

## Review Article

# Comprehensive Review on Synthesis, Applications, and Challenges of Graphene Quantum Dots (GQDs)

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Carbon-based nanomaterials are contemporary and are outpacing the technology platform. Graphene quantum dots (GQDs) had a significant impact on the subject of bioengineering, pharmaceuticals, biomedicine, biosensors, fuel, energy, etc. Depending on how quickly this field is developing, it is important to recognize the new difficulties that GQDs have to overcome. This is incredibly significant because many novel applications and innovations that have made GQD synthesis easier recently have not been systematically evaluated in prior studies. Their ability to combine the benefits of quantum dots,  $sp^2$  carbon materials (large specific surface area), and have rich functional groups at the edge makes them special. The naturally occurring inert carbon helps to stabilize chemical and physical characteristics and makes significant advancements in the creation of benchmark photocatalysts. Moreover, current challenges and potential of these rapidly developing GQDs are emphasized. The future of GQD research is limitless, according to the assessment in this review, notably if future research focuses on simplicity of purification and ecofriendly synthesis. This feature article offers a realistic summary on recent developments in the synthesis, characteristics, and uses of GQDs. Frequent review articles focusing on the progress of GQDs for specific applications are published but a thorough review article on GQDs for their numerous uses has not yet been published. The recent trends of scientific research based on new optical biosensing applications, including the comprehensive applications of different zero-dimensional nanomaterials, specially GQDs are discussed in this study.

## 1. Introduction

Fullerene, carbon nanofiber, diamond, grapheme, carbon nanotubes, and GO are all carbon nanomaterials that have been thoroughly investigated for a variety of potential applications [1–3]. Nanotechnology is the focus of contemporary

scientific and technical research, and it promises to revolutionize industries such as transportation, medicine, environment, information technology, electronics, and solar energy. Nanotechnology's outstanding skills in shaping materials structures at extremely small sizes to achieve the desired qualities, allow us to realize the true promise of this

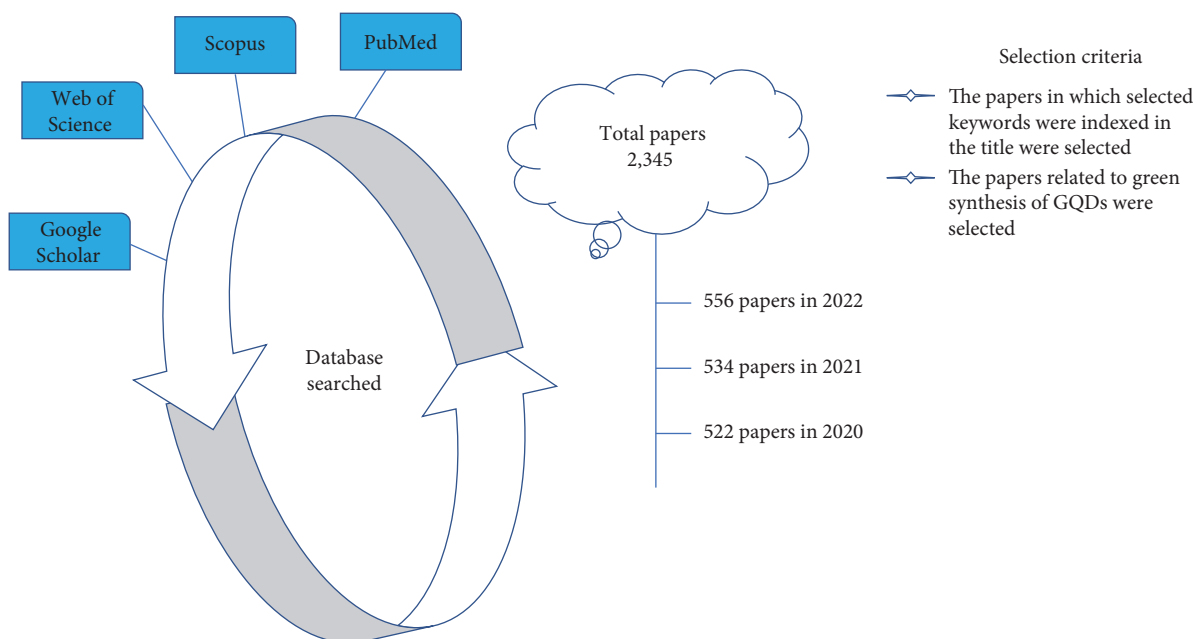


FIGURE 1: Schematic methodology adapted for review.

technology [4]. Nanomaterials science has advanced rapidly in the last 20 years, resulting in new prospects for materials design in scientific, technological, and industrial applications. Carbon-based materials have sparked the interest of researchers in a variety of potential fields because of their outstanding properties, like tunable structure, large surface area, less toxicity, high conductivity, and long life [5].

Graphene has revolutionized several realms of science and technology from a mysterious chemical to a magnificent legend. This is due to graphene's unique nanoscale properties, which include high current density, chemical stability, high optical permeability, good thermal conductivity, and outstanding hydrophobicity. Graphene has been gaining substantial attention with its two-dimensional,  $sp^2$  hybridized extended structure and zero bandgaps, which was purportedly created by mechanical exfoliation in 2004 [6]. Sanchez et al. [7] investigated the facets of graphene-based nanomaterials, such as their toxicity and adequate communication with cells, tissues, and biomolecules as the number of layers increased.

Quantum dots are crystalline materials with diameters ranging from 1 to 10 nm and consist of 100–10,000 atoms that produce light after being excited. In comparison to macrocrystalline materials, their modest size distinguishes them [8]. Quantum dots are notable for their large surface area and optical characteristics, as a result of which they have become a sensor for targeted medication administration and pharmacotherapy. Graphene quantum dots (GQDs) paired with ligands are used to target the cells or tissues in concern. With advancements in the manufacture of biocompatible GQDs, the use of GQDs for *in vivo* studies has recently gained attraction [9]. QDs can have high solubility in plenty of other solvents, such as aqueous buffers when formed as a semiconductor fundamental with a sealant and a cap [10].

Out of 2,345 articles which were available with the above-mentioned keywords only 296 of those articles were chosen using thorough inclusion and exclusion criteria, as shown in Figure 1.

GQDs, which are recent immigrants to the carbon nanomaterials family, are composed of one or few nanometers-size graphene sheets and have extraordinary electrical and optical properties [11]. GQDs are 0D nanomaterials and were initially made from graphene sheets in 2010 using a hydrothermal technique [12]. GQDs have features comparable to graphene, including surface groups like carbonyl, hydroxyl, carboxyl, and epoxy, as well as a crystal structure composed of C, O, and H [13]. GQDs are crystalline and mostly made up of  $sp^2$  hybridized carbon. GQDs are luminous due to their quantum confinement, zigzag or armchair edges, and surface defects giving them unique fluorescence features [14–16]. GQDs typically range in size from 3 to 20 nm. However, the largest size being reported is 60 nm [17].

GQDs are accolladed by many researchers due to their intriguing properties, including low cytotoxicity, excellent water solubility, high electrical conductivity, good biocompatibility, chemical stability, photoluminescence, low photobleaching, environmental friendliness, and optoelectronic properties [18]. GQDs have the potential to be used in flash memory devices, solar cells, electronic displays, packaging, LEDs, antibacterial activity, drug delivery, tissue engineering, supercapacitors, batteries, optoelectric detectors, bioimaging, photodynamic therapy photocatalysis, anticancer agent various biosensors, lithium-ion batteries and energy conversion, and theranostic applications [19, 20]. The purpose of this paper is to review the research on the efficacy of green GQDs. The biocompatible, target-specific, and biologically produced GQDs aid in the effective treatment and management of dreadful diseases.

TABLE 1: Approaches used to synthesize GQDs with its potential applications.

S. no.	Method	Source	Size (nm)	QY (%)	Applications	References
Process—top-down						
1	Chemical oxidation	CX 72 Carbon black	15–18	2.4–4.0	Bioimaging and biolabeling	[21]
2	Hydrothermal	GO	5–19	7.4	Energy applications	[22]
3	Solvothermal method	GO	5.3	11.4	Bioimaging	[23]
4	Microwave irradiation	GO	~4.5	22.9	Detection of Cd <sup>2+</sup>	[24]
5	Microwave-hydrothermal	Glucose	~3.4	7–11	Blue and white LED	[25]
6	Hydrothermal treatment	Graphene sheets	~2.5	19–29	Bioimaging	[26]
7	Microwave hydrothermal	GO	2–6	12.5	Biolabeling and bioimaging	[27]
8	Microwave irradiation	Graphite	2–5	9	Bioimaging	[28]
9	Hydrothermal treatment	1,3,6-Trinitro-pyrene	~2.5	9.2	Detection of Ag <sup>+</sup> ions	[29]
10	Sonochemical and microwave heating	GO	2–5	23.8	Metal ions sensing and bioimaging	[30]
11	Microwave assisted	Glucose and urea	15	11–32	Removal of triazine	[31]
12	Hydrothermal treatment	CA	NA	9%	Photodegradation of methylene blue	[32]
13	Solvothermal treatment	Graphite	~35	15	Biomedical applications	[33]
14	Solvothermal treatment	Graphite	2.5–50	8.8	Detection of Tb <sup>3+</sup> and Eu <sup>3+</sup>	[34]
15	Solvothermal method	CA	NA	NA	H <sub>2</sub> S gas detection	[35]
16	Electrochemical method	MWCNTs	3–8.2	5.1–6.3	Nanoelectronic and biomarkers devices	[36]
17	Electrochemical method	GO	2.4–4.6	7.8	Biosensor	[37]
18	Electrochemical	Graphite rod	5–10	14	Labeling of stem cell	[38]
19	Electrochemical method	Graphite	~20	18.95	Bioimaging and detection of Fe <sup>+3</sup>	[39]
Process—bottom-up						
20	Pyrolysis	CA	~15	9.0	Photovoltaic devices	[40]
21	Pyrolysis	CA	5–10	22.2	Biosensing	[41]
22	Pyrolysis	CA	~12.7	6.91	Biosensing and bioimaging	[42]
23	Pyrolysis	L-glutamic acid	~4.66	54.50	Cell imaging	[43]
24	Pyrolysis	Trisodium citrate	1.30	3.60	Cell imaging	[44]
25	Pyrolysis	Melamine powder	2–6	0.22–0.76	LED	[45]
26	Pyrolysis	CA	~2.0	62.8	Cellular imaging	[46]

## 2. Synthesis of GQDs

Primarily used methodologies in the synthesis of GQDs are “top-down” and “bottom-up” approaches via diverse synthetic protocols (Table 1). The top-down technique typically employs large sp<sup>2</sup> carbon domains like as fullerene, carbon nanofiber, diamond, graphene, carbon nanotubes, and GO. This approach, however, frequently results in a low quantum yield and less photocatalytic activity. The rigid cutting makes controlling the morphologies, dimensions, and the precursors are restricted to bulk carbon materials.

