

Review

g-C₃N₄-Based Nanomaterials for Visible Light-Driven Photocatalysis

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Abstract: Graphitic carbon nitride (g-C₃N₄) is a promising material for photocatalytic applications such as solar fuels production through CO₂ reduction and water splitting, and environmental remediation through the degradation of organic pollutants. This promise reflects the advantageous photophysical properties of g-C₃N₄ nanostructures, notably high surface area, quantum efficiency, interfacial charge separation and transport, and ease of modification through either composite formation or the incorporation of desirable surface functionalities. Here, we review recent progress in the synthesis and photocatalytic applications of diverse g-C₃N₄ nanostructured materials, and highlight the physical basis underpinning their performance for each application. Potential new architectures, such as hierarchical or composite g-C₃N₄ nanostructures, that may offer further performance enhancements in solar energy harvesting and conversion are also outlined.

Keywords: g-C₃N₄; photocatalysis; nanomaterials; CO₂ reduction; H₂ evolution; semiconductor; environmental remediation

1. Introduction

1.1. Background

Future energy production, storage and security, and combating anthropogenic environmental pollution, represent key global challenges for both developed and emerging nations [1,2]. Sunlight, an essentially limitless source of clean energy, has the potential to address both these challenges [3,4], and its utilization to this end entered mainstream science following breakthroughs in semiconductor light harvesting for photocatalysis by Honda and Fujishima in the 1970s [5–7]. This discovery led to extensive research into titania semiconductor photocatalysts, principally for water splitting and the degradation of aqueous or airborne organic pollutants under UV light irradiation [8–13]. However, efficient harnessing of visible light (the major component of solar radiation that reaches the Earth's surface) by photocatalysts to drive chemical transformations remains problematic [14–16] due to identifying suitable materials that possess narrow band gaps, high quantum yields, efficient charge carrier transport, and low rates of charge carrier recombination, and good thermo-, photo-, and chemical stability. The development of such low cost photocatalysts from earth abundant, and ideally non-toxic elements for visible light harvesting would unlock opportunities for their large-scale application to supplement existing renewable energy networks and pollution control systems.

1.2. Semiconductor Photocatalysis

Semiconductor photocatalysis refers to the acceleration of chemical transformations (most commonly oxidations and reductions) brought about through the activation of a catalyst, comprising a semiconductor either alone or in combination with metal/organic/organometallic promoters, through

light absorption, with subsequent charge and/or energy transfer to adsorbed species. Note that the direct activation of reactants and intermediates through light absorption is the realm of photochemistry; in establishing whether a transformation is truly photocatalytic it is therefore crucial to establish that photons are absorbed by the catalyst rather than adsorbates [17,18]. In the photocatalytic production of so-called ‘solar fuels’, photoexcited charge carriers drive the conversion of water and CO₂ into H₂, CO, CH₄, CH₃OH and related oxygenates and hydrocarbons [19–21]. Such processes parallel those in nature wherein sunlight absorbed by chlorophyll in plants promotes starch and oxygen production from carbon dioxide and water, and are hence termed artificial photosynthesis (Figure 1). Photoexcited charge carriers can also either induce the total oxidation (mineralization) of organic pollutants such as those encountered in aquatic environments, either directly, or through the creation of potent oxidants such as hydroxyl radicals [22].

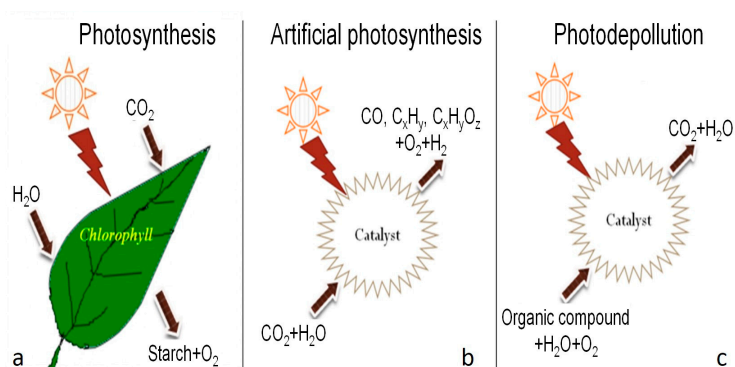


Figure 1. (a) Natural, and (b) artificial photosynthesis through water splitting and CO₂ reduction, and (c) photodegradation of aqueous organic pollutants.

1.3. Photocatalytic Mechanisms

Semiconductor photocatalysis is initiated by exciton formation following photon absorption and the excitation of electrons from the valence band into the conduction band (Step I). The resulting electron–hole pairs may recombine in either the bulk of the semiconductor, or at the surface, with the associated energy released through either fluorescence or thermal excitation of the lattice (Step II); recombination is the primary process that limits photocatalyst efficiency after photon capture. Electrons (and holes) that migrate to the surface of the semiconductor and do not undergo rapid recombination may participate in various oxidation and reduction reactions with adsorbates such as water, oxygen, and other organic or inorganic species (Steps III and IV) [9,10,23,24]. These steps are summarized below and illustrated in Figure 2:

- Step I Light absorption $SC + h\nu \rightarrow SC \times (e_{CB}^- + h_{VB}^+)$.
 Step II Recombination $e_{CB}^- + h_{VB}^+ \rightarrow h\nu + \text{heat}$.
 Step III Reduction $\text{Adsorbate} + e_{CB}^- \rightarrow \text{Adsorbate}^-$.
 Step IV Oxidation $\text{Adsorbate} + h_{VB}^+ \rightarrow \text{Adsorbate}^+$.

Oxidation and reduction reactions are fundamental to photocatalytic environmental remediation and solar fuel production, and are ultimately limited by the reduction potential of photoexcited electrons in the conduction band and oxidation potential of photogenerated holes in the valence band. The redox potential, band energies and gap of a semiconductor therefore largely determine the likelihood and rate of charge transfer, and hence are key design parameters for photocatalysts [12,25]. Although the underlying physics of space charge carriers and surface-electronic structure of photocatalysts varies between materials and applications, in essence, semiconductor photocatalysis represents interfacial reactions between electrons and holes generated through band gap excitation.

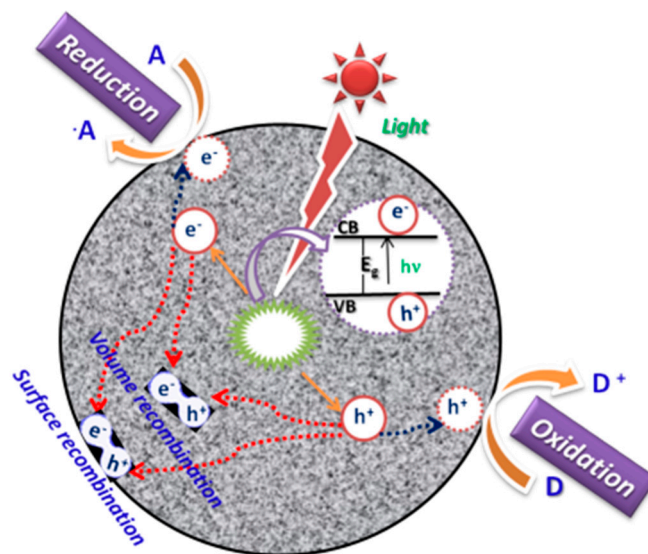


Figure 2. Principal photophysical processes for a semiconductor (SC) under light irradiation.

2. Photocatalytic Materials

The discovery of photocatalytic water splitting over titania electrodes under UV irradiation [5] has led to intensive research into explored H₂ production through this approach. Similarly, the first report on the photocatalytic oxidation of cyanide ions over TiO₂ powder [26] prompted a rapid expansion in environmental purification research and technologies, particularly for aqueous environments. In both cases, recent research has focused on identifying and developing alternative semiconductors to titania, offering superior performance under solar (rather than UV irradiation) [25]. Numerous semiconductors, including ZnO [27], Fe₂O₃ [28], WO₃ [29], SrTiO₃ [30], NaTaO₃ [31], CdS [32], Ag₃PO₄ [29], BiPO₄ [33], and g-C₃N₄ [34] are known photocatalysts, with their application dependent on their band gap (Figure 3). Despite a large body of literature, the practical utilization of such photocatalysts for solar fuels production or the degradation of organic pollutants remains a huge challenge due to poor visible light harvesting or efficient conversion of light energy to achieve chemical transformations [13,16,35].

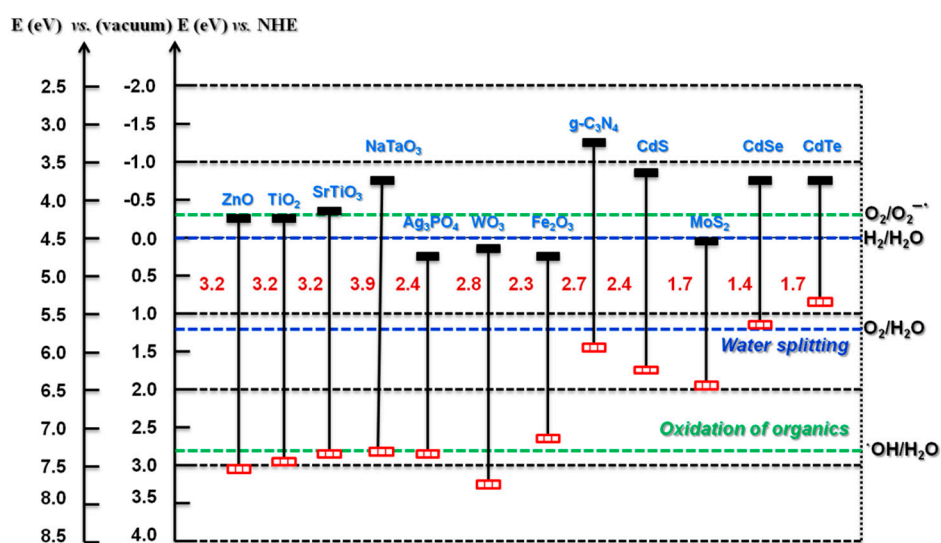


Figure 3. Band gap energy and band edge energies of different semiconductors.

3. Graphitic Carbon Nitride (g-C₃N₄)

Solar energy output reaching the Earth's surface is dominated by three regions (Figure 4) of the electromagnetic spectrum, UV (~5%), visible (~45%), and IR (~50%) [36]; visible light photocatalysis therefore offers the best opportunity to obtain maximum solar energy. However, most photocatalysts possess relatively wide band gaps, such as TiO₂ (3.0–3.2 eV) and are hence primarily active under UV irradiation (<385 nm) [8]. The quest for high performance visible light counterparts is reflected in the rapid growth of associated scientific papers and patents [8,10,15] for water splitting, CO₂ reduction and pollutants degradation [35]. Graphitic carbon nitride (g-C₃N₄) is a promising metal-free, polymeric semiconductor (Figure 5a) with a narrow band gap suited to visible light absorption (Figure 5b) [34], and amenable to large-scale synthesis. g-C₃N₄ may also be readily doped or chemically functionalized, permitting tuning of its photophysical properties, and in contrast to many other organic semiconductors, graphitic carbon nitride also exhibits high thermal and chemical stability to oxidation, even at temperatures of 500 °C. There is an extensive literature describing the synthesis of g-C₃N₄ and its derivatives for various applications [37–43]. This Review focuses on applications in photocatalytic environmental remediation and solar fuel generation, with an emphasis on emerging synthetic strategies to improve the photoactivity of g-C₃N₄-based nanostructures through controlling size, morphology, light absorption, charge separation, and ultimately surface reactions. Future research directions are also highlighted.

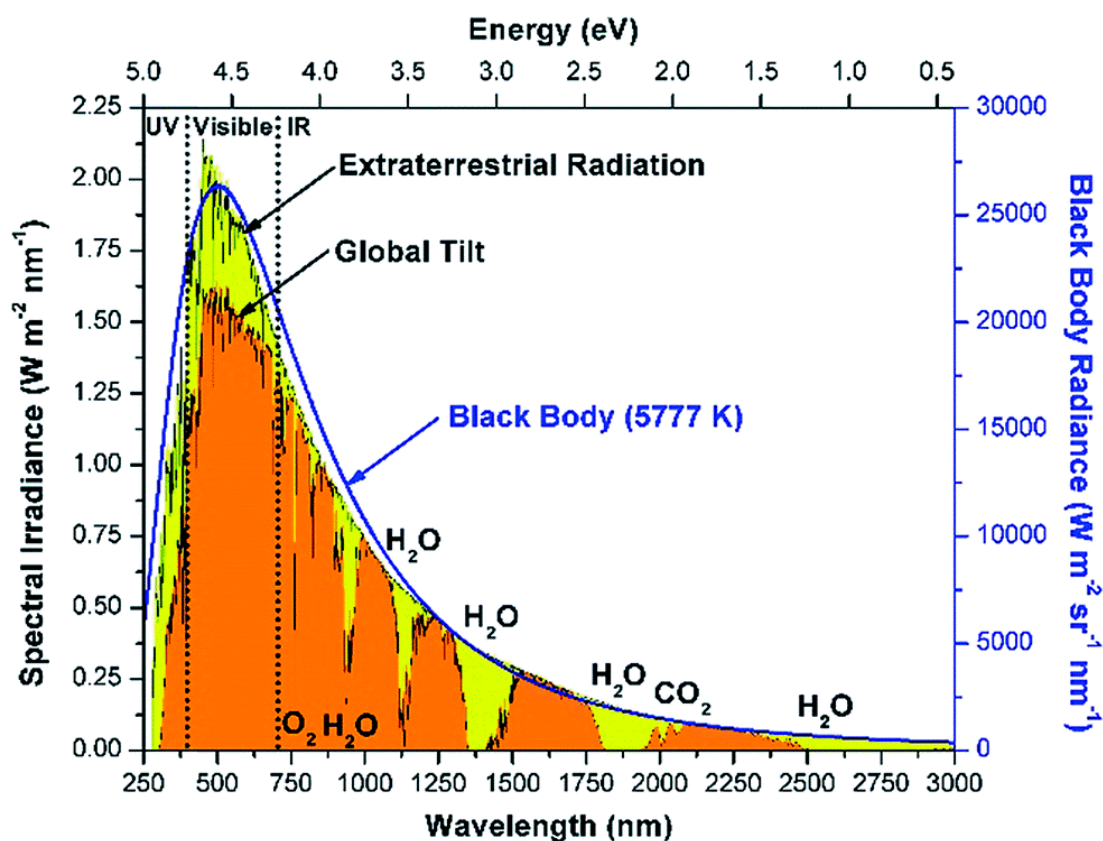


Figure 4. Spectral irradiance of solar light. Reproduced with permission from [36]. Copyright Royal Society of Chemistry, 2015.

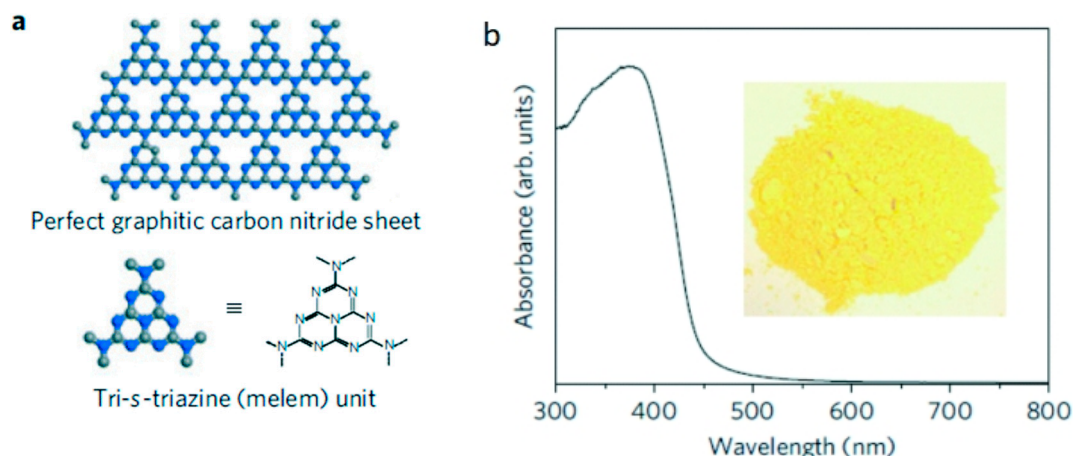


Figure 5. (a) Graphitic carbon nitride structure comprising melem units, and (b) UV-vis diffuse reflectance spectrum and image (inset) of g-C₃N₄. Reprinted with permission from [34]. Copyright Springer Nature, 2009.

