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A Review based on the Synthesis of Carbon Quantum Dots: Top-Down, Bottom-up Approaches and their Properties

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Abstract:- A novel class of nano-carbon materials called as luminescent carbon quantum dots (CQDs) has received a most of attention lately, specially in the fields of chemical sensors, bio-imaging, nano-medicine, solar cells, light-emitting diode (LED), and electro catalysis. Many methods, including the arc-discharge method, microwave pyrolysis, hydro-thermal process, and electro-chemical synthesis, can be used to make CQDs quickly and affordably. CQDs have outstanding chemical and physical characteristics, including high crystallisation, good dispersibility, and photoluminescence. Particularly, the CQDs-based composite has high electric conductivity and catalytic activity due to the small size, superconductivity, and quick electron transfer of CQDs. Additionally, CQDs have a lot of surface functional groups, which could make it easier to make multi-component electrical active catalysts. As a result of interactions within these multi-component catalysts, charge transfer, a crucial electrochemical process, may be promoted, which could further improve the catalytic performance. Recent studies on CQDs have concentrated on their photocatalytic and fluorescence features. This review summarize the different approaches for the synthesis of C-QDs and the properties in the different field of science.

Keywords *:- Cqds; Nanoparticles; Oxidation; Nanotechnology; Quantum Dots.*

I. INTRODUCTION

QDs are a class of small, highly fluorescent nano-particles that are heterogeneous [1]. It is an advanced family of materials that have been used in medicinal, imaging, and sensing applications [2]. Several kinds of tiny particles are found inside quantum dots, and these particles trap energy [3]. According to the law of quantum theory such particles have good energy level. Size of quantum dots vary from one to ten nano-meters. CQDs are just one of the numerous types of QDs that are used for various applications. These are derived from several carbon containing compounds. 15th most abundant element on earth crust is carbon [4]. Sun et al. published the first study on the quantum-sized brilliant and colourful photoluminescence of CQDs in 2007 [5].Generally CQDs possess spherical shape and Zero-dimension based nanoparticles . Core of CQDs is amorphous to nanocrystalline. CQDs have prodigious physical and chemical properties i.e. strong luminescence ability, biocompatibility, high electrical and thermal conductivity, good tensile strength, low toxicity, eco-friendly, production cost is low, superior solubility, high specific surface area, easy to functionalized. They have mainly sp^2 C-hybridization but sometime possess sp^3 C-hybridization. Ideal size of CQDs is 1 to 10 nm. Due to these

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all excellent properties, CQDs achieve enormous attention from last few years. C-QDs are consists of carbon, nitrogen, oxygen and hydrogen atoms.These nanoparticles contain many functional groups on their outermost i.e. -OH, -COOH, carbonyl and NH_2 groups (as shown in figure a). The structure of CQDs contain a huge number of amino groups on their surface so that DNA can easily bind and perform their function as gene carrier.

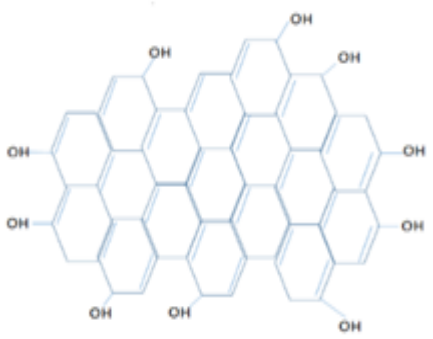

Fig. 1 . showing the structure of carbon quantum dots

These nanoparticles have many advantages in biomedical sciences. CQDs are used in bio-imaging, bio-sensors and bio-molecule, energy storage, energy conversion, in cancer therapies they used for drug delivery and also used for tumor targeting fluorescence imaging sensors. These are also used in the detection of high toxic and carcinogenic metal ions. As CQDs acquire good bio-compatibility and photo-stability properties, so it is used as excellent bio-imaging and pattering agent. CQDs are the nanoparticles which are derived from the several carbon sources. CQDs synthesize from two sources – green or agricultural source and chemical source.

In green sources only that plant or plant part is used as a carbonic precursors which contain high amount of carbohydrates. In this the main source for the production of CQDs is agricultural waste. Agricultural waste like orange peel, grape peel, lemon peel, sugar-cane bagasse, banana peel, watermelon peel, wheat husk, rice straw, corn stover, barley straw, rice barn , Pomelo peel, Willow bark, contain high amount of carbon weightage. CQDs can also be synthesized from banana or banana juice, apple juice, carrot juice, Strawberry juice, Soy milk, Garlic, Onion waste, date, grass and many plant leaves. Agricultural wastage is a excellent source for the synthesis of carbon quantum dots. Agricultural waste easy to available, less of cost. The main significance is that it is non-toxic to environment so, causing no pollution . It also act as a eco-friend by utilizing waste material of agriculture. The soil and water pollution caused by combustion of agriculture waste material can be reduced by using in synthesis of carbon quantum dots. On the other hand in chemical sources, that chemicals are used which contain high amount of carbon. There are different kinds of chemicals like Ascorbic acid, acetic acid, maltose, glucose, galactose, fructose, phenol, formaldehyde resin, silica-particle etc. which are used as a carbonic precursor. Chemical source are not easily available and cost-effective also. Different kinds of approaches are utilized for the synthesis of CQDs which includes hydro-thermal methods, chemical oxidation, microwave treatment, electro-chemical method, Laser ablation technique and ultrasonic method.

II. SYNTHESIS OF CQDS

There are many differents routes via which CQDs can be synthesized. Only route can be followed by researchers which is yield high, clean and highly efficient. CQDs can be synthesized by 2 routes – top-down route and bottom-up route [6].

