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# Review Article

## Carbon Dots: Zero Dimensional Fluorescent Material

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#### Abstract

Luminescence and fluorescence phenomena have very interesting concepts in nanotechnology. Luminescent materials have very diverse and unique properties, which attract so many scientists to do the research in this field to implement in different sectors. After the discovery year in the research of carbon dots in 2004, the immense study of carbon dots was invented, which included different fluorescent properties of carbon dots. It has various and valuable properties like water solubility, stable fluorescent property, exceptional photostability, high sensitivity with low toxicity, which can be applied in various disciplines of research areas. Many synthesis methods of carbon dots have been reported, therein hydrothermal technology is a simple and accurate method to synthesis very fined C-dots. There are many conventional research methods, which defined the fluorescent study of C-dots by using different precursors in the synthesis process. This article represents the different properties and synthesis of conventional methods of CDs. Even it describes the various important applications of CDs in different sectors. It also conveys the brief information of CDs as a fluorescent material and its various applications through some literature survey.

#### Keywords

Carbon dots; Photostability; Applications of CDs; Fluorescent material

#### Introduction

Carbon Dots (CDs) is a new class of nanostructured materials that have recently attracted general interest for their unique properties of strong tunable photoluminescence, low cost, low toxicity, and biocompatibility. These properties have led to a series of potential applications as in light-emitting diodes [1,2] solar cells [3,4] sensing [5] catalysis [6] integration in photovoltaic devices, etc.[5] and more importantly to a possible breakthrough in bioimaging and medical diagnosis [7]. Usually, CDs are quasi-spherical nanostructures 2 nm-10 nm, consisting of amorphous carbon core that contains well embedded smaller graphitic and turbostratic parts. The external surface is covered mainly by carboxylates and several other organic functional groups such as hydroxyl, amines, and amides depending on the preparation methods and the precursor molecules [8,9]. The presence of hydrophilic groups at the surface of CDs induces excellent solubility in water. The main characteristic of CDs is the relatively

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strong photoluminescence, which mainly depends on their size, the excitation wavelength, and the surface functionalization [1,9]. Carbon quantum dots, or Carbon Dots (CDs) as they are often called, were discovered in 2004 during the purification of single-walled carbon nanotubes prepared through wet-chemical synthesis. Carbon dots are small nanoparticles (generally<10 nm) that are water-soluble, highly photoluminescent, inexpensive to make, have good biocompatibility, and are believed to be nontoxic [5,10]. Strong photoluminescence, easy and low-cost preparative methods, stability, biocompatibility, and low toxicity are the main characteristics and properties of CDs that have attracted huge interest for their use in various potential applications, especially in biosensing and imaging, as well as in lightemitting devices, fluorescence probes, environmental engineering, and photocatalysis. Among a plethora of different procedures that have been presented up to now, microwave or thermal pyrolysis, electrochemical oxidation, hydrothermal treatments, and laser ablation are the most common approaches to create CDs (Figure 1). As a precursor, a variety of organic compounds from natural carbohydrates to specific chemicals have been reported [11-13]. After the formation of carbon dots, a post purification step is often required, as it appears to significantly affect the fluorescence of final products [14-16]. Incorporation of highly luminescent C dots in appropriate solid-state matrices is highly attractive to materialize their properties in real device applications. Stabilization in solid materials such as polymers, ceramics, and inorganic oxides is an important challenge since often fluorescence quenching pathways are presented due to CD aggregation or other chemical interactions of its external surface. In the literature, there are several examples where CDs were successfully incorporated into polymer matrices such as Polymethyl methacrylate (PMMA), Polydimethylsiloxane (PDMS), Polyvinylpyrrolidone (PVP), Polyvinyl Alcohol (PVA), or gel glass. In most cases, the embedded CDs in solid polymeric films showed stable photoluminescence [10,16-17].

#### **History of Carbon Dots**

CDs were discovered accidentally in 2004 at the time of purification of Single-Wall Carbon Nanotubes (SWCNTs). Two years later, in 2006, the first synthesized stable photoluminescent carbon nanoparticles of different sizes and named them "Carbon Quantum Dots" (CQDs). Within a year, water-soluble CDs passivated with polypropionylethylenimine-co-ethylenimine had been reported [18,19]. The CDs showed two photon-induced luminescence spectra and were utilized to detect human breast cancer MCF-7 cells [17]. CD is the youngest member in the family of the nanoworld. They are commonly spherical having an average diameter of less than 10 nm. CDs have only sp<sup>2</sup>-hybridized carbon framework whereas CDs are composed of both sp<sup>2</sup> and sp<sup>3</sup> hybrid carbon networks. Moreover, they can be easily functionalized with hydroxyl, carboxyl, carbonyl, amino, and epoxy groups over their surfaces thereby offering extra advantages for binding with both inorganic and organic moieties [18,19]. The functionalities specifically allow the surfaces of CDs to espouse either with hydrophilic or with the hydrophobic character which finally provides the necessary thermodynamic stabilities in deferent solvents especially in water. A further advance in this aspect occurred in 2012 when some researchers used grass as a raw material to prepare CDs through facile hydrothermal treatment for the detection of Cu<sup>2+</sup>,