4. g-C₃N₄ Nanostructures: Size and Shape

Engineering materials at the nanoscale is critical to the development of devices for the electronics [44], catalysis [45], biomedical [46], sensing [47], and smart materials [48] sectors, with nanoparticles now in widespread use across science and engineering [48–51]. A number of key aspects differentiate nanomaterials from their bulk analogues. Nanomaterials possess a high surface:bulk atom ratio, which heavily influences their thermodynamic properties resulting in, e.g., melting temperature depression, and elevated solid–solid phase transition temperature. Quantum confinement effects, which influence the electrical and optical properties of nanomaterials, arise from their evolving band structure and the emergence of atomistic like behaviour. Many heterogeneous catalysts exhibit strong size-dependencies due to quantum confinement [52], notably gold [53,54], high surface areas, and the exposure of low-coordination, high energy sites [45,55–57]. In concert, these aspects may enhance the rate of interfacial charge transfer from a photocatalyst surface to an adsorbate [58,59]. The use of nanostructured g-C₃N₄ is a fast growing area of photocatalysis research, with nanoparticles, nanorods, nanowires, nanotubes, nanospheres, and particularly nanosheets, demonstrating unique features as components of photocatalyst systems [39].

2-dimensional g-C₃N₄: 2D-based materials offer an exceptionally high specific surface area, good crystallinity, rich options for host-guest interactions, maximal light absorption, and improved charge-carrier separation over their 3D analogues [60]. Numerous 2D nanomaterials have been reported as heterogeneous catalysts in recent years, with g-C₃N₄ emerging as one of the most promising photocatalysts. Ping and co-workers developed a facile method to prepare g-C₃N₄ nanosheets by direct thermal oxidative ‘etching’ of bulk g-C₃N₄ under air (Figure 6) [61]. In this method, the hydrogen-bond strands of polymeric melon units which form the interlayers, are gradually removed by oxidation such that the thickness of bulk g-C₃N₄ can be reduced to the desired nanoscale by controlling the etch time, and hence represents a simple, low-cost, and scalable synthesis. The resultant nanosheets exhibit a blue shift of the intrinsic absorption edge in their UV-vis spectra relative to the bulk. The increase in band gap of nanosheets (2.97 eV; Figure 7A) relative to their bulk counterpart (2.77 eV) is further confirmed by a blue shift in the fluorescence emission spectrum of 20 nm (Figure 7B). This widening of the band gap reflects quantum confinement which raises and lowers the conduction and valence band edges respectively [62]. Electronic properties of the nanosheets were determined from the corresponding I–V curve, semiconducting characteristics observed for single g-C₃N₄ nanosheets, suggesting electron transport within the nanosheet plane. In contrast, no current was detected for bulk particle under an applied bias spanning –10–+10 V, evidencing extremely poor electronic conductivity

for bulk $g\text{-C}_3\text{N}_4$. The lifetime of charge carriers in the nanosheets from time-resolved fluorescence decay spectra also exceeded that of bulk $g\text{-C}_3\text{N}_4$.

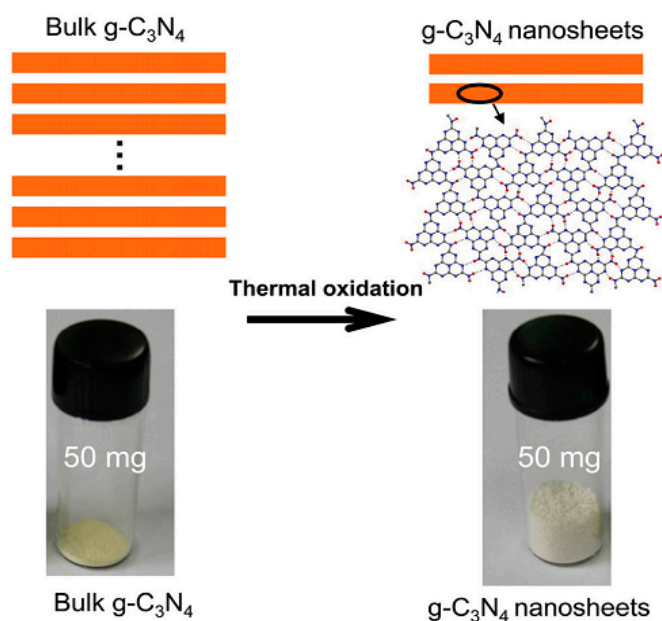


Figure 6. Thermal exfoliation as a low-cost and green method to prepare ultrathin $g\text{-C}_3\text{N}_4$ nanosheets from bulk $g\text{-C}_3\text{N}_4$ in water. Reproduced with permission from [61]. Copyright John Wiley & Sons Inc., 2012.

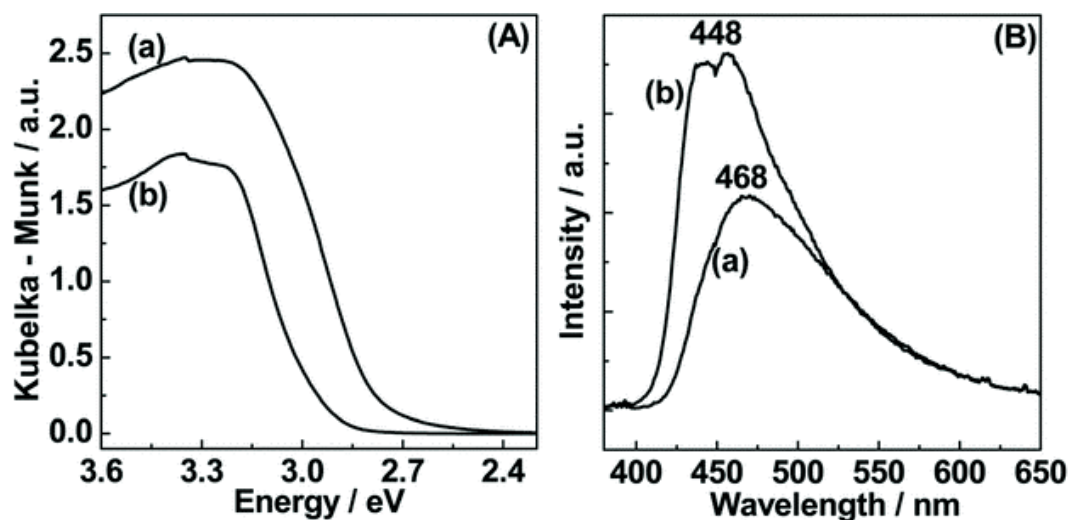


Figure 7. (A) Diffuse reflectance UV-Vis spectra (DRUVS), and (B) photoluminescence (PL) spectra of bulk (a) and exfoliated nanosheet (b) $g\text{-C}_3\text{N}_4$. Reproduced with permission from [61]. Copyright John Wiley & Sons Inc., 2012.

Xiaodong and co-workers developed a different liquid exfoliation strategy as a low-cost and green route to ultrathin $g\text{-C}_3\text{N}_4$ nanosheets from bulk $g\text{-C}_3\text{N}_4$ in water, illustrated in Figure 8 [63]. From a range of solvents, water effectively exfoliated the $g\text{-C}_3\text{N}_4$ into ultrathin nanosheets, possibly reflecting its high polarity. The morphology of the exfoliated $g\text{-C}_3\text{N}_4$ showed free-standing nanosheets 120 nm across that were almost transparent, and displayed a well-defined Tyndall effect in solution (Figure 8 inset) indicating the presence of monodisperses ultrathin nanosheets. These $g\text{-C}_3\text{N}_4$ nanosheets were very stable in acidic and alkaline environments, but exhibited pH-dependent photoluminescence.

The $g\text{-C}_3\text{N}_4$ nanosheets show superior photoabsorption to the bulk counterpart, resulting in an extremely high PL quantum yield of up to 19.6%. Liquid exfoliation of $g\text{-C}_3\text{N}_4$ in isopropanol [64] and methanol [65] resulted in nanosheets with improved photocatalytic performance for the degradation of organic pollutants relative to bulk $g\text{-C}_3\text{N}_4$.

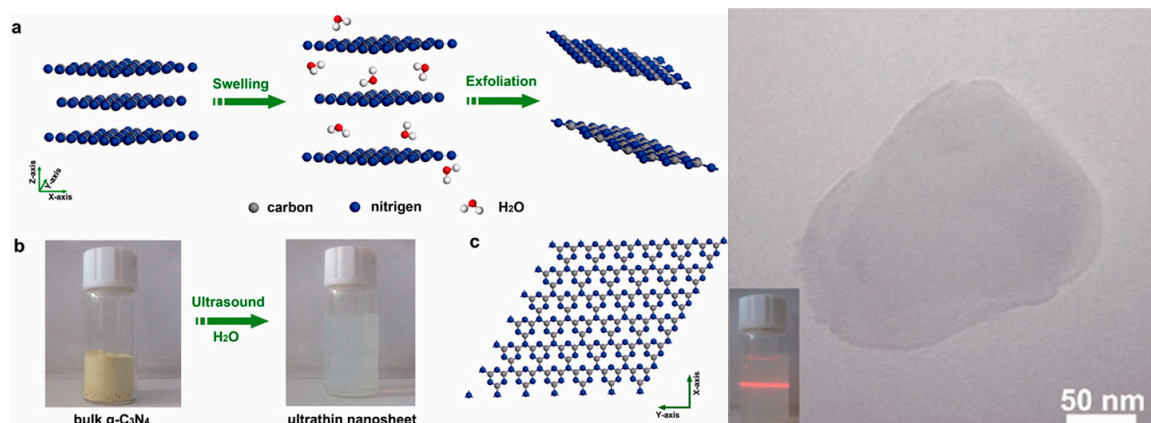


Figure 8. Liquid exfoliation route as a low-cost and green method to prepare the ultrathin $g\text{-C}_3\text{N}_4$ nanosheets from bulk $g\text{-C}_3\text{N}_4$ in water. Reprinted with permission from [63]. Copyright American Chemical Society, 2013.

1-dimensional $g\text{-C}_3\text{N}_4$: In recent years, 1D nanostructures have attracted interest as photocatalysts due to their unique morphology and photophysical properties [66,67], and hence there is interest in preparing 1D $g\text{-C}_3\text{N}_4$. 1D $g\text{-C}_3\text{N}_4$ nanorods with different aspect ratios were prepared by the reflux of $g\text{-C}_3\text{N}_4$ nanoplates as a function of solvent and reflux time [68]. The transformation from $g\text{-C}_3\text{N}_4$ nanoplates to nanorods reflects an exfoliation and subsequent re-growth process, which results in ‘rolling-up’ of individual nanosheets into rods (Figure 9a). The photocatalytic activity of the as-prepared nanorods for methylene blue (MB) degradation in water was explored under visible light ($\lambda > 420$ nm) and simulated solar irradiation ($\lambda > 290$ nm). The resulting photocatalytic activity and photocurrent response of $g\text{-C}_3\text{N}_4$ nanorods under visible and solar light were about 50–100% greater than the $g\text{-C}_3\text{N}_4$ nanoplates.

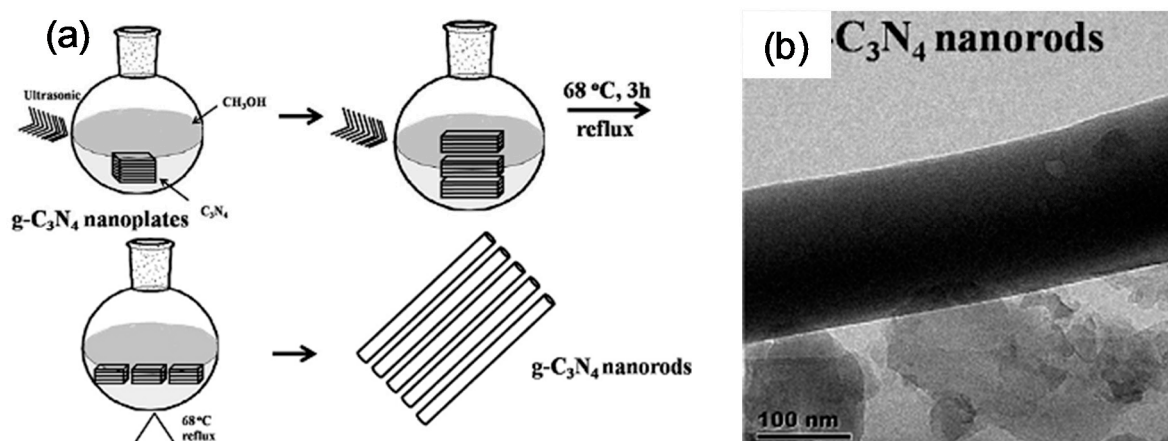


Figure 9. (a) Synthesis, and (b) Transmission electron microscopy (TEM) images of $g\text{-C}_3\text{N}_4$ nanorods. Reprinted with permission from [68]. Copyright American Chemical Society, 2013.

Zhihong and co-workers demonstrated a large-scale synthesis of well-aligned $g\text{-C}_3\text{N}_4$ nanorods via the reactive thermolysis of mechanically activated molecular precursors, $\text{C}_3\text{N}_6\text{H}_6$ and $\text{C}_3\text{N}_3\text{C}_{13}$,

under heat treatment [69]. These nanorods exhibit peculiar optical properties, evidenced by PL emission and UV-vis absorption. Uniform $g\text{-C}_3\text{N}_4$ nanorods were also synthesized via a template of monodispersed, chiral, mesostructured silica nanorods, which were easily prepared via ammonia-catalyzed hydrolysis of tetraethyl orthosilicate with F127 and cetyltrimethylammonium bromide (CTAB) surfactants [70]. The one-dimensional, hexagonal mesostructure of the porous silica nanorods enabled carbon nitride condensation within the pores. The resulting $g\text{-C}_3\text{N}_4$ nanorods demonstrated a high photocatalytic activity in hydrogen evolution from water in the presence of triethanolamine and 1 wt % Pt as a co-catalyst compared to that obtained with a conventional $g\text{-C}_3\text{N}_4$ [71]. Porous $g\text{-C}_3\text{N}_4$ nanorods were also prepared by direct calcination of hydrous melamine nanofibers, precipitated from an aqueous solution of melamine [72]. Porosity provided an enhanced interfacial area for catalysis. Oxygen atoms doped into the $g\text{-C}_3\text{N}_4$ matrix altered the band structure, resulting in more effective separation of electron/hole pairs and a corresponding excellent visible light photocatalytic activity for hydrogen evolution in the presence of triethanolamine as a hole quencher. A simple wet-chemical route was also reported for the preparation of nanofiber-like $g\text{-C}_3\text{N}_4$ structures with an average diameter of several nm and 100 nm in length [73]. The $g\text{-C}_3\text{N}_4$ nanofibers exhibited a high surface area, and low density of crystalline defects, with a slight blue shift of 0.13 eV compared to bulk $g\text{-C}_3\text{N}_4$, possibly due to more perfect packing, electronic coupling, and quantum confinement effects. The catalytic activity of $g\text{-C}_3\text{N}_4$ nanofibers for Rhodamine B photodegradation was much higher than that of bulk $g\text{-C}_3\text{N}_4$, with the nanofibers also exhibiting superior stability. An alternative approach to the synthesis of $g\text{-C}_3\text{N}_4$ nanotubes adopted the direct heating of melamine, packed into a compact configuration to favour tubular structures (Figure 10a–d) [74]. This route was advantageous since it required no additional organic templates, facilitating commercial, low-cost and large-scale application. The resulting $g\text{-C}_3\text{N}_4$ showed intense fluorescence around 460 nm, and hence has potential application as a blue light fluorescence material. These $g\text{-C}_3\text{N}_4$ nanotubes exhibited better visible light photocatalytic activity for MB degradation than either bulk $g\text{-C}_3\text{N}_4$ or a p25 TiO_2 reference (the latter is unsurprising since pure titania is a UV band gap material). Muhammad and co-workers also prepared tubular $g\text{-C}_3\text{N}_4$ by pre-treating melamine with HNO_3 before thermal processing [75]. The $g\text{-C}_3\text{N}_4$ nanotubes were again active for MB and methylene orange (MO) degradation under visible light, and were more stable than bulk $g\text{-C}_3\text{N}_4$; the superior activity attributed to the higher surface area ($182 \text{ m}^2 \cdot \text{g}^{-1}$) of the tubes and improved light absorption and charge separation/transfer of electron–hole pairs. $g\text{-C}_3\text{N}_4$ nanotubes can also be obtained through rolling-up nanosheets via a simple water-induced morphological transformation [76], avoiding the use of organic solvents and hence promoting green chemical principles.

