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A review on carbon quantum dots synthesis from food waste as a carbon source and their applications

M Ramesh babu[1*], Laxmi Mangamma J [1,2], Dharamasoth Rama Devi[3,4], Keloth Basavaiah[1*]

[1]*Department of Inorganic and Analytical Chemistry, Andhra University, Visakhapatnam-530017, India*

[2]*Govt. Degree College, Chodavaram-531036, India*

[3]*AU College of PharmacueticalSciences, Andhra University, Visakhapatnam-530017, India*

[4]*Samuel George Institute of Pharmaceutical Sciences, Markapur, Andhra Pradesh-523316, India*
**e-mail address:* ramesh.dec25@gmail.com*;* *klbasu@gmail.com*

Abstract:
Carbon quantum dots (CQDs) are a new type of carbon nanomaterial's that are novel fluorescent. They are also one of the most important carbon nanomaterial's. In the last few decades, more has been learned about how to make them, how to change them, and where they can be used. If a chemical reaction happens or not, the method of making CQDs from food waste can be divided into physical synthesis and chemical synthesis. Food waste that has enough carbon sources can also be used to make CQDs, but this hasn't gotten much attention. So, it makes sense for CQDs technology to use food waste as a starting point. Due to their good fluorescence performance, CQDs made from food waste can be used for analysis and sensing. Food waste is everywhere in the natural and living environment, and there is a lot of it. Even though a lot of work has been put into making multifunctional CQDs from biomass waste, there are still many problems to solve. First of all, scientists don't fully understand how CQDs give off light. For CQDs made from renewable biomass waste to be used in more places, its photoluminescence mechanism needs to be explained in detail and be widely known.

Keywords:
Carbon quantum dots, Food waste, photoluminescence

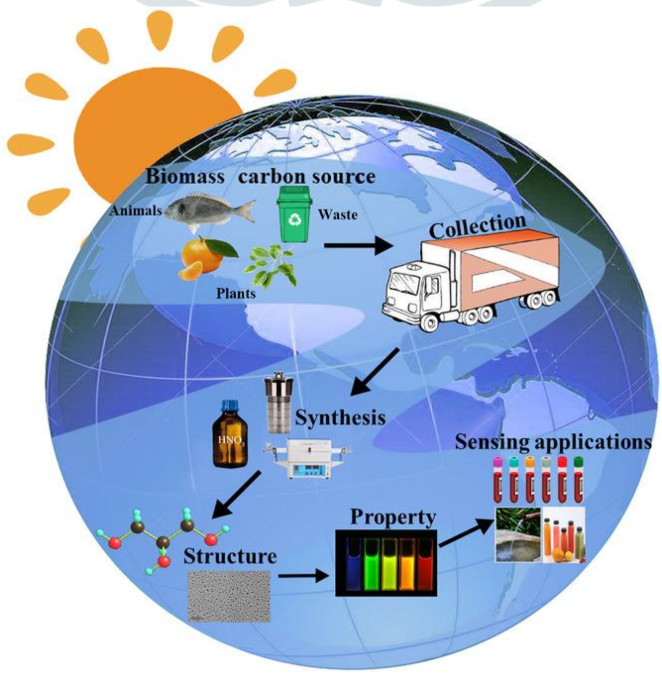
Introduction:

Carbon quantum dots (CQDs) which was discovered by Xu et al is a photo luminescent material with sizes below 10 nm [1]. CQDs are novel fluorescent carbon nanomaterial's and among the most important members of the carbon nanomaterial's family. In general, CQDs are well-dispersed spherical particles with particle size less than 10 nm. Besides the high quantum yield and adjustable emission wavelength, which are also possessed by traditional semiconductor quantum dots, CQDs have many other excellent characteristics, including good photo stability, low cytotoxicity, good biocompatibility, easy surface modification and high chemical inertness, and therefore have attracted considerable scholarly attention in recent years. Hence far, CQDs have been widely used in many fields such as cell imaging [2–4], in vivo imaging [5,6], drug delivery [7–9], fluorescence sensing [10–12], photo catalysis [13–15], multicolour light-emitting diode (LED) production [16,17], energy conversion and storage [18–20], etc. CQDs are gradually become one of the research hotspots in the above-mentioned fields and considered as a potential substitute for semiconductor quantum dots.