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which sets off a craze for the synthesis of CDs from natural substances [15]. Raw materials containing these elements enrich the materials for the preparation of CDs. Although element doping avoids the tedious modification process and could obtain CDs with excellent PL qualities and good performance. Unfortunately, the action mechanism of these doped hetero atoms is not fully and reasonably explained. Compared with other elements, the nitrogen source is easy to obtain and N-doping is an effective way with a relatively clear mechanism, to improve the properties of CDs [20]. Carbon dots, also known as carbon quantum dots or Graphene quantum dots, possess chemical structures and physical properties similar to those of Graphene oxide. CDs present several interesting and attractive chemical, biological,

and physical properties. CDs can maintain their photoluminescence property after bonding with another material [21].

#### Different Methods to Synthesis CDs

CDs can be synthesized by two general strategies: top-down and bottom-up methods (Figure 2). The former involves breaking down or cleaving larger carbon structures to smaller ones via chemical, electrochemical, or physical approaches. The top-down approach has other techniques to synthesis CDs like arc-discharge, laser ablation, electrochemical, oxidation, chemical oxidation, and ultrasonic synthesis. The bottom-up approach has microwave synthesis, thermal decomposition, hydrothermal treatment, template-based routes, and plasma treatment to fabricate CDs [22].





#### **Chemical ablation**

Strong oxidizing acids carbonize small organic molecules to carbonaceous materials, which can be further cut into small sheets by controlled oxidation. This method may suffer from harsh conditions and drastic processes. Some literature reported a simple route to prepare luminescent CDs in an aqueous solution by dehydrating carbohydrates with concentrated H<sub>2</sub>SO<sub>4</sub> followed by breaking the carbonaceous materials into individual CDs with HNO, and passivating with amine-terminated compounds (4,7,10-trioxa-1,13-tridecanediamine). The surface passivation was essential for the Photoluminescence (PL) of these CQDs [23-26]. The emission wavelength of these Carbon Quantum Dots (CQDs) can be tuned by differing the starting material and the duration of the nitric acid treatment. The multicolor emission capabilities and nontoxic nature of these CQDs enable them to be applied in life science research. Photoluminescent CQDs were synthesized in one-pot using Polyethylenimine (PEI), a cationic branched polyelectrolyte, as both a carbon source and passivating agent via HNO, oxidation. In contrast to the commonly reported pH-insensitive CQDs, the PL of these CQDs was highly pH-sensitive i.e. the PL intensity decreased with increasing pH from pH 2 to 12. In addition, the pH response of the PL behavior was reversible. This property endows them the potential to serve as proton sensors in monitoring cell metabolization processes with proton release. When incubated with HeLa cells, the CQDs could readily penetrate the cell membrane and exhibit low cytotoxicity and favorable biocompatibility, which is essential for HeLa cell imaging [22,27].

#### **Electrochemical carbonization**

Electrochemical soaking is a powerful method to prepare CQDs using various bulk carbon materials as precursors. However, there are only a few reports about electrochemically carbonizing small molecules to CQDs. Preparation of CQDs via the electro-chemical carbonization of low-molecular-weight alcohols is proposed. Two Pt sheets were used as the working and auxiliary electrode, and a calomel electrode mounted on a freely adjustable Luggin capillary was used as the reference electrode. The alcohols were transformed into CQDs electrochemical carbonization under basic conditions. The sizes and graphitization degrees of these CQDs increase with the increasing applied potential. The resultant CQDs with amorphous core exhibited excellent excitation-and size-dependent PL properties without complicated puri cation and passivation procedures. Note that the Quantum Yields (QYs) of these CQDs can reach 15.9%. CQDs can be prepared from divergently small molecular alcohols showing low toxicity to human cancer cells [27].

#### Laser ablation

In early 2006, some of the literature concluded that the CDs via laser ablation of a carbon target in the presence of water vapor with argon as a carrier gas at 900°C and 75 kPa [28-30]. Reflux system with HNO<sub>3</sub> for 12 h and passivating the surface by attaching simple organic species such as PEG1500N (amine-terminated polyethylene glycol) and poly(propionylethyleneimine-co-ethyleneimine) (PPEI-EI), the acid-treated CDs gave bright luminescence emission. The synthesis of fluorescent CDs by laser irradiation of a suspension of carbon materials in an organic solvent by selecting organic solvents, the surface states of the CDs could be modified to achieve tunable light emission. Based on control experiments, the origin of the luminescence was attributed to the surface states related to the ligands on the surface of the CDs.

