



CQDs embed g-C₃N₄ photocatalyst in dye removal and hydrogen evolution: An insight review

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ABSTRACT

Recent research has highlighted heterogeneous photocatalysts as a feasible contender for addressing energy shortages and environmental cleanup. One of the best semiconducting photocatalysts used in wastewater treatment, disinfection, and energy evolution is graphitic carbon nitrate (g-C₃N₄). Researchers have used carbon quantum dots (CQDs) to maximize and enhance the photocatalytic activity of g-C₃N₄ and to get over the material's limits due to photoinduced charges, partial surface area, and insufficient light-capturing difficulties. In this context, the fundamentals of CQDs and g-C₃N₄ are described in length, along with their structural state, synthesis, and modification techniques. The classification, manufacturing procedure, and characterization of CQDs/g-C₃N₄ are then highlighted in this paper. Following that, it is shown how CQDs/g-C₃N₄ photocatalysts are used in dye removal and hydrogen evolution studies. The discussion of CQDs/g-C₃N₄'s present hurdles, unmet needs, and future research prospects concludes while keeping in mind their practical applications. This study shows that by embedding CQDs, the influence of charges, morphological modification, and textural quality of g-C₃N₄ have been altered. It is anticipated that this review will offer a practical overview and comprehension of CQDs embedded with g-C₃N₄ photocatalysts in order to promote their utilization. The ultimate goal of this review may be to impart a fundamental understanding of photocatalysis while also providing an expository evaluation of the most recent advancements in g-C₃N₄/CQDs photocatalysts in the sectors of energy and environmental security.

1. Introduction

Currently, environmental remediation and energy security have emerged as serious problems that are harmful to both people and the environment. Physical, chemical, biological, advanced and hybrid technologies are successfully applied to overcome energy and environmental issues [1,2]. Numerous advanced oxidation processes (AOPs) are used in wastewater treatment, including fenton oxidation, ultrasonic oxidation, moist air oxidation, photochemical oxidation, ozonation, and electrochemical oxidation [2,3]. However, there are many underlying factors that limits the application and performance of the methods such as pH dependent, costly, slow process, secondary sludge and well

catalyst requires [3–5]. Consequently, researchers introduced AOPs based photocatalysis method to combat and purify the wastewater and simultaneously convert solar energy into green hydrogen [6]. Several photocatalysts such as TiO₂, ZnO, Mn₂O₃, ZnS, SnO₂, Fe₂O₃, WO₃ are commonly used in photocatalysis system that are widely functional in hydrogen (H₂) production, pollutants photodegradation, CO₂ reduction, and other related fields [7–15]. Therefore, conventional photocatalyst has some drawbacks such as low photo quantum efficiencies, high electron recombination rate, low efficiencies and poor stabilities that need to be solved [6,16]. A well functional photocatalyst should have an appropriate bandgap that satisfies efficient light absorption as well as an excellent conduction band (CB) and valance band (VB) and high

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photogenerated electron and hole separation/transfer efficiency.

Since, $g\text{-C}_3\text{N}_4$ is discovered as promising nontoxic and metal free semiconductor photocatalyst that is rapidly applied in dye wastewater purification as well as hydrogen evolution [6,17]. In comparison to other semiconductor, $g\text{-C}_3\text{N}_4$ offers strong fluorescent properties such as photostability, wide excitation spectra and tuneable photo-luminescent emission spectra [18–23]. The outstanding photocatalytic performance of $g\text{-C}_3\text{N}_4$ in water splitting mechanism under visible light irradiation was initially reported by Wang et al. [24] in 2009. Momentous interest has been shown in this prospective metal-free $g\text{-C}_3\text{N}_4$ photocatalyst because of easy synthesis, high levels of physical and chemical stability, visible light induced band gap and plentiful in nature [17,18,25,26]. More importantly, $g\text{-C}_3\text{N}_4$ can be easily fabricated by thermal polymerization of abundant nitrogen-rich precursors such as urea, melamine, thiourea, cyanamide, dicyandiamide, and ammonium thiocyanate [27–31]. High levels of polymerization and the tri-s-triazine ring structure of $g\text{-C}_3\text{N}_4$ possesses the excellent heat and chemical resistance nevertheless suitable bandgap and CB, 2.7 eV and 1.07 eV, respectively [32]. But unfortunately, photogenerated charge carriers recombine at a rapid rate due to weak van der Waals interactions between adjacent carbon nitrate layers, high exciton binding energy, small specific area, low quantum efficiency, high electron hole and unstable in solar light [33–35]. Several modified strategies of have been developed to enhance the photocatalytic performance of $g\text{-C}_3\text{N}_4$ such as morphology control, modification with co-catalyst, coupling with other photocatalyst and doping with metal and non-metal elements, modification by carbonaceous nanomaterials [6,36]. For example, $g\text{-C}_3\text{N}_4$ is coupled with SnO_2 [9], ZrO_2 [28], CoFe_2O_4 [20], ZnO [12], CdS [37], BiOBr [38] and $\text{W}_{18}\text{O}_{49}$ nanowires [39] but the ultimate result does not meet the expectation as achieved by coupled with CQDs [40–42]. Thus, $g\text{-C}_3\text{N}_4$ exposed better performance and photocatalytic activity by increasing the electron transfer and reservoir properties by coupling with CQDs [6,30,43–45]. CQDs are a fascinating class of zero-dimensional carbon nanomaterials with an average size <10 nm with exceptional physicochemical, electrical, and optical properties [46]. In addition, CQDs exhibit the anticipated advantages of high surface area, aqueous stability, low toxicity, and highly tuneable photoluminescence behaviour [47]. The photocatalytic activity and visible light absorption rate of CQDs has been enhanced and improved by coupling with different semiconductor photocatalysts [47–49]. Therefore, CQDs are auspicious nanomaterials in the field of photocatalysis, wastewater treatment and energy evolution [13]. As a result, CQDs have been used as an effective part of photocatalyst design.

In this regard, a modification strategy of $g\text{-C}_3\text{N}_4$ semiconductors using CQDs was implemented to improve their adsorption, electron-hole pair migration and separation efficiency, ultra-high specific surface area

and photocatalytic activity. A growing body of research has attempted to combine $g\text{-C}_3\text{N}_4$ with CQDs to develop a new photocatalyst in order to enhance their photocatalytic activity, taking into account the outstanding and distinctive features. For example, Methyl Blue (MB) and Rhodamine B (RhB) dyes were completely degraded by using CQDs modified $g\text{-C}_3\text{N}_4$ photocatalyst, simultaneously hydrogen evolution rate was outstanding [6]. Although, CQDs and $g\text{-C}_3\text{N}_4$ both are non-metal, non-toxic and their internal electrical mechanism in highly potential for the separation of photoexcited electron and hole pairs. In addition, this composite photocatalyst can produce highly active species, oxidizing free radicals thus converting them into water and treated product. However, $g\text{-C}_3\text{N}_4$ not only modifying and coupling by CQDs but also by graphene quantum dots (GQDs), N-CQDs, S-CQDs, Pt-CQDs and applied in different sectors as designated in Fig. 1.

Research on photocatalytic dye wastewater purification and hydrogen evolution through CQDs modified $g\text{-C}_3\text{N}_4$ photocatalyst have been increasing over the years. However, some of the published review articles only addressed photocatalytic activity, some of them hydrogen evolution and some of them are focusing pollutant removal separately [13,14,48]. Therefore, a comprehensive summary of CQDs modified $g\text{-C}_3\text{N}_4$ based photocatalyst for dye wastewater treatment simultaneously hydrogen evolution is required that offers an updated progress. The aim of this review is to condense the recent progress of CQDs modified $g\text{-C}_3\text{N}_4$ photocatalyst in associated with fundamentals, synthesis process, structure, properties with the application and performance in dye removal and hydrogen evolution. In Section 2, recent studies are summarized by assessing the fundamentals, synthesize, structure, application and mechanism of $g\text{-C}_3\text{N}_4$. Subsequently, the fundamentals, properties, fabrication and application of CQDs is discussed in Section 3. Classification, synthesize process and characterization of $g\text{-C}_3\text{N}_4$ /CQDs photocatalysts is addressed in Section 4, followed by the mechanism of dye wastewater purification synchronously hydrogen evolution is herein discussed. The performance of $g\text{-C}_3\text{N}_4$ /CQDs photocatalysts in photocatalytic dye wastewater treatment and hydrogen evolution is displays in a tabular form. After carefully analysis of the previous published research articles, there hasn't been a comprehensive review which is addressed both dye wastewater treatment and hydrogen evolution together. In Section 5, the photocatalytic performance of $g\text{-C}_3\text{N}_4$ /CQDs is described by considering social, economic and environmental sustainability through SWOT analysis. Recent observations, growth, prospects, challenges and future perspectives of CQDs modified $g\text{-C}_3\text{N}_4$ photocatalyst are discussed in Section 6.

Finally, future research advances and expectations for $g\text{-C}_3\text{N}_4$ /CQDs heterogeneous photocatalysts are presented in a constructive way. This study will be providing a valuable overview and insight for the promotion of applications of CQDs modified $g\text{-C}_3\text{N}_4$ based-photocatalysts.

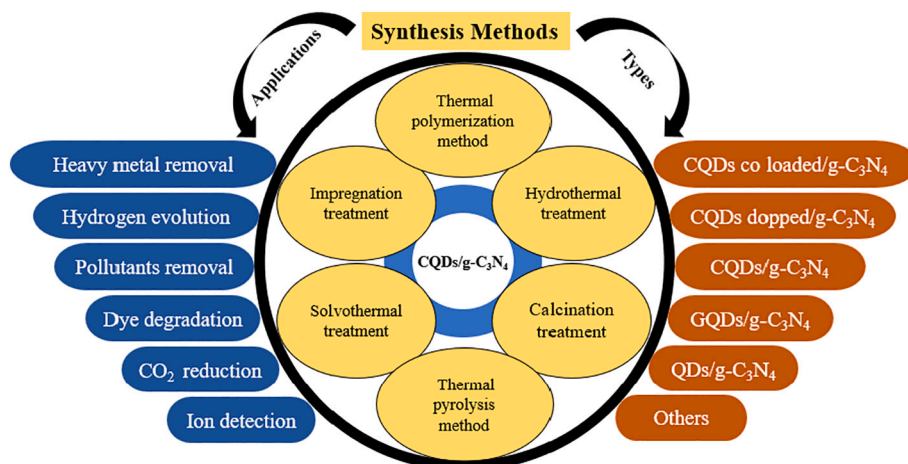


Fig. 1. Compendium illustration of the types, synthesis methods, and applications of CQDs/ $g\text{-C}_3\text{N}_4$ photocatalysts.

In addition, offers direction and guideline for young researchers to improve the photocatalytic performance of CQDs modify g-C₃N₄, not only in dye wastewater treatment and hydrogen evolution but also medical, bio sensing, biotechnology sectors and so on. This research is the evidence of CQDs embedded g-C₃N₄ photocatalyst fabrication and its importance in the field of energy and environmental sustainability. This research might support the development of highly effective g-C₃N₄/CQDs photocatalyst and their commercial use.

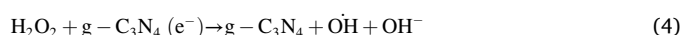
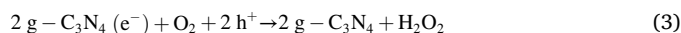
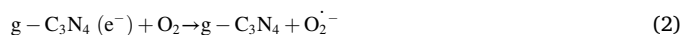
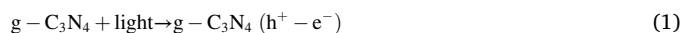
2. Fundamentals of g-C₃N₄ photocatalyst

g-C₃N₄ is a promising semiconductor due to its excellent characteristic such as metal-free, lightweight and highly stable in ambient conditions [50]. This semiconductor can be applied a variety of photocatalytic applications such as H₂ and O₂ evolution, solar cell application, air purification, biomedical, and ammonia synthesis [50–52]. Recently, g-C₃N₄ has gained more attention as a non-metallic and visible-light activated photocatalyst and talented material for a variety of environmental remediation applications like degrading pollutants in water and wastewater [53]. Fig. 2 shows the application and controlled fabrication of g-C₃N₄.

Subsequently, g-C₃N₄ has exceptional properties due to its structural condition. For instant, g-C₃N₄ can work in a variety of situations due to its narrow bandgap, physical-chemical-thermal stability, effective visible-light absorption, strong optical characteristics [51,54]. And it is feasible to combine with other materials due to the amino group and triazine ring on the surface. g-C₃N₄ is the most stable carbon nitride phase under ambient temperature, with a two-dimensional layered allotrope comparable to graphite. As a result, when compared to other materials, carbon has the biggest number of documented allotropes. These include graphene, buckminsterfullerene, carbon nanotubes, and many more [55]. g-C₃N₄ is made up of s-triazine or tri-s-triazine units that are connected together using tertiary amines to produce a π-conjugation system [53]. The structure of g-C₃N₄ is relatively flexible, which aids doping with various elements and makes it more compatible and efficient. g-C₃N₄ is the most effective photocatalyst for photodegradation of carbon-based contaminants due to the features described above. Fig. 3 shows the structure and Fig. 4 bandgap position of g-C₃N₄.

Yet, pure g-C₃N₄ has a number of drawbacks, including a low efficiency in visible-light, a high electron hole recombination rate, delayed charge transport, low quantum yield, and lower charge conductivity, ineffective visible-light absorption (below 460 nm), and a small surface area [33,34]. To regulate these limits, various procedures are used, such as heteroatom doping in the bulk form of g-C₃N₄ to establish an impurity level in its forbidden band, which restricted its bandgap energy for

visible light absorption. It has stated that the design of g-C₃N₄ is based on surface area, morphology, reactive sites, abundance, size, and most importantly prolonged light absorption capability. Recently, g-C₃N₄ energy gap engineering by metal and non-metal doping has shown to be quite beneficial in terms of regulating light acquisition ability, modifying redox band potentials, and changing luminescent and electrical features [51]. The photogenerated electron-hole pairs can explain the g-C₃N₄ photocatalytic mechanism under adequate light irradiation. The generation of reactive oxygen species (ROS) and overall electron transport is depicted in Eqs. (1)–(4) [57].



Three phases are involved in the photocatalytic process on g-C₃N₄. Firstly, electrons on the VB of g-C₃N₄ are energised, jump to the CB and VB is left with positive holes when exposed to light. The divided electrons and holes then travel to photocatalyst surfaces, where they initiate redox processes. Dissolved oxygen and H₂O/OH⁻ can operate as electron acceptors and donors, respectively, in aqueous solutions. Finally, the produced superoxide anion (O₂^{·-}) and hydroxyl radical (·OH) have the ability to mineralize organic contaminants and convert them to innocuous chemicals [58]. Table 1 shows the general information regarding crystallite size, surface area, band gap and visible-light degradation rate of g-C₃N₄.

g-C₃N₄ is cost effective due to it is synthesized from numerous low-cost nitrogen precursors e.g., melamine, urea, cyanamide, thiourea and dicyandiamide [25,29,48,59,60] as displays in Fig. 5. g-C₃N₄ photocatalyst has been prepared by using a variety of physical and chemical methods. The top down and bottom-up techniques have widely been accepted. Breaking up larger g-C₃N₄ blocks into smaller units, such as g-C₃N₄ nanosheets, is part of the top-down method. On the other hand, smaller molecules are incorporated into bigger complex compounds via a bottom-up strategy. In addition, sol-gel process, thermal polymerization, solvothermal technique, precipitation, hydrothermal, template assisted techniques were frequently used for the synthesis of g-C₃N₄ photocatalyst [27,61,62]. There have been numerous attempts to produce the necessary g-C₃N₄, where doping has been deemed to be the most effective method. The choice of precursor and synthesis process is crucial for obtaining the desired properties (physical and chemical) in nanohybrids.