“Bottom-up” synthesis has attracted a lot of interest due to its simple and straightforward preparation process. GQDs are often synthesized from tiny organic molecules including phenyl compounds using a bottom-up approach that involves hydrothermal or solvothermal treatment. Dehydrogenation and carbonization advances are the major reaction processes. This approach can normally produce high-quality GQDs, but the starting ingredients are usually highly toxic, posing a

severe environmental risk [47]. To summarize, the bottom-up method is thought to be a suitable choice for synthesizing high-quality GQDs, but the shortcomings of precursors must be overcome.

The science community understood that to achieve the “sustainable green synthesis goals,” it is important to manage the plentiful waste biomass that might be used to make graphene-like materials. Fabrication of innovative GQDs with fascinating features from low-cost, renewable plant biomass has been a hot issue [48]. Bioinspired synthesis is more favorable than other traditional approaches to nanomaterial synthesis due to the eco-friendly method and easier availability of biological entities [49]. Plant biomass is primarily constituted of cellulose, hemicellulose, and lignin, the latter of which contributes significantly to structural integrity and chemical resistance [50]. Even though biomass is an eco-friendly, renewable, cost-effective, reliable, and natural source of carbon, it can now be used in the mass production of GQDs [51]. Recently, biomass like rice grain [52], wood charcoal

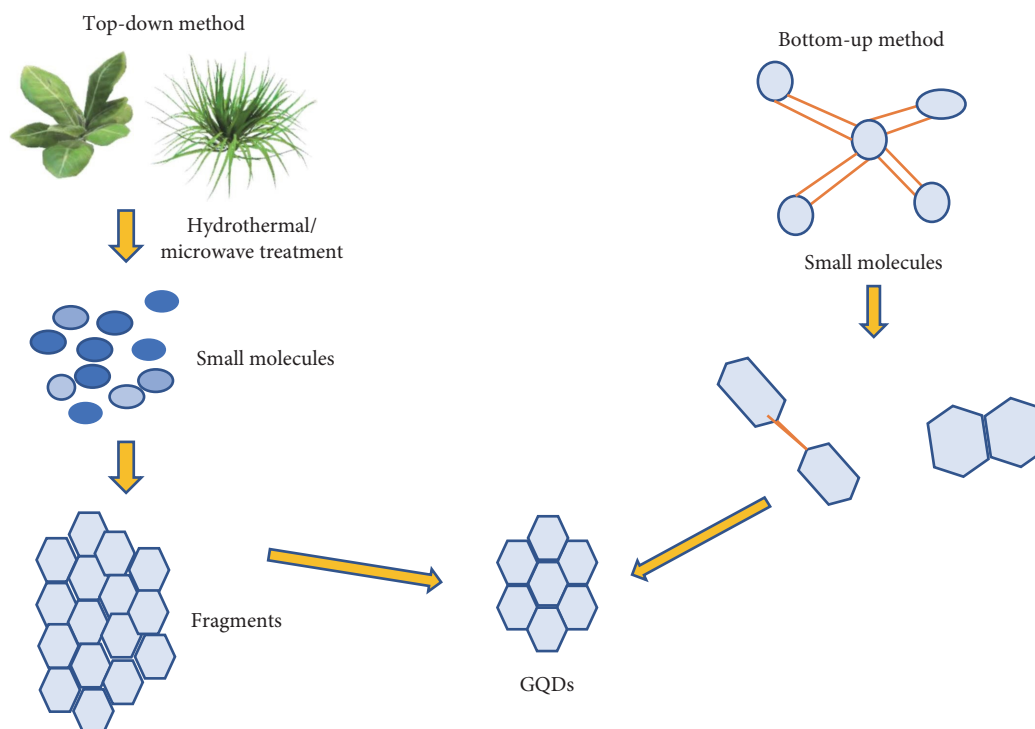


FIGURE 2: Approaches for synthesis of GQDs.

TABLE 2: Comparison of synthesis method for GQDs.

Types	Methods	Advantages	Disadvantages
Top-down	Hydrothermal	Easy, quick, and ecofriendly	It is necessary to use treated carbon material
	Electrochemical oxidation	Stable and regular size GQDs formed	Production yield is low
	Microwave assisted	Reaction time decreased	It is expensive method
	Oxidative cleavage	Large scale production	Oxidizing agent may cause explosion
Bottom-up	Controllable synthesis	Stable and regular size GQDs formed	Time consuming process
	Carbonization	Facile and ecofriendly method	Polydispersed GQDs may obtained

[53], tea waste [54], rice husk [55], plant leaves [56], flowers [57], durian, lignin [58], cow milk [59], and molasses [60] have been used as precursors to synthesize GQDs. However, GQDs with high quantum yield is hard to procure directly from biomass material, and investigators enhanced GQD quantum yield by doping of heteroatoms, which complicate the synthesis process. A simple, efficient, environmentally, ecofriendly, and green starting material are desperately needed to produce the best quantum yield GQDs. Figure 2 shows the process of synthesis of GQDs via top-down and bottom-up approaches and Table 2 shows the comparison of both top-down and bottom-up approaches.

GQDs can be synthesized from glucose [61], citric acid [41], and dead neem leaves [62] by pyrolysis; from graphene oxide [63], corn powder [64], cane molasses [65], coconut husk [66] by hydrothermal method; from graphite rod [38], wood charcoal [53] by electrochemical method; from GO [23], guava leaves [67], and dimethylformamide by solvothermal method; from bamboo timber waste [68], neem, fenugreek leaves [69], and cotton cellulose [70] by hydrothermal

method, Arjuna Bark [67], *Opuntia* [19], and mango leaf [71] by microwave-assisted method.

**2.1. Green Synthesis.** In recent years, both industrial and academic researchers have turned toward a more broad perspective focused on pollution, sustainable sources, and waste minimization, resulting in the arrival of a new chemistry approach known as green chemistry. The major issues of the modern world include energy scarcity, restricted availability, and overconsumption of nonrenewable resources, as well as increasing degradation of the natural environment [48, 69]. The concerns about global warming and pollution have prompted researchers to look for new functional materials that are clean, sustainable, renewable, and ecologically benign [48, 64].

Biological sources are now used as a source of clean, organic, feasible, cheap, and productive carbon source in the optimized fabrication of GQDs (Table 3). Because biomass is plentiful and cost-effective, and there is no publication on the overall cost of GQD components, the cost of biomass precursors is predicted to be lower than the value

TABLE 3: GQDs synthesized from biological sources and its applications.

S. no.	Source	Method	Result	References
1	Dead neem leaves	Pyrolysis	Detection of Ag <sup>+</sup> ions	[62]
2	Mango leaf	Microwave assisted	Detection of intracellular temperature	[71]
3	Cow milk	Microwave	Bioimaging	[59]
4	Wood charcoal	Electrochemical synthesis	Detection of 200 <sub>2</sub> and glucose	[53]
5	Corn powder	Hydrothermal	Lower charge recombination while increasing free charge carriers	[72]
6	Sugarcane molasses	Hydrothermal	Bioimaging	[73]
7	Cooking palm oil	Double thermal chemical vapor deposition	Carbon source for the fabrication of graphite	[74]
8	Rice husk	Hydrothermal method	Extraction of Pb(II) and La(III) from real water	[55, 75]
9	Sugarcane bagasse	Hydrothermal carbonization	Preparation of better value-added biomass materials	[76]
10	Turmeric powder	Hydrothermal method	Anti inflammatory activity	[77]
11	Cotton cellulose	Hydrothermal	Low cytotoxicity	[70]
12	Mango peels	Hydrothermal	Effective removal of Pb(II)	[78]
13	Marigold petals	Pyrolysis	Detection of Fe <sup>3+</sup> ions	[79]
14	Popcorn powder	Hydrothermal	Antibacterial activity	[80]
15	<i>Ziziphus mauritiana</i> seeds	Microwave assisted	Monitoring of ammonia	[81]
16	Rice grains	Pyrolysis	Bioimaging	[52]
17	Cane molasses	Hydrothermal treatment	Bioimaging	[73]
18	<i>Passiflora edulis</i>	Hydrothermal treatment	Cell imaging	[82]
19	Neem leaf	Hydrothermal method	Bioimaging	[83]
20	Bamboo timber waste	Hydrothermal method	Detection of curcumin	[68]
21	Corn powder	Hydrothermal	Engineering application	[64]
22	Coffee grounds	Hydrothermal cutting	Bioimaging	[84]
23	Grape seed extract	Microwave assisted	Bioimaging	[85]
24	Sugarcane bagasse	Hydrothermal method	Antibacterial activity	[86]
25	Miscanthus	Hydrothermal carbonization	Detection of Tri-channel sensitive Fe <sup>3+</sup> ions	[87]
26	Miscanthus	Ultrasound assisted	Synthesis of graphene materials	[88]
27	Cocoa bean	Hydrothermal method	Bioimaging	[89]
28	Opuntia	Microwave assisted	Detection of phytic acid	[19]
29	Marigold flower	Hydrothermal method	Synthesis of a sustainable supercapacitor electrode	[57]
30	Tea waste	Hydrothermal treatment	Detection of Fe <sup>3+</sup>	[54]
31	Molasses	Carbonization	Thermal degradation is observed around 140°C	[60]
32	Pineapple leaf fiber	Hydrothermal	Detection of Hg <sup>2+</sup>	[90]
33	Pinto beans	Hydrothermal	Antibacterial activity	[91]
34	Lemon leaves	Carbonization	Synthesis of GQDs	[92]
35	Corn straw	Hydrothermal method	Detection of PO <sub>4</sub> <sup>3-</sup>	[93]
36	Dhruva grass	Solvothermal method	Photoluminescence property	[94]
37	Paddy straw	Hydrothermal	Enhancement of dielectric property	[95]
38	Red onion	Hydrothermal	Electrochemical hydrogen storage	[96]
39	Garlic extract	Pyrolysis	Chelating agents	[97]
40	Potato amylose	Hydrothermal	Detection of tetracycline	[98]
41	Orange peel waste	Hydrothermal	Detection of Fe <sup>3+</sup>	[99]
42	Watermelon rind waste	Hydrothermal	Detection of Fe <sup>3+</sup>	[100]
43	Tamarind shell powder	Hydrothermal	Detection of uric acid	[101]
44	Leaves of curry tree	Hydrothermal	Detection of aflatoxin B1	[102]
45	Pistachio shells	Hydrothermal	Detection of cysteine	[103]