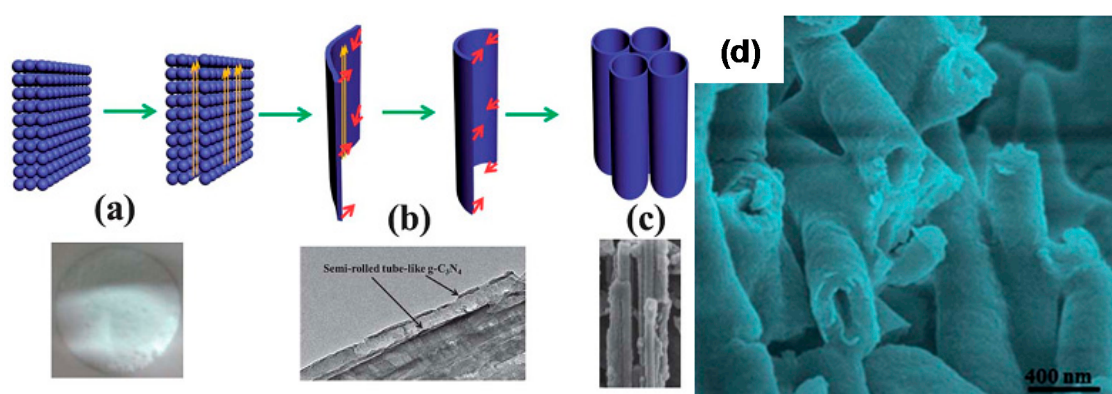


Figure 10. (a–c) Synthetic strategy, and corresponding (d) TEM image of $g\text{-C}_3\text{N}_4$ nanotubes. Reproduced with permission from [74]. Copyright Royal Society of Chemistry, 2014.

Ribbon-like $g\text{-C}_3\text{N}_4$ nanostructures have been prepared employing dicyandiamide (DCDA) and NaCl crystals as structure-directing agents [77], with a possible mechanism shown in Figure 11. These ribbon-like $g\text{-C}_3\text{N}_4$ nanostructures exhibit interesting optical and electronic properties, including a large blue shift in their absorption spectrum corresponding to an increased band gap from 2.7 eV to 3.0 eV. The latter may reflect the incorporation of some Na^+ ions within the nitride pores, and functionalization by cyano groups. The ribbon-like $g\text{-C}_3\text{N}_4$ emits blue light at around 440 nm under 365 nm excitation, whereas bulk $g\text{-C}_3\text{N}_4$ exhibited a broad emission spanning 460–520 nm, i.e., yellow-green light. Unfortunately, these ribbon-like $g\text{-C}_3\text{N}_4$ nanostructures have not yet been tested for as photocatalysts.

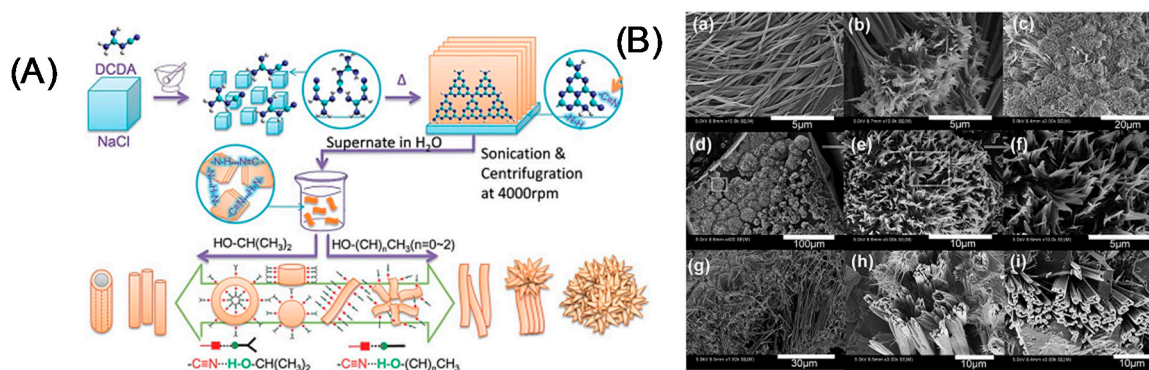


Figure 11. Synthesis strategy (A) Ribbon-like $g\text{-C}_3\text{N}_4$ nanostructures (B) TEM image. Reproduced from with permission from [77]. Copyright Royal Society of Chemistry, 2014.

0-dimensional $g\text{-C}_3\text{N}_4$: 0D materials such as quantum dots are of great interest in photocatalysis [78]. $g\text{-C}_3\text{N}_4$ quantum dots have been prepared from bulk $g\text{-C}_3\text{N}_4$ by thermochemical etching [74]. This tunable multi-step preparation involves thermal exfoliation of 3D bulk $g\text{-C}_3\text{N}_4$ into 2D nanosheets, followed by acid etching with concentrated H_2SO_4 and HNO_3 to produce 1D nanoribbons. In this second step, some C–N bonds which connect the tri-s-triazine units are oxidized, resulting in the introduction of oxygenate functional groups, such as carboxylates, at edges and on the basal plane. Cleavage of the nanosheets along preferential orientations yields nanoribbons with diameters <10 nm and several tens of nm in length. In a final step, nanoribbons are converted to 0D quantum dots of 5–9 nm across by hydrothermal treatment (Figure 12) that are highly soluble in water, and stable in solution under ambient conditions for almost eight months. These quantum dots exhibited light ‘up-conversion’ when excited by long wavelength light, for example, irradiation with 705–862 nm light resulted in 350–600 nm emission, encompassing a large portion of the visible-light spectrum. This up-conversion was proposed to occur via a multiphoton process involving anti-Stokes photoluminescence. The ability of $g\text{-C}_3\text{N}_4$ quantum dots to convert NIR to visible light renders them a promising universal energy transfer component in a photocatalytic system, able to harness long wavelength solar energy. This was demonstrated for water splitting, in which quantum dots were added to promote photocatalytic H_2 production by platinumized bulk $g\text{-C}_3\text{N}_4$ and P25, with dramatic rate-enhancements (up to 52-fold) observed for the latter under visible light irradiation in the presence of a methanol sacrificial hole scavenger. Single layered $g\text{-C}_3\text{N}_4$ quantum dots were also prepared by Guoping and co-workers, although in this instance for two-photon fluorescence imaging of cellular nucleus [42]. They again adopted a multi-step synthesis involving acid treatment of bulk $g\text{-C}_3\text{N}_4$ to form a porous material and subsequently ultrathin nanosheets, with subsequent ammonia addition, hydrothermal treatment, and ultrasonication of the porous $g\text{-C}_3\text{N}_4$ nanosheets liberating aqueous suspensions of $g\text{-C}_3\text{N}_4$ quantum dots.

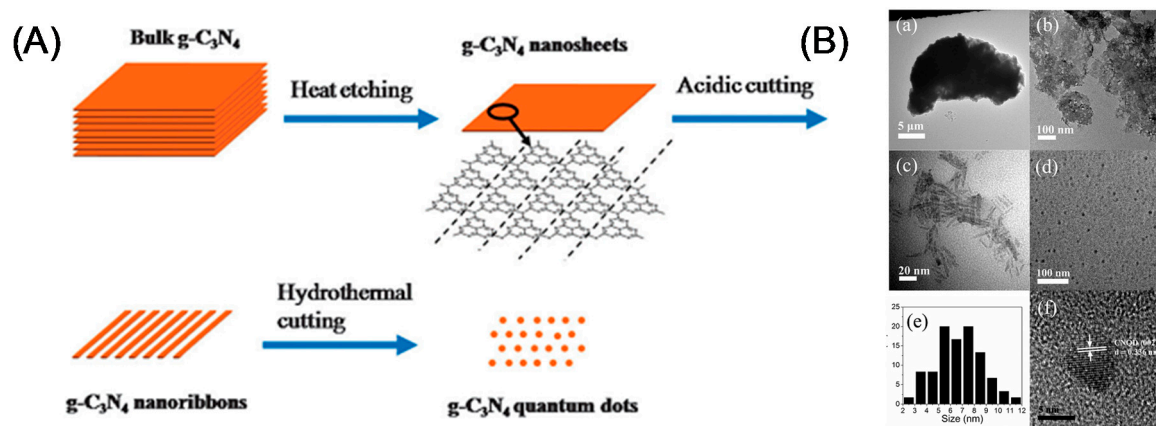


Figure 12. (A) Synthesis, and (B) TEM images of $g\text{-C}_3\text{N}_4$ quantum dots. Reproduced from with permission from [74]. Copyright Royal Society of Chemistry, 2014.

3D-dimensional $g\text{-C}_3\text{N}_4$: 3D nanomaterials unlock a vast and complex design space for constructing novel and efficient photocatalytic systems [79], such as hierarchical 3D nanoporous $g\text{-C}_3\text{N}_4$ microspheres using a template-free solvothermal synthesis [80]. In this example, a two-step synthesis was adopted: (i) amorphous and nanoporous $g\text{-C}_3\text{N}_4$ microspheres were prepared from melamine and cyanuric chloride in acetonitrile; and (ii) subsequently subjected to thermal processing at 550 °C under argon to transform the amorphous microspheres into hierarchical $g\text{-C}_3\text{N}_4$ microspheres (Figure 13). Surprisingly, the hierarchical $g\text{-C}_3\text{N}_4$ microspheres exhibited a red-shift relative to the bulk counterpart, and uncalcined microspheres, attributed to the high degree of condensation and packing between the layers within the microspheres. The photoluminescence emission intensity of hierarchical $g\text{-C}_3\text{N}_4$ microspheres was low compared to bulk and uncalcined $g\text{-C}_3\text{N}_4$ microspheres indicating that calcination suppresses radiative charge recombination in the hierarchical structure. These porous $g\text{-C}_3\text{N}_4$ microspheres also exhibit a narrowed band gap (2.42 eV), lower electrical resistance and a higher photoresponse than the bulk material, facilitating visible-light harvesting and more efficient transport and separation of photo-induced charge carriers. Hierarchical $g\text{-C}_3\text{N}_4$ nanospheres, comprised of nanosheet assemblies, were also prepared by Jinshui and co-workers, but employing high area silica nanospheres as sacrificial templates [81]. The silica template offered efficient cyanamide adsorption, and a framework for the formation of interconnected 2D $g\text{-C}_3\text{N}_4$ nanosheets during self-polymerization on heating. The excellent thermal and mechanical stability of silica spheres enabled high temperature construction of the hierarchical $g\text{-C}_3\text{N}_4$ nanospheres, and could subsequently be removed through etching by NH_4HF_2 solution, with the hierarchical $g\text{-C}_3\text{N}_4$ retaining a spherical morphology. These hierarchal nanospheres are constructed of flat nanosheets emanating from the center (sphere surface) and then interconnecting to form a mesoporous shell (Figure 14), this structure may favour both charge separation and mass transport in photocatalysis. The nanospheres had a wider band gap than bulk $g\text{-C}_3\text{N}_4$, possibly due to quantum size effects, but superior light harvesting across the optical spectrum, especially between 430–590 nm. This may arise from multiple reflections (and hence opportunities for absorption) of incident light within the hierarchical architectures, and/or presence of a high density of defective sites associated with exposure of low-coordination sites at the ‘sharp’ edges of the constituent nanosheets.

Hollow $g\text{-C}_3\text{N}_4$: Hollow nanostructures are another promising morphology for energy storage and conversion applications, with significant research efforts devoted to the design and synthesis of hollow nanostructures with high complexity through manipulating their geometry, chemical composition, building blocks, and interior architecture to, e.g., enhance their electrochemical performance [82,83]. Hollow $g\text{-C}_3\text{N}_4$ nanospheres have been synthesized using silica nanoparticles as templates [83,84]. Careful control over the shell thickness of such polymeric $g\text{-C}_3\text{N}_4$ hollow nanospheres prevents deformation of the core–shell arrangement (Figure 15), even after 400 °C

processing. Although a blue shift in the band gap accompanying their synthesis is undesirable, and attributed to either quantum effects or enhanced H-type interlayer packing, further chemical methods, such as extending the pi system by anchoring aromatic motifs, exist to improve visible light absorption, for example, through co-polymerization; such chemical modification and extended p-conjugation can red-shift optical absorption, and improve charge separation in the shell, without damaging the hollow polymeric architectures [85]. This strategy has been adopted to tune the semiconductor properties of the shell in the hollow g-C₃N₄ nanospheres to enhance photocatalytic activity for hydrogen evolution under visible light. A simple, molecular cooperative assembly of low cost triazine molecules into hollow g-C₃N₄ is also reported by Young-Si et al. [86], with this precursor enabling simultaneous optimization of the textural and photophysical properties of g-C₃N₄.

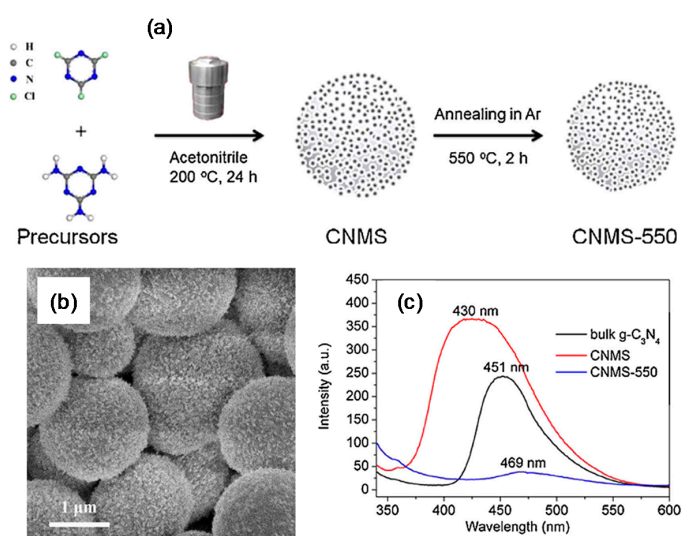


Figure 13. (a) Synthetic strategy, (b) TEM image, and (c) room-temperature photoluminescence spectra of porous g-C₃N₄ microspheres. Reprinted with permission from [80]. Copyright Elsevier, 2015.

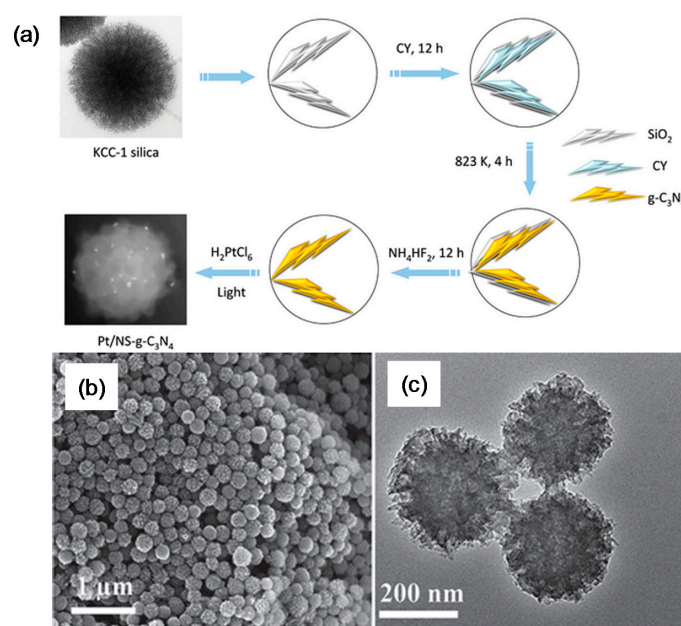


Figure 14. (a) Synthetic strategy, and (b,c) Scanning electron microscopy and TEM images of hierarchical g-C₃N₄ microspheres. Reproduced with permission from [81]. Copyright John Wiley & Sons Inc., 2014.

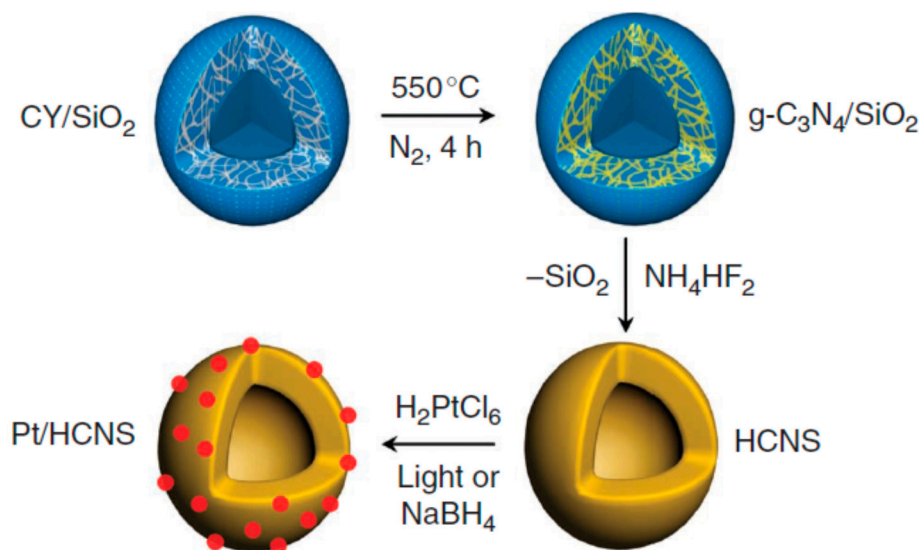


Figure 15. Synthetic strategy for fabricating hollow $g\text{-C}_3\text{N}_4$ nanospheres. Reprinted with permission from [84]. Copyright Springer Nature, 2012.