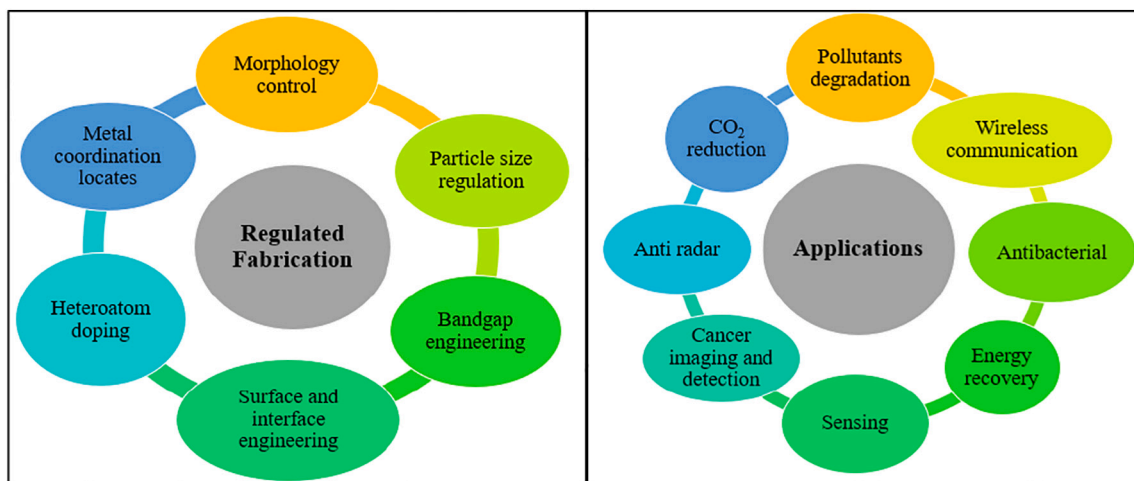


Fig. 2. Potential applications and modulations of g-C₃N₄.

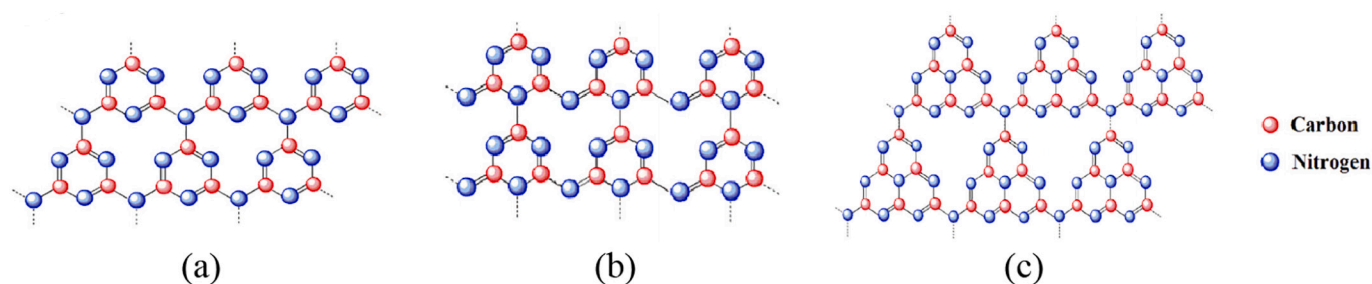


Fig. 3. Structure of $g\text{-C}_3\text{N}_4$ (a) *s*-triazine based hexagonal structure, (b) *s*-triazine based orthorhombic structure, (c) tri-*s*-triazine-based structure [56].

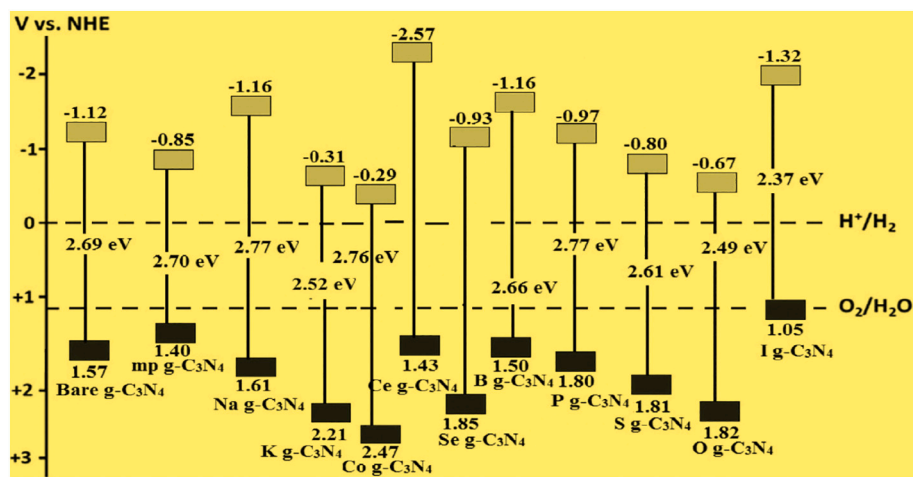


Fig. 4. Band gap positions of pure $g\text{-C}_3\text{N}_4$ and doped $g\text{-C}_3\text{N}_4$ [52].

Table 1

Crystallite size, surface area, band gap and visible-light degradation rate of $g\text{-C}_3\text{N}_4$ derived from different precursors [58].

Precursor composition	Surface area (m^2g^{-1})	Crystallite size (nm)	Band gap (eV)	Rate constant for Acid Orange (AO) 7 (k) (min^{-1})
Melamine	11.24	9.18	2.58	0.034
Urea	55.10	8.39	2.69	0.257
Melamine and urea	18.54	10.02	2.54	0.118
Thiourea	13.10	8.81	2.61	0.009
Thiourea and urea	24.35	9.67	2.77	0.196
Melamine and urea and thiourea	16.48	9.61	2.64	0.043
Melamine and thiourea	9.42	9.36	2.60	0.030

An improved highly thermal polymerization treatment method was applied for the synthesis of $g\text{-C}_3\text{N}_4$ through melamine as a precursor by Feng et al. [48]. A certain amount of melamine was kept into an alumina crucible with proper cover and nonstop heated at $550\text{ }^\circ\text{C}$ for 4 h with a heat rate of $55\text{ }^\circ\text{C}/\text{min}$ in a furnace. The resultant sample kept to cool under room temperature and found bulk yellow colour product that appraised as $g\text{-C}_3\text{N}_4$ (Fig. 6a). Same precursor has been considered by several researchers for fabrication of $g\text{-C}_3\text{N}_4$ photocatalyst under different temperature and duration [59,60,63,64]. $g\text{-C}_3\text{N}_4$ was synthesized by using thiourea as a precursor with a continuous heat at $550\text{ }^\circ\text{C}$ for 2 h in a muffle furnace with a heating rate of $5\text{ }^\circ\text{C}/\text{min}$ [46]. Finally, subsequent yellow colour product is obtained as $g\text{-C}_3\text{N}_4$ (Fig. 6b). A simple thermal heating technique was conducted by Hak et al. [65] to synthesize the $g\text{-C}_3\text{N}_4$ by urea as a precursor. Initially, urea was kept in a

crucible with suitable cover and heat at $80\text{ }^\circ\text{C}$ for 12 h in an oven. Thereafter, the crucible was entered into a muffle furnace and seated at $550\text{ }^\circ\text{C}$ for 3 h towards further heat with a heating ramp of $10\text{ }^\circ\text{C}/\text{min}$. The resulting yellow colour outcome was then rinsed with nitric acid and distilled water. At the end of the process, the wet sample was centrifugation for 10 min at 3500 rpm to remove unwanted residual and dried at $80\text{ }^\circ\text{C}$ overnight to obtain pure $g\text{-C}_3\text{N}_4$ (Fig. 6c). Several studies have developed $g\text{-C}_3\text{N}_4$ using the same precursor but at various heating rates and duration [6,17,23,44,59,61,66].

Thermal decomposition treatment was executed to synthesize $g\text{-C}_3\text{N}_4$ through dicyandiamide as a precursor. Certain amount of dicyandiamide was placed in a crucible and loaded into a muffle furnace for uninterrupted heating at $450\text{ }^\circ\text{C}$ for 2 h with a heating ramp $2\text{ }^\circ\text{C}/\text{min}$ followed by maintaining additional heat for 4 h at $550\text{ }^\circ\text{C}$ [35]. Subsequently, the substance was cooled under room temperature, washed properly with distilled water and filtered to eliminate undesirable ingredients. Finally, the sample was placed inside an oven and heat at $100\text{ }^\circ\text{C}$ to obtain dry $g\text{-C}_3\text{N}_4$ powder (Fig. 6d). The synthesis of $g\text{-C}_3\text{N}_4$ through same precursor utilizing varied temperatures and period of time has been investigated in numerous research [59,60,67]. $g\text{-C}_3\text{N}_4$ was fabricated through a thermal heating process by using cyanamide as a precursor. Cyanamide was placed in a crucible with good cover and loaded into a muffle furnace for constant heat at $500\text{ }^\circ\text{C}$ for 4 h, followed by continuing further heat for 2 h at $520\text{ }^\circ\text{C}$ with a heating ramp $20\text{ }^\circ\text{C}/\text{min}$ [60]. Lastly, the crucible was cooled under room temperature and the obtained yellow colour sample is considered as cyanamide induced $g\text{-C}_3\text{N}_4$ (Fig. 6e).

However, researchers also applied two or more precursors to synthesize the non-metal $g\text{-C}_3\text{N}_4$ photocatalyst. $g\text{-C}_3\text{N}_4$ was prepared by Shi et al. [31] through the mixture of melamine and urea as a precursor under thermal heating process. A specific quantity of melamine and urea were mixed properly by grinding in a certain period. Subsequently, the

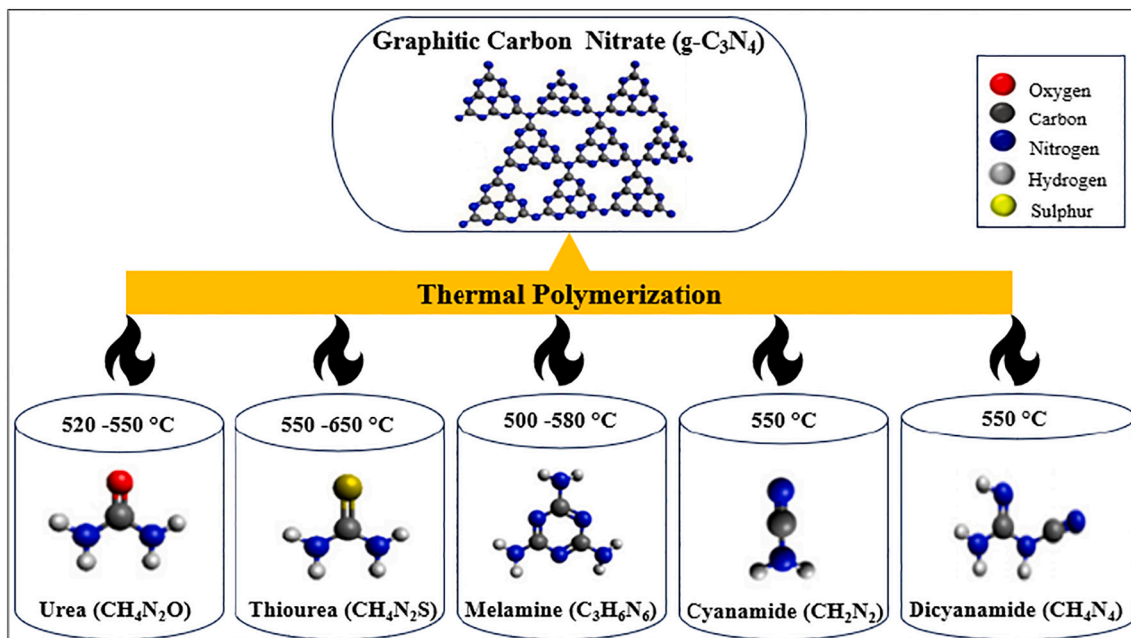


Fig. 5. Development and synthesis process of g-C₃N₄ through thermal polymerization.

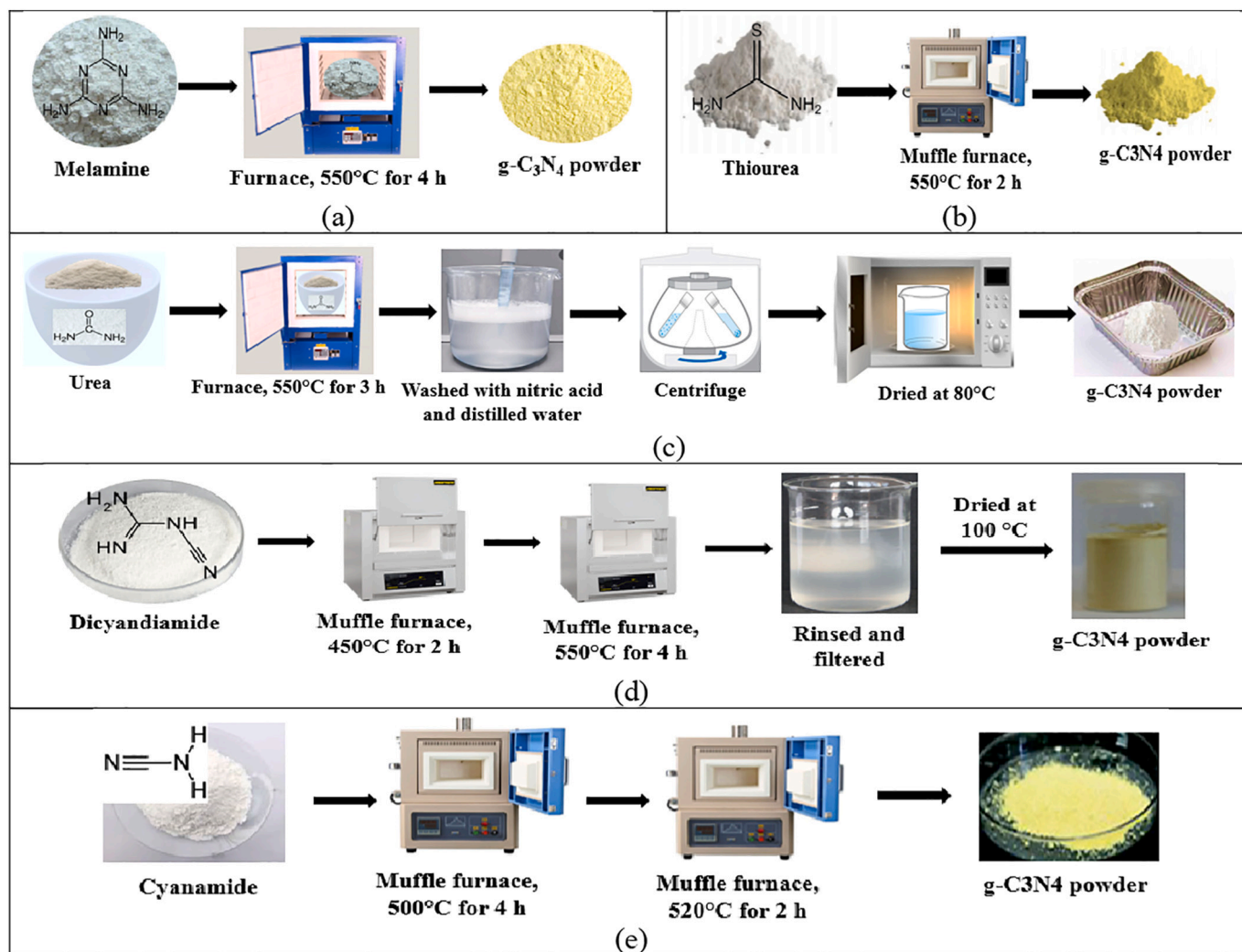


Fig. 6. Synthesis of g-C₃N₄ from (a) Melamine; (b) Thiourea; (c) Urea; (d) Dicyandiamide; (e) Cyanimide.

mixture is placed in an alumina crucible and continuous heat at 550 °C for 4 h with a heating ramp 2.3 °C/min. Finally, the obtained yellow colour powder sample is found as g-C₃N₄ photocatalyst. Furthermore, the same precursors were applied by Xu et al. [68] to synthesize the g-

C₃N₄ with diverse duration and temperature. A combination of urea and thiourea was polymerized for 2 h at 550 °C with a heating ramp rate of 3 °C to fabricate g-C₃N₄ [69]. On the other hand, Yao et al. [67] synthesized g-C₃N₄ by utilizing the mixture of dicyandiamide and urea. The