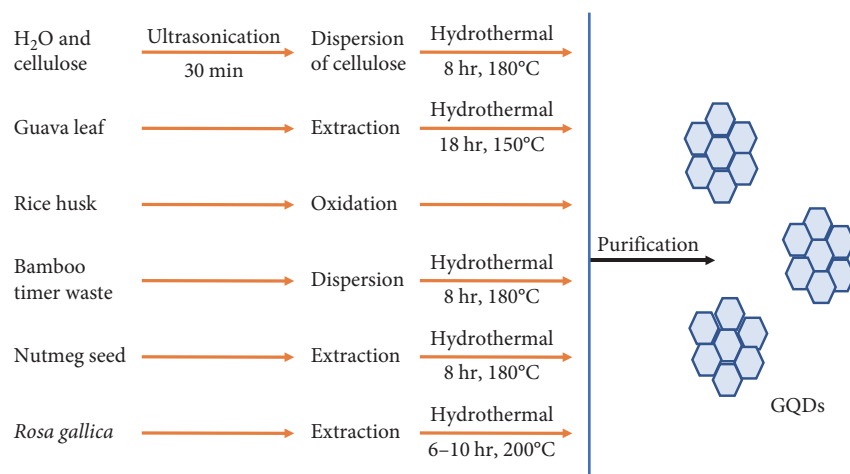


FIGURE 3: Green synthesis of GQDs from various plant parts [55, 56, 8, 68, 70, 104, 98].

of other precursor chemicals (graphite powder, carbon fiber, glucose, CNTs, and CA). Using various types of biomass, like tea waste [54], plant leaves [56], flowers [57], rice grain [52], coffee grounds [84], miscanthus [88], and wood charcoal [53] GQDs were synthesized, as shown in Figure 3. GQD processing with an extraction yields is more conceivable than the costly graphene-based precursors. GQDs obtained from biomass have a higher quantum yield than graphene and its derivatives. Plant-derived bioinspired GQDs demonstrate promising drug delivery and anticancer activity while posing low cytotoxicity. Plant sources with bioactive compounds as anticancer nanomaterials have various advantages over chemical medications, including the ability to work at lower concentrations, ability to pass the blood–brain barrier, and target specificity. The fundamental benefit of bioinspired nanomaterial is ability to use a wide range of precursors and a variety of technological techniques [64]. Belletti et al. [105] showed cytotoxic effect of curcumin-loaded PLGA nanoparticles with quantum dots on HBL6 and BCBL-1 cell lines. The bioavailability of curcumin and the activity were improved as a result of the loading process. Developing ecofriendly routes to procure GQDs obtained not only from biodegradable materials such as biomass waste, as well as from other natural ingredients found in food or agricultural waste (i.e., lignin, carbohydrates, proteins) without contending with food suppliers without use of any organic solvents, oxidizing, reducing, or passivating agent is a hot topic of research in the twenty-first century.

**2.2. Microwave-Assisted Method.** When compared to other approaches, microwave-assisted nanoparticle production has numerous advantages. The advantage of this method over the hydrothermal method is that it is faster and has a lesser fabrication temperature. Microwave-assisted reactions have a number of advantages, including: (1) low levels of impurities in the products, (2) easy temperature and pressure control, (3) high product efficiency, (4) selective heating (i.e., reduced energy costs), (5) environmental friendliness, (6) high security, (7) reproducibility, and (8) easy control of product size [106, 107]. Microwaves have been proposed

by Ayele et al. [9] as bioinspired method for synthesis of CdSe quantum dots [19]. Gu et al. [108] suggested a simple and rapid process for the manufacture of nitrogen-doped GQDs utilizing microwave produced from the root of cedar tree without any surface modification.

**2.3. Electrochemical Oxidation Methods.** Controlling the current–voltage ratio with electrochemical methods allows nanostructures to be adjusted. Electrochemical corrosion methods of carbon reagents occur when a controlled voltage applied to a mass of precursor materials, leading to the formation of nanomaterials. A high temperature does not involve in this process and it can be carried out rapidly on a large scale utilizing any solvents. It is the quickest method to generate graphene sheets [109–111]. Wong et al. [112] proposed using the electrochemical technique to synthesize nitrogen-doped GQDs by the bottom-up approach. This approach is, therefore, simple, clean, and green, which is favorable for larger synthesis having 95% interest rate. The particles have a quantum gain of 0.71 [112].

**2.4. Hydrothermal Method.** The hydrothermal method is a quick and easy way to generate bioactive GQDs, which includes one-step process involving heating a natural precursor to high temperatures and pressures in a teflon line autoclave. The bonds between carbon nanomaterials are disrupted because of high pressure and high temperatures, resulting in bioactive GQDs (Figure 4). Different precursors and temperature optimization can be used to change the electrical–optical characteristics of the particles. To modify the electrical–optical characteristics of the particles, different precursors and temperature optimization can be applied. Using a hydrothermal technique, Liang et al. [113] produced extremely luminous quantum dots from gelatin with ease. Liu et al. [114] created a simple, clean, and cost-effective method for producing luminous quantum dots using hydrothermal processing. They looked into the use of produced quantum dots in Fe<sup>3+</sup> detection and cell imaging. Wang et al. [93] synthesized GQDs from corn straw to detect phosphate ions.

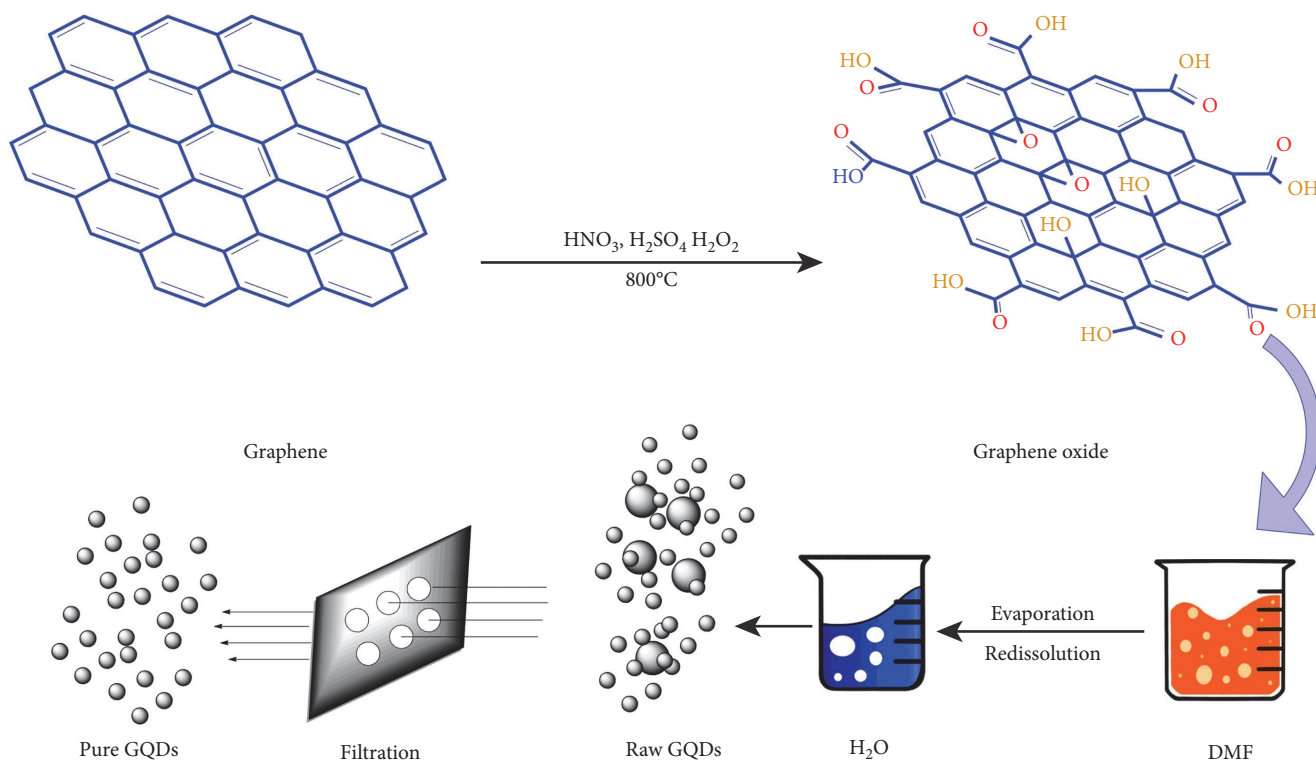


FIGURE 4: Synthesis of GQDs from graphene oxide [33].