A simple laser ablation approaches to prepare CDs using nanocarbon materials as the starting material and a simple solvent as the liquid media. In a typical procedure, 0.02 gm of nano-carbon material was dispersed in 50 mL of solvent (such as ethanol, acetone, or water). Ultra-sonication of 4 mL of the suspension was dropped into a glass cell for laser irradiation. Nd:YAG pulsed laser with a second harmonic wavelength of 532 nm was used to irradiate the suspension. In laser irradiation, the solution was centrifuged to obtain the supernatant containing the CDs [29].

#### Ultrasound-assisted method

In 2011, Li and his co-workers treated the aldohexose in an acid or base environment and with success obtained PL CQDs with a diameter of 5 nm and QY of 7% [31]. This ultrasound-assisted methodology for CQDs synthesis employs the high energy of an inaudible wave to crack carbon materials into Nanoparticles (NPs) at the presence of acid, alkali, or oxidizer, which is taken into account as a replacement pathway of CQDs synthesis. The employment of high energy of inaudible wave avoids the advanced post-treatment method, thereby realizing the facile synthesis of CQDs with small size. Figure 3 has illustrated the ultrasound-assisted synthesis method and also the formation mechanism of CQDs with inexperienced luminescence from potato starch [32]. These ready CQDs acts as associate degree effective fluorescent sensing looked for sensitive and selective detection of Zn<sup>2+</sup> in solution. The oxygen-rich teams on the surface of CQDs will be changed victimization different materials, which has further vital applications within the aspects of sensing and chemical action [33].

Huang has rumored associate degree ultrasound-assisted synthesis strategy of a thiol-terminated synthetic resin glycol (PEG-SH) functionalized fluorescent CQDs [34]. By scrutiny the morphology and performance of CQDs and PEG-SH-CQDs, it's incontestable that the introduction of hydrophilic PEG not solely improves its dispersibility in the liquid section however additionally endows the CQDs with wonderful biocompatibility, which has exceptional significance for the fabrication of CQDs with designable properties and functions. Lu et al. [35] synthesized blue fluorescent N-CQDs by unhearable treatment of monoamine neurotransmitter in dimethylformamide (DMF), which exhibited smart stability of mixture and light-weight in solution. Compared with the CQDs ready by Huang, albeit identical solvent (DMF) is employed within the synthesis method, the obtained sizes of the CQDs area unit different, which verifies the importance of the carbon supply employed in the preparation method. From the present analysis progress, the ultrasound-assisted technique has the characteristics of low requirements for instrumentation, straightforward operation, and time-saving however the QY of CQDs is sometimes low, which is tough to satisfy the wants of high fluorescent performance. Therefore, it's necessary to control the preparation method strictly [36].

#### **Microwave irradiation**

Microwave irradiation of organic compounds is a rapid and lowcost method to and Diethylene Glycol (DEG) as the reaction media, green luminescent CQDs were obtained within one minute under microwave irradiation. These DEG-stabilized CQDs (DEG-CQDs) could be well-dispersed in water with a transparent appearance. With an increase in the excitation wavelength, the intensity of the PL first increased to a maximum (360 nm excitation) and then decreased. However, no perceptible of the PL peak over an excitation range from



320 nm to 380 nm could be observed. Moreover, these DEG-CQDs could be efficiently ingested by C6 glioma cells and exhibited low cytotoxicity, suggesting their potential in bioimaging. Microwavemediated pyrolysis of citric acid can make the bond with various amine molecules to synthesize highly luminescent CQDs. The amine molecules, especially primary amine molecules, play a dual function as N-doping precursors and surface passivating agents for the CQDs, which enhanced the PL performance. The QY values greatly increased with an increase in N content for the CQDs fabricated from citric acid and 1,2-ethylenediamine, showing a QY up to 30.2%. The resultant CQDs are highly biocompatible and have great potential for biomedical applications [37,38].

#### Hydrothermal/solvothermal treatment

Hydrothermal Carbonization (HTC) or solvothermal carbonization is a low cost, environmentally friendly, and non-toxic route to produce novel carbon-based materials from various precursors. Typically, a solution of an organic precursor is sealed and reacted in a hydrothermal reactor at high temperature. CQDs were prepared via HTC from many precursors such as glucose, citric acid, chitosan, banana juice, and protein. Highly photoluminescent CQDs with a QY of 26% can be synthesized in one step by hydrothermal treatment, followed by centrifugation. These CQDs with sizes of 1.5 nm-4.5 nm were applied in bioimaging due to their high photostability and low toxicity. One-step synthesis of amino-functionalized fluorescent CQDs can be formed by hydrothermal carbonization of chitosan at 180°C for 12 hours. The amino-functionalized fluorescent CQDs can be used directly as novel bioimaging agents [15,39]. Solvothermal carbonization followed by extraction with an organic solvent is a popular approach to prepare CQDs. Carbon-yielding compounds were subjected to heat treatment in high boiling point organic solvents, followed by extraction and concentration. Two kinds of CQDs can be synthesized by this method, hydrophobic and hydrophilic with diameters less than 10 nm from the carbonization of carbohydrates. The hydrophobic ones were produced by mixing different amounts of carbohydrate with octadecylamine and octadecene before being heated up to 70°C-300°C for 10 min-30 min. The hydrophilic ones can be synthesized by heating an aqueous solution of carbohydrate within wide pH ranges. The hydrophilic CQDs with yellow and red emissions can also be synthesized by mixing an aqueous solution of carbohydrate with concentrated phosphoric acid followed by heating at 80°C-90°C for 60 min [40,41].

