CHAPTER – II

THE PRESENT WORK

CONTENTS

2.1 GRAPHITIC CARBON NITRIDE

The general formula of graphitic carbon nitride (g-CN) is g-C_3N_4. It is a material that contains carbon and nitrogen with little hydrogen as impurity. These are connected through tris- triazine-based pattern. It is very important organic material which is used mostly as semiconductor. It has very high stability even at more than temperature 500 $^{\circ}$C and it is hard enough. The band gap of graphitic carbon nitride is low [2.7 eV]. The graphitic carbon nitride is pale yellow in colour. The structure of graphitic carbon nitride with the tri-S-triazine is most stable, (Fig. 2.1).

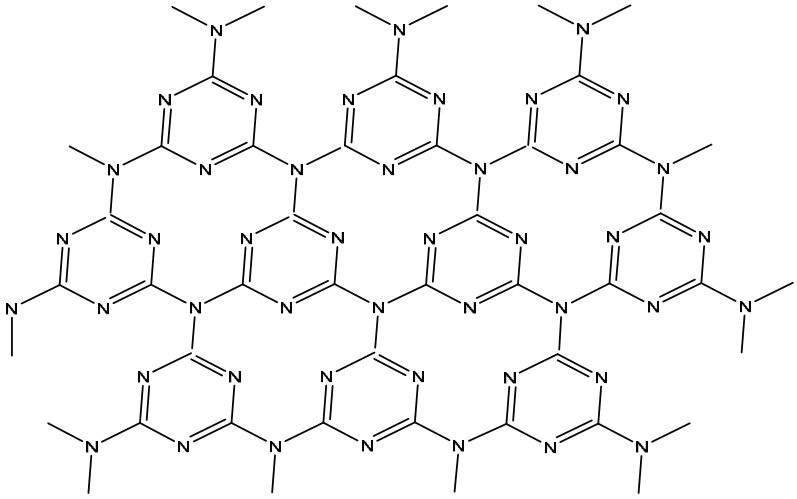

Fig. 2.1: Structure of g-C_3N_4

Graphitic carbon nitride is a metal-free conjugated polymer having two-dimensional (2D) nanosheets like structure. It is electron-rich in properties. It can be made by polymerization of melamine, cyanamide or dicyandiamide.

As it consists of layers, it can be converted into different morphologies (nanosheets, quantum dots, spherical, nanotubes. bulky, tubular and flaky. It is prepared by hydrothermal, solvothermal and thermal polymerization methods.

The graphitic carbon nitride has gained attention all over the globe as it can find applications as sensors for heavy metals, organic contaminants, pharmaceuticals and biomolecules due to its larger surface area and biocompatibility (**Fig. 2.2)**

