Chapter 10

Structural Modifications of Carbon Nitride for Photocatalytic Applications

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Abstract

The current research on photocatalysis is totally focused on the designing and innovation of various low cost materials. For an efficient photocatalyst, there are some aspects which are to be assessed before practical use, such as optical activity, thermal and chemical stability, easy and availability of raw material, biocompatibility, etc. Fortunately, g-C₃N₄ offers most of these qualities to behave as a star photocatalyst. g-C₃N₄ could be easily prepared from low cost precursor materials such as urea, melamine, cyanimide and dicyandiamide by simple thermal treatment. Furthermore, larger surface area and two-dimensional planar conjugation structure of g-C₃N₄ can provide a large platform for anchoring various substrates. Various researchers have utilized g-C₃N₄ for varieties of applications such as green energy production, energy storage devices, biomedical application, wastewater treatment via photocatalysis and adsorption, photo sensors, etc. Although there are some disadvantages associated with use of g-C₃N₄ when utilized for various applications. To overcome such hitches various structural modifications have been applied to g-C₃N₄. The current chapter summarizes a wide mode of applications of g-C₃N₄ along with various structural modifications which were recently applied to improve the photocatalytic efficacy.

Keywords

g-C₃N₄, Modification, Energy, Wastewater, Photocatalysts

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1. Introduction

The current era is demanding clean energy production and environmental remediation. Various aspects of environment and health related concern have triggered the research towards exploring novel hybrid materials which can be utilized and address these issues without altering the environment quality. Photocatalysis is a greener approach that has proven to be cost effective and highly efficient in the field of clean energy production, wastewater treatment, air pollution remediation, catalytic process, etc.[1–4]. But efficacy and cost effectiveness of photocatalysis process is extensively dependent on the material that is being utilized as “photocatalyst”. Now these photocatalysts are the key material as well as center of curiosity and research which attract attention of the worldwide scientist’s community. Various metal based and metal free photocatalysts have been explored for the varieties of photocatalytic processes [5]. However, metal based and metal free photocatalysts have their own limits of advantages and disadvantages e.g. Ag, Au, Pt, Ru, etc. based photocatalysts are highly active but their expensive nature limits their exploration for large scale applications [6]. At the same time, metal free photocatalysts such as g-C₃N₄, reduced graphene oxide (RGO), graphene oxide (GO), carbon-quantum dots (CQDTs) and other organic photocatalysts offer an alternate material for being utilized for various photocatalytic applications [7–10]. However chemical, thermal stability and synthesis strategies are various parameters on which the efficacy of a metal free photocatalysts depends.

1.1 g-C₃N₄ as tremendous metal-free photocatalysts

g-C₃N₄ is a Nobel metal-free, stable, nontoxic and inexpensive polymeric photocatalyst [11]. g-C₃N₄ holds many advantages, such as good thermal and chemical stability, metal-free and tunable structure which are the essential requirements. The abundance and low cost of its precursor material such as urea, melamine, cyanimide and dicyandiamide make it approachable for the entire researchers to explore it for high quality research. Furthermore, larger surface area and 2-D planar structure of g-C₃N₄ can provide a large sites for anchoring various substrates [12]. g-C₃N₄ is quite stable up to 600 °C and starts decomposing when temperature is above 700 °C [13]. Due to strong interlayer stacking interaction g-C₃N₄ is almost insoluble in solvents like water, ethanol, methanol, THF, diethyl ether and toluene [14].

1.2 Chemical structure of g-C₃N₄

The structure of g-C₃N₄ consists of tri-s-triazine of units which are interconnected with each other with tertiary amine [15] units as referred with figure 1. It possesses the π -conjugated graphitic planes which are formed due the sp^2 hybridization between the carbon and nitrogen atom. The band gap energy for g-C₃N₄ is calculated to be ≈ 2.7 eV which corresponds to the absorption of wavelength ≈ 460 nm.

Figure 1: Chemical structure of g-C₃N_{4q}.

