DE GRUYTER Open Chemistry 2023; 21: 20220285

Review Article

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Graphene quantum dots: A comprehensive overview

https://doi.org/10.1515/chem-2022-0285
received October 28, 2022; accepted February 1, 2023

Abstract: Because of their prospective applications and exceptional features, graphene quantum dots (GQDs) have gotten a lot of recognition as a new class of fluorescent carbon materials. One of the carbon family's newest superstars is the GQD. Due to its exceptional optoelectrical qualities, it has sparked a lot of curiosity since its debut in 2008. Two of the most important traits are a band gap that is not zero, biocompatibility, and highly changeable characteristics. GQDs have several important characteristics. GQDs have shown potential in a variety of fields, for instance, catalysis, sensing, energy devices, drug delivery, bioimaging, photothermal, and photodynamic therapy. Because this area constantly evolves, it is vital to recognize emerging GQD concerns in the current breakthroughs, primarily since some specific uses and developments in the case of GQDs synthesis have not been thoroughly investigated through previous studies. The current results in the properties, synthesis, as well as benefits of GQDs are discussed in this review study. As per the findings of this research, the GQD's future investigation is boundless, mainly if the approaching investigation focuses on purifying simplicity and environmentally friendly synthesis, as well as boosting photoluminescence quantum output and manufacturing output of GQDs.

Keywords: energy-related applications, biomedical applications, dots, graphene quantum, sensing

1 Introduction

Carbon and its allotropes have attracted the attention of physicists, chemists, as well as general biologists, and are among the primarily available and biologically essential elements. Carbon-based materials are critical for the industry's long-term growth, bring innovative solutions to our progressive community and address several of the world's most pressing health and energy problems. Carbon's modern past commenced in the mid-nineteenth century with the synthesis of graphite oxide, which was pursued by an era of intensive investigation into its structural and electrical properties [1].

As a result, Smalley [2] discovered the zero-dimensional (0D) football-shaped fullerene in the year 1985, while Iijima [3] discovered the 1D helical carbon nanotube (CNT) in the year 1991 in which both represent new as well as brilliant carbon allotropes. These breakthroughs catapulted carbon research to new heights. Smalley, Curl, and Kroto were bestowed with the Nobel Prize in the year 1996 for their fullerenes discovery, which sparked interest in carbon-based nanomaterials in business and science. For synthesizing 2D graphene sheets with a few atomic layers thickness, in the year 2010, the Nobel Prize in Physics was awarded to Novoselov et al. [4]. The amazing and one-of-a-kind properties of this new carbon family member sparked a surge of current research and development innovation. At room temperature, 2D graphene sheets have chemical stability, large specific surface area, high carrier mobility, excellent mechanical strength, and thermal conductivity. Sensors, pollutant absorbents, energy storage devices, touch screen displays, solar cells, medicine delivery, and other applications can all benefit from graphene-based materials. In addition, the honeycomb structure of graphene exhibits a zero bandgap due to the significant delocalization of electrons [5].

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This combined with graphene's low spectrum absorption and dispersibility in common solvents, greatly restricts its use in various sectors, including optoelectronics, semiconductor technology, and bioimaging [6]. In the broad family of carbon nano-allotropes, graphene quantum dots (GQD) are among the most recent superstars. Given its exceptional optoelectrical, electrical, and optical characteristics, it has drawn considerable interest following its revelation in 2008. Because of substantial edge effects and quantum confinement, GQDs differ from existing carbon materials' family members (for instance, CNTs, fullerene, as well as graphene) in regards to optical as well as physical properties [7].

In comparison to other carbon-based materials and graphene, GQDs were discovered to absorb more active sites (e.g., functional groups, edges, as well as dopants) due to their ultra-fine particle size and enormous specific surface area. Non-zero bandgap has produced enormous edge effect and amphiphilic nature, whereas high dispersibility has given the massive edge effect and amphiphilic nature. Moreover, quantum confinement is another major trait that has generated considerable obstacles between the use of graphene and GQDs. In conjunction with high surface grafting, persistent photoluminescence, biomolecule-like size, low toxicity, biocompatibility, and chemical stability, these properties facilitate some specific benefits and thus potential applicability [8].

The special properties of GQDs enable the development of high-performance electrical, photonic, and energy-related devices, for instance, photodetectors, light-emitting diodes (LEDs), rechargeable batteries, solar cells, flexible memory systems, as well as supercapacitors. In addition, GQDs have been employed in biomedical engineering (for example, chronic disease treatment, anticancer therapy, drug delivery, and bioimaging), catalysis, sensors, nanofluids, and environmental clean-up [9]. Moreover, carbon nanodots (CNDs) and carbon quantum dots (CQDs) are two more 0D luminous carbon-based nanomaterials with multiple applications. Therefore, in-depth characterization approaches should be used to resolve the morphological and structural disparity between GQDs and these equivalently structured nanomaterials.

In a nutshell, GQDs have a graphene lattice structure made up of at least one graphene sheet [10]. As a result of this, GQDs are tiny graphene fragments having various chemical moieties inside or at the interlayer fault edges. GQDs acquire their bandgap and fluorescence features from quantum confinement and edge effects. The GQDs size, as well as the conjugated-domains confinement, which are separated by layers' diverse in-plane defects, are both regulated by the conjugated-domains confinement. Enhancing the oxygen element or adding heteroatom doping in the GQDs structure generally increases fluorescence centres.

CQDs are a partly amorphous carbon sphere with substantial sp_3 bonding. On the surface, they have visible crystal lattices and chemical groups. The CQDs electronic bandgap is highly influenced by extrinsic state luminescence (e.g., vacancy defects, heteroatom doping, and surface energy traps created by structural defects) as well as intrinsic state luminescence, including as the CQDs size's quantum confinement effect. Apart from that, the photoluminescence wavelength and the CQDs' bandgap may change these nanomaterials' sizes [11]. Besides, CNDs are heavily carbonized and contain surface chemical groups. There are no crystal lattices visible in the CNDs structure. The CNDs photoluminescence behaviour is correlated to subdomain state and surface defects within the graphitic carbon core in the absence of the particle size's quantum confinement effect.

Although there are some similarities between CNDs, CQDs, and GQDs, some GQD-specific applications and procedures may not suit CNDs and CQDs, such as dimensional printing and injectable conductive hydrogels for tissue engineering applications. The focus of this review study is solely on GQDs. Insulator quantum dots and non-carbonic semiconductors have distinct optical and physical properties than GQDs. Surprisingly, GQDs have been shown to outperform other quantum systems. Dots are commonly used because of their unusual photostability, chemical inertness, as well as high solubility. Moreover, non-carbonic quantum dots are challenging to functionalize and frequently emit hazardous materials [12].

Many of these insulator quantum dots and semiconductors, however, display distinctive near-infrared (NIR) photoluminescence activity (e.g., Ag_2S and PbS quantum dots) or appealing optical characteristics (e.g., extremely high photoluminescence quantum generate [PLQY]) (e.g., CdSe as well as CdTe quantum dots). As a consequence of their small size, some may claim that GQDs are unlikely to display PLQY as significant as any of these non-carbonic quantum dots. Furthermore, unlike PbS or Ag_2S quantum dots, GQDs may not exhibit NIR photoluminescence. However, recent research has shown that heteroatom doping and surface modification can enhance the PLQY of GQDs to an acceptable level (30–70%) [13].

Furthermore, GQDs NIR photoluminescence activity has been established in several studies and is being exploited for a variety of applications, including bioimaging [14]. However, novel uses of GQDs, for instance, have been breakthroughs in the purification's ease of the PLQY and the manufacturing GQDs yield that have not been thoroughly investigated in previous research. This review article further addresses a variety of new GQD dimensions, such as GQD-based heterostructures for vertical tunnelling single-electron transistors (SET), z-scheme GQDs for photocatalysis, current breakthroughs in GQD-based imaging and therapeutic applications (Table 1).

2 Literature review

2.1 GQD's properties

2.1.1 Structural properties

The graphene-based 0D substance GQD is described as one or more graphene layers having a lateral dimension of lesser than 30 nm. In addition, due to the honeycomb graphene lattice, GQDs exhibit a high crystallinity. Every carbon atom establishes a trigonal planar orbital geometry having three other carbon atoms. Also, the presence of various structural flaws, functional groups, as well as heteroatom dopants can all influence the crystalline structure of GQDs.

To this point, GQDs comprising a variety of functional groups (e.g., amide, amine, epoxy, hydroxyl, carboxyl, and carbonyl) have been synthesized [15]. Single, double, and even multiple heteroatoms (e.g., sodium, potassium, chlorine, fluorine, selenium, sulphur, oxygen, phosphorus, nitrogen, and boron) have also been documented for various GQDs in the literature. Given a large number of dopants as well as functional groups available, the GQDs final structure is complicated and case-dependent. Furthermore, when most heteroatom dopants may establish diverse bonding configurations within these 0D nanomaterials lattice structures, the structural attributes of GQDs appear to be much more complex. As a result, the only generalization that can be made regarding the structure of GQD is that it is very crystalline. Otherwise, when determining the exact form of each GQD, the synthesis technique, functional group, dopant composition, edge geometry, as well as the final concentration of edge and surface defects should all be considered.

Table 1: Some of the substantial applications and properties of GQDs

Properties of GQDs	
1. Low toxicity	
2. High surface area	
3. Non-zero bandgap	
4. Stable photoluminescence	
5. Excellent dispersibility	
6. Tunable properties	
Application of GQDs	
1. Nanofluid	10. LEDs
2. Batteries	11. Photodetector
3. Gene delivery	12. Supercapasitor
4. Heavy metal removal	13. Bioimaging
5. Photothermal therapy	14. Drug delivery
6. Photodynamic therapy	15. Nano-coolant
7. Sensors	16. Solar cells
8. Catalyst	17. Anti-microbial materials
9. Memories devices	18. Surfactants

2.1.2 Electronic properties

Prior to delving into the GQDs' electronic properties, it is critical to first understand graphene's electronic behaviour. This is because every carbon atom in graphene possesses a trigonal planar orbital geometry having three sp_2 hybridized bonding orbitals that form stable bonds with its neighbours. Consequently, in graphene's honeycomb lattice, three of four valence electrons with respect to each carbon atoms are tightly bound to the adjacent atoms. Furthermore, in the p orbital, on the other hand, the fourth electron is still unpaired.

Since it is perpendicular to the graphene's planar structure, this undamaged p orbital can establish a half-filled covalent bond with the p orbitals of neighbouring carbon atoms. Because of graphene's peculiar electrical structure, the bandgap between the conduction band (produced by lone electrons of bonds) and the valence band (formed by electrons engaging in bonds) is zero. Due to the unpaired electrons' high mobility in half-filled connections, graphene conducts electricity like metals. While this scenario assumes a fully new 2D carbon lattice with no structural flaws, it has been demonstrated that changing the graphene's lateral dimension and introducing chemical functional groups, edge states, heteroatom dopants, including defects may alter its bandgap and electrical properties. GQDs, unlike graphene, have a semiconductor property that enable them to change their bandgap from terahertz to ultraviolet. It is crucial to note that the GQDs bandgap opening is driven by size quantization and the insertion of different edge states, functional groups, and defects into the honeycomb lattice of graphene. Various experimental and theoretical experiments have been presented to better understand the GQDs electrical properties. For example, Ponomarenko et al. [16] discovered the size quantization effects on the GQDs electrical properties in depth.

They assert that if the GQDs linear size is less than 100 nm, quantum confinement takes control and a local electrostatic confinement process replaces the charge carrier transit technique. In addition, Ji et al. [17] examined the GQDs electronic structure having lateral widths of less than 2 nm utilizing spectro-electrochemistry. As per their findings, state densities, the band gaps, and GQDs excitation binding energies are all associated with their size. Even while the real data did not match the classic Dirac Fermion model, computer models that included electron–electron and electron–hole interactions came close. The crystallographic origin of edges, in addition to size quantization, is anticipated to possess a substantial influence on GQD electrical characteristics. The bandgap opening and electrical characteristics of GQDs with armchair (AM) and zigzag (ZZ) edges were also studied by Li et al. [18]. ZZ- and AM-edged GQDs bandgap and binding energy both diminish as the lateral dimension of the GQDs is increased. The bandgap opening of GQDs is influenced by edge geometry, with ZZ-GQDs exhibiting a smaller bandgap as well as lower excitation binding energy in comparison to AM-GQDs. Edge design has a considerable impact on the electrical characteristics of GQDs, according to Ritter and Lyding [19]. Metallic electrical features were identified in ZZ-GQDs having an average dimension of 7–8 nm, which differed from the energy gap-size scaling rule, due to their metallic zigzag edge states.