A. Top-down route

Top-down approaches begin with a complicated structure. Large structures are crumbling into smaller ones along this route. The precursor has a bulk structure that is broken down into materials with nanostructures using various techniques. Graphites, nano-diamonds, carbon-soot, carbon-nanotubes are complex structure of carbon which are converted into quantum dots. Large carbonaceous materials can be broken down into nano-structures using this processes like as acidic oxidation, arc-discharge, laser-ablation, and ultrasonics.

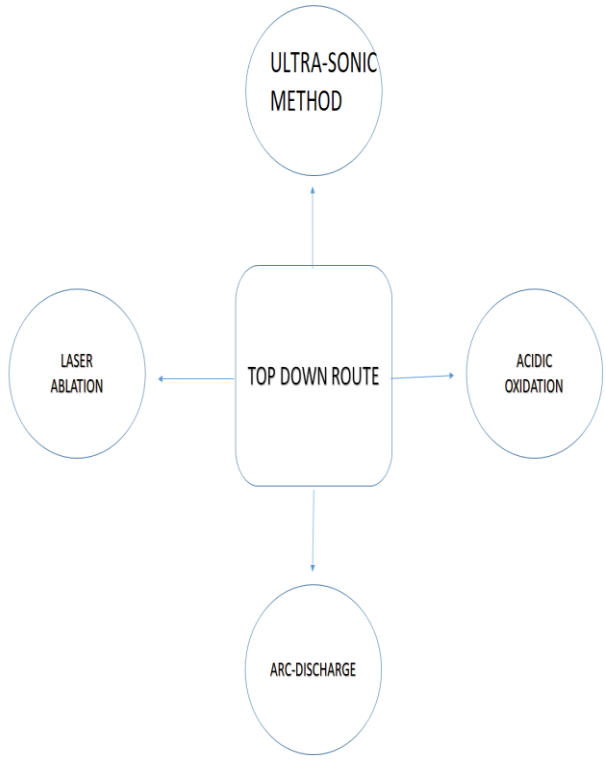

Fig. 2. Showing different top down approaches for CQDs synthesis

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a. Acidic oxidation

As complex organic structures are broken down into simpler forms by the process of acidic oxidation, various types of acids are utilised. The raw carbon used to create carbon nanoparticles was transformed into quantum dots. Different kinds of oxidizing agents are used for the oxidation of carbon precursor. Oxidizing agents such as chlorine-dioxide , hydrogen-peroxide, hydroxyl-radicals, ozone and others are used for oxidation of substances. Acids like HNO_3 , H_2SO_4 , $NaClO_3$ and toluene are also used in acidic oxidation. These substances have the ability to instantly and completely destroy saturated organic molecules. Aromatic compounds like (benzene) and trichloroethylene(TCE) are used for unsaturated organic-compounds. Acidic oxidation also convert toxic elements into non-toxic elements [7]. Acid oxidation results hydro-philic groups (e.g.-OH group or -COOH group) on the surface of obtained C-QDs [8]. These groups are benefited for CQDs to enhance the water solubility and characteristics of fluorescence . This method is extremely effective at destroying the dissolved state of non-aqueous phase liquids, which are difficult to dissolve. Acidic oxidation is cost effective, fast performing and low maintenance technology [9]. Calcium peroxide (CaO_2) nano-particles were synthesized via acidic oxidation method [10]. In this experiment hydrogen peroxide as a oxidative agent.

Yellow Fluorescence emitting CQDs were prepared from fullerene carbon-soot (FCS) by Zhang in 2017 [11]. In this, the concentrated HNO_3 and H_2SO_2 were pre-mixed by cup-sonication for 40 minutes. The volume-to-volume ratio for combining the two acids is 1:1. Following the addition of FCS, the mixture was cup-sonicated for 1 hour at 80 to 120 °C and then continuously stirred for 12 to 36 hours. Mixture was cool down and K_2CO_3 was added to neutralize the acidic mixture. The final product was obtained after the centrifugation and dialysis processes. By utilising these two concentrated acids, FCS was dispersed into quantum dots. Both of these oxidising agents separate the functional groups that are present on the surface of FCS and disrupt the conjugate structural bonding of FCS [11].

Yang reported the production of C-QDs from Chinese ink in 2014. [12] . Chinese ink was oxidized by using HNO_3, H_2SO_4 and $NaClO_3$. After preparing a mixture of all the acids, CNPs were added to it. . Stirred the above mixture at 5 °C for 1h. After this mixture was maintained at 15 °C for 5h. Cold ammonia was used to neutralise the obtained solution. These acids simultaneously chemically cut the Chinese ink, and oxidized-CQDs were produced as a precursor. Oxidized CQDs were obtained after this process . From these oxidized CQDs , heavy doped CQDs was prepared. Di-methylformamide (DMF), sodium-hydrosulfide (NaHS), and sodium-selenide (NaHSe) mixture was added in Teflon autoclave with above oxidized-CQDs. Above solution was hydro-thermally treated at 240 °C for 12h. DMF, NaHS and NaHSe act as a nitrogen, sulfur and selenium source respectively. Nitrogen-CQDs, Sulphur-CQDs and Se-CQDs was obtained with high quantum yield and longer fluorescence life-time as compared to the pure CQDs.

