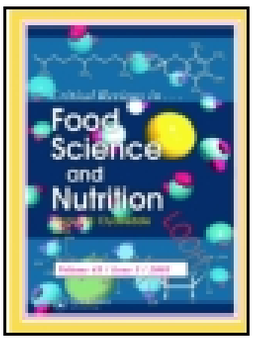

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Carbon Dots: Principles and Their Applications in Food Quality and Safety Detection

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Abstract

In the past ten years, as a novel and prospective nanomaterials, carbon dots have acquired tremendous attention for their unique optical and physicochemical properties, high compatibility and low cost, as well as great potential in sensing area. This review aims to present the current detecting principles based on carbon dots and other nano biological technologies, involving fluorescence quenching and recovery mechanisms. The synthetic and modificatory approaches in making carbon dots including top-down and bottom-up methods, as well as surface passivation and heteroatom doping ways are introduced. Their applications in food area, concerning detection of nutrients, restricted or banned substances as well as foodborne pathogenic bacteria and the toxins secreted are discussed. Finally, the difficulties to be overcome or problems to be solved are presented, and other novel techniques to combine with carbon dots to obtain more stable and specific nanosensors in various fields are proposed. Although carbon dots based sensors have shown the potential in sensing aspect of food area, as food samples are complex in compositions that may cause interferences, more novel techniques are needed to combine with carbon dots to develop sensitive and specific sensing probes.

Keywords: Carbon quantum dot; fluorescence quenching; fluorescence recovery, sensing; food safety

1. Introduction

Fluorescent carbon dots (CDs) were first isolated and characterized by Xu *et al.* (2004) as a new class of carbon nanomaterials with sizes below 10 nm. Since then, dramatic developments have taken place mainly in the improvement of their synthetic methods and optical properties as well as the extension of their application aspects. Carbon dots are composed of quasi-spherical carbon quantum dots (CQDs), complanate graphene quantum dots (GQDs) and polymer dots (Miao *et al.*, 2015; Zhu *et al.*, 2015a),

with CQDs being the most extensively studied one in food quality and safety detection. Therefore, in this review, only CQDs are discussed, and CDs are referred to quasi-spherical carbon quantum dots.

The term of quantum dot originates from the quantum confinement effect as the physical dimensions of nanodots are smaller than the exciton Bohr radius, manifesting the fluorescent optical properties (Zuo *et al.*, 2016). In addition, surface defects seem to be another reason in the photoluminescence (PL) mechanism (Du & Guo, 2016; Zhu *et al.*, 2015a). Up to now, the actual mechanism of CDs photoluminescence is still controversial due to various structures and synthetic principles. In order to make CDs sufficiently fluorescent for further use, their sizes and surface chemical groups should be carefully controlled and modulated during the synthetic process. CDs can be synthesized by two routes: top-down nano-cutting and bottom-up organic routes. Top-down nano-cutting refer to the cutting of carbon materials into carbon nanoparticles. On the other hand, bottom-up organic route contains dehydration and carbonaceous aggregation from small molecules known as precursors. The reacting conditions and purifying operations significantly influence the size distribution of the prepared CDs. Besides, surface functionalization and doping are commonly used to tune and enhance the PL properties as well as to modify the physicochemical properties for specific applications (Georgakilas *et al.*, 2015).

As a new family of versatile nanomaterials, CDs have been in the spotlight of tremendous potential applications, including optronics, biology, medicine, and sensing issues. Sensing is an arresting application in the field of chemical and microbial detections. Compared with conventional semiconductor QDs comprised of heavy metals such as cadmium or lead, CDs are more environmental friendly in synthesis and therefore can be favourably consumed as alternatives in further development of biological, environmental and food areas attributing to their low-toxicity compared with semiconductor quantum dots, water solubility and high photostability (Lim *et al.*, 2015). In particular, CDs have gained initial achievements in the field of food quality and safety detection with high

sensitivity and selectivity, involving the measurement of nutrients (Liu *et al.*, 2015), pesticides (Li *et al.*, 2016), pathogenic microorganisms (Wang *et al.*, 2015a), mycotoxins (Wang *et al.*, 2016a), banned additives (Xu *et al.*, 2015) and so on. Recently, CDs have been created by natural carbonaceous materials using green process. Therefore, many researches have been conducted to study the feasibility of CDs in food quality and safety evaluations, involving nutrients, pesticide residues, illegally used additives and pathogenic microbial organism or chemicals. Facts proved that CDs are potential for on-line and specific detections based on different principles to constitute multifarious sensors. As many review papers have comprehensively summarized the properties and applications of CDs (Du & Guo, 2016; Li *et al.*, 2013; Zheng *et al.*, 2015), the current review will focus on CDs in the food field, including the testing object, detecting environment and the mechanisms employed in the sensors. It is hoped that the current review will encourage more studies of the technology in quality and safety evaluation for the food industry.

