ISSN: 2476- 2334

Research Article

International Journal of Nanotechnology & Nanomedicine

# Structure and Photoluminescence of Carbon Dots: Different Opinions

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**Submitted:** 2023, Dec 30; **Accepted:** 2024, Jan 18; **Published:** 2024, Feb 02

**Citation:** Garg, P. (2024) Structure and Photoluminescence of Carbon Dots: Different Opinions. *Int J Nanotechnol Nanomed, 9*(1), 01-10.

**Abstract**

*Carbon Dots (CDs), a novel type of carbon-based nanomaterial, have drawn board research interest for years, due to its inherent physical and chemical properties and favourable characteristics, such as high stability, different optical features, outstanding compatibility, low cost, and eco-friendly. In this review, we look on the classification of CDs which are based on their different mechanism and also describe their various synthesis processes as well as their optical properties, such as fluorescence and photoluminescence. There are numerous review articles with differing viewpoints on the synthesis of CDs, but there is no clear picture of CDs; the topic is an open for debate. In the last, there is a conclusion of different opinions given by different authors who provided the synthesis of CD and photoluminescent.*

**Keywords:** Carbon dots, Photoluminescence, Quantum Yield, Fluorescence, Graphene quantum dots

## Introduction

"Carbon" is the most richest elements in this world. There are wide variety of carbon that are found in our nature which exists in the form of its allotropes like diamond, graphite. Nowadays, there are new types of carbon materials are formed because of its more potential use such as graphene, carbon nanotubes, and fullerenes [1] [2]. But these days, Carbon Dots (CDs) are also attracting a lot of attention. A novel type of multicoloured luminous carbon nanoparticles called CDs.

CDs are also known as carbon quantum dots [3] which are zero-dimensional photoluminescent nanocarbon having size less than 20nm [4]. Wang et al [5] proposed that there is a new class of CDs which have unique properties with size less than 10nm. In 2004, CDs was first prepared by the purification of SWNCTs (single-walled carbon nanotubes)[6] [7] [8] and then many scientists gave their different opinions about their synthesis of CDs.

### Synthesis of CDs

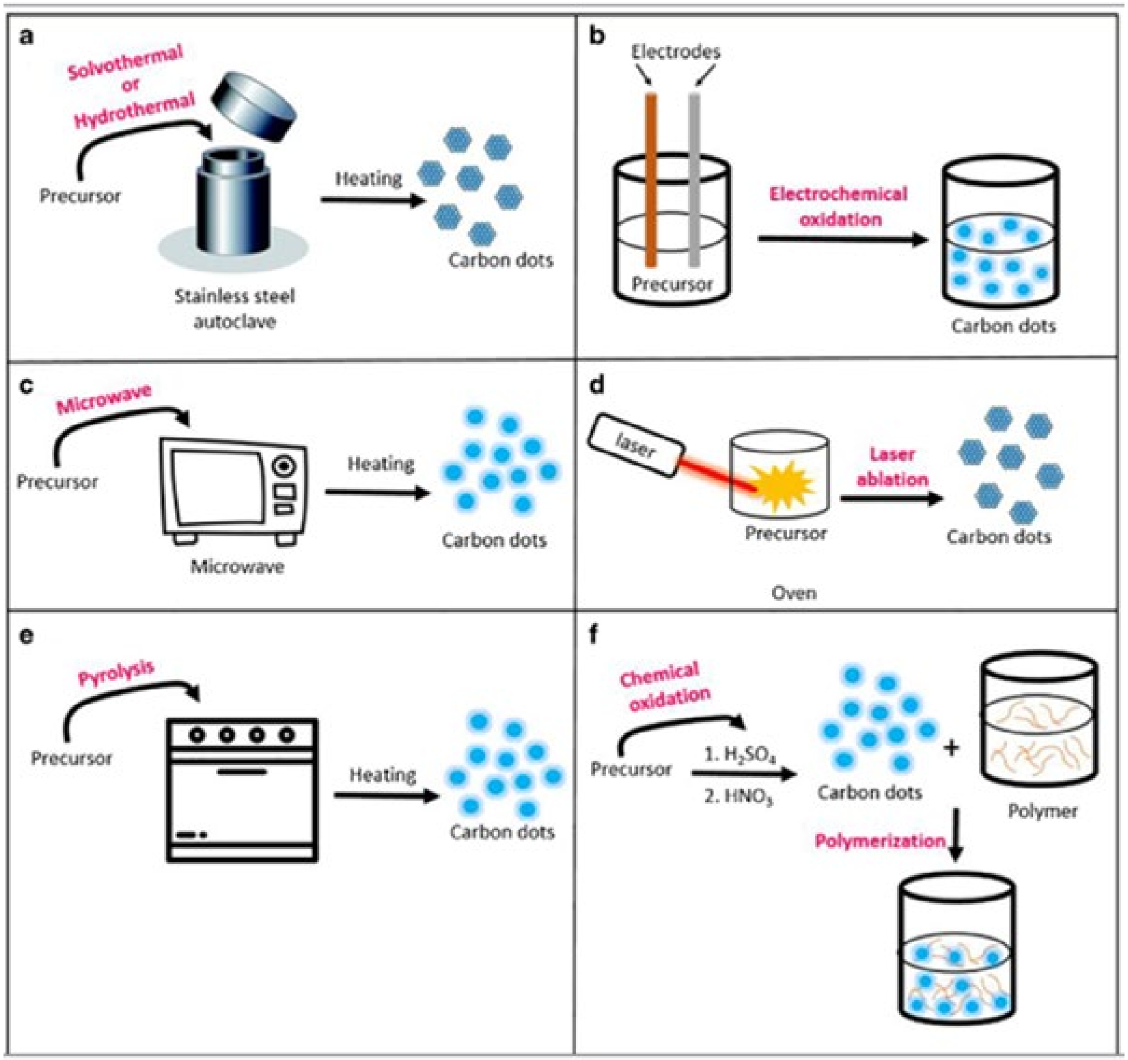
There are several methods to prepared CDs which can be divided into two approaches: top-bottom approaches & bottom-up approaches[9][10][11][12][13][14][15][16][17][18] as shown in table 1.In top-down approaches, CDs are prepared by fragmentation of carbon materials by using laser ablation, chemical oxidation & electrochemical oxidation etc.