**2.5. Ultrasonic Method.** In recent decades, plenty of approaches to develop photocatalyst materials to be used in solar cells have been introduced. Moreover, from the standpoint of green chemistry, the advantages of synthesizing these minerals in unique methods are intriguing. In aspects of science and technology, the concept of producing incredibly effective photocatalytic activity via ultrasound is both exciting and vital, and it holds great potential for developing photocatalysts in the coming future. This method is an encouraging method for controlling material size, structure, and dimension and photocatalytic activity [115]. Zhu et al. [116] used ultrasonic irradiation to produce high GQDs using potassium permanganate and graphene oxide and used them for alkaline phosphate detection test. Kir et al. [92] used lemon as a precursor to create GQDs that were both rapid and ecologically friendly. This method creates nontoxic quantum dots that can be used as optical imaging devices.

**2.6. Liquid Peeling Process.** Liquid peeling (LP) of the 0D materials has a tremendous attention because of their scalability. Additionally, LP is the finest method for producing nanofilms because it offers a number of benefits including easy use, low processing costs, and minimal environmental impact. This procedure converts graphite into graphene sheets, enabling the LP process to produce GQDs with excellent crystallinity. Carbon acetylene powder or graphite powder has also been employed as precursors of low and high flaws in this procedure to create GQDs. Recently, after the intercalation, LP of graphite formed graphene, which has attracted a lot of attention. GQDs were synthesized by using LP probe and by sonication of graphite powder by high-intensity ultrasonic waves.

These waves split layers of graphene into ultrafine particles. The process is carried out by the ultra-fine particles, or GQDs, created when these waves separate layers of graphene [117].

**2.7. Method of Soft Template.** In comparison to more conventional synthetic methods, this method was generally an easy and practical way to create nanostructures. The main advantage of using this technology to clarify the properties of any nanoparticles is the ability to efficiently oversee the form, size, and surface texture of graphene nanomaterials. The template system is separated into hard templates and soft templates based on its distinctive form. The soft template method is far more suitable for GQD output in contrast to the hard template method. It may enhance the characteristic nanoscale reaction vacuum smoothly in operations of separation, purification, and mass processing [117].

### 3. Applications

**3.1. Sensors.** QDs are an enthralling collection of materials. It is a class of materials that have unusual fluorescence properties. The ability to fluoresce efficiently is the primary reason why QDs offer so much potential in a variety of sensing applications [118]. GQDs have intriguing optical absorption properties, with a peak ranging from 260 to 380 nm, making them perfect candidates for the production of photodetectors or optoelectronic devices [119]. They have enzymatic activity similar to peroxidase, which makes them ideal for label-free biosensing [120]. Furthermore, biosensors based on GQDs offer a wide range of applications in illness diagnosis, prognosis, and therapy (Table 4). These biosensors are categorized

TABLE 4: Applications of QGDs in sensors.

S. no.	Source	Method	Application	Linear range	LOD	References
1	L-glutamic acid	Pyrolysis	Rapid detection of H <sub>2</sub> O <sub>2</sub>	0.1–10 Mm	20 mM	[43]
2	Glycine	Thermolysis	Determination of Fe <sup>3+</sup>	0.5–500 μM.	100 nM	[121]
3	SBA-15 template	Vapour cutting method	Determination of Fe <sup>3+</sup>	3–60 μm	0.3 μM	[122]
4	CA	Carbonization	Detection of microRNAs (miRNAs)	0.1–200 nM	100 pM	[123]
5	CA	Pyrolysis	Determination of sunitinib	0.05–20.00 μmol L <sup>-1</sup>	0.03 μmol L <sup>-1</sup>	[124]
6	GO	Visible-fenton reaction	Detection of lipovitellin	0.001–1,500 ng/ml	0.9 pg/ml	[125]
7	GO	Electrochemical	Detection of glucose	0.25–50 μM	0.1 mM	[126]
8	Watermelon rind waste	Hydrothermal	Detection of Fe <sup>3+</sup>	NA	0.28 μM	[100]
9	Tamarind shell powder	Hydrothermal	Detection of uric acid	10–100 μM	401.72 pM	[101]
10	1,3,6-Trinitropyrene	Hydrothermal	Fluorescent detection of Ag <sup>+</sup> ions	0.1–130.0 nM	30 nM	[29]
11	Glucose	Pyrolysis	Optical sensor for glucose	4–40 mM	3.0 mM	[61]
12	CA	Pyrolysis	Detection of catechins	0.01–30 μM	0.005 μM	[127]
13	Wood charcoal	Electrochemical method	Detection of glucose and H <sub>2</sub> O <sub>2</sub>	0.01–0.6 mM	0.006 mM	[53]
14	CA	Photoelectrochemical synthesis	Zeatin detection	0.1–100 nM	0.031 nM	[128]
15	Glucose powder	Thermal pyrolysis	Determination of cholesterol	0.08–300 μmol L <sup>-1</sup>	35 nmol L <sup>-1</sup>	[129]
16	GO	Sonochemical and microwave heating	Detection of Fe <sup>3+</sup>	10–120 μM	10 × 10 <sup>-6</sup> M	[30]
17	CA	Pyrolysis	Hepatitis B virus DNA detection	10–500 nM	1 nM	[130]
18	Graphite rods	Electrochemical synthesis	Detection of tumor cells	2–64 nM	1.19 nM	[131]
19	CA	Pyrolysis	Antibodies detection	0.16–125 U/ml	0.11 U/ml	[132]
20	GO	Sonoe fenton reaction	Detection of CFP-10 protein	0.0050–500 μg ml <sup>-1</sup>	0.330 ng ml <sup>-1</sup>	[133]
21	CA	Pyrolysis	Detection of catecholamine neurotransmitters	1–120 μM	83 nM	[134]
22	Glucose	Hydrothermal	detection of copper ions	NA	5.6 nM	[135]
23	CA	Pyrolysis	Fluorescent detection of microRNA	1 × 10 <sup>-18</sup> –1 × 10 <sup>-12</sup> M	1 × 10 <sup>-18</sup> –1 × 10 <sup>-12</sup> M	[136]
24	CA	Pyrolysis	Topotecan determination	0.35–100 μM	0.1 μM	[137]
25	GO	Pyrolysis	Detection of cardiac Troponin I	0.17–3 ng ml <sup>-1</sup>	0.02 ng ml <sup>-1</sup>	[138]
26	Graphite flakes	PLA process	Detection of Fe <sup>3+</sup>	500 nM–50 μM	~5.36 μM	[139]
27	CA	Hydrothermal method	Detection of carcinoembryonic antigen	0.5–1,000 ng ml <sup>-1</sup>	0.01 ng ml <sup>-1</sup>	[140]
28	Graphene	Electrochemical exfoliation	Detection of Fe <sup>3+</sup>	Wide range	≈7.22 μM	[141]
29	Coal	Ultrasonic	Detection of Cu <sup>2+</sup>	0–8 μmol L <sup>-1</sup>	0.29 μM	[142]
30	Leaves of curry tree	Hydrothermal	Detection of aflatoxin B <sub>1</sub>	5–800 ng ml <sup>-1</sup>	0.158 ng ml <sup>-1</sup>	[102]
31	GO	Hydrothermal	Detection of Hg <sup>2+</sup>	0–4.31 μM	23 nM	[143]
32	GO	Hydrothermal	Detection of paraquat	0.05–2.0 μg ml <sup>-1</sup>	19 μg l <sup>-1</sup>	[144]
33	Marigold petals	Pyrolysis	Detection of Fe <sup>3+</sup> ions	NA	41.1 nM	[79]

(continued)



TABLE 4: Continued.

S. no.	Source	Method	Application	Linear range	LOD	References
34	Waste toner	Hydrothermal	Detecting of specific DNA sequence	0.5–30 nM	0.17 nM	[145]
35	CA	Hydrothermal	Detection of Cobalt ions	0–40 mM	1.25 mM	[146]
36	CA	Hydrothermal	Detection of Fe <sup>3+</sup>	0–85 μM	0.26 μM	[147]
37	<i>p</i> -coumaric acid	Hydrothermal	Detection of Cu <sup>2+</sup>	0–10 μM	57 nM L <sup>-1</sup>	[148]
38	GO	Electrochemical synthesis	Sensing soil moisture with response time of 180 s	NA	NA	[110]
39	CA	Pyrolysis	Detection of Hg <sup>2+</sup> and ClO <sup>-</sup>	0.25–5.0 μM	22.1 nM	[149]
40	CA	Hydrothermal	Detection of cysteine	0.5–5 μM	0.1 μM	[150]
41	Lactose	Hydrothermal	Detection of Fe <sup>3+</sup> ions	2.8–11.2 nM	NA	[151]
42	CA	Hydrothermal	Detection of neuron-specific enolase	0.1–1,000 ng ml <sup>-1</sup>	0.09 pg ml <sup>-1</sup>	[152]
43	CA and thiourea	Solvothermal method	Detection of bisphenol A	0.12–5 μM and 5–40 μM	0.04 μM	[153]
44	Starch	Hydrothermal	Detection of <i>O. tsutsugamushi</i>	NA	0.002 ng/μl	[154]
45	Anthracite and bituminous coals	Chemical route	Detection of glutathione	NA	27 μM	[155]
46	CA	Pyrolysis	Detection of glutathione	0.5–7 μM	0.5 μM	[156]
47	GO	Hydrothermal	Detection of D-phenylalanine	0.1–5 μM	0.023 μM	[157]
48	CA	Microwave irradiation	Detection of isoniazid	0.19–750 μM	10.91 nM	[158]
49	CA	Hydrothermal	Detection of chloride ion	8.5–300 μmol L <sup>-1</sup>	0.1 μmol L <sup>-1</sup>	[159]
50	Pistachio shells	Hydrothermal	Detection of cysteine	0–150 nM	2.38 nM	[160]
51	Pistachio shells	Hydrothermal	Detection of homocysteine	0–100 nM	1.94 nM	[161]
52	Xylan	Hydrothermal	Detection of chromium	3–75 μM	0.10 μM	[162]
53	CA	Carbonization	Detection of copper ion	0–2.5 mM	NA	[163]
54	Starch	Hydrothermal	Detection of clenbuterol	5 × 10 <sup>-10</sup> –5 × 10 <sup>-7</sup> M	2.083 × 10 <sup>-13</sup> M	[164]
55	Lignin	Hydrothermal	Detection of ascorbic acid	NA	1.62 μmol/l	[58]
56	Graphite flakes	Oxidation method	Detection of picric acid	0–200 μm	1.2 μM	[165]

into numerous groups based on the type of transducer used like optical, electrochemical, photoelectrochemical, etc. Liu et al. [166] synthesized PEHA-GQD-His, which are often used as fluorescent probes for microRNA fluorescence platforms for biosensors using the compiled molecular beacon double-cycle amplification strategy.