In the past few decades their synthesis routes, modification methods and application fields have been further developed [21]. Thus, in this review we aimed to explore the use of waste and by-products from different sources in the CQDs synthesis, their potentials, applications in various fields and advantages, as well as presenting current challenges in the field of study. CQDs' synthesis methods include top-down and bottom-up synthetic routes. The top-down synthetic route breaks down the feedstock with a larger carbon structure. The bottom-up method preferred for synthesis of CQDs because it uses simple natural materials such as fresh tomato [22], garlic [23] green tea [24] or some food waste material as sources without any toxic substances.

Synthesis method:

The synthesis method of CQDs from food waste can be divided into physical synthesis and chemical synthesis according to whether the chemical reaction takes place or not. Physical synthesis mainly includes vapour deposition, grinding, ultrasonic synthesis, molecular epitaxy, and photo etching. Compared with the chemical synthesis, the physical synthesis may cause many defects on the material surface, the destruction of the morphology and the easy contamination of the material itself in the preparation process. Therefore, chemical synthesis has been more used by Researchers. The following will focus on the different chemical synthesis methods including hydrothermal method, solvothermal method, chemical oxidation method and pyrolysis method.



Schematic representation of carbon source from biomass.

Hydrothermal and solvothermal method:

Hydrothermal synthesis is a chemical reaction method in aqueous solution under the conditions of temperature of 100 ∼ 1000 ◦C and pressure of 1 MPa ∼ 1 GPa. The advantage of hydrothermal synthesis is that the ions are evenly mixed in aqueous solution, so the product has high purity, good dispersion, and easy particle size control. Solvothermal method is developed based on hydrothermal method. It differs from hydrothermal method in that the solvents used are organic solvents rather than water. Under liquid phase or supercritical conditions, the reactants dispersed in the solution become more active and the products are formed slowly. The process is relatively simple and easy to control, which can effectively prevent the volatilization of toxic substances and prepare air-sensitive precursors in closed systems. However, the disadvantages of the method are low yield, insufficient product purity, and unsatisfactory uniformity of product size and morphology. Generally speaking, hydrothermal has no negative effect on the properties of materials, but solvothermal has a significant influence on the properties of materials due to different solvents, so hydrothermal method is more common than solvothermal method in the synthesis

Other methods:

In addition to the two methods introduced above, chemical oxidation and pyrolysis methods have also been reported for the preparation of carbon dots from biomass. Chemical oxidation is the method in which the oxidant oxidizes the target by losing electrons. The advantages of this method are mild reaction conditions, easy to control, convenient operation and high selectivity.

Commonly used experimental design to synthesis CQDs:

Drying process should be followed by ashing, which involves subjecting the dried food sample to (300-600 ° C) for 2-3hrs They can be performed by treating 50W or more microwave for 45min-120min or performing under 5W or more ultrasonic irradiation for 45min-180min.Then it should be stirred for 10-30min.before stirring it should be dissolved with any of the solvents like water, alcohol, ethanol & ether. The amount of the applied alcohol is preferably 5 times to 10 times an amount of the food waste residues. The filtering step includes performing vacuum filtering which should be stirred together with the solvent, using a filter paper, continued by filtering through a membrane filter. The concentrating step is carried out by the filtrate using a rotary evaporator.

Methods of synthesis of CQDs using various food waste materials:

Nowadays food losses and food waste (FLW) has turned into a worldwide serious problem. According to the Food and Agriculture Organization (FAO) statistics, about one-third of the world's food for human consumption (or 1.3 billion tons) is lost or wasted ever year. Food waste which possesses enough carbon sources is also a good material for synthesis of CQDs but has not received much attention. Hence it is reasonable to utilize food waste as starting material in CQDs technology.

