resonance energy transfer, photo-induced charge transfer, photo-induced electron transfer, and inner filter effect, which have been applied for sensing. The fundamental principle of the development of sensors for ions ( $\text{Fe}^{3+}$ ,  $\text{Pb}^{2+}$ ,  $\text{Hg}^{2+}$ ,  $\text{CN}^-$ , and  $\text{PO}_4^{3-}$ ) is the quenching of the fluorescence of r-CNDs by using some ions. Among them,  $\text{Fe}^{3+}$  is the most commonly detected. Biosensing application is an important application of organic products derived-r-CNDs, due to the highly selective and is commonly used.<sup>22</sup>

Sachdev *et al.*<sup>108</sup> reported coriander leaves as a carbon precursor to prepare organic products-based r-CNDs through a facile treatment. Black and green tea, honey, papaya, egg white, goose feathers, and other organic resources are extensively used to synthesize r-CNDs to detect  $\text{Fe}^{3+}$ .

Xu *et al.*<sup>177</sup> prepared water-soluble and dispersible r-CNDs with bright fluorescence from potatoes through a one-step hydrothermal approach. The r-CNDs prepared from cheap and cost-effective carbon sources demonstrated strong blue fluorescence with QY up to 15%. The result strongly confirmed that the synthesized r-CNDs could be used as a novel sensing probe for the label-free, selective, and sensitive detection of  $\text{Fe}^{3+}$ . The detection limit was  $0.025 \mu\text{mol}^{-1}$ , corresponding to different sensitivities of  $1.0\text{--}5.0 \mu\text{mol}^{-1}$  and  $5.50 \mu\text{mol}^{-1}$  at least and a wide range of concentration. Thus, the as-prepared r-CNDs can detect the  $\text{Fe}^{3+}$  ions in real-life water samples.

Jayaweera *et al.*<sup>178</sup> demonstrated the green and low-cost r-CNDs from durian shell waste using a microwave hydrothermal approach for label-free detection of  $\text{Fe}^{3+}$ . The durian shell contains carboxylic acids, esters, and amino compounds that required improving the r-CNDs surface. The resulting quantum yield was 6.2%. The blue-green emitting r-CNDs showed stability over a range of pH and chloride concentrations and indicated photobleaching resistance. There was a concentration range of 0–20 nM with the detection limit of up to 128 nM. In this way, the synthesized r-CNDs could be applied in sensing  $\text{Fe}^{3+}$  in real-life water systems, as indicated in Fig. 16a.

In addition to the detection of  $\text{Fe}^{3+}$ , Bhamore *et al.*<sup>169</sup> synthesized green r-CNDs from *Acacia concinna* seeds (a carbon source) for fluorescence sensing of  $\text{Cu}^{2+}$  ion. These r-CNDs comprise various phytochemicals, such as alkaloids, acacic acid, lactone, lupeol, spinasterol, ascorbic acid, citric acid, tartaric acid, succinic acid, and sugars. These phytochemicals, belonging to *Acacia concinna* seeds, could produce ultra-small fluorescent r-CNDs with intrinsically multifunctional surface groups, making them robust, selective and sensitive sensing probes. As a result, the optical properties of ultra-small r-CNDs (0.5 mL) exhibited fluorescence emission wavelengths with different metal ions ( $\text{Hg}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Zn}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Ba}^{2+}$ ,  $\text{Cd}^{2+}$ ,  $\text{Pb}^{2+}$ ,  $\text{Al}^{3+}$ ,  $\text{Fe}^{3+}$ , and  $\text{Cr}^{3+}$ ). However, (0.7 mL;  $500 \mu\text{M}$  of  $\text{Cu}^{2+}$ ) was individually analyzed by fluorescence spectrometry (Fig. 17b). The optical spectrum of ultra-small r-CNDs solution showed the wavelengths in the range of  $\lambda_{ex}/\lambda_{em} = 390/468 \text{ nm}$ , but a single fluorescence intensity of  $\text{Cu}^{2+}$  ion significantly quenched. Thus, the higher concentration of  $\text{Cu}^{2+}$  ions could remarkably quench the strong blue emission of ultra-small r-CNDs (Inset of Fig. 16b). These observations verified that the quenching of the significant fluorescence was connected to the  $\text{Cu}^{2+}$  ion addition, favourably confirming that the ultra-small r-CNDs are useable for the sensing of  $\text{Cu}^{2+}$  ions as a selective probe. Moreover, there are considerable studies of r-CNDs prepared from respective organic resources for the detec-



