

Review

Outstanding Graphene Quantum Dots from Carbon Source for Biomedical and Corrosion Inhibition Applications: A Review

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Abstract: Graphene quantum dots (GQD) is an efficient nanomaterial composed of one or more layers of graphene with unique properties that combine both graphene and carbon dots (CDs). It can be synthesized using carbon-rich materials as precursors, such as graphite, macromolecules polysaccharides, and fullerene. This contribution emphasizes the utilization of GQD-based materials in the fields of sensing, bioimaging, energy storage, and corrosion inhibitors. Inspired by these numerous applications, various synthetic approaches have been developed to design and fabricate GQD, particularly bottom-up and top-down processes. In this context, the prime goal of this review is to emphasize possible eco-friendly and sustainable methodologies that have been successfully employed in the fabrication of GQDs. Furthermore, the fundamental and experimental aspects associated with GQDs such as possible mechanisms, the impact of size, surface alteration, and doping with other elements, together with their technological and industrial applications have been envisaged. Till now, understanding simple photo luminance (PL) operations in GQDs is very critical as well as there are various methods derived from the optical properties of manufactured GQDs can differ. Lack of determining exact size and morphology is highly required without loss of their optical features. Finally, GQDs are promising candidates in the after-mentioned application fields.

Keywords: graphene quantum dots; synthetic approaches; biosensors; energy storage; corrosion inhibitor applications



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1. Introduction

Carbon is an outstanding material and a more abundant element in the form of coal. It is considered as one of the world's major sustainability matching with the green approach. The graphene shape has recently astonished the scientific community [1,2], as did the football fullerene shape, which was discovered with small needle-shaped carbon nanotubes (CNTs) in 1985 and characterized for the first time in 1991 [3,4]. These recent findings of unique carbon allotropes have given scientists, from all disciplines great interest and fascination. Zero-dimensional (0D), one-dimensional (1D), two-dimensional (2D), and three-dimensional (3D) graphite are included in the classification of carbon into graphite forms [5–8], as shown in Figure 1. Because of the van der Waals force between layers, 2D graphene is a single-layered nanomaterial that differs from multilayer graphite [9]. In the universe, the strongest and thinnest substance ever weighed is an atom-thick carbon material [10].

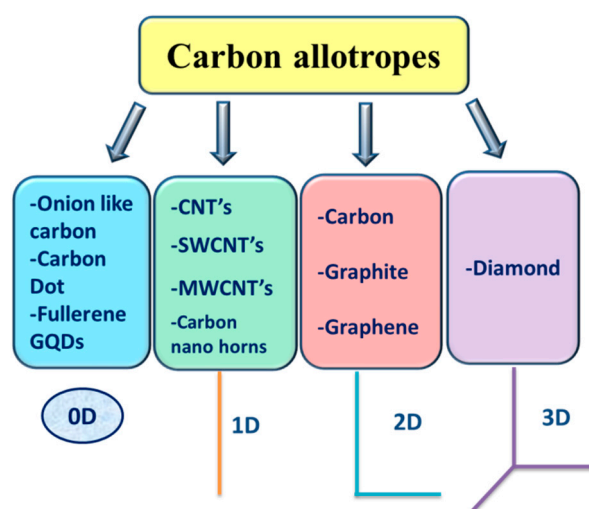


Figure 1. Carbon Materials Category.

Graphene was discovered at the University of Manchester in 2004 and since this time it is considered the marvelous substance of the 21st century [11]. The spectrum of materials bonded to graphene and graphene-based nanostructures are similar, but with different nomenclature, meaning that the carbon content contains one or more monolayers of graphene [12,13]. Furthermore, for the delicate handling and processing of graphene and its derivatives, many advanced techniques are now available. These techniques are used to manufacture items of various sizes from waste contents, such as C, O, H, or by manipulating surface groups, like hydroxyl, epoxy, carbonyl, and carboxyl [14,15]. In principle, there are four groups of different carbon materials, including carbon quantum dots (CQDs), carbon nanodots (CNDs), polymer carbon dots (CPDs), and graphene quantum dots (GQDs) with carbon dots (CDs) being used as a generic term [16]. Importantly, CDs families can be categorized based on their surface groups, properties, and basic structures of the carbon core. In this respect, CQDs have a crystal lattice accompanied by a spherical shape with surface chemical groups and also they have a quantum confinement effect (QCE) with luminescence features [17]. Moreover, it is possible to adjust the photoluminescence wavelength provided by the CQDs by changing its size [18]. Although GQDs have an apparent graphene lattice, they consist of one or more layers of fragments of graphene. Usually, the height of these GQDs is less than ten graphene layers with a transverse dimension of less than 100 nm. The surface groups in GQDs are adjacent to the defects or edges of the intermediate layers, impacting the different QCE and edge properties. QCE in GQDs are not only based on their size but also described in the planes of graphene by isolated conjugated π -domains, chemical groups, and a high carbonation degree [18]. QCE does not play a part in the properties of photoluminescence and is predominantly determined in the carbon core of graphite by the subdomain states and defect states. Carbon polymer dots (CPDs) are another class that contains a hybrid carbon/polymer structure in which the surface and center of the carbon are connected to a large number of functional/polymer classes [19–21]. Additionally, the key culprits for photoluminescence properties of CPDs are surface states, subdomain states, molecular states, and the crosslinking effect of radiation [22].

A comprehensive study of the assessment and classification of carbon dots has been reported based on properties and structure, with a particular focus on designing 0D GQDs in 2D lines [23,24]. This material form has many outstanding properties, such as good chemical inertia, excellent biocompatibility, high solubility parameter, fluorescent activity, photostability, emission of luminescence, long-term resistance to photobleaching, wide surface area, and better surface grafting [25,26]. In turn, these characteristics allow the study of new structural, optical, and electrical phenomena that are not present in other materials.

Over the past few years, a variety of carbon nanomaterials, such as high-surface-area, compatibility, prominent electron transport, excellent mechanical strength, and hydrophilic-

ity have received extensive interest with graphene and graphene oxide (GO). The existence on its surface of different functional oxygen groups (carbonyl and carboxylate) which make GO with high hydrophilicity. The GO nanosheets have a high surface area that may also be an effective place to decorate and disperse other inorganic nanoparticles such as TiO_2 ; in a recent study, reduced graphene oxide/ TiO_2 nanocomposite was used for modification of thin-film nanocomposite reverse osmosis membranes. The rGO/ TiO_2 nanocomposite, as a hydrophilic additive, was synthesized using a facile hydrothermal method the result demonstrates that The membrane containing 0.02 wt. % rGO/ TiO_2 nanocomposite observed excellent RO performance involving 51.3 L/m² h flow of water, 99.45% NaCl rejection and exceptional chlorine good resistance [27,28]. Furthermore, graphene oxide sheet has also used with Ce ions in the photocatalytic efficiency of magnetite for photodegradation of oxytetracycline [29].

These unique features make this structural carbon category a promising candidate for different utilizations, such as biosensors, energy storage, bioimaging, and in redox electrochemical reactions by modified graphene surface of the TX-100 to understand its Electrochemistry interface [30–32].

GQDs demonstrate premium solubility in inorganic solvents such as THF, DMF, and further acetone. Further, DMSO and ethanol are commonly used solvents that provide good solubility. However, the applications of GQDs in bioimaging and selective drug delivery systems have been greatly affected by the increased solubility in aqueous solvents [33]. Moreover, the potential to be water-soluble is due to the units containing hydroxyl and carboxyl attached to the GQDs edges since the hydrophilicity of GQDs is enhanced by surface functionality, and it can be controlled by synthetic methods and hence, tuning the chemical properties [34].

Highly reliant upon the technique of GQDs synthesis was obtained, research is at a relatively early stage, considering the many superior advantages and properties; hence, many of the shortcomings of GQDs have yet to be overcome. Although many significant advantages and promising applications are possible, more research is required to enhance material properties and resolve some constraints. Moreover, electrical behavior resulting from the small size of GQD exhibits a quantum size effect. In addition, several obstacles must be met to take advantage of these specific properties [35]. The synthetic methodology of GQDs by the chemical route results in a remarkable heterogeneity in the size and the functionality of the surface. With such a broad variety of chemical features and measurements, the function of its particular properties is difficult to study [36]. Moreover, GQD's photoluminescence and quantum confinement characteristics are highly dependent on size [37].

Various methods of synthesis can account for the major difference in chemical structure and scale. Thus, it is important to recognize the connection between dimensional variation depending on the synthesis process and optical properties in terms of applied and fundamental perspectives [38,39]. A fascinating report of the GQDs synthetic process was primarily evaluated on the size-dependent photoluminescence properties and the quantum size effect of GQDs [40]. Furthermore, because of their superior properties, the benchmark shows continuous improvement in functionalized and critical GQDs applications [41].

Carbohydrates are one of the most diverse and important classes of biomacromolecules in nature and provide well-defined chiral scaffolds ready for modification of the anomeric position and functionality of alcohol. Therefore, the use of carbohydrates as a starting material for GQDs synthesis is extremely attractive not only for their large quantity, availability, and heterogeneity, but also for their high-water solubility, low carbonization temperatures, low cost, and lack of toxicity. Not surprisingly, with all of these tuning options for GQDs synthesis, researchers have already begun to see the benefits of carbohydrates when they consider synthesizing new GQDs with improved properties. For example, simple monosaccharides such as glucose, glucosamine, mannose, fructose, and their common derivatives and disaccharides, such as sucrose, lactose, and maltose, have been used to create GQDs by various methods. Likewise, important natural biopolymers based on carbohydrates such

as cellulose, dextran, β -cyclodextrin, chitin, chitosan, and hyaluronic acid, which vary not only in their basic composition but also in their physical and chemical properties which have been successfully used to obtain GQDs [42–44].

Polycyclic aromatic hydrocarbons (PAHs) are an organic hydrocarbon with two or more fused benzene rings. PAHs are aromatic compounds mainly produced by the natural manner and they are mostly toxic. The significant effort to degrade and track potentially such dangerous substances is desperately needed [45]. So, by using commercially available PAHs as precursors, we present a simple and efficient approach to PL GQDs based on the bottom-up approach. The PL GQDs obtained have 5–10 nm sizes and 0.5–2 nm thicknesses and shows better solubility of water and tunable fluorescence. We also show that, because of their stable fluorescence and low toxicity, the PL GQDs are not only promising for bioimaging but also effective for Fe^{3+} and hydrogen peroxide sensing [46].

In this review, in terms of size, convexity, surface, and solvent compliance, the methods currently proposed for the functionalization of GQDs and their properties were examined. Furthermore, it addressed pristine and modified GQDs applications in sensors, energy storage, biological fields, and corrosion inhibitors. This review will eventually provide data and create a new insight to understand the properties/application relationship of GQDs.