b. Arc-discharge

The primitive and highly effective way for creating high-quality carbon quantum dots is arc discharge. This arc-discharge technique was invented by Iijima in 1991 to synthesise and reorganise carbon atoms. [13]. The arch discharge approach for the creation of modified carbon atoms is discussed by N. Arora and N.N. Sharma in 2014 [14]. The electrical breakdown of the bulk materials is accomplished via the arc-discharge process. Arc is produced with electrical current. Arc-discharge devices have a chamber that is made up of two electrodes. The catalyst and powder form of the carbon precursor are placed within the anode electrode. A pure graphite rod typically serves as a second cathode electrode. Gas usually fills in the chamber. An arc is produced when the electrodes are powered. It is essential to keep the current flowing through the electrodes constant in order to produce a non-fluctuating arc. If the current was unstable, the arc would start to fluctuate, which would make the plasma unstable. It has an impact on the final product's quality. Arc current causes high temperature, which attracts carbon-precursor to the anode electrode. High temperatures cause the formation of carbon vapours, which gather in the gaseous phase and flow towards the cathode electrode. These collections of carbon vapour cool down since the temperature on the cathode side is typically low. Arc discharge is stopped after a shortest time period, and modified carbon is recovered from the cathode electrode.

The synthesis of carbon nano-structures utilizing the arc-discharge method is discussed by Yatom in 2017 [15]. This approach produced a vast population of nano-particles with a diameter of 20 nm. Single-walled C-nanotubes were made via the arc-discharge method in 2004 by Xu et al. [16]. Three separate carbon nano-particles with distinct fluorescence characteristics and varying relative molecular masses were discovered throughout the experiment.

c. Laser-ablation

By intensely applying a laser beam, a thermal process called laser-ablation removes atoms from solid materials [17]. This approach is highly helpful in the production of nanoparticles because it allows for the break-down of the bulk structure of carbon into tiny particles. The carbon precursor gets heated when a laser is introduced to it because it absorbed the laser energy. This method was utilised by Kuzmin P. in 2010 to create silicon nanoparticles. Kuzmin concluded that as the laser pulse's duration is decreased, the size dispersion of the nanoparticles also reduces.

Hu et al. carried out a study in 2008 to create C-quantum dots through laser ablation [18]. Hu created a one-step procedure for the laser-induced irradiation of carbon powder in organic solvents to produce fluorescent carbon quantum dots. A suspension of carbon powders in an organic solvent was laser-irradiated to create fluorescent carbon nanoparticles

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(CNPs). The CNPs' surface modification was completed concurrently with their creation, and by choosing the right solvents, adjustable light emission could be produced. Carboxylate ligands on the surface of the CNP were thought to be the cause of the luminescence. The CNPs' surface change happened simultaneously with their creation, and by choosing the right solvents, luminescent surface states could be produced.

Xiangyou Li presented this approach to manufacturing the carbon quantum dots with high luminescence properties in 2010 [19]. The performance of the synthesised CQDs was determined to be visible, stable, and controllable. In a typical technique, 50 ml of a solvent (like as ethanol, acetone, or H_2O) was used to disperse 0.02 g of nano-C material. 4 ml of the suspension was added to a cell of glass for laser-irradiation after ultrasonication. The cell has a crystal window over it. Without focusing, the suspension was exposed to a Nd:YAG pulsed laser with a 2nd harmonic wave-length of 532 nm. The raw carbon nanoparticles were kept from gravitationally settling during irradiation by using a magnetic stirrer. Following laser irradiation, the solution was centrifuged for 30 minutes at 6000 rpm to extract the supernatant, which was then used to test for PL data using a spectrofluorophotometer. Obtained supernatant contain the carbon quantum dots. By irradiating carbon nanoparticles with a laser in straightforward, common chemical solvents, C-QDs with visible, stable, and controllable PL performance were created. The unfocused laser beam in this process makes it simple, easy, and highly efficient than existing techniques for creating carbon QDs. The morphology was observed by HR-TEM, and the results of an XPS investigation showed that passivation by laser irradiation was crucial in the development of PL. This technique can be applied to many different fields, including biology and imaging, and has a significant potential for creating new luminous materials.

Cui Liu used the laser-ablation approach to create the silica nanoparticles in 2015 [20]. The silicon and carbon dots began forming from the silica nano-particles as the temperature and pressure increased as a result of the immediate laser irradiation. Liu was able to determine that a fast and efficient way to create carbon nanoparticles would be through laser ablation. Strong lasers' instantaneous induction of high temperatures and pressures resulted in the reduction of some silica and the pyrolysis of C-chains inside of silica nano-particles, whereupon the rapid crystallisation of silicon and carbon vapour produced si-dots and diamond-like C-dots in the silica matrix.

Donate developed C-QDs for fluorescence imaging in 2018 using the laser irradiation method [21]. It was found that the CQDs yield is extremely high and also has a compact size following a 4 hour exposure to laser irradiation. High Fluorescent Semiconductor Quantum Dots can be produced using a laser. This type of carbon dots have a very high photoluminescence [5]. With the system's simplicity, the carbon microparticles more effectively capture laser energy, increasing the fabrication efficiency by 15% and the fluorescence of C-QDs by an order of magnitude compared to the traditional batch method. The flow jet-synthesized C-QDs are employed for fluorescence imaging of transparent healthy and malignant epithelial human cells. Their typical size is 3 nm. Without the use of any additional additives or processing of the cell culture, complete internalisation is seen after a brief incubation period of 10 min. According to a study of the photo-luminescence, C-QDs displayed an emission behaviour that was excitation-dependent, with greater emission wavelengths for high excitation wave-lengths. The strongest fluorescence emission intensity is related to excitation wavelengths about 287 nm. The presence of C-O, C-OH, C-O-C, and C-O-OH groups on the outerward has been studied using XPS and FTIR, and these groups offer a lot of potential for biological applications in conjugating drugs or targeting moieties.

d. Ultra-sonic method

Ultrasound is a cutting-edge method that allows for the functionalization, activation, and excellent dispersion of nanoparticles during sono-chemical synthesis [22]. Ultrasonication is a critical step in the process of nanoparticles. This technique uses an ultrasonic sound wave frequency (>20kHz) to apply homogenous dispersion of nanoparticles to the solution. These sound waves disrupt the intermolecular force between the nanoparticles, which reduces their tendency to collect or take aggregate forms. The direct ultra-sonic approach works well for creating nanoparticles.