**Figure 16** (a) Images of fluorescence emission spectra of ultra-small CNs treated with the addition of different concentrations of  $\text{Fe}^{3+}$ . Inset represents the linear calibration plot for  $F_0/F$  versus various concentrations of  $\text{Fe}^{3+}$ . The image is reproduced with permission from reference [178]. Copyright © 2017 Elsevier. (b) Photograph of fluorescence spectra of ultra-small CNs of excitation and emission wavelengths ( $\lambda_{ex}/\lambda_{em} = 390/468 \text{ nm}$ ) with different metal ( $\text{Hg}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Zn}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Ba}^{2+}$ ,  $\text{Cd}^{2+}$ ,  $\text{Pb}^{2+}$ ,  $\text{Al}^{3+}$ ,  $\text{Fe}^{3+}$ ,  $\text{Cr}^{3+}$ , and  $\text{Cu}^{2+}$ ,  $500 \mu\text{M}$ ). Inset: photograph of their respective solutions under the condition of UV light at  $365 \text{ nm}$ . The image is reproduced with permission from reference [169]. Copyright © 2018 Elsevier.

tion of  $\text{Cu}^{2+}$ . To further describe them more, their detection limits were comparatively provided in Table 5.

Gu *et al.*<sup>132</sup> prepared CNs from the raw material of lotus root as a precursor for detecting  $\text{Hg}^{2+}$  ions using the microwave method without any surface passivating agents. The fluorescent nitrogen-doped CNs have a nitrogen content of 5.23%. In particular, the synthesized lotus root-based CNs (LR-CNDS)

**Table 5** Analytical performance of the presented result of detection of  $\text{Cu}^{2+}$  ion compared to the other reported results. The table is used with the permission from reference [169].

Probe	Linear range/ $\mu\text{M}$	Detection limit/nM
Polymer nanodots	0–50	1
Graphene Quantum Dots (GQDs)	0–15	226
Bamboo leaves CNDS	0.333–66.6	115
Coal CNDS	0.05–0.5	2.0
Waste polyolefins-CQDs	1–8	6.33
DNA-Au NPs	0.5–10	250
Starch-stabilized Ag NPs	0.1–10	500
Single-stranded/DNA Au NPs	0.625–15	290
Aminophenylboronic acid CNDS	1–25	300
Cross-linked chitosan	$0.15 \times 10^3$	$7.7 \times 10^3$
Petroleum coke CQDs	0.25–10	29.5



**Ultrasensitive: 55 pM  $\text{Pb}^{2+}$**

**Figure 17** Schematic diagram of fluorescent nitrogen-doped CNs from lotus root and the applications in detecting the  $\text{Hg}^{2+}$ . The image is reproduced with the permission from reference [132]. Copyright © 2016 Elsevier.

can remarkably serve as a selective fluorescent probe to detect  $\text{Hg}^{2+}$  with the linear concentration range from 0.1 to  $60.0 \mu\text{M}$ . As indicated in Fig. 17, the detection limit was evaluated to be 18.7 nM.

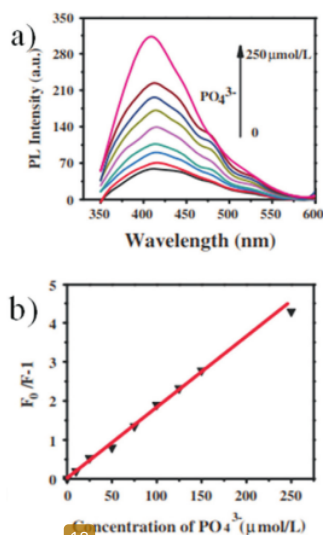
Xu *et al.*<sup>179</sup> reported the flavonoid-based CNs (Fla-CNDS) from the starting material of flavonoid extracts of *Ginkgo biloba* leaves to detect toxic heavy metal ion  $\text{Pb}^{2+}$  through a one-pot hydrothermal route. They found that Fla-CNDS can detect the quantitative range (0.1–20.0 nM) of  $\text{Pb}^{2+}$  following an ultrahigh sensitivity of 55 pM. The selective efficacy of Fla-CNDS for  $\text{Pb}^{2+}$  was almost one order of targeting extent larger than that for other appropriate metal ions. More interestingly, agarose hydrogel was loaded onto Fla-CNDS (AHG-CNDS) for better optically luminescent detection and elimination of  $\text{Pb}^{2+}$  from water. They concluded that the  $\text{Pb}^{2+}$  adsorbed AHG-CNDS could be re-established in HCl solution. This new exploration contributed to the synthesis of high-cost effective materials for targeting and detecting heavy metal ions.

In addition to metal ions, organic products-based CNs can also be used to detect anions. Yin *et al.*<sup>106</sup> synthesized CNs, using a low-temperature carbonization route, from sweet pepper. The fluorescence properties of these CNs were quenched remarkably depending on the concentration of added  $\text{ClO}^-$ .

Xu *et al.*<sup>180</sup> synthesized blue-fluorescing CNs from potatoes as raw materials through a hydrothermal approach. The  $\text{PO}_4^{3-}$  explores the sensing performance's sensitivity concentration followed by a detection limit of  $0.8 \mu\text{mol L}^{-1}$  indicated in Fig. 18. They found that the photoluminescence of CNs can be quenched successfully by  $\text{Fe}^{3+}$ . If the  $\text{PO}_4^{3-}$  concentration was increased, the fluorescence could gradually be restored. These CNs could be a selective and sensitive probe for sensing and detecting  $\text{PO}_4^{3-}$ . Besides, the obtained CNs showed intense brightness, good biocompatibility, and well-dispersed water; that is why these promising features are effective for bioimaging applications.