Plant By-products

Plant by-products are suitable CQDs' starting materials due to their biocompatibility, low cost and low toxicity [25]. The most common parts of plant by-products are plant peels and plant shells [26].

Fruit by-products

There are numerous studies which used fruit by-products as starting materials, such as preparation of CQDs from mangosteen peel [27](Aji, Susanto, Wiguna, & Sulhadi, 2017), pineapple peel [28](Vandarkuzhali, et al., 2018) and grapefruit peel [29](Xiao, et al., 2018). JJ Zhou, et al. published the synthesis of CQDs using watermelon peel[30]. *Carica papaya* waste pulp is a good source to prepare CQDs. Pooja D and colleagues produced CQDs from papaya waste[31]. In another study fresh yellow banana peels as CQDs starting material[32].

Vegetable by-products

Some experiments used the by-products of vegetables to synthesize CQDs. Bandi et al. used onion waste to produce CQDs[33].

Cereal by-products

Grains for human food are ground into powder to refine cereals without bran and germ in order to meet consumers' food needs. Thus fine processing will bring tons of by-products and they are also suitable natural resources to synthesis CQDs.

Nut by-products

The peanut shell waste is cheap, renewable and green resource which is suitable for the production of CQDs as the starting material [34]. Palm shell was also a splendid starting material to prepare different kinds CQDs [35].

Coffee by-products

Zhang, et al., used mild molecular aggregation method to prepare CQDs from coffee bean shells-derived [36].

By-products of Animal Food

Most of the by-products from fish, livestock and poultry are discarded because their high water content lead to deterioration. By-products of animal principally include skin and shell which contain abundant carbon. Therefore, they are appropriately regarded as starting materials to participate in preparation of CQDs.

Although a small amount of egg shells are used as a fertilizer, most of them are abandoned as waste [37]. In another study egg-shells as starting material were used to prepare CQDs [38]. Crab shells are rich in proteins and nitrogen which is suitable for preparing CQDs [21]. The waste of shrimp was regarded as another excellent starting material to prepare CQDs.

By-products in Food Processing

The synthesis of CQDs from daily food waste are widely used nowadays. Carbon dots from yogurt, honey was synthesized via microwave heating [39,40]. Apart from these, De and Karak reported the synthesis of carbon dots from banana juice via heating 150°C for 4 h [41] and Baruah et al. showed the synthesis of carbon dots from Assam tea via heating at 200°C for 10 h totally [42].