By using an ultrasonic process with glucose as a carbon precursor, Haitao Li produced carbon nanoparticles in 2010 [23]. Ultra small sized carbon nano-particles were produced using this one step process. These particles have extremely appealing photoluminescent characteristics. A one-step alkali or acid aided ultrasonic process was used to directly synthesise mono-dispersed water-soluble fluorescent C-nanoparticles (C-NPs) from glucose. TE-microscopy, optical fluorescent microscopy, fluorescent spectrophotometry, FT-IR , and UV-spectrophotometry were used to characterise the CNPs. The suitable quantity of glucose was dissolved in 50 mL of deionized water to generate a transparent solution. The glucose solution was mixed with either a 50 ml NaOH solution or a 50 ml HCl solution, and the combined solution was subjected to an ultrasound treatment for 4 hours. The unprocessed glucose/HCl CNP solution was dried in oven at 80 ^oC for 6 hours. The pH of the another raw samples made from glucose/NaOH was first brought down to 7 with HCl, and then 100 mL of ethanol was gradually added while agitating the mixture. After that, to remove the salts and water from the CNPs solution, a suitable amount of $MgSO_4$ was added, stirred for 20 minutes, and then stored for 24 hours. The findings demonstrated that the particle surfaces were very hydrophilic due to their abundance in hydroxyl groups. The full visible-to-near infrared (NIR) spectral range could be covered by the photoluminescence that the CNPs were capable

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of emitting. Notably, NIR excitation might be used to produce the CNPs' NIR emission. Additionally, the up-conversion fluorescence characteristics of these CNPs were good.

B. Bottom-up route

Bottom up is the route in which carbon quantum dots are synthesized from molecular precursor. This method use the small molecules as precursor to obtained the carbon-quantum dots through chemical reaction. Bottom up route includes hydrothermal method, combustion, electrochemical and Microwave synthesis.

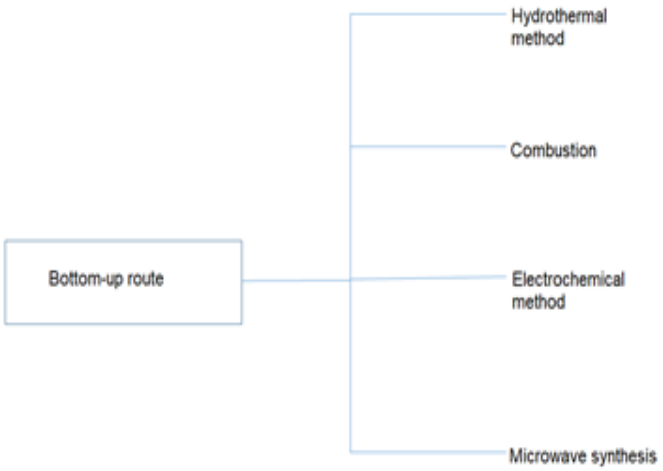

Fig. 3. Showing different bottom-up approaches for CQDs synthesis

a. Hydrothermal method

The hydrothermal method uses an aqueous solution as a reaction operator in a completely sealed vessel. High pressure and temperature are required for this process. The vessel must be set up in a system where high temperatures will be used. Vapours generated pressure on their own. The hydrothermal reaction occurs above 100 ^{0}C [24]. Schafhasutl published the first account of a hydrothermal-based reaction outcome in 1845. The solvent in a hydrothermal reaction is water. In 1978, Franck reported on the physical and chemical characters of water during the hydrothermal process. The hydrothermal process is crucial in the production of CQDs nanoparticles.Teflon-lined hydrothermal autoclave had been used throughout this process. The material of teflon is stainless steel. Lemon juice has been used as the starting material for the hydrothermal synthesis of C-QDs [25]. Lemon juice was used to make lemon extract. A Teflon autoclave made of stainless steel was filled with 40 ml of lemon essence. kept this Teflon reactor at a high temperature between 120 and 280 ^{0}C for 12 hours. The system is subsequently cooled down to RT. The white solution's transformation into a dark one reveals the production of carbon quantum dots. Use filter paper to filter the above-mentioned solution. Under ultraviolet light, a mild blue-green color appears. Images of the HRTEM results reveal carbon quantum dot dark spots. The particle size varies between 3 and 5 nm at 240 ^{0}C. The presence of carboxylic and -OH groups on the surface of carbon quantum dots is indicated by FT-IR peak patterns. According to DLS data, the particle size is 50 nm, and the UV vis absorption peak is 283 nm. Carbon quantum dots have a 21.37 percent quantum yield. Using a hydrothermal process, CQDs were produced from tulsi leaves in 2018 [26]. The fresh tulsi leaves should be cleaned with distilled water before usage. 2.5 g of tulsi leaves were chopped up into tiny bits. 35 ml of distilled water was added to it, and the above-mentioned solution was transferred there. maintained 200 ^{0}C in the hydrothermal unit for 4 hours. The solution is then cooled to RT and passed via membrane filter. The obtained yellow fine particles were kept at 4 ^{0}C for storage. Particles of a 5 nm size display spherical morphology in the TEM results. Strong peaks of the amino, carboxylic, carbonylic, and hydroxyl groups can be detected under FT-IR. Fluorescence that is bluish is visible under Ulta-Violet light. The quantum yield was 3.06%.