Ananthanarayanan et al. [141] demonstrated first that GQDs could be used for Fe<sup>3+</sup> detection due to differential fluorescence quenching of Fe<sup>3+</sup> ions. Gupta et al. [167] created a glucose sensor using a GQD and functionalized graphene composite. Raeyani et al. [168] demonstrated GQDs as a viable optical-based CO<sub>2</sub> sensor. Dong et al. [169] reported GQDs sensor for free chlorine detection in drinking water. Sun et al. [170] demonstrated amine functionalized GQDs for the detection of Cu<sup>2+</sup> ions.

**3.1.1. Electrochemical Sensor.** According to the calculation of the applied electric signal, the various electrochemical sensor

types are divided into potentiometry, conductometry, and amperometry or voltammetry. By using pulse differential voltammetry, sensors for electrochemically detecting bisphenol A in water have been developed. Composite electrodes made of polypyrrole (PPy) and GQDs make up this sensor. The sensor demonstrated a good response, with detection limits of 0.01–50 and 0.04 M, respectively [33]. A glucose sensor system based on chemically reduced graphene oxide (CR-GOx) has been reported to have improved amperometric responses for monitoring glucose, with a large linear range (0.01–10 mM) and a 2.0 μM limit of detection (LOD) [171]. The results of the electrochemical study revealed that the electrode with the GNR/Co<sub>3</sub>O<sub>4</sub> coating had a good electrocatalytic activity for the oxidation of H<sub>2</sub>O<sub>2</sub> at 0.925 V. From 10 to 200 M, this sensor responded linearly to H<sub>2</sub>O<sub>2</sub> oxidation. According to calculations, the LOD is 1.27 M [172]. The use of oxygen plasma treatment, which produces oxygenated functions, edge plane sites, and defects, has

improved the performance of graphene-based electrochemical sensors. The treated films showed improved dopamine, ascorbic acid, uric acid, and NADH detection responses [173]. An electrochemical sensor based on Ni/RGO/CCF modified electrode has been reported for uric acid determination with a linear range 10–60  $\mu\text{M}$ , low limit of detection of 5.083  $\mu\text{M}$  [174]. Later on, an electrochemical sensor was also developed by using CuO/rGO nanocomposite for determination of ascorbic acid with a linear range from 500 to 2,000  $\mu\text{M}$  and the limit of detection was 189.053  $\mu\text{M}$  [175].

**3.2. Bioimaging.** Bioimaging is among the most important fields where QDs provide numerous advantages in a favorable manner [176, 177]. Bioimaging is a technique for seeing, observing, and detecting desired molecules or tissues in the body avoiding intrusive procedures [178]. It allows for a more thorough insight of the body's biological pathways [179]. In 1896, Wilhelm Roentgen was able to acquire the first X-ray image. A window has opened for bioimaging applications to track and detect ailments and symptoms like tumor imaging [180], carcinoma [181], Parkinson's disease [182], bone fractures [183], and so on. As a result, GQDs have proven to be an effective bioimaging targeting or evaluating candidate for a diverse array of tumor cell lines. HeLa cell lines have frequently been investigated for *in vitro* imaging [184]. Other cells such as dermal fibroblast cells, A549 cells, T47D, HEK293A cells, MDCK cells, MCF-7 cells, CHOek1 cells, and MC3T3 cells have all been investigated. GQDs are effective and convenient agents in bioimaging because of their remarkable controllable PL features, chemical inertness, photostability, and high biocompatibility [5], it was observed that because of their crystalline form, GQDs perform better than CNDs in terms of reliability and bleaching activity when exposed to a xenon lamp. During the imaging process, GQDs were found to be sustainable particles that had no effect on the results. Fluorescence imaging of NIR spectra is a common research method [71].

QDs have long been used as imaging technique and are effectively accumulated into bioimaging systems seeing as they overcome the challenges of traditional dyeing procedures. Nonetheless, their toxicity later became a constraint to biological applications. GQDs are justified for having no or very few properties in living tissue, regardless of the toxicity caused by other semiconductor QDs [184]. These GQDs, on the other hand, could have improved photostability and lighting, as well as size-tunable characteristics [185]. Notably, the initial showing of GQDs (>10 nm) could produce "blue-PL," as compared to graphene nanoribbons, marked the start of a new era. This accelerated research in labeling and optoelectronics, as well as the advancement of GQD-based bioimaging applications. It is essential to know that the extreme luminous emission of GQDs is caused by the closely packed, free zigzag edges shown in small structure arising from the size >10 nm. Furthermore, it has been demonstrated that PL emission is highly sensitive to moderate pH, that is, alkaline mediums can produce strong PL but acidic mediums cannot, implying that PL can be altered when the pH of the medium changed. Because GQDs have

superior nontoxicity and bioapplicability when compared to other synthetic QDs, new approaches that use GQDs for bioimaging applications have emerged rapidly [186].

Zhu et al. [187] described a single-step solvothermal strategy for extremely green-PL GQDs with a QY of 11.4% for bioimaging, highlighting that due to their biomedical properties, GQDs are also used as tagging. Conjugations of GQDs with various several other materials enhanced their efficiency and convenience over time. The surface modification of GQDs has been revealed to alter the PL. Although a photochemical reduction approach for boosting QY as well as cell uptakes of GQDs has been proposed [188]. Surface-functionalization of GQDs with tiny organic compounds proved to be an effective technique for PL tuning by changing bandgaps and lowering cellular toxicity [189].

Luo et al. [190] reported a microwave-assisted method to prepare A-GQDs having high fluorescence with QY 21.36%. A-GQDs was found to be a suitable candidate for bioimaging because its high fluorescence quantum yield and their cytotoxicity was examined using MTT viability assay on A549 cell as a test organism. A-GQDs show excellent cell viability and have good biocompatibility at the concentration >2 mg/ml. The cell viability was 94%, even after incubated for 24 hr with A-GQDs at a concentration 2 mg/ml.

For bioimaging purposes, various color emissions of GQDs were generated using various synthetic techniques. Hydrothermal treatment of GO, for instance, led to the formation of N-GQDs with blue luminescence. Biomedical applications of N-GQDs were adequate to illustrate HeLa cells under this study [191]. Ecofriendly luminescent GQDs can be mass-produced from graphite powder using a simple fabrication method for imaging of human liver cancer cells. Afterward, water-soluble, uniformly GQDs with red fluorescence revealed a high bioimaging applicability as a potent biological marker for progenitor or stem cells [192]. Due to their biocompatibility and superior-sized tunable emission properties, it was demonstrated that bioinspired synthesis of GQDs may conquer cytotoxicity. Research using mango leaves for green synthesis of GQDs discovered total cellular absorption and viability for 24 hr after GQD treatment, even at high concentrations, with NIR emissions ranging from 650 to 750 nm. The above NIR excitation-independent fluorescent emissions could meet the demand for *in vivo* administrations requiring deep tissue penetration [71].

Yan et al. [193] presented a method for producing highly biocompatible GQDs using only glucose and no acids or oxidizers in the regard of cytotoxicity. The hydrothermal one-pot approach was used to make HGQDs by sterilizing the solution of glucose at 200°C for 10 hr. HGQDs revealed lower levels of apoptosis than conventionally synthesized GQDs with ~60% less cytotoxicity as well as 2.24% QY, demonstrating greater cytocompatibility than commonly synthesized GQDs. The structure of the as-prepared HGQDs was encountered to be similar to that of glucose, which could improve their cellular uptake. *Ex vivo* experiments demonstrated that HGQDs accumulated in mice's liver, kidney, and brain. Furthermore, sugar-based HGQDs were tested to see if they increased malignant cell survival. When cancer cells were imaged, it

was discovered that CGQDs could damage them, whereas HGQDs could provide greater cytocompatibility for bioimaging. *Ex vivo* imaging of isolated organs from rats that had already been handled with HGQDs for 20 days revealed the existence of HGQDs in the brain, liver, and kidney. Their aggregation in the brain, in particular, illustrated that GQDs pass through blood–brain barrier. The above method may describe future brain-related researches for imaging. Chen et al. [194] were able to effectively functionalize the GQDs with sugar moieties, dubbed “sweet GQDs,” which were used for monitoring of precisely tagged carbohydrate receptors to examine authentic complexities. GQDs synthesized above exhibited a QY of 31% and had 5.40 nm size. Similarly, mannose receptors were found to be upregulated in breast cancer cell lines, and the synthesized man-GQDs were found to be capable of recognizing mannose receptors in various body areas. Similarly, mannose receptors were discovered to be upregulated in human breast cancer cells, and man-GQDs were able to recognize mannose receptors in various body areas. Furthermore, it included both biosensing and bioimaging tests, above substantial GQDs were effectively internalized by the MCF-7 cancer cell line and demonstrate significant bioimaging potential. The above heterocyclic hydrocarbon-derived GQDs with sizes ranging from 5 to 10 nm have been found to be great bioimaging probes due to their excellent solubility, high PL, low toxicity, and chemical stability [195]. Chen et al. [196] observed that by increasing the concentration of GQDs from 0.078 to 1.250 mg/ml, cell viability remains more than 80%. This finding suggests that GQDs have minimal cytotoxicity and high biocompatibility, making them appropriate for bioimaging applications.

**3.3. Cytotoxicity Studies.** GQDs’ cytocompatibility opens up a world of biological possibilities and is already desirable in many circumstances. In terms of biosafety, the potential cytotoxicity of GQDs on living cells is to be considered [126, 178]. Furthermore, researchers are actively researching the maximum-suited therapy and alternate solution techniques [197].