1.3 Band structure of g-C₃N₄

The lone pair of electrons on nitrogen is supposed to be contributing for band structure of g-C₃N₄. The overlap of 2p orbitals of N together with the overlap carbon 2p orbitals are mainly depot to VB and CB of the g-C₃N₄ [16].

Figure 2: Band structure of g-C₃N₄.

The extra stability for lone pairs of nitrogen has been provided by π -bonding states which are present in conjugation. Therefore, the lone pair electron of nitrogen plays a key role in structure of g-C₃N₄ [17]. Zhang et al. reported that the CB edge and VB edge lies at -1.3 and 1.4 eV (vs NHE) respectively [18] which fulfills the requirements of reduction and oxidation reaction directed by sun light. The band structure of g-C₃N₄ is shown in Fig. 2. The CB & VB edge potential of g-C₃N₄ located at sufficient potentials which enable it for various photo reduction and oxidation reactions.

2. Synthesis approaches via thermal annealing/Pyrolysis

Thermal annealing is the most common and popular technique used for any modification for the fabrication of template-based and template-free g-C₃N₄. Various precursor materials such as urea, melamine, cyanamide and dicyandiamide etc. are thermally treated at high temperature (400-700 °C) with a heating rate of 2 to 5 °C min⁻¹ [19]. Thermal treatment allows the precursor materials to condense into the polymeric form of tri-s-triazine units as shown in fig. 3.

It has been reported that when g-C₃N₄ is prepared in bulk it possesses comparatively low surface area (< 20 m²g⁻¹) [4]. Thus various effective strategies have been employed to exfoliate thin layer of g-C₃N₄ such as oxidation exfoliation [20], ultrasonic liquid exfoliation [21] chemical exfoliation [22] etc. The exfoliated thin sheets of g-C₃N₄ have high surface area as compared to bulk g-C₃N₄. The condensation of precursor materials such as urea, thiourea and melamine have been represented in fig. 4.

Figure 3: Method of thermal condensation.

Figure 4. Proposed thermal condensation of various precursor materials to g-C₃N₄.

3. Strategies followed for the modification of g-C₃N₄

However, g-C₃N₄ has been continuously explored for various photocatalytic applications but there are still some shortcomings and bottlenecks which have to be dealt with, such as rapid charge recombination rate, slightly low absorption in visible spectrum, etc. [23]. To overcome such limitations various modifications have been carried out in g-C₃N₄ to effectively increase the efficiency.

3.1 Doping

Doping is a techniques which focusses on deliberately introducing impurities to the core structure of a photocatalyst is known to be an adequate approach to tune the band gap of g-C₃N₄, which appreciably broadens the light absorption range and reduces the photogenerated charge recombination rate too [24]. Doping has been carried out either by interstitial or substitutional to modify texture and electronic structure of g-C₃N₄ for enhancing the photocatalytic activity [25].

3.2 Metal/non-metal based doping

Doping with metals and non-metals has proven to be an effective approach to increase photo-activity in visible spectrum. Doping not only narrows the band gap but efficiently

increases the active sites on the surface of host material. Various metal based doping experiments have been carried out to increase photocatalytic performance of g-C₃N₄. Table 1 & 2 refer to the scientific work carried out in recent years concerning the doping of g-C₃N₄ for enhanced activity.

Table 1: Metal based doping in g-C₃N₄ along with efficiency.

Sr.no	Dopant	Performance	Ref
1.	Fe	Apparent quantum efficiency of 78.5% for Photocatalytic water splitting	[26]
2	Co	Exhibits 11 times in rhodamine B degradation than that of pure g-C ₃ N ₄	[27]
3	Ti ³⁺	Exhibits 7.6 times higher degradation efficiency for methylene blue h	[28]
4	Fe	1.7 & 6 times higher efficiency for MB degradation and H ₂ production respectively.	[29]
5	Mn	Photocatalytic activity for Cr(VI) reduction and RhB degradation increased to 76.5% & 8.9% respectively	[30]
6	K	Efficiency increased to 5.6 times for photocatalytic hydrogen evolution	[31]
7	Cu	High efficiency for H ₂ -Evolving Electro catalysis	[32]
8	Ag	Two times higher efficiency for photocatalytic oxidation of methylene blue	[33]
9	Zr	High efficiency for the RhB degradation	[34]
10	Y	Higher degradation efficiency for RhB degradation	[35]
11	Eu(III)	Degradation efficiency of RhB increased to 1.71 fold	[36]
12	Pd	15.3 times higher efficiency toward Hydrogen Evolution Reaction	[37]