Green synthesis of C-QDs from grass was performed through hydro-thermal method [27]. The grass was cleaned and allowed to air dry. The grass was crushed, and 100 ml of water was then added. Put this solution in a hydrothermal reactor and ran it for two hours at 180 ^{0}C. Once this powder had been obtained, it had been cleaned with milli.Q. water. The above powder was dried at 80 °C for 24 hours. Some particles smaller than 10 nm in size can be seen in SEM pictures. The particles' uniform surface is shown by AFM. The surface of the particles has OH, C-H, C=O, and COOH groups, according to the FT-IR results. Corn stalk was also used for the production of C-QDs [28]. Water was used to wash the corn stalk in order to remove the dust. The stalks are broken into little pieces after air drying. 500 ml of deionized water was combined with 20 g of this. Put the solution mentioned above in a hydrothermal unit and set the temperature at 270 ^{0}C for 10 minutes. After cooling, a membrane microporous filter was used to filter the resulting solution, and 24 hours of dialysis were then performed. Applying freeze drying, solid carbon quantum dot particles were produced. Under UV light, blue fluorescence was observed. FT-IR results confirmed the presence of OH, C-N, C=O, C-O, and C-H groups. XRD broad peak at 22.50. 16% quantum yield was obtained. Additionally, the leaves of water hyacinth were utilised as a carbon precursor in the development of carbon quantum dots [29]. The leaves were first cleaned in water and then allowed to air dry. Powder was produced after the grinding. A teflon reactor was filled with a mixture of 5 g of powder and ultrapure water. for 12 hours, this reactor was kept at 180 ^{0}C. The solution should be cooled at room temperature. The above-mentioned solution was centrifuged at 8000 rpm for 20 min. After the supernatant was filtered, 2 hours of dialysis against purified water was conducted. Brown solution was produced, demonstrating how carbon quantum dots are formed. Storage begin at 4 ^{0}C. Based on HR-TEM data, particles have an average diameter of 2.44 nm and a range in size from 1.2 to 4.2 nm. On the carbon quantum dot surface, C-H, C=O, C=C, O-H, C-N, and C-O

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groups can be seen using FT-IR. 3.3% quantum yield was obtained.

For the hydothermal method of producing C-QDs, Azadirachta indica (Neem) leaves were utilized [30]. Fresh leaves were cleaned in distilled water and allowed to air dry. 10 g of freshly ground leaves were combined with 10 ml of distilled water. Place the above-mentioned solution in a Teflon reactor at 150 ^{0}C for 4 hours. Centrifuging the obtained brown solution at 15000 rpm for 10 min. Supernatant was collected and dialyzed for a further 24 hours in distilled water. Carbon quantum dots were created as a clear brown solution that was kept at 4 ^{0}C. TEM results revealed that the size of the particles ranges from 1-5.5 nm, and a yield of 27.2% was attained. Maple tree leaves was also used as a carbon precursor [31]. The maple leaves were first cleaned in water and then allowed to air dry. Teflon reactor with 50 ml of extract had been operating at 190 ^{0}C for 8 hours. The obtained brown solution was then centrifuged one more for 20 minutes at 15,000 rpm. The supernatant was gathered and purified. Particles range in size from 2-10 nm, according to TEM. Results from AFM demonstrate homogeneously dispersion. C-H, C=O, C=C, O-H, and C-O groups can be seen on the carbon quantum dot surface thanks to FT-IR. Particles have a yellow color when exposed to UV radiation.

Although banana peel is a waste product, it can be utilised as a carbon precursor for generating carbon quantum dots [32]. Peel from a banana was cleaned and dried. They were chopped into tiny pieces and combined with milli Q water. Put the above-mentioned solution in a Teflon reactor at 200 °C for 24 hours. The result was a brownish-yellow tint that was further filtered and kept at 4 ^{0}C. According to TEM/HRTEM measurements, particle size ranges from 4-6 nm. A 5 nm average size diameter and a 20% quantum yield were used.

Comparison between the different sources of C-QDs obtained from hydrothermal method at different temperature (TABLE I.)

Sr. no.	Carbon precursor	Temperature	Particle size	References
1	Lemon extract	240 ^{0}C	50 nm	[25]
2	Tulsi leaves	200 ^{0}C	5 nm	[26]
3	Grass	180 ^{0}C	0 – 10 nm	[27]
4	Corn stalk	270 ^{0}C	1 – 3 nm	[28]
5	Water hyacinth leaves	180 ^{0}C	1 – 4 nm	[29]
6	Neem leaves	150 ^{0}C	1 – 5.5 nm	[30]
7	Maple tree leaves	190 ^{0}C	2 – 10 nm	[31]
8	Banana peel	200 ^{0}C	4 – 6 nm	[32]
9	Apple juice	180 ^{0}C	1 – 3 nm	[33]
10	Pomelo peel	200 ^{0}C	2 – 4 nm	[34]

b. Combustion

Combustion [35] is the process which occur at high temperature. This is an extremely efficient, inexpensive approach for producing many kinds of nanoparticles [36]. The major reaction in the conventional approach, which happens at a high temperature of more than 2000k, is the breakdown of the covalent connection between the carbon precursor and the creation of quantum dots [37]. Combustion synthesis is of two types;1) Solid state combustion 2) Solution combustion. All initial reactant, intermediates, and end products in solid state combustion are solid [38]. On the other hand solution combustion have liquid components [38]. A variety of nano-sized particle types are generated via solution combustion. The manufacture of oxide particles and carbon quantum dots is currently carried out using this technology all over the globe.