As a consequence of the vacant third orbital abundance, phosphorus atoms can establish a variety of bonds inside the GQDs structure, influencing the electronic characteristics and spectra of phosphorus-doped GQDs (P-GQDs) in a variety of manners. Furthermore, when phosphorus atoms were discovered to be sp_3 hybridized, the expected HOMO–LUMO gap for P-GQDs was reported to be minimized. Phosphorus doping in tetrahedral-like structures has a greater impact on the electrical characteristics of P-GQDs, according to researchers. Pyramidal-like configurations, on the other hand, offer no regulated charge-transfer capacity during the absorption process. As a result, Qian et al. [20] proposed that phosphorus doping may be used to tune the semiconductive behaviour of GQDs.

2.1.3 Photoluminescence properties

The ability of a GQD to absorb photons, excite electrons to a greater energy level, which then release photons as the electrons return to their stable energy level is referred to as photoluminescence. Several GQDs with various photoluminescence colours in the range of ultraviolet to visible light and even NIR were synthesized to date [21]. Despite this, the literature does not agree on the photoluminescence's origin in GQDs. The surface functional groups, size, as well as GQDs heteroatom dopants are hypothesized to affect their photoluminescence. The extrinsic and intrinsic emission routes are now being used by authors to describe the photoluminescence behaviour of GQDs based on these parameters. The intrinsic emission route is centred on bandgap opening and quantum confinement in GQDs, which are controlled by the aromatic rings' quantity and size in their structure.

Extrinsic or surface-related emission, on the other hand, is associated with charge transfer between the sp_2 carbon network and surface states (for instance, chemical moieties, edge states, and heteroatom dopants). The surface functional groups' effect on GQD emission wavelength has been explored extensively. GQDs' emission intensity and photoluminescence colour alter as the surface functional groups concentration is oxidized, reduced, or modified. The pH-dependent photoluminescence activity of GQDs also illuminates the extrinsic emission route. The surface functional groups protonation as well as deprotonation changed the emission strength and, in some cases, the emission wavelength of GQDs at both low and high pH.

2.1.4 Biocompatibility and cytotoxicity

Biocompatibility is among the most critical features of GQDs for many biological applications, including drug administration, biosensing, and bioimaging. Biocompatibility refers to an agent's ability to perform its job in tissue without triggering unwanted biological responses, such as toxic reactions. The biocompatibility idea should be assessed relying on the material's specific application. Examining the cytotoxicity of a substance is one technique to determine its biocompatibility (the quality of being hazardous to cultured cells).

Furthermore, in comparison to other metal-based nanoparticles and carbon-based nanomaterials, GQDs have been shown to have decreased cytotoxicity. GQDs have excellent water stability and might be regarded as a non-toxic alternative to some other carbon nano-allotropes that display increased cytotoxicity leading from aggregation. Furthermore, due to their carbon-based architectures, GQDs eliminate hazardous metallic residues compared to metal-based nanoparticles. Several theoretical and practical studies on GQD cytotoxicity show that GQDs with small lateral widths do not damage lipid membranes in cells and can thus be used in a variety of biomedical applications [22]. Moreover, Chong et al. [23] also performed *in vivo* bio-distribution investigations on GQDs, which revealed no material aggregation in the major mice organs and rapid GQD removal via the kidney.

Additionally, research on the enzymatic breakdown of GQDs has demonstrated that human myeloperoxidase and eosinophil peroxidase are capable of degrading these nanoparticles [24]. Both enzymes have the potential to degrade GQDs in a few hours. Nevertheless, within several days of treatment, considerable degradations might be noticed. Surface functionalization and doping of GQDs having different dopants as well as functional groups may improve their biological properties. Whereas amide, amine, and carboxylic-modified GQDs have been demonstrated to have extremely little toxicity at levels up to $200\,\mu g\,mL^{-1}$, hydroxyl-modified GQDs have been proven to exhibit significant cytotoxicity at concentrations of more than $100\,\mu g\,mL^{-1}$. Moreover, reactive oxygen species (ROS) formation has long been implicated in the increased cytotoxicity of hydroxylated GQDs.

However, the latest study contradicts this theory, demonstrating that ROS production happens only when reduced cell viability [25]. As a result, the cytotoxicity of functionalized GQDs cannot be solely associated with their propensity to establish ROS. The embedded GQDs in the polyethylene glycol (PEG) matrix showed decreased cytotoxicity and greater cell survival than unmodified GQDs. The loading and intracellular therapeutic delivery of these GQDs polymer nanocomposites were also improved, making them useful platforms for chemotherapeutic delivery and intracellular imaging. Heteroatom-doped GQDs (for instance, N, S co-doped GQDs, N, P co-doped GQDs, S-GQDs, and N-GQDs) have shown to be biocompatible and cytotoxic. Notwithstanding the results, there is still a dispute on GQDs biocompatibility and cytotoxicity, and more study is necessary to establish the particular mechanism of GQDs intracellular ROS formation. Moreover, the impact of structure and different chemical moieties with respect to GQD *in vivo* toxicity as well as photo-induced ROS production should be explored deeper.

2.2 Synthesis of GQDs

There exist two types of GQDs synthesis strategies: bottom-up and top-down. In synthesizing nanoscale GQDs, bottom-up and top-down methodologies have been devised, mainly containing oxidative cleavage, electrochemical witting, and several other top-down methods, such as potassium intercalation, physical grinding, and electron beam lithography. In addition, approaches that include stepwise organic synthesis, soft template synthesis, microwave-assisted synthesis, and hydrothermal or solvothermal synthesis are described.

In the top-down procedure, bulk graphitized carbon materials (e.g., multi-walled carbon nanotubes (MWCNTs), graphene oxide, graphene, graphite, etc.) are employed as antecedents. The carbon precursors are subsequently exfoliated and sliced into required GQDs using chemical, thermal, or physical processes. The top-down synthesis pathway includes pulsed laser ablation (PLA), electrochemical cutting, and reductive/oxidative cutting. In the reductive/oxidative cutting process, proper chemicals such as reducing or oxidative agents are used like scissors to slice through bulk carbon precursors and form small-sized GQDs.

Chemical scissors are commonly used with high-temperature treatments, such as hydrothermal/solvothermal procedures or high-energy technologies like microwave and ultrasonic, to speed up oxidative/reductive processes. In electrochemical cutting, the applied electric potential drives charged ions into the precursors' graphitic layers. Then, these ions operate as electrochemical scissors, dividing GQDs from their predecessors. The PLA approach, which uses no strong acidic chemicals and hence suggests a practical and environmentally safe pathway to GQDs, is another promising top-down synthesis option.

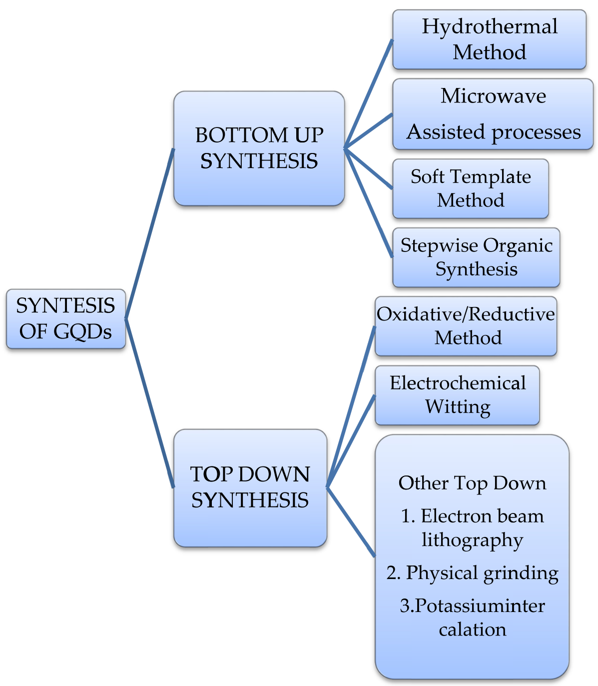
In this technique, nanosecond pulsed laser irradiation develops plasma plumes, cavitation bubbles, as well as thermal destruction of bulk carbon-based precursors [26]. Because it relies on widely available bulk precursors, the top-down technique is popular. Top-down efforts to alter GQDs features like size and brightness are progressing. GQDs with remarkable water stability and high oxygen functional groups can also be made using particular top-down techniques. Nevertheless, including heteroatoms into the structure of top-down synthesized GQDs (primarily into the basal plane) is tricky. Moreover, harsh and caustic chemicals are used in several of the most frequent top-down GQD synthesis methods, limiting their use. On the contrary to top-down methods, the bottom-up method consists fusing small precursor molecules (such as 1,3,6-trinitropyrene, glucose, citric acid, and others) into larger structures to create GQDs.

Hydrothermal, soft-template, microwave-assisted, as well as stepwise organic synthesis is the most common bottom-up synthesis methods. To crystallize and generate GQDs, the hydrothermal synthesis technique employs high-temperature and generally high-pressure solutions of small precursor chemicals [7]. Hydrothermal techniques make it very simple to induce heteroatom passivating as well as doping the chemical moieties of GQDs. Nevertheless, this method is time-consuming and costly.

As a result, various research studies have concentrated on the microwave-assisted synthesis procedure, which may efficiently minimize precursor fusion time while permitting the inclusion of heteroatoms and chemical moieties to the GQDs structure [27]. Microwave synthesis speeds up chemical reactions and enables simultaneous heating, leading to GQDs with a homogeneous distribution of particle size. The bottom-up synthesis of GQDs has recently progressed, resulting in impressive reaction control. Stepwise organic synthesis, for example, necessitates the fusing of tiny organic molecules via a sequence of organic reactions.

The soft-template technique is a relatively intriguing bottom-up synthesis strategy with great response control. This method creates nano-scale reaction chambers where a small number of precursor molecules fuse uniformly. The soft-template method produces monodispersed GQDs without the need for complex separation and purification procedures, which could lead to a slew of new GQDs industrial applications in the upcoming years. Lastly, the bottom-up synthesis method yields impressive GQDs functionalization and doping pathways. In addition, certain bottom-up approaches are significantly more accessible than most top-down alternatives. This could explain why there are so many papers about bottom-up GQDs synthesis and its applications (Table 2).

Table 2: Synthesis of GQDs



2.2.1 Bottom-up synthesis of GQDs

2.2.1.1 Hydrothermal method

In the hydrothermal process, small precursor molecules are crystallized and developed into GQDs in high-temperature, high-pressure solutions. Moreover, citric acid has been utilized as a precursor in the hydrothermal method of making GQDs since 2012. For example, Dong et al. [28] made GQDs by paralyzing citric acid directly and changing the carbonization degree. However, the GQDs produced were mainly nano-sheets with a thickness of 15 nm and a PLQY of only 9%.

In addition, Qu et al. [29] implemented a simple hydrothermal synthesis approach to synthesize N-GQDs utilizing urea as the nitrogen source and citric acid as the carbon source. Others have used a similar source material to make N-GQDs for a number of purposes [30]. Alternative nitrogen sources have been introduced to the hydrothermal technique of synthesizing N-GQDs, for instance, tris (hydroxymethyl) amino methane and ethylenediamine. Thiourea and citric acid were also used as source materials to make S, N co-doped GQDs. As a result, they possess different photoluminescence spectra than N-GQDs. Qu et al. [29] also introduce sulphur and nitrogen atoms into the GQDs structure, resulting in wider visible absorption bands and multicolour emission when exposed to visible light.

Additional hydrothermal precursors, for instance, amino acids and polycyclic aromatic hydrocarbons, were employed to make GQDs in addition to citric acid. Amine-functionalized GQDs were made by carbonizing 1,5-dinitronaphthalene in an aqueous ammonium solution, to state a few instances. In another study, pyrene was utilized to synthesize GQDs under green hydrothermal and mild conditions. Pyrene is nitrated in this technique, and the resulting 1,3,6-trinitropyrene is then hydrothermally treated in an alkaline aqueous solution.

The GQDs have a high manufacturing rate, excellent optical qualities, as well as a PLQY of 23%, which is satisfactory. For instance, Wu et al. [31] also showed that L-glutamic acid could be pyrolysed in a single step to yield highly fluorescent GQDs with NIR fluorescence in the 800–850 nm range. This NIR activity has significant implications for imaging and sensing *in vitro* and *in vivo*. Hydrothermal techniques make it very simple to induce heteroatom doping and passivate the GQDs chemical moieties. As a result, certain GQDs generated using these synthesis techniques have a PLQY of >25%, adequate for various fluorescence-based sensing applications and i maging. One drawback of the hydrothermal approach is the lengthy and expensive synthesis and purification operations. Additionally, the GQDs produced utilizing this synthesis method possess small particle sizes and negligible agglomeration. Lastly, this synthesis approach is suitable for biomedical or other applications that necessitate a small number of GQDs having a high PLQ.