According to Tabish et al., [126] GQDs with small diameters and concentration levels in the microgram and milligram range are found to be lesser toxic to rat and mouse cell lines. However, it should be remarked that various studies have found GQDs to be threatening [126]. Wang et al. [47] first time investigated mechanism of cytotoxicity by N-GQDs. The communication between RBCs and carbonic structures was examined because nonspecific adherence of nanomaterial to the cell membrane increased cytotoxicity. As a result, the N-GQDs were found cytotoxic to RBCs, despite the fact that they had no more harmful effects than GO [198]. However, it was also stressed that the way of communication between the living compartment and GQDs must be carefully investigated to gain a complete understanding of the effect mechanism. PDT applications of GQDs, on the other hand, are still pending for comprehensive cytotoxicity data.

Lee et al. [199] demonstrated that GQDs’ excretion pathway is another key component of their toxicity. GQDs were eliminated from mice via the renal system after being degraded by numerous enzymes, such as HRP. Furthermore,

human myeloperoxidase and eosinophil peroxidase enzymes were found to degrade GQDs [200]. PEGylation has been proposed as a viable strategy for reducing the toxicity caused by GQDs [201]. Another study using the idea of PEGylation found that PEGylation may really reduce cytotoxicity because GQDs did not trigger considerable cell death in HeLa cells *in vitro* at concentrations of 160 g/ml. In the same report, GO-PEG and GQD-PEG *in vivo* comparisons were performed with the GQD-PEG conjugation demonstrating exceptional biocompatibility. The small size and ease of elimination from their bodies, as well as the high oxygen content, were factors for their desirable biocompatibility [202]. One gene on hematopoietic stem cells have been appear, this is a small number among the 20,800 genes [203]. At the subcellular, cellular, protein and genetic levels, graphene has biological effects. Graphene’s toxicity is determined by its absorption in numerous organs as well as chemical and physical interference. The buildup of graphene in these organs has an impact on cellular performance. After entering a cellular system, dispensation, their impeachment and excretion acquire information about their cytotoxicity. Mitochondrial imaging has previously been accomplished using a variety of approaches, such as having GQDs gather mitochondrial and lysosomal sites using a simple alteration of aptamer AS1411 to tag tumor cells. The specific mechanism of uptake of AS1411-GQDs, however, was unknown and was found that it was similar to that of AS1411. Fan et al. [204] glanced into GQDs-TPP as a specific target agent for mitochondria imaging, expecting the lipophilic TPP to accumulate inside mitochondria for targeted imaging. Mitochondrial and nuclear imaging were performed with no adverse effects, as expected [204]. GQDs are undeniably promising in a variety of applications today, as noted; nonetheless, it is necessary to assess the negative consequences of GQDs in order to ensure biosafety before being used in medical applications. In the study of toxicology, SEIRA has proven to be a highly effective instrument for assessing the connection between GQDs and their environment [204]. Table 5 shows cytotoxic studies of GQDs against various cell lines.

**3.4. Antimicrobial Activity.** One of the most persistent risks to humanity’s well-being is the growth of antibiotic-resistant microorganisms [46]. Functionalized GQDs have been discovered to be an efficient antibacterial substance (Table 6). The huge  $\pi$ -conjugated system present in GQDs can interact with the cell wall of bacteria by  $e^-$  transfer, exacerbating membrane stress, and the generation of ROS, that eventually causes cell death [226, 205]. Luo et al. [190] reported a microwave-assisted method to prepare A-GQDs and the antimicrobial effect of A-GQDs against *E. coli* was demonstrated using the propidium iodide (PI) stain by white light irradiation [190]. Kashani et al. [46] discovered that the Ag@S-GQDs nanocomposite outperforms AgNPs and S-GQDs in antibacterial activity, with MIC levels as low as 35 and 70 mg/ml, respectively, to resist the growth of *S. aureus* and *P. aeruginosa*.

**3.5. Drug Delivery.** GQDs having graphene’s nanoscale layered structural motif are rapidly expanding as a prospect for designing and implementing efficient drug delivery systems, which include theragnostic drugs [209]. GQDs have been

TABLE 5: Cytotoxicity study of GQDs.

S. no.	Source	Method	Cell lines	Assay	Cell viability	References
1	GO	Solvothermal method	MG-63 MC3T3	MTT	>80%	[205]
2	CA and 3-mercaptopropionic acid	Pyrolysis Ag@S-GQDs	HEK 293	MTT assay	Reduction in cell viability by 21.4%, 1.54%, and 0.35%	[190]
3	GO	Oxidation method	THP-1	WST 1	Cell viability reduced by 20%	[206]
4	Carbon fiber	Hydrothermal method	KB, MDA-MB231, A549 MDCK	MTT, LDH	>95%	[207]
5	GO	Photo-Fenton reaction	MGC-803 MCF-7	MTT	GQDs < GO	[208]
6	CA	Carbonization	Saos-2	MTT	45.4%	[209]
7	Graphite powder	Oxidative cutting method	HeLa cells and A549 cells	WST-1	>95	[210]
8	GO	Hydrothermal	A549	MTT	>80%	[211]
9	Pyrene	Hydrothermal	4T1 cells	CCK-8	>90%	[212]
10	Aspartic acid	Pyrolysis	SW 480	CCK-8	Low cytotoxicity	[213]
11	Rice grains	Pyrolysis	HeLa cells	SRB assay	90%	[52]
12	Neem leaf	Hydrothermal method	HeLa, MCF-7, and MCF-10A	MTT	>95%	[89]
13	GO	Hydrothermal method	HeLa cells	CCK-8	>90%	[214]
14	Graphite	Hydrothermal	THP-1 macrophages	MTT	82.5%	[215]
15	Graphite rods	Electrochemical oxidation	SH-SY5Y	MTT	NA	[216]
16	GO	Microwave	HeLa	MTT	87%	[217]
17	GO	Solvothermal	RBC	MTT	Low cytotoxicity	[218]
18	Coffee grounds	Hydrothermal cutting	HeLa cells	MTT	>88%	[88]
19	Grape seed extract	Microwave assisted	L929 cells	MTT	Sensor applications and nucleus imaging	[90]
20	Graphite plate	Laser ablation	MCF-7	MTT	Real time tracking and bioimaging	[219]
21	Adenine modified	Microwave assisted	A549	MTT assay	94%	[203]
22	GO	Sonochemical and microwave heating	HeLa	MTT	Cell viability $\geq$ 90%	[220]
23	CA	Hydrothermal	4T1	CCK-8	>90%	[221]
24	Trisodium citrate	Pyrolysis	HeLa	MTT	>80%	[222]
25	Lignin biomass	Hydrothermal	RAW 264.7	MTT	>85%	[223]
26	CA	Carbonization	B16F10 and MCF-7	SRB assay	Photodynamic therapy	[224]
27	CA	Pyrolysis	ACHN	MTT	Negligible toxicity	[225]
28	Cocoa bean	Hydrothermal method	MCF-7	MTT	Low cytotoxicity	[93]

studied in a variety of applications (Table 7) due to their tuning PL, abundance of peripheral  $-\text{COOH}$  groups, thermal and chemical stability, and low cytotoxicity nature. GQDs have been used in field of biomedicine and pharmaceuticals applications like phototherapy, drug delivery, biosensors, supercapacitors, and bioimaging. Wang et al. [212] were the first to illustrate GQDs' extraordinary ability not only to deliver the anti-cancer drug DOX but also to contribute to the drug's anticancer activity against breast cancer cells. When compared to pristine pharmaceuticals, the GQDs-DOX combination was seen to utilize substitute cellular and nuclear internalization mechanisms, which improves drug delivery efficiency [104]. Remarkably, such GQDs-DOX conjugates increased DOX nuclear absorption and cytotoxicity in drug-resistant cancer

cells, indicating that combining anticancer drugs with GQDs could be able to improve inadequate anticancer drug chemotherapeutic effectiveness due to drug resistance [104, 210]. Figure 5 illustrates GQDs as nanocarrier in drug delivery.

**3.6. Phototherapy.** It is a noninvasive therapy that employs fluorescent light to treat a wide range of medical conditions. "Photodynamic therapy" and "photothermal therapy" are two types of phototherapies. During diagnostics, the targeted agent is administered to the disease part and the therapeutic agent is then photoexcited at specific wavelengths. Photothermal agent consumes near-infrared light which generate heat, which causes cell ablation [63]. When used against tumors, the main concern with conventional chemo- and radiotherapies

TABLE 6: Antibacterial study of GQDs.

S. no.	Source	Method	Microbial strain tested	Result	References
1	CA and 3-mercaptopropionic acid	Pyrolysis	<i>S. aureus</i> and <i>P. aeruginosa</i>	MIC value of Ag@S-GQDs was 35 mg ml <sup>-1</sup> for <i>S. aureus</i> and 70 mg ml <sup>-1</sup> for <i>P. aeruginosa</i>	[46]
2	MWCNTs	Hydrothermal	<i>P. aeruginosa</i> , <i>S. aureus</i> , <i>E. coli</i> , and <i>B. subtilis</i>	MICs value for <i>E. coli</i> and <i>B. subtilis</i> were found to be 512 µg ml <sup>-1</sup> . And the MICs for <i>S. aureus</i> and <i>P. aeruginosa</i> were ~256 µg ml <sup>-1</sup>	[226]
3	PEG	Pulsed laser	<i>S. aureus</i> and <i>P. aeruginosa</i>	MIC value for <i>P. aeruginosa</i> was 25 µg ml/1 And the MIC value for <i>S. aureus</i> was found to be 50 µg ml <sup>-1</sup>	[205]
4	Graphite	Microwave assisted	<i>E. coli</i>	Excellent antibacterial activity against <i>E. coli</i> Produced <sup>1</sup> O <sub>2</sub> destroy cell membrane of <i>E. coli</i> .	[190]
5	Coconut husk	Hydrothermal carbonization	<i>E. coli</i>	Antibacterial activity	[190]
6	CA	Hydrothermal method	<i>E. coli</i>	85% bacterial survival by GQDs alone 50% bacterial survival by ZnO 0% bacterial survival by ZnO-GQDs upon UV irradiation for 5 min	[54]
7	GO sheet	Ultrasonic shearing reaction	<i>E. coli</i>	Inhibits bacterial cells growth	[206]
8	Nickel oxide	Laser ablation method	<i>M. luteus</i> and <i>E. coli</i>	<i>M. luteus</i> and <i>E. coli</i> were inactivated after irradiation of 5 min	[207]
9	GO	Microwave	<i>S. aureus</i> and <i>E. coli</i>	Inhibit bacterial cells growth by generating free radical	[208]

TABLE 7: Drug delivery applications of GQDs.