In 2018, an experiment was performed by S.Zhang [39]. In this experiment C-dots was prepared through combustion method. Ash was prepared by burning the organic solvent at high temperature in air. Different kinds of flammable solvents like ethanol , n-butanol, domestic candle and benzene are used for synthesis of carbon dots-1, carbon dots-2, carbon dots-3 and carbon dots-4 respectively. 2g of soot or ash mixed with solvent on stirrer for 30 mintues. After stirring, solution was sonicated for 2h. Centrifugation was performed at 5000 rpm for 3 minutes . Light brown supernatant was collected after centrifugation. It was concluded that though CDs prepared via different framework but possess similar photo-luminescence and u.v. via absorption. After characterization it was observed that methyl and carbonyl groups present on CDs and have 2-4 nm in size. FTIR result revealed that C=C bonds and oxygen containing groups present in CDs.

L.A. Diaz-Torres in 2018 [40], works on the synthesis of luminescent, structural and morphological properties of C-dots via combustion method. In this experiment author describe the devlopment of ZnO and C-QDs. To change the concentration of CDs samples were annealed at various temperature i.e. 600, 700, 800 and 900 ^{0}C. $Zn(NO_3)^3$, $Al(NO_3)_3$ and citric acid was used as carbon precursor. All are dissolved in a beaker . A solution of deionized water and ethanol in a ratio of 3:1 was added in same beaker followed by continuous stirring for 2 h. A small amount of ethylene glycol was added. After 30 mintues, NaOH was added to the solution and heated at 80 ^{0}C until high viscous solution was obtained. Cool down the solution at room temperature and after this placed the obtained solution in furnace at high temperature (400 ^{0}C). 15 mintues later combustion reaction ended and blackish fluffy powder form was obtained . The obtained powder grinded and burn in air at 600, 700, 800 and 900 ^{0}C.Timing for annealing process

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was 10 ^{0}C/minute. Once the temperature reached at final state then kept it for 8 h. Then furnace was turned off and sample was cooled down. Finally it was concluded that carbon dots was obtained from this method have good fluorescence and optical property. Carbon dots annealed at 600 and 700 ^{0}C show green and yellow emission respectively after excitation at 405 nm. Same annealed carbon dots when excited at 455 nm then they show orange (annealed at 600 ^{0}C) and green (annealed at 700 ^{0}C) emission respectively. Chemical bonding was characterized by the XPS method. SEM images show that C-dots annealed at various temperature show round shape and very few one show oval like shape. Average size of the smallest particles was around 0.5 to 9 nm.

C-QDs was developed by Himani in 2019 by the using of sugarcane baggase [41]. Sugarcane baggase used as carbon precursor. Sugarcane baggase was dried for 6 days in sunlight and then cut it into small pieces. Burn the sugar-cane baggase at 60 ^{0}C for 4 h. 500 mg ash of sugarcane baggase was dissolved in 50 ml toluene. Stirred the above solution for 1st day for completly dispersion. After this sonicate the above solution for 1 hour and then centrifuge it at 8000 rpm for 30 min. After the filteration, supernatant was collected. FT-IR revealed that carboxyl, -OH, methyl and epoxy groups are present on the surface of C-QDs. Under the SEM images spherical dots was observed. TEM result images show that 86% particles have 2-9 nm diameter, 10 % have greater then 9 nm diameter and few have 2 nm diameter. Average size of the particles in between 3 to 5 nm. Under the U.V., blue emission of carbon quantum dots was obtained .

So overall conclusion was that combustion is also a versatile , simpular and rapid process for the synthesis of C-nanoparticles or carbon-quantum dots. It is less of cost and good method for synthesis of various kinds of nano-particles. The advantages of this method is purity of the product, low energy requirement, relative simplicity and rapid process.

c. Electrochemical method

Electrochemical method is very effective method for the synthesis of C-QDs. In this method the chemical reaction for the synthesis of nano-particles occurs at very normal temperature and low pressure. Carbon quantum dots particle size and photo-luminescence property was reported by Deng[42], Ahirwar [43] and Anwar [44]. Electrochemical synthesis of CQDs was carried out in 2015 by Yuxin Hou [45]. In an appropriate proportion sodium citrate and urea was added in ultrapure 10 ml water. Mixed the solution properly. For positive and negative electrode 2 platinum sheets was employed about at 1 cm distance. The electrochemical reaction occur at 5V potential for 1 h. Transparent solution turned into brown colour. Dialysis was performed against ultra-pure water for 6 h. Fluorescent Carbon quantum dots was obtained. It was observed that average size of carbon QDs is 2.4 nm. Carboxyl and hydroxyl groups are observed under FT-IR.

In alkaline conditions, photoluminescent C-nanodots have been made from glycine using an electrochemical method [46]. Electrooxidation, electropolymerization, carbonization, and passivation are the processes that lead to the creation of C-dots. The photoluminescence (PL) features of the as-prepared Carbon-dots are excitation-wavelength-dependent and pH sensitive, and they are stable in solutions with high salt concentrations. The use of Carbon-dots with the fluorescence resonance energy transfer (FRET) method to detect haemoglobin has been proven. With a limit of detection (SNR3) of 30 pM, the PL intensity of Carbon-dots is inversely proportional to haemoglobin concentration over the range of 0.05-250 nM (r=0.99). By measuring the haemoglobin concentrations in 5 representative diluted blood samples, we have validated our test. The results are in good agreement with those obtained by using a commercial haemoglobin metre. Blood-stained pictures and fingerprints have been obtained using the water-dispersible and photo-stable Carbon-dots.