2. Synthetic Strategies and Functionalization

2.1 Top-down synthetic route

As one of the two synthetic strategies for CDs, the top-down method indicates cutting down larger carbon materials like graphite powder, graphite oxide, carbon nanotubes, carbon rods and carbon soot into nano fragments, aiming to produce defect-mediated fragments by adding oxygen- or nitrogen-containing functional groups like hydroxyl, carboxyl and amino groups to create defects on CDs. For CDs synthesis, Xu *et al.* (2004) provided a new technique of CDs serendipitously when they used top-down route called arc-discharge to purify single-walled carbon nanotubes (SWCNTs) by gel electrophoresis from arc-discharged soot. Subsequently it was found that laser ablation was feasible for CDs synthesis using a carbon target in the presence of water vapour with argon as carrier gas (Sun

et al., 2006). Moreover, the photoluminescence of the obtained CDs was obviously improved after passivation by acid oxidation. Besides, electrochemical methods were also used to generate CDs (Zhou *et al.*, 2007) with low costs and improved production efficiency (Ming *et al.*, 2012). However, the top-down method is not commonly used to produce CDs.

2.2 Bottom-up synthetic route

Bottom-up approaches are used to make quasi-spherical carbon quantum dots (CQDs). These approaches create CDs from molecular precursors with thermal or pyrolytic treatments, which have the advantages of precise manipulation of the size distribution of CDs by selecting the appropriate organic precursors and controlling the reaction conditions. Bottom-up strategy has been more extensively used for CDs synthesis using hydrothermal or solvothermal treatment with a high temperature span from inexpensive and biocompatible starting materials, such as citrate (Yang *et al.*, 2014), saccharides (Wang *et al.*, 2011a), chitosan (Yang *et al.*, 2012), ascorbic acid (Wu *et al.*, 2012), gelatine (Liang *et al.*, 2013) and even some green materials including proteins (Wang *et al.*, 2012), fruit juice (Huang *et al.*, 2013) and peels (Lu *et al.*, 2012). Song *et al.* (2015) mixed citric acid and ethanediamine in hydrothermal treatment. Fig. 1 shows different products and their relationships during this synthetic process (Song *et al.*, 2015). In addition, with the aid of microwave, the synthetic process has become more rapid and cost effective (Zhai *et al.*, 2012).

2.3 Surface passivation and heteroatom doping approaches

In order to enhance the quantum yields (QYs) and improve the PL property of carbon dots so that they can be better adapted to different applications, passivation can be performed on the surface of carbon dots via connection with other chemical groups. Various chemical groups such as diamine, alkylamine and hydrazide can be attached to the surface of CDs during or after synthesis. Polymer modification is another way to introduce functional groups using polyethylene glycol (PEG), polyethylenimine or other polymers. Besides, by controlling the oxygenated degree with strong acids, carbon dots with diverse oxygen-containing groups such as carboxyl, hydroxyl and epoxy can be prepared with chemically reactive sites, endowing them with excellent water dispersion ability and improved PL performance. On the other hand, heteroatom doping, including N, S and P, has been commonly used to tune the PL properties, which can be realized by adding nitrogenous precursors during synthesis involving ethanediamine, ammonium hydroxide, trimethylamine, etc. Both the above approaches can synthesize CDs with good optical and biocompatible properties (Zhu *et al.*, 2015a; Zuo *et al.*, 2016).

3. Optical and Physicochemical Properties

In spite of various structures, the optical properties of CDs are similar in terms of absorption, photoluminescence (PL) and up-conversion luminescence, which have been frequently studied and improved for further applications. CDs are typical in possessing strong but narrow optical absorption in the UV region around 260-320 nm (Zhang & Chen, 2014), with a tail extending to the visible range. After surface passivation, the wavelength range may be broadened. As one of the most appealing features of CDs, PL emission with a spectral range from UV to visible light and even near infrared region can be tuned by various synthetic approaches, in order to make the CDs suitable for different applications (Wang *et al.*, 2016b). QY is the number of emitted photons referring to the number of absorbed photons and it is used to evaluate the fluorescent efficiency of the prepared CDs. The PL photoluminescence quantum yields of bare CDs without passivation and element doping is too low (typically < 10%) to apply to real applications, owing to the emissive traps on the surface. Thus, effective surface passivation is indispensable to improve the PL properties. Moreover, the emission peak position of CDs is always associated with the excitation wavelength and this phenomenon is known as excitation-dependent behaviour, which can result in the production of multi-colour CDs for more flexible applications by modulating size distribution and surface functionalization. Apart from down-conversion fluorescence emissions, certain CDs have been proved to exhibit up-conversion fluorescence, in which the fluorescence emission wavelength is shorter than the excitation wavelength. Up-conversion fluorescence CDs are particularly appealing in the applications of vivo bioimaging due to the improved photon tissue penetration and reduced background interference at longer wavelengths especially in the NIR region. Several researchers (Huang *et al.*, 2013; Jia *et al.*, 2012; Ming *et al.*, 2012) have successfully obtained such CDs by adjusting the reagents and synthesis conditions. Cao *et*

al. (2007) happened to acquire up-conversion fluorescence CDs when they observed visible light from the CDs excited by an 800 nm femtosecond pulsed laser.

As mentioned above, CDs can be synthesized by various routes using different chemicals. As a result, the physical and chemical structures of CDs are diverse. However, in terms of the physicochemical properties, CDs are similar in many aspects. First, CDs possess a high photostability with stable fluorescence emission intensity during a long-time continuous excitation, which explains the application of CDs in bioimaging. The fluorescence intensity is highly sensitive to pH, due to that the local surface environment can be affected by the ionization of carboxylic-acid groups. Hence, the PL can be switched off and on by varying the pH values from neutral to strong acid or alkaline (Li *et al.*, 2011). Jia *et al.* (2012) synthesized a kind of CDs by heating ascorbic acid solution directly, which represented a linear relationship between the fluorescence intensity and the pH values (4.0 - 8.0) of the obtained solution. This also shows the potential for CDs to be used for pH measurement.