S. no.	Source	Method	Drug	Drug loading	% Drug release (%)	Release time (hr)	Cell viability (%)	Assay	Cell line	References
1	GO	Photo-Fenton reaction	DOX	50 µg/ml	70	24	35	MTT	MGC-803 and MCF-7 cells	[211]
2	CX-72 carbon black	Chemical oxidation	DOX	68 wt%	NA	8	>80	MTT	A549 and HEK293A cells	[212]
3	L-glutamic acid	Pyrolysis	DOX	15 wt%	70	48	>35	CCK8	BT-474	[213]
4	GO	Thermally exfoliated	DOX	54.6 wt%	40	72	60.8	MTT	U251	[214]
5	CA	Pyrolysis	DOX	10 mg/ml	42.5	24	17	MTT	Nucleus	[210]
6	GO	Solvothermal method	DOX	101 µg/mg	60	72	45–60	CCK8	MC3T3-E1, DU-145, and PC-3	[215]
7	Oxidized graphene Sheets	Amino-hydrothermal treatment	DOX	80 wt%	12	6	>90	MTT	HeLa cells	[216]
8	MWCNTs	microwave-assisted hydrothermal	Methotrexate	10 mg/ml	60	9	~80	MTT	A549 and MCF-7 cells	[217]
9	GO	Hydrothermal	DOX	2.5 mg/ml	42	6	NA	NA	NA	[66]
10	CA	Carbonization	Sodium salicylate	3 mg/ml	74	7	NA	NA	In vivo	[218]
11	Cow milk	Microwave	BHC	0.2 mg/ml	50	24	50	MTT	MDA-MB-231, HeLa, and L929 cells	[59]

is the increase in side effects. Due to their superior qualities, much more efficient treatment activity, and fewer side effects, nanomaterials produced for PTT and PDT have shown considerable promise [219, 220] (Table 8).

PDT uses photosensitizers that, when it is activated by light, it generates reactive oxygen species that harms cells [220] (Figure 6). With the development of next-generation therapeutic molecules, phototherapy has the potential to

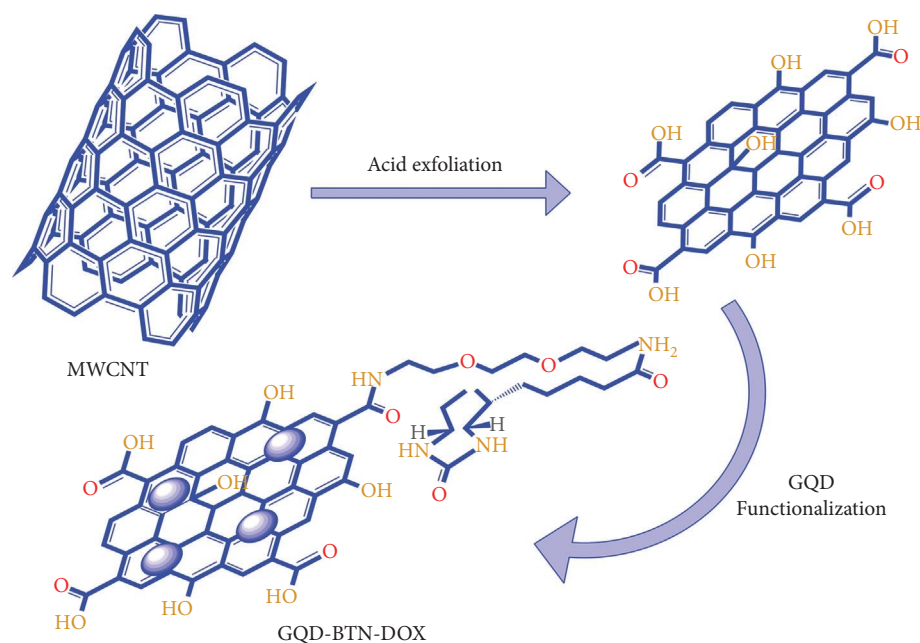


FIGURE 5: GQDs as agent for drug delivery.

TABLE 8: Phototherapy study of GQDs.

S. no.	Source	Method	Model	Finding	References
1	Graphite	Electrochemical	<i>Staphylococcus aureus</i> and <i>Escherichia coli</i>	When photoexcited at wavelength of 470 nm, it produces reactive oxygen species, and kill harmful methicillin-resistant bacteria, <i>S. aureus</i> and <i>E. coli</i>	[221]
2	Graphite	Electrochemical	<i>Escherichia coli</i>	Produce single oxygen upon UV illumination increase in photoluminescence quantum yield	[222]
3	Carbon black	Chemical oxidation	HepG-2, A549, MCF-7, and HeLa	Generates heat from light absorption which leads to death of the cancer cells	[223]
4	Polythiophene (PT2)	Hydrothermal treatment	HeLa cells	Generate $^1\text{O}_2$ via a multistate sensitization process.	[224]
5	CA	Hydrothermal	87.9% photothermal conversion efficiency	Upon 808 nm laser irradiation it completely eradicates tumors upon	[225]
6	GO sheet	Ultrasonic shearing reaction	<i>E. coli</i>	$^1\text{O}_2$ and $\text{O}_2^-$ were effectively generated	[227]
7	Chemical	Chemical	4T1 cells	Convert the near-infrared irradiation into the heat	[228]

become an optimal treatment for malignancies. The phototherapy treatment kills cancer cells and improves the efficacy of other therapeutic modalities such as radiotherapy, chemotherapy, and gene therapy. However, these of photothermal therapy enhances the efficacy of radio and chemotherapies. Furthermore, enhancing the permeability of cell membranes at tumor areas will improve medication cellular uptake and release [14, 63, 219].

**3.7. Photocatalysis.** Photocatalysis is a process in which a catalyst can greatly improve the pace of a chemical reaction in the presence of light. The main issue in photocatalysis is to first produce a catalyst which may efficiently absorb sunlight, resulting in improved chemical reactions and then to synthesize effective photocatalyst using a cheap approach. GQDs unique features such as high stability, increased

surface area, high solubility, nontoxicity, and superior conductivity, make them a viable photocatalysis and electrocatalysis material. For photocatalysis, heteroatoms such as N, S, and P can be used to optimize the characteristics of GQDs toward the sun absorption property, resulting in improved performance. GQDs have been employed in a variety of photocatalysis reactions, including  $\text{H}_2$  evolution [229–231], organic pollutant degradation [176, 232, 233],  $\text{CO}_2$  reduction [144, 234], and so on, either alone or in conjunction with other inorganic materials.

Zhuo et al. [235] devised a unique ultrasonic synthesis method for GQDs with excitation-independent up-and-down conversion of photoluminescent properties. They also created GQD composites with the anatase and rutile phases of titanium oxide and used the composites to degrade methylene blue dye in visible light. On illumination with light  $>420$  nm

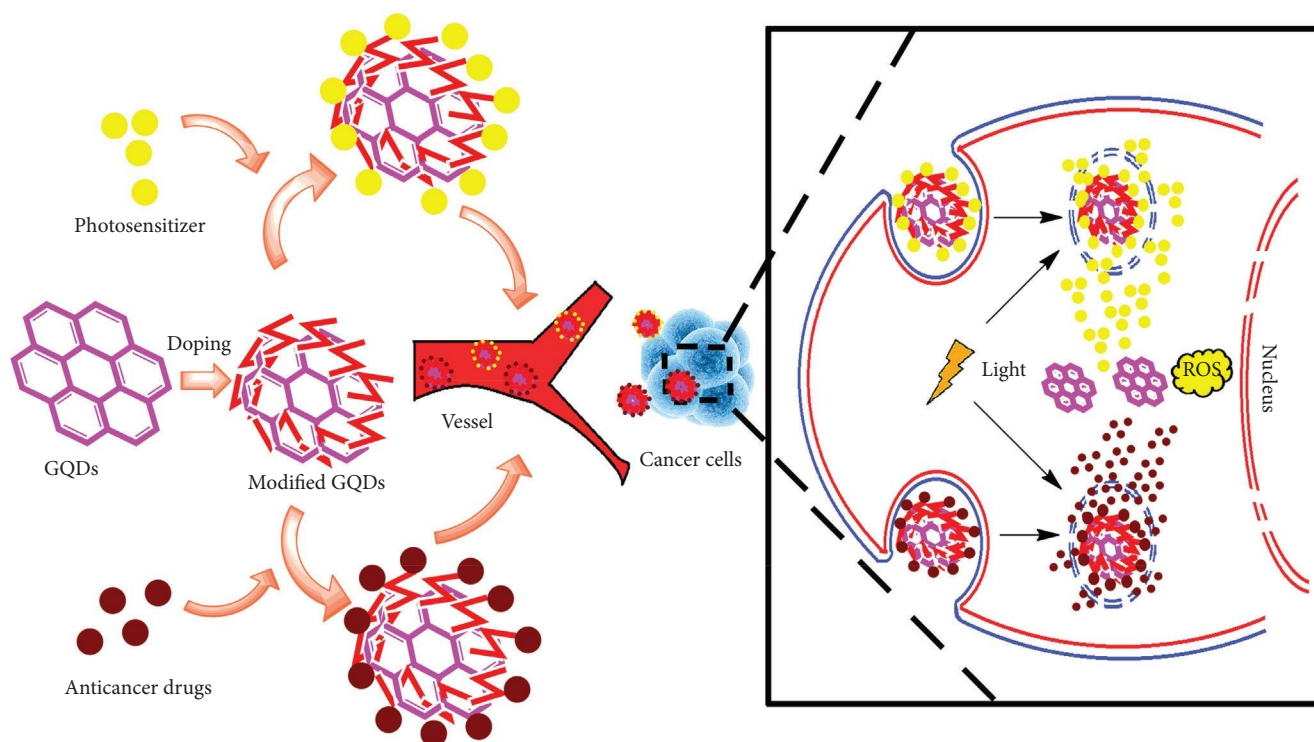


FIGURE 6: Mechanism of action for photothermal therapy.