**Laser ablation: -** Since their invention, Laser has been thoroughly explored and utilised in a discovery, particularly in Laser ablation, which is the process of using laser irradiation to remove material from a surface. Many researchers have employed the Laser ablation method to create CDs. For the purpose of producing carbon nanoparticles and thin films of carbon Suda et al [19] proposed a pulsed laser deposition approach.

**Chemical oxidation: -** In this process there is an oxidation take place in the presence of some chemical oxidant gives CDs. It also produces high-purity compounds that are thermodynamically stable. Carbonaceous substances found in nature, such as carbohydrate food material, activated charcoal, carbon black, wood, coal, candle soot, etc. is simply ignite to produce CDs. Liu et al [20] used candle soot for the production of CDs and used a chemical oxidative acid treatment. Candle soot combined with an oxidant, such as nitric acid, gives centrifuged CDs.

**Electrochemical oxidation: -** It focuses on the investigation of chemical processes occurring in solutions at the interface between an electrode and an electrolyte. Numerous research teams have employed graphite, carbon nanotubes, and graphite nanopowder as carbon precursors in electrochemical synthesis processes. Zhou et al [21] synthesis of luminous CDs multiwalled carbon nanotubes that were grown on carbon paper using the chemical vapour deposition method were subjected to electrochemical treatment.

**Table 1: Schematic synthesis of CDs**




In the bottom-up approaches [22], molecular precursor materials are carbonized via microwave methods, thermal pyrolysis & hydrothermal methods as shown in table 1 it gives the brief description about the synthesis method of CDs [23].

**Microwave synthesis: -** The advantages of the microwave-assisted technologies include their simplicity, usability, speed, energy efficiency, and industrial scalability. As a result, they are frequently utilised in material synthesis and preparative chemistry. As was previously said, sugars are a plentiful and convenient supply that can be employed as a carbon precursor.

**Thermal pyrolysis: -** Small organic molecules are heated to cause dehydration, polymerization, carbonization, and the subsequent creation of larger CDs. This simple pyrolysis or carbonization of small organic molecules has been frequently described in the synthesis of CDs [7].

**Hydrothermal method: -** The hydrothermal approach is one of many chemical processes that holds great promise for producing nanoparticles with predictable forms, sizes, surface modifications, and stability. The word "hydrothermal," which derives from the Greek words "hydro" (water) and "thermal" (heat), describes a heterogeneous chemical reaction that takes place in the presence of aqueous solvents at high pressures and ideal temperatures in order to dissolve and recrystallize substances.

Recently, taking ultrapure water as a solvent, many researchers were achieved an electrochemical approach over large synthesis of HQCDs (high quality of CDs). In recent years, there was a reported of P-doped GQDs for free radical scavenging with the help of electrochemical synthesis. Many researchers have obtained GQDs by using different methods like chemical oxidation using strong acids [24].

## Properties of CDs

CDs have certain chemical, physical, electronic and optical properties because of their specific structure [13]. Due to its different properties, CDs are used in varies platform like in medicinal industry because of their good biocompatibility, and have high quantum yield (QY), also used as optoelectronics and as a good catalysts due to its strong absorption and stability [4].