Foe applications in food or biological systems, low cytotoxicity of CDs is a prerequisite. Therefore, the cytotoxicity of luminescent CDs *in vitro* and *in vivo* is an attractive issue and has been studied by many researchers (Gao *et al.*, 2014; Havrdova *et al.*, 2016). Wang *et al.* (2011b) conducted an evaluation of the biocompatibility and cytotoxicity *in vitro* and *in vivo* on fluorescent CDs prepared by various carbon precursors and passivation molecules. Their results showed that the CDs were not intrinsically cytotoxic in terms of their nanoscale structures and the carbon elements. Other researchers (Baker & Baker, 2010; Tao *et al.*, 2012) have achieved similar conclusions that CDs possess excellent biocompatibility and low cytotoxicity, except the cytotoxicity concerns relating to surface passivation molecules. Therefore, the consideration should be given in selection of surface passivation agents for use with live cells in imaging studies.

4. Principles in Using Various CDs-Based Sensors

CDs can be applied to many areas of detection as a main element to constitute various biosensors, as CDs possess a good ability to serve as either electron donors or acceptors. In other words, the established biosensors can be used in detections according to the fluorescence quenching or recovery properties of CDs as shown in Table 1, and by combining with different bio-macromolecules, their specificity and sensitivity can be further enhanced.

4.1 Fluorescence quenching (on-off)

CDs have been widely used for detection of different ions involving metal ions (Cu^{2+}, Hg^{2+}, Ag^{2+}, Fe^{3+}, Pb^{2}, etc) (Cayuela *et al.*, 2016; Lai *et al.*, 2013; Liu *et al.*, 2014; Wang *et al.*, 2014; Wee *et al.*, 2013; Zhang & Chen, 2014) and other ions (Cl^{-}, H^{+}, etc) (Ding *et al.*, 2016; Dong *et al.*, 2012; Kong *et al.*, 2012) based on a similar mechanism. The functional groups on the surface of CDs exhibit distinct affinities to diverse target ions, which results in the specific interaction between CDs and the ions based on the high selectivity to ions, as well as the quench of PL intensity via an energy or electron transfer process. Zhang and Chen (2014) used N-CDs-based sensor to determine the concentration of Hg^{2+} in tap water and real lake water with a limit of detection (LOD) of 0.23 μM. The sensor was based on the fluorescence quenching mechanism, induced by the surface states in the CDs or the Hg^{2+}-induced conversion of the special functional group (-CONH-) from spirolactam structure to an opened-ring amide. Besides, silver ions (Ag^{+}) were first detected using luminescent passivated carbon quantum dot (p-CQDs) and hydrogels, with different molecular groups on the surface. Results showed that the sensing system with more carboxylic groups was more specific, mainly due to the strong Ag-O interaction, which can quench the fluorescence via charge transfer. Moreover, the application of hydrogels in p-CQDs turned to be noticeable for the selective detection of Ag^{+} ions (Cayuela *et al.*,

2016). Additionally, Qian *et al.* (2014) developed N-doped CQDs sensing platform for multifunctional detection of Fe^{3+}, Ag^{+}, pH values and H_2O_2, based on their distinctive fluorescence influence on the system. Other chemical compounds can be also determined by CDs nanosensor based on their direct quenching properties on CDs, including a large number of chemicals such as ascorbic acid, nucleic acid, hydroxyl radicals, blood sugar, tartrazine, iodide, quercetin, and other substances (Ren *et al.*, 2014; Shen & Xia, 2014; Xu *et al.*, 2015a; Zhang *et al.*, 2013; Zhu *et al.*, 2015b; Zou *et al.*, 2015). Zhu *et al.* (2015b) prepared nitrogen-doped carbon dots (NCDs) for sensitive and selective detection of ascorbic acid (AA). The NCDs could effectively decrease the fluorescence intensity due to the spatial effect and hydrogen bond between AA and the groups on the surface, which were resulted from the inner filter effect (IFE) and the static quenching effect (SQE). The linear detecting range was found to be from 0.2 μM to 150 μM, with a LOD of 50 nM (Zhu *et al.*, 2015b). Moreover, as the fluorescence of CDs can be quenched by nanoparticles, some researchers as below attempted to use CDs biosensor for quantifying nanoparticles directly. Based on IFE, Cayuela *et al.* (2015) employed amine-modified CDs to detect citrate-silver nanoparticles (cit-AgNPs), which act as the energy donor and receptor, owing to the aggregation of cit-AgNPs caused by the free amine groups at the surface. More interestingly, the CDs sensor could be reused with excellent performances by simply removing the agglomerated cit-AgNPs via filtration (Cayuela *et al.*, 2015).

Apart from direct quantification of the objects that can quench the CDs fluorescence, CDs sensor has also been used for detecting other substances based on indirect quenching mechanism via chemical

reaction. Wang *et al.* (2013) developed a fluorescent quenching assay for measuring acetylcholine (ACh) using reduced graphene oxide decorated with carbon dots (C-dots@RGO) and the principle is illustrated in Fig. 2. The fluorescent intensity of C-dots@RGO can be quenched linearly by reactive oxygen species (ROS) generated from betaine and H_2O_2, which are the oxidative products of the reaction by adding acetylcholinesterase (AChE) and choline oxidase (ChOx) into the ACh solution. Thereby, the concentration of ACh can be detected indirectly.