**4.4. Solar Cell**

In the last few years, the organic products derived-CNDS have been investigated to be used in solar cells, because they exhibited excitation-independent, excitation-dependent fluorescence wavelength emissions, pH and size-dependent or independent fluorescence emissions.<sup>181</sup> Marinovic *et al.*<sup>3</sup> investigated the usefulness of mesoporous  $\text{TiO}_2$ -based solar cells loaded with green CNs synthesized from organic sources, including actual food waste of lobster shells. These promising CNs were intended to be used for optoelectronic materials in a solar cell. They were further separated for purification, subsequently performed by hydrothermal carbonization of the other starting materials. They indicated that the solar cells sensitized with lobster shells derived-CNDS exhibited a somewhat lower efficiency of 0.22%, suggesting that optoelectronic materials for



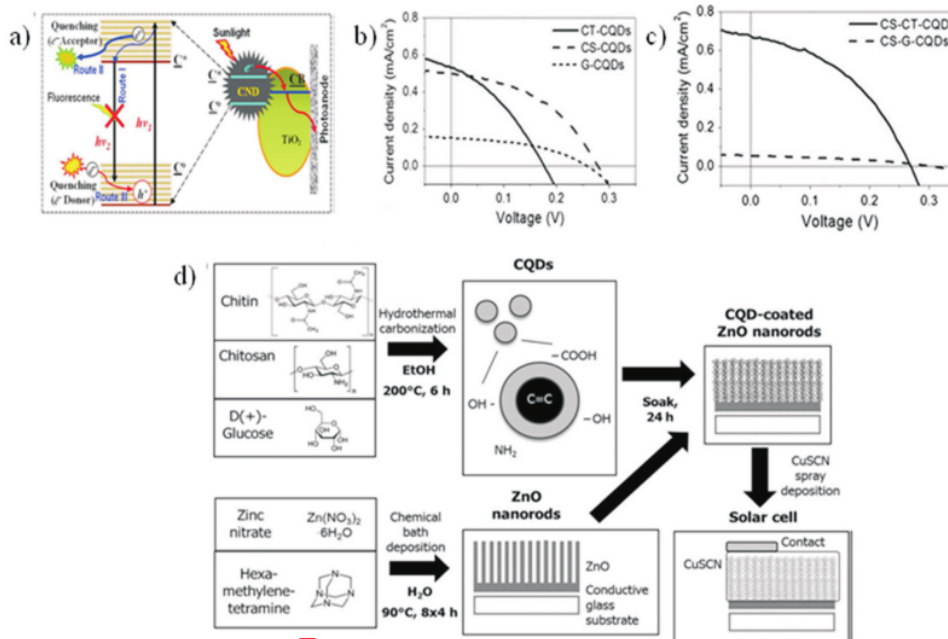
**Figure 18** (a) Image of fluorescence emission spectra of CNDs-Fe<sup>3+</sup> with the addition of different concentrations of PO<sub>4</sub><sup>3-</sup> (0, 10, 25, 50, 75, 100, 125, 150, 250 μmol L<sup>-1</sup>). (b) Calibration curves for 'a'. Images are reused with the permission of reference [180]. Copyright © 2014 John Wiley & Sons, Ltd.

the application in renewable energy storage cell technologies generated from organic waste products were possible to some practical extent.

Marinovic and co-workers suggested that the efficiency of the

solar cells was also connected to structural, chemical, and electrical properties of the synthesized CNDs.<sup>3</sup> Moreover, there are considerable studies of respective starting materials which derived CNDs for functional optoelectronic materials to be used in solar cells. Their average internal efficiency was comparatively studied with lobster-CNDs provided in Table 6.

Zhang *et al.*<sup>181</sup> prepared a fluorescence dimming mechanism that remarkably developed the conversion efficiency of CNDs responsive to aqueous solar cells. Organic products-based CNDs were prepared from grass as a carbon source material. The produced CNDs were significantly reliable for fluorescence quenching development. Their findings verified the concept that was extensively applicable for increasing solar cells' efficiency using other fluorescent quantum dots or nanodots sensitizers. A schematic diagram shows that an electron acceptor or donor could make the photo-excited electron quenched (Fig. 19a). Briscoe *et al.*<sup>139</sup> explored CNDs synthesized from organic sources, such as chitin, chitosan, and glucose. Since the functionalization of the as-prepared CNDs was performed by the other surface functional groups in the starting materials, these loaded CNDs would potentially sensitize ZnO nanorods to sunlight, agreeing well to be applied in solid-state nano-structured energy storage cells. A single-layer CNDs and a 4-layer layer-by-layer (LbL) ZnO/CNDs/CuSCN solar cells coating to illuminated current-voltage data are shown in Fig. 19b,c, respectively. Shi *et al.*<sup>182</sup> prepared carbon quantum dots through bottom-up synthesis. These TiO<sub>2</sub> photoanode loading CNDs can enhance photo-to-electrical conversion and the short-circuit current density efficiency in a solar cell device. The CNDs-sensitized solar cells prepared by HTC (high-temperature condition) of organic product sources and the applicable materials of chem-