2. Synthetic Routes of GQDs

The primary and key method before using a material for a specific application is material synthesis. The variance in application outcomes usually depends on the morphology and material properties, which are primarily determined by the method of synthesis [47]. GQDs consisting of carbon-rich materials used as precursors, such as graphite, polysaccharides, fullerene, graphene oxide (GO), CNT, and carbon fiber (CF) [48,49]. To synthesize GQDs, there are two main methods used, namely, top-down and bottom-up strategies. These two techniques are difficult to synthesis the quantum dots in traditional semiconductors. Subsequently, as shown in Figure 2, carbonization or controlled synthesis techniques have been implemented to produce GQDs from acceptable organic molecules or polymers [50,51]. Furthermore, the different between top-down and bottom-up approaches were illustrated in Table 1

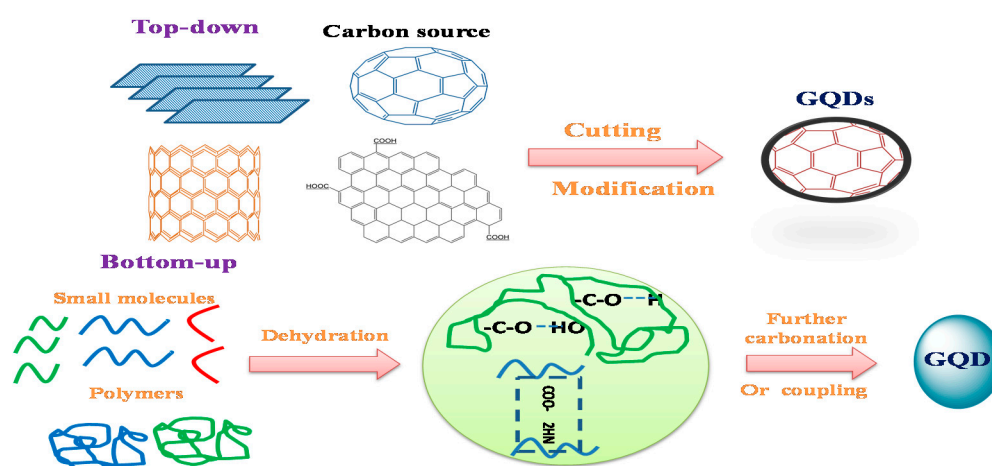


Figure 2. Two techniques for the production of fluorescence graphene quantum dots (GQDs): “top-down” breaking from complex particles and “bottom-up” from small molecules.

To obtain broad GQDs, controlled synthesis is precise but complex and involves several processing steps. However, tiny molecules or polymers suitable as GQDs are obtained by dehydration or combination when using the carbonation technique [52]. Sometimes, these procedures are out of balance, resulting in non-uniform GQDs proportions. Fortunately, due to the use of non-toxic reagents, GQDs are biocompatible. Further, as far as we know, the degradation of the carbonaceous material includes much of the top-down

synthesis process. However, in terms of low efficiency, unforeseen structural damage, and heterogeneous morphology, these approaches have major drawbacks [53].

Table 1. Comparison between top-down and bottom-up approaches.

Basis for Comparison	Top-Down Approach	Bottom-Up Approach
Basic	Successive cutting or grinding of bulk material to get nanoparticles	The buildup of material from bottom: atom or molecule to get nanoparticles
Starting materials	Solid-state	The starting material is either gaseous or liquid
Processing method	Physical method	Physical and chemical methods
Advantages	• Large scale production: • Deposition over a large substrate is possible • Chemical purification is not required	• Ultra-fine nanoparticles • Deposition parameters can be controlled • Cheaper method
Disadvantages	• Broad size distribution • Varied particle shape • Control of deposition parameters is very difficult • Expensive technique	• Large scale production is difficult • Chemical purification of nanoparticles is necessary

Oxidative degradation, hydrothermal/solvothermal processes, microwave/ ultrasonic processes, electrochemical oxidation, and chemical vapor deposition (CVD), pulsed laser ablation (PLA) are the most important top-down approaches mentioned [54–56]. The bottom-up methods, on the other hand, provide a controlled synthesis and provide good carbonation with a reasonable size range, high brightness, and satisfactory properties of the synthesized GQDs [36,51].

Recently, the effective use of starch as an innovative material for the synthesis of GQDs. Biocompatibility and imaging potential of synthesized GQDs were evaluated using MTT assay and CaSki cell lines, respectively [57]. Further, GQDs can be synthesized from fructose as a precursor in which hydrochloric acid and ethylenediamine were found to be strong chlorine and nitrogen donors for chlorine and nitrogen co-doped GQDs, respectively, which showed excellent stability [58].

On the other hand, the carbonization of sugar resulted in the breakdown of glycosidic linkages via dehydrogenation into elemental carbon. In this study, when subjected to serial MW heating with hydrothermal treatment glucose pyrolyzed to form GQDs and the control on GQDs size can be achieved by MW heating time [59].

In another work, investigated an effective utilization of the potential industrial by-product such as sugarcane molasses (SMs) for single-crystalline sulfur-doped (S-GQDs) synthesis via the hydrothermal method, the remarkable property of S-GQDs demonstrated by labeling the cytoplasmic area of HepG2 cells in-vitro with minimum uptake by normal DF-1 and HEK 293 cells [60]. Furthermore, the emulsion-template carbonization (E-TC) method has been used for the synthesis of GQDs by using honey and n-butanol water in oil emulsion by simple heating offering a quantum yield (QY) of about 3.6% [61].

In another example, efficient synthesis of GQDs using rice grains as a carbon source. Heating of starch powder has resulted in the formation of glucose oligomers, further heating of these oligomers offers nucleation and pyrolysis resulted in black carbonaceous powder comprising GQDs [62].

In a recent study it was designed a fluorescence-responsive sodium hexametaphosphate sensor depend on a reduced graphene quantum dot/chitosan formula for ALP, the

bright blue emissions rGQDs with high negative charged hydroxyl group was prepared using NaBH_4 . In the production of the probe for ALP detected was fabricated by the combination of rGQD and chitosan via auto assembly. The chitosan charged biopolymer simultaneously displays the transformation-induced fluorescence quenching of rGQDs as well as electrostatic appeal. The method established shows good ALP selectivity and has promising results when applied to real test samples. This low-cost toxin-free test offers a new approach to the biosensing of the r GQD and biopolymer systems [63].

2.1. Top-Down Technique

2.1.1. Liquid Peeling Process

Owing to their scalability, liquid peeling (LP) of the 2D materials has a great focusing. Further, LP is the best way to manufacture nanofilms with various advantages such as low-cost processing, ease of use, and reduced environmental effect [64]. Graphite is exfoliated into graphene sheets during this process as a precursor, so the LP process can acquire GQDs with strong crystallinity. Moreover, precursors of low and high defects (edge and surface defects) such as graphite powder or carbon acetylene powder have been used for the production of GQDs in this process [65]. Lately, after the intercalation, graphene was produced by LP of graphite and has gained a lot of attention. Inspired by this synthesis of the GQDs by LP probe sonication using a graphite powder by high-intensity ultrasonic waves. These waves split layers of graphene into ultra-fine particles, or GQDs are involved in the reaction. The resulting GQDs were extracted with different solvents based on the water and DMFs; the particle size was determined as shown in Figure 3 [44,45]. The GQDs obtained in water is comparatively smaller than DMF extracted particles. The mean GQDs size and height were 4.1 nm and 1.2–1.7 nm. By calculating the number of layers, it is composed of 2–3 graphene layers with 0.34 nm layer spacing. This process does not involve a carbon source as a precursor compared to other approaches, so there is a benefit to this approach. Further, by adjusting the parameters of the LP process, the physicochemical properties of the GQDs might be regulated [66].

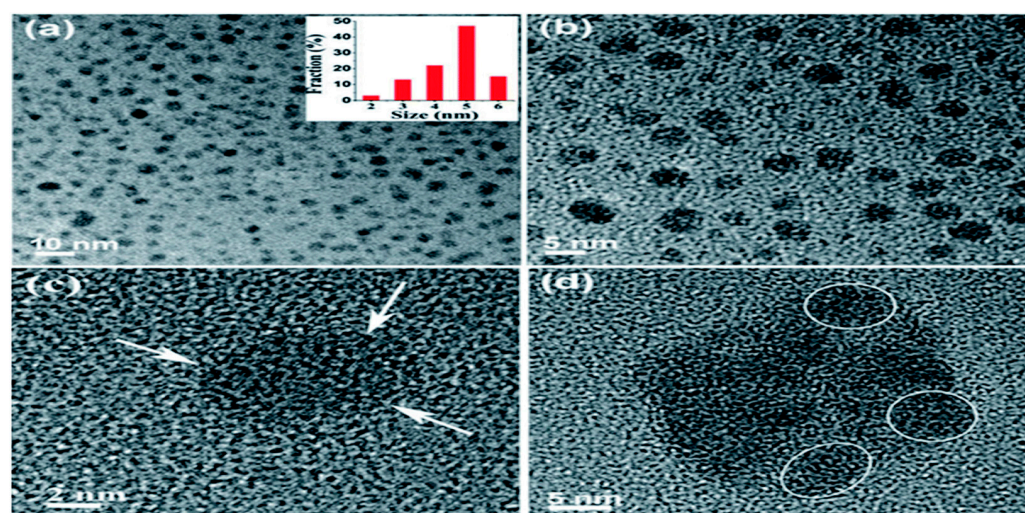


Figure 3. (a,b) TEM photos of GQDs in water, the figure displays the distribution of GQDs in water. (c) HRTEM single-GQD image. (d) Minor agglomeration of GQDs in DMF. Reproduced with permission from ref [67], copyright 2016, The Royal Society of Chemistry.

2.1.2. Hydrothermal Method

The decomposition of carbon nanoparticles into GQDs from carbon raw materials by the hydrothermal method is a favorable way of getting GQDs from powerful oxidants such as HNO_3 , H_2SO_4 , and H_2O_2 [66]. A benefit of hydrothermal GQDs synthesis is the ability to adjust the size of the GQDs particles by applying various hydrothermal temperatures as well as the hydrothermal temperature rose, the GQDs particle size decreased. For example,

it was reported that the hydrothermal technique is validated for the synthesis of GQDs from powdered GO as a source of carbon and H_2O_2 as a reagent. In this process, H_2O_2 split up into OH radicals at elevated temperatures. Followed the hydrothermal reaction, the graphite plates were thermally breaking into small fragments. These findings show that at elevated temperatures, graphite disintegrates easily [68]. Recently, a hydrothermal method was developed to generate blue luminescence GQDs. In short, using a mixture containing controlled oxidants (H_2SO_4 and HNO_3) under the action of ultrasound, layers of graphene were cut into tiny bits. Under hydrothermal conditions, the leaves were regenerated at a high temperature using a lined autoclave filled with Teflon to exhibit oxidized tiny graphene. The resulting GQDs had a mean diameter of 39.6 nm, composed of 1–3 graphene layers, and used quinine sulfate as a guide to display a quantum yield of 6.9% [69].

On the other hand, the one-pot hydrothermal process is used to synthesis functionalized GQDs using polyethylene glycol (GQDs-PEG). In this process, GO and PEG plates were the starting materials, and a uniform diameter ranging from 5 to 25 nm was shown by the resulting monodispersing of GQDs-PEG. Compared to naked GQDs, the resulting GQD-PEGs displayed PL quantum yield with 360 nm emission using rhodamine B which is used for comparison by about 28% and also showed excellent luminescence properties [70].

