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Controllable Synthesis of Fluorescent Carbon Dots and Their Detection Application as Nanoprobes

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Abstract: Carbon dots (CDs), as a new member of carbon nanomaterial family, have aroused great interest since their discovery in 2004. Because of their outstanding water solubility, high sensitivity and selectivity to target analytes, low toxicity, favorable biocompatibility, and excellent photostability, researchers from diverse disciplines have come together to further develop the fundamental properties of CDs. Many methods for the production of CDs have been reported, therein, hydrothermal and solvothermal technology needs simple equipments, and microwave synthesis needs less reaction time, hence these methods become current common synthesis methods, in which many precursors have been applied to produce CDs. Due to their excellent fluorescence, CDs have made impressive strides in sensitivity and selectivity to a diverse array of salt ions, organic/biological molecules and target gases. The development of CDs as nanoprobes is still in its infancy, but continued progress may lead to their integration into environmental and biological applications. Hydrothermal, solvothermal, and microwave synthesis of fluorescent carbon dots and their detection applications as nanoprobes in salt ions, organic/biological molecules, and target gases will be reviewed.
Keywords: Carbon dots; Hydrothermal; Solvothermal; Microwave; Nanoprobe

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Introduction

Heavy metals are essential elements in the conventional semiconductor quantum dots (QDs), which are under-utilized concerning about their toxicity, stability and environmental hazard, so their biological application is subject to restriction. Fluorescent carbon dots (CDs) are a fascinating class of carbon family [1,2] recently discovered with a size below 10 nm and have drawn great research interests due to their excellent photostability, favorable biocompatibility, low toxicity, outstanding water solubility, high sensitivity and excellent selectivity to target analytes, tunable fluorescence emission and excitation, high quantum yield (QY) and large Stokes shifts [3,4]. A variety of synthesis methods on CDs have been developed, including laser ablation [5], electrochemical oxidation [6], combustion/thermal microwave heating [7], and supported synthesis [8], but some methods need complex equipments or complex treatment processes. Due to their cheap devices and easy operating steps, hydrothermal, solvothermal, and microwave synthesis methods have been emerging on. Although some reviews have been reported on CDs about their synthesis and applications [4,9-12], there are no reviews about recent systematic advances on controllable synthesis of carbon dots and

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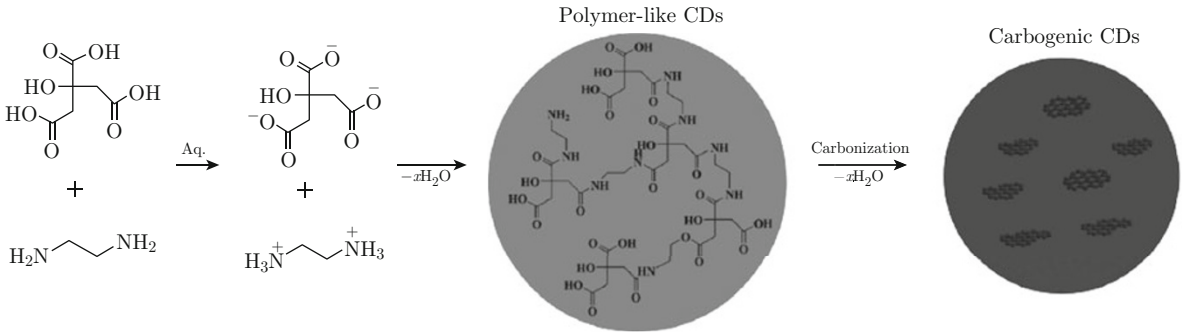


Fig. 1 Reaction mechanism of CDs synthesized by citric acid and ethylenediamine.