#### **Different Properties of CDs**

#### **Optical absorption**

CQDs typically show obvious optical absorption in the UV region (260 nm-320 nm), with a tail extending to the visible range. For example, CQDs produced from the electrochemical oxidation of Multi-Walled Carbon Nanotubes (MWCNTs) show an absorption band at 270 nm, with narrow Full Width at Half Maximum (FWHM) of 50 nm [42,43]. In addition, recent studies have shown that the absorbance of CQDs can be red-shifted after specific surface modification. For instance, the absorption wavelength of the CQDs passivated by TTDDA is increased in the 350 nm to 550 nm range, thus inducing increased intensity of emission in this region. CQDs

obtained after the treatment of PEG-200 and NaOH clearly show a distinct absorbance peak centered at 262 nm [37] (Figure 4).

#### Photoluminescence (PL)

PL is one of the most fascinating features of C-dots. Studies of the optical properties of small-sized C-dots are controversial due to the exact mechanisms of PL, which remain unsettled and require further clarification. Nevertheless, some aspects are well recognized and established, and are briefly reviewed here for relevance to the design of C-based luminescent probes and photocatalysts. One interesting feature of the PL of C-dots is the clear lex dependence of the emission wavelength and intensity, whether it is due to differently sized nanoparticles (quantum effect) and/or different emissive traps on the C-dot surface, or a mechanism currently unresolved. Similarly, the requirement for surface passivation is little understood but appears to be linked to the synthetic method. For example, only after surface passivation treatment by certain organic moieties do C-dots produced by laser ablation show bright PL emission. The resulting PL emission spectra were generally spectrally broad, ranging from the visible into the NIR and depending upon lex [17]. The optical behavior may reflect not only effects from particles of different sizes in the sample but also a distribution of different emissive sites on each C-dot. Mechanistically, the PL to the presence of surface energy traps, which become emissive upon surface passivation. They explained that there must be a quantum confinement effect of emissive energy traps on the surface for C-dots to exhibit strong PL upon surface passivation, a similar effect having been observed in Si nanocrystals. Surface passivation was essential for the C-dots prepared by the supported method to attain PL emission the C-dots also showed lex-dependent PL emission [39].

#### Photo-induced electron transfer property

CQDs can be quenched efficiently by either electron acceptors or electron donors in solution, that is, photo-induced CQDs are both excellent electron donors and electron acceptors. Although this photo-induced electron transfer feature of CQDs has been widely reported recently, direct evidence and essence of the photo-induced charge separation in CQDs have not been accomplished [44-46].



Some indirect experimental proof was obtained through certain redox processes. Furthermore, it was explained that photo-excited CQDs have redox-active nature which results in the reduction of metal ions in an aqueous solution. Specifically, the irradiation of CQD solution with a noble metal (silver, gold, or platinum) salt results in the formation and deposition of the noble metal on the surface of CQDs. Because the noble metal has high electron affinity, it takes electrons from the attached CQDs, again disrupting the radiative recombination, to result in the observed extremely efficient static quenching of fluorescence emissions [20].

#### Tailoring the properties of CQDs

Surface functionalization with various molecules such as organic or inorganic molecules and polymers is an effective way of tailoring the properties of CQDs by controlling their surface states. Hola et al. reported the synthesis of alkyl chain passivated CQDs with controlled size and surface functionalization [47]. The alkyl chains on the CQD surface are readily converted to carboxylate groups via mild base (NaOH) hydrolysis and induced red-shifted emission (from 402 nm to 440 nm) due to CQD surface states. Surface functionalization with para-substituted anilines endowed CQDs with new energy levels, exhibiting long-wavelength (up to 650 nm) PL of very narrow spectral widths with high quantum yields of 20%. The observed redshift of CDs in emission peaks graph from 440 nm to 625 nm was ascribed to a gradual reduction in their bandgap with the increasing incorporation of oxygen species into their surface structures. Recently, another effective way, doping has been evidenced to tune the properties of CQDs by changing their electronic structures. The Raman spectra demonstrate the intrinsic characteristics of SP<sup>2</sup> carbon with the disorder. Therefore, the N dopants heteroatoms inevitably induce increased La ((ID/IG)1) and Ld ((ID/IG)1/2) by disrupting the conjugated sp<sup>2</sup> cluster and accelerating point-like defect density through generating disordered structures. However, excessive N dopants could lead to PL quenching, possibly due to the deactivated zigzag-edge sites and effective energy transfer between N dopants and Graphene [23].