2.2.1.2 Microwave-assisted method

Microwaves are known as electromagnetic waves yielding a steady stream of high-intensity energy to speed up chemical reactions. Compared to hydrothermal synthesis, microwave-assisted synthesis can drastically reduce the time it takes for precursors to fuse. This approach also allows for simultaneous heating, which results in a uniform particle size distribution in GQDs. Tang et al. [32] published the first systematic study on the GQDs microwave-assisted synthesis. They used a household microwave to irradiate an aqueous solution of glucose to manufacture self-passivated GQDs. The size and fluorescence characteristics of synthesized GQDs are directly influenced by changes in microwave power, solution volume, source concentration, as well as heating duration. The microwave-assisted technique has been employed to generate heteroatom-doped GQDs in a number of researches.

A mixture of 1,3,6-trinitropyrene and hydrazine hydrate, for instance, was utilized to make ultra-bright N-GQDs with a PLQY of 35%. A mixture of triethanolamine and sodium citrate, an aqueous solution of glucosamine-HCl [33], and a mixture of glucose and ammonia are also utilized to generate N-GQDs. Moreover, by replacing di-methyl sulphoxide for water under microwave irradiation, Jeon et al. [33] were able to synthesize N-GQDs and N, S co-doped GQDs.

The N, S co-doped GQDs and N-GQDs produced have distinct physical and photocatalytic characteristics. The influence of content, composition, and dopant type on the photoluminescence behaviour of GQDs was examined by Campbell et al. [34]. In a microwave-assisted procedure, N, S co-doped GQDs, B, N co-doped GQDs, and N-GQDs were made utilizing glucosamine as the primary precursor and benzene boronic acid or sulphur thiourea as doping agents. With quantum yields varying from 22 to 60%, the synthesized GQDs generated bright light in visible and NIR spectrums.

F-GQDs were synthesized using a microwave-assisted method employing hydrofluoric acid as the fluorine source and glucose as the carbon source in another investigation [27]. F-GQDs are commonly the essential component for producing nanomedicines among all GQD derivatives. GQDs containing a fluorine atom in their structure have improved contact and selectivity with specific cells, rendering them excellent for drug administration. As a result, synthesizing F-GQDs is critical from a medical standpoint. Future study should focus on developing novel, long-lasting F-GQD synthesis techniques that do not use hydrofluoric acid as a fluorine source.

Microwave-assisted synthesis, which is attained from quick heating, simultaneous, and homogenous is within the best options for large-scale manufacturing of GQDs. The doping and functionalization of GQDs may also be done employing this synthesis technique [34]. Nonetheless, the minimal GQDs manufactured with this technique limit their application in several industries. Furthermore, because microwave irradiation cannot penetrate most substrates deeply, the maximum size of reaction samples is limited. Essentially, the GQDs that are created using this bottom-up process may possess a low crystallinity. Also, some studies have reported synthesizing CQDs using the same methods described in this section. To assure the GQDs synthesis with outstanding crystallinity, the power as well as microwave heating duration should be accurately regulated in this process. The crystalline structure of nanoparticles created utilizing the microwave-assisted technique has to be investigated further.

2.2.1.3 Soft-template method

The soft-template method produces nanoscale reaction chambers where a small number of precursor molecules fuse uniformly [13]. Without the need for time-consuming separation and purification operations, this approach provides monodispersed GQDs. The soft-template method is perfect for producing low-cost GQDs on a big scale. In comparison to microwave and hydrothermal-assisted synthesis, the soft-template method provides more control over GQD growth and nucleation. The GQDs created by this technique, on the other hand, have a higher inclination to aggregate caused by the low concentration of surface functional groups and stacking. Alternatively, Li et al. [35] used a soft-template technique using the template and lone precursor 1,3,5-triamino-2,4,6-trinitrobenzene (TATB) to make N-GQDs. Using the recommended process, which involves symmetric intermolecular carbonizations of TATB, GQDs are pyrolysed, nucleated, and generated uniformly. The TATB was initially annealed using a thermal breakdown technique, which resulted in chemical bonds shattering and expanding gases forming. The multilayers of graphitic-like TATB were subsequently ruptured, producing single-layer graphene sheets. Ultimately, oxidative exfoliation was employed to create N-GQDs. In a different research, Dong et al. [28] used a combination of microwave-assisted and soft-template synthesis approaches to develop a novel method for making GQDs. Under microwave radiation, a small number of glucose molecules were uniformly carbonized, nucleated, crystallized, and developed in each template. As a result, self-surface passivated crystalline GQDs were ultimately made possible. It must be highlighted that the GQDs diameter generated was said to be easily controlled by modifying the heating duration alone, without the requirement for a complicated purification or separation method.

2.2.1.4 Stepwise organic synthesis

The solution chemistry technique, also defined as stepwise organic synthesis, includes a series of organic reactions fusing small organic molecules. This method gives the GQDs much control over their ultimate structure, size, and photoluminescence capabilities. Furthermore, Zheng et al. [36] revealed that the stepwise organic approach could produce GQDs with specific sizes and shapes via the oxidative condensation of aryl groups of polyphenylene dendritic precursors. This approach has synthesized nitrogen-doped and functionalized GQDs in a few intriguing investigations. Nonetheless, the stepwise synthesis method is plagued by time-consuming and overly intricate reactions. Because the nanoparticles created using this process are primarily stable inorganic solvents, their uses in domains like environmental remediation and biological applications are severely limited. Together with the lack of scalability potential, these drawbacks suggest that for GQDs, the stepwise organic synthesis strategy is no longer a feasible synthesis route.

2.2.2 Top-down synthesis of GQDs

2.2.2.1 Oxidative/reductive cutting

Oxidative/reductive cutting, which employs bulk graphitized carbon-based materials including graphene oxide, coal, carbon fibre, graphene graphite, as well as carbon black to generate GQDs, is one of the most prevalent methods. H_2O_2 or oxidative (e.g., dimethylformamide [DMF], ammonia, alkyl amines, and hydrazine) or reducing agents (e.g., DMF, ammonia, alkyl amines, and hydrazine) or oxidative (e.g., DMF, ammonia, alkyl amines, and hydrazine) or oxidative (e.g., DMF). Using higher temperatures in a hydrothermal/solvothermal approach or high-energy technologies like microwave and ultrasonic to accelerate oxidative/reductive processes has been demonstrated to be highly efficient.

These modified oxidative/reductive cutting methods have previously been classified as several synthesis techniques, such as the microwave/ultrasonic-assisted technique or the bottom-up hydrothermal/solvothermal technique in earlier research. All of the efforts in this sector were studied because all of these tactics are based on reductive or oxidative processes. Strong acids are widely utilized as oxidants to break down carbon–carbon bonds in large graphitized carbon-based materials. The solution is purified through centrifugation and dialysis after excess acid is neutralized. To create GQDs, Chen et al. [28] were among the first to apply graphene oxide as a precursor and a mixture of H_2SO_4 and HNO_3 as an oxidative agent. Note that strong acidic media and intercalating reagents were utilized to cut through diverse carbon sources, including SWCNTs, MWCNTs, C60, graphite nanoparticles, carbon fibres, and coal. Coal is one of the most acceptable possibilities among these ancestors because of its different framework as well as structure cost-effectiveness.

Coal is composed of graphitized carbon domains joined by amorphous carbon bridges efficiently cleaved by strong oxidants. Strong acids almost always form GQDs with a significant number of oxygen-containing groups (e.g., epoxy, carboxyl hydroxyl, and carbonyl) and surface defects developed during the oxidative processes. As a result, these water-soluble GQDs can be used for drug administration, heavy metal scavenging, sensing, as well as bioimaging. Strong acids, on the other hand, are a major environmental problem and demand a lengthy and complex purification process. Therefore, when utilizing solid acids to synthesize GQDs, large-scale manufacture of these 0D carbon-based nanomaterials is impracticable. Moderate oxidants like H_2O_2 and O_3 have also been used to exfoliate GQDs. These gentle oxidants are less harmful to the environment than solid acids, and they do not necessitate nearly as many purification steps.

The hydrogen peroxide solvothermal reaction with expandable graphite in the DMF solvent has also been described as a source of water-soluble GQDs. During the whole preparation process, this synthesis technique reveals remarkably low impurity and removes the need for a time-consuming purifying phase. Another study used hydroxyl radicals created by H_2O_2 catalytic interaction with tungsten oxide nanowire catalysts to cleave graphene oxide nanoparticles into GQDs. With a simple centrifugal separation, the catalyst was eliminated following the hydrothermal treatment, and the resulting suspension of GQDs required no further purification. Unexpectedly, the manufactured GQDs' claimed production yield and PLQY were 77.3 and 69.3%, respectively, significantly greater than most past findings. Shim et al. [37] also demonstrated the use of an oxone oxidant to synthesize GQDs from diverse carbon sources (e.g., charcoal, carbon fibres, MWCNTs, and graphite). This new synthesis approach eliminates the need for a lengthy purification process to remove a significant amount of salt created during the strong acid neutralization. This research also provided a method for recovering the initial carbon-based precursors, which significantly increased the recommended process' manufacturing output. Wen et al. also described a method for making green GQDs that used graphene oxide as the major precursor and an oxidant combination of H_2O_2 and O_3 as the oxidant. They mixed an aqueous suspension of graphene oxide with H_2O_2 solution in a quartz tube and purged it with O_3 under ultrasonic irradiation. The large amount of carboxyl groups found in the synthesized GQDs gave them a distinct advantage in metal ion electrochemical detection. In addition to oxidative cleavage, some ongoing studies have been conducted on the reductive cutting of large graphitized carbon-based materials.

To manufacture surface passivated GQDs from graphene oxide nanoparticles, Bacon et al. [38] employed PEG diamine as a passivation agent and hydrazine hydrate as a reductant. Frayed graphite was also cut amidatively with a mixture of oleylamine, 1-octadecene, and hydrazine hydrate to produce GQDs. Simply modifying the amine concentration can alter GQDs photoluminescence behaviour as well as size generated. To create amino-functionalized GQDs, another study used a moderate amino-hydrothermal treatment of graphene oxide utilizing ammonia solution. Note that the amino-functionalized GQDs in this study had a PLQY of 19–29% and a few carboxylic and epoxide groups that may behave as non-radiative electron–hole recombination sites.

The oxidative/reductive synthesis of heteroatom-doped GQDs has only been described several times in the literature. From one study, graphene and reduced graphene oxides were solvothermal treated in a DMF solution to produce N-GQDs [39]. Moreover, Zuo et al. [40] synthesized F-GQDs utilizing cotton as the carbon source, which is very interesting. Degreasing cotton was annealed at high temperatures to make carbon fibres in this experiment. Hydrofluoric acid was used to hydrothermally treat the generated fibre, resulting in fluorine-doped carbon fibre. The fluorine-doped carbon fibre was ultrasonically treated in *N*-methyl-pyrrolidone solvent to produce the F-GQDs.

When compared to the bottom-up synthesis processes mentioned in the preceding sections, the reductive/oxidative cutting process is one of the worst synthesis strategies for generating heteroatom-doped GQDs. Moreover, because cleavage of the bulk materials occurs at poor intercalation sites that are randomly dispersed throughout the bulk carbon-based precursor's structure, the reductive/oxidative cutting strategy does not permit for precise control over the edge and size states of synthesized GQDs. Despite these disadvantages, oxidative/reductive cutting could open the way for large-scale GQD manufacture if further study can provide a straightforward purification technique and environmentally acceptable synthesis.

2.2.2.2 Electrochemical cutting

For creating high-quality and uniform-sized GQDs, electrochemical cutting of bulk carbon materials (MWCNTs, coke, wood charcoal, carbon fibre, reduced graphene oxide, graphite, including graphene) is advantageous. By applying an electric potential to precursors, charged ions are pushed into the graphitic layers of bulk carbon materials. Apart from that, these guest ions also act as electrochemical scissors, cutting carbon–carbon bonds and allowing nanoscale GQDs to develop.

The electrochemical synthesis technique is specifically exciting compared to typical oxidative/reductive cutting processes since it eliminates the requirement for harsh chemical oxidants. By changing the utilized electric potential as the driving force of intercalation and exfoliation, electrochemical cutting allows for selective oxidation of precursor materials and adjustable characteristics in final products. Moreover, Li et al. [41] were the first to employ an electrochemical technique to make GQDs. Using a thin graphene sheet as the working electrode, they electrochemically cut GQDs in a 0.1 m phosphate buffer solution. Thus, Lee et al. [42] have recorded GQDs electrochemical generation of employing MWCNTs thin layer as a supporting electrolyte and $LiClO_4$ (3 mm) as a supporting electrolyte with propylene carbonate as a supporting electrolyte.