Table 2: Non-metal based doping in g-C₃N₄ for better photocatalytic efficiency.

Sr.no	Dopant	Performance	Ref
1.	N	4.3 times higher efficiency for enhanced photocatalytic H ₂ evolution	[38]
2	N	Enhanced efficiency for tetracycline degradation	[39]
3	S	2.5 fold higher efficiency for photocatalytic CO ₂ -reduction performance	[40]
4	S	Photoreduction of CO ₂ to acetaldehyde and methane increases to 8.3 & 9 times	[41]
5	P	High photocatalytic efficiency for H ₂ production	[42]
6	P	High efficiency for photocatalytic oxidation of aromatic alcohols	[43]
7	P	2.9 times higher efficient for photocatalytic activity for H ₂ evolution and Rhodamine B degradation	[44]
8	O	5 times higher Photocatalytic CO ₂ Reduction activity	[45]
9	O	2.5 times higher H ₂ evolution photocatalytic activity	[46]
10	B	2.4 times higher efficiency for photocatalytic H ₂ evolution	[47]
11	B	17 times higher photo degradation efficiency for MB dye	[48]
12	C-I	9.8 times higher efficiency for photocatalytic hydrogen evolution	[49]

3.3 Fabrication of g-C₃N₄ based binary/ternary heterojunctions

g-C₃N₄ based binary/ternary heterojunctions offer propitious and worthwhile strategies to harness solar radiation which sufficiently overcomes the disadvantages associated with other modification strategies. Over the last decades, various g-C₃N₄ based binary/ternary heterojunctions have been successfully fabricated by keeping primary concern such as light response, appropriate lowering in band gap energies, reduced e⁻/h⁺ pair charge recombination, etc. [50].

Figure 5: Band structure for the g-C₃N₄ based binary and ternary hetero-structures.

Fig.5 refers to the band structure and effect on photoluminescence when junction formation takes place between g-C₃N₄ and other semiconductors in various based binary and ternary hetero-structures. However, narrowing of band gap for g-C₃N₄ is not the sole concern to boost its photo activity, no doubt that narrow band gap semiconductors can absorb visible light more efficiently but additionally they suffer from the complications such as high rate of charge recombination, photo corrosion, low quantum yield etc. All these disadvantages are mainly associated with single-component photocatalysts which can be tackled by fabricating the heterojunction of different semiconductors which not only improves the photo-activity but also increase the surface area and life time of photo induced charge carriers [51,52].

Table 3: Binary coupled modification of g-C₃N₄ along with utilization and efficiency.