Mesoporous $g\text{-C}_3\text{N}_4$: Mesoporous photocatalysts have attracted attention for their (comparatively) high quantum efficiency associated with high surface areas, superior molecular mass transport in-pore [87], and opportunities for enhanced light harvesting through the internal scattering of incident light. An atomically thin mesoporous mesh of $g\text{-C}_3\text{N}_4$ nanosheets was recently prepared by solvothermal synthesis (Figure 16) which exhibits outstanding photocatalytic activity for H_2 production [88].

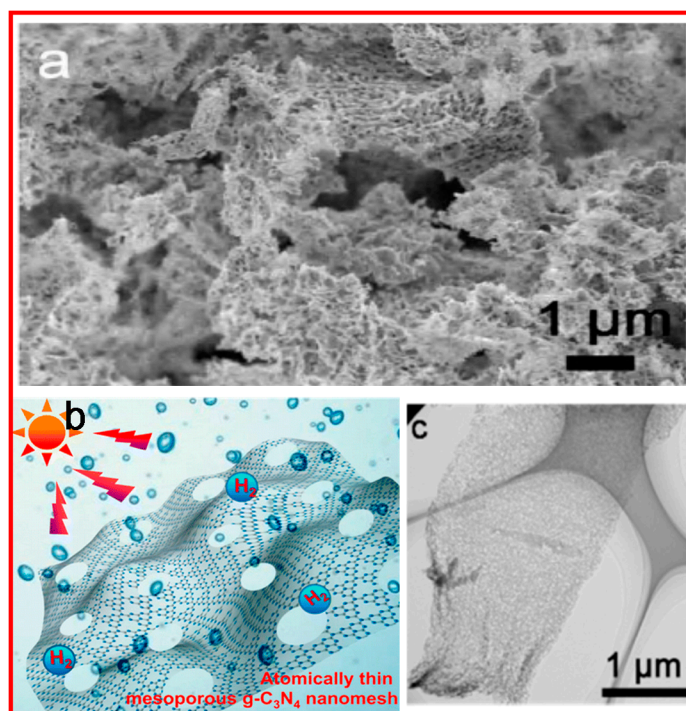


Figure 16. (a) SEM images, (b) cartoon of photocatalytic H_2 from water splitting, and (c) TEM image of atomically thin, mesoporous $g\text{-C}_3\text{N}_4$ nanosheets. Reprinted with permission from [88]. Copyright American Chemical Society, 2016.

5. Photocatalytic Applications of g-C₃N₄ Nanostructures

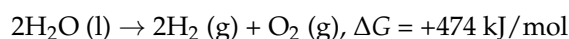
g-C₃N₄ nanostructures have proven excellent catalysts in diverse applications [39,85,89,90] including hydrogen production from water splitting [88,91], CO₂ reduction to fuels and chemicals [92], environmental remediation [39], fuel cells [93], and organic synthesis [89]. Here we focus on photocatalytic applications.

5.1. Solar Fuel Generation

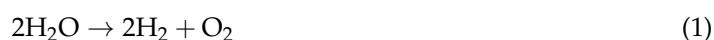
Solar fuels production from CO₂ and water via artificial photosynthesis is one of the promising strategies to deliver H₂, syngas and hydrocarbons as sustainable energy and chemical feedstocks [19]. g-C₃N₄ offers the promise of metal-free and scalable photocatalysts for visible light use.

5.1.1. H₂ Evolution

Hydrogen is one of the most promising alternative energy sources to fossil fuels; however, the large energy barrier to water splitting still presents a challenge to practical photocatalytic systems [35]:



Advanced materials are hence sought that are amenable to harnessing sunlight for either direct photochemical, or photoelectrochemical water splitting. For photocatalytic water splitting, the conduction band (CB) energy must be sufficiently negative (relative to normal hydrogen electrode (NHE)) such that photoexcited electrons are sufficiently energetic to reduce water [94,95]:



The redox potential for the overall reaction at pH = 7, $E_{\text{H}} = -1.23 \text{ V}$ (NHE), with the corresponding half-reactions of -0.41 V (Equation (4)) and 0.82 V , giving an overall $\Delta G^0 = +237 \text{ kJ}\cdot\text{mol}^{-1}$.

Most single component photocatalysts exhibit poor activity for visible light-driven H₂ production. However, the combination of g-C₃N₄ with a metal co-catalyst and hole scavenger can afford high visible-light photoactivity. Shubin and co-workers [64] prepared g-C₃N₄ nanosheets by thermal exfoliation which demonstrated a superior hydrogen production from a water/triethanolamine solution relative to the bulk nitride; nanosheets with a thickness as low as 2 nm were optimal, achieving rate-enhancements of 5.5- and 3-fold under UV-vis and visible light irradiation respectively. Single atomic layer g-C₃N₄ nanosheets prepared by a chemical exfoliation [96] also display better photogenerated charge transport and separation than bulk g-C₃N₄, presumably due to the improved H₂ evolution. Atomically thin, mesoporous g-C₃N₄ nanomesh prepared by solvothermal routes exhibits an exceptional photocatalytic activity for H₂ evolution [88] of $8510 \mu\text{mol}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$ (with an apparent quantum efficiency of 5.1% at 420 nm), far higher than the $1560 \mu\text{mol}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$ achieved over non-porous 2D g-C₃N₄ nanostructures or $350 \mu\text{mol}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$ observed for bulk g-C₃N₄ (apparent quantum efficiency 3.75% at 420 nm); the porous g-C₃N₄ nanomesh possessed a high surface area and better alignment of conduction and valence band edges. g-C₃N₄ nanorods also show high photocatalytic activity for hydrogen production from water in the presence of triethanolamine (TEOA) and a 1 wt % Pt co-catalyst [68] wherein the platinum nanoparticles uniformly decorate the g-C₃N₄ nanorods. Such materials are also superior to mesoporous analogues [70]. TEOA is the most common hole scavenger for g-C₃N₄ photocatalysts wherein it confers superior activity to methanol (a 14-fold rate enhancement); although the origin of this difference remains poorly understood, Jones and co-workers speculated that the nitrogen lone pair is responsible for the enhanced activity [62]. P25 also

exhibited superior activity for photocatalytic hydrogen production when TEOA was employed as a hole scavenger (versus methanol), albeit to a lesser extent than for carbon nitride. $g\text{-C}_3\text{N}_4$ nanotubes synthesized through a rolling-up mechanism by water-induced morphological transformation also display superior visible-light H_2 production bulk $g\text{-C}_3\text{N}_4$ or $g\text{-C}_3\text{N}_4$ nanosheets [76]. $g\text{-C}_3\text{N}_4$ quantum dots [77] prepared from bulk $g\text{-C}_3\text{N}_4$ by thermochemical etching were three times more active than bulk $g\text{-C}_3\text{N}_4$ under visible irradiation when promoted by 1 wt % Pt and using 10% triethanolamine as a sacrificial agent, possibly due to up-conversion of NIR to visible light and concomitant increased light harvesting. Tuning of the electronic band structure of $g\text{-C}_3\text{N}_4$ quantum dots [97] to optimize their visible or NIR light response, further enhances photocatalytic H_2 evolution. P-doped $g\text{-C}_3\text{N}_4$ nanosheets also exhibit promising visible-light photocatalytic H_2 productivity of $1596 \text{ mmol}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$ (apparent quantum efficiency of 3.56% at 420 nm) superior to other metal-free $g\text{-C}_3\text{N}_4$ nanosheet photocatalysts [98]. The excellent photocatalytic activity originates from P-doped macroporous analogues arises from empty mid-gap states (-0.16 V vs. NHE) which extend light harvesting up to 557 nm. Macropores also increased the surface area to $123 \text{ m}^2\cdot\text{g}^{-1}$, and shortened the charge-to-surface migration length to only 5–8 nm.

Hierarchically 3D nanoporous $g\text{-C}_3\text{N}_4$ microspheres [80] have also been exploited for water splitting in aqueous solution with 15 triethanolamine and 3 wt % Pt as a co-catalyst under visible light. These $g\text{-C}_3\text{N}_4$ microspheres showed H_2 productivity 2.5 times higher than that of bulk $g\text{-C}_3\text{N}_4$, and good stability over five consecutive recycles. Hierarchical $g\text{-C}_3\text{N}_4$ nanospheres [81] comprised of nanosheets with 3 wt % Pt co-catalyst showed significant improvements in H_2 production, with an apparent quantum yield of 9.6% at 420 nm, far superior to that for individual $g\text{-C}_3\text{N}_4$ nanosheets of 3.75%. Monodispersed, hollow $g\text{-C}_3\text{N}_4$ nanospheres are also reported to exhibit high photoactivity for water splitting, and a high apparent quantum yield of 7.5% [84]. H_2 evolution over these hollow $g\text{-C}_3\text{N}_4$ spheres was significantly enhanced by addition of a MoS_2 co-catalyst, with the formation of the $\text{MoS}_2/g\text{-C}_3\text{N}_4$ heterojunctions (Figure 17) improving light-harvesting, and fast charge separation [99].

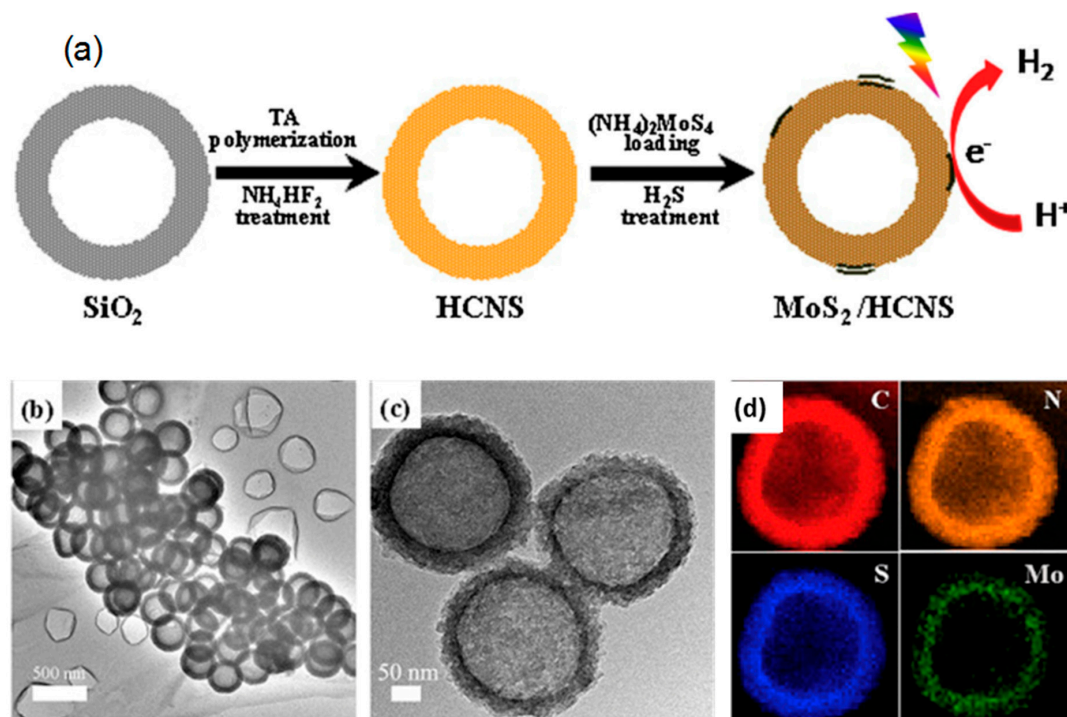


Figure 17. (a) Synthetic strategy and (b,c) TEM images and (d) Energy dispersive X-ray spectroscopy (EDX) elemental maps of MoS_2 @hollow $g\text{-C}_3\text{N}_4$. Reprinted with permission from [99], copyright Elsevier, 2016.

$g\text{-C}_3\text{N}_4$ has also been coupled with semiconductors and metal nanoparticles that exhibit visible light surface plasmon resonances to extend their spectral range. Such heterojunction materials offer enhanced separation of photoexcited charge carriers, and hence suppressed recombination and energy loss through fluorescence [39,91]. Noble metal-promoted $g\text{-C}_3\text{N}_4$ offers improved UV and visible light harvesting, fast molecular diffusion, and a high density of photoactive sites [100–102]. $\text{TiO}_2/g\text{-C}_3\text{N}_4$ heterojunctions have been fabricated by a two-step hydrothermal-calcination route from melamine, followed by an in-situ solid-state reaction [103]. The resulting $\text{TiO}_2/g\text{-C}_3\text{N}_4$ heterostructures possess a narrow band gap and good photoactivity ($556 \mu\text{mol}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$) for H_2 evolution under visible-light irradiation compared to pure $g\text{-C}_3\text{N}_4$ ($108 \mu\text{mol}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$) or TiO_2 ($130 \mu\text{mol}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$). Core@shell heterojunction nanocomposites have additional advantages due to a high interfacial contact area between the shell and core components [104]. For example, $\text{CdS}@g\text{-C}_3\text{N}_4$ core/shell nanowires [104] with different $g\text{-C}_3\text{N}_4$ contents were prepared by a combined solvothermal and chemisorption method (Figure 18) in which $g\text{-C}_3\text{N}_4$ uniformly adsorbs over CdS nanowires resulting in enhanced improved photocatalytic H_2 production of $4152 \mu\text{mol}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$ for 2 wt % $g\text{-C}_3\text{N}_4$. A one-step self-assembly route was recently developed to fabricate core-shell architecture comprising carbon spheres decorated by $g\text{-C}_3\text{N}_4$. These composites showed extended light absorption and high mechanical stability, with enhanced conductivity for charge transport [105], delivering hydrogen evolution rates of $129 \text{ mol}\cdot\text{h}^{-1}$, and 8-fold improvement over pristine $g\text{-C}_3\text{N}_4$ ($16 \text{ mol}\cdot\text{h}^{-1}$). Other $g\text{-C}_3\text{N}_4$ nanocomposites were investigated with a range of materials and morphologies [39,82,91,106–128], to access different charge transfer mechanisms between $g\text{-C}_3\text{N}_4$ and the other components. These include a $g\text{-C}_3\text{N}_4$ -based type II heterojunction [103], $g\text{-C}_3\text{N}_4$ -based p-n heterojunction [91,129], $g\text{-C}_3\text{N}_4$ -based Z-scheme heterojunction [113,130], $g\text{-C}_3\text{N}_4$ /metal heterojunction [100,102], and a $g\text{-C}_3\text{N}_4$ /carbon heterojunction [131]. The design of $g\text{-C}_3\text{N}_4$ heterojunction photocatalysts is an attractive strategy to tune the electronic structure and redox potentials for visible-light absorption photocatalytic H_2 generation. Table 1 compares the performance of different $g\text{-C}_3\text{N}_4$ photocatalysts.

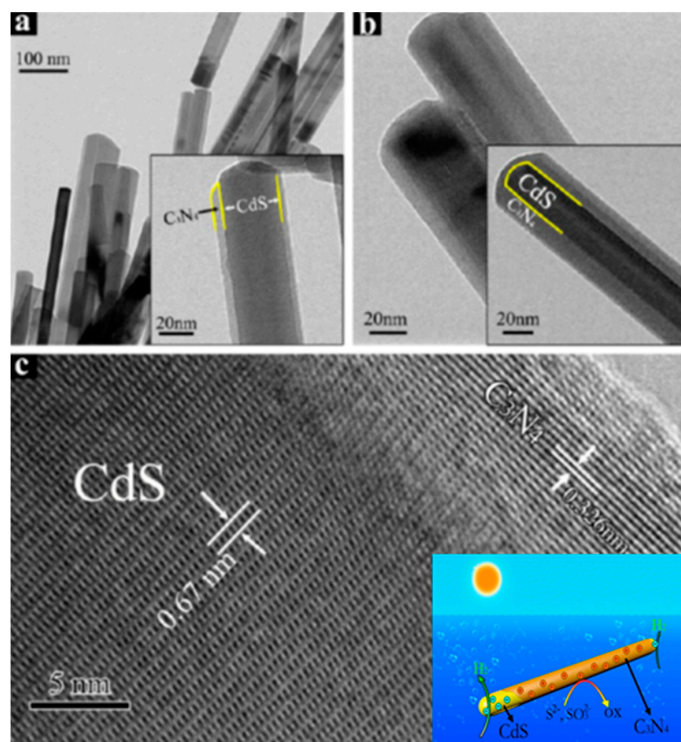


Figure 18. (a,b) TEM and (c) HR TEM image of core-shell $\text{CdS}@g\text{-C}_3\text{N}_4$ heterojunction nanocomposite. Reprinted with permission from [104]. Copyright 2013 American Chemical Society.

Table 1. Photocatalytic H₂ production over g-C₃N₄ nanostructured catalysts.