2.1.3. Electrochemical Method

Recently, using an accessible electrochemical technique to formulated PL yellow-green emission GQDs was assessed. An aqueous solution of NaOH is prepared as an electrolyte through the GQDs synthesis process. Then, the graphite rod and platinum foil, which acted as both the anode and counter electrode were soaked in an aqueous solution and applying the 5.0 v voltage for 6 h. The color of the homogeneous solution was changed from brownish to dark black along the reaction time and by filtration, GQDs were eventually obtained [71]. The GQDs synthesis by the electrochemical oxidation procedure has good stability, but perhaps the pre-treatment of raw materials and the cleansing of GQDs take a very long time, and the quantum yield is considerably low, making it difficult to achieve large-scale production of GQDs [72]. However, LP is among the most promising method for synthesis of 2D nanosheets with unique benefits, such as low production costs, ease of operation, or minimal environmental impact. Furthermore, the graphite is exfoliated to graphene layers during the long LP phase and is mostly very thin (quantum size) with low-defect Nano graphite powder and highly defective acetylene carbon powder were prepared via LP [73].

2.2. Bottom-Up Methods

2.2.1. Hydrothermal Method Using Microwaves

To produce GQDs, the hydrothermal process takes more time in most cases. A fast, microwave-assisted method, namely the microwave-assisted hydrothermal method (MAH), has therefore now been chosen to synthesize GQDs which share the hydrothermal and microwave advantages methods when assisting with a microwave oven [74]. Glucose as a precursor for producing water-soluble GQDs was used in the presence of a microwave oven. Here, microwave heating support uniform, stable, simultaneous, and rapid heating which accelerates the development of a uniform distribution of quantum dots across the size. The GQDs radiation energy detected was 4.1 eV, which is the maximum radiation energy compared to other QDs at the shortest radiation wavelength [75].

It was reported that GQDs was excited before by a laser of 197 nm. Figure 4 shows that the MAH method using glucose as a precursor to produces GQDs. From Figure 4b the GQDs formation process was attained by different function moieties without surface passivation or required inorganic additives anymore [59]. Furthermore, to form nucleation crystals and reactive functional groups bound to the GQDs surface, glucose molecules were dehydrated. The molecules of glucose are pyrolyzed and quickly mutated into GQDs [76].

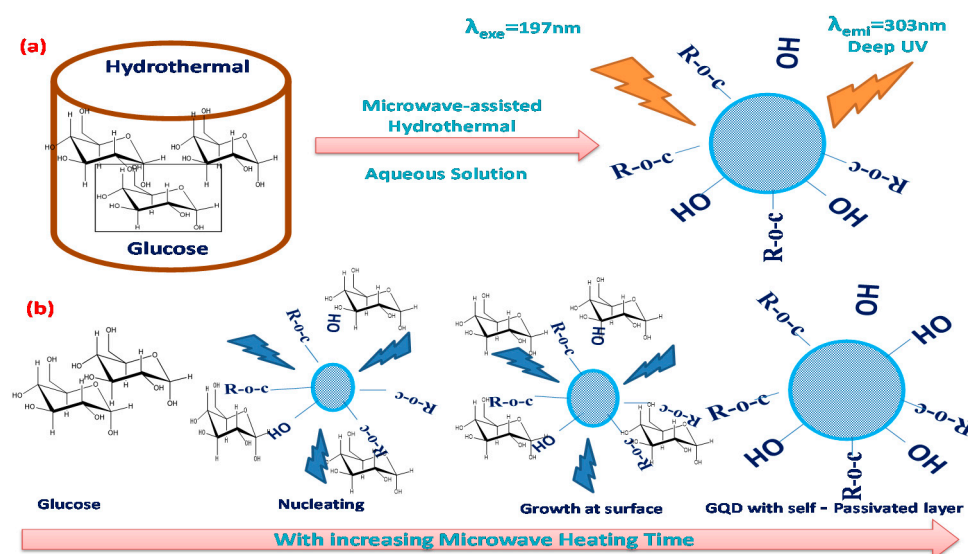


Figure 4. (a) Schematic illustration of GQDs synthesis through the microwave-assisted hydrothermal (MAH) technique and (b) schematic diagram for GQDs formation process with functional groups.

Monodispersed GQDs were synthesized in another study using a GO as a preliminary material after subjecting to microwave irradiation for 5 min at more than 190 °C. Then using sodium carbonate, the pH was neutralized, and the solution was centrifuged. The resultant supernatant was eventually isolated as GQDs. By induction and doping, the GQDs solvents can also be synthesized [77].

2.2.2. Method of Soft Template

In general, this approach was a facile, convenient process to develop nanostructures rather than traditional synthetic routes. This technique effectively enables to monitor the shape, size, and surface texture of nanomaterials, which is the key benefit for elucidating the characteristics of any nanoparticles. Based on its distinct form, the template system is divided into soft templates and hard templates [78,79]. The soft template method is very appropriate for GQDs output in contrast to the hard template method. In processes of separation, purification, and mass processing, it can promote the typical nanoscale reaction vacuum smoothly.

In another study, 1,3,5-triamino-2,4,6-trinitrobenzene (TATB) was inserted as a highly symmetrical planar structure with 6 heavy bonds of intramolecular hydrogen amongst the functional groups $-\text{NH}_2$ and $-\text{NO}_2$. As a carbon source, the structure of a graphite-like sheet was used and the model TATB was originally hardened during the thermal phase to crack chemical bonds and produce gases such as NO, NO_2 , and H_2O . Then, because of gas expansion, the graphite-like TATB was mounted in one layer [80].

Further study was obtained 60 nm wavelength disc-shaped monodispersed GQDs using Hexa-peri-hexabenzocoronene (HBC) as a precursor and prototype. The HBC molecules were folded during the pyrolysis stage into a faulty graphite structure in this process. Using a modified Hummers process, the graphite was oxidized, exfoliated, and then reduced with hydrazine hydrate to give a pure form of GQDs [77,81].

2.2.3. Metal Catalyzed Method

This method is an unusual method for processing economically and ecologically GQDs. Recently, ruthenium (Ru) was used as the catalyzed metal and C60 was used as a precursor for GQDs synthesis. Surprisingly, this catalyzed metal process was used to observe the shapeshift of the GQDs at various annealing temperatures, Figure 5a by scanning tunnel microscopy (STM), triangular, and hexagonal GQDs shapes were observed when the samples were recycled at different temperatures for 2 min, as shown in Figure 5b,c.

However, the metal catalyst and the particular structure of the starting material are rarely used in this process to obtain the GQDs [82].

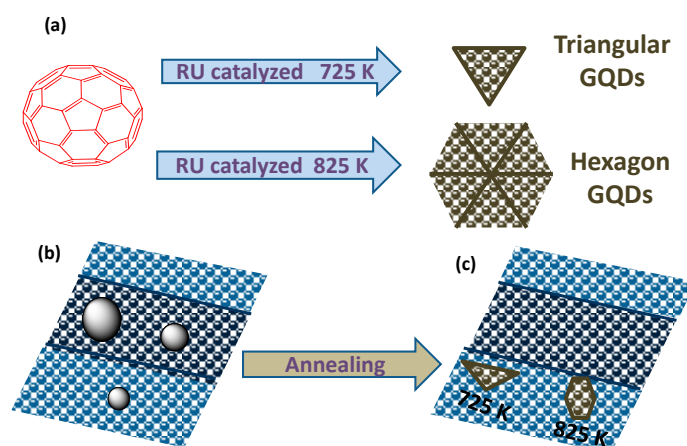


Figure 5. The shapeshift of the GQDs at various annealing temperatures. (a) C₆₀ was catalyzed cage-opening under different temperatures as a precursor for GQDs synthesis using Ru metal (b,c) C₆₀ molecules adsorbed on the terrace and growth of GQDs with various equilibrium structures from the diffused carbon cluster.

2.3. Green Synthesis

In addition to the numerous techniques described in the aforementioned sections, scientists sought to detect and acquire greater GQD development methods. To achieve specific features and properties, GQD synthesis was highly concerned. All the described methods used for preparing the semiconductor quantum dots are a beneficial effort to develop greener and low-toxicity solutions. Although its efficiency is high, the real battle is to remove by-products accompanied by the reaction such as minerals, acids, and inorganic salts. In the large-scale processing of preparing material with a strong crystallinity pattern, it is impossible to apply these approaches. Otherwise, GQDs are environmentally benign, virtually non-toxic, and photostable compared with QDs and organic dyes in semi-conductors. GQDs are synthesized using green-synthesized methods from various sources of carbon such as peels, food waste, algal blooms, yeast, human urine, etc. The green chemistry methods have many differences in morphology and UV-vis absorption in contrast to traditional synthesis [83–86]. In literature, many materials are validated for adsorption and photocatalytic degradation of toxins [87–98]. It was reported that GQDs were synthesized using corn powder as a precursor and green material and the photodegradation against rhodamine B (RhB) by UV radiation was tested. In this study, GQDs/TiO₂ exhibits superior photocatalytic activity in RhB degradation compared to conventional TiO₂ material, which is about 53% degradation efficiency within 80 min [99]. In another study, GQDs are processed using hydrothermal methods that are effective and environmentally safe, using polymeric macromolecule starch. The obtained products were GQDs, water, and carbide precipitate was also obtained, and the GQDs had a diameter of between 2.2 and 3.3 nm [100].

In a recent study, GQDs at temperatures between 150 and 200 °C for 6–10 h were obtained using nutmeg seeds via a hydrothermal green process and its general properties were investigated. The obtained GQDs manifest heavy optical absorption from 260 to 320 nm in the UV region [101]. Moreover, the primary benefit of using these precursors is that they are readily available, simple to use, and often non-toxic. Depending on the functionality present on the surface, functionalization and carbonization can be accomplished using biomass with low reaction temperatures as a carbon source with distinctive fluorescent properties using the green chemistry process [102,103].

Chitosan is a natural biopolymer with different applications with high bioavailability and biodegradability. In a recent study, GQDs can be green synthesized by cross-linked

for chitosan (CS) using citric acid. This prepared GQDs-crosslinked CS hybrid bio-nano composite beads were filled with sodium salicylate (SS) as a drug model (CS-GQD/SS) and protected with pH-sensitive biopolymeric carboxymethylcellulose (CMC). CMC and CS synergistic effects have long improved the stability of drug-dose regulation in the conditions of the gastrointestinal tract [104].

3. Photoluminescence (PL)

The optical properties of GQDs were studied by PL spectroscopy and determine their absorption in the near-infrared area which exhibits various absorption and luminescence properties. GQDs are most commonly represented as a high absorption peak resulting from π - π^* transition. In addition, it is possible to find slight absorption peaks associated with the n - π^* transition at longer wavelengths [105]. GQDs keep the center of graphene and indeterminate chemical groups on their surface, so the graphene center and neighboring chemical groups coordinate photoluminescence. Furthermore, the basic characteristics of GQDs depend mainly on shape and size which represent the main factors in the localization of absorption peaks [106]. Moreover, the edge structure, functional groups, solvents, and temperature plays an important part which is affected by the fundamental properties and other determining factors [107].

