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Graphene Quantum Dots: Sustainable and Greener Synthetic Approaches

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Abstract

Honeycomb crystal lattice graphene has proved to be a subject of a great deal of investigation over the previous decades owing to its intriguing electrical, optical and physical properties. Graphene identification imparts a significant lift and vital dimensions to nanotechnology and materials science. Graphene quantum dots (GQDs), a nanomaterial with zero dimensions (0D) demonstrated encouraging potential in biomedicine because of their really tiny and harmless nature, water solubility, outstanding biological compatibility, adjustable fluorescence, and photo resilience thereby gaining a great deal of interest in the biomedical sector. GQDs wide bandgap and their flexibility to engineer its bandgap over an extensive range and carrier selectivity play a major role in catalysis and energy applications. There are wide range possibilities for new applications due to their unique structure – related features such as optical, electrical, physiochemical qualities and exceptional biocompatibility when compared to other nanomaterials. They have gained tremendous attention along with other graphene derivatives in the last few years. Therefore, the development of GQDs became a remarkable catalyst for the utilization of graphene. Top down as well as bottom-up methodologies are applied for production of GQDs. Sustainable synthesis of GQDs has a variety of advantages, including the least cost and non-corrosive material, faster reactions, environmentally conscious resources, and simpler post-processing procedures than with synthetic methods. In this review, we have compiled a few of the green methods utilized by various research groups worldwide, for the green synthesis of GQDs and some of its biomedical applications.

Keywords

Graphene quantum dots, graphene, green methods, biomedical applications.

1. Introduction

Numerous studies on carbon and its derivatives have been conducted over the previous several decades. Among them grphene has played a vital role. Due to delocalized electrons, graphene, which is essentially a single, densely packed sheet of carbon atoms inside a honeycomb-shaped lattice, exhibits numerous unusual features. Although it has many exceptional qualities, such as high conductivity and superior mechanical strength, it still has several challenges before it can be used in industry. The development of graphene quantum dots (GQD's) and their discovery have greatly aided in overcoming the obstacles.[1]

Graphene has an unlimited Bohar radius of excitons [2]. By transforming into two-dimensional graphene, GQD becomes a zero-dimensional material. These nanomaterials differ from graphene in a number of important ways, including their hydrophilic nature, minimal cytotoxicity, good biocompatibility, and superior photoluminescent (PL) capabilities [3-6]. As a result, GQDs have a wide range of exciting potential uses in drug administration, bioimaging, solar cells, biosensors, and electronic devices [7-10]. Due of GQDs' exceptional qualities and possible uses, a great deal of study has been done on their synthesis, and numerous techniques have been put forth [11-14]. Because of their exceptional biocompatibility, GQDs can safely interact with biological systems. This is an essential characteristic for any substance looking to be used in medical applications.

Targeted drug delivery and particular interactions with cellular structures are made possible by surface functionalization of GQDs with different biomolecules. The various approaches used for GQD synthesis, their potential functionalization, the synthesis of their nanocomposites, and the uses of GQD-derived nanoparticles in various biomedical domains, such as bioimaging, gene delivery, tissue engineering, antimicrobial and biosensors, have all been covered in this review [15,16].

GQD's are synthesized utilizing top-down and bottom-up methodologies. They can also be produced by reducing the size of carbon predecessor such as graphene [17], graphene oxide (GO) [18], and carbon fiber [19], by hydrothermal treatment [20,21] under severe conditions, which calls for the use of powerful oxidizers like sulphuric acid (H_2SO_4) or nitric acid (HNO_3) [22]. Some hurdles of these routes include possible safety hazards, degradation of the environment, increased expenses, and difficult fabrication and post-processing steps. Therefore, it is imperative in this field to identify creative greener antecedents as well as environmentally and eco-friendly ways.[23]

In table 1 and table 2 below, we have summarized few remarkable works performed by various research groups using variety of naturally occurring, easily available and sustainable sources for the green synthesis of GQD's as well as CQD's.