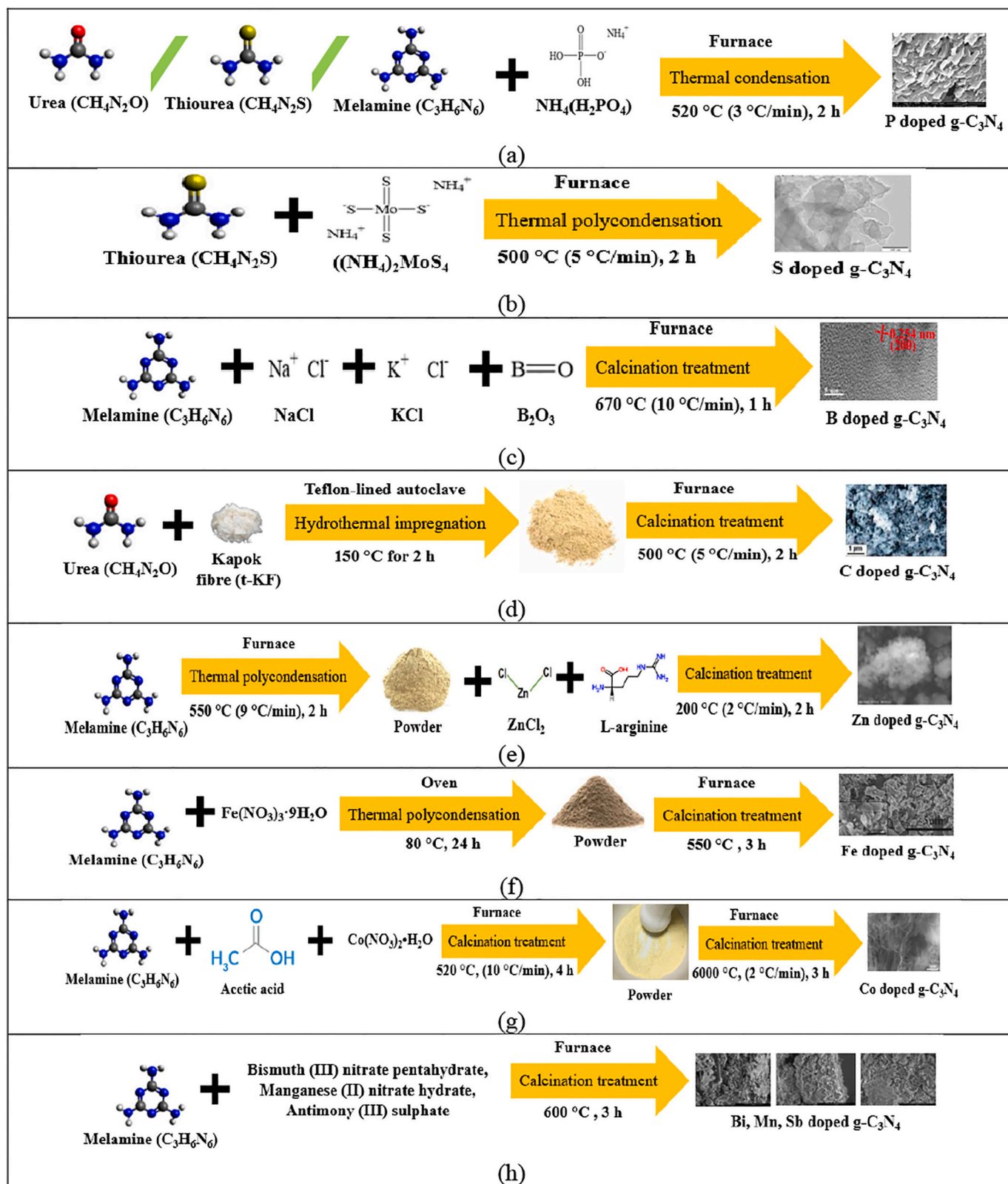


Fig. 7. Synthesis routes for the preparation of non-metal (a) Phosphorous (P) [25]; (b) Sulphur (S) [73]; (c) Boron (B) [74]; (d) Carbon (C) [75] and metal (e) Zinc (Zn) [76]; (f) Iron (Fe) [77]; (g) Cobalt (Co) [78]; (h) Bismuth (Bi), Manganese (Mn), Antimony (Sb) [79] doped g-C₃N₄.

mixture was then nonstop heated at 550 °C for 4 h, followed by keeping the heat at 500 °C for additional 2 h to obtain the yellow colour substance considered as g-C₃N₄.

Although, a number of techniques have been established for the fabrication of nanocomposites based on g-C₃N₄. The preparation of g-C₃N₄ based catalysts can be split into three categories: (i) physical (physical vapor deposition, ultrasonic treatment, chemical vapor condensation, solid state reaction, gas condensation), (ii) chemical (template, thermal polymerization, microwave-assisted heating, pyrolysis, sol-gel, hydrothermal/solvothermal method) and (iii) mechanical synthesis methods (mechanical grinding) [56,70]. Physical procedures do not disrupt the crystal lattice structure of g-C₃N₄, but they are hardly used in practical production because of the necessity for high-purity raw materials and specialised equipment [71]. It has been discovered by the scientific community that doped g-C₃N₄ is more effective than pure g-C₃N₄ [23,44,72] (Fig. 7).

Based on the above discussion, it can be summarized that the precursor has a significant importance in the quality and performance of g-C₃N₄. The precursors have the potentiality to enhance the morphology and optical properties of any composites in conjunction with g-C₃N₄ [48]. That particularly improve the application and overall performance of the particular composite photocatalyst. Among the various precursors of g-C₃N₄, urea possess optimum quality and highly efficiency in the field of pollutants removal and energy evolution [6,65]. In addition, as a precursors urea is cheap, abundant and has the capability to prepare more effective composite photocatalyst modifying with various biological and chemical synthesized CQDs [80]. On the other hand, some precursors of g-C₃N₄ need to be rinsed with acid and distilled water prior to use or combined with other photocatalyst due to the removal of any unwanted particles, residual and impurities [65].

3. Fundamentals of CQDs photocatalyst

CQDs are a novel form of 0-dimensional carbon material with a size <10 nm [81,82]. CQDs are made up of discrete nanoparticles and quasi-spherical particles with layered of graphene-based structures. Researchers from a different fields have used CQDs for a variety of purposes such as biomedical and biotechnological applications, dye-synthesized solar cells, light-emitting devices, imaging and bio-imaging, electrochemical studies, semiconductor devices, chemical sensing, photo-catalysing, and electrocatalysis [81,83–88]. CQDs have gotten a promising attention in several applications, particularly in wastewater treatment. Because of its effective characteristics such as low toxicity, high dispersibility of water, low-cost manufacturing, stable Photoluminescence (PL), superior chemical stability, outstanding photostability and electron reservoir features [81,82,89]. Due to quantum

confinement effect and optical stability qualities, CQDs provide a good photoluminescent signal, strong fluorescence activity, chemical inertness, and durability [81]. In energy conversion applications, CQDs can be used as a substitute for semiconductor quantum dots. The properties of CQDs are depicted in Fig. 8a.

CQDs are a type of conjugated π structure material that is made up of amorphous to nanocrystalline graphite carbon or graphene [44]. CQDs have excellent electron transport and storage capabilities. CQDs also have outstanding PL capabilities for both up and down conversion, as well as strong electrical and optical functionality [44,82]. The conjugated π -domains and PL emission related with the surface state, conjugated structures, and specific structure locations are credited with CQDs capacity to fully use the sunshine spectrum. Furthermore, CQDs can transform long-wavelength into short-wavelength because of having various nanoparticle sizes and emissive trap states on the CQD surface [86]. The carbon precursors emissivity used as raw materials also affect the luminous properties of CQDs [90]. Fig. 8b depicts the fundamental distinctions between CQD surface functionalization and passivation procedures.

CQDs can be synthesized via breaking bulk carbon structures and the chemical reactions of precursors. This process is conducted under two main methods named top-down and bottom-up approach. However, there are five sub approaches are commonly considered under both approaches. For example, the top-down methods are laser ablation, arc discharge, electrochemical oxidation, ultrasonic treatment, chemical ablation and bottom-up approaches are hydrothermal, template method, plasma treatment, thermal decomposition and microwave irradiation techniques. Among the entire synthesis approach of CQDs, hydrothermal and microwave treatments are common, facile, and widely applicable [45,91,92].

CQDs were successfully synthesized by Remli and Aziz [91] through watermelon rinds as a carbon source in a simple hydrothermal synthesis method. Watermelon rinds juice was mixed with ethanol (4:3 ratio) and stirred for a certain period for homogenous suspension. Thereafter, the mixture was transferred into a stainless-steel Teflon autoclave and continuous heating for 2.5 h at 120 °C in an oven. In order to separate the less fluorescent deposit and obtain highly fluorescent CQDs, the resulting dark brown solution is centrifuged and rinsed with dichloromethane and acetone (Fig. 9a). CQDs were synthesized by improved sonication technique by using glucose, distilled water and sodium hydroxide solution [48]. 2 h ultrasonic treatment was applied to the combined and colourless solution. After that, HCl was used to neutralise the brownish yellow solution and further dialysis to obtain the purified CQDs solution [93] as represented in Fig. 9b. Therefore, different metal and non-metal dopes CQDs offers outstanding performance in various application by overcoming monochromatic fluorescence and less

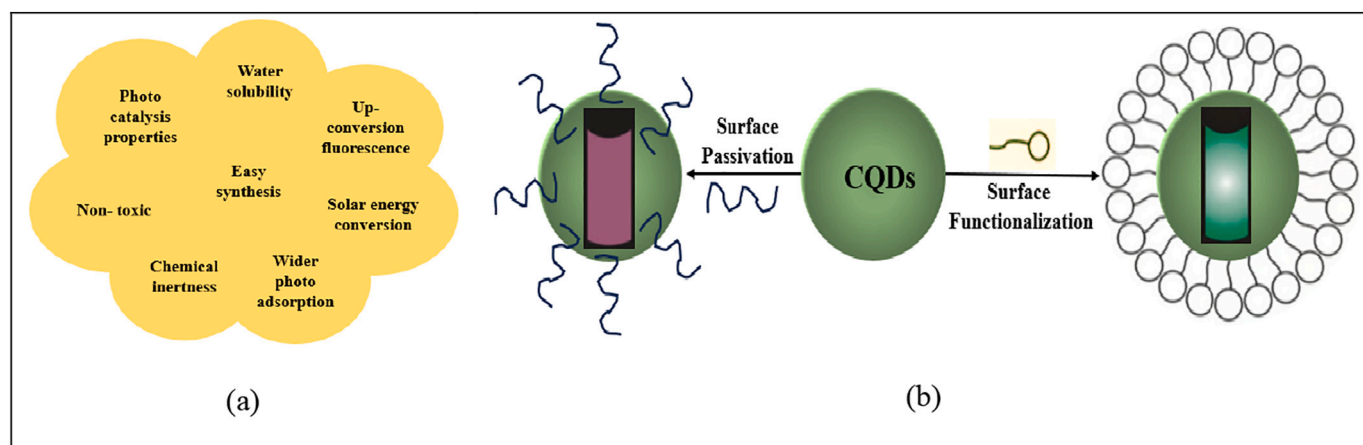


Fig. 8. (a) Properties, (b) Surface functionalization and passivation of CQDs [90].

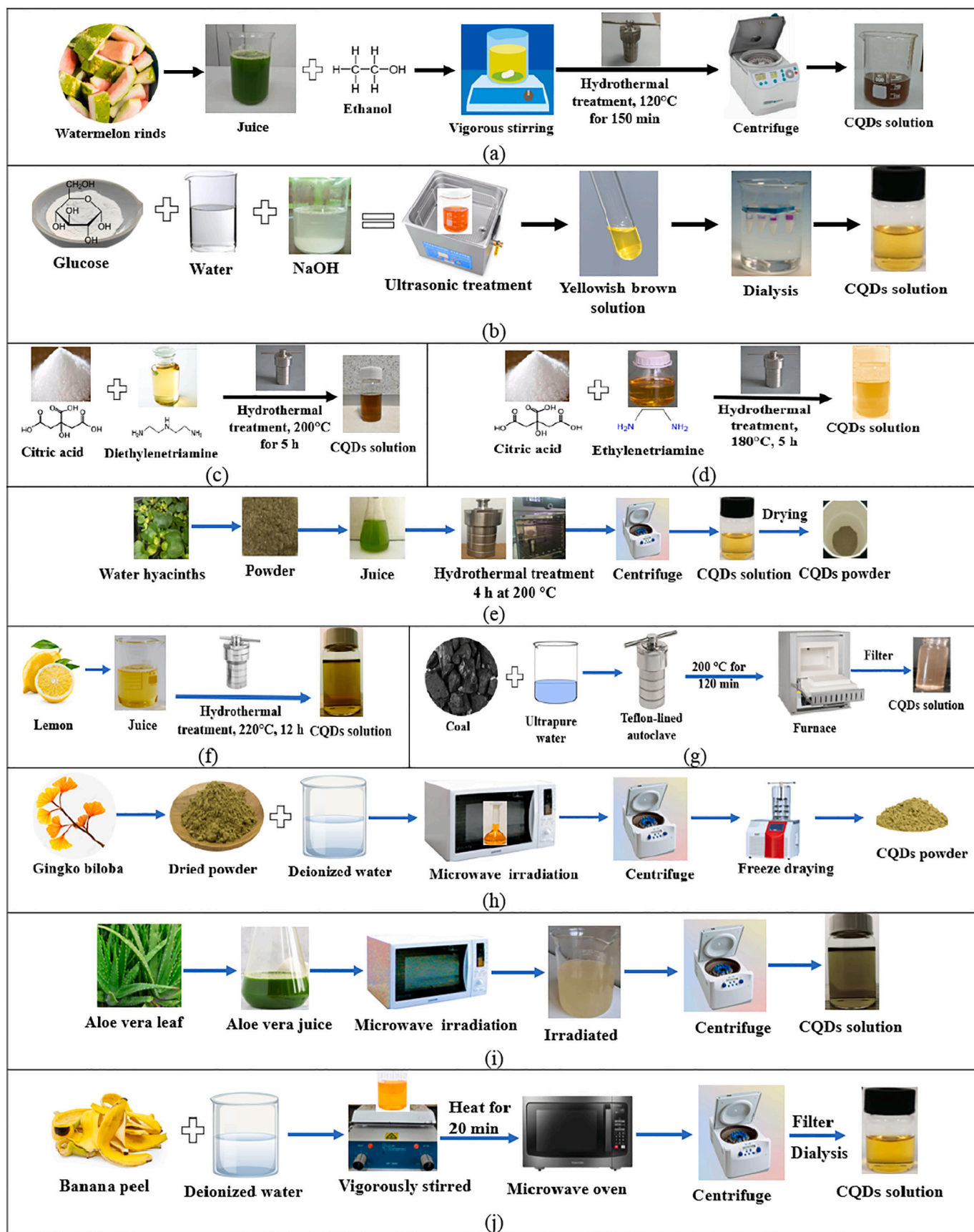


Fig. 9. Synthesis of CQDs from (a) watermelon rinds; (b) glucose; (c-d) citric acid; (e) water hyacinths; (f) lemon; (g) coal; (h) ginkgo biloba; (i) *Aloe vera* leaf; (j) banana peels.

fluorescence yield of CQDs [44,94]. Aghamali et al. [95] fabricated nitrogen doped CQDs (N-CQDs) through the hydrothermal synthesis method by using citric acid. Certain amount of citric acid and diethylenetriamine was added with distilled water and shifted to autoclave with a temperature at 200 °C in an oven for 5 h. A transparent, light-yellow solution was formed once the process completed and considered as N-CQDs (Fig. 9c). Numerous researchers applied the same precursors for the development of N-CQDs [31,44,61,96] with different dose, duration and temperature. On the other hand, Hong et al. [30] also synthesized CQDs by applying advance hydrothermal method with citric acid, distilled water and ethylenediamine solution. The solution was moved into an autoclave and constant heated for 5 h at 180 °C. The reaction was followed by a natural cooling of the autoclave to room temperature. A semi-permeable membrane was used to perform dialysis on the product in order to get the CQDs as described in Fig. 9d. Similar method and process were followed by several researchers [6,17,45,64] to synthesize CQDs, however the heating temperature and duration was different.