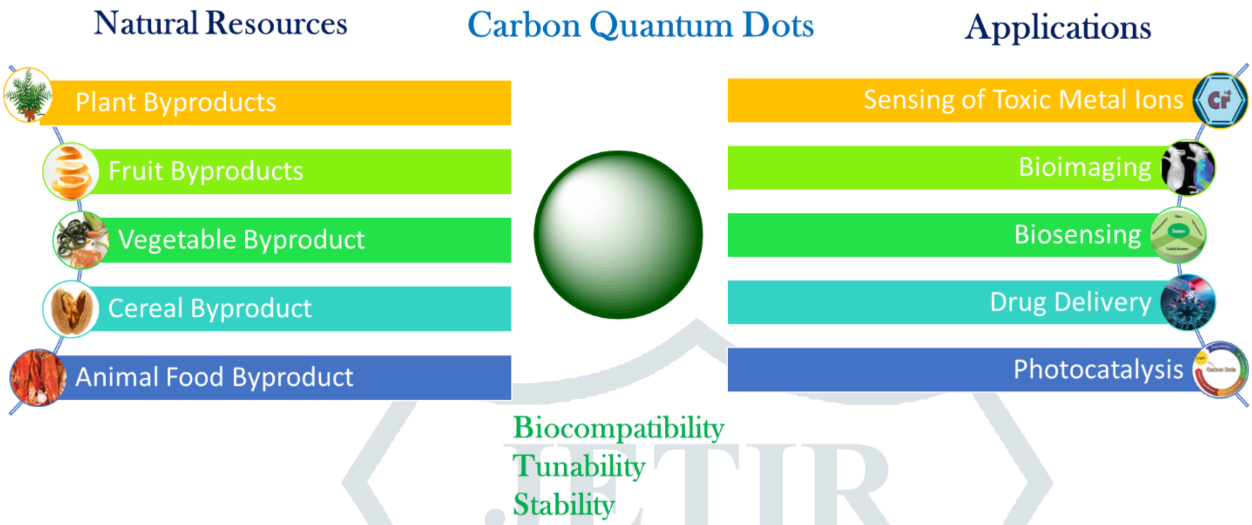


Fig.2: Schematic representation of carbon quantum dots from natural sources and applications.

Table-1: Carbon quantum dots synthesised from various food waste materials as carbon sources and their properties.

S. No.	Carbon source	Method	Size [nm]	Ref.
1.	Orange waste peels	Hydrothermal	2.9	[43]
2.	Cane sugar	Solvothermal	9.5	[44]
3.	Mushroom	Microwave	20	[45]
4.	Lemon peel	Hydrothermal	6–14	[46]
5.	Tea, Peanut Shell	Hydrothermal	7–9	[47]
6.	Fenugreek seed	Hydrothermal	5.6	[48]
7.	Rice grains	Pyrolysis	2–6.5	[49]
8.	Roasted fish	Hydrothermal	2.73	[50]
9.	Cow milk	Hydrothermal	2.8	[51]
10.	Cow milk	Microwave	5	[52]
11.	Crab Shell	Microwave - hydrothermal	4.0	[53]

Applications of CQDs from Food Waste in Food Safety Detection:

CQDs synthesised from food waste can realize its value in analysis and sensing due to excellent fluorescence performance. They have many functional groups on the surface compared with non-biomass carbon dots, some substances including metal ions and non-metal molecules can interact with these groups to change their fluorescence properties. Based on the quenching mechanism, the quantitative analysis of various target detection can be achieved.

Sensing of metal ions:

Iron (Fe) is an indispensable element for haemoglobin. Iron deficiency will reduce haemoglobin synthesis and suffering from anaemia for the human body. At present, more and more researchers have applied the synthesized biomass carbon dots to the detection of metal ions. The carbon dots prepared using poplar leaves,[54] lemon juice,[55] pear juice,[56] corncob,[57] and alkali lignin[58] as carbon sources could be used to detect iron ions.

Detection of Heavy Metal Ions:

As heavy metal pollution escalates, toxicity to humans is also of particular concern. Athika et al. fabricated a kind of CQDs from spoiled milk [59]. The result exhibited this CQDs had ability to detect Cr^{6+}. Sugarcane molasses was extracted from industrial waste residue after sugar purification. Huang et al. prepared the CQDs with this waste [60]. The result suggested the decrease of PL intensity had a great linear relationship with Fe^{3+} concentration

Biocompatible carbon dots synthesized from yogurt via microwave synthesis successfully used in imaging of colon epithelial cells [61]. Toxicological assessment of yogurt carbon dots showed no toxic effects on healthy CoN cells and MCF-7 breast cancer cells up to 7.1 mg/mL carbon dot concentration. Fluorescent carbon dots synthesized from pomegranate fruits using hydrothermal method were used in bio imaging of *Pseudomonas aeruginosa* and *Fusariumavenaceum*. Carbon dots from citric acid and glucosamine using domestic pressure cooker. Resulting carbon dots with bright, stable and wavelength-dependent fluorescence were utilized in imaging of mice embryonic fibroblast cells with almost no cytotoxic effect. Carbon dots originated from honey exhibited successful performance in sensitive and selective detection of Fe^{3+}[62].

