

Carbon nano-dots from natural resources as optical sensors for iron ions/Fe³⁺: a review

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Abstract: Carbon nanodots (CDs) are a new category of carbon nanomaterials with sizes under 10 nm, displaying a range of fascinating properties. Broadly speaking, they can be described as small, surface functionalized carbonaceous nanoparticles characterized by an intense and tunable fluorescence, a marked sensitivity to the environment and a range of interesting photochemical properties. CDs are currently the subject of very intense research, motivated by their possible applications in many fields, including bio-imaging, nano-sensing and photo-catalysis. Also, CDs have really been synthesized from a large variety of precursors, natural resources and some waste materials. There are many methods for the synthesis of CDs for example Arc Discharge, Laser ablation, Chemical Oxidation and Hydrothermal Synthesis. Hydrothermal Synthesis method was the most commonly used in the synthesis of CDs. Since the setup is simple and with high Quantum Yield, the result particle is almost uniform in size. In this work, we searched for the different natural resources of CDs and identify their synthesis methods, properties and some of their applications, 69 natural sources of CDs were found. We found nearly 21 natural resources of CDs for sensing iron ions Fe³⁺, that has a great importance in monitoring iron levels in the water, whose excess may cause many diseases.

Keywords: carbon nanodots; fluorescence ; synthesis methods; hydrothermal ; natural resources ; bio-imaging ; sensing.

1. Introduction

Issues regarding global energy and environment have driven the development of new materials and techniques in order to minimize financial and environmental costs [1]. The speedy development of nanomaterials has now not solely resulted in numerous benefits to many science and engineering fields, however has moreover brought some new issues. Among these troubles is that the synthesis of nanomaterials commonly requires high energy consumption and use of toxic chemical reagents, which no longer solely increases production prices, however is the opposite of the core standards of sustainable development. Therefore, it is critical to make use of environmental-friendly and renewable raw resources to handle the challenges confronted via the use of the sustainable manufacturing of nanomaterials without the use of poisonous chemicals [2][3]. Carbon nanodots (CDs) are a new category of carbon nanomaterials with sizes under 10 nm [4] They first received throughout the purification of single walled carbon nanotubes via preparative electrophoresis in 2004 [5]. CDs have step by step grow to be a rising celebrity in the nano carbon family, due to their benign, plentiful and cheaper nature [4]. Carbon is oftentimes a black material, and until these days was generally viewed to have low solubility in water and vulnerable fluorescence [6]. The most important purpose why such tiny CDs have currently

attracted vast interest is due to the fact of their strong fluorescence, for which they are referred to as fluorescent carbon [7].

Over the last decade, a broad range of techniques have been proposed for the preparation of CDs [8]. Synthesis methods for CDs are based on cutting larger carbon materials (top-down) or fusing smaller (bottom-up) precursor molecules [9].

Top-down approach refers to breaking down larger carbon structures via chemical oxidation, arc discharge, laser ablation and extraction [10]. However, drawbacks of this approach includes the requirement of expensive materials, harsh reaction conditions, and long reaction time [8]. On the other hand, the bottom-up approach refers to the conversion of smaller carbon structure (polymerization and carbonization by chemical reaction) into CDs of the desired size. This bottom-up approach is consisting of combustion routes, microwave pyrolysis, hydrothermal synthesis and electrochemical methods to synthesize CDs [11].

CDs have really been synthesized from a large variety of precursors, mostly man-made, such as graphite [12], fullerene [13], or natural resources such as orange juice [14], coffee grounds [15], egg white [16], banana [17]. In addition, some waste materials have also been well used for the synthesis of CDs for instance, paper pieces [18], which not only reduces CDs production costs but also abates environmental pollution. CDs have become a hot topic of numerous scientific studies since their discovery due to their obvious advantages over conventional semiconductor quantum dots whose notable toxicity and environmental hazards [19]. Compared to florescent organic dyes and genetically engineered fluorescent proteins, CDs excel themselves with significant advantages like high PL quantum yield, photo-stability, resistance to metabolic degradation and so on, which endows the huge potential of carbon dots based bio-applications [20]. CDs have many physical and chemical properties such as Components and structure [21], Dispersibility [22], Cytotoxicity [23] and Optical Properties [21] for example Absorbance [24] and Fluorescence [4]. Dual functional fluorescence Nano sensors have many potential applications in biology and medicine such as Optronics [25], Bio-imaging [26], Photocatalysis [21] and Sensing [27]. Where monitoring temperature with higher precision at localized small length scales or in a Nano cavity is a necessity in various applications. As well as the detection of biologically interesting metal ions using low-cost and sensitive approach is of great importance in bio analysis [28], A wide variety of biological/chemical sensors were developed based on the fluorescence properties and surface functional groups of CDs, such as the detection of Hg^{2+} , Cu^{2+} , Fe^{3+} , Ag^{1+} , Cd^{2+} , etc. [29]. Iron homeostasis problems are one of the utmost everyday diseases of people and cowl an expansive range of diseases with a number of signs and symptoms, starting from anemia to excesses of iron, liver and kidney diseases, diabetes Mellitus, cardiovascular sickness and very probably to neurodegenerative disorders [28].