Many published works reported that the GQDs disturbance is caused by its size and thus, the size of the GQDs decreases based on increasing in bandgap value, it may adjust its PL. Further, due to a shift in the bandgap and chemical functionalization, PL emissions can change. Moreover, GQDs display greater photoluminescence than other materials based on carbon [108]. Basically, GQDs showed PL due to changes in synthesis parameters, such as concentration, size, pH, solvent, and the wavelength of excitation, while PL was largely dependent on quantum confinement, edge effect, composition, structure, and form. Furthermore, different PL spectra and GQDs excitation, respectively, at different pHs were discussed before. The PL spectra color is related to the GQD sizes and reflects the energy bandgap for different GQD sizes. The GQDs PL intensity is also shown to be sensitive to solvents. In acetone, DMF, THF, and water, the peak moved from 475 to 515 nm. In general, the shifting wavelength of excitement depends on the emission wavelength and in photoluminescence spectrums, the peak intensity is typically found. The changing wavelength of arousal often changes to a higher wavelength of the wavelength of emissions [109].

4. Application of GQDs

4.1. Sensors

A sensor is a machine, device, or module that intended to determined events or environmental changes and transmits information to other electronic devices, often the processor of a computer. Because of their unique properties, such as a fixed bandgap GQDs showed great interest in many applications. It is proved that GQDs serve as a high sensing material owing to greater affinity of electron movement with a steady reaction rate which gives it excellent candidates in sensing utilization. The existence of field-effect transistors, electrochemical sensors, PL sensors, Electrochemiluminescence (ECL) sensors, glucose sensors, and bioimaging has previously been investigated in GQDs [67].

The glucose sensor is the general system of detection that uses different GQDs containing polar -COOH and -OH surface recoveries and cationic corrosive bipyridinium (BBV) salt to operate under natural conditions [110,111]. GQDs in this sensor provided as fluorescent components and BBV act as a rapid glucose receptor and good fluorescence quencher [112]. A decline in the PL intensity of the GQDs was prompted by the obsession between the GQDs and BBV. Boronic acids were turned over to tetrahedral anionic glucoboronate esters at the time of glucose processing, which sufficiently destroyed the cationic bipyridinium net charge. The extinguishing output was therefore reduced and the PL of GQDs efficiency was reduced. Therefore, sensitive and precise identification of glucose was found using the observed PL shift [113,114].

Recently, biocompatible polysaccharide chitosan/Thiolated graphene quantum dots cross-linked and modified by gold nanocomposite which used for immobilization of ractopamine (RAC) aptamer [115]. Further, chitosan-based N-doped GQDs nanocomposite explored the first time for selective electrochemical sensing of dopamine in human urine. Chitosan charges play an interactive role by hindering the ascorbic acid interference and enlarging the peak potential separation in both dopamine and uric acid [116].

4.1.1. Sensor Photoluminescence

GQDs have an impressive blend of graphene and QDs properties. In the chemistry and biology field, GQDs produced vast scales of study. The photosensitive approach was recently used as an enabling technology to detect metallic ions by investigating raw and surface adjusted GQDs fluorescence. While several advantages of GQDs have been successfully achieved as sensor components for the detection of metal ions, GQDs are relatively low quantum efficiency which hindered their detection sensitivity. The concept of doping heteroatoms in GQDs, such as N, B, sulfur, and P were introduced to increase quantum yield and to overcome these limitations. In sensing applications selectivity is also a critical variable during application. The selectivity and the functionality of GQDs have been evaluated using a lot of metal ions, such as Ag^+ , Cu^{+2} , Mn^{+2} , Co^{+2} , Cd^{+2} , and Ni^{+2} [117,118]. Advances in the creation of sensors based on the special properties of GQDs for the detection of biomaterials, small organic molecules, and metal ions are common. Doping agents or functionalized GQDs have been proposed for the improvement of sensitivity, selectivity, and specificity. Furthermore, a change in the optical properties of GQDs, such as surface adsorption of ions or molecules, can alter the bandgap, resulting in a saturation effect or an increase in PL hopefully to build sensors [119]. GQDs have been extensively studied as a possible candidate for the identification of a large range of higher sensitivity and selectivity of different analytes. Furthermore, GQDs are a non-toxic substance, in contrast to other hazardous QDs. They are readily soluble and have optical and electronic properties that are comparable [120].

Using GQDs as a heteroatom-doped detection portion, the identification of metal ions and nitro compounds can be harmful to general health and the surrounded media. To evaluate the existence of metal ions, most researchers have used the fluorescence quenching effect process [121,122]. On another hand, an effective method for determining ascorbic acid (AA) in human serum based on the fluorescence intensity of GQDs was suggested by Liu et al. In the detection range and borderline area of 1.11–300 μM and 0.32 μM , respectively, the tailored results showed a significant and satisfactory linear response for AA. The presented theory was simple, inexpensive, and more sensitive and selective than other methods [123].

The GQDs (N, S-GQDs) sensor doped with nitrogen and sulfur has been synthesized and studied for elucidating nitro-explosives. Moreover, sulfur-doped GQDs (S-GQDs) fluorescence quenching metal ion sensor to detect silver ions. In a further study, the authors achieved a successfully synthesized nitrogen doped GQDs (N-GQDs) to determine Fe^{+3} based on the quenching effect of GQDs fluorescence intensities with a good linear range between Fe^{+3} concentration and fluorescence intensity over a broad range of 1.0 μM to 2.0 mM In this study, the detection limit was 70 nM, which an excellent response was observed [124–126].

4.1.2. Electrochemiluminescence (ECL) Based Sensor

ECL is an effective method owing to the merge between electrochemistry and chemiluminescence to recognize Electrochemiluminescence. It depends on the emission of light during the transfer of electrons from an excited state, and it is produced in the medium of radical cations and luminophore anions. It transforms radiative energy from electrochemical energy through an applied potential on the electrode surface. The light signals from the exciting status of the ECL luminophore provided by the electrode are found during the electrochemical reaction [127]. The most surprising benefit of the ECL is that it does not re-

quire external sources of light. By adjusting the electrode potential, the position and timing of the ECL emission are changed. Further, high sensitivity, cheap and compact instruments are other significant factors associated with this. Moreover, ECL can be more effective than chemiluminescence because, by adjusting the electrode potential, the production of states can also be selectively coordinated [128,129]. The benefits of this are the lack of a simple mechanism, optical context, refined selectivity, sensitivity, and a wide variety of responses. GQDs are used based on their ECL activities to detect such dangerous metals.

An ECL sensor in GQDs/peroxodisulfate (GQDs/S₂O₈²⁻) for the detection of naturally toxic hexavalent chromium (Cr (VI)) was reported in a recent study. The authors developed an ECL sensor based on GQDs/S₂O₈²⁻ with different parameters and achieved a linear range of 50 to 60 nM. To detect Cr(VI) in displaced river water, similar types of sensors have been developed [130]. A new and simple method for ECL determination has also been developed by another study which uses the N-GQDs/chitosan polymer film to record a high sensitivity and convenience of nitroaniline (NA). In this study, the authors found that NA signals with HCL and NaNO₂ were successfully amplified with a detection limit of 0.005 $\mu\text{mol L}^{-1}$ for linear detection of NA over the range 0.01–1 $\mu\text{mol L}^{-1}$ [131].

4.1.3. Electrochemical Sensor

Environmental pollution is caused by the vast spread of further organic and inorganic contaminants, especially air pollution, which contains unknown chemicals that affecting on agriculture, drinking water, and human health that is the biggest environmental issue at present. The development of analytical instruments such as electrochemical sensors is motivated by growing concerns about the diffusion and effects of chemicals from the environment. A chemical sensor included an electrochemical sensor is a type that the electrical signals are obtained from the transformation of the chemical reactions of electrode analytes. According to the types of electrochemical sensors, more information is provided about their environment.

Further, the methods of electrochemical detection and analysis are cheap, sensitive, and facile to use, so that data can be accessed in remote areas. Currently, many electrochemical sensors are compliant and environmentally friendly. These types of electrochemical sensors consist of molecular sensing signal conversion with very small sizes and have a broad spectrum of applications in the areas of biological monitoring, clinical diagnosis, food science analysis, and environmental detection [132].

Layered nanomaterials are the most talented immobilized molecules and have recently attracted attention due to their remarkable properties such as costless, ease of use, high sensitivity, and online estimation. To greatly increase the stability of the resulting sensors and the sensitivity, behavior of the immobilized molecules, these layered nanomaterials are mixed with other polymers or nanocomposite. The electrochemical sensors are distinguished, along with the optical mass and heat sensors, by their great recognizability, simplicity, and low cost. So, in recent decades, electrochemical have generated a great deal of interest in analytical chemistry and today hold a prominent place among the currently available sensors [133].

The various types of electrochemical sensors are classified into potentiometry, conductometry, and amperometry, or voltammetry, according to the calculation of the used electric signal. An electrochemical sensor for the detection of bisphenol A (BPA) in water was adopted in water by pulse differential voltammetry (PDV). This sensor consists of polypyrrole (PPy) and GQDs composite electrodes. With a good linear range and detection limits of 0.01 to 50 μM and 0.04 μM , respectively, the sensor showed a good response [134].

A facile and ultrasensitive electrochemical biosensor for the detection of miRNA-155 using GQDs has been developed by Hu et al. with a low detection limit of 0.14 fM and direct detection of miRNA-155 in human serum albumin. A great promising criterion in the clinical detection was obtained for this type of sensor which exhibited great potential in sensitivity and selectivity [135].

4.1.4. Humidity Sensor

Confidence in low-power portable sensors has grown tremendously. Therefore, under different conditions, scientists have developed various sensors and designed them according to the climate. In various fields, such as everyday life, health, medicine, nature observation, biology, automobiles, meteorology, prescription, food preparation, etc., humidity sensors have found support for their various applications and play a central role in stimulating human life. It also has many disadvantages, particularly for corporations and innovations. The vapor comprising incredibly intuitive dipolar particles that are consolidated or disappeared from the surface was such a crucial location (as a result of shifts in the electron negativity of the atoms of hydrogen and oxygen), even with a small distinction between temperatures. Therefore, it is easy to track and regulate the humidity of the ground. The constant monitoring of moisture in a variety of fields is important, such as soil sticking control, bundling business, semiconductor manufacture, nutrition preparation and medicine industry, structural construction, electronics, residential devices, and chilling frameworks [136,137]. Material choice is very necessary and very difficult in the development of humidity sensors. A large number of materials have been used, such as polymers, metal oxides (MOs), and materials based on carbon and their compounds [138,139]. Over the entire spectrum of relative humidity (RH), the analysis of these materials must be highly sensitive. Moreover, due to their moisture safety, the electrical parameters associated with their ties are subject to change.

The humidity-sensitive properties of graphene-bonded materials have recently been discovered and improvements in strength, ability, and water adsorption have been observed. In detecting humidity, oxygen-containing molecules on the surface are major in monitoring an excellent function. A great number of these can cause the material to act as an electrical insulator, as in the case of GO. As for resistive transducers, a high resistance is undesirable for the sensors.