Entry	Photocatalyst	Co-Catalyst (Loading)	Experimental Details	H ₂ Productivity/ $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	Reference Material/ $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	Enhancement Relative to Conventional g-C ₃ N ₄	Apparent Quantum Efficiency/%	Reference
1	g-C ₃ N ₄ nanosheets (thermal exfoliation)	Pt (6 wt %)	10 vol% TEOA 300 W Xe ($\lambda \geq 400$ nm)	170	bulk g-C ₃ N ₄ 31.48	5.4		[61]
2	g-C ₃ N ₄ nanosheets (liquid exfoliation)	Pt (3 wt %)	10 vol% TEOA 300 W Xe ($\lambda \geq 420$ nm)	93 μmol	bulk g-C ₃ N ₄	10		[64]
3	g-C ₃ N ₄ nanosheets (thermal treatment)	Pt (0.5 wt %)	15 vol% TEOA 300 W Xe ($\lambda > 420$ nm)	1400	g-C ₃ N ₄ 450	3	2.6 (420 nm)	[132]
4	g-C ₃ N ₄ nanosheets	Pt (1 wt %)	10 vol% TEOA full sunlight and $\lambda > 400$ nm	1395	bulk g-C ₃ N ₄ 250	5.6		[133]
5	Single layer g-C ₃ N ₄	Pt (3 wt %)	10 vol% TEOA 500 W Xe ($\lambda > 420$ nm)	230	bulk g-C ₃ N ₄ 90	2.5		[96]
6	Urea derived g-C ₃ N ₄	Pt (3 wt %)	~10 vol% TEOA 300 W Xe ($\lambda \geq 395$ nm)	3327.5	DCDA derived g-C ₃ N ₄ thiourea derived g-C ₃ N ₄	7 10	26.5 (400 nm)	[134]
7	Nano Spherical-g-C ₃ N ₄	Pt (3 wt %)	10 wt % TEOA 300 W Xe ($\lambda > 420$ nm)	14,350	Pt/bulk g-C ₃ N ₄ 318	45	9.6 (420 nm)	[81]
8	g-C ₃ N ₄ nanostructure	Pt (3 wt %)	15 wt % TEOA 300 W Xe ($\lambda > 420$ nm)	689	bulk g-C ₃ N ₄ 8	8.6		[135]
9	Porous g-C ₃ N ₄ microspheres	Pt (3 wt %)	15 wt % TEOA 300 W Xe ($\lambda > 420$ nm)	180	bulk g-C ₃ N ₄ 7.8	2.3	1.62 (420 nm)	[80]
10	Silica templated mesoporous g-C ₃ N ₄	Pt (3 wt %)	10 vol% TEOA $\lambda > 420$ nm	237.4 ($\mu\text{mol}^{-1}\text{m}^{-2}$)	g-C ₃ N ₄ 9.16 ($\mu\text{mol}\cdot\text{h}^{-1}\cdot\text{m}^{-2}$)	25.8		[136]
11	macroscopic 3D porous g-C ₃ N ₄ monolith	Pt (3 wt %)	10 vol% TEOA 300 W Xe ($\lambda > 420$ nm)	29	g-C ₃ N ₄ 10.2	2.8		[137]
12	hollow g-C ₃ N ₄ nanospheres	Pt (3 wt %)	10 wt % TEOA 300 W Xe	15,000	pure g-C ₃ N ₄ 5000	3		[85]
13	Iodine doped-g-C ₃ N ₄	Pt (3 wt %)	10 vol% TEOA 300 W Xe ($\lambda \geq 420$ nm)	890	bulk g-C ₃ N ₄ 98.8	9		[138]
14	P doped-g-C ₃ N ₄	Pt (3 wt %)	10 wt % TEOA 300 W Xe	2082	pure g-C ₃ N ₄ 226.3	9.2		[139]
15	O-doping supramolecular porous g-C ₃ N ₄	Pt (3 wt %)	10 vol% TEOA 300 W Xe ($\lambda \geq 420$ nm)	1204	bulk g-C ₃ N ₄ 3D porous g-C ₃ N ₄	6.1 3.1	7.8 (420 nm)	[140]
16	K-g-C ₃ N ₄	Pt (0.5 wt %)	10 vol% TEOA 300 W Xe ($\lambda > 400$ nm)	1028	pure g-C ₃ N ₄ 73.4	14		[141]
17	AuPd/g-C ₃ N ₄	Au and Pd	10 vol% TEOA 300 W Xe ($\lambda \geq 400$ nm)	326	Au/g-C ₃ N ₄ Pd/g-C ₃ N ₄	3.5 1.6		[142]

Table 1. Cont.

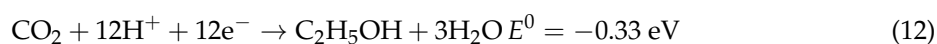
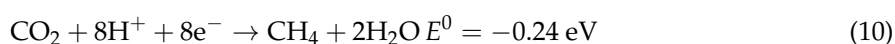
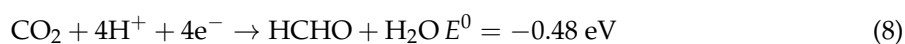
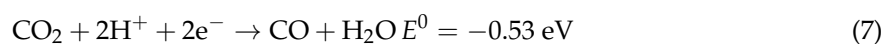
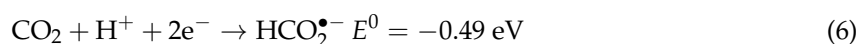
Entry	Photocatalyst	Co-Catalyst (Loading)	Experimental Details	H ₂ Productivity/ $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	Reference Material/ $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	Enhancement Relative to Conventional g-C ₃ N ₄	Apparent Quantum Efficiency/%	Reference
18	Hydrogenated g-C ₃ N ₄	Pt (3 wt %)	10 vol% TEOA 350 W mercury arc lamp ($\lambda > 420$ nm)	900	bulk g-C ₃ N ₄ 132.3	6.8		[143]
19	Surface alkalization of g-C ₃ N ₄	Pt (1 wt %)	20 vol% aq. methanol 300 W Xe	2230	urea derived g-C ₃ N ₄ 159.3	14	6.67 (400 nm)	[144]
20	dye sensitized g-C ₃ N ₄ nanosheets	Pt	5 vol% TEOA 300 W Xe ($\lambda > 420$ nm)	6525	Pt/g-C ₃ N ₄ 466	14	33.4 (460 nm)	[145]
21	2-Aminobenzonitrile-mp-g-C ₃ N ₄	Pt (3 wt %)	10 vol% TEOA 300 W Xe ($\lambda \geq 420$ nm)	229	mp-g-C ₃ N ₄ 127	1.8		[146]
22	PPy-g-C ₃ N ₄	Pt (3 wt %)	No sacrificial reagent 350 W Xe ($\lambda > 400$ nm)	154	Pt-g-C ₃ N ₄	49.3		[147]
23	Cu ₂ O@g-C ₃ N ₄ core@shell		10 vol% TEOA 300 W Xe	202.28	Cu ₂ O 35.08	5.7		[148]
24	CdS/g-C ₃ N ₄ core/shell	Pt (0.6 wt %)	0.35 M Na ₂ S and 0.25 M Na ₂ SO ₃ 300 W Xe ($\lambda \geq 420$ nm)	4152	pure CdS 2001	2.1	4.3 (420 nm)	[104]
25	Core-shell Ni/NiO-decorated g-C ₃ N ₄	Ni/NiO	10 vol% TEOA 300 W Xe	10	pure g-C ₃ N ₄ 1.01	10		[149]
26	MoS ₂ /g-C ₃ N ₄	N/A	10 vol% TEOA 300 W Xe ($\lambda > 400$ nm)	252	pure g-C ₃ N ₄ 31.5	8		[150]
27	CdS QD/g-C ₃ N ₄	Pt (0.5 wt %)	0.1 M L-ascorbic acid (pH = 4) 300 W Xe ($\lambda > 420$ nm)	4494	pure g-C ₃ N ₄ 299	15		[151]
28	CdS nanorods/g-C ₃ N ₄	NiS	10 vol% triethanolamine 300 W Xe ($\lambda \geq 420$ nm)	2563	pure g-C ₃ N ₄ 1582	1.6		[152]
29	CaIn ₂ S ₄ /g-C ₃ N ₄	Pt (1 wt %)	0.5 M Na ₂ S and 0.5 M Na ₂ SO ₃ 300 W Xe	102	CaIn ₂ S ₄ 34	3		[153]
30	BiPO ₄ /P-g-C ₃ N ₄	N/A	Na ₂ S (0.1 M) 300 W Xe ($\lambda \geq 420$ nm)	1110	P-g-C ₃ N ₄ 676	1.6		[154]
31	AgQCs/g-C ₃ N ₄	Pt (1 wt %)	25 vol% methanol simulator AM 1.5 G	5.59	pure g-C ₃ N ₄ 3.29	1.7		[155]
32	Al ₂ O ₃ /g-C ₃ N ₄	Pt (1 wt %)	25 vol% TEOA 300 W Xe ($\lambda \geq 420$ nm)	52.10	pure g-C ₃ N ₄ 20.75	2.5		[156]
33	MoS ₂ /mp-g-C ₃ N ₄	Pt	10 vol% lactic acid 300 W Xe ($\lambda \geq 420$ nm)	1030	Pt/mp-g-C ₃ N ₄ 239.5	4.3	2.7 (420 nm)	[157]
34	carbon black/g-C ₃ N ₄	Pt (3 wt %)	25 vol% methanol $\lambda > 420$ nm	689	pure g-C ₃ N ₄ 215	3.2		[158]
35	graphene/g-C ₃ N ₄	Pt (1.5 wt %)	25 vol% methanol 350 W Xe ($\lambda > 400$ nm)	451	g-C ₃ N ₄ 150	3		[107]
36	carbon black/NiS/g-C ₃ N ₄	NiS	15 vol% TEOA 300 W Xe ($\lambda \geq 420$ nm)	992	g-C ₃ N ₄ /NiS 396	2.5		[159]

Table 1. Cont.

Entry	Photocatalyst	Co-Catalyst (Loading)	Experimental Details	H ₂ Productivity/ $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	Reference Material/ $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	Enhancement Relative to Conventional g-C ₃ N ₄	Apparent Quantum Efficiency/%	Reference
37	N,S-TiO ₂ /g-C ₃ N ₄	N/A	10 vol% methanol 125 W Hg lamp	317	g-C ₃ N ₄ 125	2.5		[160]
38	N-CeO _x /g-C ₃ N ₄	Pt (1 wt %)	10 vol% TEOA 300 W Xe ($\lambda \geq 420$ nm)	292.5	g-C ₃ N ₄ 134.5	2		[161]
39	g-C ₃ N ₄ (2D)/CdS (1D)/rGO (2D)	Pt (1 wt %)	10 vol% TEOA 300 W Xe ($\lambda \geq 420$ nm)	4800	pure g-C ₃ N ₄ g-C ₃ N ₄ /rGO g-C ₃ N ₄ /CdS	44 11 2.5		[122]
40	Au/(P3HT)/Pt/g-C ₃ N ₄	Au and Pt	10 vol% TEOA 300 W Xe ($\lambda > 420$ nm)	320	g-C ₃ N ₄ /Au; 73 and g-C ₃ N ₄ /Pt; 82	4		[162]

5.1.2. CO₂ Reduction

Rising atmospheric levels of carbon dioxide and the depletion of fossil fuel reserves raise serious concerns about the continued reliance on the use of fossil fuels for both energy and chemicals production [3,163], to which the photocatalytic reduction of CO₂ to light oxygenates and hydrocarbons could provide a sustainable solution. CO₂ reduction involves multi-electron transfer and hence the reaction kinetics for, e.g., formic acid, carbon monoxide, formaldehyde, methanol and methane production are intrinsically slower than for H₂ production. CO₂ photoreduction begins with molecular adsorption at the catalyst surface, wherein the anion radical is generated by the transfer of electrons photoexcited across the semiconductor band gap following light absorption. In the case of aqueous phase CO₂ reduction, charge-compensation occurs through concomitant water splitting and the transfer of photoexcited holes in the valence band onto hydrogen atoms, with the resulting protons migrating to the CO₂ anion. The reduction potentials for CO₂ photoreduction with water to various products are described below (relative to NHE at pH = 7) [11,164]:



Key factors influencing CO₂ photocatalytic reduction include band energy matching, efficient charge-carrier separation, kinetic of e- and hole transfer to CO₂ and the reductant, and the basicity of the photocatalyst and hence strength and coverage of CO₂ adsorption [164]. In recent years, the g-C₃N₄ nanostructured materials have been studied for CO₂ photoreduction [92,165], due to their excellent stability, sufficiently negative CB energy and narrow band gap. Many strategies are reported to promote g-C₃N₄ with condensed matter and molecular sensitizers [166,167], such as doping with metals [168,169] and non-metal [170–172], heterojunction construction [173–176] and Z-scheme composites employing co-catalysts [165–167,173,175]. Pengfei et al. reported ultrathin C₃N₄ nanosheets for enhanced photocatalytic CO₂ reduction [177] in which surface functionalization and textural modification by NH₃-mediated thermal exfoliation enhanced light harvesting, charge-carrier redox potentials, and the surface area for CO₂ adsorption (to 0.2 mmol·g⁻¹), resulting in CH₄ and CH₃OH productivities of 1.39 and 1.87 μmol·h⁻¹·g⁻¹ respectively, a five-fold increase over bulk g-C₃N₄. Jiaguo and co-workers [168] demonstrated that Pt promotion significantly influenced both the activity and selectivity of g-C₃N₄ for CO₂ photoreduction to CH₄, CH₃OH, and HCHO; Pt nanoparticles improved charge separation across the metal/semiconductor interface, and lowered the overpotential for CO₂ reduction. Qingqing et al. reported Pd nanoicosahedrons with twin defects promoted CO₂ reduction into CO and CH₄ over C₃N₄ nanosheets [126]. CO₂ conversion reached 61.4%, with an average CO productivity of 4.3 μmol·g⁻¹·h⁻¹ and average CH₄ productivity of 0.45 μmol·g⁻¹·h⁻¹, indicating the presence of highly reactive sites for CO₂ adsorption and activation.

Hierarchical, porous O-doped g-C₃N₄ nanotubes prepared via successive thermal oxidation exfoliation and condensation of bulk g-C₃N₄ also show promise for photocatalytic CO₂ reduction under visible light [171]. As-prepared O-doped g-C₃N₄ nanotubes comprise interconnected, multi-walled nanotubes with uniform diameters of 20–30 nm, which evolve methanol at 0.88 μmol·g⁻¹·h⁻¹, five times faster than bulk g-C₃N₄ (0.17 μmol·g⁻¹·h⁻¹). Heterojunction composites of g-C₃N₄/ZnO

synthesized by a one-step calcination route [165] are also superior to bulk $g\text{-C}_3\text{N}_4$ (2.5-fold enhancement), ascribed to a direct Z-scheme mechanism reflecting efficient $\text{ZnO} \rightarrow g\text{-C}_3\text{N}_4$ electron transfer occurring the interface. Zhongxing et al. reported that CeO_2 -modified C_3N_4 photocatalysts produced by a simple hydrothermal route were effective for the selective photocatalytic reduction of CO_2 to CH_4 [178], with a CH_4 productivity of $4.79 \text{ mmol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$, about 3.44 times that of $g\text{-C}_3\text{N}_4$. Wang et al. prepared a 2D-2D $\text{MnO}_2/g\text{-C}_3\text{N}_4$ heterojunction photocatalyst by an in-situ redox reaction between KMnO_4 and MnSO_4 adsorbed at the surface of $g\text{-C}_3\text{N}_4$ [179] for photocatalytic CO_2 reduction to CO ($9.6 \text{ mmol}\cdot\text{g}^{-1}$), in which band matching facilitated efficient separation of photogenerated charge-carriers. Photocatalytic CO_2 reduction reaction is also reported over a direct Z-scheme $g\text{-C}_3\text{N}_4/\text{SnS}_2$ catalyst [180] which yielded both CH_3OH ($2.3 \text{ }\mu\text{mol}\cdot\text{g}^{-1}$) and CH_4 ($0.64 \text{ }\mu\text{mol}\cdot\text{g}^{-1}$), with electrons in SnS_2 combining with holes in $g\text{-C}_3\text{N}_4$. Another Z-scheme mechanism is invoked for a $\text{MoO}_3/g\text{-C}_3\text{N}_4$ composite [181]. Ryo and co-workers adopted a different approach, attaching Ru(bipy)complexes to $g\text{-C}_3\text{N}_4$ nanostructures; these displayed improved activity for CO_2 photoreduction to formic acid, with a high apparent quantum yield of 5.7% at 400 nm under visible light (Figure 19). Anchoring of polyoxometalate clusters to C_3N_4 also creates active photocatalysts for CO_2 reduction [179]. Here, noble-metal-free Co_4 polyoxometallates were used to achieve a staggered band alignment, with the $\text{Co}_4@g\text{-C}_3\text{N}_4$ hybrid photocatalysts achieving $107 \text{ }\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$ and 94% selectivity for CO production under visible light ($\lambda \geq 420 \text{ nm}$); cumulative CO production reached $896 \text{ }\mu\text{mol}\cdot\text{g}^{-1}$ after 10 h irradiation, far exceeding that for unpromoted $g\text{-C}_3\text{N}_4$.