In order to improve the selectivity for specific determination of objects, some novel techniques have been developed in recent years, including the combination with antibodies, aptamer (Ma *et al.*, 2015) and molecular imprinting (Mao *et al.*, 2012). Ma *et al.* (2015) grafted antibodies and aptamer onto the surface of CDs via surface amino groups. The addition of biomarker mucin 1 (MUCI) induced the aggregation of CDs, leading to a fluorescence decrease. Based on this principle, the contents of MUCI can be successfully detected with a LOD of 2 nM in real serum samples, regardless of the interference of other proteins in the samples. Mao *et al.* (2012) constructed a type of CDs anchored with molecularly imprinted polymer (MIP) matrix (CDs@MIP) on the surface. The resulting composite exhibited excellent fluorescent properties and can be used to sense the concentration of dopamine, which can decrease the fluorescent intensity after removal of the original templates of MIP with a detecting range of 25-500 nM and a LOD of 1.7 nM. Furthermore, the developed method was also successfully used in biological fluids such as human urine samples without other interference (Mao *et al.*, 2012).

4.2 Fluorescence recovery (on-off-on)

As discussed above, the fluorescence of CDs can be quenched via chemical reactions or energy transfer based on nanoparticles, and the quenching mechanism can be used to build a fluorescence recovery system by breaking the state of fluorescence quenching. Up to now, many researches (Gao *et al.*, 2015; Gong *et al.*, 2015; Hou *et al.*, 2015; Lan *et al.*, 2015; Qian *et al.*, 2015; Qu *et al.*, 2013; Xu *et al.*, 2015b; Zheng *et al.*, 2013; Zhou *et al.*, 2012) have been performed to study the feasibility of CDs sensing platform based on the fluorescence recovery, and their testing substances cover a wide range including phytic acid, ascorbic acid, methyl parathion, hydrogen peroxide, alkaline phosphatase activity, dopamine, glutathione, biothiols, etc. Specifically, Gong *et al.* (2015) utilized the fluorescent quenching and recovery mechanism of CDs to quantify AA. The fluorescence was quenched by Fe^{3+} and then recovered with the addition of AA due to its reduction of Fe^{3+} to Fe^{2+} that has no quenching impact on CDs. Similarly, AA was detected using the complex of CDs and chromium (VI) (Cr^{6+}) as Cr^{6+} can quench the fluorescence based on IFE and Cr^{6+} can be reduced to Cr^{3+} by AA (Zheng *et al.*, 2013). As shown in Fig. 3, the fluorescence intensity of the CD-Cr^{6+} mixture can respond to AA at different concentrations following a linear equation with R equal to 0.993. It could be inferred that it may be possible to detect strong reducible agents using the CDs-Cr^{6+} mixture. Hou *et al.* (2015) prepared L-tyrosine methyl ester functionalized carbon dots and the fluorescence was quenched by tyrosinase by catalysing the oxidation of tyrosine methyl ester on the surface of carbon dots. The sensing platform established was then successfully used to detect methyl parathion (a type of organophosphorus pesticide) as it could suppress the enzyme activity, thereby causing a fluorescence recovery.

Moreover, nanoparticles are frequently used in the fluorescent recovery systems including the introduction of MnO_2 (Cai *et al.*, 2015; He *et al.*, 2015; Wang *et al.*, 2015b; Yang *et al.*, 2015), AuNPs (Deng *et al.*, 2015; Mandani *et al.*, 2015; Wang *et al.*, 2011c) and others in the CDs solutions. In these studies, the researchers constructed CDs-MnO_2 nano complex by adding MnO_2 nanosheets into the

CDs colloid solution for trace detection of glutathione (GSH). As the fluorescence of CDs could be efficiently quenched by MnO_2 via fluorescence resonance energy transfer (FRET) and then restored when GSH was introduced, resulting from the reduction of MnO_2 to Mn^{2+} by GSH, the fabricated nanocomposite exhibited excellent performance with the best LOD of as low as 10 nM. More importantly, the MnO_2-CQD mixture has the potential in biological applications such as tracing GSH variations in living cells due to its good membrane permeability and excellent biocompatibility. In addition, AuNPs were introduced to combine CDs for measuring sulfhydryl compounds due to that the thiol moiety could effectively ligand the surface of AuNPs with the formation of Au-S bond (Deng *et al.*, 2015; Mandani *et al.*, 2015). First, a fluorescent quenching system was built in the construction of stabilized Au NPs with a thin continuous CDs layer around the nanoparticle surface, and then the biothiol rescued AuNPs from the coverage of CDs and released the fluorescent intensity of CDs. Based on this principle, biothiols could finally be detected according to the fluorescent recovery rate. Moreover, the above studies demonstrated the static and dynamic nature of quenching, which were caused not only by the ligand displacement but also by the changes in FRET (Deng *et al.*, 2015; Mandani *et al.*, 2015).

In complicated testing environments, aptamer can be employed to specifically target the objects. Wang *et al.* (2016a) modified sulfhydryl group on specific aptamer for aflatoxin B_1 (AFB_1), which can connect AuNPs via Au-S bond. The prepared CDs were then assembled on aptamer/AuNPs by electrostatic interactions. Thus AFB_1 could be quantified based on the above system, which provided a feasible way of detecting most compounds, including non-sulfhydryl compounds, as shown in Fig. 4.