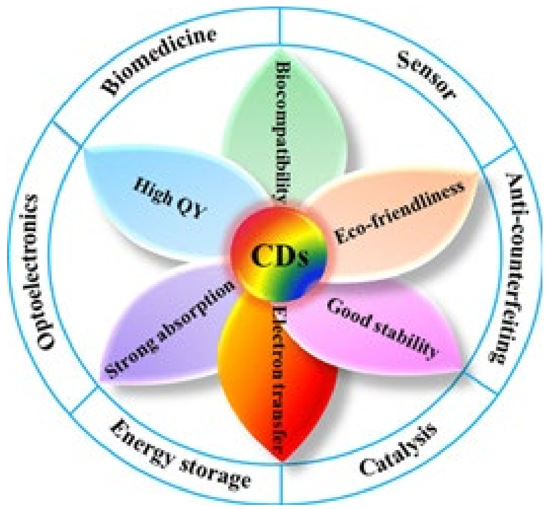



**Figure 1:** Applications of CDs

CDs have good optical properties, which can be explained by a photoluminescence (PL) and fluorescence. Firstly, we should know about the luminescence. Light is a form of energy that generally called luminescence i.e generally a "cold light" which can be emitted at very low temperature. In luminescence, when a source of light is falling on an electron then the electrons are jump and go to excited state and then return back to ground state and is explained with the help of Jablonski energy diagram and seen the origin of fluorescence and phosphorescence. There are different varieties of luminescence called Fluorescence and PL [25].

Generally, Fluorescence is most commonly found in fluorescent lights, movie special effects, in amusement park and, the redness of rubies in sunlight, "day-glow" or "neon" colour. Jelinek et al [26] proposed that the CDs properties-fluorescence explained that there is a excitation dependent emission which is only possible in case of smaller size of nanoparticles but the larger GQDs (graphene quantum dots) exhibited different colour of CDs which is depend on their excitation [27] as shown in fig.2

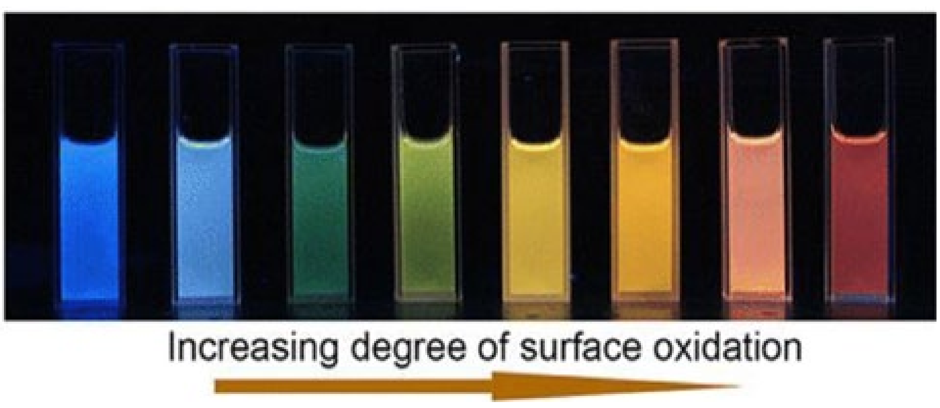
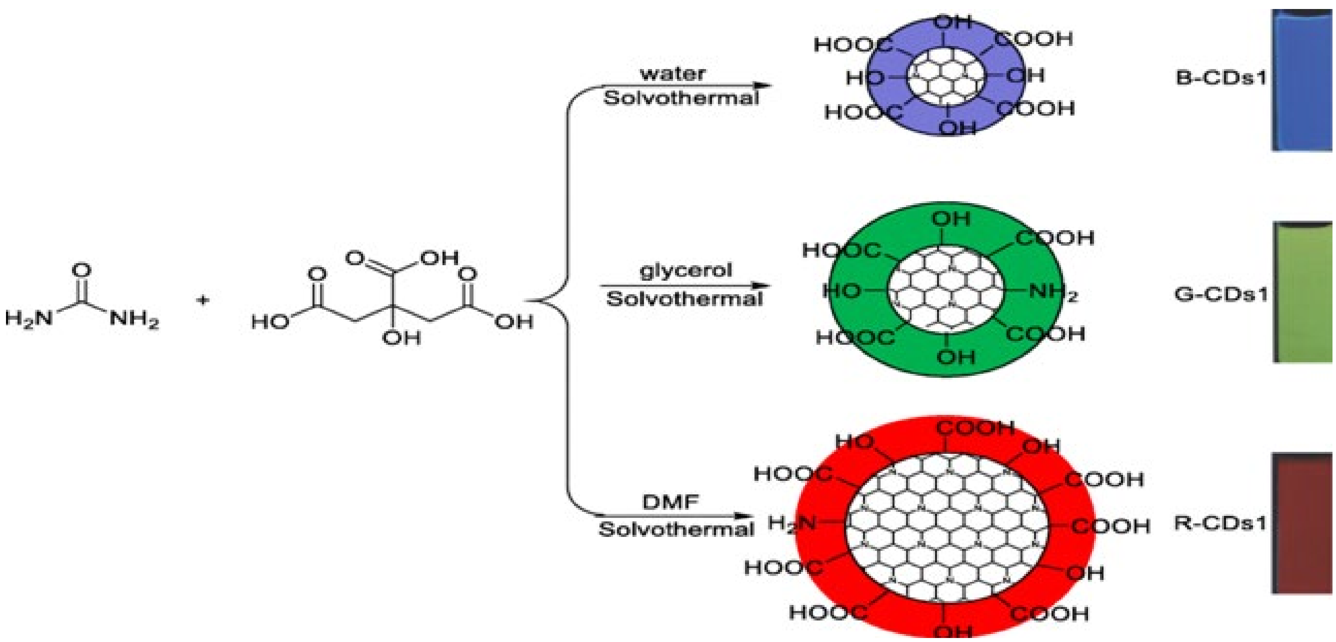