Fabrication and development of the CQDs by using biomass is modern innovation by researchers. Hak et al. [65] formed CQDs by facile hydrothermal technique through dried water hyacinths leaves. The mixture of distilled and water hyacinths leaves powder were vigorously stirred to make the homogenous solution. Later on, the solution was shifted into an autoclave for continuous heating at 200 °C for 4 h in the oven. The autoclave was cooled at room temperature and centrifugation to remove the large particle to obtain yellowish-brown solution considered as CQDs. Finally, the solution is dried at 100 °C in an oven to secure solid form of CQDs as shows in Fig. 9e. Lemon juice was exert to synthesize CQDs with a certain volume of ethanol via hydrothermal treatment by Jin et al. [46]. The pre mixture solution was flushed into an autoclave and a continuous heated for 12 h at 200 °C in an oven to get the CQDs solution as illustrates in Fig. 9f. Lemon juice also employed to synthesize CQDs by Olmos-Moya et al. [97] through microwave assisted technique under diverse temperature and duration. Coal and distilled water were mixed to fabricate CQDs through hydrothermal treatment method by Saikia et al. [88]. Initially, the mixture was placed in a Teflon-lined autoclave reactor and later on loaded into a furnace for continuous heat for 2 h at 200 °C (Fig. 9g). After air cooling, the pH was determined and the solution was undergone for filtration with a dialysis membrane kit to preserve the CQDs.

Furthermore, CQDs have been synthesized through microwave assisted technique from dry ginkgo biloba as a carbon precursor by Genc et al. [92]. Certain amount of ginkgo biloba powder was mixed with distilled water and loaded into a domestic microwave oven at various period of time scale. Thereafter, the dark-brown mixture was centrifugated and the supernatant was collected by filtering. Finally, the supernatant was dried in a vacuum freeze to obtain powder is considered as CQDs as displayed in Fig. 9h. Another research has been conducted by Malavika et al. [98] for the preparation of CQDs from *aloe vera* leaf juice under microwave assisted synthesis process. Fresh and smashed *aloe vera* leaf was merged with deionized water and shifted to a conical flask for microwave treatment. After a certain period, the reaction is completed and sample is collected to allow cool at ambient temperature. Subsequently, the sample was centrifuged to avoid unwanted particles and contaminants. Finally, CQDs are collected by a separation method named silica gel followed by proper dialysis as exhibited in Fig. 9i. It is mentioned that CQDs were formed from the biomass by the acquired fluorescence emission under UV light [6,85,97,98]. Huang et al. [99] synthesized CQDs through the mixture of banana peels and distilled water under microwave assisted method. The pre-mix solution of banana peels and distilled water was placed to a domestic microwave oven for 20 min. The resultant solution was cooled under room temperature and centrifuge to eliminate undesired large particles. The resultant solution is filtered and dialyzed to obtain CQDs solution (Fig. 9j). Finally, the solution is lyophilized to secure the powder considered as CQDs.

4. Design of CQDs modified g-C₃N₄ photocatalyst

Although, there are some drawbacks of pure g-C₃N₄ such as limited surface area, poor quantum efficiency and high electron hole. These limitations are responsible to reduce the performance of g-C₃N₄ in various environmental applications. Numerous studies indicate that CQDs modified g-C₃N₄ has more photoactivity than pure g-C₃N₄ by encountering and improving the surface area, quantum efficiency, electron transfer and reservoir properties [6,30,31,44,52,85]. There are several classes of metal free CQDs modified g-C₃N₄ ternary and binary photocatalysts and significantly enhance the photocatalytic performance. There is a schematic representation of the classification of CQDs embedded g-C₃N₄ as displays in Fig. 10.

Presently, a number of strategies have been devised to fabricate CQDs modified g-C₃N₄ nanocomposites because of their momentous application for the abolition of pollutants and alternative green hydrogen energy evolution. However, the physical, chemical and optical characteristics of the modified photocatalysts are critically influenced by the synthesis techniques and conditions. For example, Li and co-workers investigated the photocatalytic efficiency of CQDs modified g-C₃N₄, fabricated through a direct calcination process by the combination of melamine, and cyanuric acid [100]. A certain amount of cyanuric acid was mixed with melamine and distilled water for 12 h under ardent stirring, wherever melamine and cyanuric acid have a 1:1 M ratio. Thereafter, the milky solution is vaporising in an oil bath drying oven at 90 °C. The powder was then put into an alumina crucible with a cover and heated for 4 h at 500 °C in a muffle furnace with a 2.3 °C/min. Once it has naturally cooled to ambient temperature, the obtained solid powder form is considered as CQDs modified g-C₃N₄ as displayed in Fig. 11a. According to this study, the pre-prepared CQDs/g-C₃N₄ in an exceptional multifunctional photocatalyst hydrogen evolution through water splitting and dye degradation. This research demonstrated incredibly high hydrogen evolution rate of 24,760 µmol/h/g and completely degradation of MB within 240 min. Most recently, Liu et al. [23] successfully synthesized CQDs/g-C₃N₄ composite through citric acid and urea under impregnation approach. The synthesized nanocomposite demonstrated superior light harvesting, a narrow band-gap, and workable electron transport capabilities, which led to increased photocatalytic activity for the degradation of RhB. Furthermore, the nanocomposite completely degraded RhB within 120 min and offered a hydrogen evolution rate of 626.93 µmol/h/g. The simultaneous operation of the oxidation and reduction processes is thought to be the cause of the improved photocatalytic efficiency and increasing the number of electrons and holes produced for redox processes as well as the effectiveness of charge carrier separation. On the other hand, Zhang et al. [6] fabricated CQDs/g-C₃N₄ composite through the same precursors nevertheless under the hydrothermal treatment approach. The efficiency of producing hydrogen as well as degrading MB and RhB was significantly improved by CQDs/g-C₃N₄ composite under visible light. The result displayed that the MB and RhB were completely degraded by 20 min and 110 min, respectively. Simultaneously, the hydrogen evolution rate was 1291 µmol/g/h under 300 W Xe lamp. However, charge transfer and separation efficiency of the e⁻/h⁺ pairs is critically increased by adding CQDs on the surface of g-C₃N₄.

Hak et al. [65] synthesized CQDs/g-C₃N₄ by the mixing of water hyacinths induced CQDs and urea induced g-C₃N₄ under simple hydrothermal method. Following that, the mixture was placed in a Teflon-lined autoclave and nonstop heated at 100 °C for 2 h. The autoclave then allowed to cool in room temperature and the resultant samples were rinsed with distilled water. Thereafter, the washed solution is undergone for centrifugation and the precipitate was dried at 80 °C to secure the CQDs/g-C₃N₄ composite as shows in Fig. 11b. Concurrently, CQDs/g-C₃N₄ composite has been fabricated by using thiourea tempted g-C₃N₄ and lemon synthesized CQDs [46]. The mixture was then effectively stirred before being put into an autoclave for continuous heat for 12 h at 220 °C in an oven. The precipitant was assembled by centrifugation and

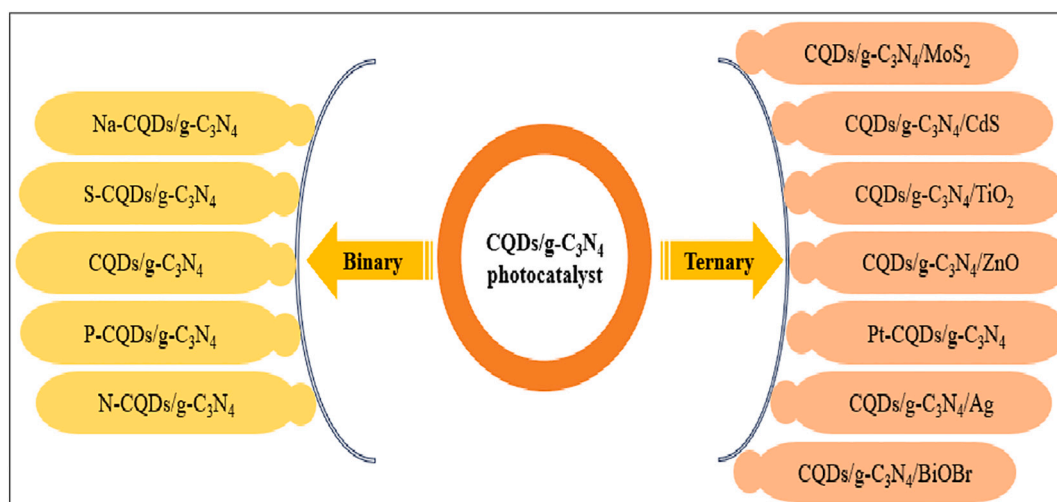


Fig. 10. Graphical representation of the classification of CQDs/g-C₃N₄ based photocatalysts.

washed with distilled water and ethanol before being dried at 60 °C in an oven. Finally, the obtained powder is considered as CQDs/g-C₃N₄ nanocomposite as shows in Fig. 11c. Hong and coresearchers [30] synthesized CQDs/g-C₃N₄ heterojunctions by low temperature method with the presence of citric acid and melamine as precursors [64]. The mixture is simply nonstop stirred for 12 h and CQDs/g-C₃N₄ composite was collected by drying 80 °C in an oven as expressions in Fig. 11d. Urea and citric acid induced bulk g-C₃N₄ and CQDs has been applied to fabricate CQDs/g-C₃N₄ composite [29,85]. Different volume of CQDs and g-C₃N₄ were merged by magnetic stirring and dried at 60 °C to obtain as powder form. Thereafter, the powder was heated for 3 h at 550 °C with a heating ramp of 15 °C/min as shows in Fig. 11e-f. Finally, the sample was collected and considered as CQDs/g-C₃N₄ composite. Similar precursors has been utilized by numerous researchers to synthesize noble CQDs/g-C₃N₄ nanocomposite [17,44,45,61,66]. Wang and coworkers [63] prepared CQDs/g-C₃N₄ photocatalysts through L-Ascorbic acid and melamine as precursors by simple hydrothermal technique. Certain amount of melamine induced g-C₃N₄ was mixed into the L-Ascorbic acid and distilled water solution by sonication and vigorously magnetic stirrer. Subsequently, the mixture was placed into a Teflon-sealed autoclave for continuous heated for 4 h 180 °C and then allowed to cool at room temperature. The sample was centrifuged and washed by distilled water and ethanol, dried at 60 °C to collect the precipitants considered CQDs/g-C₃N₄ composite as illustrates in Fig. 11g.

4.1. Characterization of CQDs-based g-C₃N₄ photocatalytic materials

4.1.1. Fourier transform infrared (FTIR) spectroscopy

The functional and fingerprint group of CQDs, g-C₃N₄ and CQDs/g-C₃N₄ composite are acknowledged by FTIR spectroscopy as shows in Fig. 12. According to Hak et al. [65], an extensive and slight peak of water hyacinths leaves synthesized CQDs is noticed at 3400 cm⁻¹ and 2932 cm⁻¹ that attributable to the stretching vibration mode of O—H and C—H, respectively (Fig. 12a). However, outstretched carbonyl groups of C=O is confirmed due to 1616 cm⁻¹ band and vibrational band peaks (1052 cm⁻¹ and 1112 cm⁻¹) is correspondent to C—O—C vibrations stretching. On the other hand, urea synthesized g-C₃N₄ offers numerous absorption peaks at 1241 cm⁻¹ and 1630 cm⁻¹ that dispensed to C—N (sp³) and C=N (sp²) stretching that matches with the typical stretching modes of the CN heterocycles. Thus, the triazine breathing mode of g-C₃N₄ was attributed to the strong peak at 808 cm⁻¹. When CQDs is merged with g-C₃N₄, the intensity of these distinctive bands is reduced in all composites that is may be due to low doses of CQDs as shows in Fig. 12a. The frail CQDs absorptions peak was obscured by the robust g-C₃N₄ absorptions peak. As CQDs dose is increased by 40 wt%,

the stretching mode of CN heterocycles shows a little blue-shift from 1241 cm⁻¹ to 1238 cm⁻¹ (Fig. 12a) that is due to reaction between CQDs and g-C₃N₄. Li and peers [85] synthesized CQDs/g-C₃N₄ photocatalyst by using urea and citric acid as carbon precursor where N—H group is shown by the large band at 3000–3400 cm⁻¹ (Fig. 12b). In contrast, the O—H band is caused by surface-adsorbed hydroxyl groups and uncondensed amino groups. Peaks at 1200–1600 cm⁻¹ and 807 cm⁻¹ are associated with the triazine unit stretching and breathing modes, respectively [23,100]. It can be can infer that the initial g-C₃N₄ structure was not materially reformed by the addition of a slight quantity of CQDs. Consequently, L-ascorbic acid and melamine synthesized CQDs, g-C₃N₄ and CQDs/g-C₃N₄ photocatalyst by Wang et al. [63] shows outstanding functional group through FTIR. The position of the primary distinguishing absorption peaks of CQDs at 786, 1023, 1384, 1621, 1704, 2868, 2970, and 3419 cm⁻¹ which allocated the functional groups of C—O, C=O, —CH₃, C=C, C=O, —CH₂, —CH₃ and —OH as shows in Fig. 12c. Different absorption peaks are associated with the functional groups of the tri-s-triazine structure (808 cm⁻¹), heptazine heterocycles (1635 cm⁻¹, 1410 cm⁻¹, and 1242 cm⁻¹) and the unreacted amino group (3100 cm⁻¹ - 3300 cm⁻¹) in g-C₃N₄. Interestingly, CQDs/g-C₃N₄ photocatalyst also offers the similar spectrum with g-C₃N₄ because of low dose of CQDs on the surface of g-C₃N₄ and the intersection of peak positions [85,101]. Several functional groups including OH, CH, NH, C=C, C=O, C—H, O—H, C—O reflecting the abundance of carboxyls, hydroxyls, and amines functions [23,96].