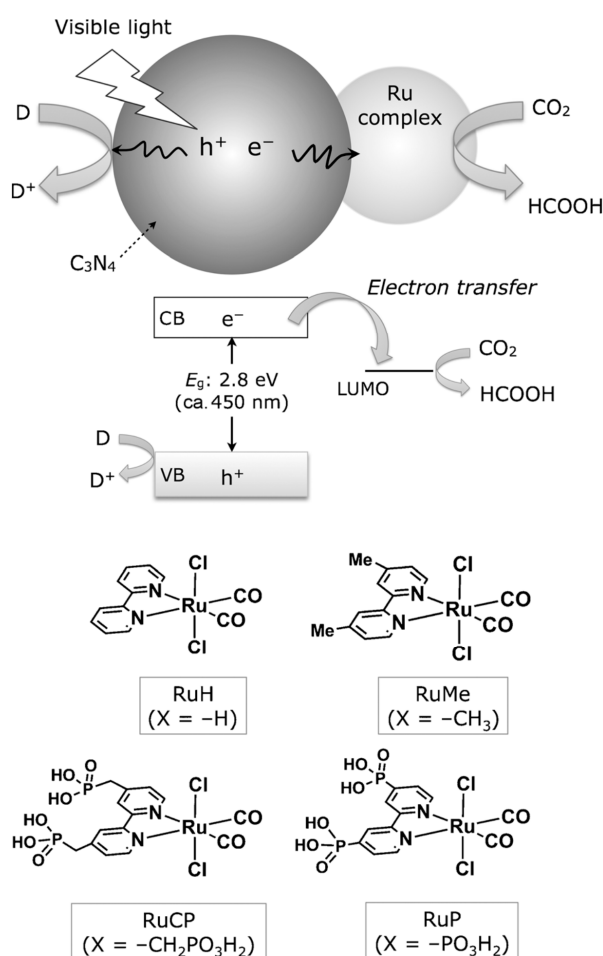


Figure 19. CO_2 reduction using a Ru complex/ C_3N_4 hybrid photocatalyst, and structures of the Ru complexes. CB = conduction band, VB = valence band. Reproduced with permission from [166]. Copyright John Wiley & Sons Inc., 2015.

A multicomponent heterostructure, termed an intercorrelated superhybrid, comprising AgBr supported on $g\text{-C}_3\text{N}_4$ decorated in turn on N-doped graphene (prepared by wet-chemical synthesis) has also shown excellent activity for the photocatalytic reduction of CO_2 to methanol and ethanol (Figure 20) [174]. Oluwatobi et al. reported $g\text{-C}_3\text{N}_4/(\text{Cu}/\text{TiO}_2)$ [182] nanocomposites prepared by pyrolysis and impregnation for enhanced photoreduction of CO_2 to CH_3OH and HCOOH under UV-vis irradiation wherein maximum productivities of CH_3OH and HCOOH under visible light were 2574 and 5069 $\text{mmol}\cdot\text{g}^{-1}$ respectively. Enhanced photoactivity was attributed to the location of the metal within the composite and consequent distribution of photoexcited electrons. Hailong et al. also studied $g\text{-C}_3\text{N}_4/\text{Ag-TiO}_2$ hybrid photocatalysts [183], wherein CO and CH_4 were preferentially formed, with a maximum CO_2 conversion of 47 $\mu\text{mol}\cdot\text{g}^{-1}$, and product yields of 28 $\mu\text{mol}\cdot\text{g}^{-1}$ CH_4 formation and 19 $\mu\text{mol}\cdot\text{g}^{-1}$ CO . Enhanced activity was proposed to arise from the transfer of photoexcited electrons across the $g\text{-C}_3\text{N}_4/\text{TiO}_2$ heterojunction, and subsequently from $\text{TiO}_2 \rightarrow \text{Ag}$ nanoparticles due to the lower Fermi level; this spatial separation of charge greatly suppressed the electron-hole recombination, with electrons accumulating on the Ag nanoparticles on the TiO_2 surface.

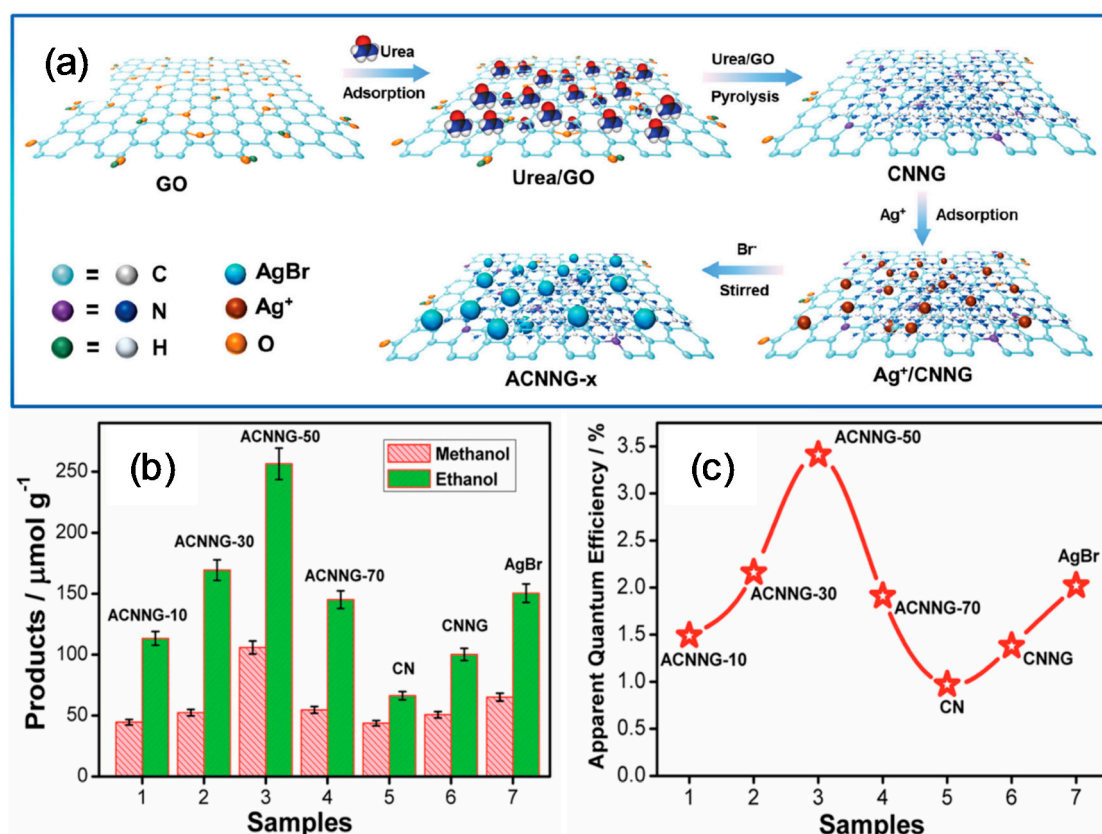


Figure 20. (a) Synthetic strategy, and (b,c) photocatalytic performance for CO_2 reduction of intercorrelated superhybrid $g\text{-C}_3\text{N}_4$ nanocomposites under visible light and corresponding apparent quantum efficiencies. Reproduced with permission from [174]. Copyright John Wiley & Sons Inc., 2015.

Table 2 compares the performance of different $g\text{-C}_3\text{N}_4$ photocatalysts for photocatalytic CO_2 reduction.

Table 2. Photocatalytic CO₂ reduction over g-C₃N₄ nanostructured catalysts.

Entry	Photocatalyst	Experimental Details	Productivity/ $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	Reference Material/ $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	Enhancement Relative to Conventional g-C ₃ N ₄	Apparent Quantum Efficiency/%	Reference
1	g-C ₃ N ₄ nanosheets	300 W Xe ($\lambda > 420$ nm), 15 °C and 25 kPa CO ₂ , catalyst in 80 mL of H ₂ O	CH ₄ : 0.94	Bulk g-C ₃ N ₄ : 0.30	3.1		[184]
2	g-C ₃ N ₄ nanosheets	300 W Xe (400 nm), 200 mW/cm ² . 20 mg catalyst in 0.1 mL H ₂ O, CO ₂ bubbled to 0.06 MPa	CH ₄ : 1.2 CH ₃ OH: 0.2	Bulk g-C ₃ N ₄ CH ₄ : 0.28 CH ₃ OH: 0.24	CH ₄ : 4.3		[185]
3	Ultrathin g-C ₃ N ₄ nanosheets	300 W Xe, 100 mg catalyst, 0.084 g NaHCO ₃ + H ₂ SO ₄ to release CO ₂	CH ₄ : 1.39 and CH ₃ OH: 1.87	Bulk g-C ₃ N ₄ CH ₄ : 0.14 and CH ₃ OH: 0.35	CH ₃ OH: 5.34		[177]
4	Thiourea and urea derived g-C ₃ N ₄	300 W Xe/420 nm, 40 mg catalyst	Urea derived g-C ₃ N ₄ CO: 0.56, CH ₃ CHO: 0.44, CH ₄ : 0.04 thiourea derived g-C ₃ N ₄ CO: 0.36, CH ₃ CHO: 0.26, CH ₄ = 0.025	N/A	N/A		[186]
5	Melamine and urea derived g-C ₃ N ₄	300 W Xe (420 nm), 0.2 g and 1.0 M NaOH solution (100 mL)	Urea derived g-C ₃ N ₄ CH ₃ OH: 6.28, C ₂ H ₅ OH: 4.51, O ₂ : 21.33 melamine derived g-C ₃ N ₄ CH ₃ OH: TRACE, C ₂ H ₅ OH: 3.64, O ₂ : 10.29	N/A	N/A	Urea derived g-C ₃ N ₄ : 0.18, melamine derived g-C ₃ N ₄ : 0.08	[172]
6	Thiourea, urea and DCDA derived g-C ₃ N ₄	300–795 nm KG1 filter, 40 mW cm ² illumination, 0.5 mg catalyst per mL in CH ₃ CN/TEOA/H ₂ O (3:1:1), $t = 2$ h, [Co(bpy) ₃] ²⁺ as a co-catalyst	Urea derived g-C ₃ N ₄ CO: 460, H ₂ : 138 μmol thiourea derived g-C ₃ N ₄ CO: 22, H ₂ : 86 μmol DCDA derived g-C ₃ N ₄ CO: 92, H ₂ : 94 μmol	N/A	N/A		[167]
7	Sulfur-doped g-C ₃ N ₄	300 W simulated solar Xe and 200 mL Pyrex reactor, 100 mg 1 wt % Pt co-catalyst, 0.12 g NaHCO ₃ and 0.25 mL 4 M HCl solution	CH ₃ OH: 0.37	Bulk g-C ₃ N ₄ CH ₃ OH: 0.27	1.37		[170]
8	Pd/g-C ₃ N ₄	300 W Xe/UV420 cut-off filter	CO: 0.5, CH ₄ : 0.05, CH ₃ OH: 1 $\mu\text{mol}\cdot\text{g}^{-1}$	Bulk g-C ₃ N ₄ CO: 4, CH ₄ : 0.15, CH ₃ OH: 2.5 $\mu\text{mol}\cdot\text{g}^{-1}$			[187]
9	Pt-loaded g-C ₃ N ₄	15 W energy-saving daylight bulb, flow rate of CO ₂ fixed at 5 mL·min ⁻¹	CH ₄ : 1.3	Bulk g-C ₃ N ₄ CH ₄ : 0.25	5.2		[188]
10	Pt-g-C ₃ N ₄	200 mL Pyrex reactor, 300 W simulated solar Xe, 100 mg catalyst, NaHCO ₃ (0.12 g) and HCl aq. solution (0.25 mL, 4 M)	CH ₄ : 0.25, CH ₃ OH: 0.25, HCHO: 0.125	Bulk g-C ₃ N ₄ CH ₄ : 0.07, CH ₃ OH: 0.11, HCHO: 0.06	CH ₄ : 3.57		[168]

Table 2. Cont.

Entry	Photocatalyst	Experimental Details	Productivity/ $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	Reference Material/ $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	Enhancement Relative to Conventional g-C ₃ N ₄	Apparent Quantum Efficiency/%	Reference
11	Amine-functionalized g-C ₃ N ₄	300 W Xe, Pyrex 200 mL, 100 mg catalyst, 0.084 g NaHCO ₃ + 0.3 mL of 2 M H ₂ SO ₄	CH ₄ : 0.34 CH ₃ OH: 0.28	Bulk g-C ₃ N ₄ CH ₃ OH: 0.26 CH ₄ : trace	CH ₄ : 1.3		[189]
12	SnO ₂ -coupled B and P co-doped g-C ₃ N ₄	300 W Xe (420 nm), 0.2 g catalyst in 3 mL water/100 mL NaOH purged with CO ₂	CH ₄ : 30	Bulk g-C ₃ N ₄ CH ₄ : 3.5	8.57	2.02 (420 nm)	[190]
13	g-C ₃ N ₄ -Ru complex	400 W Hg lamp (400 nm) 11 mL reactor containing 4 mL 20 vol % TEA in acetonitrile and 8 mg catalyst purged with CO ₂	HCOOH: 4.6	Bulk g-C ₃ N ₄ HCOOH: trace	N/A		[191]
14	Ag ₃ PO ₄ /g-C ₃ N ₄	500 W Xe/420 nm, stainless-steel reactor 132 mL, 10 mg in 4 mL H ₂ O, 0.4 MPa CO ₂ at 80 °C	CO: 44, CH ₃ OH: 9, CH ₄ : 0.2, C ₂ H ₅ OH: 0.1	Bulk g-C ₃ N ₄ CO: 4, CH ₃ OH: 0.35, CH ₄ : 0.09, C ₂ H ₅ OH: 0.01	CO: 11		[175]
15	AgX/g-C ₃ N ₄ (X = Cl and Br)	15 W energy-saving daylight lamp, 100 mg catalyst, CO ₂ flow of 5 mL/min	CH ₄ : 1.282	Bulk g-C ₃ N ₄ CH ₄ : 0.388	3.3		[192]
16	B ₄ C/g-C ₃ N ₄	300 W Xe (UV/IR filter), 100 mL photoreactor, 6 mg catalyst, CO ₂	CH ₄ : 0.84	Bulk g-C ₃ N ₄ CH ₄ : 0.14	6		[193]
17	BiOI/g-C ₃ N ₄	300 W Xe (400 nm), 0.10 g catalyst, CO ₂ bubbled through water.	CO: 3.58, O ₂ : 1.96, H ₂ : 0.4, CH ₄ : 0.2	Bulk g-C ₃ N ₄ CO: 0.2, O ₂ : 0.56, H ₂ : 0.92	CO: 17.9		[194]
18	g-C ₃ N ₄ /C	500 W Xe lamp, 0.1 g catalyst, CO ₂ + H ₂ O mixture flow 20 mL min ⁻¹ , 30 °C and 110 KPa CO ₂	CO: 2.5 CH ₄ : 1.4	Bulk g-C ₃ N ₄ CO: 1.1 CH ₄ : 0.72	CO: 2.27		[195]
19	CeO ₂ /g-C ₃ N ₄	300 W Xe, reactor volume 500 mL, 50 mg catalyst, CO ₂ bubbled through water	2 wt % CO: 11.8 and CH ₄ : 9.08 3 wt % CO: 10.16 and CH ₄ : 13.88	Bulk g-C ₃ N ₄ CO: 6.78 CH ₄ : 0.2	CH ₄ : 69.4		[196]
20	Graphene/g-C ₃ N ₄	15 W energy saving daylight bulb, CO ₂ 5 mL min ⁻¹	CH ₄ : 0.59 $\mu\text{mol}\cdot\text{h}^{-1}$	Bulk g-C ₃ N ₄ CH ₄ : 0.25 $\mu\text{mol}\cdot\text{h}^{-1}$	2.36		[197]
21	g-C ₃ N ₄ /NaNbO ₃	300 W Xe, reaction volume 230 mL, 50 mg catalyst, reactor purged with CO ₂ , then 2 mL H ₂ O injected	CH ₄ : 6.4	Bulk g-C ₃ N ₄ CH ₄ : 0.8	8		[173]
22	g-C ₃ N ₄ /N-TiO ₂	300 W Xe lamp, reaction system vol 780 mL, 0.1 g catalyst, flow rate of CO ₂ 15 mL min ⁻¹	CO: 14.73 μmol	Bulk g-C ₃ N ₄ CO: 4.20 μmol ; P25: 3.19 μmol	3.5		[198]
23	rGO/g-C ₃ N ₄	15 W energy-saving daylight lamp, CO ₂ at a flow rate of 5 mL/min, 100 mg catalyst	CH ₄ : 14	Bulk g-C ₃ N ₄ CH ₄ : 2.5	5.6	0.56 (420 nm)	[199]

Table 2. Cont.