**Figure 2:** Different colours of CDs, Copyright (2016), American Chemical Society

But in actual as for many studies have been reported that the origin of fluorescence of CDs still an open debate but CDs are produced by various ingredients and gives different structures and components this means that the CDs are synthesized by using different precursors and different synthetic approaches and this makes the difference in their optical properties. Hence it is impossible to compare the CDs properties that are available in many review paper to define a compiled theory [12]. As we discussed earlier, there are one more variety of luminescence i.e. photoluminescence.

Photoluminescence (PL) is the process of reading the electronic structure of different materials. There are several methods for the origin of PL by using different precursors. But the Jablonski tells little bit about the PL emission. Yan et al [28] proposed that CDs shows the tunable PL emission and its wavelength dependent excitation. The authors described the CDs with different emission colours of fluorescence as shown in fig.3 are developed because it is affected by the electronic bandgap of conjugated π-domains.




**Figuer 3:** Schematic representation of different CDs

As we discussed above there are many properties which are attributes to define the CDs but there is no clear picture, so that many scientists gave about their opinions on the behalf of synthesis of CDs and its PL of CDs. In this review, we would compile the whole data about their different structure of CDs that are given by different scientists. Now we will be discussed about the different opinions that are given by different scientists.

Ehrat et al [29] proposed that the synthesis time of CDs which is obtained from citric acid & ethylenediamine that gives the molecule fluorophores for the blue luminescent of CDs that is shown in fig.4. But still there is no clear picture about the origin of its optical properties and its structural properties.

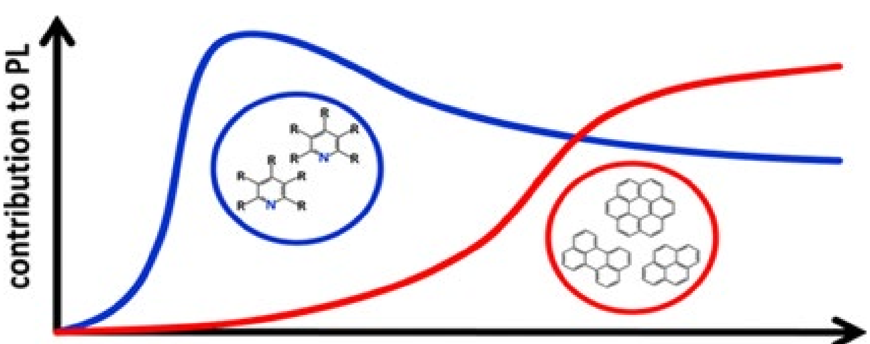



**Figure 4:** Graph between PL and synthesis time. Copyright (2017), American Chemical Society.

Righetto et al [30] proposed the structure of CDs are having sp2 hybridized domains embedded within sp3 scaffold that it is made up of carbogenic core and fluorescence correlation spectroscopy (FCS). TREPR provides deep studied into their structural form of carbon cores, where the domains of Csp2 are inserted within Csp3 as shown in fig.5