Table 1. Source, Synthetic Approach and Size of Carbon Quantum Dots

Sources	Synthetic Approach	Size	References
Apple Juice	Hydrothermal	4.5 nm	[24]
Carrot	Hydrothermal	2.3 nm	[25]
Mango Leaves	Microwave	2-8 nm	[26]
Sugarcane Molasses	Hydrothermal	1.9 nm	[27]
Strawberry	Hydrothermal	5.2 nm	[28]
Winter melon	Hydrothermal	4.5-5.2 nm	[29]
Sweet Potato	Hydrothermal	3.39 nm	[30]

Table 2. Source, Synthetic Approach and Size of Graphene Quantum Dots

Sources	Synthetic Approach	Size	References
Cotton Cellulose	Hydrolysis followed by cyclic condensation of glucose	5nm	[32]
Coal tar pitch (CTP)	Mixture of H_2O_2 and CTP is refluxed	1.7-0.4nm	[31]
Cow milk	One-pot microwave assisted	5nm	[34]
Starch	One-pot hydrothermal reaction	2.25-3.50nm	[23]
Citric acid 3,4-dihydroxy-L-phenylalanine	Facile one-pot solid-phase method	12.5nm	[36]
Silica supported Silver nanoparticle and GQD	Greener photochemical approach	1-4nm	[37]
Coffee grounds	Hydrazine hydrate-assisted hydrothermal cutting	2-8nm	[38]

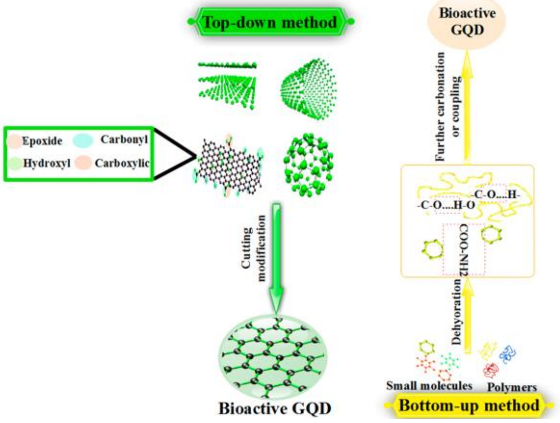

Figure 1. The top-down and bottom-up methods

2. Sustainable and Eco-Friendly Preparation of Graphene Quantum Dots (GQD's)

The production of GQD's frequently uses corrosive acids or organic solvents, and there are still several obstacles in the way of their sustainable green production. Thus, non-toxic synthetic approaches that utilize natural, renewable raw resources as precursors and have simple separation and no complex post-processing should be developed. Here we have discussed some of those methods.

2.1. From Cotton Cellulose

The initial advances in the synthesis of GQDs is the hydrolysis of cellulose, followed by cyclic condensation. This is the reaction mechanism that is described. The produced GQDs have a limited distribution and uniform diametQers between 1.0 and 5.0 nm. They also have excellent hydrophilicity, high photoluminescence emission, favorable biocompatibility and cytotoxicity. Moreover, the synthesized GDs are a great photoluminescent biomaterial that may be used in cell imaging. The current procedure is among the greenest ways to fabricate GQDs when compared to other hydrothermal procedures.[32]

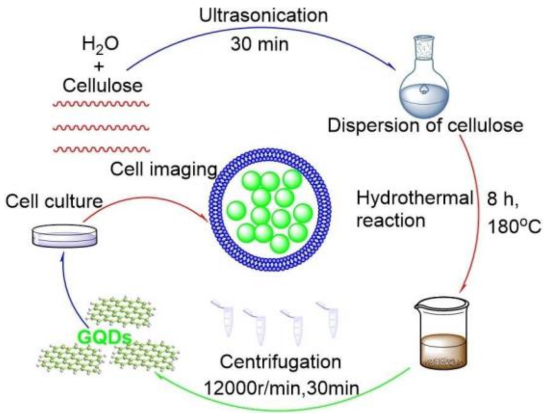

Figure 2. The green synthesis of GQD using cellulose [32]

2.2. Coal Tar Pitch (CTP)

Coal tar pitch (CTP), a derivative of the food-processing industry, is a molecule with a graphene-like structure that includes an aromatic nucleus and many bonds with branches on it. This structure is structurally comparable to GQD's. A method has been reported with a high yield of more than 80 weight percent for converting coal tar pitch into monodispersed GQD's of approximately (1.7-0.4) nm by using H2O2 for oxidation in moderate circumstances. In an aqueous solution, these graphene quantum dots demonstrated high fluorescence and substantial solubility [31].