The most frequent oxidation states of iron are Fe^{3+} and Fe^{2+} vigorously altering to one another, which makes it challenging detecting Fe^{3+} or Fe^{2+} ions only. However, considering an easy, precise, and field-appropriate detection method for total iron (Fe^{3+} and/or Fe^{2+} ions) probing is via a ways from super significance for the analysis of iron elaborating in medical diagnosis, environmental monitoring of water nice and other quality control examinations [23-25]. The most ordinary method for the detection of iron ions consists of voltammetry, electron paramagnetic resonance, spectrophotometry, atomic absorption spectrometry, inductively coupled plasma-atomic emission spectroscopy and inductively coupled plasma mass spectrometry. The cited techniques favor elaborate instrumentation, tedious sample preparation, lengthy procedures, and professional personnel that restricts their hobbies contrast and are inapplicable for most laboratories [21-26]. Among all detection methods, fluorescence spectrometry has obtained a whole lot undertaking and is a dominant optical approach for trace assessment of excellent sized biological samples due to

the fact of its immoderate sensitivity, simple operation, being reproducible and speedy implementation [28].

In this work, we searched for the different natural resources of CDs and identify their synthesis methods, properties and some of their applications where the applications of CDs had been based on the principle that the interactions between analytes and CDs either limit the fluorescence by quenching. The focus in this review was on the sensing of iron ions Fe^{3+} that come from natural resources. we have identified limit of detection (LOD) for Iron and their sensing range, the applications of CDs had been based on the principle that the interactions between analytes and CDs either limit the fluorescence by quenching mechanism or increase fluorescence by suppressing the quenching effect.

2. Sensing

Two distinct techniques were produced through CD-based nanosensors: the nanosensors were essentially pure" CDs, as-synthesized or passivated by unique target groups; and (ii) the practical sensing medium was a nano-composite that was fabricated by pairing CDs with other nano- or micro-materials. Differently prepared CDs can be used, also in a biological setting, to detect different metal cations such as Hg^{2+} , Cu^{2+} , Fe^{3+} or small molecules or macromolecules in solution [27]. Usually, as CDs are immersed in a solution with a particular analyte, their luminescence is quenched (decreases), allowing high sensitivity and also high selectivity analytes to be detected, as obtained by correctly designing the functional groups of the surface, reacting only with a certain form of metal cation. The sensitivity to detection can be very high and the minimum detectable ion concentration can be as low as femtomolar [34]. The fundamental causes of quenching, however are widely studied and can include multiple fundamental interactions, such as a shift of charge from the dot to the ions, which hinders radiative recombination [35], or an energy transfer from the dots to the acceptor, quenching the CD emission and increasing the latter's emission [36][37].

Table 1 summarizes some of the natural resources of CDs with their synthesis method and application in bioimaging or sensing.

Table 56. Natural Resources of CDs and their applications.