Table 1 Hydrothermal and solvothermal synthesis of CDs

T(°C)	Time (h)	FC	Size (nm)	Reactants	Ref.	QY (%)[a]
200	3	Blue	2-4	Pomelo peel	15	6.9
200	10	Blue	2.94	Bagasse's carbonaceous blocks, NaOH	16	4.7
180	3	Blue	13-40	Soy milk	17	2.6
200	3	Blue	1-4	Willow bark	18	6.0
120	2.5	Green	1.5-4.5, 50-60	Orange juice, Ethanol	19	25.6, 19.7
200	3	Blue	35	Giant Knotweed Rhizome	20	11.5
200	3	Blue	1.7	Getatin	21	31.6
180	6	Blue-Green	3-5	Dopamine	22	6.4
180	12	Blue	6.8	BSA, Ethanol	23	7
180	12	Blue	2-6	BSA, TTDDA	24	11
300	2	Blue	1-5	APTMS (AEAPTMS, TEOS, APMDES)	25	42.6
150-300	5	Blue	2-6	Citric acid, Ethylenediamine	13	80
250	2	Blue	2.0	Citric acid, $OHCH_2CH_2OCH_2CH_2NH_2$	26	19.2
180	4	Blue	1.59	Sodium citrate, NH_4HCO_3	27	68.22[b]
180	12	Blue	4-7	Chitosan, Acetic acid	28	43
200	12	Blue (Green)	1.83 (3.83)	Glucose, Monopotassium phosphate (N_2 filled)	14	2.4 (1.1)
140	12	Green	15-70	Glucosamine hydrochloride	29	/
160	4	Blue	70-100	Carbohydrates, Acid (Alkali)	30	0.55-17
160	4	Blue	100, 76	Cellulose (Cyclodextrin), NaOH	31	7.47, 4.49
300	2	Blue	2.6, 3.3, 3.0, 7.9	Glycine (TRIS, EDTA, cadaverine)	32	30.6, 26.0, 26.6, 5.4
220	24	Blue-Green	3	EDTA·2Na	33	15
180	4	Blue	2.0-2.5	L-ascorbic acid, Ethanol	34	6.79
160	7/6	Green	5	L-ascorbic acid, Glycol	35	/
160	24	Blue	< 1	PEG200, NaOH	36	1.95
210	360	Blue	7-12	CTAB, HCl, $Na_2S_2O_8^c$	37	9.8
200	2	Blue	3-5	CCl_4, Quinol, NaOH, Ethanol	38	3.4
150	2	Blue	/	CCl_4, EDA	39	/
200	1,8	Blue, Cyan, Kelly, Yellow	1-2, 2.5-4	CCl_4, $NaNH_2$	40	22
200	15	Blue	/	Soot	41	4.96

a. Quinoline sulfate as a standard.
b. Rhodamine 6G as a standard.
c. Oxidizing by nitric acid, then reacting with PEG600.
T. stands for temperature. FC stands for fluorescence colour. Ref. stands for references. BSA stands for bovine serum albumin. TTDDA stands for 4, 7, 10-trioxa-1, 13-tridecanediamine. APTMS stands for 3-aminopropyltrimethoxysilane. AEAPTMS stands for 3-(2-Aminoethylamino) propyltrimethoxysilane. TEOS stands for tetraethylorthosilicate. APMDES stands for 3-aminopropylmethyldiethoxysilane. TRIS stands for 2-amino-2-hydroxymethyl-propane-1, 3-diol, EDTA stands for ethylene diamine tetraacetic acid. CTAB stands for cetyltrimethylammonium bromide. EDA stands for 1,2-ethylenediamine. "/" stands for no information.

their detection applications. In this review, we focused on recent works about hydrothermal, solvothermal, and microwave synthesis methods of CDs and their application as nanoprobes in salt ion, organic/biological molecule, and target gas detection.