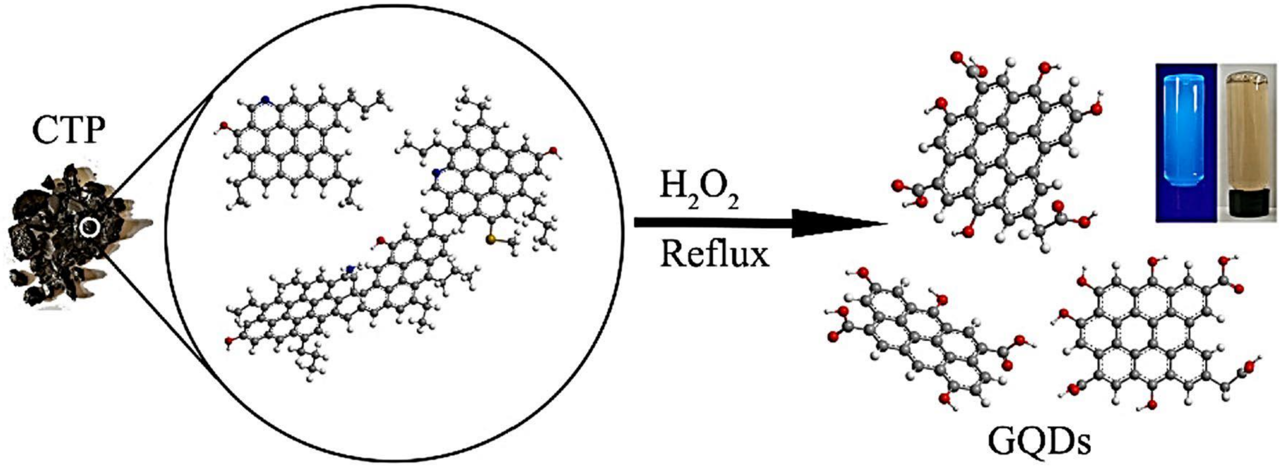

Figure 3. Green synthesis of GQD using Coal Tar Pitch [31]

2.3. From Starch as a Precursor

A simple, environmentally benign, one-pot hydrothermal synthesis using starch as a derivative has produced GQD's of approximately 2.25nm to 3.50nm. The preparative step involves hydroxylation first, then ring-closure condensation. The generated GQD's were utilized for cell imaging due to their suitable hydrophilicity, low cytotoxicity, and high photoluminescence emission [23]. Furthermore, a straightforward and highly productive hydrothermal method was presented for creating GQD's from glucose; the resultant GQD's demonstrated noteworthy green photoluminescence and excitation-independent photoluminescence emission characteristics, with a fluorescence yield of 44.3%.[33]

2.4. From Cow Milk

Water-soluble GQDs for concurrent administration of medications and cancer imaging have been synthesized from cow's milk using a more cost-effective and eco-friendly technique. These multi-fluorescent, spherical quantum dots (less than 5 nm) were created using a one-pot, microwave-supported heating techniques. It was discovered that the photoluminescence (PL) properties of them are highly influenced by the ionic strength and heating time. On graphene quantum dots, berberine hydrochloride was bonded (88% efficacious drug loading) [34]. As a result, the GQD's were shown to be biologically compatible with L929 cells. However, the generated companion therapeutic complex had significant lethal outcome on various cancer cell lines under examination. The produced complex's drug transport and bioimaging properties demonstrated in vitro companion therapeutic applications in cancer therapy.[34]