**Figure 19** (a) Schematic diagram of quenching the photoexcited electron with an electron acceptor and donor. The image is reprinted with the permission of reference [183]. Copyright © 2015 Elsevier. (b) Illuminated current-voltage data of ZnO/CNDs/CuSCN solar cells using a single layer of organic product-based CNDs. (c) Images of illuminated current-voltage data of ZnO/CNDs/CuSCN solar cells using 4-layer LbL coating. (d) Schematic diagram of the preparation of CNDs sensitized solar cells by hydrothermal carbonization of organic product precursors to go binding with chemically grown ZnO nanorods. Images of (b), (d), and (d) are reproduced with permission of reference [139]. Copyright © 2015 Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.



**Table 6** Light-harvesting efficiency (LHE), power conversion efficiency (PCE), and mean internal efficiency of as-prepared solar cells with commercial TiO<sub>2</sub> (P25). The table is used with the permission of reference [3].

Sample	LHE (400–800 nm) %	Average internal efficiency %	PCE/%
Chitosan CNDs+ P25	16.3	1.02 ± 0.05	0.167 ± 0.008
Chitin CNDs+ P25	11.6	1.20 ± 0.02	0.139 ± 0.002
D-Glucose CNDs+ P25	50.0	0.20 ± 0.02	0.103 ± 0.008
L-Arginine CNDs+ P25	23.6	1.53 ± 0.03	0.362 ± 0.007
L-Cysteine CNDs+ P25	24.5	1.39 ± 0.04	0.34 ± 0.01
Lobster CNDs+ P25	20.8	1.04 ± 0.04	0.216 ± 0.008

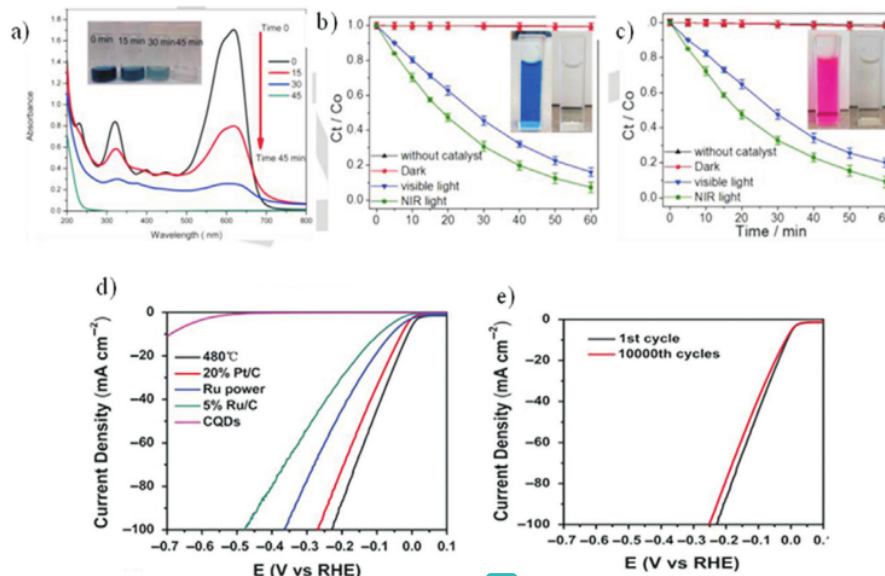
ically grown ZnO nanorods are displayed in Fig. 19d. The efficiency of the devices relies on which functional groups were present in the synthesized CNDs surface. This precursor-dependent solar cell efficacy also indicated that the functionalization signifies the binding of CNDs to the ZnO surface and the characteristic transport of the charge in the layer, which affects the solar cell efficiency.

#### 4.5. Catalysis

In addition to their bioimaging, sensing, and solar cells applications, the organic products derived-CNDs have played significant roles in the catalysis and electrolysis fields. Photoactive CNDs have been shown to serve as both excellent electron donors and acceptors, as revealed by electron transfer with CNDs, which can be induced by photoexcitation.<sup>184</sup> The organic products-based CNDs can enhance the photocatalytic production of hydrogen. CNDs doped with other additives, namely