GQDs provide not only properties but also a bandgap of graphene that can be modified [140]. Edge effects and quantum confinement increases from the value of the bandgap of the resulting GQDs in the UV range by minimizing the size to several nanometers. GQDs have already been an ideal substrate for applications such as photovoltaic, LED, and deep UV photodetectors because of the induced adjustable bandgap, high stability, and high optical absorption. When exposed to room temperature and under photon illumination, the conductivity of GQDs suddenly decreases because on their surface the oxygen and water molecules can be adsorbed. This phenomenon is defined as negative photoconductivity (NPC) and is primarily observed in the simultaneous carrier detection processes caused by surface adsorbents and the removal of electronic photographic traps. GQDs are therefore an exceptional substance that is resilient to distinct ambient humidity [141].

The single electron transistor (SET) was originally used primarily for GQDs. Furthermore, electronic pressure and humidity sensors have been developed with detection control in SET and GQDs [142]. Recently, it was developed highly versatile and humidity sensors with high sensitivity based on GQDs with good response (390 at 99% RH), selectivity, wide detection range (1–100% relative humidity), short response, and recovery times (12 and 43 s, respectively), and versatility [143].

A GQDs-based humidity sensor was also designed by citric acid carbonation and found that GQDs have an exceptional ability to detect the ratio between the quantity of water vapor pressure in the air at a selected temperature and the same amount needed to be saturated at the same temperature at lower relative humidity levels for different humidity levels of RH [144].

In a further analysis, PEDOT: PSS and CNT were assembled with GQDs to obtain a pure composite working as humidity sensors. The composite sensor is said to respond well to humidity in the range of 60% to 80% at room temperature and atmospheric pressure. Response and recovery times were roughly 30 s or 50 s, with minor improvements compared to naked GQDs in response and recovery times [141].

A more appealing Fabry–Perot interferometer (FPI) was suggested by Yong et al., considering the GQDs-PVA compound used as a material for assessing sensitivity to relative humidity. Water absorption alters the assessment of relative humidity. The interaction of PVA with GQDs and GQDs-PVA with water molecules is shown in Figure 6 [145].

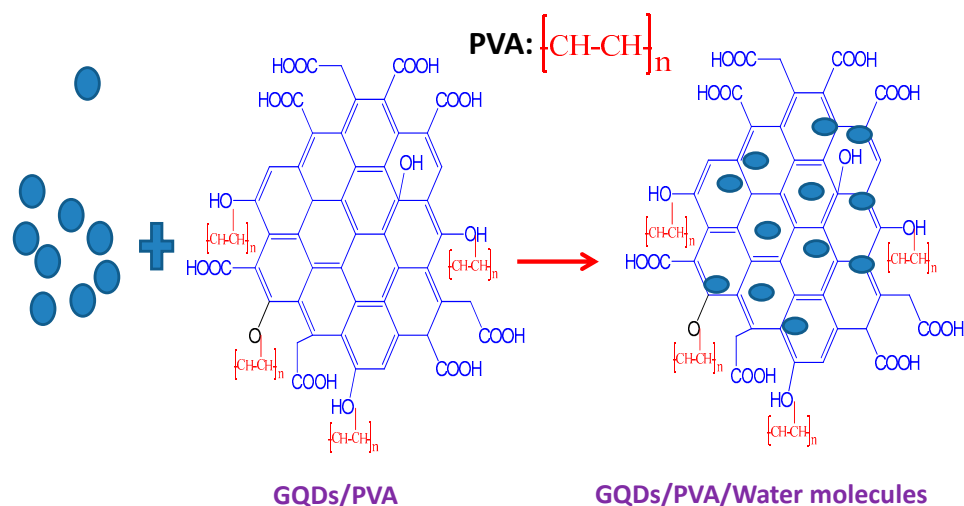


Figure 6. The method of mixing PVA with GQDs and water molecules with GQDs-PVA.

At 25 °C, the experiment was conducted. Information on the propagation of incoming light is explained in Figure 7 broadband (amplified spontaneous emission (ASE)) propagated by a sensitive RH sensor that analyses the obtained output light from an optical spectrum analyzer (OSA) with a spectral resolution of 0.02 nm. When the plastic jugs were filled with different classes of soaking salts, such as magnesium chloride, magnesium nitrate, lithium chloride, and potassium chloride, which were separately immersed, the characteristic state of relative humidity occurred. A hygrometer was used to record the relative humidity to make unambiguous estimates of relative humidity and the hygrometer was attached to the ROTRONIC brand. The sensor test used for the relative humidity and the hygrometer test were mounted in plastic containers in the same positions to ensure the precision of the calculation. The direct-current voltage source (DCVS) provides the hygrometer liveliness. The comparison of modern graphite-based relative humidity sensors reveals that the advantages of high sensitivity and a large number of measurements are combined with GQDs-PVA FPI. The relative humidity sensor of the GQDs-PVA fiber optic also has a higher link ratio than the graphite-based sensor. Fiber adhesive and several standard-based contrast sensors, meanwhile, show that GQDs-PVA-based FPI has many interesting points, such as ease of production and high sensitivity [145–147].

In addition, the high reversibility and repeatability of the functional applications provide higher research potential. In several applications, GQDs humidity measuring properties have been discovered and the uniqueness of the materials has been successfully exploited. GQDs are highly sensitive to the presence of air humidity and to increase relative air humidity with varying resistance. Even at very low relative humidity levels, such sensor devices can work with immediate response times and have a great activity for the production of low-power humidity sensors with ultra-compact features [145].

Summarizing the new humidity sensor based on the graphene quantum dots as mentioned above, the sensor was extremely sensitive to broad range changes in relative humidity. Other than its higher sensitivity, this sensor's ability to be active at lower relative humidity values can be cited as an advantage of this sensor over most of the humidity sensors as previous publication [148]. The sensor displayed a fast response time, and no significant sensor hysteresis was observed. As two different sensing ranges were seen for this sensor, two sensing mechanisms were thought to dominate the sensor's response to relative humidity [149].

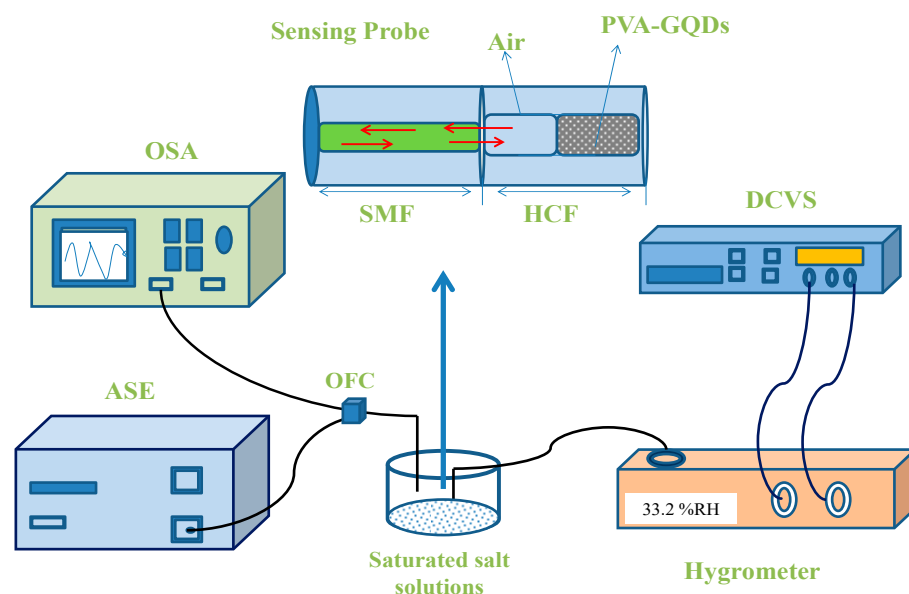


Figure 7. The graphical representation of the proposed Fabry–Perot interferometer (FPI) and the GQDs-PVA filling preparation process.

4.1.5. Gas sensor Type

Sensing is one of the environmental remedies used to track and catch the detection of gases from a rising society in particular. The prevalence of volatile organic compounds (VOCs) throughout these years, with the greenhouse gas emissions and climate change, poses risk [150]. GQDs have been developed as a sensor substance because they have more atoms on the edge than 2D materials because it contains more atoms on the surface and thus there is more adsorption [151,152]. Two separate GQDs (neutral and acidic) were synthesized and were used to manufacture NH_3 gas sensors called sensors A and B. After exposure to different concentrations of NH_3 gas, representative studies of sensors A and B were observed. When sensors are exposed to NH_3 gas, the current response is 14.9% and 5.9% respectively. Furthermore, the sensors have been stated to experience various electrical reactions with the same concentration of gas molecules, and the responses of sensors A and B to 10 ppm NH_3 over three cycles produced a resistance that can return to its original states after the GQDs sensor material has been stripped of NH_3 . This means that a high stability and high response towards A and B sensors at different concentrations of NH_3 . Moreover, the response and recovery times strongly vary with increasing the contents NH_3 [153]. Recently, drop-casting hydroxyl-functionalized graphene quantum dots (OH-GQDs) on a conductive nickel electrode has created an alternative room-temperature gas sensor. The OH-GQDs was built based on a bottom-up method using pyrene hydrothermal treatment; this study found that edge fictionalization and modification of GQDs is a promising approach for achieving high-efficiency gas sensors with high specificity for a specific target gas [154]. In another approach, for carbon dioxide gas detection, a room temperature graphene quantum dots (GQDs) based optical gas sensor. GQDs have been produced using a hydrothermal process then using a drop-casting technique for deposit a quartz substrate material [155].

4.2. Biomedical Applications

4.2.1. Bio Images

In both scientific and clinical applications, bioimaging plays an important role and facilitates the identification and study of biological processes from the subcellular level to processes in animals. Using suitable bioimaging sensors, the researchers were able to identify the early stages of the disease and determine behavioral patterns. GQDs have recently been described as a class of fluorescent nanomaterials with optoelectronic properties and can provide excellent data in diagnostic and diseased treatment in different

biological systems. Thanks to GQDs 0D structure with non-toxicity and bioimaging results in elevated solubility, biocompatibility, and inertness of chemicals. The most common materials today under physiological conditions are GQDs [156]. For instance, the previous study has performed an experiment in which up to 400 μg were added along with 150 mL of culture medium (104 cells) that did not attenuate cell activity as indicated by the MTT assay as shown in Figure 8. At excitation 405 nm, the GQDs were observed across the cell membrane using a confocal fluorescence microscope by tracking the light green region inside the cells. Therefore, bioimaging relies on GQD's arousal activity, resulting in different outcomes of PL-dependent performance. The resulting ratio consisted of excitation shifts from green to yellow at 488 nm [157].