#### **Applications of Carbon Dots: A Fluorescent Material**

#### **Bio-imaging**

Traditional QDs, such as CdTe and related core-shell nanoparticles, have been used in various in vitro and in vivo optical imaging experiments. As the QDs contain toxic heavy metals, their application has raised health and environmental concerns. Due to their excellent PL properties and low toxicity, C-dots may be an attractive candidate for bioimaging applications. The C-dots were used in bioimaging applications. C-dots passivated with PPEI-EI for twophoton luminescence microscopy using human breast cancer MCF-7 cells. The bioimaging application of GQDs by incubating a solution of MG-63 (human osteosarcoma) cells with suspensions of GQDs from the stock solution with Dulbecco's Phosphate Buffer Saline (DPBS). The pig kidney cell was used (LLC-PK1 cells) to test the practicality of the as-prepared C-dots for cell imaging. C-dots were localized in the cell membrane and cytoplasm of LLC-PK1 cells. The C-dots were likely internalized into the LLC-PK1 cells through endocytosis. When excited at longer wavelengths (510 nm-530 nm), red fluorescence was observed from LLC-PK1 cells that had been cultured in a medium containing 1.2 mg mL<sup>-1</sup> C-dots for 24 h. There was no autofluorescence from cells when excited at the same wavelength. The strong PL intensity in the cells demonstrated the stability of the C-dots, confirming their strong and stable fluorescence in high ionic strength

media. The cell viability was measured after cells had been treated with different amounts of C-dots (Mg mL<sup>-1</sup>-2.4 mg mL<sup>-1</sup>). The average cell viability was greater than 95% at a C-dots concentration of up to 1.8 mg mL<sup>-1</sup>. The results revealed that C-dots are more biocompatible than QDs. A549 human lung adenocarcinoma cells were used to evaluate the cytocompatibility of the amino-functionalized C-dots obtained by hydrothermal carbonization of chitosan [1, 48].

The cell viability of the C-dots was determined by a Methylthiazolteterazolium (MTT) assay. MTT assays of cell viability studies suggested that the C-dots exhibited low cytotoxicity and posed insignificant toxic effects. These results suggested that C-dots can be used in high concentrations for imaging or other biomedical applications. C-dots were introduced into A549 cells to show their bioimaging capabilities using a confocal microscopy test in vitro. The results showed that the photoluminescent spots were observed only in the cell membrane and cytoplasmic area of the cell, but were very weak in the central region corresponding to the nucleus, indicating that C-dots easily penetrated the cell but did not enter the nuclei. The observation is consistent with previous studies on the interaction of living cells with nanomaterials, in which genetic disruption did not occur. The results further confirmed the low cytotoxicity of C-dots. In vivo, optical imaging using C-dots produced by laser ablation has been demonstrated differently. Recently, C-dots made from MWNTs were chosen for in vivo fluorescence imaging studies [49]. In most cases, heavy metal quantum dots like Cd, Te, and Se are used to achieve high performing QDs for bio-imaging purposes. However, an excess amount of heavy metal causes diseases like Wilson's disease, Minamata disease, Itai-Itai, etc. Carbon quantum dots have many advantages distinguished by multicolor emission, low cytotoxicity, excellent biocompatibility, and prominent photostability, allowing them as a quintessential candidate for bio-imaging. Carbon dots are extensively utilized for cellular imaging of many kinds of cells like HEK293, BGC823 cells, E. coli cells, pig kidney cell lines, Caco-2 cells, etc [50].

#### Drug delivery

The excellent biocompatibility and clearance from the body of carbon dots desirably meet the pre requisition of in vivo applications. The rich and tunable function groups such as amino, carboxyl, or hydroxyl, etc. endow carbon dots the ability to carry therapeutic agents, generating the theranostic nanomedicine. The bright emission of carbon dots provides the opportunity to dynamically and real-time monitor the drug distribution and response. Carbon dots synthesized by thermal pyrolysis of citric acid and polyene polyamine have been employed by Zheng and coworkers to carry oxaliplatin. One of the platinum-based drugs, where the platinum-based drugs are the most efficient anticancer drugs used in more than 50% chemotherapeutic treatment for clinical cancer patients. One of the oxaliplatin (IV) derivatives Oxa(IV)-COOH was employed to link with carbon dots, forming CD-Oxa, and the linkage was finished by the reaction between the amino groups on carbon dots surface and the activated COOH group of Oxa(IV)-COOH, where the COOH group was activated by using EDC/sulfo-NHS, The newly appeared Pt 4f XPS signal confirmed the successful bonding of Oxa(IV) to carbon dots. Inductively coupled plasma mass spectrometry (ICP-MS) analysis unveiled that the platinum element of carbon dots-Oxa was 1.5 wt%, corresponding to a 4.2 wt% oxaliplatin (IV). In addition, TEM data indicated that the size of carbon dots-Oxa was 2.71 nm ± 0.43 nm, larger than the diameter of pure carbon dots (2.28 nm  $\pm$  0.42 nm). The synthesized Carbon dots-Oxa was injected into the hepatocarcinoma 22 cell line (H22) liver cancer of Chinese Kun Ming (KM) mice by means of intralesional injection and the dynamical distribution of the drug were observed from *in vivo* optical imaging system using the blue light as the excitation source. At the instant of the exposure, a highlight point was observed from the *in vivo* imaging due to the high concentration of carbon dots, and the following delivery of Carbon dots-Oxa resulted in the fluorescent intensity decrease of initial position and a broaden intensity distribution [49, 51].