4.1.2. Transmission electron microscopy/high resolution TEM (TEM/HRTEM)

Internal structure of CQDs, g-C₃N₄ and CQDs/g-C₃N₄ nanocomposite, their size distribution and morphology can be described by TEM. Citric acid induced CQDs offers a homogenous distributed circular shape with a size of 4 nm in TEM image [29] as displays in Fig. 13A1. TEM of Ethylenediaminetetraacetic acid synthesized pure CQDs offers a size of 5–10 nm that are well diversified (Fig. 13A2) [102]. However, the size of citric acid synthesized CQDs are around 10 nm [66] as shows in Fig. 13A3, that may be due to the CQDs solution not having undergone proper filtering. On the other hand, citric acid synthesized CQDs offers a well distributed carbon dots with average size of 5 nm. In addition, the lattice gap of a single CQDs particle in the HRTEM image is approximately 0.21 nm (Fig. 13A4) [31]. Alike, TEM image of citric acid synthesized CQDs displays a uniform distribution of carbon dots primarily in sizes between 2 and 5 nm (Fig. 13A5) [6]. So, the size of CQDs and their distribution can be varied not only for precursors but also synthesis duration, temperature, environment and methods. TEM image of g-C₃N₄ offers several tremendously folded paper sheet with micron in size as

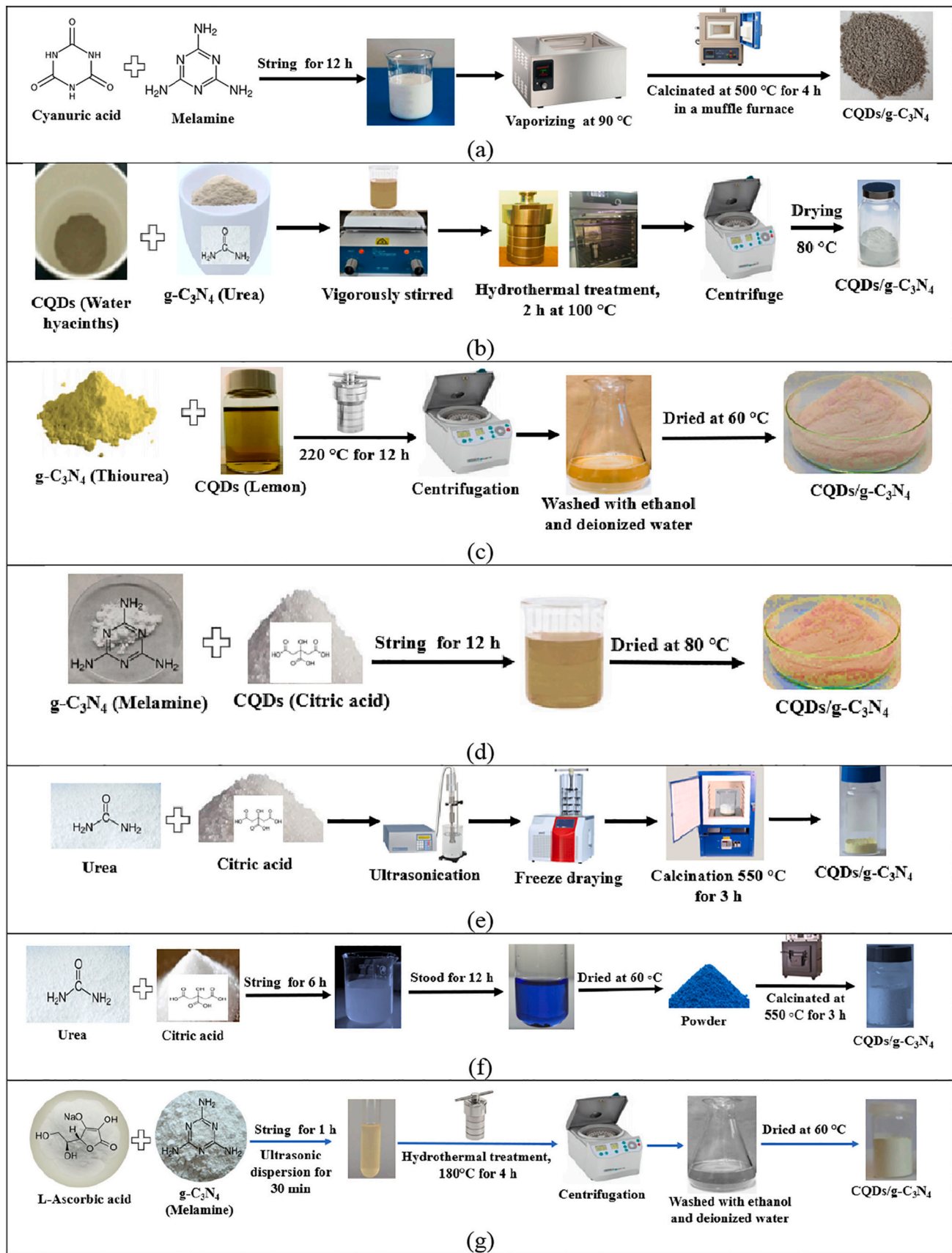


Fig. 11. Synthesis of CQDs/g-C₃N₄ composite from (a) cyanuric acid and melamine; (b) water hyacinths and urea; (c) lemon and thiourea; (d) melamine and citric acid; (e-f) urea and citric acid; and (g) L-ascorbic acid and melamine.

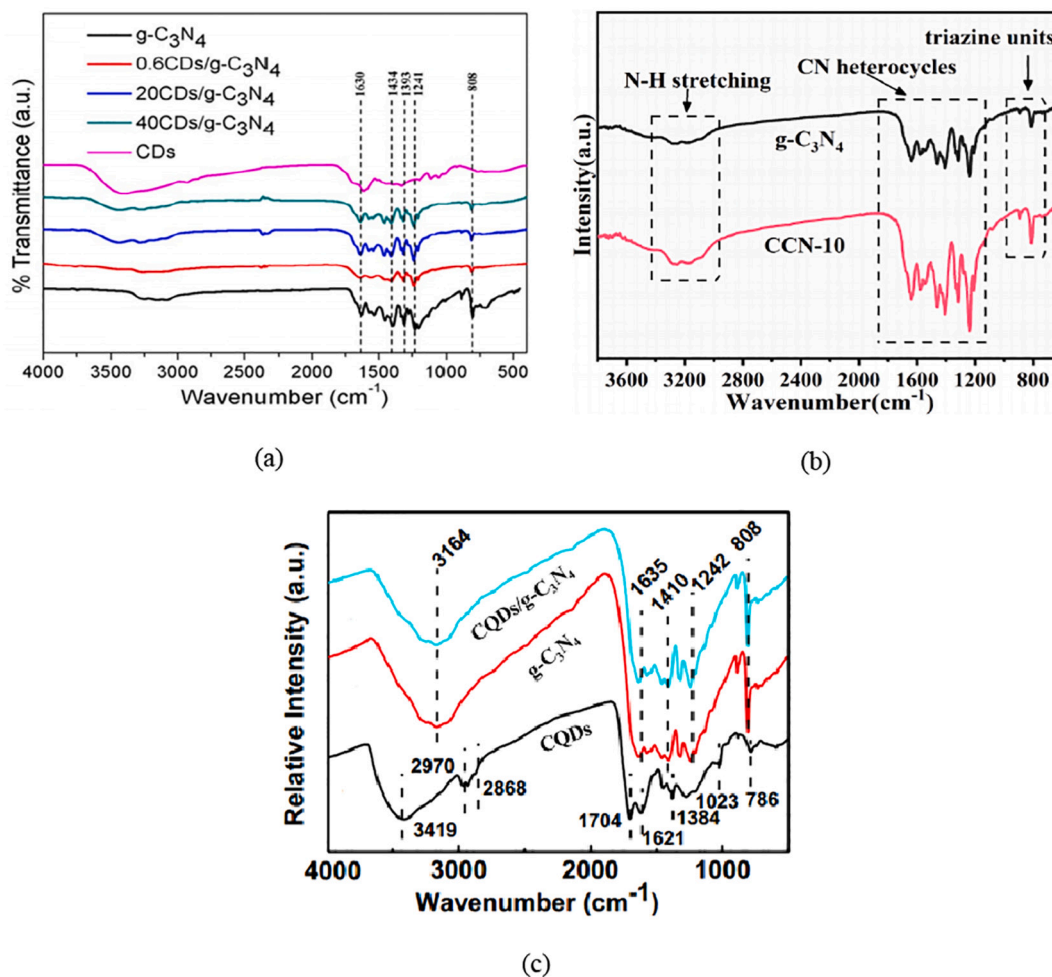


Fig. 12. FTIR spectra of (a) CQDs, g-C₃N₄, CQD/g-C₃N₄ composites [65]; (b) g-C₃N₄ and CQD/g-C₃N₄ composite [85]; (c) CQDs, g-C₃N₄ and CQDs/ g-C₃N₄ composite [63].

shows in Fig. 13B1 [29]. Additionally, TEM image of g-C₃N₄ revealed a two-dimensional laminated structure [102] in Fig. 13B2 and a flat surface with a sheet-like two dimensional structure [30] as shown in Fig. 13B3. Moreover, a lamellar morphology structure of g-C₃N₄ is found according TEM image in Fig. 13B4 [63]. In the TEM image of g-C₃N₄ (Fig. 13B5), ripples and wrinkles resembling chiffon were also observed in a two-dimensional layered structure [6].

The successful and uniform distribution of CQDs on the surface of g-C₃N₄ is well-defined by the TEM image as displays in Fig. 13C1 to C7. Yet, it is observed from Fig. 13C1 that the surface of acid synthesized CQDs modified g-C₃N₄ looks bumpy that indicating a significant loading of foreign substance onto the composite [29]. However, the little shady spots in Fig. 13C2 are expressed the homogenous distribution of biomass synthesized CQDs on the surface of g-C₃N₄ [65]. It was also discovered in Fig. 13C3 that CQDs had loaded on the surface of g-C₃N₄ and their size harmonised that of pure CQDs [102]. The CQDs/g-C₃N₄ samples in Fig. 13C4 similarly exhibit the two-dimensional sheet-like structures, with the exception of a few black dots that are visible on their surfaces [30]. On the other hand, acid synthesized CQDs are uniformly inserted in the porous tubular g-C₃N₄ and are identified by a white circle in Fig. 13C5 [100]. Li and coworkers [85] observed that urea synthesized g-C₃N₄ is equally dispersed with citric acid synthesized CQDs with an average size of 8 to 10 nm. However, Zhang et al. [6] also applied the same precursors to fabricate CQDs/g-C₃N₄ nanocomposite but the composite is not uniformly distributed even though the size of CQDs was 2 to 5 nm (Fig. 13C7). So, synthesize method, temperature, duration and chemicals has the potentiality to enhance the distribution and size of

CQDs.

According to the HRTEM image of CQDs/g-C₃N₄ (Fig. 13D1), black specks with widths of 2 to 5 nm were seen dispersing uniformly on the surface of g-C₃N₄, though some of them consolidated into bigger particles after thermal polymerization [29]. The HRTEM image of CQDs/g-C₃N₄ further demonstrated the presence of the usual lattice stripes of CQDs onto the g-C₃N₄ surface. HRTEM image of CQDs/g-C₃N₄ (Fig. 13D2) further confirms the presence of nano sized (1.2 to 4.9 nm) CQDs with lattice fringes of 0.213 nm, which correspond to (100) in-plane lattice spacing of graphene [23,65,93]. It has been discovered in Fig. 13D3 that few black dots are exist in CQDs/g-C₃N₄ composite with a size of around 5 nm, and that clearly discernible lattice spacing of 0.321 nm corresponds to the (002) crystal plane of CQDs [30]. Similar lattice spacing (0.326) and crystal plane (002) was observed in Fig. 13D4 but the size of CQDs are in between 2.5 and 3.5 nm that's why CQDs/g-C₃N₄ composite is observed as uniformly distributed [46,101]. The interplanar spacing of the CQDs crystallite in the HRTEM pictures (Fig. 13D5—D6) are 0.24 and 0.22 nm, which is equal to the (101) spacing of g-C₃N₄ demonstrating their close proximity to one another which favours charge transfer at the interface [23,85,100]. Hydrothermally treated L-Ascorbic acid and melamine synthesized CQDs/g-C₃N₄ composite shows a uniform distribution of CQDs (5 nm) on the lamellar surface of the g-C₃N₄ [63]. The CQDs are virtually amorphous and have weak crystallization [6], as shown by the HRTEM image that follows in Fig. 13D7. Finally, it can be summarized that there is a major influence of the size of CQDs for the proper distribution on the surface of g-C₃N₄ to

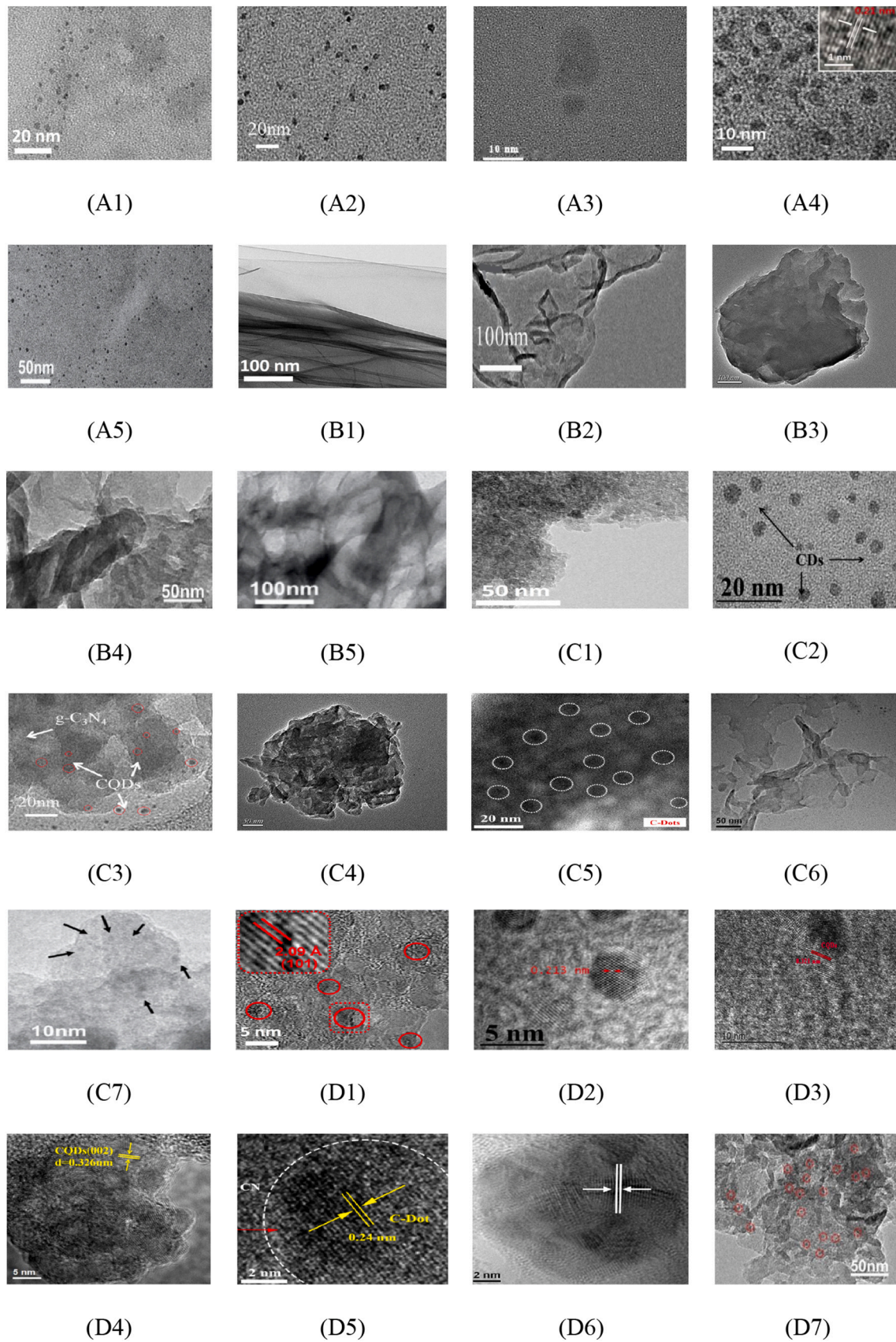


Fig. 13. TEM images of CQDs (A1) [29], (A2) [102], (A3) [66], (A4) [31], (A5) [6]; g-C₃N₄ (B1) [29], (B2) [102], (B3) [30], (B4) [63], (B5) [6]; CQDs/g-C₃N₄ (C1) [29], (C2) [65], (C3) [102], (C4) [30], (C5) [100], (C6) [85], (C7) [6] and HR-TEM image of CQDs/g-C₃N₄ (D1) [29], (D2) [65], (D3) [30], (D4) [46], (D5) [100], (D6) [85], (D7) [63].

prepare the ideal CQDs/g-C₃N₄ composite. Nevertheless, the discussion shows that CQDs were successfully settled on the surface of g-C₃N₄.

4.1.3. X-ray photoelectron spectroscopy (XPS)

Elemental composition and chemical states of CQDs/g-C₃N₄ nanocomposite is identified by XPS. Based on elemental percentage in Fig. 14a, CQDs/g-C₃N₄ composite has a greater C and N atom ratio than g-C₃N₄ [85], which is probably because of the presence of CQDs. Liu and peers [23] also found C, N and O on the surface of CQDs/g-C₃N₄ nanocomposite (Fig. 14b). The fundamental heterocycle edifice of g-C₃N₄ has been verified by the C1s and N1s XPS spectra [101]. According to Zhang et al. [6], CQDs/g-C₃N₄ nanocomposite is mostly formed by carbon, nitrogen, and oxygen with photoelectron peaks at 288 (C 1s), 401 (N 1s), and 530 eV (O 1s) (Fig. 14c). CQDs/g-C₃N₄ photocatalyst is free of other unintended contaminants and there is only CQDs and g-C₃N₄ states is present. Similar photoelectron peaks were observed by Hak and coworkers [65] in their article about biomass synthesis CQDs/g-C₃N₄ photocatalyst preparation. The C1s peak was divided into three peaks at energies of 284.77 eV, 287.97 eV, and 288.83 eV, which were assigned to graphitic carbon (C—C), carbonyl (C=O), and sp² carbon (N—C=N), respectively (Fig. 14d). The major peak at 398.51 eV was attributed to CN=C coordination while the other two faint peaks were attributed to tertiary (N-(C)3) and amino functional groups (C-N-H) at binding energies of 399.80 eV and 400.77 eV, respectively (Fig. 14e). The absorbed water was credited with producing O 1s signal at 531.91 eV (Fig. 14f) that revealing of C—O and C=O bond groups [96].