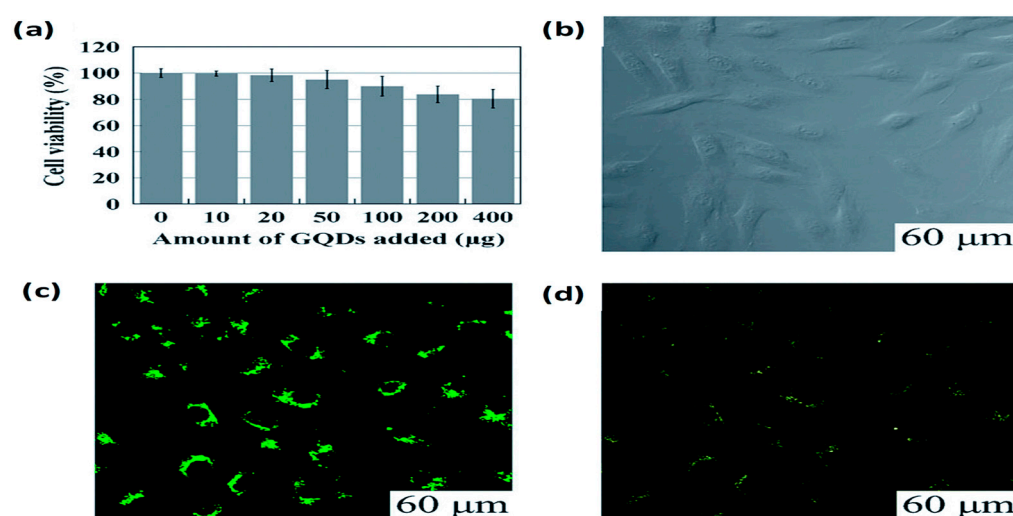


Figure 8. Cellular toxicity and cellular imaging of GQDs (a) Impact of GQDs on the viability of MG-63 cells (b–d) brilliant field washed cells, 405 nm, and 488 nm excitations, respectively. Adapted from ref [67] copyright 2019, RSC publications.

In a further study, the trifunctional-themed peptide structure is seen by non-covalent collaboration as a previous barrier to the creation of a new multifunctional protein nanofiber (PNF) and also paired with strongly fluorescent GQDs. Sufficient bioavailability and biocompatibility of PNF-GQDs nanohybrids were found in cell proliferation experiments. These PNF-GQDs nanohybrids demonstrated the ability to concentrate and visualize cancer cells simultaneously using a sensing component and an imaging test. Furthermore, PNF-GQDs nanohybrids have extraordinary potential, particularly when sensitive traceability and efficient labeling are evaluated [158,159]. GQDs-dependent operator is another photodynamic treatment (PDT), that can produce singlet oxygen through a multi-state refinement method and provide PDT experts with a quantum output of 1.2 with the highest description. The GQDs also exhibit a wide retention band with a heavy dark red discharge covering both UV and visible regions. In vitro and in vivo the researches have shown that GQDs can be used as PDT operators, thus offering highly trained treatment and imaging for cancer. This study also ushers in a new age of carbon nanomaterial PDT specialists who surpass conventional technicians in terms of quantum yield, water dispersibility, pH stability, and bioavailability [160].

In another example, hydrothermal treatment was used to prepare Cl-GQDs with HCl and fructose as a source of Cl and carbon, respectively. The oxygen and hydrogen groups inside fructose have been dehydrated, while the carbon forms the nucleus of GQDs, at the same time HCl has facilitated the reaction to the creation of a Cl dopant. These GQDs has considerable advantages to actively replace this fluorophore due to tunable and strong PL, photostability, excellent biocompatibility, and effective renal clearance, thus offering unprecedented opportunities for bioimaging [161].

4.2.2. Biosensors

Biosensors rely on the detection by the GQDs PL of the emitted photon in addition to bioimaging. The GQDs biosensor uses the association between the biomolecule analytes and the functional group of the GQDs to detect the presence of biomolecules. Ions are liable for chronic exposure and therefore should be transported and controlled efficiently at the cellular level. Consequently, it is very important to have in vitro biosensor ion sensitivity. On a comparable hand, the GQDs-based ethylenediamine-modified Ni²⁺ (E-GQDs) sensors showed a strong yellow PL emission that was substantially suppressed in the presence of Ni²⁺. The limit of detection for Ni²⁺ was 3×10^{-8} min with a quantum yield of 83% and its ability to be detected in vitro was shown by processing adipocyte stem cells in rats. The increase in hydrogen sulfide content (H₂S) contributes to diseases associated with cancer and Alzheimer's. A GQDs-based sensor of a functionalized (2,4-dinitrophenoxy) tyrosine (DNPTyr) is identified for HS detection with a detection limit of only 2×10^{-9} min [67,162].

In further analysis, to detect 2,4,6-trinitrophenol (TNP), nitrogen-doped GQDs (NGQDs) were modified with tris (hydroxymethyl) aminomethane. Owing to the overlapping of the NGQD emission spectrum and the TNP absorption spectrum, the sensor showed substantial photo-age in the presence of TNP. The use of nitrogen-doped GQDs as a catalyst has also been documented in a colorimetric biosensor for hydrogen peroxide and glucose. Moreover, for sensitive and selective detection of dopamine (DA), a label-free fluorescence-based technique was developed using graphene quantum dots (GQDs) as effective probes, this approach has been successfully applied to the determination of DA in biological samples with adequate recovery (98.8–106.4%) [163]. Moreover, GQDs can be used in other sensing applications such as the environment, agriculture, and food safety [164]. In addition, GQDs are ideal candidates for in vivo studies, because of their water solubility and biocompatibility [165]. Withstanding this, few other researchers have analyzed the capabilities of in vivo sensing of GQDs.

4.2.3. Drug Delivery Formulating Systems

GQDs are a very promising substrate for drug delivery systems because of their excellent tunable chemical and physical properties, as well as the simple surface functionalization. Otherwise, there are dozens of published articles used as antimicrobial and anticancer treatments [20,39,166–169]. The size-dependent coupling of graphene nanosheets with DNA molecules was reported that GQDs have a limited lateral dimension and the capacity of DNA molecules to intercalate is most apparent. Moreover, Doxorubicin (DOX) is a frequently used therapeutic agent for human cancers such as sarcoma of the soft tissue, aggressive non-Hodgkin's lymphoma, and breast cancer [170]. The targeted drug binds to the GQDs functional groups and the drug is loaded onto the surface through the π - π interaction for precise DOX delivery into tumor cells. In the medication of breast cancer cell lines (MCF-7), the DOX/GQDs conjugate mechanism has been demonstrated as successful drug delivery [171]. From a recent study, folic acid as a ligand in GQDs has been reported to release DOX against cancer cells, GQD's intrinsic fluorescence enables real-time tracking, targeted distribution of drugs, and selective labeling of cells [172]. A fluorescent, traceable, and pH-sensitive GQDs-based DOX drug delivery system was documented by Qiu et al., using the arginine-glycine asparagine ligand. The targeted release of biocompatible and localized GQDs-based DOX using biotin has recently been documented as a ligand molecule. Similarly, it was reported that GQDs are anchored with hyaluronic acid (HA) as a target agent [173]. To research the delivery of DOX-targeted drugs using GQDs, a revolutionary real-time monitoring system was developed using Forster resonant energy transfer (FRET) [174].

Hyaluronic acid is a natural polysaccharide and a major component of the extracellular matrix and synovial fluids of the body. It can be used with human serum albumin (HSA) nanoparticles functionalized with GQDs were produced (HA/HSA/GQDs) and used for the targeted delivery of gemcitabine to pancreatic cancer which achieved a delayed-

release and increases its efficacy compared to freely use [175]. Further, HA was used with a bifunctional system based on mesoporous silica nanoparticles coated N-GQDs as a targeting unit, for the selective release of DOX to cancer cells. Furthermore; HA with the carboxymethyl functionalized inulin was conjugated to Ag-GQDs and as a targeting moiety [176]. In another example, supramolecular β -cyclodextrin (β -CD) encapsulated with GQDs by the hydrothermal method to obtain an effective drug molecules delivery system for Age-related macular degeneration [177].

4.3. Energy Storage

4.3.1. Supercapacitor

Two key factors that require specialized high-performance technology and energy storage are huge energy consumption due to advancements in technology [178]. The transformation and accumulation of electrochemical energy systems is therefore an excellent prospect, and therefore a new subject of study between the manufacturing sector and academia. In order to store energy that is converted from chemical energy into electrical power, electrochemical energy storage systems (EESS) are widely used. EESS is an important prerequisite for any energy storage unit, which has attracted a lot of interest because of its high charge/discharge rates and long life. The supercapacitor is the key device of the EESS that, in the current situation, revolves around great caution and major sources of energy. It gives fast charge/discharge, cyclic stability with long-term and high-power density, also known as the electrochemical capacitor. These properties allow EESS to be one of the superior performance materials for use in electric vehicles, portable electronics, and emergency power systems [179].

Depending on the structure and composition of the probe materials, the electrochemical characteristics of the supercapacitor, as well as the power and stability of the cycle limited resources of transition metal oxide. Conjugated polymers and carbon structures have so far been confirmed as energy storage carriers [180]. Numerous studies have been performed in recent years on the properties of the EESS GQDs and their future use as electrode materials. GQDs would be an encouraging backdrop for the mass-scale production in the area of advanced energy storage due to (i) the 0D conjugated carbon structure is highly elastic for complex and conductive architectures, (ii) a large number of energy storage active sites can be obtained through the improved edge structure and functional groups, and (iii) strong chemical reactivity and migration makes for fast assembly or process. Recently, some researchers have found that GQDs includes the necessary properties of EESS that can significantly improve the performance of a supercapacitor [181].

From a recent study, integrated GQDs impact was investigated on supercapacitor via a one-step hydrothermal process in 3D graphene (3DG). The SEM 3DG and GQDs/3DG pictures of the bare joints are shown in Figure 9. The present pores in pure 3DG size ranging from submicron to microns as shown in Figure 9 which indicates that there are bigger pores in the GQDs/3DG-40 composite. Due to the lower GQDs levels, (GQDs/3DG-80) demonstrates a more compact structure than pure 3DG the GQDs/3DG-40 had a greater surface area compared to the GQDs/3DG-80. Moreover, a comparison of the electrochemical properties of pure 3DG and GQDs/3DG Just in the cyclic voltammetry curves indicate that the redox peaks were dark at different scan speeds are almost rectangular CV curves as shown in Figure 10a. Figure 10b shows the charge/discharge curve at a current density of approximately 1 A g^{-1} , due to the specific surface of GQDs, the composite material had the optimum specific capacity and better electrical conductivity than pure 3DG. Figure 10c indicates that the specific capacitance decreases with increasing current density. In Figure 10d the cyclic stability showing 93% capacity retention after 10,000 revolutions due to high-quality GQDs [182].