GQDs were made by applying a thin coating of MWCNTs on a glassy carbon electrode and then exposing it to an oxidation–reduction process. Because graphene and MWCNT precursors are so expensive, recent research into the electrochemical synthesis of GQDs has concentrated on low-cost, highly likely starting materials, including carbon fibres, coke, and graphite. Ahirwar et al. also employed graphite rods as a precursor and a supporting electrolyte of alkali hydroxide and citric acid in water to create GQDs. The graphite rod was heated to 1,050°C before electrolysis to create additional flaws in the graphitic layers and expedite the exfoliation process. Changing NaOH concentrations in the electrolyte has led to several GQDs formations with multiple oxygen functional groups as well as optical characteristics. Graphite rods electrolysis in various electrolytes, for instance, NaOH/ethanol solution, sodium methoxide aqueous solution, as well as NaOH aqueous solution, has also yielded high-quality GQDs.

Increased manufacturing provides large-scale GQDs synthesis, which has emerged from recent insights into the GQDs electrochemical cutting. To enhance GQD production yield, Huang et al. [43] devised an electrochemical approach based on a weak electrolyte. In their study, they electrolysed graphene paper in both weak (ammonia solution) and strong (NaOH solution) electrolytes. Also, the number of GQDs created in the weak electrolyte was nearly 28 times that of the strong electrolyte. Potential graphene paper electrolysis process in both weak and strong electrolytes. The graphitic sheets of the precursors detach fast given the robust electrolyte system's intensive electrochemical exfoliation, culminating in a poor manufacturing yield. The weak electrolyte's production yield is boosted by the delayed oxidation process and lower intercalation (up to 28 wt%).

Deng et al. [44] created a system that used two platinum sheets as electrodes and graphite oxide water dispersion as an electrolyte and source material, in contrast to classic electrochemical techniques that utilized massive carbon materials as electrodes. Water is electrolysed to produce oxygen and hydroxyl radicals in this technique. GQDs are formed when these radicals slice through the graphite oxide structure. The recommended method not only increases GQD output by 65.5%, but also removes the need for raw material pre-treatment in the fabrication of carbon-based electrodes. Researchers used a graphite rod as the starting material and an aqueous glucose solution as the supporting electrolyte to produce GQDs using a microplasma-assisted electrochemical approach.

The microplasma discharge isolates glucose and water molecules in the electrolyte by reacting with exfoliated nanoparticles, resulting in hydrogen, oxygen, and carbon atoms forming surface-passivated GQDs. This breakthrough makes it possible to change the photoluminescence spectra from blue to red using a programmable manner. The electro-chemical cutting process is used here to give exceptional functionalization and doping capabilities for GQDs. The electrochemical approach can be used to make heteroatom-doped and functionalized GQDs in two ways. When making GQDs, the initial step is to dope and functionalize the precursor materials. In the second method, supporting electrolytes are used to further functionalize or dope exfoliated GQDs. Electrolysis of a graphite rod in a solution containing NaOH, Na_2S, and sodium phytate produced P, S co-doped GQDs [35]. Electrochemically cutting graphite rods in aqueous NaOH solutions with semi-carbazide as a supporting electrolyte has also been claimed to make N-doped GQDs.

2.2.2.3 Other top-down approaches

Other top-down approaches (e.g., PLA, potassium intercalation, physical grinding, as well as electron beam lithography) have been designed for creating nanoscale GQDs in addition to oxidative/reductive and electrochemical cutting processes. However, with considerable success, the PLA approach has been utilized to create excellent GQDs from graphene oxide, carbon nano-onions, MWCNTs, and graphite. Kang et al. [45], for instance, presented a quick PLA method for synthesizing perfect GQDs with homogenous shapes and sizes using MWCNTs as a precursor. With a moderate PLQY of 12%, the as-prepared GQDs emitted a noticeable blue emission.

Laser ablation of graphene oxide in diethylenetriamine produced N-doped GQDs having an average diameter of 3.4 nm and a nitrogen-to-carbon atomic ratio of 26% (DE-TA). The photoluminescence intensity, nitrogen-to-carbon atomic ratio, as well as generated GQDs' C–N bonding topologies can all be affected by changing the diethylenetriamine concentration throughout the synthesis process. GQDs were also made by physically grinding graphite in an ionic liquid. Note that the GQDs diameters created varied from 9 to 29 nm, with a production yield of 20%, which was considered satisfactory. This approach proved it was unable to control the size and number of GQD lateral layers, despite the fact that it could synthesize pure GQDs with almost no oxygen-containing functional groups. Another top-down approach worth mentioning is electron beam lithography. The research group of *Novoselov* and *Geim* was the first to use this technology to make GQDs [36]. This procedure necessitates a large amount of expensive machinery and produces just a tiny number of GQDs. Nonetheless, electron beam lithography opened the way for GQDs, a sector that has seen a lot of study and development in recent years. Li et al. [43] utilized the potassium intercalation method to create high-quality GQDs having low oxidation and high yield. MWCNTs and graphite have potassium atoms introduced into their graphitic layers. Furthermore, the energy released by potassium atoms interacting violently with water was employed to cleave bulk materials into GQDs.

PH-assisted selective sedimentation, enzyme catalytic oxidation of graphene oxide, catalytic cracking of fullerene, and diamond-edge-induced nanotomy were other synthetic techniques employed to generate GQDs. Nonetheless, these approaches are constrained by time-consuming, expensive, and intricate reaction processes and low production yields, which are not widely used.

2.3 Applications of GQDs

2.3.1 Energy-related and electronic applications

2.3.1.1 Solar cells

Due to their promise to provide a clean and sustainable energy source, solar cells, which are platforms for converting light energy to electricity, have piqued the interest of both academics and industry. Furthermore, charge carriers, for instance, hole pairs and electrons are created in light-absorbing materials and separated by a conductive contact that conducts electricity in a solar cell device [37]. As a result, GQDs can boost the efficiency of a wide range of solar cells. This is due to its distinguishing characteristics, which include intense UV absorption, powerful photoluminescence, outstanding down conversion properties, the ability to tolerate a wide range of electron-withdrawing or electron-donating moieties, as well as a customizable bandgap.

Because of their unique electrical transfer capabilities and band topologies, GQDs have mostly been utilized as an electron–hole transport layer in various solar cells. Furthermore, in a variety of studies, these 0D nanomaterials have been used as the energy down shift layer of various photovoltaic devices or as an active layer additive in dye-sensitized and organic solar cell donor/acceptor blends. GQDs-inorganic hybrid photovoltaic devices have been and continue to be a vital GQDs-based solar cells component for a long time. Hybrid silicon/GQDs solar cells have caught researchers' interest because of GQDs' remarkable light absorption and photon-down conversion capabilities, as well as silicon's lower rates and benign nature. With a perchloroethylene (PCE) of 12.35%, Ganganboina et al. [46] constructed a highly efficient GQDs/silicon heterojunction.

They expressed that the GQDs layer acts as both a hole-transport and an electron-blocking layer at the anode, enabling for more effective separation of photo-generated electron–hole pairs. Lee et al. [47] increased the PCE by 2.7% by using a GQD energy downshift layer on the surface of a crystalline silicon solar cell. This study's findings emphasize the need to utilize kinetic spray coating to establish a thin and homogenous layer of GQDs on solid substrates like silicon. Other hybrids of GQDs having inorganic materials, e.g., semiconductor quantum dots, TiO_2, and ZnO, have been reported to develop the PCE of optimized solar cells in addition to silicon.

For instance, Hasan et al. [48] constructed a solar cell with good performance using ozone-treated nitrogen-doped GQDs (Oz-NGQDs) and TiO_2 thin films. The oxygen-containing functional groups of Oz-NGQDs are improved by treating N-GQDs with ozone, resulting in improved charge transfer between the GQDs active layer and the TiO_2 active layer in the device state. Perovskite solar cells have also been shown to benefit from GQDs. GQDs aid charge extraction at the anode and cathode interfaces, reducing charge recombination considerably in these solar cells.

Furthermore, it has been demonstrated that GQDs in flexible perovskite solar cells improve perovskite flexible film crystallization and hole extraction in the structure. In addition, for flexible perovskite solar cells, a combination of GQDs and SnO_2 was employed as an efficient electron transport layer. Compared to pure SnO_2, the produced GQDs/SnO_2 composites showed greater electron mobility, greater energy level alignment, as well as enhanced film coverage. The PCE of the perovskite solar cell incorporating GQDs/SnO_2 composites was 17.7%, and the mechanical durability was outstanding, with nearly 91% of the baseline PCE value remained after 500 bending cycles.

An electron-donating phenylenediamine was employed to functionalize N-GQDs having a narrow bandgap between the $CH_3NH_3PbI_3$ and TiO_2 layers of a perovskite solar cell in another research, leading to improved charge collection. They found that adding N-GQDs to perovskite solar cells increases photo-generated electron separation and transport from the $CH_3NH_3PbI_3$ absorber (up to 6-fold). Improved organic solar cells have previously used amino-functionalized GQDs as cathode interfacial layers (CIL). The CIL that was created effectively reduced the cathode's work function, increased their built-in potential, as well as reduced the organic solar cells' series resistance (Rs). GQDs were also employed in flexible solar cells to improve the form of polymer domains. Sung et al. [49] utilized GQDs to create bulk heterojunction polymer/fullerene polymer-based solar panels. When GQDs are used as the third component in a polymer-based solar cell system, the crystallinity of the polymer domains may be changed. As a result, more effective photon conversion and charge transfer may be performed, potentially resulting in higher power conversion efficiency. Gebreegziabher et al. [50] developed a polyaniline-GQDs (PANI/GQDs) nanocomposite by polymerizing PANI in the presence of GQDs *in situ*, owing to the interaction between the conjugated structure of PANI and the electrons of GQDs. The active layer of photovoltaic cells made of this plastic-based nanocomposite system showed good light absorption, electrical, and thermal properties.

2.3.1.2 Photodetectors

Photodetectors with cutting-edge technology are at the heart of a wide range of developing technologies, which include missile approach warning systems, environmental monitoring, flame detection systems, optical communication, and real-time monitoring, to list a few. As per experimental research, photodetectors can gain from the unique advantages of GQDs, such as uncommon ease of functionalization, photostability, non-zero bandgap, and excellent light absorption characteristics. To date, GQDs have been utilized as photosensitizers in a variety of photodetectors, either alone or in combination with standard semiconductors. Additionally, Zhang et al. [51] presented a GQD-based deep ultraviolet (DUV) photodetector with good performance and low cost. This device demonstrated good photoresponsivity utilizing asymmetric Ag–Au electrodes at a comparatively low light intensity of 42 μW cm^{-2}.

It is worth mentioning that GQDs' absorption peak appears in the ultraviolet range in regards to the π–π* transition. As a result, most scientists believe photodetectors with pure GQD active layers can only detect short wavelengths (DUV photodetectors). On the other hand, current research has shown that adding heteroatom dopants and chemical moieties to GQDs can successfully extend their absorption wavelengths. Hence, pure GQDs may be utilized to create broadband photodetectors. N-GQDs, for example, were used to create a photodetector that covered the DUV to NIR spectral range. Apart from that, they attributed the broadband absorption of the novel photodetector to the layered structure of N-GQDs, which comprises a massive conjugated system with widespread delocalized π-electrons.

Below is a description of N-GQDs' broadband emission (which consists of the DUV, NIR bands, and visible). During the electron transition, localized electrons in double bonds (mostly C═C) were presumed to absorb DUV and create an electron–hole pair (exciton). DUV light (radiative recombination after vibration relaxation) or visible light can be emitted by the exciton (via inter-band transition from a more extraordinary conduction band to a smaller conduction band). Moreover, partly conjugated electrons in N-GQDs were predicted to absorb visible light and release it as excitons. In the end, the conjugated electrons in N-GQDs' layered structure were expected to aid with NIR absorption. Since the wavelength of the created NIR light was longer than the wavelength of the absorbed NIR photons, vibration relaxation was detected in this circumstance. Because of their massive layered structure with more conjugated electrons, only N-GQDs greater than 3 nm were capable of absorbing and generating NIR light.