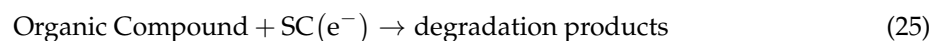
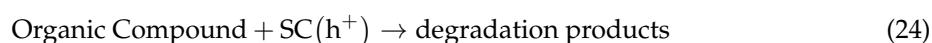
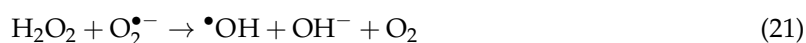
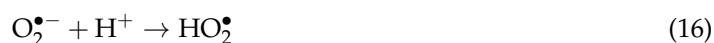
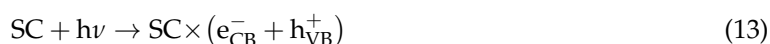
Entry	Photocatalyst	Experimental Details	Productivity/ $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	Reference Material/ $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	Enhancement Relative to Conventional g-C ₃ N ₄	Apparent Quantum Efficiency/%	Reference
24	g-C ₃ N ₄ and a Ru(II) complex	400 W high-pressure Hg lamp, 8 mg catalyst, DMA (containing 20 vol % TEOA) 4.0 mL	CO: 2.9 $\mu\text{mol}\cdot\text{h}^{-1}$, HCOOH: 1.5 $\mu\text{mol}\cdot\text{h}^{-1}$; H ₂ : 0.13 $\mu\text{mol}\cdot\text{h}^{-1}$	Bulk g-C ₃ N ₄ Only trace	N/A		[200]
25	Ru complex/mp g-C ₃ N ₄	450 W Xe lamp, 8.0 mg catalyst, acetonitrile and triethanolamine (4:1 v/v) 4 mL mix in 11 mL Pyrex test tube	CO: 0.6, H ₂ : 0.25, HCOOH: 4 $\mu\text{mol}\cdot\text{h}^{-1}$	Bulk g-C ₃ N ₄ HCOOH: trace	N/A		[201]
26	SnO ₂ /g-C ₃ N ₄	500 W Xe, 20 mg catalyst, 4 mL water injected into the bottom of the reactor, 0.3 MPa CO ₂ , 80 °C	CO: 19, CH ₄ : 2, CH ₃ OH: 3	Bulk g-C ₃ N ₄ CO: 2.4, CH ₄ : trace, CH ₃ OH: 2.8, P25: CO: 3.5, CH ₃ OH: 1	CO: 7.9		[202]
27	Brookite TiO ₂ /g-C ₃ N ₄	300 W Xe, 60 mg catalyst, CO ₂ produced from reaction of NaHCO ₃ (1.50 g) and H ₂ SO ₄ solution (5.0 mL, 4 M)	CO: 0.84, CH ₄ : 5.21	Bulk g-C ₃ N ₄ CO: 7.10, CH ₄ : 1.84	CH ₄ : 2.83		[203]
28	TiO ₂ /g-C ₃ N ₄	8 W Hg lamp ($\lambda = 254$ nm; intensity = 0.5 mW/cm ²), vol of SS reactor 355 cm ³ , 0.1 g catalyst, 140 kPa CO ₂	CO: 2.8, CH ₄ : 8.5, H ₂ : 41	Bulk g-C ₃ N ₄ CO: 0.93, CH ₄ : 4.75, H ₂ : 16.25	CO: 3		[204]
29	g-C ₃ N ₄ /WO ₃	LED ($\lambda = 435$ nm) at 3.0 mW cm ² , 3 mg catalyst in 5 mL ion-exchanged water	CH ₃ OH: 1.1 μmol , 0.5 wt % Au and Ag 2.5 and 1.5 μmol , resp.	Bulk g-C ₃ N ₄ CH ₃ OH: 0.6 μmol	1.83		[205]
30	g-C ₃ N ₄ /ZnO	300 W Xe lamp, 200 mL Pyrex reactor, 100 mg catalyst CO ₂ and H ₂ O vapor produced by NaHCO ₃ (0.12 g) and HCl (0.25 mL, 4 M)	CH ₃ OH: 0.6	Bulk g-C ₃ N ₄ : CH ₃ OH: 0.26 Pure ZnO: CH ₃ OH: 0.37	2.3		[165]
31	ZnO/g-C ₃ N ₄	500 W Xe/420 nm, steel reactor 132 mL, 10 mg catalyst in 4 mL H ₂ O, 0.4 MPa CO ₂ and 80 °C	CO: 29, CH ₃ CHO: 9, CH ₄ : 3.5, C ₂ H ₅ OH: 1.5	Bulk g-C ₃ N ₄ CO: 4.5, CH ₃ CHO: 4.3, CH ₄ : 0.5, C ₂ H ₅ OH: trace P25CO: 4.5, CH ₃ CHO: 3, CH ₄ : 2, C ₂ H ₅ OH: trace	CO: 6.4		[206]
32	Co-porphyrin/g-C ₃ N ₄	300 W Xe (UV/IR cut-off filter), 1 mL of TEOA and 4 mL of MeCN were mixed and injected into the cell, 80 kPa CO ₂	CO: 17	Bulk g-C ₃ N ₄ CO: 1.4	12.14	0.80 (420 nm)	[207]
33	Co-(bpy) ₃ Cl ₂ /g-C ₃ N ₄	300 W Xe lamp with a 420 nm cut-off, 50 mg catalyst, MeCN (4 mL), TEOA (2 mL), CO ₂ (1 bar), 60 °C	CO: 37 H ₂ : 6		N/A		[176]

Table 2. Cont.

Entry	Photocatalyst	Experimental Details	Productivity/ $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	Reference Material/ $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	Enhancement Relative to Conventional g-C ₃ N ₄	Apparent Quantum Efficiency/%	Reference
34	g-C ₃ N ₄ /Bi ₂ WO ₆	300 W Xe/420 nm cut-off filter, reactor 500 mL, 0.1 g catalyst, CO ₂ and H ₂ O vapour mixer	CO: 5.19	pure g-C ₃ N ₄ CO: 0.23 Bi ₂ WO ₆ CO: 0.81	22		[208]
35	g-C ₃ N ₄ /Bi ₄ O ₅ I ₂	300 W Xe lamp with 400 nm cut-off filter, 0.10 g catalyst, Pyrex glass 350 mL, 5 mL H ₂ SO ₄ (4 M) with NaHCO ₃ to achieve 1 bar CO ₂ , 15 °C	CO: 45.6	Bulk g-C ₃ N ₄ CO: 5.8	7.86		[209]
36	Core-shell LaPO ₄ /g-C ₃ N ₄ nanowires	300 W Xe lamp, reactor volume 500 mL, 30 mg catalyst, CO ₂ and water vapor	CO: 14.43	0.41	10		[210]
37	CdIn ₂ S ₄ /mp g-C ₃ N ₄	300 W Xe lamp with 420 nm cut-off filter, 0.1 g catalyst in 100 mL water containing 0.1 M NaOH, ultrapure CO ₂ was continuously bubbled through	CH ₃ OH: 42.7	pure CdIn ₂ S ₄ CH ₃ OH: 23.1	1.84	0.14 (420 nm)	[211]
38	Mesoporous phosphorylated g-C ₃ N ₄	300 W Xe lamp, Pyrex glass 350 mL, 0.2 g catalyst, 5 mL of 4 M H ₂ SO ₄ with NaHCO ₃ (1.0 g) to give 1 bar CO ₂ 10 °C	CO: 20, CH ₄ : 40, H ₂ : 3, O ₂ : 10	CO: 4.5, CH ₄ : 4, H ₂ : 0.5, O ₂ : 1.75	CH ₄ : 10	0.85 (420 nm)	[212]
39	Pt-g-C ₃ N ₄ /KNbO ₃	300 W Xe lamp with 420 nm cut-off filter, 0.1 g catalyst, CO ₂ , 2 mL of H ₂ O	CH ₄ : 2.37	CH ₄ : 0.62	3.8		[213]
40	g-C ₃ N ₄ /BiOBr/Au	300 W Xe lamp ($\lambda = 380$ nm), 350 mL Pyrex glass, 0.1 g catalyst, 5 mL H ₂ SO ₄ (4 M) + 1.3 g NaHCO ₃ to give 1 bar CO ₂	CO: 6.67 CH ₄ : 0.92	N/A	N/A		[214]
41	g-C ₃ N ₄ /Ag-TiO ₂	300 W Xe, 50 mg catalyst, CO ₂ flow rate of 3 mL·min ⁻¹ , 45 °C	CH ₄ : 9.33 and CO: 6.33	N/A	N/A		[183]

5.2. Environmental Remediation

Many large-scale processes operated by the petrochemical, textile and food industries discharge polluted water into the aquatic environment [215]. Organic dyes are often used in textile, printing, and photographic industries, and a sizable fraction of these are lost during the dyeing process into effluent wastewater streams. Even low concentrations of such dyes pose serious risks to human and animal health, and their bio- or chemical degradation is challenging [216,217], hence the development advanced oxidation processes (AOPs) to treat contaminated drinking ground and surface waters, and wastewaters containing toxic or non-biodegradable compounds are sought [218,219]. Semiconductor photocatalysis offer an effective and economic approach to the treatment of recalcitrant organic compounds at low concentrations in wastewater [220–223]. Photoexcited holes are the key active species in such photocatalytic environmental remediation, being powerful oxidants in their own right, or reacting with water to produce hydroxyl radicals ($\bullet\text{OH}$) which are themselves powerful oxidants with an oxidation potential of 2.8 eV (NHE). Reactively-formed $\bullet\text{OH}$ can rapidly attack adsorbed pollutants at the surface of photocatalysts or in solution, to achieve their mineralization as CO_2 and water. Mechanisms for the photocatalytic oxidation of organic pollutants in water are widely discussed in the literature [4,221,222]. Briefly:



A variety of active radicals, including $\text{O}_2^{\bullet-}$, $\bullet\text{OH}$, HO_2^{\bullet} , in addition to H_2O_2 have been invoked as the oxidants responsible for mineralization, with $\bullet\text{OH}$ the most likely candidate Equation (23). Direct oxidation of carboxylic acids by photoexcited holes to generate CO_2 Equation (24) has also been evidenced, termed the ‘photo-Kolbe reaction’. Reductive pathways involving photoexcited electrons Equation (25) are considered unimportant in dye degradation; however, thermodynamic requirements for semiconductor photocatalysts dictate that the VB and CB should be positioned such that the oxidation potential of hydroxyl radicals $E_{(\text{H}_2\text{O}/\bullet\text{OH})}^0 = +2.8 \text{ eV(NHE)}$ and reduction potential of superoxide radicals $E_{(\text{O}_2/\text{O}_2^{\bullet-})}^0 = -0.3 \text{ eV(NHE)}$ lie well within the band gap. In other words, the redox potential of photoexcited holes must be sufficiently positive to generate $\bullet\text{OH}$ radicals, and that of photoexcited electrons sufficiently negative to generate $\text{O}_2^{\bullet-}$.

Considerable efforts have been devoted to developing photocatalysts for water purification under solar irradiation. g- C_3N_4 based nanostructures are potential photocatalysts for the degradation of various pollutants [39,42], with photophysical properties of the parent nitride modified through

doping with heteroatoms, heterojunction formation with other materials, and textural improvements to enhance surface area and porosity. For example, ultrathin g-C₃N₄ nanosheets derived from bulk g-C₃N₄ by exfoliation in methanol exhibit enhanced photocatalytic performance for methylene blue (MB) degradation [65]. g-C₃N₄ nanotubes show superior photoactivity under visible light for MB degradation than bulk g-C₃N₄ or P25 [74]. Tahir and co-workers also employed tubular g-C₃N₄ for MB and methyl orange (MO) photocatalytic degradation under visible light, observing better stability and activity than bulk g-C₃N₄, attributed to the high surface area (182 m²·g⁻¹) and improved light absorption and charge separation/transfer [75]. 1D g-C₃N₄ nanorods with different aspect ratios have been screened for MB degradation under visible light ($\lambda > 420$ nm) and simulated solar irradiation ($\lambda > 290$ nm) [68]. The resulting photocatalytic activity and photocurrent response of g-C₃N₄ nanorods under visible light were 1.5–2.0 times that of g-C₃N₄ nanoplates. A simple chemical route was reported for preparing nanofiber-like g-C₃N₄ structures which showed promising activity for Rhodamine B (RhB) photodegradation [73].

g-C₃N₄ doping is a common strategy to broaden spectral utilization and band alignment to drive separate photogenerated charge carriers. Doping by metals such as Cu and Fe [224–226], non-metals such as B, C, O, or S [224,227–231], and co-doping [232–234] have all been employed for environmental depollution applications. For example, S and O co-doped g-C₃N₄ prepared by melamine polymerization and subsequent H₂O₂ activation prior to trithiocyanuric acid functionalization (Figure 21a) enhanced the photocatalytic degradation of RhB (Figure 21b) 6-fold relative to the parent g-C₃N₄ nanosheet [235]. Doping resulted in a strongly delocalized HOMO and LUMO that increased the number of active sites and improved the separation of photogenerated electrons and holes.

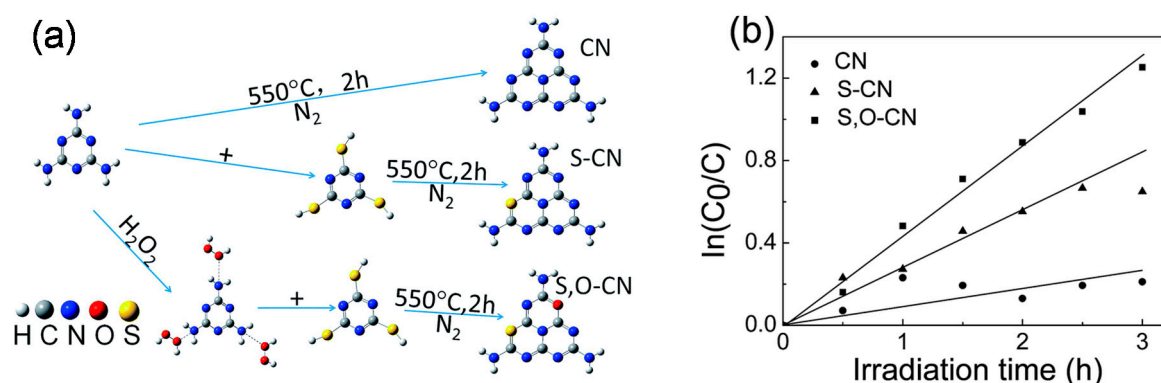


Figure 21. (a) Synthetic strategy, and (b) photocatalytic activity of S and O co-doped g-C₃N₄ for RhB degradation. Reproduced with permission from [235]. Copyright Royal Society of Chemistry, 2017.

Plasmonic photocatalysts have also been exploited for environmental remediation, for example, 7–15 nm Au and Pt nanoparticles photodeposited on g-C₃N₄ are promising for the photocatalytic degradation of tetracycline chloride as a representative antibiotic whose uncontrolled release is of concern [236]. The Au surface plasmon resonance broadens the optical adsorption range, while Pt acts as a sink for photoexcited electrons. The combination of noble metals and g-C₃N₄ enables tunable heterojunctions with improved charge transport than traditional nanocomposites [237–243], and such multicomponent heterostructures are a promising solution to environmental depollution [39,40,42], for example g-C₃N₄/Ag₃PO₄ systems for MO degradation [242,243]. Ag₃PO₄@g-C₃N₄ core-shell photocatalysts have also been applied to MB degradation under visible light, achieving 97% conversion in 30 min compared with only 79% for a physical mixture of the Ag₃PO₄ and g-C₃N₄ components, and 69% for pure Ag₃PO₄. The g-C₃N₄ shell may protect Ag₃PO₄ from dissolution in the composite, conferring superior stability. Core-shell g-C₃N₄@TiO₂ photocatalysts synthesized by a sol-gel and in situ re-assembly route and subsequently applied to phenol removal under visible light were seven times more photoactive than bulk g-C₃N₄. Increasing the g-C₃N₄ shell thickness from 0 to 1 nm

increased the photodegradation rate constant from 0.0018 to 0.0386 h⁻¹; however, thicker shells slowed charge transport to the external photocatalyst surface, lowering activity. Z-scheme N-doped ZnO/g-C₃N₄ hybrid core-shell nanostructures (Figure 22Aa,b) were successfully prepared via a facile, low-cost, and eco-friendly ultrasonic dispersion method [244]. The g-C₃N₄ shell thickness was tuned by varying the g-C₃N₄ loading. Direct contact between the N-doped ZnO core and g-C₃N₄ shell introduced a new energy level into the N-doped ZnO band gap, effectively narrowing the band gap. Consequently, these hybrid core-shell nanostructures showed greatly enhanced visible light photocatalysis for RhB degradation compared to pure N-doped ZnO surface or g-C₃N₄ components (Figure 22Ac) [240]. A facile, reproducible, and template-free synthesis has also been demonstrated to prepare magnetically separable g-C₃N₄-Fe₃O₄ nanocomposites (Figure 22Ba) [37]. Monodispersed Fe₃O₄ nanoparticles with 8 nm diameter were uniformly deposited over g-C₃N₄ sheets (Figure 22Bb) and exhibited enhanced charge separation and photocatalytic activity for RhB degradation under visible light irradiation (Figure 22Bc). These g-C₃N₄-Fe₃O₄ nanocomposites showed good stability with negligible loss in photocatalytic activity even after six recycles, and facilitated magnetic catalyst recovery (Figure 22Bd). Xiao et al. demonstrated that the excellent stability of g-C₃N₄ towards photocatalytic oxidation in the presence of organic pollutants reflects strong competition of the latter for •OH radicals under practical working conditions, resulting in preferential decomposition of the pollutants rather than the carbon nitride [245].