Furthermore, some researchers (Liu *et al.*, 2014; Shi *et al.*, 2014; Wang *et al.*, 2016b) have developed ratiometric fluorescent nanosensor based on the CDs with dual-emission properties for the detection of Cu^{2+}, GSH and optical thermometry. These new types of sensors not only provide more sensitive measurements but also exhibit more specific responses towards the target objects in complex environments.

5. Applications of CDs as Biosensors in Food Quality and Safety Detection

There are many applications for the sensing systems based on CDs in the food area as listed in Table 2, including detecting nutrient substances, pesticides, banned or limited additives, pathogenic microorganisms and microbial toxins, except from detecting metal ions in water solutions as discussed previously.

Liu *et al.* (2015) fabricated a CQD-MnO_2 sensing platform based on fluorescent recovery for measuring AA, which were then successfully used in the detection of some fresh fruits, vegetables and commercial fruit juices. Ahmed *et al.* (2015) employed CDs for direct measurements of tannic acid (TA) in red and white wine samples with a LOD of 0.018 mg/L as TA has the ability to quench the fluorescence. As a kind of restrictedly used synthetic food colorant, tartrazine can result in a strong fluorescence quenching of the prepared CDs. Therefore, Xu *et al.* (2015a) was able to use a CDs sensor to detect steamed buns, honey and candy, which were spiked with different amounts of tartrazine ranging from 0.25 to 32.50 μM. Also Dai *et al.* (2014) constructed a FRET system using CDs and AuNPs for the detection of melamine with a LOD of 36 nM, and the proposed method was then successfully used in milk samples with satisfactory results.

Pesticide residual is a serious problem in the agricultural industry. Some studies (Li *et al.*, 2016; Wang *et al.*, 2016c) have investigated the feasibility of CDs in pesticide detection in agricultural and food

samples. Wang *et al.* (2016c) established a sensitive and selective sensor using carbon dot labelled with antibody for specific determination of glyphosate with a LOD of 8 ng/mL and the proposed method was generalized to river water, tea, and soil samples with satisfactory recovery ratio between 87.4% and 103.7%.

More importantly, CDs has found its applications in sensing detrimental microbial counts and toxins, including *Salmonella typhimurium* and aflatoxin B1 (AFB1) (Wang *et al.*, 2016a; Wang *et al.*, 2015a). Wang *et al.* (2015a) built carbon dot–aptamer complexes (CD-apt) for quantitative detection of *Salmonella typhimurium* specifically in eggshell solutions and tap water, free from the interference of *Escherichia coli* O157:H7 and *Staphylococcus aureus*, with a testing range of 10^3 to 10^5 cfu/mL and a LOD of 50 cfu/mL. Many other studies (Mandal & Parvin, 2011; Wang *et al.*, 2016a; Zhao *et al.*, 2008) have confirmed the practicality of microbial detection using CDs, GQDs, carbon nanotubes and semiconductor (QDs) in simple or complicated food environments. On the other hand, CDs can be used to detect toxins in food products. For example, based on the fluorescence on-off-on principle, Wang *et al.* (2016a) assembled CDs and AuNPs with the assistance of aptamer for specific detection of aflatoxin B_1 (AFB_1) and achieved a LOD of 5 pg/mL (16 pM). This proposed strategy has proved to be successful in real samples and has been extended to the detection of other objects by changing the corresponding aptamer. Besides, carbon nanotubes and GQDs have also been used for sensing other toxins including *staphylococcal enterotoxin* B (Tang *et al.*, 2010; Wang *et al.*, 2016d). These studies confirmed that CDs is feasible for target detection of harmful microorganisms and their secreted toxins, and generally, aptamer or antibody should be employed in such a sensing system for enhancing the specificity.

6. Conclusions and Outlook

Carbon dots (CDs) based nanosensors and other related nanomaterial or biological techniques are emerging technology, which has been developed very fast in the past ten years. This review summarizes CDs in the aspects of synthetic approaches and modification as well as optical and physicochemical properties. Moreover, various detecting principles based on different CDs-based sensors are presented, which has been seldom discussed in other review papers. In addition, focuses are given on different applications of CDs based sensing systems due to its low-toxicity, high-biocompatibility, simple preparation procedure and low cost, especially in the area of food quality and safety.

Fluorescence quenching and recovery are the main research directions in constructing sensing platform for accurate quantification. In addition, it is critical to explore proper chemicals or nanomaterials to combine with CDs and constitute a feasible, specific and efficient sensing mechanism, such as redox reagents, AuNPs, MnO_2, aptamer or antibodies, or other potential substances for specificity. CDs are expected to have more applications in detecting different types of foods. However, as food samples have complicated chemical and biological components, it is thus a challenge to detect a specific constituent under the interference of complex environments. Therefore, apart from specific reactions with certain chemicals, the proper use of aptamer or other substances with high affinity and selectivity is vital important. Moreover, it is essential to construct more stable CDs and/or to combine them with other nano techniques in order to establish a reliable fluorescence quenching or recovery system for more accurate quantification.