Sr no	Binary coupled composite	Enhanced photocatalytic activity evaluated by	Efficiency	Ref
1	WO ₃ /g-C ₃ N ₄	RhB degradation	91.3% degradation after 150 min	[59]
2	LaNiO ₃ /g-C ₃ N ₄	Tetracycline (TC) degradation.	3.9 and 3.8 times larger than those of sole components respectively	[60]
3	g-C ₃ N ₄ /BiVO ₄	RhB degradation	2.3 times higher than that of the pure g-C ₃ N ₄	[50]
4	(SiC)/g-C ₃ N ₄	Degradation of dyes rhodamine B (RhB) and methyl orange (MO)	1.2 time higher than sole g-C ₃ N ₄	[61]
5	g-C ₃ N ₄ /gC ₃ N ₄	Removal of NO at ppb levels	47.6% high as compared to host substrates	[62]
6	CoTiO ₃ /g-C ₃ N ₄	Photocatalytic H ₂ -evolution	~ 2 times higher than that of the pure g-C ₃ N ₄ under artificial sunlight illumination	[63]
7	Bi ₂ O ₂ CO ₃ /g-C ₃ N ₄	Photocatalytic oxidation removal of NO at ppb levels	15.4 times higher rate for NO removal	[64]
8	TiO ₂ /g-C ₃ N ₄	Potocatalytic reduction of CO ₂ and photocatalytic decomposition of N ₂ O	57% N ₂ O conversion in 16 h	[65]
9	g-C ₃ N ₄ /Bi ₂ O ₂ CO ₃	rhodamine B degradation	3.1 times higher than that of individual C ₃ N ₄	[66]
10	Co ₃ O ₄ -g-C ₃ N ₄	degradation of methyl orange	7.5-folds higher than that of pure g-C ₃ N ₄	[67]
11	BiOCl-C ₃ N ₄	degradation of methyl orange (MO)	36.2 times higher than sole g-C ₃ N ₄	[68]
12	g-C ₃ N ₄ /Bi ₂ MoO ₆	Degradation of Rhodamine B	3 times higher those of pure g-C ₃ N ₄	[69]
13	ZnS/g-C ₃ N ₄	photocatalytic H ₂ evolution	30 times higher photocatalytic H ₂ evolution rate than that of pure g-C ₃ N ₄	[70]
14	α-Fe ₂ O ₃ /g-C ₃ N ₄	Photocatalytic CO ₂ conversion	2.2 times higher	[56]
15	Ag ₂ O/g-C ₃ N ₄	Degradation of Rhodamine B (RhB), phenol, imidacloprid, methylene blue (MB) and methyl orange (MO) under visible and NIR light irradiation	RhB degradation is about 26 and 343 times higher than that of pure g-C ₃ N ₄ in visible and NIR respectively	[71]

Table 4: Ternary coupled modification of g-C₃N₄ along with utilization and efficiency.

Sr no	Ternary coupled composite	Enhanced photocatalytic activity evaluated by	Efficiency	Ref
1	g-C ₃ N ₄ /ZnO@ α -Fe ₂ O ₃	Photodegradation of tartrazine	6.8 folds	[72]
2	g-C ₃ N ₄ @Bi/BiOBr	Photo degradation of tetracycline and rhodamine B	Highest efficiency 98%.	[73]
3	C ₃ N ₄ /ZnS/SnS ₂	Photodegradation of methylene blue (MB)	8.74 times higher	[74]
4	BiOCl/ CdS/g-C ₃ N ₄	Degradation of rhodamine B (RhB)	12 times higher compared to bared g-C ₃ N ₄	[75]
5	g-C ₃ N ₄ /Fe ₃ O ₄ /MnWO ₄	Rhodamine B, methylene blue, methyl orange, and fuchsine removal	7, 10, 25, and 31 time as compared to g-C ₃ N ₄ respectively	[76]
6	g-C ₃ N ₄ /Ag ₃ VO ₄ /AgBr	RhB degradation	116 times higher	[77]
7	g-C ₃ N ₄ /Ag ₃ PO ₄ /Ag ₂ MoO ₄	Solar oxygen evolution from water splitting	2.2 times higher	[78]
8	CdS/g-C ₃ N ₄ /CuS	photocatalytic H ₂ -production activity	5 times higher	[79]
9	g-C ₃ N ₄ /Ag/MoS ₂	Organic pollutant degradation and production of H ₂	9.43 & 8.78 time higher for RhB degradation and H ₂ production respectively	[80]
10	g-C ₃ N ₄ /CeO ₂ /ZnO	Methylene blue (MB) degradation.	11.46 times higher	[81]
11	ZnO-Ag ₂ O/pg-C ₃ N ₄	Degradation of ciprofloxacin	3.83 times higher	[82]
12	C ₃ N ₄ /Fe ₃ O ₄ /BiOI	Degradation of RhB	21-fold higher	[83]
13	g-C ₃ N ₄ /Fe ₃ O ₄ /Ag ₂ CrO ₄	Degradation of rhodamine B	6.3 folds higher	[84]
14	g-C ₃ N ₄ /Fe ₃ O ₄ /AgI	Degradation of RhB	8.7 times higher	[85]
15	g-C ₃ N ₄ /Fe ₃ O ₄ /Ag ₃ VO ₄	Degradation of RhB	14 fold higher	[86]