The carbon dot surface was negatively charged at pH=7.4 which corresponds to the normal physiological condition, while the surface reversed to positively charged when pH decreased to 6.8, mimicking the tumor microenvironment. Negatively charged carbon dot surface in the normal cell microenvironment suppressed the drug absorption of normal cells by means of electrostatic repulsion between the carbon dots surface and the negatively charged cell membrane, thus reducing the side effect. On the contrary, the positively charged carbon dots surface in tumor cell microenvironment could promote the internalization and cancer drug absorption because of the electrostatic interaction with negatively cell membrane both the in vitro and in vivo demonstration indicated that carbon dots could be used as an effective imaging-guided drug carrier with increased therapeutic efficiency and deceased side effect [52]. Swarup Krishna Bhattacharyya et al. studied the carbon dots, which crosslinked to the gelatine nanocomposites hydrogel. In their study, novel CDs embedded gelatin nanocomposite hydrogel is fabricated by simple solvent casting technique for the intestinal drug delivery and nearneutral pH sensing. The pH-responsive photoluminescence behavior of the GNC hydrogel in the near-neutral pH range can be useful for in vivo imaging, bio-sensing applications, and quantitative measurement of the system pH [53].

#### LEDs

The direct white light emission from carbon dots, which offered the possibility to fabricate carbon dots, based on white light-emitting devices. Later, they carried out the white light-emitting devices by using the white light carbon dots as the emissive layer. Briefly, the carbon dots were synthesized by thermal pyrolysis method using citric acid as the carbon source and 1-hexadecylamine as the surface passivation agent, which showed a relatively high quantum yield of 60% with an average diameter of 5 nm. The device structure, energy alignment, and organic molecular structure were adopted. The architecture of the device was constitutive of three layers. The buffer layer consisted of 40 nm poly(3,4-ethylenedioxythiophene)poly(styrene sulfonate) (PEDOT:PSS) in the anode, which has two functions:

- 1. Increase the work-function from 4.7 eV (ITO) to 5.0 eV
- 2. Increase the smoothness of the anode

The carbon dots emissive layer was fabricated by a spun-cast method with an optimized thickness of 20 nm. A thicker film e.g. 35 nm shall lead to the bad charge transport in the carbon dot film, but a thinner film e.g.10 nm shall increase the density of voids, grain boundaries, etc, both of which shall decrease the device EL efficiency. AFM measurement indicated that the surface roughness of the carbon dot film was less than 3 nm, which implied good compatibility between the carbon dots and the buffer layer [54-55].

The electron transport layer was employed by 40 nm thick 1,3,5-tris(N phenylbenzimidazol-2-yl) benzene (TPBI) since it's well-behaved electron transport ability. The electrode was consist

of 1 nm LiF and 120 nm Al fabricated by thermal evaporation. The pure carbon dots exhibited broad PL with the excitation dependent emission feature, while the LED device displayed stable emission even if the voltage was increased to 10 V. The Commission International d'Eclairage (CIE) coordinates of the emitted light was (0.40, 0.43) ((0.33, 0.33) for pure white light), with a color-rendering index CRI of 82. The WLED had a turn-on voltage of 6 V, with the maximum external quantum efficiency (EQE) reaching to 0.083% at a current density of 5 mA/cm<sup>2</sup>, and the maximum brightness output was 35 cd/m<sup>2</sup> with the condition of the current density of 160 mA/cm<sup>2</sup> and voltage 9 V. By modifying the device structure, color-switchable CD LEDs were demonstrated using the same carbon dots by Zhang's group [23,56]. According to the excitation dependent PL and time-resolved measurement, the authors claimed that there existed three emission channels whose lifetimes were 2 ns, 5-6 ns, and 14-15 ns, contributing to 420 nm, 460 nm, and 580 nm band emission respectively [49].