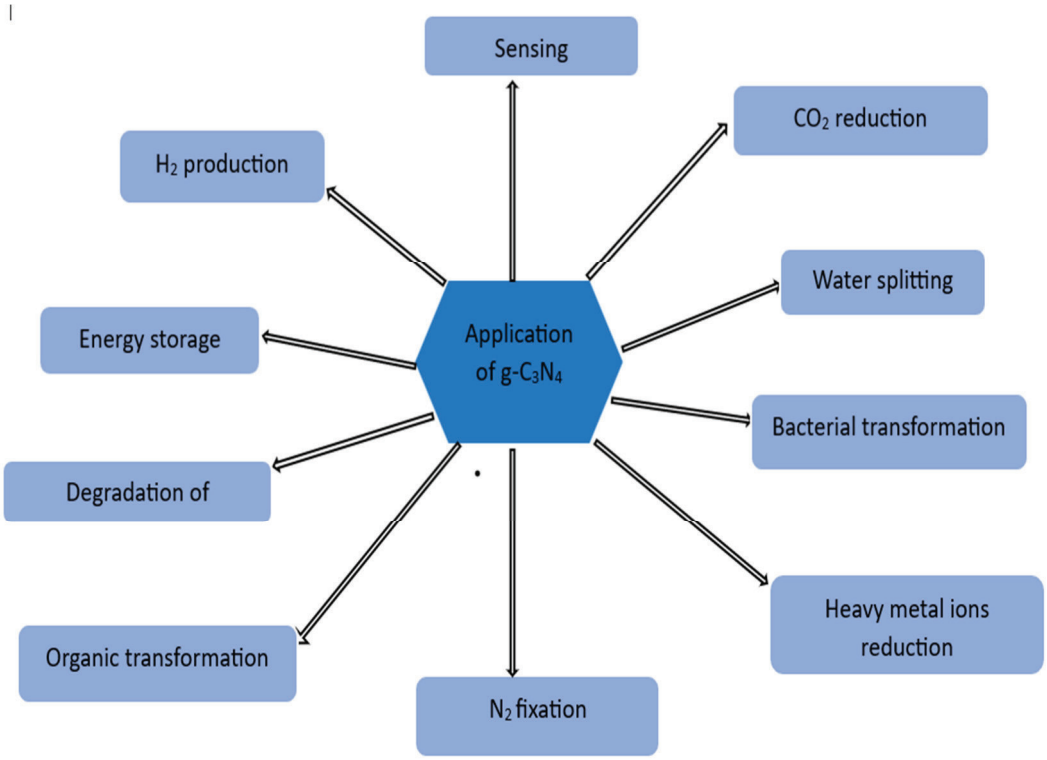

Fig. 2.2: Applications of g-C_3N_4

Graphitic carbon nitride is a metal-free semiconductor, serving as a medium band gap photocatalyst with a commendable response to visible light, extending up to 460 nm. However, pristine g-C_3N_4 is typically hindered by insufficient solar light absorption, low surface area, and rapid recombination of photogenerated electron-hole pairs; thereby, limiting its photocatalytic efficiency. Various types of modifications can be implemented to enhance the photocatalytic performance of g-C_3N_4.

- Coupling with other semiconductors,
- Conjugation with carbon nanomaterials,
- Element doped g-C_3N_4 (X-g-C_3N_4),
    (i) Metal doping, and
    (ii) Non-metal doping
- Core-shell structured g-C_3N_4, and
- Vacancy engineered g-C_3N_4.

2.2 PHOTOCATALYTIC DEGRADATION

2.2.1 Dyes

Ganesan et al.[1] tuned physiochemical properties of g–C_3N_4 by direct thermal exfoliation (TE–g–C_3N_4) Later, they used it for photocatalytic degradation of rhodamine B (RhB), methylene blue (MB), and methyl orange (MO). It was reported that degradation efficiencies for RhB, MB and MO were $95 \pm 0.4\%$, $92 \pm 0.18\%$ and $93 \pm 0.31\%$, respectively in an hour under UV light irradiation. It was also revealed that TE–g–C_3N_4 had higher surface area (48.20 m^2g^{-1}) as compared to bulk g–C_3N_4 (5.03 m^2g^{-1} It was also confirmed that superoxide radicals played an important role in the photodegradation of dye, as compared to a hydroxyl radical ($^{\bullet}OH$) and the photo-induced holes (h^+), This as-prepared TE–g–C_3N_4 also displayed excellent stability for up to five cycles with only a slight decrease in its activity from 92 to 86.2%.

Sewnet et al.[2] synthesized graphitic carbon nitride using mixtures of thiourea and urea at temperatures of calcination ranging from 500 to 650 °C for 3 h in air. It was reported that 94.83% rhodamine B (RhB) could be removed using g-C_3N_4 calcined at 600 °C within 3 h under visible LED light irradiation.

Nguyen et al.[3] synthesized water-dispersible graphitic carbon nitride using some nitrogen-rich organic precursors such as thiourea, urea, melamine and dicyandiamide via thermal treatment. The photocatalytic degradation of methylene blue (MB) was observed in presence of as-prepared g-C_3N_4**.** It has high surface area, increased band gap, and reduction in the recombination rate of photogenerated electron-hole pairs.

de Lima et al.[4] synthesized mesoporous graphitic carbon nitride using surfactant Tween-40. It was used as a visible-light active photocatalyst in degrading rhodamine B. It was reported that 93.0% rhodamine B could be degraded by as-prepared catalyst after 1 h under visible-light irradiation, while mesoporous graphitic carbon nitride and bulk graphitic carbon nitride with Pluronic F127 could degrade only 86.0 and 30.5% of rhodamine B, respectively.