Researchers created C dots with a significant Stokes shift using o-phenylenediamine (OPD) as the carbon source in an electrochemical technique for sensing Fe3+ and ascorbic acid (AA) [47]. At the platinum anode during the electrolysis, OPD experiences polymerization, carbonization, and passivation to create C dots. Transmission electron microscopy, Raman spectroscopy, X-ray photoelectron spectroscopy (XPS), and FT-IR spectroscopy were used to characterise these C dots. They are made up of the atoms C, N, O, and H, and have a Stokes shift of 309 nm. Their greatest emission occurs at 570 nm while their maximum excitation occurs at 261 nm.

d. Microwave synthesis

Microwave synthesis open the best approach for the development of small sized nano-particles [48]. Nowdays the use of the microwave equipment is strongly recommended. Microwave synthesis is the very fast technique for the development of nano-particles. The first use of microwave technique was reported in 1986 [49,50]. Microwave radiation provides a efficient and controlled heating, which is very beneficial for the synthesis of nano-particles and nano-structures . Heating that was provided by a microwave is reduced the time of chemical reaction. This one is very big advantage of the microwave synthesis. It improves the yield and repro-ducibility of the nano-particles. Chemistry of the microwave based on the desired heat of matter produced by micro-wave di-electric heating or on the absorption of microwave energy by the specific material (solvent or any reagent) and convert it into heat [51,52]. In the microwave, the frequency of electromagnetic radiation ranges in between 0.3 to 300 GHz, having wavelength of 1 mm to 1 m. In this frequency region, the microwave energy converts into thermal energy [53]. 1.0×10^{-5} ev is at the frequency 2.45 GHz able to cleave the chemical bonding between the particles of carbon precursor [54]. Heating mechanism in microwave involves two main process - Di-polar polarization and Ionic conduction [51]. The advantages of microwave irradiation is that it produces efficient heating (internal) and also increases

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the temperature of whole volume uniformly and simultaneously.

In 2015, carbon quantum dots was synthesized by Naralia R. Pires from raw cashew gum [55]. Solution was made from the raw gum and filtered. The above solution was heated in microwave for 30 – 40 minute. on rotating plate. Due to heating effect the carbonization reaction occur of the particles and a light brown solid was obtained. Cool down the solution and dissolved it in water. Centrifuge the above solution at 25,000 rpm for 15 min. for removal of the less luminescent particles. Pale brown solid was obtained which contains C- dots composite. Normally the C-dots are light yellow brownish in colour and under the U.V. light blue emission was observed. The result of EFTEM show images with spherical dots having 3 to 9 nm size. FT-IR result show the –OH, C-C, C-O-C, C-O groups stretching on the carbon quantum dots surface.

CQDs was developed by Adams in 2017 via microwave method from starch [56]. Firstly fresh starch was obtained from the cassava. 1g of cassava starch powder was added in 25ml deionized water. In the above solution 7ml (2M) sulphuric acid was added in above solution. Kept the beaker in microwave for 3-7 min. Carbonization reaction takes place . Solution colour was changed from white to light brown colour and finally dark brown colour was obtained . For clear solution filtered the solution with filter paper. FT-IR result revealed that hydroxyl, carboxyl groups are present on the surface of obtained carbon quantum dots. Under the U.V. light , blue green or yellow light luminescence shown.

Hasan Eskalen in 2019, prepared the C-QDs from the wastage of linter [57]. Linter waste is used as a carbon precursor. Closed microwave system was used for 5 min. Linter waste was taken with water in a quatz-tube and this tube was placed in a Teflon reactor . After then Teflon was placed in the microwave. Obtained product was filtered with the whatman filter paper . Centrifuge the above solution over 15000 rpm. pH of the solution was set to 7.03. Characterization of the above nano-particles for the study of morphology and optical properties was performed. TEM result show that particles are spherical in shape, size ranges from 1.8 nm to 22 nm and 10.14 nm is the average particle size diameter. FT-IR result show that carboxylic, hydroxylic groups are present on the surface of carbon-QDs. U.V. vis absorption and photo-luminescence spectra was investigated to evaluate optically properties of carbon-QDs .

Neetu in 2021, used the microwave for the synthesis of carbon-QDs from Calotropis gigantean [58]. Fresh leaves of Calotropis gigantean was collected and cleaned by the distilled water for the removal of dirt. Air dried the leaves. 10 gm extract of the leaves was mixed with 10 ml of distilled water. Filter the solution with whatmann paper before the microwave treatment. After filteration , placed the solution in the microwave until brown fluid was obtained . Centrifuge the obtained solution for 15 min. at 15,000 rpm. After the filteration of the above solution water suspended carbon quantum dots was obtained. Store the nano-particles at 4 ^{0}C for further use. 4.24% average yield of carbon-QDs was obtained. TEM-images show that the carbon-QDs are spherical in shape with 5.7 nm average size and size lies between the range of 2.7 to 10.4 nm. Zeta-potential value was around -13.8 mV. According to FT-IR OH, C-H, C=O, C-O, COOH groups are present on the surface of carbon quantum dots.

In TABLE II. different kinds of methods are compared which show the comparison between quantum yield that was obtained.

TABLE II.