#### Sensing

The fluorescence intensity of carbon dots is closely related to the surrounding environment. The interaction between carbon dots and chemicals results in the quenching/enhancement of carbon dots emission. Thus carbon dots can be used as a king of the fluorescent probe to detect the quantity of the items. Various sensors based on the fluorescence of carbon dots have been reported and can be generally categorized into the following groups: ion sensing as Intracellular Lysine, Ascorbic Acid, guanosine 3 -diphosphate-5 -diphosphate (ppGpp), enzyme (e.g. Thioredoxin Reductase (TrxR)), DNA, etc., and temperature sensing. However, the single fluorescent wavelength intensity change-related sensing requires strict conditions. The fluctuation of the light source, the concentration of fluorescence probes, different optical paths, and even the aggregation of the probes could devalue the reliability of the sensing results. Thus ratiometric sensing as an alternate has been proposed which simultaneously collected the intensity change of two separated emission bands. The intensity ratio of the two wavelengths is the indicator of the sensing item and is considered as the output. Compared to a single emission sensor, ratiometric sensing provide self-calibration for both the light source and sensing environment, thus can vastly improve the sensing accuracy and reliability [6,4].

The carbon dots were synthesized by thermal pyrolysis of citric acid with the 4,7,10-trioxa-1,13-tridecanediamine (TTDDA) as the surface passivation agent. Both pH-sensitive Fluorescent Fluorescein Isothiocyanate (FITC) and pH insensitive Rhodamine B Isothiocyanate (RBITC) have been utilized to post-treat the carbon dots, endowing the carbon dots two emission bands located at 515 nm and 575 nm (excited by 488 nm), respectively. The pH value changed from 5 to 9 which covered the full range of physiological environment of the human body, the intensity of the 515 nm emission band increased substantially, while the 575 nm only increased a little, thus it was clearly shown that the carbon dots exhibited a linear ratiometric response to pH input. In addition, the authors also demonstrated that both the in intracellular genres (such as ions, saccharides, and proteins) and redox substances have neglected effect on the carbon dots ratiometric response, which verified the selectivity of the carbon dots based ratiometric sensing. The small molecular linked carbon dot ratiometric sensors possess high accuracy, while the ease of photobleaching and the leaching of small molecular from carbon dots also limited their practical applications [58,59].

Proper waste disposal from households and restaurants is becoming an important and recurring waste-management concern.

Preparation of C-dots from a highly abundant carbon source of waste refusals is highly effective in the commercial aspect as well as in reducing the immense environmental pollution. The multifunctionality of COC-dots has been established in their versatile area of applications in novel sensing probes for sensitive and selective detection of Fe<sup>3+</sup> ion in aqueous solution as well as in environmental samples even at very low concentration. The sensing mechanism for the COC-dots could be ascribed to the formation of the complexes between the Fe<sup>3+</sup> ion and the surface hydroxyl groups of the carbon nanodots. The versatile application of fluorescent ink and lightemitting polymer composite can be applied to UV-active marker and sunlight conversion film [59]. Similarly, Sayan Ganguly [60]. studied that Hexavalent chromium (Cr<sup>+6</sup>) has been considered a terrible pollutant because of its toxicity and carcinogenic features. But carbon dots can trace the pollutant by determining the 'on-off' fluorescence behavior. Moreover, they proved that the nanodots' surface ligands are quite susceptible to capture drug molecules which also have been tested by pH-tunable cumulative drug release method.

Carbon Dots (CDs) is a relatively new carbon nanomaterial that has been extensively studied in the last decade, due to their unique characteristics and properties (Figure 5). Strong photoluminescence, easy and low-cost preparative methods, stability, biocompatibility, and low toxicity are the main characteristics and properties of CDs that have attracted huge interest for their use in various potential applications, especially in biosensing and imaging, as well as in lightemitting devices, fluorescence probes, environmental engineering, and photocatalysis. Among a plethora of different procedures that have been presented up to now, microwave or thermal pyrolysis, electrochemical oxidation, hydrothermal treatments, and laser ablation are the most common approaches to create CDs. After the formation of carbon dots, a post purification step is often required, as it appears to significantly affect the fluorescence of final products [61,62]. Apostolos Koutsioukis et al [14] and Apostolos Koutsioukis et al. [61] studied carbon dots. They synthesized carbon dots with the help of hydrothermal method and fabricated the ink formulation with it. They formed the water-based ink/gel for the gravure printing technique with 451 nm of excitation wavelength on a different substrate. They also used several precursors to study optical properties.



They got blue fluorescence color of the liquid with yellow color of the same liquid without UV rays. Same as Cai-Feng Wang et al [62] and Jyoti Sharma et al [63] studied carbon dot-based latent fingerprinting with ink-free printing. They similarly synthesized carbon dots with help of hydrothermal method. They formed Polyvinyl Alcohol (PVA) based ink formulation to make printing ink and also used laserjet printing technique to print the fluorescence ink on bond papers with the same excitation wavelength.