Source	Method	Size (nm)	QY (%)	Application	Ref.
Winter melon	Hydrothermal	4.5-5.2	7.51	Bio-imaging	[38]
Onion	Hydrothermal	9	28	Bio-imaging	[39]
Coriander	Hydrothermal	1.5-2.98	6.48	Detecting	[40]
Pomegranate	Hydrothermal	2.5-6.5	7.6	Bio-imaging	[41]
Bagasse	Hydrothermal	2.94	4.7	Bio-imaging	[42]
Grass	Hydrothermal	3-5	2.5-6.2	Solar cells	[43]
Flaxseed	Hydrothermal	4-8	14.2	Biosensors	[44]
Fish scale	Hydrothermal	4-9	9	Detecting	[45]
Peach gum	Hydrothermal	2-5	28.46	Detecting	[46]
Black tea	Hydrothermal	4.6		Detecting	[47]
Prawn shell	Hydrothermal	6	54	Drug	[48]
Oatmeal	Hydrothermal	20-40	37.4	Bio-imaging	[49]
Shiitake	Hydrothermal	2-6	5.5	Bio-imaging	[50]
Lentil	Hydrothermal	5	10	Detecting	[51]
Egg white	Hydrothermal	2.1	64	Detecting	[16]
Sweet pepper	Hydrothermal	2-7	19.3	Detecting	[52]
Yam	Hydrothermal	2.7	9.3	Detecting	[53]
Carrot	Hydrothermal	3~8	5.16	Bio-imaging	[54]
Cucumber	Hydrothermal	>10	3.25	Detecting	[55]
Durian	Hydrothermal	2-6	79	Bio-imaging	[56]
Prunus avium	Hydrothermal	7	13	Bio-imaging	[57]
Citrus pectin	Hydrothermal	2.7	1.1	Bio-imaging	[58]
Dragon fruit	Hydrothermal	2.5		Bio-imaging	[59]
Gelatin	Hydrothermal	1.7	31.6	Bio-imaging	[60]
Bamboo	Hydrothermal	2-6	7.1	Detecting	[61]
Waste paper	Hydrothermal	3-7	10.8		[62]
Aloe	Hydrothermal	5	10.37	Detecting	[63]
Kitchen waste	Hydrothermal	50		Drug	[64]
Lignin	Hydrothermal	2-6	21	Bio-imaging	[65]
Starch	Hydrothermal	2.25-3.5	21.7	Bio-imaging	[66]
Corn flour	Hydrothermal	2-6	7.7	Bio-imaging	[67]
coriander	Hydrothermal	2.98	6.48	Detection	[68]
Humic acid	Hydrothermal	4	5.2	Bio-imaging	[37]
Beef meat	Hydrothermal		40	Cell	[69]
Red pepper	Hydrothermal		19.3		[69]
Rose heart	Hydrothermal		13.6	Fe ³⁺	[69]
Paper ash	Hydrothermal		6	Bio-imaging	1. [69]
Coffee bean	Hydrothermal	1-5		Bio imaging	[69]
Wheat straw	Hydrothermal		9.2	Labeling	[69]
Pumpkin	Hydrothermal		9.42	PH sensing	[69]
Chitosan	Hydrothermal		13	Nitro	[70]
Egg shell	Microwave	5	14	Biosensor	[7]
Goose feathers	Microwave	21.5	17.1	Detecting	[71]
Coconut water	Microwave	1-6	54	Detecting	[72]
Lotus	Microwave	18.7	19	Hg ²⁺	[73]

Rose	Microwave	4-6	13.45	Detecting	[74]
Silk worm	Microwave	19	46	Bio-imaging	[70]
Serum	Microwave	2.4-5	14	Detecting	[75]
Eutrophic	Microwave	8	13	Bio-imaging	[76]
Natural	Microwave	2-3		Bio-imaging	[77]
Peach gum	Microwave		28.46	Au ³⁺ , Fe ³⁺	[78]
Starch	Microwave			Imagined	[69]
Fingernail	Microwave		42.8	Sunset	[69]
Flour	Microwave	1-4	5.4	Sensing	[79]
Crab shell	Microwave				[69]
Silkworm	Microwave	13-26	46	Cell	[69]
Feathers	Microwave		17.1		[69]
Lotus root	Microwave		23	Hg ²⁺	[70]
Activated	Chemical	4.5	12.6	Bio-imaging	[80]
Candle soot	Chemical	1			[81]
Rice husk	Pyrolysis	3-6	15	Bio-imaging	[82]
Lychee seed	Pyrolysis	1.12	10.6	Bio-imaging	[37]
Lychee	Pyrolysis	1.4		Bio-imaging	[83]
Eggs	Pyrolysis	2.15	5.96	Bio-imaging	[9]
Urine	Pyrolysis	20.6	14	Detecting	[84]
Peanut shell	Pyrolysis	0.4-2.4	9.91	Bio-imaging	[85]
Peanut skin	Pyrolysis	10-40	7		[86]
Olive solid	Pyrolysis	10	10	Detecting	[87]
Wool	Pyrolysis	10	22.5	Cell	[88]

We note in this table, the different natural sources of carbon nano-dots with different synthesis methods, where the hydrothermal method is the most common method, and the parameters of each differ, such as size and QY%, and their applications such as bio-imaging and detecting Fe³⁺.