2.5. Facile One-Pot Solid- Phase Method

Nitrogen-doped GQD's with oxygen-rich functional groups were produced uniformly with an average diameter of 12.5 nm using a one-pot solid-phase method that reported using citric acid as the carbon source and 3,4-dihydroxy-l-phenylalanine as the nitrogen source. These N-graphene quantum dots served as an extremely effective fluorosensor for Hg^{2+} detection due to the effect of metal ions quenching through non-radiative electron transfer. With a detection limit of 8.6 nm, this selective fluorosensor demonstrated strong sensitivity toward Hg^{2+} and was effectively utilized for the successful discovery of Hg^{2+} in riverwater samples [35]. Nnitrogen-GQD's with a high yield of 28.10% were prepared using an environmentally friendly method that demonstratedhigh emission of blue fluorescence, peaking at 450 nm and 355 nm, respectively, due to excitation. Designed with a relatively low detection limit of 0.032 µm, they were intended for the effective and sensitive detection of Hg^{2+} by utilizing the effective quenching effect of Hg^{2+} on Nitrogen-GQD's. With the addition of biothiols, the fluorescence of the Nitrogen-graphene quantum dot/Hg system was restored based on the specific coordination of biothiols and Hg^{2+}. A sensitive response to biothiols was demonstrated by this fluorescent "Off-On" method, which has a detection limit of 0.036 µm for cysteine and 0.034 µM for glutathione. Hg2+ in actual water samples and biothiols in serum samples have both been successfully monitored using the Nitrogen-GQD's based fluorescence technique.[36]

2.6. Using Coffee Grounds

A method for creating extraordinarily fluorescent GQD's from coffee grounds was published. It involved hydrothermal cutting assisted by hydrazine hydrate and functionalizations with polyethylenimine. The graphene quantum dots functionalized with polyethyleneimine showed intensify band-edge photoluminescence with single exopnential decay along with their bioimaging applications were also reported [38].

2.7. Using Hydrogen peroxide

Graphene oxide (GO) was employed as a starting material in a greener synthetic process that produced fluorescent graphene quantum dots without the use of corrosive reagents. Hydrothermal synthesis, which took around two hours to complete, was made possible with the use of hydrogen peroxide.

Cell viability assessments revealed that the prepeared graphene quantum dots exhibited remarkable biocompatibility and high photostabliity. Because of the interactions between the cell and GQD's, these quantum dots have the capacity to be used as fluorescent nano-probes for bioimaging, medication delivery, and diagnostics [39].

2.8. Using Silica Supported Silver Nanoparticle and GQD's Compound

A more environmentally friendly photochemical method and an electrophoresis deposition technique have been utilized to design and produce GQD's and silica-supported silver nanoparticles, which togethercreate a Raman scattering substrate with an increased surface that is very active. This method created in situ silver-GQD substances exposed to UV radiationby using the electrochemically generated graphene quantum dot aqueous solution as both a reducing agent and solvent. These substances were gathered using the electrophoresis deposition method on a SiO_2-supported Si substrate. Because graphene quantum dots are distributed in the spaces between neighboring silver nanoparticles and have an appropriate size of ~1-4 nm, they may function as "hot spot" locations for Raman scattering signal illumination. Along with the improved adsorption of Rhodamine 6G (R6G) molecules via π-π stacking, the SiO_2 template's increased specific surface area, and the electrostatic interactions from GQD's, the as synthesized substrate demonstrated a powerful and highly reproducible surface-enhanced Raman scattering signal, pushing the R6G limit of detectionto 8.0×10−14 M [37].

3. Conclusion

The above discussion emphasized novel results from research concerning green quantum dots (GQDs) developed from sustainable and regenerative raw sources with ecologically favorable methods. Indeed, compared to conventional carbon-based materials, synthesized carbon-based nanoparticles offer a viable, cost-effective, and biocompatible solution. The utilization of biomass has produced high yields of carbon nanostructures has garnered greater scrutiny in the preceding five years. Although numerous kinds of biomass processing methods have been implemented and environmentally friendly techniques have been constructed, only a few of these procedures are capable of guaranteeing substantial yields. Looking ahead, GQDs offers a stimulating future, as advances in nanotechnology will enable an array of biological sensors based on simple, non-toxic synthetic approaches in addition to very stable, precise electrochemical detectors.

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