Various strategies has been approached for fabrication of g-C₃N₄ based binary/ternary heterojunctions such as hydrothermal/solvothermal, electrodeposition process, chemical bath deposition, sol-gel process, chemical precipitation, self-assembly method etc. [53]. Over the last decades, g-C₃N₄ based binary/ternary heterojunction are being extensively used in the various fields such as photodegradation and mineralization of noxious pollutants [54], water splitting into H₂ [55] CO₂ conversion into fuels [56], photo-treatment of some harmful metals Cr(VI), As(III), U(VI) [57,58] etc. Table 3 and 4 referred

previously fabricated binary and ternary coupled heterojunction utilized in the various filed to evaluate their photoactive potential.

Very recently Kumar et al., 2018, fabricated various g-C₃N₄ based nano composite such as biochar supported g-C₃N₄/FeVO₄ [54], coal char/g-C₃N₄/RGO [87] for various environmental applications etc. fig. 6 a & b. referred the XRD and photoluminescence spectra for the g-C₃N₄/FeVO₄. The intimate contact between both of semiconductors and formation of hetro-junction has been shown by XRD result. Furthermore, the PL spectra (referred in fig. 6 b) indicated the reduced charge recombination rate confirmed by the absence of any sharp peak in the composite part.

Figure 6: (a) XRD, and (b) Photoluminescence of g-C₃N₄/FeVO₄ (Reproduced with permission from [54] copyright Elsevier).

Figure 7: (a) XRD, and (b) Photoluminescence of RPC (Reproduced with permission from [87] copyright RSC).

Fig. 7 a & b refers to the XRD spectra of RPC (coal char/g-C₃N₄/RGO) as fabricated by Kumar et al. The XRD confirms the junction formation between the reduced graphene oxide and g-C₃N₄ on the coal char surface. The increased intensity at 27.2° for (002) graphitic plane observed due to increased staking of conjugation present in aromatic system of RGO and g-C₃N₄ [88]. The PL spectrum of RPC shows absence of sharp peak owing to successful formation of heterojunction between RPC & g-C₃N₄.

3.4 Coupling with metal-free substrates

Introduction of metal free substrate is a novel strategy employed by various researchers owing to their cost effective, ecofriendly nature and abundance of precursor materials. Modification of g-C₃N₄ with carbonaceous materials based motif has proven to be an efficient approach to alter its optical properties [89]. However g-C₃N₄ possesses relatively negative conduction band potential that can able to reduce many pollutants, such as Cr (VI), CO₂, and organic dyes but shows lower oxidation and mineralization activities [90,91].

Figure 8: Carbonaceous material as electron sink.

Incorporation of metal free components such as GO, RGO, polyaniline and activated carbon may enhance the conductivity, strength and surface area of as fabricated g-C₃N₄ based photo catalysts [92].

Recently Chen et al., 2016 [93], has synthesized g-C₃N₄/activated carbon composite (g-C₃N₄/AC) by in-situ thermal condensation method. The activity of fabricated composite

was evaluated by photodegrading phenol under the visible light radiation. A complete removal of phenol was achieved after 160 min of photodegrading experiment. Fig. 8 shows the route for charge transfer from g-C₃N₄ surface to the carbonaceous material such as RGO, GO etc. under photoexcitation. Some recent metal-free modification that was done on g-C₃N₄ by various researchers has been referred in table 5.