Hydrothermal and solvothermal synthesis of carbon dots

CDs are conjugated systems which have sp^2 and sp^3 hybridized carbons atoms with plenty of oxygen-containing groups. Zhu et al. [13] provided the highest QY (up to 80%) of CDs obtained by the hydrothermal treatment of citric acid and ethylenediamine (as shown in Fig. 1). The highest value is almost equal to that of fluorescent dyes. The reaction contains ionization, condensation, polymerization, and carbonization by bottom-up method. Yang et al. [14] implemented monopotassium phosphate (KH_2PO_4) as a fluorescent color reagent. When high concentration of KH_2PO_4 was used, the CDs showed blue fluorescence. While low concentration of KH_2PO_4 was used, the CDs showed green fluorescence. It is interesting that the use of inorganic salts can adjust fluorescence colour. Biological materials, like pomelo peel [15], bagasse [16], soy milk [17], willow bark [18], and orange juice [19] have been used as carbon sources in smart hydrothermal synthesis for CDs. These synthesized CDs show blue or green fluorescence. What's more, lots of works about using small organic compounds like dopamine [22], citric acid [13,26], carbohydrates [14,28-31], and L-ascorbic acid [34,35] as carbon sources have been done, because they are easy to be carbonized under hydrothermal conditions. It is found that most of these works show blue or green fluorescence. There is no report on long wavelength fluorescence, like yellow or red fluorescence. The actual reasons behind the high photoluminescence (PL) of CDs are still a matter of debate, probably caused by the emissive traps, quantum confinement, zig-zag sites and the defect sites [4,9]. The incorporation of carboxyl and hydroxyl functionality onto the surface of CDs was probably responsible for their high PL property. Meanwhile, radiative recombination of excitons trapped within the defects contributed to the most intense PL bands [4]. Most works on hydrothermal and solvothermal methods synthesizing CDs are summarized in Table 1.

Microwave synthesis of carbon dots

Microwave synthesis methods can be carried out by exploiting a domestic microwave oven with less reaction time needed. Many works have been reported by microwave methods in recent years (as shown in Table 2). Qu et al. [42] reported that fluorescent-dependent CDs were obtained by using citric acid and urea in 700 W for 4 $\sim$ 5 min. These synthesized CDs can emit light in dry and aggregate states. They can be applied to coat