wavelength, they observed that composite of rutile  $\text{TiO}_2/\text{GQDs}$  has a 9% higher degradation rate than the composite of anatase  $\text{TiO}_2/\text{GQDs}$  for degradation of methylene blue [236]. Using the electrostatic interaction approach, Wang et al. [212] illustrated the synthesis of hydroxyl-GQDs and the composite of hydroxyl-GQDs and mesoporous carbon nitride [240]. In the presence of visible light, the composite was found to greatly increase the photodegradation of contaminants such as Rhodamine B and tetracycline hydrochloride. The photocatalysis reaction proceeds by generating  $\text{O}^{2-}$  species as well as producing holes, according to electron spin resonance spectroscopy data. Wang et al. [237] designed a hydrothermal synthesis technique to obtain GQD-decorated ZnS nanobelts, which they then employed for Rhodamine dye photodegradation.

**3.8. Antibiotics Detection.** Antibiotics can be obtained from both natural and synthetic sources, which are used to avoid bacterial infections and divided into numerous classes of antibiotics penicillins, tetracyclines, fluoroquinolones, sulfonamides cephalosporins, aminoglycosides, phenolics, and macrolides antibiotics [176]. The common detected antibiotics by QDs fluorescent nanosensor include tetracyclines and sulfonamides to minimize accumulation in body of human by food chain which promote food safety [238]. Antibiotics have been detected using a variety of approaches based on GQDs materials (Table 9). They have been extensively researched as a fluorescence sensor for detecting a variety of compounds, including RNA [251], antibiotics [139], metal ions [252], pesticide [253], DNA [160], vitamin [254], and protein [255].

**3.9. Supercapacitor.** Huge energy consumption as a result of technological improvements and the need for energy storage are two essential concerns. Therefore, there is great potential for the accumulation of electrochemical energy systems, which opens up a brand-new area of research for both the industrial industry and academia. Electrochemical energy storage systems (EESS) which is transferred from chemical energy to electrical power frequently used to store energy. EESS, which has drawn a lot of interest due to its rapid discharge/charge rates and extended life, is a crucial prerequisite for any energy storage unit. The main component of the EESS is supercapacitor, which in the current context solves extreme caution and significant energy sources. It is also referred to as the electrochemical capacitor and offers quick discharge/charge, with long-term and high-power density. These characteristics make EESS one of the best-performing alternative materials for the use in emergency power systems, portable electronics, and electric vehicles [117].

**3.10. Challenges and Future Perspective.** Although there has been considerable success in recent years, GQDs still face significant obstacles in pollutant degradation and industrial production, necessitating further study in the future. Some of challenges faced by GQDs are discussed as follows:

- (1) Enhance the synthesis process—the large-scale output cannot be addressed by the present synthesis method
- (2) The van der Waals heterostructure should be strengthened

TABLE 9: Antibiotic detection applications of GQDs.

S. no.	Source	Method	Application	Linear range	LOD	References
1	Graphite	Hydrothermal	Detection of tetracycline	40–90 ng ml <sup>-1</sup>	45 ng ml <sup>-1</sup>	[239]
2	CA	Pyrolysis	Detection of norfloxacin	1.0–100.0 µg l <sup>-1</sup>	0.35 µg l <sup>-1</sup>	[240]
3	CA	Pyrolysis	Detection of ceftazidime	0.10–10.0 µg l <sup>-1</sup>	0.05 µg l <sup>-1</sup>	[241]
4	CA	Pyrolysis	Cefazolin detection	0.10–10.0 µg l <sup>-1</sup>	0.10 µg l <sup>-1</sup>	[242]
5	CA	Pyrolysis	Determination of sulfadiazine	0.04–22.0 µM	10 nM	[204]
6	MWCNTs	Hydrothermal	Detection of the streptomycin antibiotic	0.1–700 pg ml <sup>-1</sup>	0.033 pg ml <sup>-1</sup>	[243]
7	Poly (aminophenol)	Electropolymerization	Determination of levofloxacin	0.05–100 µM	10 nM	[244]
8	Graphene	Acid hydrolysis	Detection of ofloxacin	500–1,000 ng ml <sup>-1</sup>	10.7 ng ml <sup>-1</sup>	[245]
9	CA	Hydrothermal	Detection of sulfamethoxazole	1–100 µM	1 µM	[56]
10	Citric acid	Pyrolysis	Detection of tetracyclines	0–20 mM	8.2 nM	[246]
11	Passion fruit juice	Microwave treatment	Detection of tetracycline	0.04–70 µM	1 nM	[247]
12	CA	Microwave	determination of chloramphenicol	0.00250–0.020 mg ml <sup>-1</sup>	0.0018 mg ml <sup>-1</sup>	[82]
13	CA	Pyrolysis	Detection of levofloxacin in milk	0.10–25.0 g l <sup>-1</sup>	0.03 g l <sup>-1</sup>	[248]
14	Potato amylose	Hydrothermal	Detection of tetracycline	2.5 × 10 <sup>-10</sup> –5 × 10 <sup>-6</sup> M	9.735 × 10 <sup>-13</sup> M	[98]
15	CA	Hydrothermal	Determination of piroxicam	2.0–35.0 nmol l <sup>-1</sup>	0.11 nmol l <sup>-1</sup>	[249]
16	GO	Hydrothermal	Detection of tetracycline	1.0–104 µg·l <sup>-1</sup>	1 µg·l <sup>-1</sup>	[250]

- (3) Prevent second-hand contamination
- (4) Increase the use of GQDs in more contexts

GQDs have garnered a lot of attention in recent decades due to their qualities and use in a variety of environmental and health domains. GQD fabrication, size, repeatability, and limited quantum efficiency are all areas that need improvement in the perspective of their biomedical application. GQDs are also suited for usage in a variety of *in vivo* applications due to their low toxicity. As a result, by addressing the issue of their poor quantum efficiency by creating GQD nanocomposites by surface factorization their prospective applications in diverse domains can be expanded. GQDs as well as their synthesis methods were introduced in this work. Bioimaging, biosensors, drug delivery, gene therapy, photodynamic therapy, detection of antibiotics, and removal of dyes were among the biomedical applications of GQDs considered. The broad surface and functional groups present on GQDs allows mixing of various medicines and ligands. Therefore, GQDs are used as a nanocarrier in targeted drug delivery.

The photoluminescence of GQDs is also being exploited to bioimaging approaches for identifying different biomolecules, which could lead to a variety of new disease diagnosis tools. As a result, new ways for achieving high performance, cost-effective, and simple purification procedures that do not need the removal of raw components are required.

This review detailed recent research developments of GQDs and its composites, with an emphasis on their synthesis and biomedical applications, such as bioimaging, biosensing, drug delivery, gene therapy, photodynamic therapy, removal of dyes, and detection of antibiotics. Finally, the review suggests that further developing GQD and its composites for

numerous unresolved therapeutic usages has a bright future. The practical usage of GQDs necessitates careful consideration of chemical and electrochemical stability.

## Abbreviations

GQDs:	Graphene quantum dots
GO:	Graphene oxide
LEDs:	Light-emitting diodes
QDs:	Quantum dots
MWCNTs:	Multiwalled carbon nanotubes
CNDs:	Carbon nanodots
LOD:	Limit of detection
LR:	Linear range
CA:	Citric acid
PEHA-GQDs-His:	Pentaethylenhexamine and histidine-functionalized graphene quantum dots
AS1411-GQDs:	26-base guanine-rich short oligonucleotide
MDCK:	Madin–Darby canine kidney
MCF-7:	Breast cancer cell line
CHOek1:	Chinese hamster ovary
A-GQDs:	Adenine-modified grapheme quantum dots
Ag@S-GQDs:	Silver–sulfur doped graphene quantum dot
MIC:	Minimum inhibitory concentration
GQDs-DOX:	Doxorubicin conjugated graphene quantum dots
QY:	Quantum yield
CNDs:	Carbon nanodots
NIR:	Near infrared region
MTT:	3-(4,5-Dimethylthiazol-2-yl)-2,5-Diphenyltetrazolium bromide



HGQDs:	Highly biocompatible graphene quantum dots
Man-GQDs:	Graphene quantum dots conjugated with mannosamine
HRP:	Horseradish peroxidases
GQD-PEG:	PEGylation of GQD
GO-PEG:	PEGylation of graphene oxide
AS1411-GQDs:	Aptamer AS1411 conjugates with graphene quantum dots
TPP:	(3-carboxyl) phenyl bromide phosphine
SEIRA:	Surface-enhanced infrared absorption spectroscopy.

## Data Availability

All the data related to this study are available in the manuscript.

## Additional Points

**Highlights.** Short review on synthesis of GQDs is presented. Precursors like biomass waste, plant extracts, and biomolecules are identified for GQDs production. The cytotoxicity, bioimaging, sensors, and antibiotics detections applications have been explained. Phototherapy and its mechanism of action have been discussed.

## Conflicts of Interest

The authors declare that they have no conflicts of interest.

## Authors' Contributions

Pooja Kadyan and Rohit Malik: writing—original draft; Saurabh Bhatia, Ahmed Al Harrasi, Syam Mohan, Mansi Yadav, and Sunita Dalal: writing—review and editing, literature review; Sudhir Kumar Kataria and Thillai Arasu: conceptualization, writing—review and editing.

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