To improve the performance of the photodetectors, GQDs were combined with other classic semiconductors, for instance, MoS_2, GaN, WSe, Si nanoparticles, and ZnO. When contrasted to a pure monolayer WSe_2 photodetector, a monolayer hybrid structure of WSe_2 covered with N-GQDs was found to possess 480% greater photoresponsivity. Outstanding performance is justified by excellent light absorption and electron transport from N-GQDs to WSe_2. Fascinatingly, this photosensor maintained good stability even after 30 days of exposure to ambient conditions.

Another study created a high-performance GQD-decorated ZnO nanorods/GaN film for UV monitoring. GQDs operate as electron donors in this heterojunction, enabling the active carrier concentration to grow. Furthermore, high-performance photodetectors were made using GQDs compressed between 2D graphene sheets. Because of its great carrier mobility and mechanical strength, graphene is good for photodetectors. However, its gapless nature and low light absorption make this material have low photoresponsivity. As a result, combining 2D graphene sheets and GQDs to create high-performance photodetectors has inspired a lot of interest in overcoming the limitations stated previously. A sandwich structure composed of multilayers of GQDs and graphene sheets, for instance, has been found to possess high directivity and photoresponsivity over a wide spectral range (from UV to NIR). Charge carriers are supposed to tunnel via the energy states of GQDs, resulting in this photosensor's remarkable performance. In addition, it has gotten more adaptive and stable recently.

The DUV photodetector was created using layering GQDs between graphene sheets on PETs/3-aminopropyl triethoxysilane (APTES) substrates [51]. The APTES was inserted between the bottom graphene layer and the PET substrate to improve the desired structure's mechanical flexibility. After 1,000 bending cycles, this unique photodetector retained roughly 87% of its initial photoresponsivity.

2.3.1.3 Supercapacitors and batteries

Over the last three decades, there appears to have been a surge in the market for energy storage devices due to the introduction of innovative renewable energy technology. For instance, supercapacitors and batteries for future energy systems that are more ecological. They appear to be equivalent, though, because they both store and emit electrical energy. Furthermore, they are capable of storing energy in a variety of ways. Supercapacitors, on the other hand, store energy directly in an electric field, whereas capacitors do it indirectly. In addition, chemical reactions are used to store potential energy in batteries. Regardless, both of these technologies are said to be beneficial from GQDs.

Due to their versatility in creating diverse nanocomposites and a large specific surface area that enables for more reactions between active materials and electrolytes, GQDs have been suggested as a viable material for supercapacitors. Furthermore, due to surface functional groups, the substantially increased pseudo-capacitance of GQDs is expected to promote supercapacitor performance (for instance, hydroxyl, carboxyl groups, and amino), dopants, and edge effect. For example, Jia et al. [52] constructed a high-performance supercapacitor using GQDs/MnO_2 heterostructure.

Due to the produced electric field, the potential window and specific capacitance of the GQDs/MnO_2 electrode were reported to be 1.3 V and 1,170 F g^{-1}, respectively. GQD-based micro-supercapacitors are recommended as a driving source for microelectronic devices due to their high power density and cycling stability. As a result, a highly flexible and transparent micro-supercapacitor was created using an interdigitated graphene pattern generated with GQDs.

Electrophoresis was employed in another investigation to implant N, O co-doped GQD on the gold electrode of a micro-interdigital supercapacitor's finger. The N, O co-doped GQD micro-supercapacitors' volumetric capacitance, high loading density, and cycle stability were all improved. N, O co-doped GQDs film chemical structure was investigated using XPS. Note that the pseudo-capacitance performance and surface wettability of GQD-based micro-supercapacitors were improved by increasing the amount of N (17.8%) and O (21.3%) in the film. The bonding topologies of nitrogen atoms in the N, O co-doped structure.

GQDs were studied to understand the manufacturing procedure of micro-supercapacitors better. Pyrrolic nitrogen can conduct reversible redox reactions in an aqueous acid media, providing excellent pseudo-capacitance for micro-supercapacitors is a nitrogen substitution in the graphene that improves the GQDs film's charge transfer. However, graphitic nitrogen is a type of nitrogen found in graphite as a substitution for graphene. As a result, the GQDs film's charge transfer has been enhanced.

Highly flexible all-solid-state supercapacitors can be produced by adding GQDs and doped-GQDs into the supercapacitor architecture. Hsiao and Lin [53] also described a simple acid treatment method that uses a combination of HNO_3 and H_2SO_4 acids to make extremely flexible carbon fibre-based all-solid-state stand-alone supercapacitors with no additional active components. To synthesize GQDs from amorphous carbons in the framework of carbon fibres, they used an accurate acid ratio to add essential functional groups and enhance the carbon fibres specific surface area.

They may be employed in the construction of wearable devices since the manufactured stand-alone supercapacitors created with this technique are highly stable at a variety of bending degrees and after over 100 times repeated bending up to the bending angle of 180°. Furthermore, GQDs in the backbone of these carbon fibre flexible supercapacitors can operate as both the active material and current collector. As a result, these structures can achieve excellent electrical conductivity and energy storage without employing any extra active material.

More fascinatingly, these structures can attain 100% Coulombic efficiency after 5,000 charge/discharge cycles. Moreover, GQDs may supply express lanes for ion diffusion and quick charge transfer in Li and Na batteries due to their vast surface area open to the electrolyte and capacity to facilitate multiple active sites [54].

GQDs may also aid the electrochemical properties of these batteries while also limiting their structural growth. A blend of GQDs and sulphur was used to create a new generation of high-performance Li–S cathodes. GQDs with oxygen functional groups were identified to boost Li^+ ion transport while minimizing active material loss and improving Li–S battery cycling performance. In a $Li_4Ti_5O_{12}$ based Li-ion battery that lasted for an extended period, GQDs were employed as the charge transport layer.

GQDs have also been used in the development of all-solid-state and wearable batteries. More recently, Liu et al. [55] created a 3D N-GQDs/$NiCo_2S_4$ nano-array for flexible Zn–air batteries as a bifunctional catalyst. As the air cathode of a flexible Zn–air battery with improved electrochemical features, the scientists used N-GQDs/$NiCo_2S_4$ nano-arrays produced on carbon cloth substrates. The existence of bifunctional sites inside the structure of N-GQDs/$NiCo_2$ electrodes, as well as the synergistic coupling effects between N-GQDs and $NiCo_2S_4$, may be credited to the device's superior electrocatalytic activities in both oxygen reduction, as well as oxygen evolution reactions, including its long-term durability. Furthermore, carbonaceous electrodes for batteries and supercapacitors were developed by incorporating GQDs into the porous structure of metal-organic frameworks (MOFs).

Li-ion batteries with porous electrodes demonstrated outstanding cycle stability after 200 cycles, having capacities as high as 493 mA h g^{-1} at 100 mA g^{-1}. The extraordinary electrochemical capabilities of Li-ion batteries are due to the synergistic impact of MOF-derived porous carbon and GQDs. Nanocomposite electrodes' micro/mesoporosity and large specific surface area have been reported to speed up lithium-ion de-embedding and embedding in Li-ion batteries. Furthermore, the inclusion of GQDs to this porous structure dramatically increased electron transit speed and lithium storage capacity, as previously indicated. The electrodes were utilized to build supercapacitors having consistent capacity retention rates of roughly 130 F g^{-1} at 2 A g^{-1} after around 10,000 charge/discharge cycles. Furthermore, due to the electrode's porosity nature and the small size of the GQDs, the electrolyte was able to permeate the active surface of the electrode efficiently, reducing ion transport pathways.

2.3.1.4 Photocatalysis

GQDs stand for photocatalysis applications due to their photostability, outstanding light absorption capabilities, adjustable bandgap structure, and ability to create stable heterojunctions having other light-harvesting materials. Apart from that, they also have high electron mobility in photocatalytic processes, which increases charge transfer and extends the lifetime of electron–hole pairs. Moreover, GQDs' photocatalytic characteristics have been used in the past for dye degradation, CO_2 reduction, water splitting, and H_2 evolution, among other things. For example, Yan et al. [56] devised two alternate techniques for CO_2 reduction as well as photocatalytic water splitting by lowering the GQDs bandgap.

When the π* orbital is decreased, the bandgap of GQDs is reduced. Therefore, GQDs were adjoined with poly-aromatic moieties in the first strategy to boost sp_2-carbon systems and lower the GQDs' bandgap by reducing the π* orbital. The second method lowered the bandgap by functionalizing GQDs with electron-donating moieties that served as an intermediate n-orbital between the n and * orbitals. Finally, the z-scheme electron transport that arose from the coexistence of p- and n-type domains was employed to investigate the photocatalytic activity of functionalized GQDs.

Electron-donating moieties like amine functional groups (n-type domain) and nitrogen atoms are related to electron-withdrawing moieties like carboxyl groups (p-type domain) in this structure's ohmic contact by sp_2 clusters. In this intermolecular p–n type photochemical diode, couple redox reactions, as well as the dissociation of photo-generated electron–hole pairs are preferred. Slightly oxidized S-GQDs and ammonia-treated N-GQDs were utilized in comprehensive testing for photocatalytic H_2 evolution.

GQDs have also been combined with other nanomaterials (TiO_2, ZnS, ZnO, $BiOI/MnNb_2O_6$, g-C_3N_4, and CdS) to improve photocatalysts. For example, ZnS nanobelts comprising GQDs have been used to photocatalyst the breakdown of rhodamine B. The photo-degradation performance of GQDs/ZnS nanocomposites was discovered to be 14 times larger compared to the pure ZnS nanobelts by the cause of their increased light absorption capabilities and capacity to separate photo-generated electron–hole pairs. For the photocatalytic degradation of methylene blue, a blend of GQDs and ZnO nanowires with a shallow electron–hole recombination rate and increased photoabsorption capabilities was previously created.

The optimal GQD loading on ZnO nanowires (0.4 wt%) resulted in a solid interfacial chemical bond having lower bandgap energy and higher photo-generated electron–hole pair segregation efficacy. Another work used a new nanocomposite of stainless steel nanotubes (SSNTs) and GQDs to photocatalyse the breakdown of phenanthrene under visible light [42]. Due to the relatively quick recombination of photo-generated electron–hole pairs, SSNTs are not suited for photocatalysis. Charge segregation efficiency was increased by coating SSNTs with GQDs and using persulphate as an external electron acceptor. Sulphate radicals (SO_4), superoxide radicals (O_2), and photo-generated holes were identified as key reactive species during the photocatalysis of phenanthrene utilizing SSNTs/GQDs and persulphate under visual irradiation.

Furthermore, hydroxyl radicals (OH) have been discovered to play a minor function in the breakdown of phenanthrene. Three alternative pathways for photocatalytic degradation of phenanthrene have been hypothesized based on the creation of these reactive species (e.g., $SO_4^{\cdot}/^{\cdot}OH$ might target phenanthrene's side aromatic rings orphenanthrene's aromatic ring, meanwhile O_2. may attack phenanthrene's aromatic ring). SSNTs/GQDs nanocomposite structure could maintain its photocatalytic performance despite over five photocatalysis cycles, demonstrating reusability and good stability for practical uses.

2.3.1.5 Electrocatalysis

GQD-based nanomaterial development for electrochemical energy storage and electrocatalysis applications, such as CO_2 reduction, water splitting, and fuel cells, has recently commenced and is quickly developing. The method of GQD synthesis and the structural features that arise (e.g., doping, size, surface and edge states, and defects) significantly influenced the finished product's electrocatalytic activity. Due to their quick electron transport, high surface area, and many functional groups on the surface, for instance, –OH and –COOH, GQDs are an appealing option for electrocatalyst systems. In addition, GQDs have been employed in electrocatalysis for a variety of applications, including electrochemical biosensors, biofuel cells, CO_2 electro-reduction, hydrogen evolution reactions, oxygen evolution reactions, and oxygen reduction reactions (ORRs). ORRs are one of the purported benefits of GQDs in electrocatalysis.

Due to their electronegativity variations, heteroatoms, for instance, sulphur (S), phosphorous (P), boron (B), and nitrogen (N), polarize the sp_2 carbon atoms of GQDs, facilitating oxygen molecule adsorption. It then uses a hydrothermal treatment to self-assemble GQDs on graphene, followed by high-temperature annealing to co-dope the hybrid nanoplatelets having N and B, lowering the oxygen dissociative energy barrier in catalysts.

Combining GQDs' large surface area, abundant edges, and B, N doping sites having graphene's high electrical conductivity enabled the creation of hybrid nanoplatelets that can propose outstanding ORR activity having a 15 mV higher positive onset potential and comparable existing density than commercial platinum/carbon (Pt/C) electrodes. Then, utilizing a one-step simultaneous reduction procedure, ORR catalysts based on GQDs supported by graphene nanoribbons (GNRs) were developed. The created GQDs/GNR hybrids' ultrahigh electrocatalytic performance was attributed to effective charge transfer between the tightly packed GQDs and GNRs, including GQDs numerous flaws on the edges, hybrid surfaces, and interfaces, all of which functioned as active sites.