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Table 1. Principles of CDs probes in various applications

Fluorescent detection principles	Detecting target	Quenching or/and recovery principle	Carbon sources	LOD	Excitation wavelength	Emission wavelengths	References
Fluorescence quenching	Hg^{2+}	Interaction of Hg^{2+} and surface states	Folic acid	0.23 μM	390 nm	470 nm	Zhang & Chen (2014)
Fluorescence quenching	AA	IFE, SQE	Sodium alginate	50 nM	270 nm	440 nm	Zhu *et al.* (2015b)
Fluorescence quenching	Cit-AgNPs	IFE	Tribasic sodium citrate	5.17×10^{6} mol/L	355 nm	440 nm	Cayuela *et al.* (2015)
Fluorescence quenching	Ach	Oxidation-reduction reaction	Catechin	30 pM	365 nm	440 nm	Wang *et al.* (2013)
Fluorescence recovery	AA	DQE (Fe^{3+}), oxidation-reduction reaction	Citric acid	50 nM	330 nm	410 nm	Gong *et al.* (2015)
Fluorescence recovery	GSH	FRET (MnO_2), oxidation-reduction reaction	Citric acid	300 nM	360 nm	435 nm	Cai *et al.* (2015)
Fluorescence recovery	Biothiols	FRET (AuNPs), formation of Au-S bond	B-carotene	50 nM	370 nm	470 nm	Mandani *et al.* (2015)

Note: LOD - limit of detection, IFE - inner filter effect, SQE - static quenching effect, DQE - dynamic quenching effect

Table 2. Applications of CDs-based sensors in food area

Detecting object	Food categories	Carbon sources	Detecting principle	LOD	Excitation wavelength	Emission wavelengths	References
Ascorbic acid	Fresh fruits, vegetables and fruit juices	Sodium citrate	Fluorescence recovery (IFE)	42 nM	360 nm	441 nm	Liu *et al.* (2015)
Tannic acid	Wine	6-bromohexylboronic acid	Fluorescence quenching (electron transfer)	0.018 mg/L	362 nm	440 nm	Ahmed *et al.* (2015)
Tartrazine	Steamed buns, honey, candy	Aloe	Fluorescence quenching	73 nM	441 nm	503 nm	Xu *et al.* (2015a)
Melamine	Milk	Histidine	Fluorescence quenching (FRET)	36 nM	350 nm	438 nm	Dai *et al.* (2014)
Pesticide residues	Apple	N-Methylethanolammonium thioglycolate	Fluorescence quenching (enzyme reaction)	5 ppb	350 nm	520 nm	Li *et al.* (2016)
Salmonella typhimurium	Eggshell, tap water	Citric acid	Fluorescence recovery (aptamer)	50 cfu/ml	365 nm	442 nm	Wang *et al.* (2015a)

Aflatoxin B1	Peanut, corn	Pancreatin	Fluorescence recovery (FRET, aptamer)	5 pg/ml	365 nm	425 nm	Wang *et al.* (2016a)

Note: FRET - fluorescence resonance energy transfer, IFE - inner filter effect, LOD - limit of detection