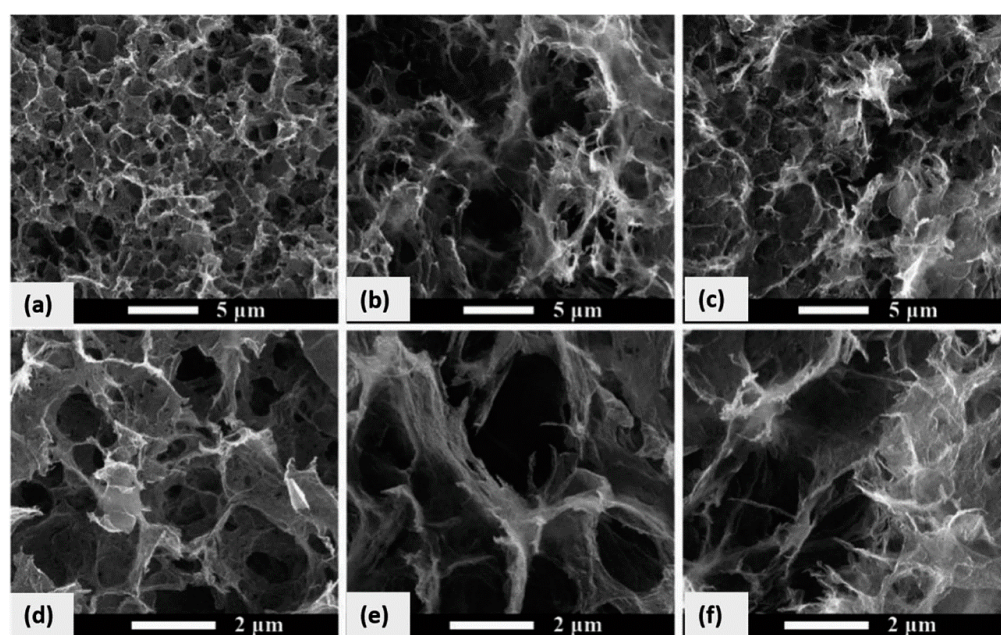


Figure 9. (a–f) GQDs/3DG SEM pictures with varying composition. Adapted from ref [182] copyright 2019, MDPI publications.

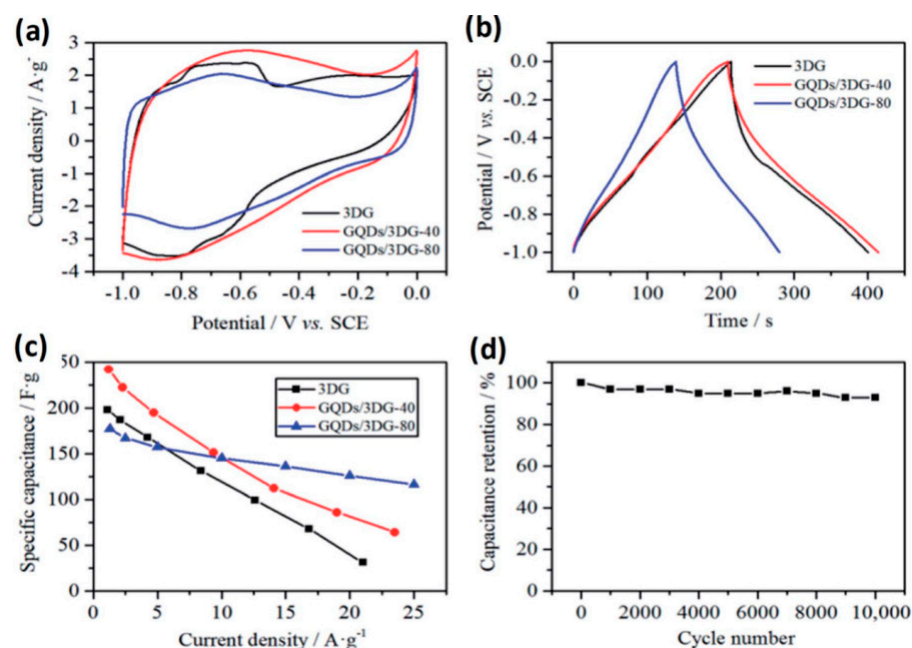


Figure 10. (a) Cyclic voltammetry curve at a scanning rate of 10 mV s⁻¹ (b) charging/discharge curves at various current densities with various proportions; (c) specific capacitance versus current density; and (d) cyclist stability at the current density of 24 A g⁻¹ for composite material. Adapted from ref [182], copyright 2019, MDPI publications.

In addition to metals, conductive polymers (CPs) play an important role in supercapacitors due to their specific characteristics, such as flexibility, high electrical attraction, and redox properties. GQD's and CP's singular benefits will enhance supercapacitor operation. Recently, GQDs and their electrochemical properties were doped with stable conductive polyaniline (PANI). Changes in GQDs @ PANI's electrochemical characteristics strongly depended on the quantity of GQDs in the PANI. For applications such as supercapacitor electrodes, the maximum specific capacity of 3632.0 F g⁻¹ was given [183].

Supercapacitor electrode includes PVA-GQDs/poly (3, 4-ethylenedioxythiophene) (PEDOT) nanocomposite was further investigated. The PVA-GQDs/PEDOT nanocomposite electrode displays a higher current potential reaction due to its large GQDs surface area. This leads to improved load build-up and load storage. Moreover, from the CV curves of the PVA-GQDs/PEDOT nanocomposite at different scanning speeds, it can be concluded that the fast spread of the electrolyte is caused by increased scanning speed on the surface of the active electrode. As the scan rate increases, the actual capacitance (C) decreases as the ion is not sufficient to move to the active electrode at a high scan rate [184].

4.3.2. Lithium-Ion Batteries

Increasing worries about the decline of fossil fuels and environmental concerns have alerted governments to the need for energy that is crucial to developing clean energy. So, finding renewable sources of energy such as tides, and human society's wind and solar energy are appreciated. Therefore, several researchers have turned their attention to the development of reliable energy storage (EES) devices. For growth, reliable ESSs such as batteries and supercapacitors are essential elements. These systems of energy Lithium-ion (Li-ion) batteries are probably the prevalent substitute, in addition to conventional lead-acid, Ni-Cd, Ni-MH, and supercapacitors. Because of their lightweight, high energy, and good efficiency, they can minimize the current demand and encourage the use of major energy sources. In recent years, many improved lithium-ion batteries have appeared, such as sodium–aluminum, lithium–air–sulfur, and metal-ion batteries. In addition, great efforts have been paid to improve their overall performance for future continuous practical applications [185].

A lithium-ion battery usually contains a cathode, an anode, an electrolyte, an outer jacket, and parts for sealing. A variety of types of lithium-containing cathode materials, such as lithium–manganese, lithium–cobalt oxide, lithium-ion phosphate, and conductive polymers, etc., have been researched to date. Carbon graphite, lithium metal is widely used to manufacture the anode and widely available on the industrial market in different forms. Because of their environmental friendliness and higher energy density, lithium-ion batteries are commonly used to drive a wide variety of portable electronic devices. While certain hazardous organic electrolytes and heavy metals, such as cobalt and flammable organic solvents, are still used in lithium-ion batteries, they can cause significant environmental pollution. Nowadays, rechargeable lithium-ion batteries are commonly used and can play a very important role in power supply devices that, in today's situation, are portable electronic systems. Many researchers, however, face the challenge of achieving high power density and energy density that is not adequate to meet rising energy-consuming applications, such as electric vehicles and energy storage at the grid level. Many researchers are trying to find an enhanced battery device that is superior to current technology to solve these issues. Graphene-based materials have recently been used, due to their special properties, made other electrode materials alternative to EESS, e.g., H. batteries with lithium, supercapacitors, etc. They have decreased dramatically and have gained a lot of interest [186,187].

A finite bandgap in the material is caused by quantum containment in GQDs, which internally affects the electronic conductivity. Quantum confinement has been reported to affect the diffusion coefficient of lithium and to affect a battery's electrochemical efficiency and long-term electrochemical cycle. Surprisingly, the functional oxygen groups on their surface boost GQDs, such that special properties as excitation luminescence and a non-zero bandgap are well understood. Due to their small scale, GQDs are also required to be able to permanently protect the target material. Indeed, GQDs have been reported to serve as a coating or composite for energy storage [188].

The implementation of GQDs speeds up the transfer to the electrode of vast quantities of electrons and electrolytes, thereby enhancing the electrochemical efficiency of lithium-ion batteries. The major benefit of using GQDs as an electrode coating material is the broad ion transport surface area ability between the electrolyte and the active material,

allowing ultra-fast storage and release of energy. The high-speed performance and cyclic stability of lithium-ion batteries are excellent because of the above characteristics of the composite coated GQDs with another metal as an electrode. For example, a GQDs-coated VO₂ material was prepared as an electrode in a lithium-ion battery and generated highly electrochemical characteristics. A sensitizer and surface defense may serve as the GQDs layer. The electrode, therefore, has a power of more than 420 mA h g^{−1} and a retention rate of 94 percent at 18 A g^{−1} after 1500 cycles [189].

4.4. Corrosion Protection

4.4.1. Corrosion Protection by Functionalized GQDs

Since metal corrosion has detrimental effects on the economies of nations, opposing arguments have been recognized to boost the corrosion resistance of metallic components. Polymer coatings and epoxy resin are represented as the most common material used to distinguish metallic surfaces from corrosive conditions as protective coatings [190,191]. While epoxy coatings are regarded as thinking leads to the provision of an effective, convenient and economical process, but it suffers from certain drawbacks, such as corrosive agent permeability, hydrolytic degradation, and can therefore not provide long-term protection against corrosion [192]. Throughout this regard, nanoparticle coatings have shown considerable interest in boosting the effectiveness of epoxy coatings for corrosion protection [193]. Graphene-based materials including graphene nanosheets (GNS) [194], graphene oxide (GO) [195], chemically functionalized graphene oxide (f-GO) [196], be excellent candidates in preceding research to enhance the mechanical and corrosion resistance properties of polymer composites. In addition, the strengthened properties of nanocomposite coatings based on polymer/graphene can be related to 2D geometry properties, high specific surface area, corrosive agent impermeability, and improved interfacial graphene-polymer interaction due to rumpled surface morphology [197]. However, to the best of our knowledge, the potential use of GQDs as nanofiller to improve the corrosion resistance properties of polymer composites has been explored. This is because of the key characteristics of GQDs for formulations in corrosion-resistant coatings such as low toxicity, simplicity to use, and costless methods that can be compatible with other graphene-based materials. GQDs as aforementioned formed from one-layer or few-layer graphene nanometer-scale debris consisting of sp² carbon atoms edged with heteroatom functional groups [198]. GQDs have now gained more focusing as a new category of 0D graphene-based materials with sizes below 20 nm due to its exceptional mentioned physicochemical properties [199].

This part briefly focusing on the corrosion resistance reliability of GQDs, The GQDs prepared are water-soluble, and their uniform dispersion in solvent-based epoxy coatings and homogeneous dispersion of GQDs in the polymers is a key obstacle. Nanocomposite coatings with improved corrosion resistance properties, similar to other graphene-based nanofillers, must therefore be prepared [200]. To boost the interfacial bonding of GQDs with polymer matrices, some modifications have also been made to the surface of GQDs with silane coupling agents. As a bridge between GQDs and the polymer matrix, the silane binding agent improves the dispersion performance of GQDs in coatings [201].

Recently, Potentiodynamic polarization tests were used to evaluate the corrosion-resistant activity of pure substrate coated by bare epoxy, and epoxy/f-GQDs composite coatings in 3.5 wt. % NaCl solutions. Figure 11 demonstrates the results of Tafel plots which indicate that composite coatings of epoxy/f-GQDs have excellent corrosion resistance compared with the control epoxy owing to their substantially lower corrosion rate. In the meantime, the epoxy coating contains higher incorporation of f-GQDs (0.5 wt. %) demonstrated better safety and lower corrosion rates than epoxy coatings filled with a low concentration (0.1 wt. % f-GQDs) [200].