Despite the significant progress made in the production of GQDs, further research is needed to fully understand the complex relationships between electrocatalysis applications and GQDs structural features. GQD defects, shape, size, active sites, heteroatom doping, conductivity, edge structures, as well as synergistic effects all play a role in improving the electro-catalytic performance of GQD-based catalysts. Apart from that, this could open the way for enhanced GQD functionalization procedures when paired with polymers, carbon allotropes, metal oxides/sulphides, and metals, leading in potential materials for numerous electrocatalysis applications. Also, the rising market for hybrid electric vehicles, for example, emphasizes the necessity of developing environmentally acceptable electrocatalytic devices with high energy density, solid catalytic activity, long durability, and low cost, as well as stability.

2.3.1.6 LEDs

GQDs provide several advantages that LEDs can use, including enhanced optical characteristics, the capacity to construct stable heterojunctions out of a range of materials, non-toxicity, as well as excellent carrier transport mobility. There are two forms of GQD-based LEDs: electroluminescent and photoluminescence (phosphor-converted). In electro-luminescent LEDs, holes and electrons are injected into an active GQDs-based emission layer via an external bias voltage and undergo radiative recombination to produce photons. Nevertheless, UV or blue LED chips are widely utilized to optically pump GQD-based phosphors. This approach can effectively change LEDs wavelength and light intensity. GQD-based LEDs, whether electroluminescent or photoluminescent, may possess a greater external quantum effectiveness, are more cheaper, and have no direct environmental impact than traditional LEDs that use rare earth elements.

Kang et al. [26] described an electroluminescent white-light-emitting diode (WLED) based on GQDs doped with 4,4-bis(carbazol-9-yl) biphenyl (CBP). This WLED's white electroluminescence was produced via an efficient energy transfer between CBP and GQDs, which was preceded by GQD excitation and white light emission. The active emitting component of a WLED has just been employed with edge functionalized GQDs (EF-GQDs) dispersed in poly(9-vinylcarbazole) (PVK). The maximum luminance of the PVK:EF-GQD-based WLED is 464 cd m^{-2}, which is significantly greater than previously recorded GQD-based WLEDs. An increasing body of literature has investigated the use of GQDs as novel phosphors in photoluminescence LEDs in addition to electroluminescent devices. WLEDs colour conversion layer was made up of yellow-green emitting GQDs cast in UV-curable siloxane.

By altering GQDs concentration and the colour conversion layer's thickness, the colour–temperature behaviour of LEDs may be modified. Enclosing GQDs in a polymer matrix to control their emission properties and avoid self-quenching is also a hot issue in this study. Wu et al. [57] created melamine-formaldehyde (MF) microspheres encased in GQDs that glow brilliantly white when exposed to UV light. The MF microspheres were then disseminated in a cross-linked polydimethylsiloxane matrix to create a flexible and high-quality white-light-emitting film that could be used to create far-flung planar WLEDs. High-efficiency long-wavelength emission LEDs may be generated by modifying the aggregation state of GQDs, and they could be useful in practical applications.

2.3.1.7 Surfactant

GQDs have remarkable amphiphilic properties thanks to their hydrophobic sp_2 domain and capability to facilitate hydrophilic moieties like carboxyl and hydroxyl groups. GQDs are suitable surfactants for highly stable emulsions because of their biocompatibility and small particle size. Graphene was the focus of previous research on carbon-based nano-surfactants. GQDs smaller than graphene can fit more active sites and generate minor and more stable emulsion droplets. Xi et al. [58] recently revealed on the use of amphiphilic GQDs to wet superhydrophobic surfaces by dispersing CNTs in an ultra-light aerogel, stabilizing the Pickering emulsion, and speeding up the Pickering interfacial polymerization reaction. GQDs were also used as a new surfactant by Zeng et al. [59] to stabilize pure graphene aqueous dispersions.

Various amino, sulphonyl, and carboxylic functionalized GQDs were formed to evaluate the possibility of stacking graphene and functionalized GQDs to stabilize pure graphene aqueous dispersions. As per their observations, sulphonyl-functionalized GQDs could stabilize the maximal graphene concentration because of electron-withdrawing solid groups availability. Following that, photonic crystals were created by dropping homogenous polystyrene/GQDs/graphene suspensions onto a substrate and mixing them with polystyrene nanoparticles.

Amphiphilic GQDs can obtain diverse functions for specific applications depending on the surface functional groups' type. Hydrophilic COOH-functionalized GQDs were grafted with hydrophobic hexylamine to produce amphiphilic GQDs, which were used to stabilize water-in-cyclohexane Pickering emulsions. Surfactants like hexylamine and oleylamine were grafted onto the surface of hydrophilic GQDs to change the shape of block copolymer particles in a comparable manner. Li's group also created amphiphilic zinc–histidine–functionalized GQDs (Zn–His–GQDs), which were coated on the surface of polystyrene microspheres and used to adsorb Cu^{2+} from water. While GQDs show great potential in the development of highly effective surfactants, they have yet to be completely studied. Given their high dispersibility, non-toxic nature, and intrinsic amphiphilic properties, GQDs ornamented with varied functional groups and doped heteroatoms can serve as a completely new class of surfactants for numerous applications.

2.3.1.8 Non-volatile matter (NVM) devices

Utilizing variable energy levels and the boundary effect, GQDs were used as the charge trapping medium of the high-density NVM [60]. By altering the size and concentration of nanoparticles, GQDs provide configurable memory performance as a charge trapping material, as well as chemical and thermal stability, cost-effectiveness, and compatibility having complementary metal-oxide-semiconductor technologies. Multiple breakthroughs have been made in this field since Dong et al. [28] initially introduced GQDs as a charge trapping medium for NVMs. The charge trapping GQDs layer, for example, has been found to boost the memory window and program/erase speeds of NVMs when sandwiched between SiO_2 thin films.

GQDs sandwiched between poly(methylsilsesquioxane) layers were also used by Ooi et al. [60] to manufacture an NVM device. The constructed gadget was said to have a 1 104 ON/OFF ratio and a retention duration of higher than 104 s. In resistor-type memory devices, GQDs have also been used as a charge trapping medium (memristors) and a resistive switching material. To perform resistive switching, memristors employ resonant electron tunnelling via GQDs in the presence of an electric field. Finally, our studies revealed that GQD-based memory devices are simple to construct and have a wide range of potential applications. Hence, to summarize, the interplay between various GQD features and memory device parameters should be examined thoroughly to fully comprehend the solid-state physical behaviour of GQD-based NVMs.

2.3.1.9 Nanofluids

The name of nanofluids was introduced in 1995 and demonstrated that nanoparticles might significantly improve the thermal conductivity of fluids. This novel family of coolants ushered in a new era in thermal system research and development due to its unique thermo-physical features. Freezers, nuclear reactor thermal management, air conditioners, heat exchangers, solar energy systems, thermal car management, and micro-electrochemical systems are just a few of the technical applications for nanofluids.

A surprising amount of research has been carried out on the improved thermo-physical characteristics of various kinds of nanofluids comprising different classes of nanoparticles (e.g., carbon-based nanoparticles, oxide ceramics, metal oxides, and chemically stable metals). Most of these studies concentrated exclusively on nanofluids' thermophysical behaviour, with little attention to their long-term qualities and stability. Despite this, most current nanofluids are ineffective for industrial applications. As a result, the promise of manufacturing stable nanofluids provides a constant source of motivation for the GQDs to make stable nanofluids because of their ability to aid a considerable number of surface functional groups as well as enormous surface area.

Xie et al. [25] were the first to investigate the thermophysical properties of amine-functionalized GQDs in water-based nanofluids. According to their findings, a small amount of GQDs (0.002 wt%) added to water increased its thermal conductivity by up to 18.6%. Furthermore, when compared to pure water, the created nanofluids had no significant viscosity enhancement, significantly reducing pressure loss and pumping power in industrial applications, according to transfer specialists. A water-based nanofluid containing small GQDs was developed as a unique kind of vehicle radiator coolant in another investigation. When contrasted to the primary fluid, this nanofluid's convection heat transfer coefficient as well as thermal conductivity, which contained just 100 ppm of GQDs, was enhanced by 5.2 and 17%, accordingly. GQD suspensions predicated on the water were also used to improve two-phase closed thermosyphon heat transfer performance. In addition to water, GQDs have been used to enhance the thermophysical properties of other prevalent coolants, for instance, ethylene glycol, glycerol, and transformer oils.

Consequently, Shanbedi et al. [61] used amine-functionalized GQDs to make exceptionally stable transformer oil-based nanofluids. Compared to pure transformer oil, the generated nanofluids had much greater breakdown voltage, flash point levels, natural and forced heat transfer rates, as well as thermal conductivity. GQDs have little impact on the rheological qualities of transformer oil, resulting in low-pressure drops and pumping power.

GQDs have paved the way for the development of highly stable nanofluids. Since it works with an extensive scope of implementation, the GQD-based nanofluids research potential appears limitless. According to the scientists, further research should focus on the effect of the size, functionalization, synthesis process, and GQDs concentration on the thermo-physical characteristics and nanofluids stability. In addition, the tribological and lubricating features of GQD-based nanofluids should be investigated in the long run.

2.3.2 Sensing applications

In previous years, significant progress was initiated in developing new GQD-based sensors for several applications, including bio-analysis, chemical, and physical. The five types of sensors created with GQDs are electrochemiluminescence (ECL), photoelectrochemical, electrochemical, photoluminescence, and electronic. In the following sections, we have gathered a quick rundown of the most current developments in these innovations and their benefits and drawbacks for the use of the readers.

2.3.2.1 Electronic sensing

GQDs were used to construct charge sensors based on SETs in electronic sensing. SET is a new form of nanosensor that amplifies current by controlling electron tunnelling. Various techniques, for instance, electron beam lithography carving and oxygen reactive ion etching, can be used to construct the desired geometries of GQD-based SETs. If the bias voltage between the source and drain is sufficient, an electron may travel through the SET structure and generate a current flow to measure the charge in this transistor type.

Kim et al. [62], who built a vertical tunnelling SET, recently reported a breakthrough in the controlled growth of GQDs-hexagonal boron nitride (hBN) hetero-structures. Intriguingly, extending GQDs inside the hBN matrix lowers the number of localized states around GQD margins significantly. These surface-passivated GQDs heterostructures could open up new avenues for building novel charge sensors. In addition to charge detection, GQDs have been applied to manufacture electrical sensors for humidity sensing. Dropping a few microliters of GQD aqueous solution over a patterned metallic interdigitated microelectrode, for example, produced a low-cost, fast-responding humidity sensor. The sensing method used in this application was capillary condensation of water molecules on the surface of GQDs, which was accompanied by a change in circuit resistance.

2.3.2.2 Photoluminescence sensing

With different PLQYs, GQDs synthesized using different synthesis techniques can emit diverse colours. Surface defects and inherent energy states of GQDs are hypothesized to be responsible for their photoluminescence emission, which is particularly sensitive to interactions with other molecules as well as critical ecological factors. As an outcome, the photoluminescence feature of GQDs may be used in various sensing applications. Few ways for constructing photoluminescence sensors have been proposed so far, including photoluminescence quenching of GQDs, photon-induced electron transfer, and fluorescence resonance energy transfer.

GQD-based photoluminescence sensors were also utilized to track small organic molecules (e.g., glucose and ascorbic acid), as well as for non-metallic anions (e.g., pyrophosphate, sulphite, and sulphide). N-GQDs with blue fluorescence created a susceptible and selective trinitrophenol (TNP) sensor. The TNP sensor's detecting process was characterized by intense electrostatic interactions between TNP molecules and N-GQDs, followed by N-GQDs photoluminescence quenching.

This TNP sensor had high selectivity for metal ions and aromatic compounds in the environment. In addition, the GQDs' photoluminescence emission has been employed in the development of biosensing systems. Fluorescent sensors based on GQDs were employed to recognize a variety of bio-targets. This includes DNA, gene sequences, microRNAs, metabolites, cardiac marker\santigenTroponin I (cTnI), as well as human immunoglobulin\sG. For example, tyramine-functionalized GQDs have been applied to establish a simple sensing platform for detecting several metabolites in a blood droplet.