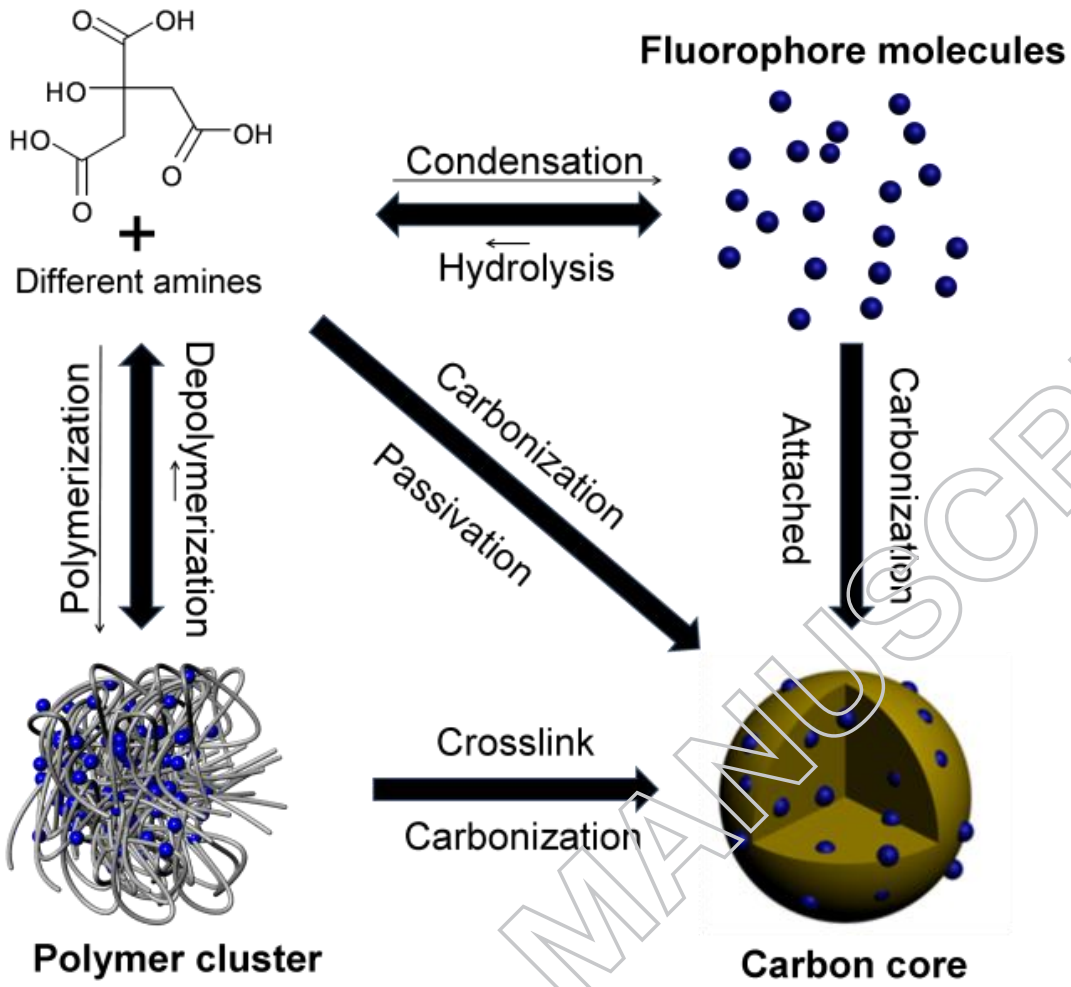

Fig. 1. Different products and their relationships during hydrothermal treatment of citric acid and ethanediamine for development of CDs (Song *et al.*, 2015).

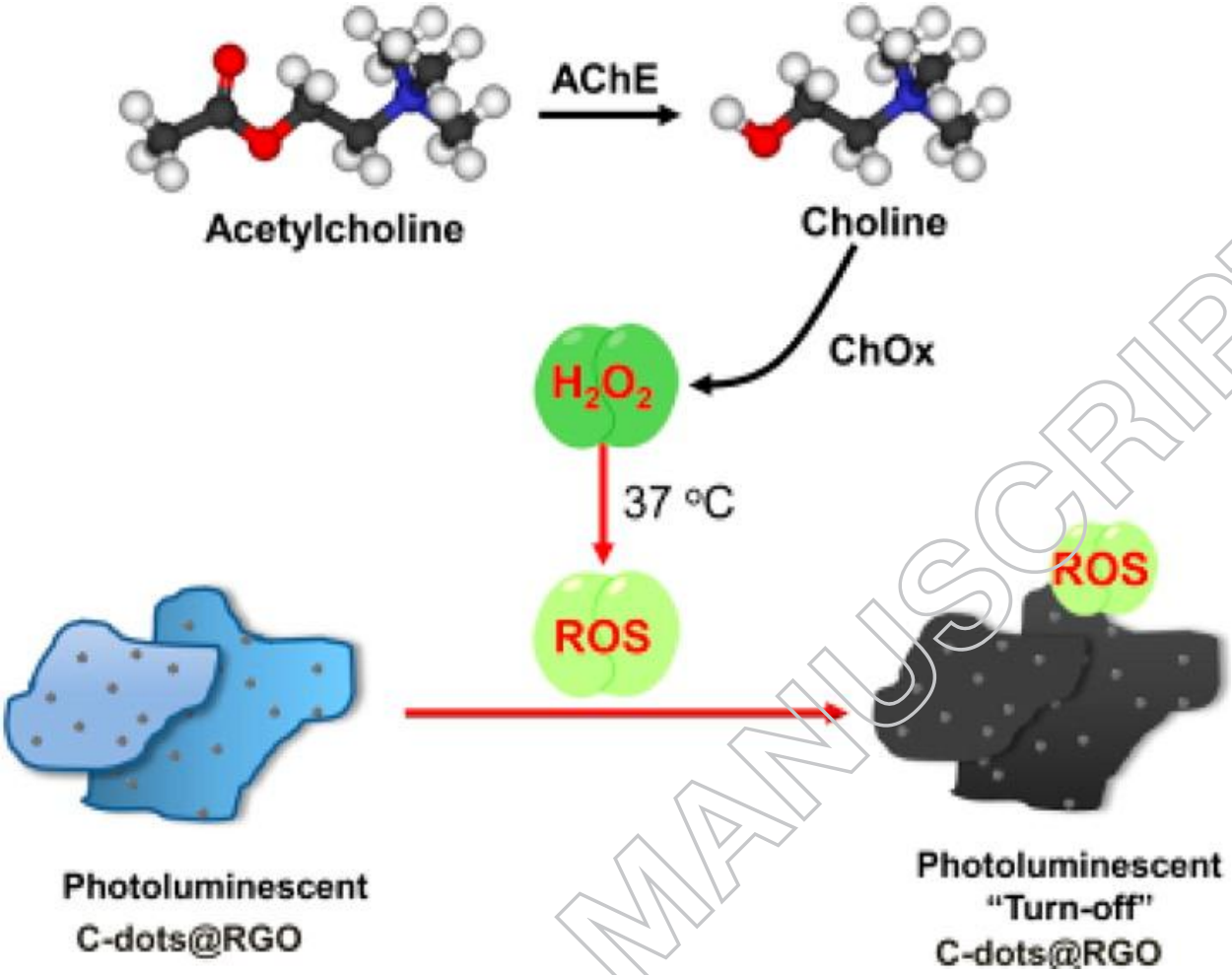

Fig. 2. ACh quantification based on a fluorescent quenching CDs system combined with redox reactions (Wang *et al.*, 2013).