Table-2: An overview of several applications of carbon quantum dots obtained from various food wastes as source

Application Field	Food Waste material	Method	Application	Ref.
Sensing	Mango peels	Hydrothermal	mesotrione detection	[63]
	Palm shell waste	ultrasonic	nitrophenol detection	[64]
	Waste tea residue	chemical oxidation	tetracycline detection	[65]
Imaging	Onion waste	Hydrothermal	multicolor imaging and Fe3+ detection	[66]
	Banana peel waste	Hydrothermal	in vivo bioimaging	[67]
	Food-waste	Ultrasonic	in vitro bioimaging	[68]
	Walnut shells	carbonization and chemical cutting	intracellular bioimaging	[69]
Drug delivery	Wheat bran	Hydrothermal	drug delivery	[70]
	Crab shells	Microwave	drug delivery and targeted dual-modality bioimaging	[71]
Photocatalysis	Waste frying oil	Hydrothermal	Photocatalysis	[72]
	Orange peels	Hydrothermal	Photocatalysis	[73]
	Bitter apple peels	Pyrolysis	Photocatalysis	[74]
	Lemon peel waste	Hydrothermal	photocatalysis and sensing	[75]
	Waste food	Hydrothermal	Light-emitting diodes	[76]
	Pineapple peels	Hydrothermal	electronic security devices	[77]

			and as a memory element	
Others	Onion peels	Microwave	accelerated skin wound healing and live-cell imaging	[78]
	Waste tea residue	Carbonization	used as growth plant stimulator	[79]

Conclusions:

Biomass waste is abundant and widely distributed in the natural and living environment. Herein we have reviewed the methods for preparing fluorescent CQDs from biomass waste, analysed the main factors affecting the fluorescence intensity of CQDs during the synthetic process, and finally discussed the properties and applications of CQDs obtained from biomass waste. The advantages of synthesizing CQDs from biomass waste include easy availability of carbon source, simplicity in preparation and feasibility of large-scale production. Compared with CQDs produced from chemical agents, some CQDs from biomass waste can be self-passivated during the synthetic process because the abundant presence of carbonaceous compounds in biomass waste enables carbonization and surface passivation to occur simultaneously, and heteroatom-containing compounds or long-chain compounds present in biomass waste can act as surface passivation agents. CQDs prepared from biomass waste have relatively high fluorescence quantum yield, good photo stability, high photo catalytic activity, excellent biocompatibility and low cytotoxicity and therefore have a wide range of promising applications in many areas such as bio imaging, drug delivery, ion sensing and photo catalysis.

Future Outlook:

Although much effort has been devoted to the preparation of multifunctional CQDs from biomass waste, many challenges still exist. First of all, the photoluminescence mechanism of CQDs has not been fully understood. A detailed and widely recognized photoluminescence mechanism of CQDs is highly needed to expand the application of CQDs from renewable biomass waste. Second, it is still a big challenge to realize the large-scale production of high-quality CQDs from biomass waste. The solubility of CQDs in solvents and their potential applications are closely related to the types and extent of surface functional groups upon CQDs. Unfortunately, how to precisely control surface functionalization of CQDs during the synthetic process is also an exasperating problem. The surface of CQDs obtained from biomass wastes are generally functionalized with carboxyl, hydroxyl and amino groups, which endow CQDs with properties of good solubility in water and rapid precipitation in other solvents but may also quench the fluorescence of CQDs. The use of CQDs as fluorescence probes in the quantitative analysis of metal ions or other substances is based on the fluorescence-quenching mechanism. However, there are many factors other than analytes of interest that can quench the fluorescence of CQDs. CQDs with excellent selectivity for the analytes of interest are therefore highly desired. The maximal absorption wavelengths of most CQDs are located in the wavelength range from the ultraviolet to the blue, which limits their application in the field of bio imaging. The preparation of far-red to near-infrared CQDs is becoming a new research hotspot.

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