Nunna et al.[5] fabricated mesostructured graphitic carbon nitride (MCN) composites from Ag-doped melamine at room temperature. It was reported that Ag nanoparticles were distributed uniformly on the surface of MCN sheets, The photocatalyst degradation of crystal, and violet, methylene blue was observed, which

was more than 96% in presence of MCN/Ag-MCN composite. Sidewise, photocatalytic hydrogen evolution (5.24 mmol. $g^{-1}.h^{-1}$) was also observed.

Utomo et al.[6] modulated particle size and morphology of ZnO by variation of pH (8.0, 9.0, 10.0, and 11.0). It was observed that at higher pH (10.0), smaller particle size and cone-like strcture was there on the edge of ZnO particle. Highest MB degradation (97.7%) could be achieved with this as-prepared composite in 150 min.

Zhao et al.[7] used 0.75 wt % 2,3-pyridinedicarboxylic anhydride (PDA) as dopant to modifiy graphitic carbon nitride with ultrarace doping (3MCN-PDA3). It was reported that photodegradation efficiency of rhodamine B (RhB) over as-prepared composite reached 92.4% in 1 h under visible light and almost complete removal (99.2%) was observed in 200 min.

Bandoh et al.[8] synthesized cobalt and graphitic carbon nitride (g-C_3N_4) co-doped ZnO through co-precipitation and used for eosin yellow degradation under visible light. It was reported that a marginal rise in absorbance was observed in the visible region due to decrease in band gap from 3.10 to 2.52 eV. It was observed that ZnO has an average crystallite size of 42 nm. The optimal photocatalytic performance was observed with 0.8 % Co-doped ZnO-g-C_3N_4, which could degrade 96 % of eosin yellow in 3 h.

Amritha and Badhulika.[9] prepared vanadium oxide and graphitic carbon nitride nanocomposite and used for degradation of methylene blue (MB) and rhodamine B (RhB). The band gap of as-prepared nanocomposite was determined as 1.89 eV. It was reported that as-synthesized nanocomposite exhibited an excellent degradation with 100 and 90% degradation of RhB and MB, respectively, within 1 h under daylight. It was also confirmed that hydroxyl and oxygen anion ($O_2^{-\bullet}$) radicals were primary oxidizing species in the degradation.

Lyu et al.[10] prepared three dimensions (3D) flower-shaped g-C_3N_4/MoS_2 composite by immobilizing molybdenum disulfide on carbon nitride. The photocatalytic performance of this composite was observed under simulated sunlight. This as-prepared composite could remove about 98 % of MB within 1 h and retained its activity to 96.4 % after 5 cycles. The apparent rate constant of this degradation was 0.0523 min^{-1}, which was 13.9, 11.5, and 8.2 times higher as compared to MoS_2, g-C_3N_4 and C_3N_4 with structural defects.

Naz et al.[11] prepared Cu and Fe doped nanocomposites $NiCo_2O_4$/g-C_3N_4 with different dopant concentrations via precipitation and ultrasonication routes. It was reported that $NiCo_2O_4$ nanoparticles have spherical shape (ranging between 18 to 32 nm). They also evaluated photocatalytic activity of doped $Ni_{0.75}Cu_{0.25}Co_{1.5}Fe_{0.25}O_4$/g-$C_3N_4$ ($NCCF_3$/g-CN) for removal of Congo red (CR) and rhodamine B (RhB) under visible light irradiation. It was observed that $NCCF_3$/g-CN nanocomposite could remove 89 % of the dye as compared to undoped nanocomposite (50 %). It was attributed to efficient separation of charge carriers by migration of photo-induced electrons from g-C_3N_4 surface to $NiCo_2O_4$.

Salehi et al.[12] prepared ternary heterogeneous Mg–Al LDH g-C_3N_4 XAg_3PO_4Y (X = wt % of g-C_3N_4 with respect to Mg–Al layered double hydroxide (LDH) and Y = wt % of Ag_3PO_4 loaded on Mg–Al LDH g-C_3N_4 30) nanocomposite and used for degradation of methylene blue (MB). The photocatalytic removal of MB under 150 W mercury lamp increased significantly in the presence of the composite of Mg–Al LDH and g-C_3N_4 as compared to Mg–Al LDH alone. It was observed that highest photodegradation activity (99%) could be obtained using this as-prepared composite. It was also revealed that effective agents in the photodegradation of MB were holes and hydroxyl radicals as evident from scavenger tests.