As a result, more research is needed to determine if these changes in selectivity are linked to structural characteristics, types of dopants, and bonding arrangements of GQDs concentrations of heteroatom dopants, doping method size and form of heteroatom dopants, and chemical moieties. Apart from the complicated processes, this sensing method has some flaws due to the exterior light's vital background source. Photoluminescence sensing also necessitates a significant financial investment.

Fluorimeters, for example, may not be used regularly, but analytical laboratories have them.

2.3.2.3 Electrochemical sensing

Because of GQDs' ability to interact with small molecules, promote heterogeneous electron transport, and speed redox processes, many electrochemical sensing platforms were developed. Apart from that, electrochemical sensors turn chemical reactions on the electrode's surface into signals, allowing them to recognize analytes and provide environmental information. Electrochemical sensors are inexpensive, easy to use, and highly sensitive. Therefore, cardiac biomarker myoglobin, micro RNAs, DNA sequence, glucose, H_2O_2, dopamine, and heavy metal ions have been detected electrochemically using GQD and its composites.

The quick dopamine as well as Hg^{2+}, Cu^{2+}, Cd^{2+} detection in a range of complex environmental and biological samples has previously been achieved using a novel electrochemical sensing platform built on nanochannel-confined GQDs [63]. GQDs were employed to enhance the signal and enable charge transfer via interactions with analytes in this platform (e.g., interactions with dopamine molecules or particular chemical moiety interactions with heavy metal ions).

On the supporting electrode, a vertically organized nanochannel film was used as an anti-interface layer to prevent non-specific molecules from invading and boost selectivity. The sensitivity and detection limits of this sensor device are rather low (9.8 pm for Hg^{2+}, 8.3 pm for Cu^{2+}, 4.3 nm for Cd^{+}, and 120 nm for dopamine). To detect H_2O_2 in living cells, another sensing technology based on N, S co-doped GQDs/graphene composite was developed. Given its wide specific surface area, multiple functional groups and doping sites, and comparably high electrical conductivity, the produced GQDs/graphene composite performed well in the electrocatalysis of H_2O_2. This H_2O_2 biosensor had a linear range of 0.4 m to 33 mm having a low detection limit of 26 nm.

2.3.2.4 Photoelectrochemical sensing

The energy and charge transfer between electron donors/acceptors and photo-responsive materials during photoirradiation is the basis of photoelectrochemical sensing. The only excitation source in this sensing method is light, and the signal readout is current. The photocurrent response of electrochemical sensors can be improved by using GQDs, which have remarkable electrical and optical features. Yan et al., for example, developed a GQDs/TiO_2 nanocomposite-based photoelectrochemical sensing platform for sensitive dopamine detection with a 6.7 nm detection limit. A sensitive photoelectrochemical sensor for detecting zeatin was developed using GQDs and a graphite-like C_3N_4 composite in another investigation.

In this semiconductor composite, GQDs were employed to boost C_3N_4's photoelectric conversion efficiency. The sensor boasted a linear detection range of 0.1–100 nm, a low detection limit of 0.031 nm, as well as good selectivity in complicated biological samples. In comparison to electrochemical and photoluminescence sensing, photoelectrochemical platforms remove the excitation source (light) from the detection signal (electrical current), significantly lowering background signals. Moreover, photoelectrochemical sensors are inexpensive, simple to miniaturize, and need relatively simple instruments. However, there are still many obstacles to overcome in developing GQD-based photoelectrochemical sensors. As an example, the majority of studies have only used photoelectrochemical sensors to quantify a single analyte. Future research will focus on the simultaneous detection of many analytes utilizing this sensor technology. Furthermore, only a few investigations on GQD-based photoelectrochemical sensors with good selectivity have been published [63].

2.3.2.5 ECL sensing

Electrochemistry and spectroscopy are combined in the ECL platform to generate a susceptible and selective sensing platform. Two of the essential advantages of this sensing technique are regulating the reaction by applying electrode potentials and the lack of a background optical signal (light source). In addition, potent oxidizing radicals are created at the surface of electrodes in GQD-based ECL sensing platforms.

These radicals subsequently undergo electron-transfer annihilation with GQDs, resulting in an excited condition which produces light. Moreover, ECL emission is quenched when specific analytes, nucleic acids, proteins, and heavy metal ions impair the interactions between radicals and GQDs.

For the detection of folic acid in human serum samples, researchers developed a new ECL resonance energy transfer platform using boron nitride quantum dots (BNQDs) as the provider and N-GQDs as the receiver. In the ECL system of NGQDs/BNQDs/$K_2S_2O_8$, the interaction between folic acid and SO_4 radicals was exploited to enhance the sensor's detecting mechanism. The linear range of this sensing platform was determined to be 10 pm to 100 m, with an ultralow detection limit of 5.13 pm. N-GQDs were also used to make a dual-potential ECL sensor for sensitive Co^{2+} detection in water in another investigation. At both negative and positive potentials, N-GQDs in contact with dissolved oxygen displayed two different ECL responses. Ratiometric detection of Co^{2+} ions having a 0.2 m detection limit was achieved using the ratio of these two ECL responses.

2.3.3 Medical and environmental remediation applications

2.3.3.1 Biological imaging

The introduction of GQD changed the fundamentals of cellular and *in vivo* imaging forever. Due to its unique vital merits, such as photostability, biocompatibility, the ability to conjugate with targeted molecules via various active sites (for instance, dopants, functional groups, as well as edges), equivalent size to biomolecules, outstanding dispersibility, and a simple photoexcitation mechanism, GQD may deliver as a universal fluorophore.

It is simple to see why GQD has such a bright future in biomolecular imaging when we compare its properties to those of semiconductor fluorophores as well as typical organic dyes. Fluorescent proteins and organic dyes, for example, are frequently utilized in bioimaging, but they quickly photobleach, rendering them unsuitable for long-term imaging. Furthermore, semiconductor quantum dots are inclined to aggregation, which can result in dangerous heavy metal leakage. Due to their gigantic size in comparison to biomolecules, semiconductor fluorophores may alter the function and trafficking of their targets, and thus they cannot be considered useful bioimaging agents [16].

GQDs were used to image T47D human breast cancer cells, MCF-7 cells, MDA-MB231, MG-63 cells, stem cells, hepatic cancer cells, L929 cells, and, more often, HeLa cells. In addition, Lou et al. [64] used highly crystallized GQDs for two-photon bioimaging of HeLa cells; meanwhile, Ding et al. [65] used N, Cl co-doped GQDs emitting at 365 nm to scan MDA-MB231 cancer cells. Aside from that, Kumawat et al. [66] made a red luminescence GQD (mGQD) out of mango leaves, which they utilized as an NIR-responsive fluorescence probe for imaging L929 cells.

A significant NIR signal was seen in the cytoplasm of L929 cells that had been labelled with mGQDs. These mGQDs demonstrated outstanding biocompatibility, selective self-localization behaviour, and 100% cellular absorption even at high concentrations. GQDs have been used for selective bioimaging to obtain accurate information about a targeted cell in conjunction with regular cellular imaging. Previously, GQDs conjugated having specific bioactive moieties, for instance, hyaluronic acid (HA), folic acid (FA), as well as arginine–glycine–aspartic acid (RGD) have been shown to be useful as target-specific bioimaging indicators.

RGD-conjugated GQDs, for example, were extremely effective in targeting U251 cells, whereas a new targeting agent for MKN cells was produced via a combination of N-doped GQDs and FA. In addition, monosaccharide-conjugated GQDs was also used to image carbohydrate receptors in living cells [67]. This excellent bioimaging marker reveals real-time carbohydrate receptor trafficking on the cell membrane, opening up a whole new world of possibilities for detecting cancer cells.

MRI has evolved as a cutting-edge imaging modality, delivering great spatiotemporal resolution and deep tissue penetration as a highly sensitive and non-invasive approach. These properties enabled quantitative probing of various cellular processes, tissue dynamics, and cellular anatomy. MRI, on the other hand, is a non-ionizing radiation-based imaging modality, as opposed to ionizing radiation-based imaging modalities such as positron emission tomography and computed tomography, avoiding radiation-induced damage and toxicity. Apart from the potential benefits, the long operational time of MRI for signal gathering, as well as its modest sensitivity (103–505 mol/L), need careful consideration for practical clinical applications. MRI uses radio waves and a magnetic field to distinguish damaged (cancer) tissue from healthy tissue by activating the local proton environment. It has the significant benefit of being simple to utilize for X-ray imaging procedures. Its shortcomings include low sensitivity and long signal recording durations. Because the proton magnetic moment of tissue varies with environment and the T1 and T2 durations may not create a superior picture, several external contrast agents are usually utilized. Gd^{3+} is employed for brilliant contrast and superparamagnetic iron oxide NPs for dark contrast.

2.3.3.2 Gene delivery

GQDs are excellent options for delivering gene to interact with a variety of small molecules and genes-conjugated sp_2 carbon network and several surface functionality groups since their favourable features, for instance, the ability to deliver multiple genes, easy cellular uptake due to their small particle size, and biocompatibility. A multipurpose device was created by Dong et al. [68]. Moreover, Wang et al. [69] employed GQDs to suppress breast cancer development genes for resistance proteins, whereas GQDs for simultaneous intercellular communication imaging and gene delivery using microRNAs (miRNAs). Both of these investigations highlighted how GQD-based platforms have been efficiently utilized to improve productivity. The efficacy of gene therapy drew a lot of attention in this research area. Micro RNA223 (miR233) was delivered into the interior of the plaque using a unique monocyte surface designed gene-delivery system based on GQDs to treat atherosclerosis-related diseases. GQDs were disulphide-bonded to the miR233 gene and then grafted onto a C189-peptide (C18P) surface with a hydrophobic end for this gene delivery approach. When the GQD–miR233 composite penetrates the interior of the plaque, the C18P bonds are digested, and the GQD–miR233 composite is liberated. In the lysosome (GILT), interferon-inducible lysosomal thiol reductase destroys the disulphide connections between GQDs and miR233. MiR233 cargos may target and infiltrate macrophages in atherosclerotic plaques *in vivo* using this technique.

2.3.3.3 Drug delivery

In today's medicine, the creation of miniature drug delivery nanosystems for delivering drugs to specific cells or organs is critical. Given the low toxicity, high water solubility, and capacity to react with specific medications via their conjugated sp_2-carbon structure and a variety of surface functional groups (carbonyl, epoxy, hydroxyl, and carboxyl), GQDs are recognized as better drug carriers.

GQD is chosen as a SCAN substrate to load the anticancer medication doxorubicin because of its high drug absorption properties (DOX). The DOX/GQD composite was coated with a pH-sensitive polymer called *N*-acetyl histi-dine-functionalized D-tocopherol PEG 1,000 succinate in the last phase of SCAN preparation. HTPGS. shows a detailed schematic representation of SCAN for hierarchical tumour targeting. SCAN accumulates in the mild acidity of the tumour environment before breaking down into the 5 nm DOX/GQD composite after NIR radiation, as shown in the image. SCAN's efficient *in vivo* targeting and tumour penetration capacity is in sync with this transitory approach.

Sung et al. [70] developed a sponge-inspired carbon composite-supported red blood cell (RBC) drug delivery system that is tumour-penetrative and stealthy. The hydrophobic drug docetaxel (DTX) was chosen for delivery via interactions in this drug delivery platform, and GQD was selected. The sponge-like mesoporous silica having huge holes was then treated with GQD/DTX before being sealed with RBC membrane and Cetuximab (Ct) (donated as Ct-RBC@GQD-D/NS).

2.3.3.4 Photodynamic and photothermal therapy

The use of a photosensitizing medication triggered by a particular wavelength of light and subsequently creates ROS that destroys adjacent cells is known as photodynamic therapy (PDT). Photothermal therapy (PTT), in contrast, is a type of photodynamic therapy that uses electromagnetic radiation (mainly infrared radiation) to treat some disorders, particularly cancer. PDT and PTT encourage the host immune system to fight tumour deposits by destroying local tumours directly through photothermal mechanisms or by generating ROS (only for PDT). Because of their capability to create radicals, outstanding dispersibility, water solubility, biocompatibility, as well as extraordinary photostability, GQD-based nanomedicines are known to improve the efficacy of numerous photo-enabled cancer ablation procedures.

Because of their tiny specific surface area and high density of surface functional groups, GQDs were employed to increase the solubility of photothermal and photosensitizer chemicals in appropriate pharmaceutical solvents and to develop high-performance anti-cancer drugs. Folic acid-functionalized GQDs, for instance, demonstrated enhanced photostability and tumour targeting capabilities when used in concert with the IR780 iodic NIR theranostic agent, allowing cancer cells to be swiftly destroyed [71]. In addition, a unique responsive nano-assembly based on GQDs was established for PDT. Hence, in this selective system, the photosensitizer chlorine 6 (Ce6) was disulphide-bonded to the surface of GQDs.