Besides, due to photoluminescent (PL) effect, CQDs not only act as a photosensitizer and an electron acceptor or mediator to control the flow of charge carriers, but they also act as a semiconducting material to produce photogenerated electron-hole pairs as displays in Fig. 15. The rapid production of charge carriers once sufficient photons has been absorbed over photocatalysts is one of the key concepts in photocatalysis

that has the potentiality to enhance the photocatalytic ability of any photocatalysts. However, CQDs has the potentiality to impede the recombination of e⁻/h⁺ pairs, generate charge carriers by converting light with longer wavelengths to light with shorter wavelengths and offers the properties for ideal electron donor/acceptor that finally contributes to potential photocatalytic activity [6,59,80]. In detail, CQDs has the potentiality to enhance the photocatalysis system when it loaded on the surface of g-C₃N₄. Due to the photoluminescent effect, CQDs serve as electron receivers or mediators to control the flow of charge carriers, photosensitizers, and electron-hole pairs produced by photosensitizers [6,13,17,65]. Additionally, morphology transformation, heteroatom doping, fabrication of interface heterostructure techniques have the potential to change the electrical structure and surface characteristics of g-C₃N₄/CQDs, improving its photocatalytic performance [14,23,103]. The photocatalytic performance of the g-C₃N₄/CQDs based heterostructure has been enhanced by the use of CQDs to delay the rate of electron-hole pair recombination. As a result, CQDs are functionalized as spectral converters due to of exceptional multi-photon illumination property. The wavelength of PL emission is always smaller than the wavelength of practical excitation, according to the up-conversion PL properties [13,88]. The efficiency of CQDs as photocatalysts may also be improved by producing photons with a narrow wavelength by exciting charge carriers at higher wavelengths.

4.2. CQDs/g-C₃N₄ photocatalyst in dye removal

Photodegradation of dye in wastewater is a research emphasis globally with the aim of preserving the environment and understanding the sustainable advancements of human life. In this context, a metal free and novel g-C₃N₄/CQDs photocatalyst is offer a potential prospect to eliminate the dye from wastewater. For example, Zhang and coworkers [6] synthesized g-C₃N₄/CQDs photocatalyst by urea and citric acid through an environmentally friendly technique and applied to

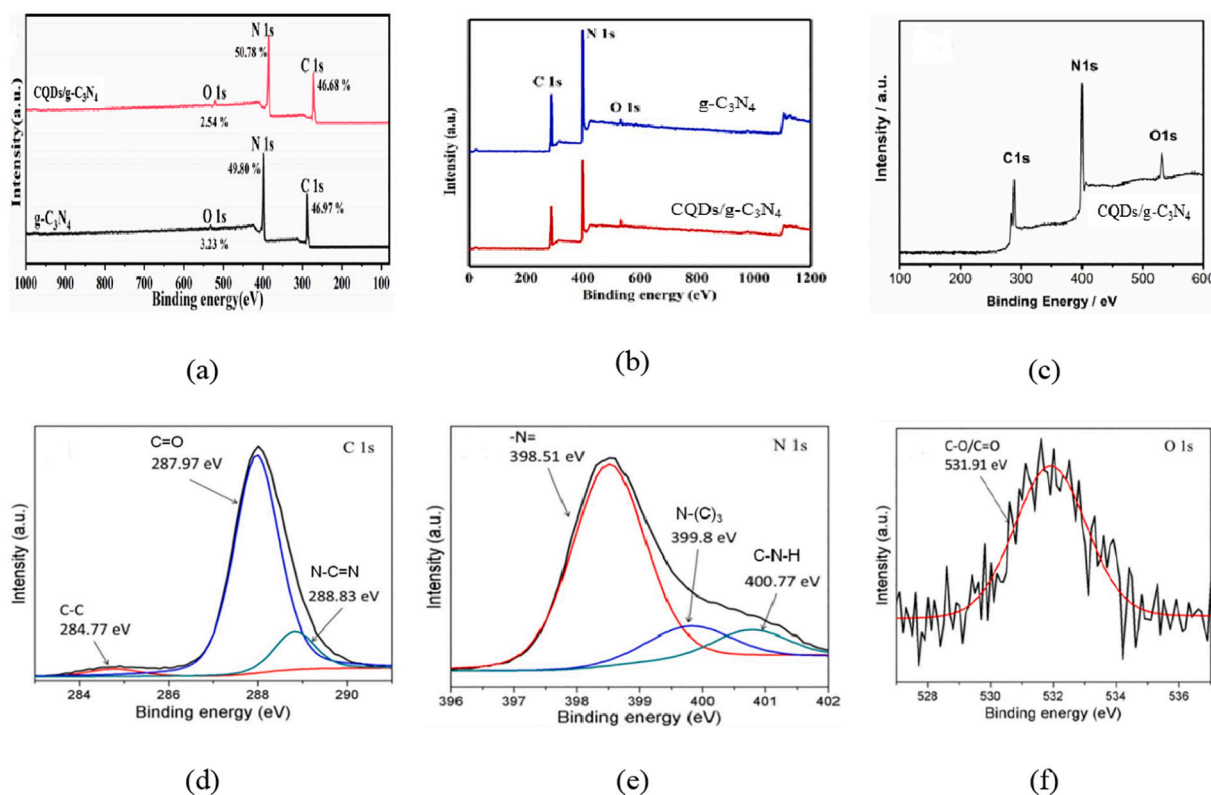


Fig. 14. XPS survey spectra of g-C₃N₄ and CQDs/g-C₃N₄ (a) [85], (b) [23], (c) [6]; (d) C1s of CQDs/g-C₃N₄, (e) N1s of CQDs/g-C₃N₄ and (f) O1s of CQDs/g-C₃N₄ [65].

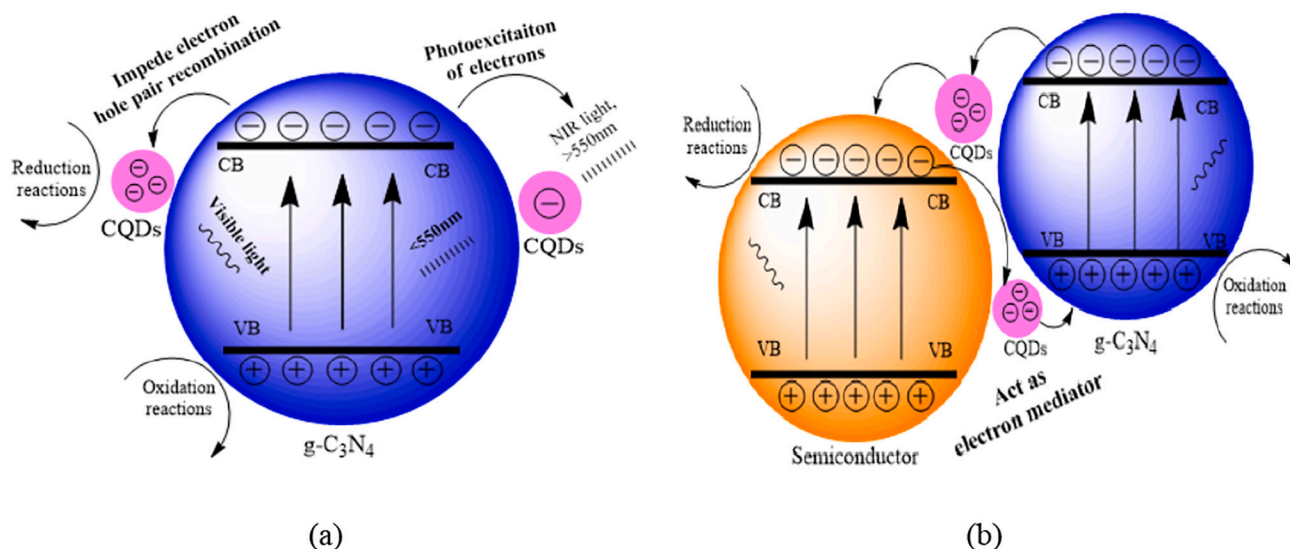


Fig. 15. Function of CQDs in pure g-C₃N₄ (a) and g-C₃N₄/CQDs photocatalyst (b) with improved photocatalytic performance.

degradation of synthetic dye. However, the latent mechanism of charge transfer by g-C₃N₄/CQDs nanocomposite for the improvement of photocatalytic performance in terms of dye removal is displays in Fig. 16a. The surface of CQDs/g-C₃N₄ is stimulated to produce photogenerated electrons and holes due to the effects of the inner electric arena. The photogenerated holes persisted on the g-C₃N₄ whereas the photogenerated electrons quickly moved to the CQDs and producing a spatial separation. The dye molecule was then effectively decomposed through oxidation using the h⁺ and converted to water, oxygen and others. In addition, while the O₂ radicals partially oxidized H₂O to create •OH active species, other radicals also reacted with the organic dye material adsorbed on the surface of g-C₃N₄. Since, the positive characteristics of CQDs/g-C₃N₄ in VB is higher than the energy level of water (H₂O) that's why photo-generated holes are unable to react with H₂O to convert the •OH radicals. Finally, existing holes are engulfed by the oxidation of dye molecules.

Likewise, Gong and peers [80] conducted the fabrication of CQDs/g-

C₃N₄ by using urea and citrus peel and successfully degraded the sunset yellow dye. They showed the pure individual g-C₃N₄ is unable to offer potential photocatalytic efficiency due to large energy gap and the quick recombination of carriers produced by photosynthesis. But interestingly, CQDs induced CQDs/g-C₃N₄ composite showed the excellent potentiality in photocatalytic dye degradation under visible light by narrowing the bandgap. According to their findings, O₂^{•-} was the most significant active radical for the photodegradation of sunset yellow dye, followed by O₂ and •OH [30,102]. The E_{VB} value of CQDs/g-C₃N₄ is lower than the conventional oxidation potential of OH/•OH and H₂O/•OH, which prevented the h⁺ on the VB from directly oxidizing OH or H₂O to generate •OH (Fig. 16b). In order to generate reactive species for CQDs/g-C₃N₄ composite during the photocatalytic removal of sunset yellow dye, the suggested reactions are as follows Eqs. (5) to (12).

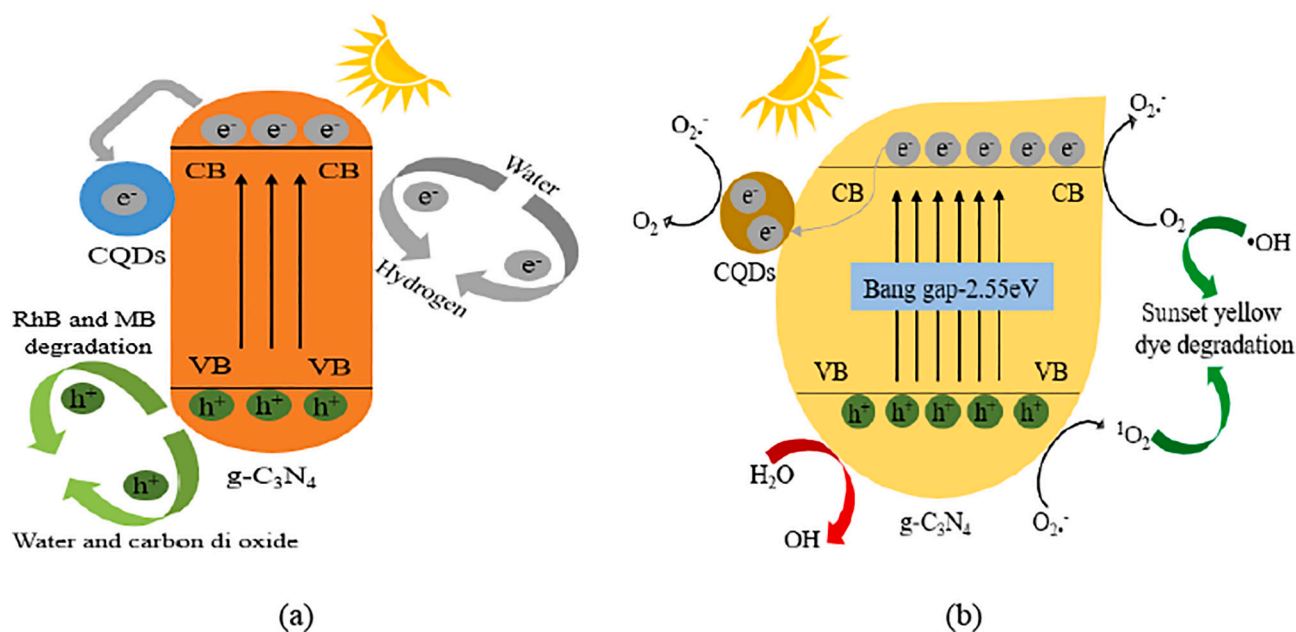
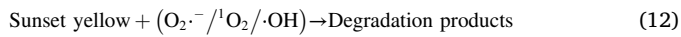
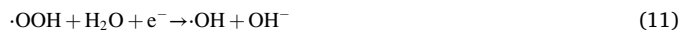
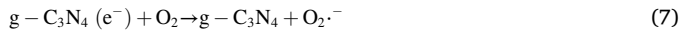
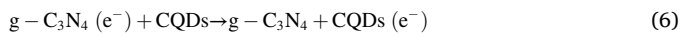


Fig. 16. Mechanism of successful dye (a) MB and RhB [6], (b) sunset yellow [80] degradation by using CQDs/g-C₃N₄ composite. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



Since, there are two processes for the inauguration and conversion of electrons and holes. Firstly, $g-C_3N_4$ is stimulated to produce

photogenerated electron-hole pairs in the presence of visible light. In order that the electrons are moving towards the CQDs and form $\cdot O_2^-$ radical by absorbing oxygen. Contaminants are then converted into tiny molecules by the potent oxidative activity of the $\cdot O_2^-$ radical. Additionally, contaminants can be directly oxidized by photogenerated h^+ that remains in the VB of $g-C_3N_4$. Secondly, the up-converted PL behaviour of the CQDs allows them to absorb light (>600 nm) and then transmute to shorter wavelengths (<460 nm). The photocatalytic activity can be further increased at shorter wavelengths by excitation of the $g-C_3N_4$ by forming electron-hole pairs [30]. In short, CQDs can boost their ability to capture visible light, which leads to an efficient breakdown of pollutants when exposed to visible light, in addition to improving the photogenerated electron-hole pair's separation. Finally, the above discussion demonstrated that CQDs/ $g-C_3N_4$ composite is highly potential than individual CQDs and $g-C_3N_4$ in photocatalytic dye

Table 2

Summary of recent published articles supported dye elimination by CQDs/ $g-C_3N_4$ photocatalyst.