3. Iron Sensing

Over latest decade, there has been widespread pastime for the detection of heavy and transition metallic ions using fluorescent chemo sensors due to the fact fluorometric technique is tremendously sensitive. So an Important application of surface functionalization is oriented towards sensing, detection of heavy metals or transition ions [89], amongst these metal ions is the iron, Ferric ion (Fe³⁺) is one amongst the most essential metallic ion which is abundantly reachable in the environment. These ions play significant role in retaining many biological processes. Where Total physique iron in adult males and ladies is usually about 50 and 34–42 mg/kg of physique weight, respectively [90]. Any change in the optimum stage of ferric ions may also disturb cell method stability and can lead to numerous ailments like Alzheimer's disease, Parkinson's disease, heart failure, inflammation, hemochromatosis etc. [91][92]. Hence sensitive and selective detection of Fe³⁺ ions are of extensive value in biological purposes [93]. And it also has great importance for monitoring its levels in water basins according to the guideline limit of Fe³⁺ concentration (5.36 μM) proposed by World Health Organization (WHO), iron in water can be determined by atomic absorption spectrometry (detection limit 1 μg/L) or by colorimetric methods (detection limit 5 μg/L), these methods are costly and time consuming, therefore, developing a sensor based on CDs which are extracted from natural resources is a necessity and a point of researchers interest [90].

Several researches have reported the use of CDs as sensing probes of Fe³⁺. almost of the researchers followed the quenching phenomena of carbon dots upon addition of ferric ions as shown in **Figure 1**, and the quenching percentage could be corresponded to the concentrations of added ions. The most important of sensor performance parameters are the sensing range which defines the range in which a signal allows quantification of the concentration of an analyte (linear range), sensitivity which is the slope of linear range, and limit of detection (LOD) is defined the minimal detectable value, where low LOD usually implies a rather high sensitivity.

Table 2 summarizes some of the natural resources of CDs Detecting Fe³⁺ Sensing with their synthesis method and applications.

Table 2. Natural Resources of CDs Detecting Fe³⁺ Sensing.

Source	Method	Size	QY	Application	Sensing	L	R
Corian	Hydrother	1.5-	6.48	Detecting Fe ³⁺	0-6	0.	[
Garlic	Hydrother	10.7	17.5	Bio-imaging			[
Papaya	Hydrother	3.4	18.9	Detecting Fe ³⁺	1-8 and	0.	[
Potato	Hydrother	0.2-2.2	6.14	Detecting Fe ³⁺			[
Black	Hydrother	4.6		Detecting Fe ³⁺	0.25-60		[
Egg	Hydrother	2.1	64	Detecting Fe ³⁺	50-250	-	[
Sweet	Hydrother	2.5-5.5	8.64	Bio-imaging	1-100	0.	[
Papaya	Hydrother	3	7	Bio-imaging			[
Prunus	Hydrother	7	13	Bio-imaging	0-100	0.	[
Cocoo	Hydrother	70	38	Bio imaging	0-6	0.	[
Honey	Hydrother	2	19.8	Detecting Fe ³⁺			[
Onion	Hydrother	9	28	Detecting Fe ³⁺	0-20	0.	[
coriand	Hydrother	2.98	6.48	Detection Fe ³⁺	0-6	0.	[
Jinhua	Hydrother		50.7	Detection			[
Rose	Hydrother		13.6	Fe ³⁺ Detection			[
Entero	Hydrother	2.75 _	8	Detecting Fe ³⁺	1-370	0.	[
Goose	Microwav	21.5	17.1	Detecting Fe ³⁺	2-7	19	[
Peach	Microwav		28.4	Au ³⁺ , Fe ³⁺			[
Chicke	Microwav	2.2-3.4	6-8	Detecting Fe ³⁺			[
Konjac	Pyrolysis	3.37	22	Detecting Fe ³⁺	0-5	-	[
Olive	Pyrolysis	10	10	Detecting Fe ³⁺	0-50	1	[

We note in this table, different natural sources of CDs detecting Fe³⁺ Sensing with their synthesis method, applications and parameters such as size, QY%, sensing range and LOD.

4. Conclusion

The main reason of these that these tiny CDs have attracted considerable attention at the moment is that they have a strong fluorescence renamed fluorescent carbon. Due the superior properties of CDs, they are used in many applications and field such as biomedical, optronics, Photocatalysis and sensing. Great improvement has been achieved in the synthesis, properties and applications of CDs. Several methods for preparing CDs have been published, which can be generally classified into "Top-down" and "Bottom-up" methods that can be adjusted during preparation or post-treatment. Most recent researches on CDs have focused on their fluorescence characteristics and photocatalytic properties. This review summarizes the primary synthesis method, physical and chemical properties, application and Fe³⁺ sensing. In this review we came up with 69 natural resources to extract CDs for different applications. 21 resources of them are for Fe³⁺ detection. As a result, the most commonly used in the synthesis of CDs is hydrothermal methods. The applications of CDs had been based on the principle that the interactions between analytes and CDs either limit the fluorescence by quenching, or increase fluorescence by suppressing the quenching effect

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