composited CNDs, are applicable in photocatalysis and electrocatalysis.<sup>22</sup> Prasannan *et al.*<sup>185</sup> used a composite of ZnO, and CNDs synthesized from orange peel using a facile hydrothermal approach to be used as a photocatalyst for the deterioration of dyes (Fig. 20a). The ZnO/CNDs hybrid materials were synthesized through the solution-dispersible method. The naphthol blue-black azo dye was nearly degraded in 45 min by the ZnO/CNDs catalyst with UV irradiation. In contrast with using only ZnO or CNDs as the catalyst, the degradation was determined to be 84.3 % and 4.4 %, respectively, after irradiation took place for the same time. The stronger photocatalytic feature of the ZnO/CNDs composite was responsible for the electronic charge interaction. The synthetic process of ZnO/CNDs was successfully completed, which might become remarkably high on the transfer of the photogenerated matters. Although other CNDs/metal photocatalyst methods have been used in the organic compound deterioration, there might be some drawbacks with these CNDs/metal hybrid photocatalyst methods. Firstly, they are potentially toxic and expensive, which confines their future uses in industry. Secondly, it is likely to have metal-containing photocatalysts aggregation in accordance with high surface energies, thereby leading to a significant loss of catalytic activity.

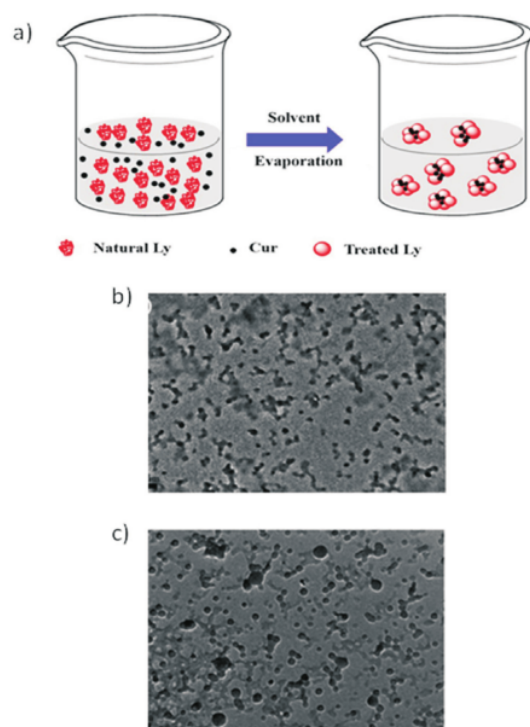
Wang *et al.*<sup>186</sup> prepared novel non-metal photocatalysts made up of CNDs, using the hydrothermal method, from glucose inserted in a carbon matrix to overcome these issues. These photocatalysts could degrade organic dyes (methylene blue and rhodamine B) in the range of UV-vis light and NIR radiation (Fig. 20b,c). The application of solar renewable energy to generate hydrogen by decomposing water in a photocatalytic process gives a favourable model to overcome worldwide energy shortage.



**Figure 20** (a) Diagrammatic model of time-course absorption variable intensity of naphthol blue-black azo dye over CNDs/ZnO composite at various irradiation intervals and correlating irradiated solutions. Image (a) is reprinted from reference [185]. Copyright © 2015 American Chemical Society. (b) Photodegraded-profile methylene blue ( $C_0 = 0.1$  mM). (c) Rhodamine B profile ( $C_0 = 0.1$  mM) was photodegraded in the absence or presence of CNDs-based carbon catalysts ( $0.02$  mg mL<sup>-1</sup>) under various irradiation conditions at 21 °C. The inset pictures in (b) and (c) show images of methylene blue aqueous solutions and rhodamine B aqueous solutions before and after photodegradation, respectively. Images (b) and (c) are reprinted from reference [186]. Copyright © 2015 American Chemical Society. (d) HER polarization curves for 20 % commercial Pt/C, Ru powder, 5 % commercial Ru/C, and the Ru-CNDs as well as CNDs. (e) Stability test for Ru-CNDs in 1M KOH before and after 10,000 cycles. Images (d) and (e) are reproduced with the permission of reference [187]. Copyright © 2018 Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

Li *et al.*<sup>187</sup> reported the novel carbon-loaded ruthenium CNDs electrocatalysts (Ru-CNDs) for hydrogen evolution reaction (HER) using CNDs prepared from ginkgo leaves. The HER electrocatalytic efficiency of CNDs loaded with Ru (Ru-CNDs) is more profitable than of CNDs loaded with other noble metals, non-noble metals or non-metallic materials. Ru-CNDs are catalytically more resistant than Pt/C in alkaline solutions. Thus, the catalytic performance of the Ru-CNDs formation was potentially explored as a new alternative design of hybridizing CNDs/metal, with remarkable potential for HER uses. HER polarization curves for Ru-CNDs and CNDs, 20 % commercial Pt/C, Ru powder, 5 % commercial Ru/C, and a stability test for Ru-CNDs in 1M KOH before and after 10000 cycles are shown in Fig. 21d–e. Qin *et al.*<sup>113</sup> also found that HTC (high-temperature condition) of cost-effective left-over of willow bark led to high photoluminescence and permeable CNDs. Such CNDs were chosen for the application as excellent photo-catalysis for the synchronous decreasing of Au complexes and graphene oxides (GOs) to produce Au nanoparticle-decorated reduced graphene oxide (AuNP/rGO) nanocomposites. Subsequently, a mixture of GO and HAuCl<sub>4</sub> aqueous solution was irradiated by UV light to find the trace of CNDs in a mixture.