Precursors	Synthesis method	Dye	Light Source	Duration	Removal rate	Reference
Citric acid + Urea	Hydrothermal approach	MB	300 W Xe lamp	20 min	100 %	[6]
Water hyacinth + Urea	Hydrothermal carbonization	RhB		110 min		
Glucose + Melamine	Thermal polymerization	DCPL	Direct sunlight	120 min	94.0 %	[65]
		MB	NIR light irradiation	120 min	86 %	[48]
		RhB		120 min	38 %	
Ethylenediamine-tetraacetic acid + Dicyandiamide	Two-step hydrothermal method	MO	300 W Xe lamp	240 min	91.1 %	[102]
Citric acid + Melamine	Low temperate hydrothermal	RhB	Visible light irradiation	210 min	95.2 %	[30]
Thiourea + Lemon juice	Induction method	RhB	Visible light	120 min	90.9 %	[46]
Melamine + Cyanuric acid	Calcination method	MB	300 W Xe lamp	240 min	100 %	[100]
Citric acid + Urea	Impregnation method	RhB	500 W Xe lamp	120 min	100 %	[23]
Citric acid + Melamine	One-pot hydrothermal treatment	Bisphenol A	Near-visible LED	300 min	90.4 %	[64]
Melamine + HNO ₃ /H ₂ SO ₄	Spot heating	Bisphenol A	450 to 850 nm	120 min	80 %	[104]
Urea + Arabian dates	Hydrothermal carbonization	DCPL	Sunlight irradiation	90 min	100 %	[105]
Citric acid + Urea	Simple calcination	RhB	250 W Xe lamp	20 min	33 %	[94]
Citric acid + Urea	Simple calcination	RhB	250 W Xe lamp	40 min	68 %	[106]
Citric acid + Cyanuric acid + Melamine	Solvothermal method	MO	300 W Xe lamp	100 min	93.6 %	[107]
Melamine + Citric acid	Hydrothermal	RhB	300 W Xe lamp	100 min	98 %	[108]
Citric acid + Urea	Hydrothermal treatment	MB	LED light irradiation	180 min	54.60 %	[44]
Phenylenediamine + Melamine	Hydrothermal approach	MB	Visible light irradiation	120 min	97.9 %	[42]
Glucose + Melamine	Simple deposition	RhB	300 W Xe lamp	210 min	100 %	[109]
Orange peel + Melamine	Simple impregnation method	MB	40 W LED bulb	75 min	98 %	[110]
		RhB			90 %	
Melamine + Citric acid	Facile decoration	RhB	50 W LED lamp	270 min	100 %	[111]
Melamine + Citric acid + Urea	In-situ calcination and hydrothermal	RhB	800 W Xe lamp	45 min	95 %	[112]
Citrus peel + urea	Hydrothermal method	Sunset yellow	300 W Xenon lamp	60 min	96.3 %	[80]
Glucose + melamine	Alkali-assisted ultrasonic	Dioxane	LED lamp	360 min	77.9 %	[93]
Citric acid + Urea	hydrothermal approach	MB	LED light irradiation	180 min	54.60 %	[44]
Dicyandiamide + Soot of alcohol burner	Calcination	RhB	3 W LED lamp	k = 0.013 mol L ⁻¹ min ⁻¹		[113]
Dimethyl sulfoxide + Melamine + Thiourea	One-pot in situ solution polymerization	MO	Visible light	80 min	94.3 %	[114]
Melamine + Glucose	Simple pyrolysis	RhB	300 W Xe lamp	40 min	100 %	
Citric acid + Melamine	Ultrasonic treatment and calcination	RhB	300 W Xe lamp	30 min	97 %	[115]
Urea + Graphite rods	Hydrothermal method	RhB	350 W LED	30 min	85 %	[116]
Graphite rods + Melamine	Simple ultrasonic dispersion	RhB	300 W xenon lamp	90 min	99.6 %	[117]
		MB	500 W Xe lamp	180 min	97.21 %	[118]
Ethylenediamine + Citric acid	Solvothermal	MB	310 W lamp	60 min	71 %	[119]
		RhB			53.8 %	
		MO			36 %	
Citric acid + Melamine	Calcination and microwave oriented self-method	RhB	LED lamp (50 W)	60 min	100 %	[120]
Melamine + Cyanuric acid + Citric acid	One-step thermal condensation	RhB	40 W LED lamp	2.04 h ⁻¹	10 mg ⁻¹	[121]
Citric acid + Melamine	Facile one-pot co-sintering method	RhB	Nature sunlight	60 min	86.1 % ~ 97.2 %	[122]
Graphite rods + Urea	Electrostatic adsorption technique	MB	Visible light irradiation	90 min	>95 %	[123]
Pyrene+ Trinitro pyrene + HNO ₃ + Cyanamide	Electrostatic interactions method	RhB	300 W Xe lamp	120 min	97 %	[124]

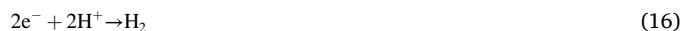
wastewater treatment whereas some examples are given in Table 2.

4.3. CQDs/g-C₃N₄ photocatalyst in hydrogen evolution

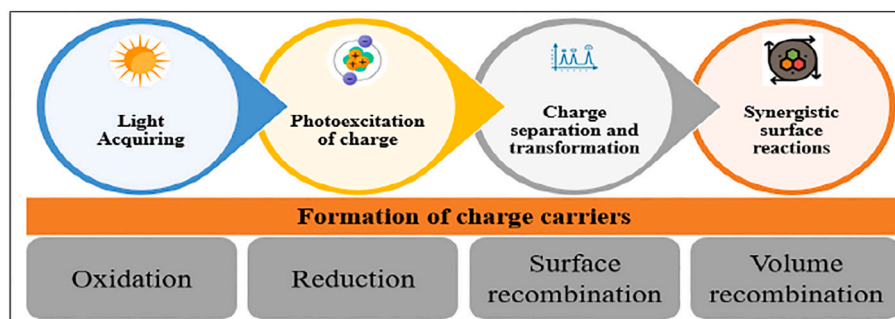
A promising alternative energy source for the future without depending on fossil fuel is the production of hydrogen from water by applying photocatalyst in the presence of solar energy. Hydrogen can be produced in two different ways: photocatalytic water splitting and photocatalytic organic improvement. In order to produce hydrogen, the photocatalytic process typically needs a photocatalyst, reactant, light energy, and photoreactor. Water can occasionally be used as a solitary reactant or in combination with sacrificial agents. Light is required for this, whether visible or UV light, but visible light ensured the absorption range for photocatalysts [23]. It would be necessary for catalyst, reactant, and light to work together effectively for proficient hydrogen generation.

Over the past few years, a significant number of g-C₃N₄/CQDs based photocatalysts with improved water splitting performance have been developed [6,101,104]. Under light illumination, the g-C₃N₄/CQDs photocatalyst is used for photocatalytic water splitting to produce oxygen and hydrogen. Since, the adsorbed H₂O molecules can become gaseous oxygen and hydrogen when the e⁻/h⁺ pairs move to the surface reaction sites on the g-C₃N₄ or CQDs without recombination. In order to separate the filled VB from the empty CB, or vice versa, a semiconductor-based photocatalyst must be illuminated with light that has energy greater than or equal to the band energy of the catalyst. This separation creates the conditions for an electron from the filled valence band to enter the CB legally, which results in the splitting of the h⁺/e⁻ pair. While electrons are used in the reduction process, photogenerated holes are used in the oxidation process [13,48]. Fig. 17a describes the significant phases of photocatalysis and Fig. 17b outlines the stepwise mechanism of photo electrocatalytic water splitting for hydrogen evolution. The symmetry between the catalyst and the carrier species is shown in Eqs. (13) and (14). Eqs. (15) and (16) represents the conversion of water (H₂O) into protons (H⁺) and hydrogen evolution,

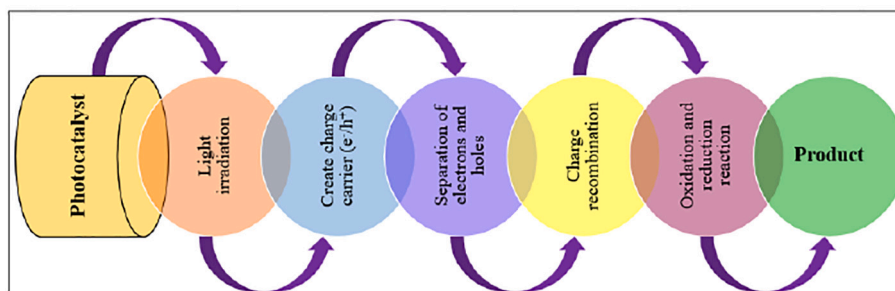
respectively. The primary duty of the photocatalysis system is to prevent photocatalytic recombination of charge carriers (e⁻/h⁺) in the water splitting process. As seen in Eq. (14), an ineffective heat loss occurs on a mixture of charge carriers. As a result, it is less profitable to produce hydrogen in an aqueous environment.



Photocatalytic hydrogen evolution mechanism via CQDs/g-C₃N₄ photocatalyst is offered and elucidated by Li and peers [85] as shows in Fig. 18a. The bandgap of CQDs/g-C₃N₄ photocatalyst in between VB and CB is condensed when CB is easily activated by the electrons in CQDs donor alignment in the presence of visible light. If visible light captured extensively during the hydrogen evolution reaction, then the photo-generated charges are increasing. However, the recombination of photo-generated charges is successfully reduced by the micro-regional heterostructures of CQDs and g-C₃N₄, which also encourage the transfer of photogenerated charges across tri-s-triazine area. As a result, additional photogenerated electrons transfer to the catalyst and the supporting platinum's surface to take part in the reaction that produces hydrogen. On the other hand, as a sacrifice reagent, triethanolamine (TEOA) can devour holes in the valence band and improve the separation of photo-generated carriers. Consequently, the H₂ evolution rates are noticeably improved by all levels of CQDs/g-C₃N₄ photocatalyst as shows in Fig. 18a1. Their result shows that CQDs/g-C₃N₄ 10 wt% has a H₂ evolution rate of 2321 μmol/g/h, which is 7.4 times advanced than pure g-C₃N₄. Simultaneously, Wang et al. [63] also found that CQDs/g-C₃N₄ 10 wt% photocatalyst offers higher hydrogen evolution rate of 2.2 μmol/h which is approximately 4.4 times that of pure g-C₃N₄ (Fig. 18b1). According to the hydrogen evolution mechanism, electron transmission

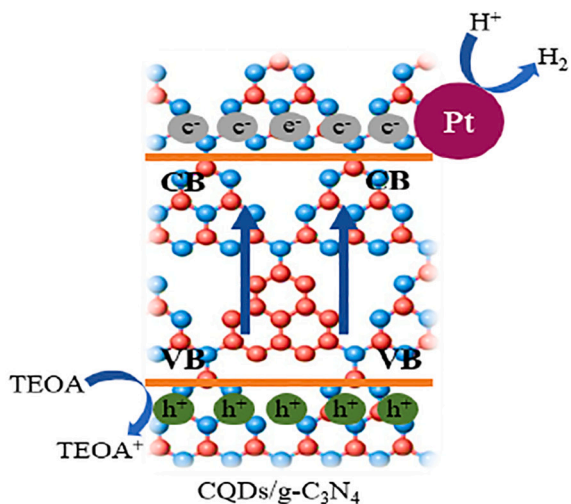


(a)

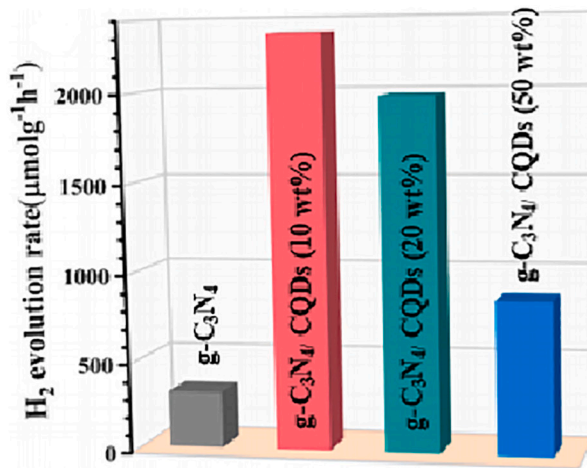


(b)

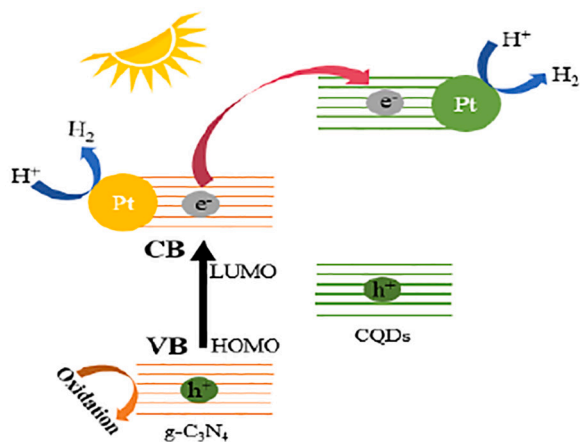
Fig. 17. Phases of (a) photocatalysis process and (b) photo electrocatalytic water splitting mechanism for hydrogen evolution.



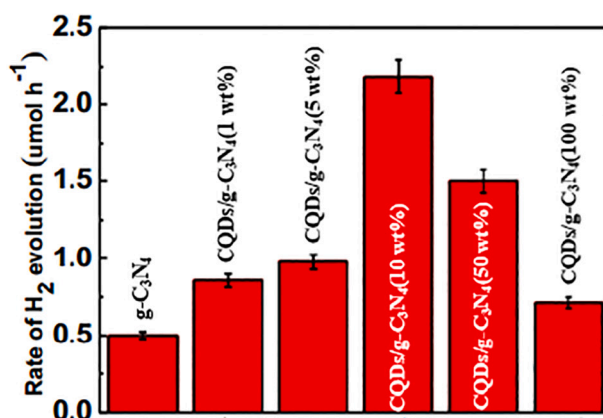
(a)



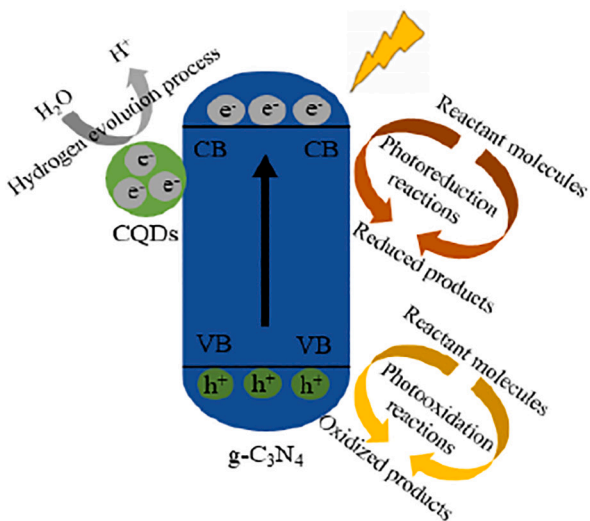
(a1)



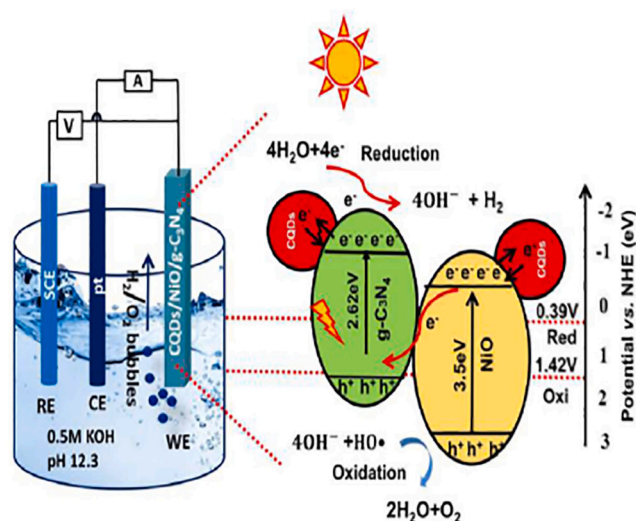
(b)



(b1)



(c)



(d)

Fig. 18. Photocatalytic hydrogen evolution mechanism and performance through CQDs/g-C₃N₄ photocatalyst (a) and (a1) [85]; (b) and (b1) [63]; (c) [6], (d) photo electrochemical water splitting on CQDs modified NiO/g-C₃N₄ photo electrode [40].

from the lowest unoccupied molecular orbital (LUMO) of g-C₃N₄ towards the LUMO of CQDs is associated with the performance of CQDs/g-C₃N₄ photocatalyst. The presence of Pt cocatalyst with CQDs/g-C₃N₄ shows the potentiality to enhance photogenerated electrons by its outstanding energetic spots and photocatalytic attribute, as a result H₂O easily diverted to hydrogen (Fig. 18b). However, a research by Zhang and team [6] applied urea and citric acid as a precursor to fabricate CQDs/g-C₃N₄ photocatalyst similarly potential to produce hydrogen effectively. The maximum hydrogen evolution rate was 664 μmol/h/g that is offered by CQDs/g-C₃N₄ 0.98 wt% photocatalyst. The photocatalytic hydrogen evolution mechanism demonstrates that the photo-generated electrons of g-C₃N₄ simply moved towards CQDs due to the effect of internally charged. Since, the photogenerated holes persisted on the g-C₃N₄ and creates a spatial gap. Subsequently, the photo-generated holes are then occupied by H⁺ to produce hydrogen (Fig. 18c).