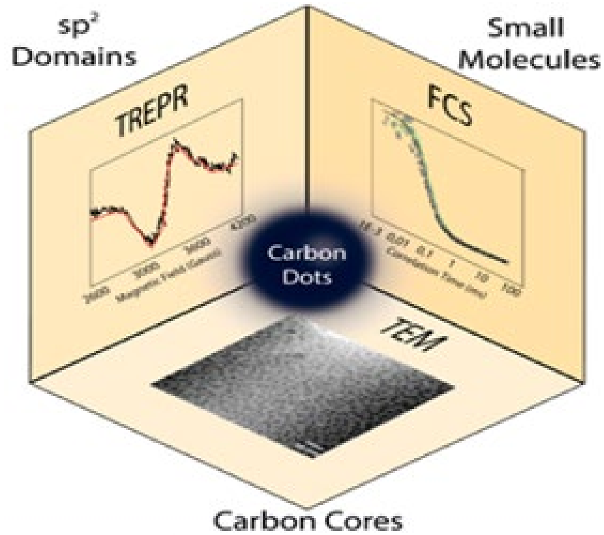
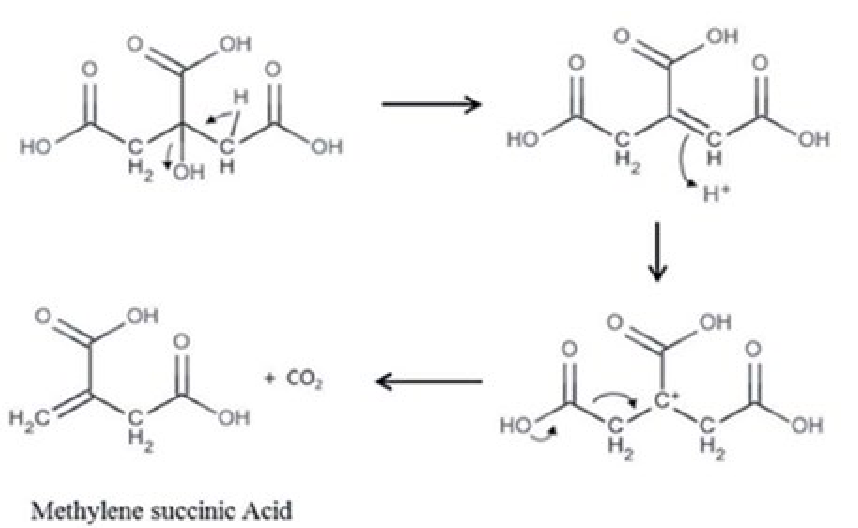



**Figure 5:** Graphical representation of CDs. Copyright (2017), American Chemical Society

Khan et al [31] prepared carbon dots of size 3-5 nm using citric acid via hydrothermal treatment and that gives methylenesuccinic acid as shown in fig.6, which gives us the nano-assemblies of H-bond with the properties like-CND. However, size of molecular fluorophore is 0.9 nm were also observed.




Strauss et al [32] proposed the thermal reaction of urea and citric acid that are formed three products namely, non- fluorescent carbonaceous particles and two molecules of fluorophores. On the basis of their result of mechanism of potential reaction which gives the nanoparticles as a product and the intermediate form i.e. fluorophores. Recently developed laser-assisted carbonization process, it was seen that the organic nanoparticle of non-fluorescent material are very good precursors as shown in fig.7

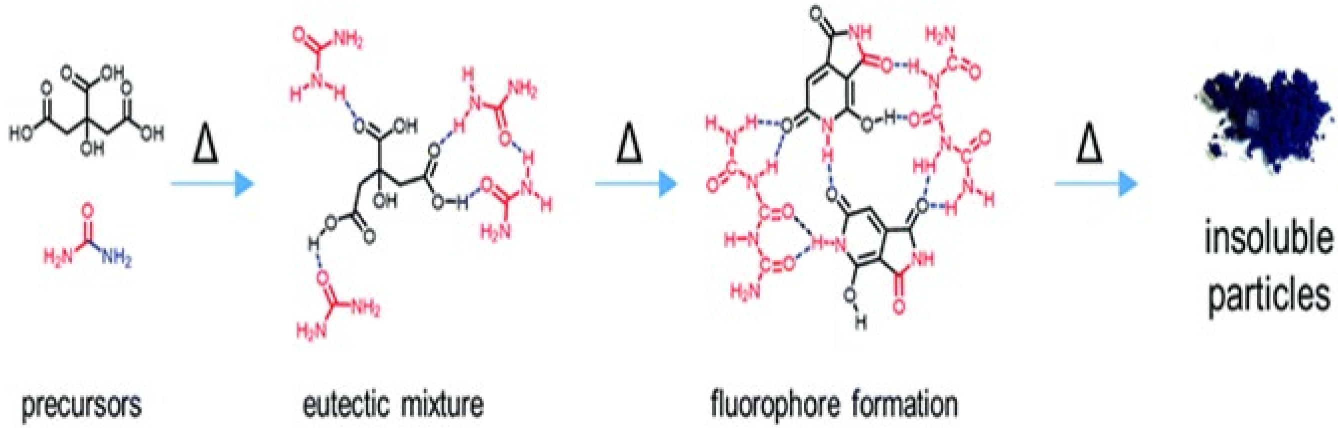



**Figure 7:** Represents the reaction between urea and citric acid and give solid nanoparticles

Das et al [33] was shown that CDs are composed of aggregated 2-pyridone derivatives employing π–π stacking & H-bonding etc. The equation followed by PL decay is single-exponential decay. The CD is suitable probe for FILM and FRET because it has long & single-exponential PL lifetime & also have high PLQY and its dependency is on the polarity. The authors could be shown that CDs behaves as a single particle which do not blink & are alive only for few minutes. These things must improve their optical properties which will make CDs as a single-particle level toward bioimaging and also a better optical emitter as shown in fig.8