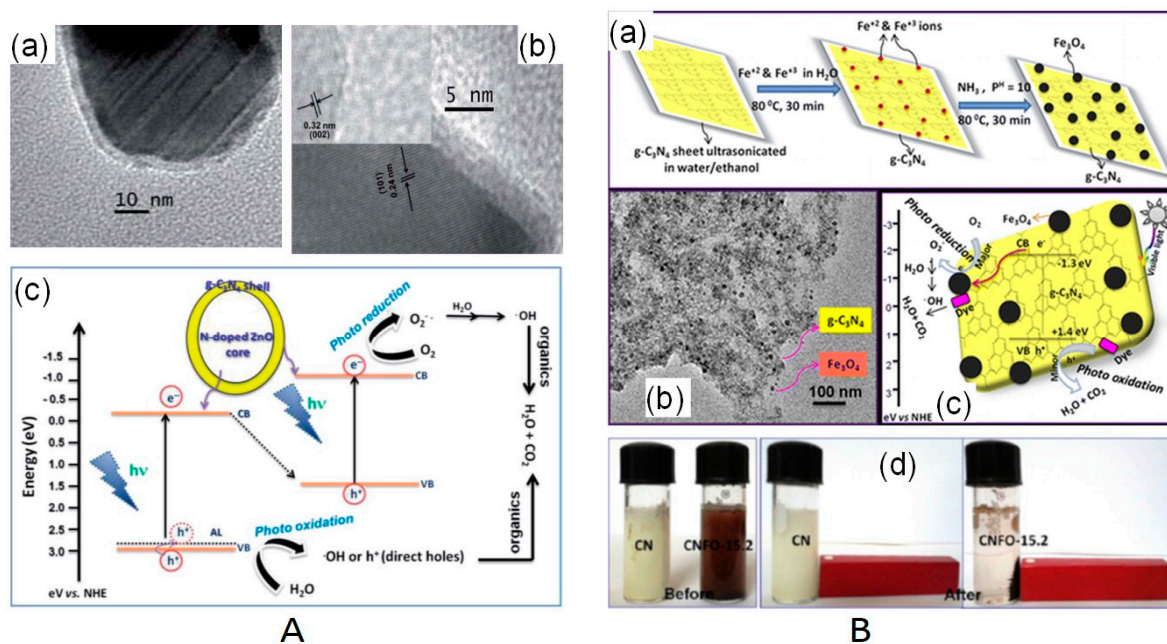


Figure 22. (A) (a,b) TEM images of N-ZnO-g-C₃N₄ core-shell nanoplates, and associated (c) Z-scheme mechanism. Reproduced from with permission from [244]. Copyright 2014 Royal Society of Chemistry. (B) (a) Synthetic strategy, (b) TEM image, and (c) photodegradation mechanism for g-C₃N₄-Fe₃O₄ nanocomposite, and (d) magnetic separation of photocatalyst post-reaction. Reprinted with permission from [37]. Copyright 2013 American Chemical Society.

Several multicomponent nanocomposites based on g-C₃N₄ nanosheets such as Au@g-C₃N₄-PANI [246], Au-NiYF/g-C₃N₄ [105], g-C₃N₄/CNTs/Al₂O₃ [247], AgCl/Ag₃PO₄/g-C₃N₄ [248], and g-C₃N₄/Zn_{0.11}Sn_{0.12}Cd_{0.88}S_{1.12} [249] are also reported; the performance of different g-C₃N₄ photocatalysts for the photodegradation of representative aqueous pollutants is summarized in Table 3.

Table 3. Photocatalytic degradation of aqueous pollutants over g-C₃N₄ nanostructured catalysts.

Entry	Photocatalyst	Organic Molecule	Experimental Details	Removal Efficiency/%	Reference Material Efficiency/%	Enhancement Relative to Conventional g-C ₃ N ₄	Reference
1	g-C ₃ N ₄ @TiO ₂ core-shell structure	Phenol	5 mg·L ⁻¹ phenol with 25 mg catalyst. 500 W Xe lamp with 420 nm cut-off filter, 23 mW/cm ² .	30	4.2	7.2	[250]
2	Ag-decorated S-doped g-C ₃ N ₄	Bisphenol A (BPA)	50 mL of 10 mg·L ⁻¹ of BPA, catalyst loading of 0.60 g·L ⁻¹ . Light source, 155 W Xe arc lamp with the solar region of 280–630 nm.	95	31.66	3	[233]
3	Ultrathin urea-derived g-C ₃ N ₄ nanosheets	p-Nitrophenol (PNP)	100 mg catalyst, aqueous PNP (10 mg L ⁻¹ , 100 mL). 300 W Xe lamp equipped with an IR cut filter and a 400 nm cut filter.	95	60	1.58	[251]
4	Mesoporous g-C ₃ N ₄ /TiO ₂	Decomposition of dinitro butyl phenol (DNBP)	25 mg catalyst added to DNBP aqueous solution (20 mg·L ⁻¹) with 500 W xenon lamp with λ < 420 nm using cut-off filter.	98.5	65	1.5	[252]
5	C ₃ N ₄ -nanosheets	Methylene blue (MB)	10 mg catalyst in 50 mL of 10 mg·L ⁻¹ MB solution. 150 W Xe lamp as the simulated sunlight source.	98	7.9	12.4	[253]
6	Z-scheme graphitic-C ₃ N ₄ /Bi ₂ MoO ₆	Methylene blue	30 mL of 10 mg·L ⁻¹ MB solution, 0.03 g catalyst. 50 W LED light with of 410 nm emission.	90	18.75	4.8	[254]
7	Sm ₂ O ₃ /S-doped g-C ₃ N ₄	Methylene blue	100 mL of MB solution (8 mg·L ⁻¹), 300 W halogen lamp with UV-stop feature.	93	27	3.5	[255]
8	Porous CeO ₂ /sulfur-doped g-C ₃ N ₄	Methylene blue	0.06–0.12 g catalyst in 6–14 mg L ⁻¹ MB, visible light (λ > 400 nm) 300 W Halogen lamp with UV stop.	91.4	25	3.65	[256]
9	ZnS/g-C ₃ N ₄	Methylene blue	200 mL MB (6 mg·L ⁻¹), 30 mg catalyst under visible light source, 100 W halogen lamp.	90	34.6	2.6	[257]

Table 3. Cont.

Entry	Photocatalyst	Organic Molecule	Experimental Details	Removal Efficiency/%	Reference Material Efficiency/%	Enhancement Relative to Conventional g-C ₃ N ₄	Reference
10	Mesoporous Carbon Nitride Decorated with Cu Particles	Methyl orange (MO)	0.07 g catalyst in 100 mL of MO (11 mg L ⁻¹) solution under visible-light, 300 W halogen lamp with UV-stop feature.	100	28	3.57	[258]
11	Plasmonic Ag-AgBr/g-C ₃ N ₄	Methyl orange	MO solution (100 mL, 10 mg L ⁻¹), 50 mg catalyst, 300 W Xe lamp with 400 nm cut-off filter.	90	14.3	6.3	[259]
12	ZnFe ₂ O ₄ nanoparticles on g-C ₃ N ₄ sheets	Methyl orange	100 mL of 10 mg·L ⁻¹ MO solution, 25 mg catalyst. 500 W Xe lamp with cold filter.	98	15.31	6.4	[260]
13	AgNPs/g-C ₃ N ₄ nanosheets	Methyl orange	50 mL 0.02 mmol/L MO solution, 25 mg catalyst. 300 W Xe lamp with a visible light reflector (350 nm < l < 780 nm) and a 420 nm longwave-pass cut-off filter (l > 420 nm).	95.2	13.8	7	[261]
14	BiOCl/C ₃ N ₄ hybrid nanocomposite	Methyl orange	15 mL of 10 mg L ⁻¹ MO solution, 10 mg catalyst. 300 W Xe lamp equipped with 420 nm cut-off filter.	84.28	14	6	[262]
15	g-C ₃ N ₄ /GO aerogel	Methyl orange	50 mL of 20 mg L ⁻¹ MO solution. 300 W Xe lamp with a cut off filter (λ > 420 nm).	91.1	33	2.76	[263]
16	g-C ₃ N ₄ nanocrystals decorated Ag ₃ PO ₄ hybrids	Methyl orange	80 mL MO, 80 mg catalyst. 500 W halogen lamp equipped with cut-off filters (420 nm < λ < 800 nm).	92	44	2	[264]
17	g-C ₃ N ₄ -NS/CuCr ₂ O ₄ nanocomposites	Rhodamine B (RhB)	250 mL of 2.5 × 10 ⁻⁵ M RhB solution, 0.1 g of catalyst. 50 W LED lamp.	98.9	30	3.3	[265]
18	Porous Mn doped g-C ₃ N ₄	Rhodamine B	100 mL of 10 mg·L ⁻¹ RhB solution, 50 mg catalyst. 300 W Xe lamp equipped with ultraviolet cut-off filter (>400 nm).	88.9	18	4.9	[266]
19	Mesoporous carbon nitride (mpg-C ₃ N ₄ /SnCoS ₄)	Rhodamine B	100 mL of 20 mg·L ⁻¹ RhB solution, 20 mg catalyst. 300 W Xe lamp equipped with an UV cut-off filter (λ ≥ 420 nm).	70	13	5.4	[267]

Table 3. Cont.

Entry	Photocatalyst	Organic Molecule	Experimental Details	Removal Efficiency/%	Reference Material Efficiency/%	Enhancement Relative to Conventional g-C ₃ N ₄	Reference
20	Iron oxyhydroxide/ultrathin g-C ₃ N ₄ nanosheets	Rhodamine B	50 mL of 10 mg·L ⁻¹ RhB solution, 50 mg catalyst. 500 W Xe lamp equipped with a cut-off filter ($\lambda \geq 420$ nm).	98	5.5	17.8	[268]
21	Two-dimensional g-C ₃ N ₄ /Bi ₂ WO ₆	Rhodamine B	100 mL of 10 mg L ⁻¹ RhB solution, 100 mg catalyst. 300 W Xe lamp with UV cut-off filter.	80	23.5	3.4	[269]
22	Ultrathin g-C ₃ N ₄ nanosheets	Rhodamine B	100 mL of 20 mg L ⁻¹ RhB solution, 100 mg catalyst. 300 W Xe lamp (>420 nm).	99	16.2	6.1	[270]
23	Z-scheme g-C ₃ N ₄ /TiO ₂ nanotube	Rhodamine B	20 mL of 5 mg·L ⁻¹ RhB solution, 2 cm × 2 cm catalyst film. 300 W Xe lamp with UV cut-off filter.	67	47.85	1.4	[271]
24	WO ₃ @g-C ₃ N ₄	Rhodamine B	50 mL of 10 mg L ⁻¹ RhB solution, 10 mg catalyst. Xe lamp with 400 nm cut-off filter, 100 mW cm ⁻² .	90	25.7	3.5	[272]
25	Mesoporous graphitic carbon nitride modified PbBiO ₂ Br	Rhodamine B	100 mL of 10 mg·L ⁻¹ RhB solution, 30 mg catalyst. 300 W Xe lamp with UV cut-off filter (>400 nm).	98	N/A	N/A	[273]
26	g-C ₃ N ₄ /CuS p-n heterojunctions	Rhodamine B	30 mL of 10 mg L ⁻¹ RhB solution, 10 mg catalyst. 300 W Xe lamp with 420 nm cut-off filter.	93	27	3.5	[274]
27	g-C ₃ N ₄ /kaolinite composites	Rhodamine B	100 mL of 10 ppm RhB solution, 200 mg catalyst. 500 W Xenon lamp with 400 nm cut-off filter.	90	21.8	4.1	[275]
28	Hexagonal boron nitride (h-BN) decorated g-C ₃ N ₄	Rhodamine B	100 mL of 20 mg L ⁻¹ RhB solution, 50 mg catalyst. 300 W Xe lamp with 420 nm cut-off filter.	99.5	13.63	7.3	[276]
29	ZnO/g-C ₃ N ₄	Rhodamine B	50 mL of 10 mg L ⁻¹ RhB solution, 50 mg catalyst. 500 W Xe lamp equipped with 420 nm cut-off filter.	51.3	24.43	2.1	[277]
30	Ag/AgO loaded g-C ₃ N ₄ microspheres	Acid Violet-7 (AV-7)	100 mL of 20 mg·L ⁻¹ AV-7 solution, 100 mg catalyst. 12 × 100 W fluorescent lamps (mainly visible light, with only 3% UV).	98	48	2	[278]

Table 3. Cont.

Entry	Photocatalyst	Organic Molecule	Experimental Details	Removal Efficiency/%	Reference Material Efficiency/%	Enhancement Relative to Conventional g-C ₃ N ₄	Reference
31	g-C ₃ N ₄ /TiO ₂ /kaolinite composite	Ciprofloxacin (CIP) antibiotic	100 mL of 10 ppm CIP solution, 200 mg catalyst. Xe lamp (90 mW/cm ²) with 400 nm cut-off filter.	92	14.48	6.4	[279]
32	Z-scheme CdS/Fe ₃ O ₄ /g-C ₃ N ₄	Ciprofloxacin	100 mL of 20 mg L ⁻¹ CIP, 50 mg photocatalyst. 300 W Xe lamp with UV filter ($\lambda > 420$ nm).	92	3.53	26	[280]
33	Carbon-Doped g-C ₃ N ₄	Tetracycline (TC)	80 mL of 10 ⁻⁴ M TC, 40 mg catalyst. Sunlight (07/10/2015, Trivandrum, India, between 11 pm and 1 pm, 78,000–80,000 lux).	95	50	1.9	[231]
34	Phosphorous-doped ultrathin graphitic carbon nitride nanosheets	Tetracycline	100 mL of 10 mg·L ⁻¹ TC solution, 100 mg catalyst. 300 W Xe lamp equipped with UV cut-off filter (>420 nm).	96.95	71.78	1.35	[281]
35	Hierarchical WO ₃ /g-C ₃ N ₄	Tetracycline hydrochloride (TC-HCl)	100 mL of 25 mg·L ⁻¹ TC-HCL solution, 50 mg catalyst. 300 W Xe lamp with 420 nm cut-off filter.	82	48	1.7	[282]
36	Co ₃ O ₄ modified g-C ₃ N ₄	Diclofenac sodium (DCF)	100 mL of 10 mg·L ⁻¹ DCF solution, 50 mg catalyst. 300 W Xe lamp with 420 nm cut-off filter.	100	17	5.9	[283]
37	silver and carbon quantum dots co-loaded with ultrathin g-C ₃ N ₄	Naproxen NPX	50 mL of 4 mg·L ⁻¹ NPX solution, 50 mg catalyst. 350 W Xe lamp with 420 nm and 290 nm light for visible and simulated sunlight sources.	87.5	8.75	10	[284]
38	g-C ₃ N ₄	Decabromodiphenyl ether (BDE209)	20 mL of 1 × 10 ⁻³ mol/L BDE209 solution, 20 mg catalyst. 300 W Xe lamp for UV-visible irradiation (>360 nm).	65	N/A	N/A	[285]
39	Metal-free sulfur doped g-C ₃ N ₄	UO ₂ ²⁺ removal	200 mL of 0.12 mM UO ₂ ²⁺ solution, 100 mg catalyst. 350 W Xe lamp with a 420 nm cut-off filter.	95	71	1.3	[286]

6. Conclusions

g-C₃N₄ nanostructures offer tunable textural, electronic and optical properties that are amenable to tailoring for solar energy harvesting and subsequent photocatalytic transformations for energy and environmental applications. Diverse synthetic methods are available to prepare pure g-C₃N₄ nanostructures of different dimensionality and porosity, and to integrate these within multi-functional nanocomposites with enhanced solar spectral utilization, apparent quantum yields, charge separation and transport, and ultimately photocatalytic activity and stability. The sustainable production of H₂ as an energy vector from water splitting is perhaps the most promising application, although issues remain regarding the use of sacrificial reagents and a lack of interdisciplinary efforts to improve photoreactor design. Photocatalytic reduction of CO₂ is at a more preliminary stage, with improvements in both activity, and the ability to select specific products for either energy (e.g., CO, CH₄, methanol, and formic acid) or chemicals (e.g., >C₂ olefins or alkanes) pre-requisites to bench scale demonstrations. Wastewater treatment using g-C₃N₄-based photocatalysts appears promising; however, a lack of standardization in either reactor design or experimental protocols hampers quantitative comparisons due to issues such as decoupling adsorption versus reaction, and photocatalysis from direct photochemical activation of chromophores.

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