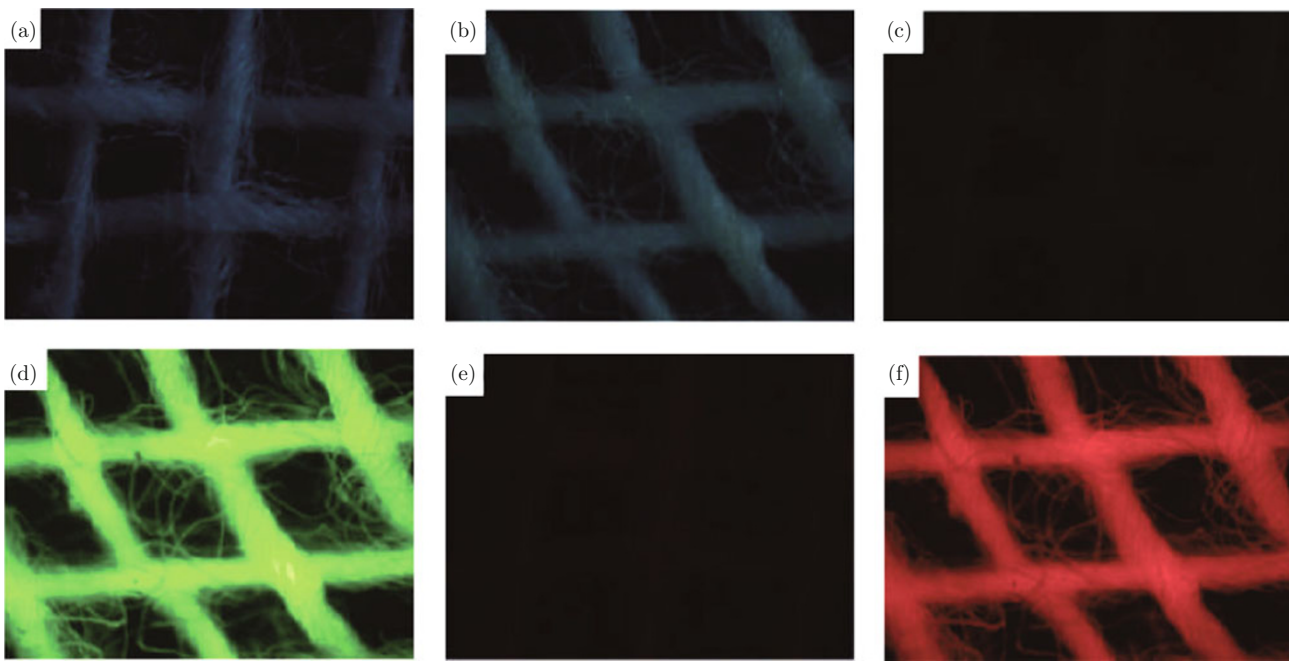


Fig. 2 Fluorescent images captured in (a) Exciter filter BP 330-385 nm, BA 420 nm, exposure time 2 ms; Fluorescent images captured in (b) Exciter filter BP 330-385 nm, BA 420 nm, exposure time 2 ms; (c) Exciter filter BP 450-480 nm, BA 515 nm, exposure time 50 ms; (d) Exciter filter BP 450-480 nm, BA 515 nm, exposure time 50 ms; (e) Exciter filter BP 510-550 nm, BA 590 nm, exposure time 260 ms of commercial gauze and (f) Exciter filter BP 510-550 nm, BA 590 nm, exposure time 260 ms of CDs-coated commercial gauze.

Table 2 Microwave synthesis of CDs

Watt or T.	Time (min)	FC	Size (nm)	Reactants	Ref.	QY (%)[a]
180°C	20	Blue	1-4	Flour	44	5.4
/	5	Blue	5	Eggshell Membrane ashes	46	14
720 W	2	Blue	/	Citric acid 1,2-ethylenediamine	52	43.8
180°C (850 W)	5	Blue	12[b]	Citric acid, PEI	53	30
700 W	2	Blue	2.2-3.0	Citric acid, EDA (BDA, DEA, TEA)	54	30.2
750 W	5	Blue	1-5	Citric acid, Urea	42	14
500 W	2-10	Blue	2.75 ± 0.45, 3.65± 0.6	PEG200, Saccharide	7, 55	3.1-6.3
900 W	10	Blue	4.5 ± 0.9	PEG200	43	16
/	10	Blue	3-4	PEG1500, Glycerine, Serine	56	12
450 W	2	Blue	6	OPPF6	57	27
750 W	14	Blue	1-4	Glycerol	58	/
700 W	10	Blue	3.5	Glycerol, TTDDA	59	12.0
750 W	14	Blue	/	Glycerol (Glycol, Glucose, Sucrose), Inorganic salts	60	3.2,9.5
700 W	5,10, 15	Blue	5.38, 5.74, 5.09	Glycerol, PEI-25k	61	9.4, 15.3, 7.0
800 W	1	Blue	27	L-arginine monohydrochloride	47	25
700 W	8/3	Blue	2 ± 0.4	Histidine, Ortho-phosphoric acid (NaOH)	48	44.9
700 W	9.5	Blue	4.6 ± 1.9	Chitosan	50	6.4
100 W	11/3	Green	3-10	Sucrose, Phosphoric acid	62	/
450 W	5-6	Blue	2-10, 2-4,1-2	Chitosan (Alginic acid, starch), Acetic acid, PEG200	51	/
800 W	2.5	Violet	3-7	Dextrin, H_2SO_4	63	5-9[d]
450 W	4	Blue	5-20	PF-68, O-phosphoric acid	64	7
700 W	1	Blue	2-8	DIA (EA, TPA), H_2SO_4 (CSA, HNO_3, HCl)	65	/
700 W	2/3	Blue	1-6	DMF, Acids	49	9
700 W	7	Blue	2.0-3.2	Acrylic acid, 1,2-ethanediamine	66	31.3
/	3.5	Blue	/	CCl_4, EDA	39	/

a. Quinoline sulfate as a standard.
b. Measure by dynamic light scattering.
c. Rhodamine 6G as a standard.
PEI stands for poly (ethylenimine). BDA stands for 1,4-butanediamine. DEA stands for diethylamine. TEA stands for triethylamine. OPPF6 stands for N-Octylpyridinum hexafluorophosphate. PF-68 stands for Non-ionic surfactant polyoxyethylene-polyoxypropylene-polyoxyethylene (PEO-PPO-PEO) block co polymer pluronic F-68. DIA stands for dimethylamine. EA stands for ethylamine. TPA stands for tripropylamine. CSA stands for chlorosulfonic acid. DMF stands for dimethyl formamide.

on commercial gauzes, vegetable fibers, animals furs, feathers, and skins as fluorescent ink (Fig. 2). These large-scale synthesized CDs can be applied in anti-counterfeit, information encryption and information storage.