According to Noor et al. [40] urea, thiourea and citric acid induced CQDs/NiO/g-C₃N₄ photocatalyst offers outstanding performance in hydrogen evolution as well as dye removal and oxygen evolution. In the application they applied photo electrochemical water splitting that shows a reduction of band-gap values from 2.62 to 1.8 eV in UV Visible spectra of nanocomposites confirmed an improvement towards red shift. Where electrochemical impedance spectroscopy (EIS) performance was evaluated in 0.5 M KOH electrolyte maintain at neutral pH ~12.3 under λ > 420 nm irradiation. On the exposure of incident light on the photoanode, electrons from VB migrate to CB of NiO/g-C₃N₄ generate the photoelectrons which further move to from CB of NiO to VB of g-C₃N₄ and to surface of CQDs for space charge separation under z-scheme mechanism as explained in Fig. 18d. Similarly Pan and team [125] and Wang and co-researcher [126] applied photocatalytic water splitting for the production of hydrogen by using g-C₃N₄/TiO₂/CQDs composite photocatalyst.

On the other hand, Vogt and team [127] applied Steam and Dry Methane Reforming (S/DMR) process for the production of hydrogen (Steam: CH₄ + H₂O → CO + 3H₂ and Dry: CH₄ + CO₂ → 2CO + 2H₂). In

addition, Nishimura and his research group [128] investigated the effect of pressure of sweep gas on a biogas dry reforming to get H₂. The highest concentration of H₂ is obtained in the case of the molar ratio of CH₄:CO₂ = 1:1. However, several challenges such as catalyst deactivation and the low H₂/CO ratio of the syngas produced are limits the performance of hydrogen evolution by applying DMR method [129]. Table 3 shows an overview of recent studies on the performance of photocatalytic hydrogen evolution by applying CQDs/g-C₃N₄ photocatalyst. In summary, the synthesized CQDs/g-C₃N₄ photocatalyst had an advanced photocatalytic efficiency for photocatalytic dye wastewater treatment and energy recovery by generating hydrogen.

5. SWOT analysis

The Strengths, Weaknesses, Opportunities, and Threats (SWOT) analysis for the use of CQDs/g-C₃N₄ photocatalyst in dye removal and hydrogen production has been carried out by considering social, economic and environmental sustainability perceptions as shown in the Fig. 19.

5.1. Strengths (S)

Mostly, carbon and nitrogen induced raw materials are applied for the synthesis of CQDs/g-C₃N₄ photocatalyst, which acts as a feasible, green and sustainable precursors [6,121,138]. Numerous studies have looked at the quick and efficient synthesis of CQDs/g-C₃N₄ using cutting-edge techniques during the last few decades. Additionally, the procedure produces a good yield, emits fewer emissions, and doesn't need any harmful solvents. It is also industry-friendly due to its simple synthesis and fast production, which opens up new possibilities for its use in a variety of applications, for example water treatment, gas capturing, pollutants degradation, biosensing, biomedical, bioimaging, CO₂ mitigation process and hydrogen production [6,44,46,111,138]. During the function of CQDs/g-C₃N₄ photocatalyst in dye removal and hydrogen evolution, electron donor-acceptor properties of CQDs is

Table 3
Summary of recent published articles supported hydrogen evolution by CQDs/g-C₃N₄ photocatalyst.

Precursor	Synthesis Method	Light Source	H ₂ Evolution Rate	Reference
Citric acid + Urea	Hydrothermal approach	300 W Xe lamp	1291 μmol/g/h	[6]
Melamine + Cyanuric acid	Calcination method	300 W Xe lamp	24,760 μmol/g/h	[100]
Citric acid + Urea	Hydrothermal method	300 W Xe lamp	2321 μmol/g/h	[85]
Citric acid + Urea	Impregnation method	300 W Xe lamp	626.93 μmol/h/g	[23]
Citric acid + Urea + Melamine	Solvothermal method	Visible light irradiation	987.4 μmol/g/h	[31]
L-ascorbic acid + Melamine	Hydrothermal approach	UV light irradiation	2.2 μmol/h	[63]
Urea + Chitosan + Acetic acid	Hydrothermal treatment	Visible light irradiation	295.9 μmol/g/h	[101]
Urea + Citric acid	Thermal polymerization	300 W xenon lamp	3538.3 μmol/g/h	[45]
Melamine + HNO ₃ /H ₂ SO ₄	Spot heating	450–850 nm	152 μmol/g/h	[104]
Urea + Glucose	Thermal polymerization	Visible light	2.34 mmol/g/h	[130]
Citric acid + Melamine	Free-template chemical method	40 W LED lamp	382 μmol/g/h	[131]
Urea + Glucose	Alkali-auxiliary sonication method	300 W Xe lamp	220.77 μmol/g/h	[132]
Sucrose + Melamine	Hydrothermal method	300 W Xe lamp	4.134 μmol/g/h	[125]
Flower bee pollens + Dicyandiamide	Hydrothermal	300 W Xenon lamp	88.1 μmol/h	[133]
Melamine + L-Ascorbic acid	Hydrothermal	3 W Four low power LEDs	183 μmol/h	[63]
Citric acid + Urea	Homogeneous thermal pyrolysis	300 W Xenon lamp	27.6 μmol/h	[134]
Dicyandiamide + Aminohexanoic acid	Solvothermal	300 W Xe arc lamp	116.1183 μmol/h	[135]
Urea + Graphite rod	Solvothermal and physical co-precipitation methods	Visible-light irradiation	9.4 μmol/g/h	[136]
Candle soot + Urea + Melamine	Impregnation precipitation methods	350 W Xe arc lamp	10.9 μmol/h	[137]
Melamine + Urea	Hydrothermal treatment	300 W Xe-lamp	70.05 10.9 μmol/h	[72]
Dicyandiamide	Calcination	3 W LED lamp	218 μmol/g/h	[113]
Urea + Graphite rods	Hydrothermal method	300 W xenon lamp	2.169 μmol/g/h	[117]
Dicyandiamide + Citric acid	One-step thermal polymerization	300 W Xenon arc lamp	3319.3 μmol/g/h	[41]
Citric acid + Urea	Colloidal template crystal method	250 W xenon lamp	8.12 mmol/g/h	[138]
Urea + Dopamine hydrochloride	Impregnated calcination	300 W Xe lamp	712.90 μmol/g/h	[139]
Urea + Thiourea + citric acid	Wet chemical method	λ > 420 nm irradiation	~ – 19 mAcM ⁻² at potential 0.39 V	[40]
Melamine + sucrose + tetrabutyl titanate	Thermal polycondensation	Xenon lamp (300 W)	6.497 μmol/g/h	[125]
Citric acid + dicyandiamide	Thermo-polymerization	Xenon arc lamp (300 W)	2.34 μmol/g/h	[126]



Fig. 19. SWOT analysis of using CQDs/g-C₃N₄ photocatalyst in energy and environmental applications.

possess outstanding performance under light illumination. CQDs have an enormously lucrative role in the charge transfer and separation process for the growth of CQDs/g-C₃N₄ photocatalyst [61]. On the other hands, due to the exhaustion of fossil fuels, water bodies are becoming more and more polluted, rendering them unfit for human consumption and causing an increase in the energy crisis. Therefore, CQDs/g-C₃N₄ photocatalyst acts as a sustainable and environmentally beneficial technology that offers decent financial aids with the favourable synthesis of hydrogen energy and fresh water. More importantly, the produced green hydrogen will be ensured the future fuel demands and the fresh water can be reuse in domestic, commercial and irrigation purposes.

5.2. Weakness (W)

Unique and advance synthesis method is one of the main drawbacks of CQDs/g-C₃N₄ photocatalyst. Appropriate synthesis process has the potentiality to prepare pure and quality products as well as highly efficient and stable photocatalyst [140]. Synthesis process can identify various morphologies, but it is unknown how they relate to one another specifically. Moreover, there is inadequate relationship between catalytic activity and morphology of CQDs/g-C₃N₄ photocatalyst. Consequently, the dye removal performance still not hundred percent successful and hydrogen evolution rate also not satisfactory. However, the presence of inappropriate bang gap in the mechanism also enhances the performance of the photocatalyst. There is also an influence of time, catalyst dose, reaction, temperature, pH and light irradiation in the performance of CQDs/g-C₃N₄ photocatalyst. Environmental and social impact assessment need to carryout prior to apply in large scale industry level. Additionally, there are currently no restrictions in national and global level that apply to carbon and nitrogen constructed nanomaterials.

5.3. Opportunities (O)

CQDs/g-C₃N₄ is a promising photocatalyst for several sectors such as

energy, environment, medical, harmful gas control, contaminants and heavy metals control [141]. One of the most important advantages of CQDs/g-C₃N₄ is workable in visible light irradiation. Which typically reduce the cost and makes the photocatalyst environmentally friendly by using nature-based power. CQDs/g-C₃N₄ offers outstanding photostability and recyclability for the degradation of pollutants. CQDs functions as a photosensitizer, causing g-C₃N₄ to absorb more visible light as a result of its up-converted photoluminescence feature and inducing g-C₃N₄ to form more electron-hole pairs and reactive •O₂ and •OH radicals. CQDs serve as electron-sinks in the g-C₃N₄ VB, where they can capture photoexcited electrons and keep them from recombining with photogenerated holes [44]. In the near future, it can be further investigated for semiconductor photocatalysis to purify water, nano-hybrid light-driven catalyst to remove heavy metal contaminants, and in the animal and human health sectors. Semiconductor photocatalysis is an auspicious technology due to its ability to directly transform solar energy into chemical energy and for being ecologically friendly.

5.4. Threats (T)

Although employing CQDs/g-C₃N₄ to generate hydrogen is a favourable strategy, it is still in the early stages of development. It's important to fully comprehend the effects of operating variables like pH, temperature, wetness and calcination prior to large scale production. On the other hand, there is a possibility to generate harmful byproducts during the application of CQDs/g-C₃N₄ in pollutants removal. Although, the use of strong acid to synthesize CQDs/g-C₃N₄ photocatalyst offers an excellent outcome in photocatalytic applications. So, toxicological effects of dye removal and hydrogen evolution process should be examined to ensure human safety and environmental sustainability. In this regard, a life cycle assessment (LCA) tools can be applied for suitability checking of a system or process and to take the proper initiatives about the use of any technology and potential to identify the risks.

6. Conclusion and future perspective

Here, this review offers a comprehensive review and detail discussion of green and non-metal g-C₃N₄/CQDs photocatalyst for photocatalysis. It also presents an assessment of current developments of CQDs/g-C₃N₄ photocatalyst in conjunction with the synthesis procedures, photocatalytic enrich mechanisms, modifications, dopant incorporation and photocatalytic applications in energy and environmental area. It was demonstrated that the optical and electrical features of g-C₃N₄ were smartly improved by the incorporation of CQDs, resulting in a notable increase in quantum efficiency for the photocatalytic properties. More importantly, CQDs modified g-C₃N₄ not only overcome the shortfalls of g-C₃N₄ but also exhibits the potentiality to prevent recombining of charge carriers, encouraged photogenerated e⁻/h⁺ separation and visible light absorption capacity by maintaining photostability which enhanced photocatalytic activity, contribution and overall performance. A stunning manifestation of CQDs/g-C₃N₄ in hydrogen evolution and degradation of dye molecules makes the composite an excellent safeguard against environmental degradation and energy crisis. In addition, CQDs/g-C₃N₄ composite is widely applied in CO₂ reduction, elimination of bacteria, biosensing, organic pollutants degradation, heavy metal detection and removal that also contributes for sustainable environment.

Although, there has undoubtedly been successful and significant achievements of CQDs/g-C₃N₄ photocatalyst, but the progress condition is still at initial phases. However, it is confessed that there are many silent challenges that must be overcome in order to better meet the demands of practical application of resolving the energy and environmental issues. As a result, the features that need for more investigation in order to effectively apply CQDs/g-C₃N₄ photocatalyst is suggested. Some crucial hushed points are explained as follow.

- In order to synthesize CQDs/g-C₃N₄, it is critical to produce simple, efficient, cheap, and environmentally friendly techniques. The suitable CQDs and g-C₃N₄ should have low costs, high fluorescence quantum yields, stable, toxic-free, and narrow particle size distributions from nature-based carbon rich biomass source. It is crucial to develop effective techniques for loading CQDs onto g-C₃N₄ photocatalysts by choosing appropriate precursors and enhancing their recyclability. In addition, the use of strong acid or base should be prohibited during the synthesis process to avoid environmental and human insecurity.
- The fabrication process of CQDs/g-C₃N₄ mostly depends on thermal carbonization in conjunction of high temperature resulting inconsistency and unregulated outcomes that has the potentiality to make barrier in photocatalytic performance. Even though the self-assembled fabricated method of CQDs/g-C₃N₄ is reasonably straightforward that possess a sloppy coupling interface that hampers operative charge carriers. The exploration of more sophisticated synthesis methods will be more demandable in current era with focusing distinguished development of photocatalytic ability.
- Laterally, the frequent shaky connection in the interface of CQDs/g-C₃N₄ typically prevents the energy level adjustment and fluent charge transfer, means that there is still room for improvement in the synthesis techniques. To maximize the efficiency of the photocatalyst, some forthright green synthesis approaches are desperately needed.
- In order to produce g-C₃N₄ with higher surface areas, several morphologies can be investigated, including nanotubes, nanofibers, nanosheets, nanorods, nanospheres structures and so on. Furthermore, it is important to investigate the initial loading volume and size of CQDs and element doping that also can be contribute in the improvements of photocatalytic performance of CQDs/g-C₃N₄.
- To date, advance prospect of CQDs/g-C₃N₄ is typically enhanced, limited and depends on the unsatisfactory hypothetical and practical exercise. The potentiality of CQDs/g-C₃N₄ can be continued in

conjunction with modern intelligence materials-oriented chemistry laboratory research discoveries.

- The relationship between charge carrier movement kinetics and structural characteristics of CQDs/g-C₃N₄ photocatalysts should be precisely demonstrated and verified in future research. To develop highly effective visible-light induced photocatalysts, organised research is required for the incorporation of various semiconductors and ingredients with CQDs/g-C₃N₄.
- It is still challenging and crucial to establish CQDs/g-C₃N₄ fabrication methods and machineries for industrial application as CQDs/g-C₃N₄ photocatalyst research is currently only at the laboratory level. The development of a multifunctional synergistic platform and the suitable integration of the related photocatalytic application systems will become popular subjects.
- Since, CQDs/g-C₃N₄ novel photocatalyst is mostly applied and concentrated in the degradation of contaminants and hydrogen generation. The application of CQDs/g-C₃N₄ photocatalyst in biosensing, biomedical, bacterial disinfection, oxygen evolution, food security and energy storage will be popular research arena. Additionally, CQDs/g-C₃N₄ photocatalyst can be highly potential for the expansion of solar cell industries.
- Subsequently, it is significantly important to prevent hazardous byproduct and gas (CO₂) during the photodegradation of contaminants and energy generation to develop an innovative research theme which will be completely green.

According to the above discussions, it is projected that the affordable, eco-friendly.

CQDs/g-C₃N₄ photocatalyst will play a significant role in a variety of photocatalytic processes. It is anticipated that the suggested outline of the development path in CQDs/g-C₃N₄ will provide scholars with complete information that will enable them to understand the main ideas and potential for the future developments of any composite photocatalysts. This review is intended to give readers a comprehensive understanding of the application and continuous growth of CQDs/g-C₃N₄ in energy and environmental sectors.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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