Table 5: *Coupling with Metal-free component modified g-C₃N₄ along with utilization and efficiency.*

Sr. no	Metal-free photocatalysts	Enhanced photocatalytic activity evaluated by	Efficiency	ref
1	g-C ₃ N ₄ /rGO	Degradation of rhodamine B (RhB) and 4-nitrophenol under visible light irradiation	3.0 and 2.7 times higher respectively	[88]
2	g-C ₃ N ₄ /rGO	Photocatalytic degradation of rhodamine B	2.6 times that of pristine g-C ₃ N ₄	[94]
3	2D RGO/pCN	Photocatalytic reduction of carbon dioxide to methane	5.4 folds higher	[95]
4	rGO/ g-C ₃ N ₄	Photocatalytic H ₂ -evolution performance	-----	[96]
5	PANI/g- g-C ₃ N ₄	Degradation of methylene blue	5.1-fold higher than that of pure g-C ₃ N ₄	[97]
6	g-PAN/g-C ₃ N ₄	Photocatalytic H ₂ -evolution	3.8 times over pristine g-C ₃ N ₄	[98]
7	g-C ₃ N ₄ /Biochar	Removal of cationic dye methylene blue (MB)	-----	[99]
8	g-C ₃ N ₄ /GO	Degradation of MO and MB and photo reduction of bromate	-----	[100]
9	SiO ₂ /g-C ₃ N ₄	Degradation of rhodamine B	3.5 times higher	[101]
10	Red phosphor/ g-C ₃ N ₄	Photocatalytic activity for H ₂ production and CO ₂ conversion	10 timers higher	[102]
11	C-Dots/ g-C ₃ N ₄	Photocatalytic hydrogen H ₂ production and RhB degradation	3 times higher	[103]
12	CNNS/CQDs	Photocatalytic generation of hydrogen	3 times higher	[104]

3.5 Chemical modification

Chemical modification of g-C₃N₄ surface is another technique to improve its photocatalytic activity. Introduction of some groups on the surface of g-C₃N₄ increase the active sites which further improves the interaction with the employed substrate. Dong G et al. 2016 [105] use perylene imides modified g-C₃N₄ for the removal of noxious NO and they found

this composite 5 times efficient for the photo generation of H_2O_2 as compared to sole $\text{g-C}_3\text{N}_4$.

Figure 9: Possible Chemical modification for $\text{g-C}_3\text{N}_4$.

Further, Wu X et al. [106] fabricated oxygen-containing groups-modified $\text{g-C}_3\text{N}_4$ and the efficiency is evaluated by photocatalytic H_2 -evolution performance. Dramatically, they found the efficiency was increased to a factor of 7 as compared to sole $\text{g-C}_3\text{N}_4$. Furthermore, the surface decoration with some functional groups such as $-\text{OH}$, $-\text{COOH}$, $-\text{SO}_3\text{H}$, $-\text{NO}_3$ etc. may become effective strategies to improve adsorption and photocatalytic activity of pristine $\text{g-C}_3\text{N}_4$. Because such groups not only alter the overall charge but also increase the static interaction between the surface and pollutants. Li et al. 2014 has fabricated hydroxyl modified $\text{Al}_2\text{O}_3/\text{g-C}_3\text{N}_4$.

The surface of $\text{g-C}_3\text{N}_4$ was modified by dipping in ammonia solution for 5h. The findings has revealed that the $-\text{OH}$ modified $\text{g-C}_3\text{N}_4$ based composite is 2.4 times more efficient than unmodified, for the photo-degradation of RhB. Fig. 9 refers to the different possible chemical modification on the surface of $\text{g-C}_3\text{N}_4$. However, a very limited research has been

carried out for the chemical modification of g-C₃N₄ but results depicts that such modification may route for new findings.