This system is photoinactive during normal circulation. When the nano-drug hits the surface of tumours, intracellular glutathione cleaves the disulphide linkage, restoring Ce6's phototoxicity. As a result, it is acceptable to state that GQDs increase the solubility and the target selectivity of the photosensitizer Ce_6 in this redox responsive nano-drug. As a result, Wu et al. [72] established a very effective nano-theranostic for cancer treatment based on functionalized photoactive GQD-based nanocomposites. The amine-terminated GQDs were conjugated with photosensitizer Ce_6 and subsequently coated with polydopamine layers to create polydopamine stabilized GQDs photosensitizer nanocomposites (GCpD). PC@GCpD(Gd) is a flexible photoimmunotherapy nano-assembly created by combining GCpD with Gd^{3+}/Cy^3 imaging probes and immunostimulatory polycationic polymer/CpG oligodeoxynucleotide (CpG ODN) nanoparticles. The photothermal activity of nano-theranostic by comparing the localized heating in the tumour region of a mouse given PC@GCpD (Gd) after laser exposure compared to an untreated mouse.

When subjected to laser light against breast cancer cells, a stable compound comprising graphene oxide nanosheets and GQDs linked with cationic polyethyleneimine (GO–PEI–GQDs) demonstrated excellent photothermal reactivity (MDA-MB231). Moreover, the GO–PEI–GQDs hybrid, which combines the intrinsic features of graphene oxide and GQDs, showed outstanding cytotoxicity and biocompatibility even at low laser power levels. As a result, this compound could pave the way for developing more potent anti-cancer therapies with fewer adverse effects.

2.3.3.5 Treatment of chronic diseases

Chronic illnesses are those that last for a long time, often incurable illnesses that are the leading cause of mortality worldwide. Aside from cancer treatment, GQD-based materials have shown potential in treating several of the world's most well-known diseases, such as HIV, hepatitis, and malaria. Diabetes, as well as Parkinson's disease, are two of the most prevalent disorders. GQDs, for example, show cytoprotective and immune modulatory characteristics that are unique because they relieve immune-mediated fulminant hepatitis. Hepatic inflammation, autophagy, and apoptosis can all be lowered [22].

F-GQDs, which are incredibly fluorescent and water-dispersible, were utilized in another investigation to prevent the production and lipids deposition, a polypeptide of human islet amyloid (hIAPP). Inhibiting the hIAPP production is a vital process in establishing type 2 diabetes medications. F-GQDs may effectively bond having hIAPP monomer/oligomers and limit the mature amyloid development due to their substantial charge density and capacity to accept multiple surface functional groups.

GQDs are important in avoiding islet amyloid polypeptide (IAPP) aggregation in embryonic zebra fishes, according to Wang et al. [73]. The IAPP helical was absorbed on the surface of GQDs sheets in the presence of GQDs and subsequently unfolded transiently following a series of thermodynamic disturbances. This breaks down IAPP's helical and sheet structures, resulting in random and developed monomers and, as a result, the prevention of amyloid disorders.

This defence could come from a variety of sources, for instance, mitogen-activated protein kinase as well as mitogen-activated protein kinase [74]. GQDs were employed as an anti-aggregation agent to avoid aggregation. As a result of their research, -synuclein (-syn) monomer accumulation and transmission in the middle of the brain GQDs were introduced.

2.3.3.6 Antimicrobial materials

Antibiotic resistance is a severe public health issue that leads to higher medical costs, longer hospital stays, and even higher death rates all across the world. As a result, developing flexible alternative drugs with high antibacterial action is critical for public health protection. GQDs have a peroxidase-like activity that could result to the creation of a new antibacterial chemicals class. Here, an antimicrobial wound dressing containing GQDs and a small amount of H_2O_2 is a good example of this. However, due to their flat surface, GQDs generated by exfoliating graphene oxide sheets (GO-GQD) lacked antibacterial characteristics, sheet form, or bacterial absorption at the least outermost layer. The bactericidal properties of nanocomposites, including GQDs as well as other materials including metal oxide nanoparticles, silver, as well as zinc, have also been found.

Due to ROS emission, Chen et al. [75] discovered that GQDs/Ag nanocomposite possessed exceptional bactericide capabilities. The synthesized nanocomposite performed admirably to destroy *Staphylococcus aureus* and cells *Escherichia coli* and eradicate drug-resistant *E. coli* germs. Another example of GQD-based nanocomposites with increased antibacterial impacts on gram-negative (*E. coli* and *P. aeruginosa*) bacteria and gram-positive (*S. aureus*) is surface disordered rutile TiO_2–GQDs hybrid. Bactericidal properties of TiO_2–GQDs nanocomposite are better in UV light than TiO_2 and GQDs only.

2.3.3.7 Heavy metal removal

The release of toxic heavy metals from aqueous solutions is among the world's most pressing environmental challenges. Due to their high toxicity and limited degradability, heavy metal ions constitute a significant hazard to human health and the environment. A high cadmium intake, for example, could lead to kidney failure or lung cancer. Long-term exposure to copper ions, on the other hand, can result in DNA damage as well as liver and pancreas failure. Furthermore, when mercury ions are eaten, they have demonstrated to cause significant damage to the neurological system and brain.

CNTs and graphene have both been extensively utilized in recent years to eliminate heavy metals and reduce their environmental impact. Nevertheless, how carbon-based nanomaterials interact with ions determines their ability to discard heavy metal ions from aqueous solutions. In terms of accepting a broad spectrum of chemical moieties and heteroatom dopants, GQD beats CNTs and graphene.

Due to the large specific surface area of these 0D nanomaterials, the active sites of GQDs may directly interact with heavy metal ions. Owing to these characteristics, as well as its biocompatibility and high dispersibility, GQD is one-of-a-kind and highly effective heavy metal absorbent. Furthermore, ionic liquid-capped GQDs (IL-GQDs) were introduced by Nagaraj et al. [76] to remove the hazardous Cr^{6+} from aqueous solutions. Chemical oxidation of graphene oxide nanosheets produced GQDs in their research. The IL-GQDs were created through an amidation reaction between an ionic liquid (methyl imidazole and 3-bromopropylamine hydrochloride IL) and carboxylic acid-functionalized GQDs. At neutral pH, the absorption of the generated IL-GQD was 934.62 mg g^{-1}, which was significantly higher compared to the absorption of other commonly identified absorbents. Note that GQDs have been coated on the surface of a variety of solids, including magnetic materials and quartz sands, in order to make recovering absorbents from aqueous solutions easier after the heavy metal extraction process. Zheng et al. [36] created a low-cost, high-efficiency heavy metal absorbent using GQDs coated on quartz sand to remove Hg^{2+} and Pb^{2+} ions from aqueous solutions. Due to its high stability, low cost, and capacity to remove heavy metals, the GQDs/quartz sand composite is appropriate for industrial wastewater treatment units.

3 Conclusion and recommendations

Due to its optoelectrical, electrical, and unique optical qualities, the future of GQD appears to be infinite in many domains of science and technology. Since 2008, numerous research projects on GQDs have progressed, and advanced applications have emerged in engineering, sensing, and biological applications of GQDs. The concept of fields has been achieved. However, there are a few fundamentals that must be addressed. Before anything else, there are concerns to be addressed and hurdles to conquer.

First and foremost, distinct physical and crystallographic peculiarities should be differentiated from those qualities of GQDs nanoparticles with similar structures. The forthcoming research should compare the morphological and physicochemical features of GQDs having CQDs, as the research has yet to address this problem that GQDs encounter. There are a variety of synthesis techniques and situations that can be used to demonstrate due to the various physicochemical properties of heteroatoms with considerable variability in edge state, shape, size, functional groups as well as dopants.

As a result, efficient GQD implementation for one type of GQD with various physicochemical and structural qualities may be impossible for another type of GQD having different structural and physicochemical characteristics. Future studies should classify GQD attributes depending on their compositional and structural aspects and then correlate them to develop a clear roadmap for GQD research. GQDs have rates that are unique to each application. Li et al. [71] published their work on engineering the GQDs bandgap. They emphasized an improved specific GQDs' catalytic application through their research, as well as a thorough knowledge of the features that generated such catalytic activity in these GQDs.

The low PLQY of these 0D carbon-based nanomaterials should not be overlooked, especially due to existing constraints and restrictions of GQDs. The bulk of GQDs published in the literature so far has PLQYs ranging from 5 to 60%, which is low when contrasted to other fluorophores (for instance, organic dyes, semiconductor QDs, as well as CQDs). GQDs are unlikely to have a similar PLQY as different fluorophores despite their small size. Numerous methods may successfully increase the PLQY of these nanomaterials, enabling them to be employed in a range of applications. Surface modification, the removal of nonradiative recombination sites, and the introduction of single or multiple heteroatom dopants, to name a few, can all enhance the PLQY of GQDs.

GQDs, on the other side, have a limited production yield and an incredibly high cost, which are two of the most critical issues for both academic and industrial operations. The bulk of current top-down or bottom-up GQD synthesis processes produces fewer than 30% by weight. These methods also demand time-consuming and costly purifying procedures, raising the ultimate cost of GQDs dramatically. Consequently, subsequent GQD research should emphasize improving manufacturing yield and PLQY and purification ease to make GQDs cost-effective for industrial use. Furthermore, Shin et al. [77] demonstrated this idea for low-cost GQD synthesis by developing a novel oxidative cutting synthesis of GQDs having a high PLQY (69%) and a high production yield (77.3 wt%) while evading time-consuming purification. Thus, it is safe to say that the GQD study has progressed toward industrial-scale synthesis methods. We anticipate that with more study, large-scale GQD production will become a reality soon, and that GQDs will join the graphene mass production trend.

Lastly, the scarcity of a well-defined photoluminescence mechanism is a problem in GQD research. There is currently no mechanism to explain how different parameters (for instance, size, heteroatom dopants, chemical moieties shape, as well as edge configurations) affect GQD photoluminescence behaviour. Future experimental studies, together with theoretical investigations that can precisely alter the chemical composition of GQDs, will throw additional light on this issue. Sung et al. [70] numerical analysis of the parameters that impact GQD photoluminescence behaviour and bandgap might be utilized as a starting point for tackling this issue. The influence of two or more factors (e.g., electron-withdrawing as well as electron-donating dopants functional groups) on the photoluminescence of GQDs will likely be studied in the future. Besides that, the outcomes of these numerical simulations must be contrasted to the features of correctly constructed GQDs. That, in my perspective, is also the most challenging barrier to confront, as it involves substantial experimental and numerical investigation.

To summarize, the absolute value of GQDs will be grasped through overcoming the hurdles mentioned earlier and utilizing GQDs' unique advantages for industrial-scale applications. The situation is still the same today. GQDs have a bright future ahead of them in terms of engineering, biological applications, as well as sense. Surprisingly, some nanofluids, surfactants, and heavy metals are examples of GQD uses. NIR bioimaging and metal scavenging are so novel that numerous individuals are not aware of them. Without a question, new applications and gadgets based on GQDs will be developed, and they will be used in a variety of industries in the coming years. GQDs will be used to develop new applications and products. They will be used in various industries in the coming years as one of the most advanced photocatalysis applications. For example, certain distinct qualities of an individual can be compared to those of GQDs to better understand the task at hand. In order to develop more advanced systems, researchers have leveraged the intrinsic attributes of GQDs by including n-orbitals in their design. These carefully engineered GQDs have demonstrated exceptional catalytic performance, which can be attributed to their unique properties.

Acknowledgments: We would like to express our gratitude to the following individuals and organizations for their contributions to this research: Universiti Teknologi MARA Negeri Sembilan Branch, Kuala Pilah Campus: We would like to thank Laborotary Department for providing the necessary resources and facilities to conduct this research. [Universitas Islam Indonesia]. We would like to acknowledge the valuable contributions of Prof Is Fatimah in providing feedback and insights that helped shape our research.

Funding information: This study was backed by Universiti Teknologi MARA (UiTM) under the International Research Matching Grant (IRMG) (600-TNCPI 5/3/DDN (05) (006/2021) and Supervisor Incentive Grant 600-RMC/GIP 5/3 (041/2022).

Author contributions: S.A.I.S.M.G.: formal analysis; I.F. and Z.N.Z: investigation; N.N.Z.: methodology; S.A.I.S.M.G. and N.D.D.: supervision; N.A.: validation; Z.N.Z. and S.A.I.S.M.G.: review and editing.

Conflict of interest: Authors state no conflict of interest.

Ethical approval: The conducted research is not related to either human or animal use.

Data availability statement: The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

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