Qiaofang et al.[13] prepared vermiculite g/C_3N_4/TiO_2 composites from TiO_2,melamine and vermiculite. It was reported that the best sample was obtained with calcination at 200 °C for 3 h. It was observed that 99.33 % degradation of methylene blue (MB) could be achieved. This composite still has good degradation efficiency even after using it four times.

Huang et al.[14] prepared Ag_3PO_4-g-C_3N_4 photocatalyst. It was reported that as-obtained Ag_3PO_4-g-C_3N_4 heterojunction exhibited excellent photodegradation performance for removal of methyl orange (MO) and rhodamine B (RhB). It was also revealed that its performance remained more than 90% after even six cycles.

Bhakar et al.[15] synthesized Z-scheme heterojunction-based ZnO-$MnWO_4$g-C_3N_4 ternary nanocomposite via coprecipitation and ultrasonication. It was observed that 94.5% methylene blue (MB 10 ppm) could be degraded under visible light irradiation using 10 mg of catalyst within 50 min. Out of all samples, ZnO-$MnWO_4$g-

C_3N_4 (1CN:1ZM) (1:1) exhibited excellent degradation (94.50%) towards MB as compared to two other ratios (1:2 and 2:1). It was found that degradation rate was 2.57 times more than the pure g-C_3N_4.

An et al.[16] synthesized zinc oxide-decorated graphitic carbon nitride composite (ZnO/gC_3N_4 or ZCN). It was found that ZnO has a spherical shape with an average diameter of 30–80 nm and it is distributed uniformaly on the CN sheets. They also used ZCN for photodegradation of methylene blue (MB), which could degrade 99.16% dye after 3 h of visible irradiation. The optimal conditions obtained were: pH 9, 50 mg of catalyst, and 10 mg L^{-1} of dye concentration. Its degradation ability for malachite green (MG) and methyl orange (MO) was about 96.42 and 57.57 %, respectively. The reusability of this composite was also ascertained as only 10 % decrease was there after five cycles.

Lan et al.[17] prepared oxygen-phosphorus-codoped graphitic carbon nitride (O, P-GCN) via thermal decomposition of oxalic acid, urea, and ammonium phosphate. This as-prepared O, P-GCN photocatalyst was used for degradation of rhodamine B (RhB). It was reported that 1 wt% oxygen and phosphorus in GCN improved its photocatalytic performance, significantly and degradation efficiency of RhB could reach up to 97.25%. It was found that various kinds of reactive species were generated such as hydroxyl radicals, superoxide radicals, and holes. There played a major role in the photocatalytic degradation of RhB.

Kumaravel et al.[18] modified graphitic carbon nitride g-C_3N_4 (GCN), with tungsten oxide WO_3/g-C_3N_4 (WGCN) as well as tungsten oxide and silver nanoparticles WO_3/Ag/g-C_3N_4 (WAgGCN) also. They used this WAgGCN composite for photocatalytic decolorization of crystal violet (95%), and methylene blue (98%). It has high chemical stability and excellent recyclability also. A decrease in total organic carbon (TOC) and chemical oxygen demand (COD) also confirmed that dyes were mineralized.

Vuggili et al.[19] synthesized nitrogen-doped g-C_3N_4/CoS (CSNG) nanocomposites by polycondensation-hydrothermal method with deferent concentrations of nitrogen-doped g-C_3N_4 (NG). It was reported that industrial real samples (RS.I and RS. II,) could be degraded by 60 and 41% in 20 and 70 min, respectively. It was also observed that a mixture of RS I with methylene blue (MB) was degraded 79 and 98% in 20 min, while

its mixture with RS II degraded 72% and 99% in 40 min. It was revealed that superoxide anion radical played an important role in the photodegradation of MB. It also exhibited excellent photostability with a loss of just 0.7% in five consecutive cycles.

Zhou et al.[20] synthesized NiS-loaded on S-doped carbon nitride photocatalysts. The activities of these photocatalysts were evaluated in photodegradation of RhB under visible-light irradiation. It was reported that the photocatalyst (SCN/NiS-1) with 0.89 wt% sulfur doping and 1 wt% NiS loading showed excellent photocatalytic activity of removing 98.5% RhB in 15 min.