Jaiswal et al. [43] performed biocompatible polymer-PEG200 as the carbon source and passivating agent to obtain blue fluorescent CDs. They modified previous report by using carbohydrate and PEG200 to synthesize nano-sized CDs [7]. They also found that PEG with molecular weight less than 800 Da can be observed by fluorescence measurements. When molecular weight of the polymer is higher, just a waxy solid can be observed. Flour [44], commercial food grade honey [45], and eggshell membrane ashes [46] as biological materials were also used to synthesize CDs under microwave condition. Nitrogen-doped CDs can also be synthesized by microwave method using nitrogen source, like nitrogen-containing amine acids [47,48], DMF [49] and chitosan [50,51]. Unluckily, the majority of synthesized

CDs in previous works showed blue fluorescence. Few works showed other fluorescence colors. So there is a long way waiting for us to modify the synthesis condition delicately.

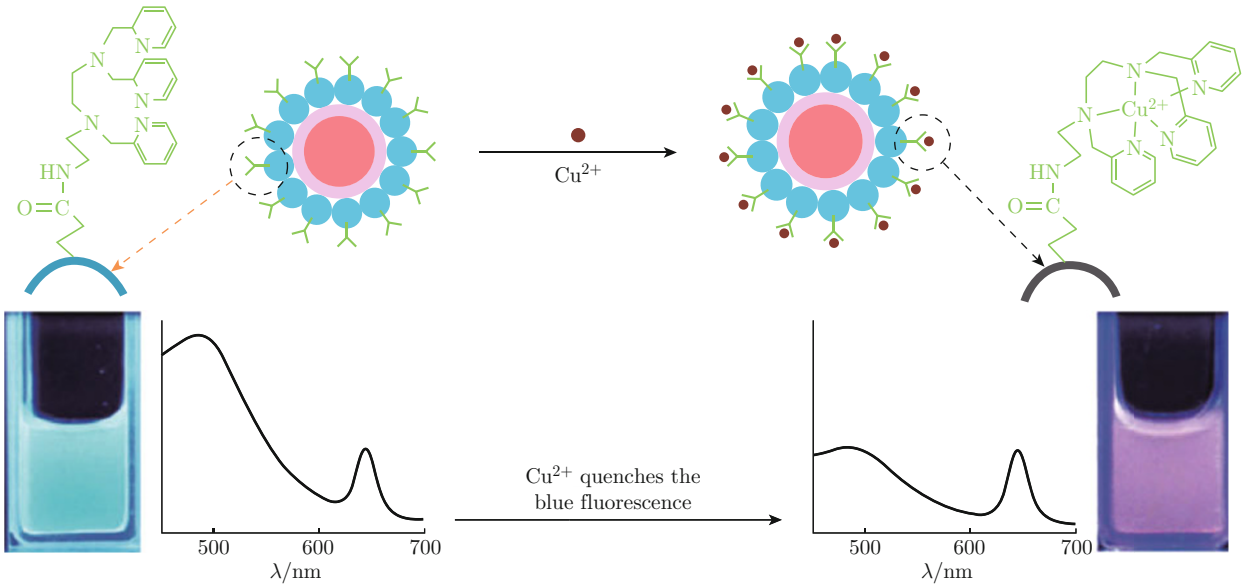
Detection application as nanoprobes

Owing to several advantages, such as high sensitivity, fast analysis and being nonsample-destructing or less cell-damaging, CDs can be used to detect metal ions, DNA assays, proteins and so on. Quenching of the fluorescence may occur through energy transfer [67], charge diverting [68], and surface absorption [69]. The quenching mechanisms can be classified as dynamic quenching and static quenching [68]. The detection of salt ions can be achieved by using an assortment of analytical methods, including atomic absorption/emission spectroscopy (AAS/AES), inductively coupled plasma mass spectrometry (ICP-MS), and spectrophotometric detection using organic dyes. The drawbacks of these techniques are that they often require a difficult-to-synthesize fluorescent-detecting probe, and sophisticated instrumentation, which limit their applications [71]. Therefore, convenient and inexpensive approaches for the sensitive and selective detection of salt ions with rapid and easy manipulation is in ever-increasing demand.