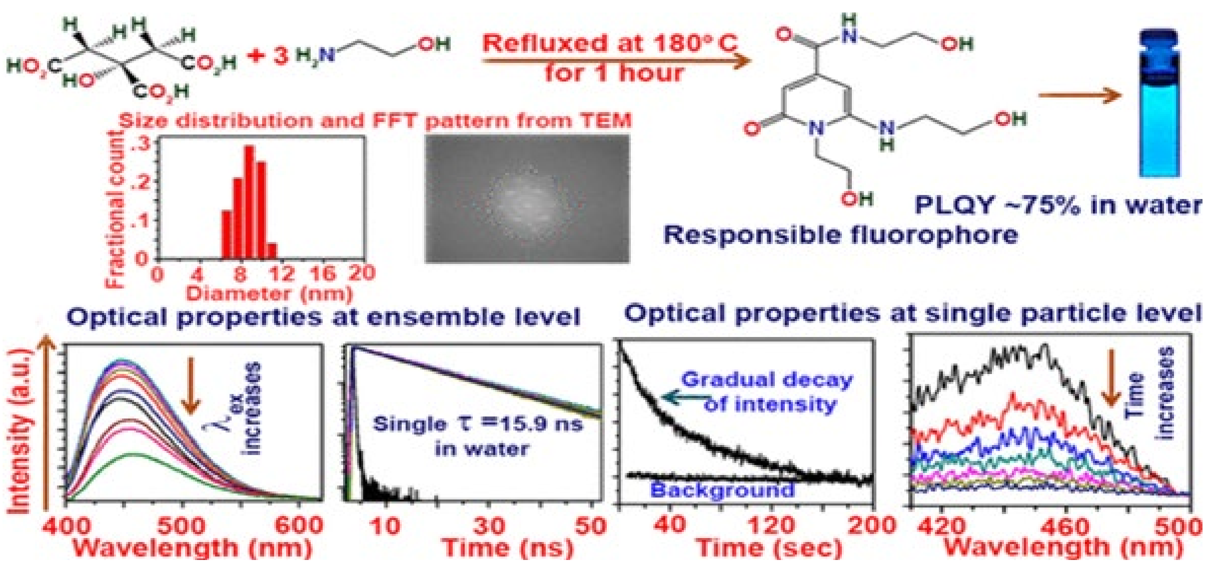



**Figure 8:** Represents the graph of optical properties at ensemble level and at single particle level, Copyright (2017), American Chemical Society.

Gude et al [34] has proposed the steady and dynamic photoluminescence behaviour of CDs. The synthesis of CDs starting from carbohydrates like sucrose, fructose and glucose employing simple synthesis method. CDs gives the aggregates of hydroxymethylfurfural (HMF) derivatives as shown in fig.9. In CDs, they exhibit excitation an independent wavelength of PL emission maxima in orange-red region. According to this, there is presence of only one type of chromophore in CDs. It shows that same chromophore unit is the fluorescing, not only the individual fluorophores. The PL emission maximum are independent on single exponential PL lifeline & also independent of polarity of the medium.