Similarly, Gomez IJ et al. [64] and Dan Qu et al. [65] formed highly luminescent carbon dots with graphene nanoparticles. They synthesized nanocomposite with high blue color fluorescence of 451 nm wavelength. They also used different precursors like TRIS and glucose with carbon source Citric Acid (CA) to study the fluorescence property of carbon dots and they got the fluorescence wavelength between 400 nm-500 nm in using each precursor. Same, Guohua Hu et al. [66], Jing Wang et al. [67] and Juan Wei et al. [68] synthesized two-dimensional fluorescence carbon dots for anticounterfeiting application by using different ink printing techniques. They also mentioned the forensic application of the carbon dots. They concluded the graphene material based nanocomposite possibility with fingerprint recognition application. They coated the carbon dot-based ink/gel formulation with the help of electrospinning coating method, which is very accurate and precise on the glassbased substrate. Lulu Pan et al. [69], Kai Jiang et al. [70] and Libin Tang et al. [71] synthesized multi-color carbon dots by using the hydrothermal method to sense multi-dimensional cellular imaging. Even they also mentioned synthesizing nanocomposites, which are very important in this application area. They concluded different excitation wavelengths between 350 nm to 650 nm with a bandwidth of 288 eV. They also concluded that carbon dots have low cytotoxicity and great potentials as a multi-color labeling reagent with delocalized  $\pi$ -electrons in double bonds of carbon (C=C). Even though with this basic feature, they mentioned other applications of carbon dot like in optoelectronic, electronic, and light-emitting devices.

Meng Li Liu et al. [72] synthesized carbon dots by using the hydrothermal method with 450 nm excitation wavelength. They experimented with different precursors like urea, glucose, TRIS, and PEG with carbon source Citric Acid (CA) and ascorbic acid to analyze different properties of carbon dots. They found light to dark blue fluorescence color with long term fluorescence quality. Ruili Liu et al. [27] and Shoujun Zhu et al. [12] synthesized 1.5 nm-2.5 nm sized carbon dots by using microwave technique. They formed nanocomposites by combining carbon dots with silica nanoparticles to boost the potential of carbon dots. They concluded that silica doped carbon dots have high quantum yields and high fluorescence spectra with 452 nm wavelength, but increasing the silica nanoparticles in carbon dot solution the fluorescence spectra is going to decrease. Similarly, Sourav Chandra et al. [73] formed multidimensional high fluorescent carbon dots by using the microwave method. They used orthophosphoric acid as a carbon source and sucrose as a precursor to synthesize carbon dots for cellular imaging and bio-imaging application. They got a very nice blue color fluorescence spectra between 340 nm to 450 nm. They fabricated ink/gel formulation by using Polyvinyl Alcohol (PVA) by mixing it in a carbon dot solution. Even, Zhi Yang et al. [37] and Zhulong. Zhou et al. [74] synthesized carbon dot-based invisible ink for bio-imaging applications. They used citric acid as a carbon source and TRIS as a precursor to obtain carbon dot nanoparticles by using the lyophilization process. They fabricated carbon dot-based nanoprobes with high fluorescence spectra of 451 nm and quantum yield of 52% and the size of the carbon dots nanoparticles are 3 nm to 10 nm. The nanoprobes can easily detect the ions in the solution by quenching the fluorescence spectra of carbon dots. Same as Yamin Liu et al. [75] Ya-Ping Sun et al. [19] and Zhu Lian Wu et al. [15] synthesized highly photoluminescent dots by using carbonization process with the use of nitric acid, sulphuric acid, and carbon nanopowder. They formed metal-doped carbon dots for anti-counterfeiting applications. They got a high quantum yield of 82% and an intense fluorescence peak of 451 nm. But increasing the metal content in the solution, the fluorescence spectra are decreased. The solution is green without UV rays and with UV rays, it gave high and light blue color fluorescence. They fabricated ink/gel formulation by using polyvinyl alcohol at the ration of carbon Dots:PVA=1:9. Similarly, Zhiping Song et al. [76] formed invisible fluorescence ink by synthesizing carbon dots with citric acid and conclude the same results.

#### Conclusion

Luminescence-based materials are of great importance in the search for sensitive and selective materials detections by fabricating different sensors. Carbon Dots (CDs) have emerged as the most precious gifts in nanotechnology because of their magical properties and applications. Carbon dots have different chemical properties, water-soluble property, electrical property, optoelectrical property, and fluorescence property; the carbon dots are very suitable and easy to synthesis material in the research area. Even the structure and component of the carbon dots determine diverse properties. Carbon dots are suitable for the chemical modification process and surface passivation process with various organic, inorganic, and biological materials with water solubility and biocompatibility. The article explained the importance of carbon dots in different sectors with diverse applications and as a fluorescent material. The article also explained the different properties of CDs..

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