Salt ion detection

Mercury (II) ion (Hg^{2+}) is one of the most dangerous and ubiquitous pollutants, which raises serious environmental and health concerns. It is demonstrated that Hg^{2+} could easily pass through skin, respiratory, and gastrointestinal tissues, leading to DNA damage, mitosis impairment, and permanent damage to the central nervous system. Zhou et al. [72] performed unmodified CDs as fluorescence probes for rapid and sensitive detection of Hg^{2+}. A good linear correlation (R^2 = 0.992) was observed over the concentration range of 0 ~ 3 μM, while the detection limit of 4.2 nM was obtained based on a 3 δ/slope. It is known that Hg^{2+} can quench the fluorescence of QDs through an effective electron transfer process by facilitating the nonradiative electron/hole recombination annihilation [73]. It is also shown that PL enhancement of CDs/Hg^{2+} solution was attributed to the interaction between Hg^{2+} and biothiols.

Intracellular Cu^{2+} plays a critical role in the physiological and pathological events. It is a catalytic cofactor for a variety of enzymes including superoxide dismutase, cytochrome c oxidase, and tyrosinase. Little alteration in the cellular homeostasis of Cu^{2+} could cause serious neurodegenerative diseases, which may be involved in the production of reactive oxygen species. Zhu et al. [74] integrated AE-TPEA (N-(2-aminoethyl)-N,N',N'-tris(pyridine-2-yl-methyl)ethane-1,2-diamine) into a hybrid system composed of carbon and CdSe/ZnS quantum dots and developed a sensitive and selective ratiometric strategy for intracellular sensing and imaging of Cu^{2+}. Upon addition of Cu^{2+}, the intensity of blue emission from the reaction CDs shows continuous quenching, where the intensity of red emission from the encapsulated CdSe/ZnS QDs still remains constant. The ratiometric probe can be easily distinguished by naked eyes (as shown in Scheme 1). Other metal ions, such as Fe^{3+}, Pb^{2+}, Sn^{2+}, Co^{2+} and anion, such as $F^-$and I^-, can also be detected by free-labeled or labeled CDs (as



Scheme 1 Dual-emission fluorescent sensing of Cu^{2+} based on a CdSe@C-TPEA nanohybrid.

Table 3 Salt ion detection application of CDs

DS	Detection Limit	Linear Range	RA	Label	Ref.	FRR
Hg^{2+}	0.23 nM	0.5-10 nM	Lake water	Free	15	Cysteine
Hg^{2+}	Submicron molar	0.1-2.69 nM	/	PEG200, NAC	75	/
Hg^{2+}	0.5 nM	0.0005-0.01 μM	Lake water	Free	44	Cys
Hg^{2+}	4.2 nM	0-3 μM	Lake water, Fountain water, Tap water	free	72	Biothiols
Hg^{2+}	10 nM	0.5-10 μM	Lake water	P_H	71	/
Hg^{2+}	10 nM	0-5 μM	Tap water, Commercial mineral bottled water	Free	27	/
Hg^{2+}	/	0.1-2.69 nM	/	PEG200,NAC	76	/
Hg^{2+}	8.2 nM	50nM-100 μM	River water	Free	20	/
Cu^{2+}	0.09 nM (UV excitation), 0.12 nM (NIR excitation)	0.3-1.6 μM	Extracellular, Intracellular	PEI	53	/
Cu^{2+}	100 nM	1-60 μM	Normal rat brain	AE-TPEA	77	/
Cu^{2+}	1μM	0.001-0.1 μM	Living cell	AE-TPEA	74	/
Cu^{2+}	6 nM	10-1100 nM	River water	BPEI	78	EDTA
Cu^{2+}	13 nM	0.001-0.1 mM	Living cells	Amino TPEA	79	/
Cu^{2+}	0.58 pM	0.002-1.5 nM	Hair and tap water	BSA, Lys	80	/
Fe^{3+}	0.32 μM	0-20 μM	Real water	Free	22	Dopamine
Fe^{3+}	1 ppm	/	/	/	13	/
Fe^{3+}	2 nM	0-1 μM	/	Free	81	/
Ag^{+}	500 pM	/	Lake water	ssDNA	82	/
Pb^{2+}	5.05 μM	0-6.0 mM	/	/	83	/
Sn^{2+}	0.36 μM	0- 4 mM	/	Free	84	/
Co^{2+a}	0.67 nM	1.0-1000 nM	HepG2 cells	CTAB	85	/
I^{-}	430 nM	0.5-20 μM	Urine	Hg^{2+}	52	/
F^{-}	0.031 μM	0.10-10 μM	Toothpaste, Water samples	$Zr(H_2O)_2$·EDTA	55	/