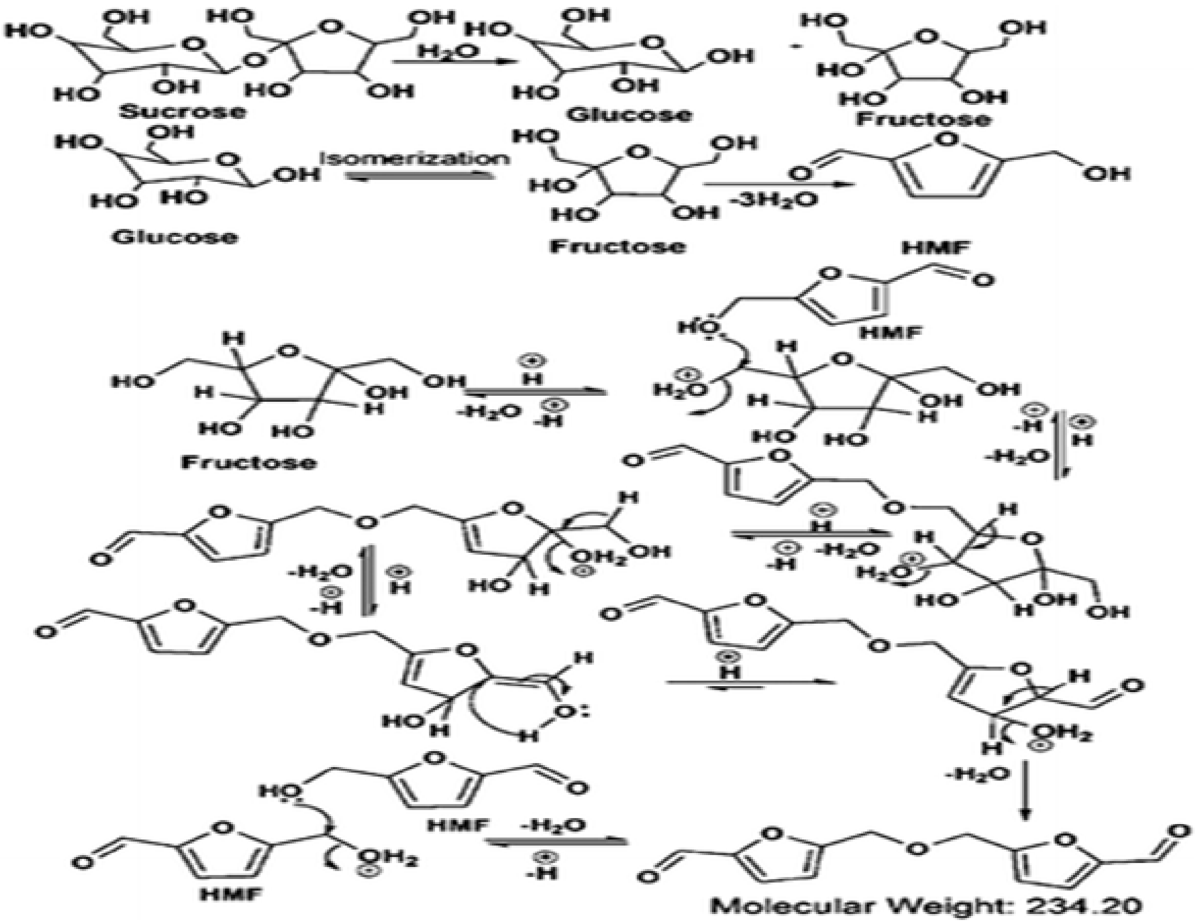



**Figure 9:** Formation of HMF derivative in CDs with the help of starting material sucrose & $H_3PO_4$

Kasprzyk et al [35] proposed that the origin of fluorescence mechanism of CDs which is obtained from urea & citric acid. The chemical structure of the fluorescent compounds is formed. The blue PL carbon dots are prepared by citrazinic acid by using hydrothermal heating and got citrazinic amide which is a source of blue fluorescence. The green fluorescence prepared from urea and citric acid without solvent using microwave irradiation and formed a fluorescent molecule of CDs which is HPPT (4-hydoxy-1H-pyrrolo[3,4-c] pyridine-1,3,6-(2H,5H)-tione i.e. shown in fig.10

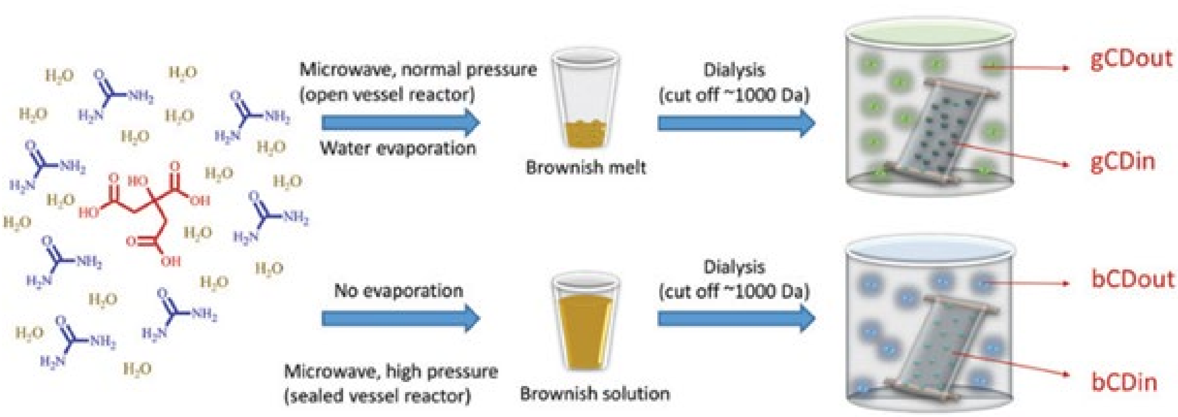



**Figure 10:** Represents the synthesis & purification of green-CDs and blue-CDs.

Meierhofer et al [36] synthesized two types of CDs by hydrothermal methods which is obtained by the reaction b/w the critic acid & 2,3-diaminopyridine (DAP) or polyethylenimine (PEI) gives the higher abundance of sp2-hybridized nitrogen based on DAP CDs, that causes a red-shift of the n-π* absorption and blue shift based on PEI CDs as shown in fig.11. The dependency of pH of the n-π* transitions help to find the both types of CDs which are fluorophores groups and functional groups.