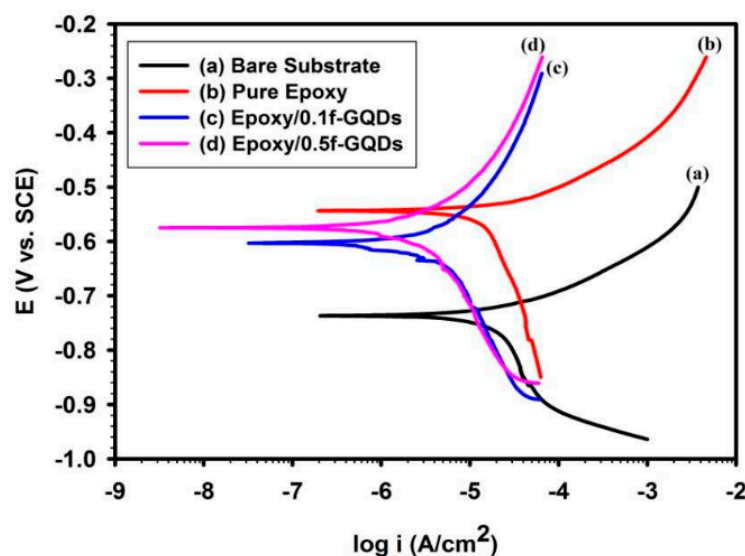


Figure 11. Tafel plots for bare substrate, pure epoxy coating, epoxy/0.1f-GQDs, and epoxy/0.5f-GQDs coatings in 3.5 wt. % NaCl solution, Adapted from ref [200], copyright 2019, Elsevier.

4.4.2. N-doping GQDs for Raising Corrosion Prevention

Magnesium alloys are perceived as renewable resources with major advancements in aerospace, electronics, and aviation manufacturing [180]. Due to their specific hardness, low density, and outstanding standard of electromagnetic shielding, magnesium alloys have low corrosion inhibition which seriously limits the life of service. Therefore, many strategies for improving magnesium alloy corrosion resistance, such as ion implantation and surface coating, have been suggested in the past few decades [202]. Surface coating is a great way to boost the corrosion protection of all these techniques by isolating the corrosion medium from the magnesium alloy, including graphene coating, double-layered hydroxide coating, polymer coating, silane coating, and composite coating [203,204]. A polymethyltrimethoxysilane (PMTMS) coating was recently formed on the magnesium AZ31 alloy by incorporating pre-treatment with micro-arc oxidation (MAO). As the PMTMS is more durable and acts as a physical barrier to prevent the corrosive solution from attacking, the composite coating of MAO/PMTMS shows greater corrosion resistance than the coating of MAO. However, the PMTMS coating is susceptible to swelling and peeling off [205].

On the other hand, graphene oxide (GO) is frequently investigated in the field of metal corrosion prevention along with its unique properties. As a physical barrier, the relatively low-density of GO coating increases corrosion resistance, while the isolated GO sheets serve as cathode locations, resulting in galvanic corrosion actions [206]. Therefore, GO nanosheets are usually hybridized with other polymers or inorganic compounds to improve corrosion resistance. Lately, the incorporation of GO into the plasma electrolytic oxidation (PEO) coating will effectively lower the number of micro-pores within the coating, and the GO composite coating could block the refracted corrosive electrolyte into the Mg surface.

However, as a result of the interface's poor bonding, the interface between the coating and the substrate has many pores and pore bands, resulting in a small increase in corrosion resistance. In addition, the graphene coating exhibits poor adhesion to the metal interface due to the direct physical contact of GO nanosheets with metal surfaces and falls off easily. Subsequently, surface-pretreatment stages are carried out to increase the adhesion force between the coating and the substrate. Long-term reliability is often unpredictable, along with inadequate and uncontrollable coverage [207].

Compared to GO nanosheets, GQDs exhibit many attractive properties resulting from their distinct structural characteristics. Firstly, GQDs are extremely stable in water dispersion and have strong film-forming properties, avoiding the insoluble GO problem in the coating [64]. Secondly, due to the tiny sizes of many nanometers, GQDs have a big number of edge structures. These edge groups, including the hydroxyl, carbonyl, and

epoxy groups, are capable of bonding with other protective coatings or substrates, and functionalizing are susceptible also to other additives [34]. More precisely, the third point is that element-doping will fine-tune the chemical structures of GQDs, such as N-doping, S-doping, and N, S-doping, according to performance requirements.

In a recent study, developing N-doped GQDs (N-GQDs)/PMTMS composite coating on the surface of the AZ91D Mg alloy. The N-GQDs/PMTMS coating preparation method is shown in Figure 12, in which the N-GQDs is electrodeposited on the surface of the Mg alloy by a methyltrimethoxysilane (MTMS) silylation reaction, and then PMTMS is coated on the surface of the N-GQDs. As an intermediate layer, the N-GQDs chemically bind the Mg substrate and PMTMS coating via the functional groups of N-GQDs.

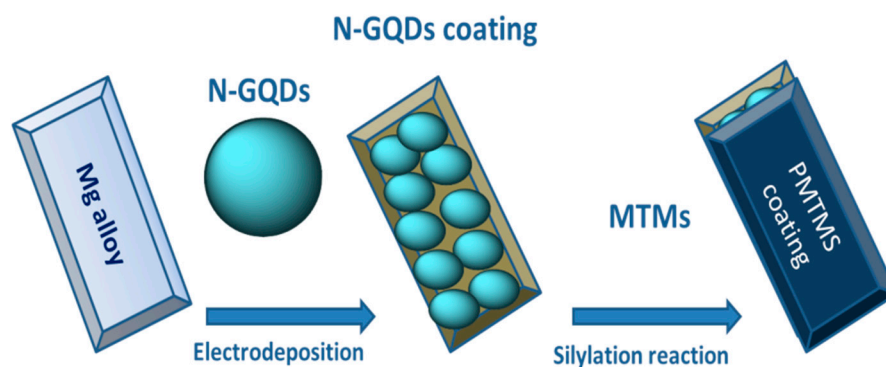


Figure 12. Scheme of the N-GQDs/PMTMS composite coating synthesis method.

Figure 13 shows the OCP curves of the bare Mg alloy, PMTMS coating, N-GQDs coating, and N-GQDs/PMTMS composite coating in 3.5 wt. % NaCl solution. The OCP of the samples are ranked in decreasing order as follows: N-GQDs/PMTMS composite coating (−1.38 V) > N-GQDs coating (−1.53 V) > PMTMS coating (−1.58 V) > Mg substrate (−1.59 V). This result implies that the N-GQDs coating and the N-GQDs/PMTMS composite coating can improve the corrosion resistance of Mg alloy [208].

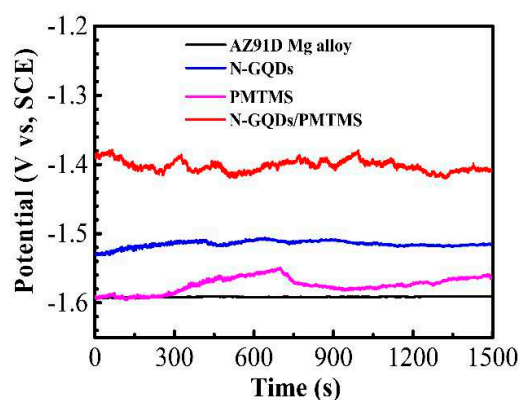


Figure 13. OCP curves of the bare Mg alloy, N-GQDs coating, PMTMS coating, and N-GQDs/PMTMS composite coating in 3.5 wt. % NaCl. Adapted from ref [208], copyright 2020, Elsevier.

4.5. GQDs in Fuel Cell

Fuel cell technology continues to occupy a place in the world of research, academic, industry, and community. Fuel cell technology has numerous applications in stationary, transport, medical, tissue engineering, and more. The rich edge of GQD is especially beneficial for electrocatalysts since the electrochemical interactions can be more easily happen on the edge planes than on the basal plane. It was successfully synthesized heteroatom-doped GQDs for fuel cell application through the hydrothermal method. The ability of GQDs as a unique material has been identified to enhance fuel cell activity as an ORR in catalytic and proton conductivity [209].

5. Challenges and Potential Opportunities

As the latest generation of carbon materials, GQDs were proposed because their exceptional properties make them ideal for many technology fields. However, for your promising future, there are some constraints to focus on. Research is underway and complete awareness of PL GQD's properties is required. Several potential mechanisms have been envisaged, such as the effect of size, alteration of the surface, and doping with other elements. The methods derived from the optical properties of manufactured GQDs can vary, so it is very important to understand basic PL operations in GQDs. GQDs have several barriers in the current scenario, including the lack of an adequate synthesis technique that results in half of the evaluation of the optical property, the lack of obtaining the apparent size required, and the morphology of the GQDs. Without loss of optical properties, PL wavelength emission problems, and PL function inconsistency in different applications, Moreover, the quantum yield of most GQDs still does not surpass 55 percent, considering the performance of different-colored PL property GQDs with PL in the NIR state. Therefore, because of its low quantum characteristics, improving GQDs is difficult due to their limited use in many areas. Robust, cost-effective, sophisticated, and robust sensors that function in all environmental circumstances are the potential benefits of GQDs. The focus should be put on manufacturing methods currently not available for GQDs in the step-by-step GQDs synthesis to enable broader applications in a variety of sensors, supercapacitors, solar cells, biomedical devices, and corrosion prevention. In addition, GQDs studies are needed to be related to environmental, bioanalysis, and energy applications. Lithium-ion batteries are only in their infancy, as GQDs are used as electrode materials. GQDs-hybrid will meet many specifications for future polymer matrix power supplies as electrode material due to high density, electricity strength, higher cyclic stability of electrodes, environmental friendliness, and high safety profile. Bare and functionalized GQDs in the various above-mentioned applications and research can affect their efficiency. This element of the challenges helps scientists working in the field of materials science to obtain a deeper understanding and firmly believes that in potential applications, GQDs will boost higher standards. There is continuing research into the physical and healing effects of GQDs with natural alternatives. It may certainly be an eye-opening material for the delivery of drugs or genes, biochemical, optical sensors, and theranostic applications.

6. Conclusions

GQDs-based materials have various technological applications in different fields such as biosensors, bioimaging, energy storage, and corrosion inhibitors. GQDs have been widely discussed based on a characteristics point of view. Top-down and bottom-up approaches were used to synthesize GQDs. Optical properties are interesting criteria for tailoring GQDs to the desired application. In particular, GQDs have an extensive research interest in sensors, owing to the edge effect and quantum confinement effect. Nonetheless, the non-toxicity, costless, ease of synthesis, and high chemical stability are advantages of GQDs rather than published nanocomposite. Significantly, PL, size, shape, electrochemical behavior, humidity sensors, supercapacitors, and other related properties/applications have been discussed in this review. A short note of GQDs application in corrosion inhibitor is also highlighted. The size and morphology of GQDs are briefly investigated since this phenomenon is still in the initial and prime platform. This review might be useful to the readers who are seeking in-depth knowledge of the synthesis, as well as the fabrication of facile, efficient, and economic GQDs together with their versatile applications in biomedical, energy storage, and corrosion resistance.

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