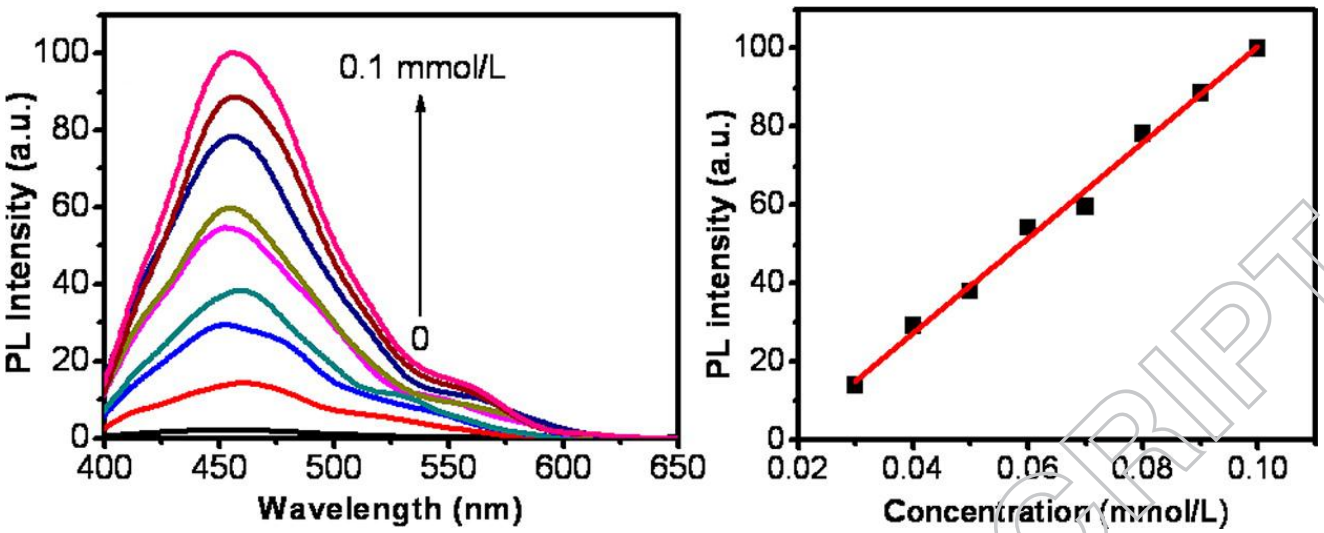

Fig. 3. Fluorescence response of the CD-Cr(VI) mixture to AA at different concentrations as well as the plot of the fluorescence intensity against the AA concentration in the range of 0.03-0.1 mmol/L based on a fluorescence recovery sensing system (Zheng *et al.*, 2013).

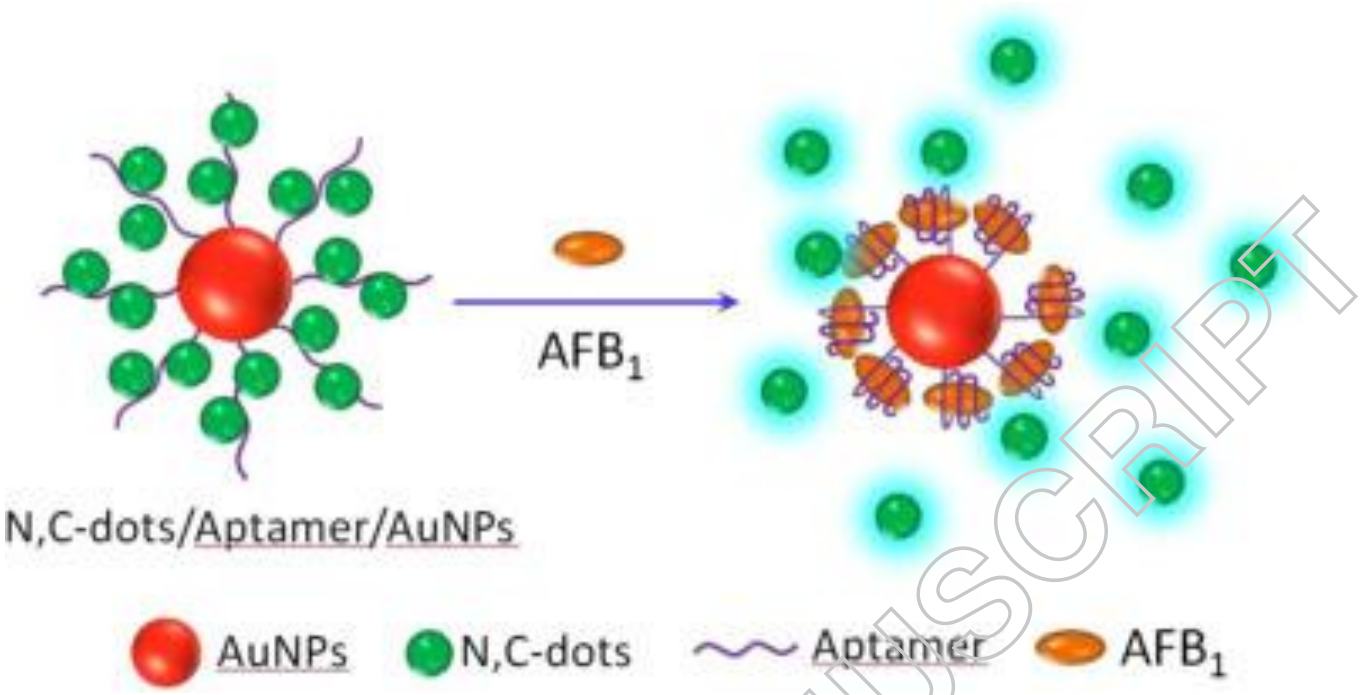

Fig. 4. Detection of AFB_1 based on the fluorescence recovery principle (Wang *et al.*, 2016a).