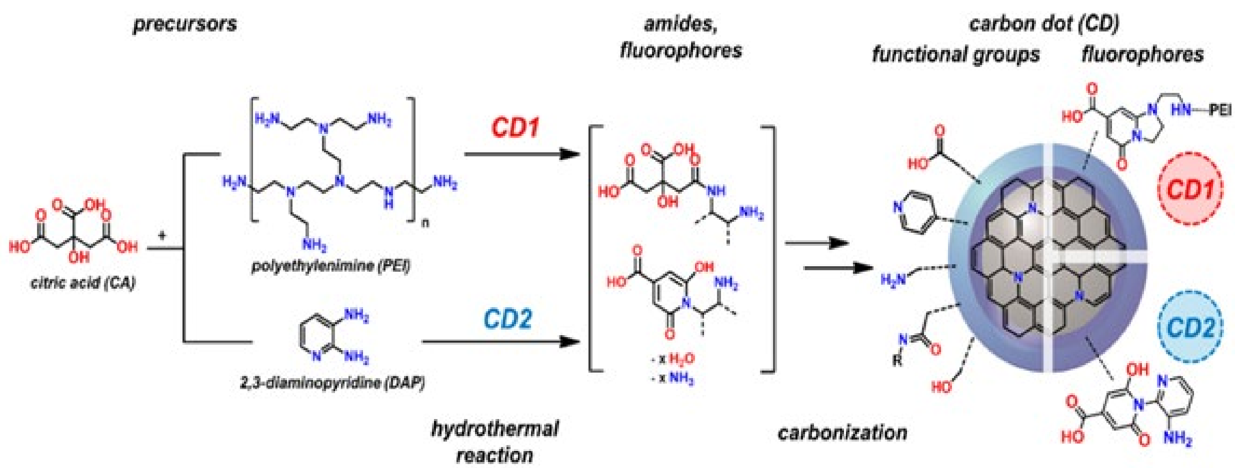



**Figure 11:** The hydrolysis of Citric Acid with PEI and DAP (amine precursors) gives CD1 & CD2 Copyright (2020) American Chemical Society

Schneider et al [14] synthesized N-CDs which is obtained from citric acid used as carbon precursor & the derivatives of citrazinic acid, gives the emission of blue spectrum. To get more knowledge about the molecular fluorescent species on their optical properties of CDs, the authors took the three samples of citric acid & three different N-containing sources: hexamethylenetetramine, ethylenediamine & triethanol- amine as shown in fig.12 and in the resulting ethylenediamine gives more QY as compared to other nitrogen sources.

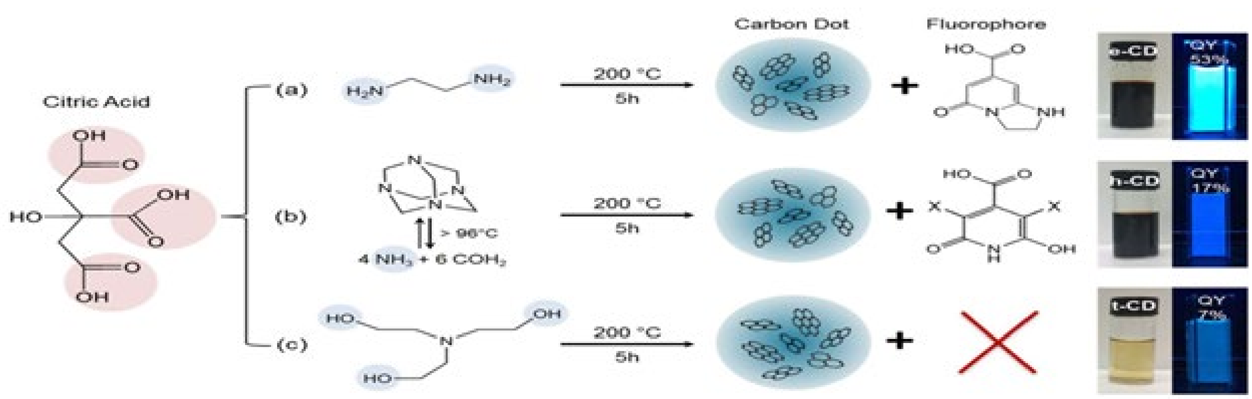



**Figure 12:** Synthesis of N-CDs by taking citric acid & three different N-containing precursors, Copyright (2017), American Chemical Society.

## Conclusion

As per in the conclusion, CDs have developed that have a broad variety of properties in all the domains, including bioimaging, in drugs, cancer therapy, medicine, and medicines. They also have a number of optical features, including PL and fluorescence. There are numerous ways to create a CD, but without a correct structure and image, it cannot be called a CDs. It is open for discussion. In this review, we describe the various techniques used to synthesise CDs and also combine all of the review paper's data in our article to make things easier to grasp. As a result, there is need to be continued improvements in the CDs that would become a widespread in their characterization, structure, functionality and their exact PL mechanism.


## Acknowledgement

I am very thankful to Chandigarh University to give me all possible help.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