Onwudiwe et al.[21] prepared graphitic carbon nitride and copper-doped bismuth sulphide heterostructure (g-C_3N_4/Cu–Bi_2S_3) by solvothermal method. Photocatalytic activity was investigated for degradation of methyl orange. It was reported that about 95% degradation of methyl orange was achived on using this heterostructure photocatalyst. It was also revealed that superoxide radicals and holes played major roles in this photocatalytic degradation.

Kumaravel et al.[22] fabricated x% Ag/g-C_3N_4 (x% AgGCN) composite with different weight percentages (x = 1, 3, 5, and 7 wt.%). It was reported that 99% photodegradation rate of methyl orange could be achieved under solar light irradiation with 5% AgGCN composite photocatalyst. It was observed that degradation with 5% AgGCN was 14.83 times more than with GCN catalyst alone. This photocatalyst could retain good efficiency up to five successive cycles. It was also indicated that $O_2^{\bullet-}$ radicals played a crucial role in degradation of MO.

Amritha and Badhulika.[23] fabricated $NaTiO_3$/g-C_3N_4 heterojunction photocatalyst and used it for removal of methylene blue (MB) and crystal violet (CV) dyes and their binary mixture under sunlight irradiation. It was reported that $NaTiO_3$ nanoparticles were aggregated on g-C_3N_4 sheet-like surface. The band gap of as-prepared sample was found to be 2.3 eV. It was observed that complete degradation of MB, and CV was observed within 25 min, while binary mixture took 20 min for complete degradation. It was also indicated that the radical species h^+ and $^{\cdot}OH$ played the major role in the photocatalytic degradation of these dyes. It was also revealed that it has good stability up to five cycles.

Chopan and Chishti[24] fabricated polypyrrole (PPy) and graphitic carbon nitride (g-C_3N_4) nanoparticles on ZnO surface as g-C_3N_4/PPy/ZnO (GPZ). The

photocatalytic activity of as-prepared photocatalyst was evaluated for degradation of RhB where it could degrade 99.0% of RhB in 1h at pH 7.

2.2.2 Pharmaceutical drugs

Falletta et al.[25] synthesized graphitic carbon nitride as powder and alginate floating beads, using two different precursors (urea and melamine). These were than used for their activity in photodegradation of diclofenac under solar light irradiation. It was reported that almost 99 and 90% degradation for diclofenac and isoproturon could be achieved. The floating photocatalysts displayed good stability during recycling experiments and it can be used up to five cycle.

Liu et al.[26] prepared layered phosphorus-doped carbon nitride ($HPCN_{0.5}$) by hydrothermal route and used for photocatalytic degradation of dinotefuran. On addition of phosphorous acid, $HPCN_{0.5}$ (tube-like structure) was obtained, where planar sheets were stacked inside these tubes. It was reported that the degradation efficiency of dinotefuran was six times higher as compared to bulk g-C_3N_4. It was observed that photodegradation proceed through demethylation and hydroxylation steps.

Pham et al.[27] synthesized and used graphitic carbon nitride in alkaline conditions for photodegradation of carbamazepine (CZ) and acetaminophen (AP) in wastewater. This was reported that it exhibited high removal efficiencies of 98.6 and 89.5% for AP and CZ, respectively. Increasing the catalyst dose facilitated the removal of antibiotic contaminants with an optimum catalyst dose of 0.1 g, achieving a photodegradation efficiency of 90.2% and 82.7% for AP and CZ, respectively. It was also revealed that as synthesized photocatalyst could remove over 98% of AP within 2 h, which was 2.14 times faster than removal of CZ. The participation of highly reactive oxidants such as hydroxyl and superoxide radical were confirmed. A good stability of g-CN was there in treating pharmaceuticals as it can be reused for three repeated cycles.

Tao et al.[28] synthesized carbon-defective carbon nitride (with pore structure) via thermal polymerization at 600°C (CCN-600) and used it for photodegradation of mepivacaine, lidocaine, and ropivacaine. It was reported that degradation followed pseudo-first-order kinetics with 5.05×10^{-2} min^{-1} rate constant, which was about 2.5 times higher as compared to g-C_3N_4. It was reported that superoxide radical played a major role in this degradation.