Sr. No.	Carbon Precursor	Method of Synthesis	Yield obtained	Refrences
1	Fullerene carbon soot (FCS)	Acidic oxidation	3-5 %	[11]
2	Chinese ink	Acidic oxidation	60-80%	[12]
3	Graphite	Arc discharge	_	[15]
4	Crude soot	Arc discharge	16%	[16]
5	Soyabean	Ultrasonic	16.7%	[59]
6	Glucose	Ultrasonic	7%	[23]
7	Graphite	Laser ablation	3-8%	[18]
8	Nano carbon material	Laser ablation	_	[19]
9	Graphite powder	Laser ablation	4-10%	[5]
10	Carbon glassy	Laser ablation	15%	[21]
11	Lemon extract	Hydrothermal method	21.37%	[25]
12	Tulsi leaves	Hydrothermal method	3.06%	[26]
13	Grass	Hydrothermal method	_	[27]
14	Corn stalk	Hydrothermal method	16%	[28]
15	Water hyacinth leaves	Hydrothermal method	3.3%	[29]
16	Neem leaves	Hydrothermal method	27.2%	[30]
17	Maple tree leaves	Hydrothermal method	_	[31]
18	Banana peel	Hydrothermal method	20%	[32]

19	Apple juice	Hydrothermal method	_	[33]
20	Pomelo peel	Hydrothermal method	6.9%	[34]
21	Raw cashew ghum	Microwave synthesis	17%	[55]
22	Starch	Microwave synthesis	_	[56]
23	Waste of linter	Microwave synthesis	_	[57]
24	Calotropis gigantean	Microwave synthesis	4.24%	[58]
25	Sugarcane baggase	Chemical oxidation	25.7%	[41]

III. APPLICATIONS OF CARBON QUANTUM DOTS

A. Biosensing

Biosensing is the process of detection of targeting molecule which is based on the principle of an immune system. In medical biosensing field carbon quantum dots play a very important role. Carbon quantyum dots role in bio-sensing is based on the anti-bodies and gene-recombinant fragments. Carbon-QDs are used as fluorescent based labelled molecules for the detection of a specific target [60]. From the comparison it was concluded that carbon quantum dots show high sensitivity as labelling agent then gold nanoparticles [61]. Carbon quantum dots have another advantages for aptamers which are selected a target point in a large nucleic acid [62]. Fluorescent Nitrogen-carbon quantum dots was synthesized from the glutamic acid via pyrolysis method for the detection of amoxicillin [63].

B. Bioimaging

The mostly important advantage of carbon-QDs is that these are non-toxic and eco-friendly. These character make them very useful in the bioimaging for the visualization of biological systems for both In vitro and In vivo [64]. Passivating agents are present on the outer surface of carbon-QDs [65] which make them non-toxic. It was reported that CQDs are uptake by cells with the help of endo-cytosis process.

C. Photocatalytic activity

CQDs have long wave-length light and high energy converter with solutions these properties make them outstanding for the use as photo-catalytic in organic synthesis. Due to good catalytic activity for H_2O_2 decomposition and electron transfer property small sized CQDs (1-4 nm) are used as NIR light driven photo-catalysts for the oxidation of alcohols into benza-aldehyde [66]. The conversion efficiency is 92 % and selectivity 100 % . CQDs can also be used as potential photo-senitizer in solar cell system.

D. Electrocatalytic properties

From last years, CQDs attain lots of interest in the area of energy conversion and storage to face the difficult environmental issues. By doping with heteroatoms , nano-composite will be prepared which are promoting the transfer of electrons . CQDs surface rich with the carboxylic , amino and hydroxyl groups due to which it is stable in water so provide good photo-chemical reaction including oxygen reduction reaction[67,68], hydrogen evolution reduction[69,70] and CO_2 reduction reaction [71,72].

IV. CONCLUSION

The development of increasingly sophisticated applications for the luminescent CQDs in the areas of chemical-sensors, bio-imaging, nano-medicine, drug-delivery, and electro-catalysis makes them an intriguing newcomer to the field of nanomaterials. This work introduces the primary synthesis techniques like top down and bottom up and photochemical characteristics of CQDs, and on the basis of that, it makes a strong case for its use in electrocatalysis. The many synthesis methods that have already been created to create Carbon-QDs with various structures and characteristics are discussed. The absorbance and PL characteristics of Carbon-QDs are unique and fascinating, and they are now a popular research area. CQD use in the electrocatalytic field is still in its infancy, though. There is an urgent need for more work on the inventive design and manufacture of Carbon-QD-based electrocatalyst. Due to its easily controllability of composition and structure through precursor optimisation, the hydro-thermal technique is a promising contender for the synthesis of Carbon-QDs used as electrocatalyst. Additionally, electrochemical synthesis of Carbon-QDs is a promising approach that can provide C-QDs with uniform particle size and, more crucially, allows C-QDs to work in tandem with other conventional electro-catalysts to manufacture products using green chemistry. C-QDs have high surface area, excellent conductivity, and quick charge transfer, which gives them a lot of potential for use in electro-catalysis. The size, shape, surface functional groups, and heter-oatom doping of C-QDs can be changed to alter their typical electrical and chemical structures. Lots of organic groups make it possible for H_2O molecules to be easily adsorbed and offer active co-ordinating sites for metal ions to create C-QDs hybridised catalyst. In addition to being crucial in shaping the electronic structures of the nearby carbon atoms in C-QDs, the hetero-atoms (i.e., Nitrogen, Sulphur, and Phosphorous) doped therein also serve as reactive catalytic sites throughout the electrocatalytic process. Additionally, C-QDs could shield the metal sites from the solution's toxicity and oxidation.

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