3.6 Dye Sensitization of g-C₃N₄

Sensitization is another popular technique to improve the photo-activity of wide band gap semiconductors. Sensitization may be carried by natural or artificial dyes. A sensitizer has ability to convert long wavelength of light to energy for the photocatalytic operation. Various dyes have been used as sensitizer to improve charge transfer mechanism and harvest more amount of solar spectrum. The charge transfer mechanism in dye sensitization is based on position of highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) of the dye. When dye sensitized C₃N₄ is subjected to irradiation, the dye gets excited and transfers its charge to CB and VB of C₃N₄. As a consequence, absorption intensity of solar spectrum gets boosted. The sensitized g-C₃N₄ has attracted much attention for DSSC application. Wang et al. 2013 [107] has fabricated g-C₃N₄ nano sheets sensitized with dye Erythrosin B and achieved enhanced activity for the H₂ evolution due to sufficient charge transfer from Erythrosin B to g-C₃N₄ surface.

The exploration of natural sensitizer to sensitize g-C₃N₄ is a greener approach because it implies the use of pigments such as anthocyanin, chlorophyll various flavonoids etc. as referred in fig. 10. These pigments are abundantly found in various plant leaves, fruits and flower and can be extracted by means of simple laboratory scale extraction procedure without involving any harsh chemical use. The modification of g-C₃N₄ with natural sensitizers may be an alternate method that can replace the use of other conventional synthetic sensitizers. Natural sensitizers are basically pigments which consist of auxochrome and chromophores. When such modification is applied to g-C₃N₄, they provided large conjugation and various functionalities which not only inhibit charge recombination but also boost solar light harvesting properties. The main advantages of such photosensitizer are that they enable the large band gap semiconductors to absorb light in the visible region. Buddee et al., 2014, have studied curcumin sensitized TiO₂ nanoparticles and found it highly effective for the cationic azo dye MB and CV photocatalytic removal [108]. Similarly Zyoud et al., 2011, fabricated Anthocyanin sensitize TiO₂ for the photodegradation of methyl orange [109].

Figure 10: Chemical structure of natural pigments anthocyanin and curcumin.

Figure 11: Hypothetical charge transfer between g-C₃N₄ and natural dye molecule.

Fig. 11 refers to the pictorial charge transfer between dye molecule and g-C₃N₄. However very limited work has been done for the sensitization of g-C₃N₄ with natural dye/pigment molecules but this area required more research to explore the use of natural sensitizer in the photocatalytic processes. Table 6 refers to some of previously reported sensitization on g-C₃N₄.

Table 6: *Dye-sensitized modified g-C₃N₄ along with utilization and efficiency.*

Sr no	Composite	Dye used as sensitizer	Enhanced photocatalytic activity evaluated by	Ref
1	MgPc/Pt/mpg- g-C ₃ N ₄	Magnesium phthalocyanine	Photocatalytic hydrogen evolution	[110]
2	Cu-Cu ₂ O/g- g-C ₃ N ₄	Erythrosin B	Hydrogen evolution	[106]
3	Xanthene Dye-Sensitized Carbon Nitride	Fluorescein, dibromofluorescein, eosin Y, and erythrosine B	-----	[111]
4	g-C ₃ N ₄	Eosin Y	Hydrogen evolution from aqueous triethanolamine	[112]
5	mpg- g-C ₃ N ₄	Eosin Y	Photocatalytic activity for H ₂ evolution	[113]

3.7 Shape specific modification

It has been well understood that the shape of a photocatalysts demonstrate a key parameter towards the photocatalytic performance. 2-D nano-sheets are the common morphological shapes of g-C₃N₄ which were fabricated by many researchers [114,115]. Moreover, nanorods [116], nano spheres [117] and flower [118] like shape controlled morphologies also has been achieved for the g-C₃N₄.

Bai et al. 2013 [119] prepared nanorods and compared the photocatalytic properties with nanoplates of C₃N₄ as synthesized by reflux method. They found that photocurrent for nanorods is approximately 2 times higher and more efficient toward the degradation of methylene blue as compared to nanoplates.

Meng et al., 2019 [120], has demonstrated the fabrication of double shell hollow g-C₃N₄.nanosphere. They had found that as fabricated nanospheres possesses outstanding optical and photoelectrical properties. The nanospheres have been prepared by thermal annealing using hallow SiO₂ as template. The activity of nanospheres has been utilized for the CO₂ conversion to formic acid.