Villanueva et al.[29] prepared coupled g-C_3N_4 /TiO_2 and used it for degradation of ciprofloxacin, tetracycline, and ibuprofen under simulated solar irradiation. They used SBA-15 silica as hard template. The photocatalytic activity was examined through the degradation of tetracycline, ciprofloxacin, and ibuprofen. It was observed that dispersion of TiO_2 nanoparticles could be obtained on using a mesoporous g-C_3N_4 sample. It was also revealed that samples with 1 wt.% g-C_3N_4 displayed highest photocatalytic performance in the degradation of ciprofloxacin, tetracycline, and ibuprofen.

Govindasamy et al.[30] constructed eggshell derived hydroxyapatite nanoparticles entrenched on two-dimensional g-C_3N_4 nanosheets. They evaluated photocatalytic performance of this as-prepared HAp/gC_3N_4 photocatalyst for degradation of doxycycline under UV–visible light exposure. It was reported that as-prepared photocatalyst exhibited excellent photocatalytic activity for degradation of doxycycline drug (83.08%) as compared to g-C_3N_4 and bare HAp nanosheets. It was indicated that this photocatalyst can be recycled for four times and reused without any significant loss in degradation performance.

Razavi-Esfali et al.[31] used SiO_2 clusters as template to fabricate mesoporous graphitic carbon nitride nano-clusters (NC MCN). It was reported that NC MCN exhibited high photocatalytic activity for degrading tetracycline under visible light irradiation and complete degradation of 15 mL solution of tetracycline (15 ppm) could be achieved in half an hour as compared to the bulk catalyst in more than 3 h. Furthermore, This NC MCN also exhibited high stability as it can be reused for eight consecutive cycles.

Minh Tri et al.[32] synthesized Ag-doped graphitic carbon nitride (Ag-GCN) and used it as a photocatalyst for degradation of hospital wastewater containing tetracycline (TC) under solar light. It was revealed that the highest photocatalytic degradation efficiency of TC (96.8%) was obtained in 2 h under solar light irradiation, when content of Ag doping was kept 3 mmol. The removal efficiency of TC was only 25.6 and 31.8% under dark conditions, respectively in presence of g-C_3N_4 and Ag-doped g-C_3N_4 (AgCN). It was also reported that AgGCN exhibited extremely high stability after six cycles without any significant loss in its efficiency.

Pham et al.[33] fabricated cobalt doped GCN (Co/GCN) and used it for the photodegradation of paracetamol (PC) in the wastewater. It was reported that as-

prepared Co/GCN exhibited a removal efficiency of 82.6% under sunlight in 1 h even at high PC initial concentration, which was 1.7 times higher than undoped GCN. The Co/GCN was capable of decomposing PC completely (100%) at low initial concentration of ≤ 1 mg L^{-1}.

Xu et al.[34] synthesized CeO_2/N-doped g-C_3N_4 (CeNCN) composite photocatalysts. It was observed that CeNCN exhibited excellent photodegradation of tetracycline (80.09%) within an hour, which is 7.86 and 2.29 times more than graphitic carbon nitride and N-doped GCN, respectively. It was reported that degradation efficiency of tetracycline is more than 75%, even after five cycles, which indicated its good stability.

Sun et al.[35] synthesized graphitic carbon nitride nanosheet modified with dodecyl benzene sulfonate (DBS). Its photocatalytic performance for removal of moxifloxacin (MOX) was evaluated. It was reported that this catalyst could remove high concentration of MOX, (100 mg L^{-1}) in half an hour of irradiation. They also observed the effect of different parameters on this degradation such as: pH, concentration of MOX and dosage of photocatalyst It was also claimed that DBS/CNNS hybrid is a potential visible light driven photocatalyst for elimination of high-concentrations of antibiotics in wastewater.

Zhang et al.[36] introduced a group on terminal amino group of g-C_3N_4 (G-CN) via in acetic acid under 80 °C to afford HAC-CN. It was reported that photocatalytic degradation efficiency of some antibiotics; sulfamethoxazole (SMZ), tetracycline hydrochloride (TC), and ciprofloxacin (CPFX)) over HAC-CN was significantly improved, which are 1.50, 1.50, and 1.99 times than that with G-CN. It was revealed that especially $O_2^{\bullet-}$ was dominant oxidized species in their degradations.