Tyagi *et al.*<sup>188</sup> reported green CNDs from the waste material of lemon peel through hydrothermal treatment. The photocatalytic efficacy of immobilized CNDs on electrospun TiO<sub>2</sub> nanofibres (TiO<sub>2</sub>-CNDs composites) was illustrated by using methylene blue (MB) dye as a model pollutant agent. The photocatalytic performance of the TiO<sub>2</sub>-CNDs composite is around 2.5 times higher than that of TiO<sub>2</sub> nanofibres.



**Figure 21** (a) Plausible encapsulated scheme of Cur-based on Ly-NPs. (b) TEM images of free Ly-NPs. (c) Cur-loaded Ly-NPs (scale bar = 0.2  $\mu$ m). Images were reprinted from reference [198]. Copyright © 2019, the authors. *Food Science & Nutrition*, published by Wiley Periodicals, Inc.

## 5. Nanoencapsulation

Recently, nanotechnology has extensively drawn interest in studying food science. The bioactive compounds encapsulated study is one of the most attractive systems for many researchers.<sup>189</sup> Nanoencapsulation consists of coating for bioactive compounds and can significantly affect their mechanical properties, bioavailability and stability.<sup>190,191</sup> Since health-giving or harmless foodstuffs are demanded by consumers, scientists or researchers have paid a lot of attention to modifying or synthesizing bioactive compounds with natural ingredients. Phenolic compounds, essential oils, essential fatty acids, carotenoids and insoluble vitamins are bioactive compounds that help human well-being.<sup>192</sup> However, these bioactive compounds, such as phenolic acid and coumarins, flavonoid derivatives are mostly hydrophobic and poorly soluble.<sup>193–195</sup> Mostly, an unpleasant or bitter sense of taste belongs to many of those molecules, subsequently consuming limited food or oral treatment. To diminish these leading issues, a nanoencapsulation of delivery systems has been extensively attractive as an alternative food science technology method.<sup>193</sup> Thus, the nanoencapsulated materials can deliver these defectively hydrophobic, soluble, and bio-available compounds into harmless foodstuffs. The nanoencapsulation enhances their absorption efficiency into cellular structures with agreeable particle compositions. The considerable properties of encapsulated form, size, and surface enable to improve nanoparticles' solubility, upgrading the rate of absorption and release, influencing gastrointestinal dispersion, and preventing metabolic deterioration.<sup>196</sup> Recently, various nanoencapsulated methods and nanocarriers of bioactive compounds, for example, nanosuspensions, nanoemulsions, liposomes and nanometer phytosomes, nanostructured lipid transporters, solid lipid nanoparticles (SLNs), biopolymer SLNs, micelles made of proteins and polysaccharides, have been studied.<sup>197</sup>

Cui *et al.*<sup>198</sup> prepared curcumin encapsulated on lysozyme nanoparticles by the de-solvation method. The synthesized lysozyme nanoparticles (Ly-NPs) was of the average size distribution of  $127.9 \pm 2.12$  nm. Curcumin is regarded as a physiologically and pharmacologically functional nutritional component. According to TEM analysis, curcumin (Cur) influenced the physical structure or size of Ly-NPs after curcumin encapsulated Ly-NPs. Significantly, the Cur packaged Ly-NPs displayed better dispersion in contrast with Ly-NPs alone, as shown in Fig. 21b,c. Moreover, packaged curcumin exhibited considerably enhanced survival rate after heat treatment in contrast with free curcumin. DPPH investigation also approved that packaged curcumin, indicating favourable antioxidant capability after 50 °C treatment. Thus, encapsulated curcumin in Ly-NPs is applicable for the encapsulation and protection of curcumin.<sup>198</sup>

Fuciños *et al.*<sup>81</sup> have reported the caffeine-encapsulated preparation into  $\alpha$ -lactalbumin nanotubes through nanotube synthesis. These caffeine-loaded nanotubes were synthesized by adding caffeine into the processing mixture before the nanotube became trapped for self-assembly. The results showed that  $\alpha$ -lactalbumin ( $\alpha$ -LA) in nanotubes prepared in the existence of Mn<sup>2+</sup> were strongly stable while freeze-drying. The encapsulated efficacy was around 100 %, meaning caffeine loaded  $\alpha$ -LA nanotubes were promising. Caffeine release from  $\alpha$ -LA nanotubes was optimal under low heating rate and strong pH.<sup>81</sup>