Zheng et al., 2014 [121], have demonstrated the fabrication of helical g-C₃N₄ by nanocasting method by using chiral mesoporous silica (CMS) template. They reported that the light absorption and band gap energies are sufficiently altered for the helical structure of g-C₃N₄ as compared to bulk g-C₃N₄. They had evaluated the photocatalytic activities of helical g-C₃N₄.by hydrogen evolution, water oxidation, and CO₂-to-CO conversion. They

found that hydrogen evolution rate is 7 folds high for helical g-C₃N₄ as compared to bulk g-C₃N₄.

3.8 Polymerization of g-C₃N₄

Polymerization of g-C₃N₄ is an important modification approach that has been adopted for the g-C₃N₄ which not only increases the specific surface area but also narrows the band gap energies [122]. Ding et al., 2018 [123], have demonstrated the fabrication of polymeric g-C₃N₄ by the thermal condensation of nitric acid pretreated melamine as the precursor material. The polymeric C₃N₄ was utilized for the photocatalytic oxidation of benzyl alcohol and they reported 68.3% conversion with 100% selectivity as compared to pure g-C₃N₄.

3.9 Persulphate activation of g-C₃N₄

The addition of some precursor materials such as H₂O₂, S₂O₈²⁻, O₃, etc. when added with photocatalysts can sufficiently trigger the generation of highly reactive radicals such as OH^{*} and O₂^{*}. Activation of g-C₃N₄ by such agents offers a cost effective and highly efficient method for the degradation and mineralization of pollutants. Liu et al., 2017 [124], have studied the degradation of Bisphenol-A by Persulphate/g-C₃N₄ and reported approximately 100% removal at a concentration of 5 mM of Persulphate. Hu et al. 2016 [125] have studied g-C₃N₄/Fe(III)/persulfate system for the photodegradation of phenol and found 240-folds high efficiency. The mechanism supposed to occur during the persulphate activation in the presence of g-C₃N₄ is shown in scheme 1. The photo excited electrons react with persulphate to produce sulfate ion and sulfate radicals which further react with water molecules present in the reaction system to generate OH^{*} radical. The OH^{*} radical once formed, undergoes series of photochemical reactions and destroys the targeted pollutant molecules.

Scheme 1: Interaction of g-C₃N₄ with persulphate.

4. Further possible modification perspectives

Various modification strategies have demonstrated that photo-activity of $g\text{-C}_3\text{N}_4$ has been greatly improved. $g\text{-C}_3\text{N}_4$ has shown enhanced efficacy for various applications such as photocatalytic hydrogen evolution, pollutant degradation and mineralization etc.

Figure 12: Various photocatalytic operations carried by $g\text{-C}_3\text{N}_4$.

But still there are some unexplored research fields for which $g\text{-C}_3\text{N}_4$ has seldom been explored such as biomedical application, catalytic cracking application for petroleum industries, etc. Cost effectiveness and abundance of its cheaper precursor material can make it as demanding material if we explore it for further modification by keeping the field specific objective in the mind. The wide mode of photocatalytic operations which has been performed by $g\text{-C}_3\text{N}_4$ has been shown in Fig. 12.

Conclusion

The chapter summarizes the various strategies adopted in the present decade for the modification of $g\text{-C}_3\text{N}_4$ to boost its photocatalytic activity. Doping, coupling with binary/ternary semiconductor system, shape specific modification, polymerization, etc. are some approaches that have been explored to augment the photocatalytic potential of $g\text{-C}_3\text{N}_4$. Moreover, all the approaches which are discussed for modification have proven to significantly enhance the activity of $g\text{-C}_3\text{N}_4$ for various photocatalytic targets such as pollutant mineralization, water splitting, CO_2 conversion, fuel energy application, etc. $g\text{-C}_3\text{N}_4$ possesses high potential for its exploration in various research fields by undergoing many modifications to its 2-D motif.

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