Rajiv et al.[37] loaded haematite on graphitic carbon nitride via hydrothermal approach and its photocatalytic activity was evaluated for degradation of ciprofloxacin (CIP). It was reported that g-C_3N_4/Fe_2O_3/UV exhibited higher degradation efficiencies than UV and g-C_3N_4/Fe_2O_3. They could achieve maximum removal of CIP (100%) and TOC (93.86%) at pH 7 in 1 h. It was also revealed that h^+ and $^{\bullet}OH$ played major role in the degradation of CIP. An excellent stability and recyclability of this composite was confirmed by five degradation cycles.

Bian et al.[38] reported S-doped g-C_3N_4 with hollow microsphere composition (SCNHM). It was reported that SCNHM had high specific surface area (~81 m^2 g^{-1}). The S dopant improved electronic structure so that band gap was narrowed down and promoted charge separation/transfer capability. The as-prepared SCNHM improved photocatalytic activity for degradation of tetracycline hydrochloride (TC) due to synergistic effect as compared to CN, SCN and CNHM samples. It was reported that it could eliminate high-concentration TC (50 mg L^{-1}) in 18 min, while removal efficiencies of 100 mg L^{-1} and 200 mg L^{-1} could reach 92 and 60 %, in half an hour.

Chin et al.[39] prepared GCN with different precursor (GCN-urea, GCN-thiourea and GCN-melamine). They also investigated abilities of these as-prepared photocatalyst to degrade oxytetracycline (OTC). It was reported that GCN-urea has high surface area 73.62 m^2g^{-1}, as compared to 5.82 and 3.99 m^2 g^{-1} GCN-thiourea and GCN-melamine respectively. The GCN-urea has the highest band gap as 2.7 eV, and GCN-melamine and GCN-thiourea have lower band gaps as 2.3 and 2.2 eV, respectively. The highest OTC degradation was achieved by GCN-urea at ~60%, which was almost 2.5 times higher than the other two

Wang et al.[40] reported the synthesis of oxygen and terminal methyl moiety co-modified g-C_3N_4 (CNNx). It was reported that synthesized CNNx (CNN25) could achieve high photocatalytic activity for photocatalytic degradation of 74, 99, 92 and 65% sulfathiazole, tetracycline, doxycycline, and ofloxacin, respectively within 30 min under visible light.

Shen et al.[41] prepared metal-free carbon/g-C_3N_4 (CNC) photocatalyst with persulfate (PDS), (CNC-PDS) and used it for photocatalytic degradation of paracetamol (PCM) under simulated sunlight. It was reported that excellent photocatalytic activity was exhibited and PCM was completely degraded in 40 min under simulated sunlight, which was about 9.5 times more as compared to g-C_3N_4 and PDS coupled systems. It was also indicated that $O_2^{\cdot-}$, h^+ and $^1O_2$ were the main active species for degradation of PCM over CNC-PDS system under simulated sunlight, and their contribution was in order the $O_2^{\bullet-} > {}^1O_2 > h^+$.

2.3 OTHERS

Zheng et al.[42] synthesized N-deficiently porous graphitic carbon nitride, particularly UMCN, which was prepared using a mass ratio of 1: 5 for melamine to urea. It was reported that high specific surface area was (183.7 m^2g^{-1}). It was observed that UMCN exhibited higher photocatalytic activity for degradation of rhodamine B (RhB) with removal rate 99.3% within half an hour under visible light. It was also revealed that excellent recyclability is there and degradation rate of RhB still remained 95.6% even in the 7th run.

Song et al.[43] prepared graphitic carbon nitride by one-step polycondensation of urea. It was observed that it has surface area of 114 $m^2\ g^{-1}$, which permitted adsorption of the rhodamine B (RhB) dye and also its photocatalytic degradation upon light irradiation in 20 min.

Poiyamozhi and Thivya.[44] synthesized a composite of g-C_3N_4 and $NiSe_2$ via ultrasonication-assisted hydrothermal method. The band gap of this hybrid composite was found to decrease from 2.72 to 2.30 eV. The photocatalytic degradation of reactive blue 5 (RB 5) and reactive violet 5 (RV 5) was observed over $NiSe_2$/g-C_3N_4 composites. The highest degradation rates for RB 5 (86%) and RV 5 (89.4%) could be obtained with 30 % g-C_3N_4 incorporated into $NiSe_2$.