Basiri *et al.*<sup>199</sup> prepared nanoencapsulated  $\alpha$ -tocopherol-loaded noisome through the modified heating system, using surfactants – span60 and tween60. Their findings showed that noisome played a crucial role in efficacy for encapsulated- $\alpha$ -TOC and combining Span 60 and Tween 60, forming a favour-

able  $\alpha$ -TOC-encapsulated nanosomia. The findings reported that  $\alpha$ -TOC loaded niosomes had on average, a narrow size distribution ( $73.85 \pm 0.6$ – $186 \pm 0.58$  nm), and high encapsulation efficiency ( $61.13 \pm 0.52$ – $98.92 \pm 0.92$ ). Since the sizeable hydrophobic moiety (HLB) decreased, the encapsulation efficiency (EE) and the niosomes durability was effectively improved.<sup>199</sup>

El-Fattah *et al.*<sup>200</sup> reported the phytosome nanoparticles-encapsulated quercetin by using thin-film hydration approach. They found a highly encapsulated efficacy of 98.4 %. Phytosome nanocapsules improved the remedial advantage of quercetin in ovariectomized rats.<sup>200</sup>

Najafi *et al.*<sup>201</sup> prepared the achillea millefolium-encapsulated nanophytosome to assess the post-thawing sperm quality and evaluate the oxidative signal of rooster semen. They aimed to analyze the content of antioxidants of achillea millefolium extract (AmE), and AmE loaded in nanophytosome under the freezing condition of Ross 308 rooster semen. They found that AmE-encapsulated nanophytosome at  $3 \text{ mg L}^{-1}$  doses can improve rooster sperm motility, feasibility and inconvenient oxidative values in the freezing.<sup>201</sup>

Arroyo-Maya *et al.*<sup>202</sup> have reported nanoencapsulation of anthocyanins-loaded biopolymer nanoparticles (nanospheres), through a complicated thermal and electrostatic method using a whey protein–pectin combination. The nano-loaded average sizes were as low as 200 nm in diameter. These nanospheres release 55 % of the moderate loading capacity before heating. These nanoencapsulated materials improved thermal feasibility and reduced antioxidant performance.<sup>202</sup>

Gunes *et al.*<sup>203</sup> prepared niosomal formulations of methanol and hydro methanol extracts produced from *N. oleander* roots using a thin-film hydration technique (as shown in Fig. 22). Total phenolic capacity in the methanol oleander extract (MOE) and hydro methanol oleander extracts (MOWE) was calculated to be  $64.51 \pm 0.945 \mu\text{g mg}^{-1}$  and  $65.05 \pm 0.37 \mu\text{g mg}^{-1}$  gallic acid equivalents, respectively. More interestingly, the vesicles' encapsulated efficacy was found to be 16.2 % for MON (in the presence of MOE) and 13.24 % for MWON (in the presence of MOWE). All of the dose concentrations of methanol oleander extract (MOE), and methanol oleander niosomal formulations (MON) inhibited cell HeLa and A549 cells, thereby reducing cell viabilities. Moreover, cell imaging studies indicated that the MON could readily enter the HeLa and A549 cells *via* endocytosis and grow around the nucleus.<sup>203</sup>

## 6. Conclusions and Future Perspectives

This review reveals different synthetic methods, properties, and applications of CNDs derived from organic products. Moreover, nanoencapsulation is also briefly studied. The current paper gives a systematic overview of converting natural organic products into green CNDs and describes the physicochemical properties of these CNDs. Furthermore, major applications of the organic products derived-CNDs, for example, sensing and detection, bioimaging, drug delivery, solar cells, and photocatalysis, nanoencapsulation of some bioactive compounds are extensively reviewed. Natural products derived-CNDs have some important advantages—firstly; the organic precursors that come from diverse carbon sources of biomass are renewable, inexpensive, easy, and abundant, produced in high yield and good biocompatibility. Secondly, the organic products of carbon sources are intrinsically doped with hetero-atoms, making it easy to prepare heteroatom-doped CNDs, which exhibit excellent photoluminescence.

Additionally, they can be used in biomedical applications because excellent optical properties and red fluorescence emis-

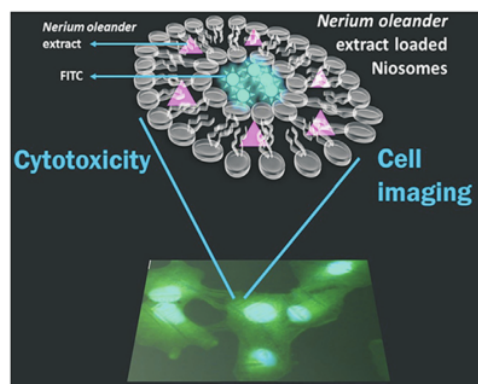


Figure 22 Schematic illustration of *Nerium oleander* extract loaded niosomes *in vitro* evaluation of bioactive nano vesicular structures. The image is reproduced with permission of reference [203]. Copyright © 2017 Elsevier.

sion of green CNDs enhanced by hetero-atom doping contents greatly attract every researcher. Lastly, the synthetic approaches used to synthesize CNDs are simple, fast, and eco-friendly to some degree. Thus, organic products are likely attractive starting carbon sources in the synthesis of green and efficient CNDs. Many studies have also paid attention to developing new natural biological wastes, new prepared approaches, new applications, and better materials and cost-effective equipment.