a. Electrogenerated chemiluminescence (ECL) sensor.
DS stands for Detection Substance. RA stands for Real Application. FRR stands for Fluorescence Recovery Reagent. P_H stands for FAM dye-labeled ssDNA probe. NAC stands for N-acetyl-L-cysteine. BPEI stands for branched poly(ethylenimine).

shown in Table 3).

Organic/biological molecular and target gas detection

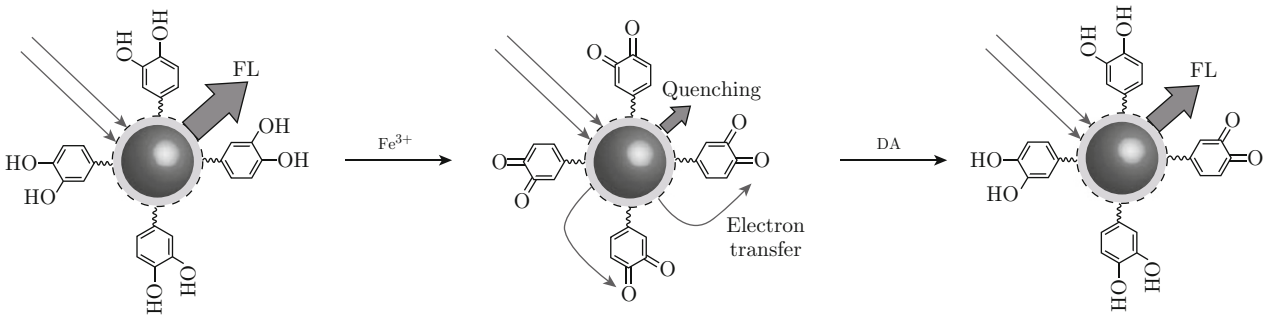
Dopamine (DA) is one of the most important catecholamine neurotransmitters in the mammalian central nervous system. Abnormal DA concentration in the brain may result in serious diseases, such as Parkinson's disease [86]. Qu et al. [22] demostrated that Fe^{3+} could oxidize the hydroquinone groups on the surface of CDs to quench the PL. When DA is added to the CDs/Fe^{3+} system, hydroquinone groups of DA can react with Fe^{3+}. It can turn on the fluorescence of CDs (as shown in Scheme 2).

Yu et al. [87] reported a naphthalimide azide anchored CDs for the selective sensing for H_2S. It is a fluorescence resonance energy transfer (FRET)-based ratiometric sensor which can ensure more accurate detection with a detection limit of 10 nM.

More related works are summarized in Table 4.