Despite having many advantages, there are still several challenges concerning organic products-derived CNDs, some of which are considered below.

*Challenge of short-wavelength absorption and emission:* currently, the organic products-based CNDs of different sizes emitting disparate fluorescence have been successfully synthesized from many organic products (foodstuffs and other valuable bioresources), using a variety of methods. The ranges of their emission spectra of CNDs are mostly in the ultraviolet/visible region's position. Consequently, those ranges are not extended enough for use in most biomedical fields. Because the short UV light harms or destroys DNA and protein, visible light cannot go through into deep tissues for photoimaging. Near infra-red (NIR) light would be better for clinical uses since NIR light can deeply enter tissues and diminish side effects concerning biological toxicity.<sup>204</sup> In future, much effort should be put into synthesizing CNDs with absorbance and emission in the NIR region. The short-wavelength absorbance of CNDs might adversely influence their photocatalytic performance. Light-harvesting ability is limited by the narrow absorption ranges of CNDs, which means that it would be advantageous to broaden the absorption length. More photons can be absorbed to increase their efficiency in photocatalytic materials, renewable energy storage cells.

*The synthesis of CNDs with multi-photon fluorescence and up-conversion fluorescence:* multiphoton photoluminescence and up-conversion fluorescence are interesting aspects, in which CNDs emit visible fluorescence emission with NIR light. However, the phenomenon of up-converting emission remains complicated, and further investigations are necessary for it to identify this in detail. The up-converting emission of CNDs is potentially applied not only for bioimaging but also for a suitable replacement for lanthanide-doped up-converting nanoparticles (UCNPs) in the manufacture of NIR-responsive matters since the organic products-based CNDs are more biocompatible than lanthanide-doped up-converting nanoparticles (UCNPs).

There are more biomedical and other applications of CNDs, such as *in vivo* bioimaging, photodynamic therapy, drug delivery, sensing, and nanocomposite hydrogels for tissue engineering.<sup>205</sup> Despite many proven advantages in natural product-derived CNDs, studies of producing the great potential of CNDs are still challenging. The authors of the current paper hope that this review will inspire exciting ideas for further applications.

The synthesis of CNDs with room-temperature phosphorescence: The spin-forbidden nature of triplet exciton transitions and non-radiative decay processes hinder the phosphorescence of CNDs. In this way, the ambient temperature environment for the synthetic process of CNDs is highly recommended to attain profitable phosphorescence. Many synthetic routes have been investigated to achieve metal-free room temperature phosphorescence (RTP). More urgently and importantly, the metal-free CNDs of RTP should be maintained by other special organic groups, such as heavy halogen, aromatic carbonyl group, and a deuterated carbon atom. Thus, organic product carbon sources limit their synthesis complexity and chemical diversity. There is a significant need for new metal-free CNDs (pure organic product derived-CNDs) with long-life RTP to be synthesized using facile synthetic methods. We strongly hope that this review will offer several advantages, especially for researchers to prepare organic products-based CNDs that are more interesting and attractive with excellent properties and potential applications.

**Nanoencapsulation advantages and disadvantages:** benefits of nanoencapsulation include improved safety against pH, light, wetness, and gastric digestive system; it covers-up bitter taste or aromatic taste, enhance releasing capacity; promotes the soluble level of lipophilic compounds in aqueous condition. It supports the long-lasting absorption of nutrients and can develop loading nanomaterials with antimicrobial and antioxidant polyphenolic compounds. However, nanoencapsulation still has drawbacks concerning their toxicological effects on the safety of human consumption and the environment. Finding the best methods to obtain them further and identifying ideal nanomaterials for bioactive compounds is still challenging.

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#### Conflict of Interest

The authors claim no conflict of interest.

#### Abbreviations

AFM	atomic absorption microscopy
BV	blood vessel
CNDs	CNDs
CBBA	4-carboxy-benzylboronic acid
Dox	Doxorubicin
ELC	effective conjugated length
FL	fluorescence
FTIR	Fourier-transform infrared spectroscopy
FRET	Förster resonance energy
GQDs	graphene quantum dots
HS	humic substances
HRTEM	high-resolution transmission electron microscopy
L-NPs	lignin-based nanoparticles
Lis-CNDs	lisinopril-loaded CNDs
LV	lymph vessel

N/S-CNDs	nitrogen/sulphur-doped CNDs
NIR	near-infrared region
OP-CNDs	organic-based CNDs
PEG	polyethylene glycol
PL	photoluminescence
QY	quantum yield
PA	photoacoustic
SLN	sentinel lymph node
TEM	transmission electron microscopy
UV-vis	ultraviolet visible

#### \*ORCID iDs

K. Siimon:  [orcid.org/0000-0001-5430-9992](https://orcid.org/0000-0001-5430-9992)  
 M. Zakki Fahmi:  [orcid.org/0000-0001-5045-1617](https://orcid.org/0000-0001-5045-1617)

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