Other detections

Chen et al. [25] reported a detection method using organosilane-functionalized CDs as temperature probes at 293-343 K. SiC-dot solutions and films both exhibited rapid temperature-dependent PL responses. The temperature induced PL quenching mechanism is related to the temperature enhanced population of nonradiative channels of surface (trap/defect) states [101]. The synthesized Gd (III)-doped CDs showed dual fluorescence/magnetic resonance imaging character presented by Bourlinos et al. [102]. Mandal et al. [103] demonstrated a rapid detection of bacteria and their counting with CDs as a fluorescent marker. In addition, Wu et al. [104] designed a ECL sensor with Ag hybrid and graphene assisted which has a limit of detection of 10 cells/mL at 3σ. It is much better than cytosensors based on CdSe quantum dots. Single-label nanobeacons have also been detected by CDs [105].



Scheme 2 Schematic representation of fluorescent CDs for detection of Fe^{3+} and dopamine.

Table 4 Organic/biological molecule and target gas detection applications of CDs

DS	Detection Limit	Linear Range	RA	Label	Ref.
Thrombin	1 nM	0-100 nM	/	TBA29, TBA15-SNPs	88
Thrombin	5 nM	0-120 nM	/	ssDNA	89
AFP[b]	0.025 pg·mL^{-1}	100 fg·mL^{-1}–100 ng·mL^{-1}	Clinical serum samples	PAMAM	90
DNA	1 nM	3-80 nM	/	Methylene blue	91
Dam MTase assay	0.1 U·mL^{-1}	0.5-100 U·mL $^{-1}$	Antibiotics and anticancer drugs	Free	92
Nucleic acid	/	/	/	Dye-labeled ssDNA	93
Phosphate	0.51 pM	0.4-15 nM	Artificial wetlands	Europium	94
Nitrite	0.53 pM	0.1-10 nM	Pond water, River water, Pure milk	H_2O_2, $NaNO_2^a$	56
H_2O_2	0.4 μM	1-100 μM	/	Free	65
Glucose	0.5 μM	1-5 μM	/	Free	65
Glucose	0.4 μM	0.001-0.5 mM	Serum	TMB, $H_2O_2^d$	95
Glucose	45 μM	2-18 mM	Human blood serum	AuNPs-rGO	18
Glucose	0.01 pmol	0.5-9 mM	Human serum	Free	96
Uric acid	/	0.1-1.8 mM	Human urine	Free	96
Ach	30 pM	0.05-10 nM	Plasma, Blood	rGO	97
Cys (HCy and GSH)	4.9 nM (6.1 nM, 8.5 nM)	0.01-5 μM (0.01-5 μM)	Fetal bovine serum	Hg^{2+}	72
PCP[a]	1.3x10^{-12} g·L^{-1}	10 pg·L^{-1}–1.0 μg·L^{-1}	River water, Tap water	Free	98
Dopamine	68 nM	0.1-10 μM	Human urine and serum	Fe^{3+}	22
Dopamine	11.2 nM	0.1-30.0 μM	Injection solution	Chitosan	99
Ferrous succinare	11.2 μM	0.05-0.5 μM	Commercial Tablets	Free	26
NO	3 nM	/	Living cell	Phenylenediamine-containing naphthalimine	100
H_2S	10 nM	/	Living cell	Naphthalimide azide	87

a. Electrogenerated chemiluminescence detection.
b. Electrochemical immunosensor.
c. Co-reactant.
TBA29: 5'-NH_2-TTTTTTAGTCCGTGGTAGGGCAGGTTGGGGTGACT. TBA15: 5'-NH_2-TTTTTTGGTTGGTGTGGTTGG. SNPs stands for silica nanoparticles. AFP stands for alpha-fetoprotein. ACh stands for acetylcholine. PAMAM stands for polyaminoamine dendrimers. TMB stands for 3,3',5,5'-tetramethylbenzidine. AuNPs-rGO stand for Au nanoparticles-reduced graphene oxide. PCP stands for pentachlorophenol. "/" stands for no information.

Conclusions

Due to their high optical absorptivity, tunable fluorescence emission and excitation wavelength, excellent photostability and high sensitivity and selectivity to target analytes, CDs have wide applications in aspects of sensor, cell imaging, and drug delivery. In this review, we summarized recent advances on hydrothermal, solvothermal, and microwave synthesis of CDs, as well

as their applications as nanoprobes. In the near future, more novel works about synthesis methodology of multicolor CDs and new detection applications will be further developed.

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