Khan et al.[45] immobilized $Ag_3PW_{12}O_{40}$ on graphitic carbon nitride (2D sheet) through grinding and prepared separately $C_3N_4Ag_3PW_{12}$ heterojunction. It was reported that C_3N_4-Ag_3PW_{12}-30% composite photocatalyst displayed efficient photocatalytic activity and removed 99 and 99.4% methylene blue and rhodamine B in 75 and 60 min respectively, whereas 26.3 and 37.7 % of methylen blue and 43.54 and 53.38 % of rhodamin B was removed by Ag_3PW_{12} and C_3–N_4 respectively.

2.3.1 Pesticides

Ejeta and Imae[46] synthesized oxygen-doped graphitic carbon nitride (Og-CN). It was then used for photocatalytic degradation of 2,4-dichlorophenoxyacetic acid (2,4-D) under UV and visible light irradiation. It was observed that degradation efficiency of Og-CN was better as compared to g-CN, and UV light was more effective than visible light. The Og-CN has good stability as there was only 4% decrease in its activity after three

photocatalytic cycles. A high total organic carbon removal (88 %) was observed in 2 h, which confirmed mineralization of 2, 4-D.

Vigneshwaran et al.[47] used graphitic carbon nitride (g-C_3N_4) incorporated into chitosan for photocatalytic degradation of an organic pesticide, chlorpyrifos (O, O-diethylO-3,5,6-trichloro-2-pyridyl phosphorothioate (CPFS)). It was reported that degradation of this insecticide showed good efficiency of about 85% on using CS/g-C_3N_4.

Liu et al.[48] used graphitic carbon nitride as a photocatalyst for photodegradation of imidacloprid which has become a research hotspot, due to its high toxicity to bees and other nontarget organisms. Photodegradation is a common method for removing imidacloprid in an aquatic environment. The g-C_3N_4 was used photodegradation of imidacloprid under visible-light illumination, which resulted in about 90% removal in 5 h. They also identified 4,5-dihydro-N-nitro-1-(3-pyridinylmethyl)-1H-imidazol-2-amine and 1-((6-chloropyridin-3-yl) methylhydroxy) imidazolidin-2-ylidenenitramide) as main photoproducts as evident from LC-MS analysis.

Zong et al.[49] synthesized modified carbon nitride (TEA-CN-30) via photochemical reduction using triethanolamine. Then it was used for photocatalytic degradation of atrazine. It was reported that about 90% atrazine degradation could be achieved in an hour. The possible degradation pathways were suggested as hydroxylation, dealkylation, and dichlorination.

2.3.2 Polycyclic Aromatic Hydrocarbon

Halim et al.[50] prepared composite of graphitic carbon nitride from different combinations of precursors through thermal polycondensation at 600°C. They used these for photocatalytic degradation of polycyclic aromatic hydrocarbons (pyrene). It was reported that best performance of composite g-C_3N_4 photocatalyst was obtained, when it was prepared from a wet mixture of guanidine carbonate and dicyandiamide precursors (mass ratio of 1:1), which could degrade 43.9 % pyrene under visible light irradiation in 4 h. It was observed that as-prepared photocatalyst was stable and can be reused up to five cycles. The holes and hydroxyl radicals were identified as the primary and secondary dominant reactive species based on scavenging studies.

Azmoon et al.[51] synthesized graphitic carbon nitride with nanorod structure (Nr-GCN). They prepared polystyrene (PS) fibrous mat and used it as a support for stabilizing nanoparticles. The performance of as-synthesized nanoparticles and photocatalytic fibers (PS/Nr-GCN) was evaluated in treatment of oilfield-produced water under visible light irradiation. It was reported that 96.6 and 98.4% removal of chemical oxygen demand (COD) could be achieved by Nr-GCN and PS/Nr-GCN, respectively at pH 4 in 150 min.

Bharathi et al.[52] synthesized g-C_3N_4–Ni nanocomposites and used for photocatalytic degradation of benzopyrene the most potent polycyclic aromatic hydrocarbons (PAH). It was reported that these nanocomposites are nanospheres with an average particle size of 22 nm. It was observed that degradation of benzopyrene increased with increasing pH till 3 h (incubation period) but afterward its percentage declined. It was also revealed that UV light enhanced degradation of benzopyrene more than sunlight.

2.3.4 Phenols

Jilani et al.[53] prepared sulfonated polyaniline, (s-PANI) graphitic carbon nitride (*g*-C_3N_4) and its nanocomposites with graphene. Different weight ratios of GN were used (1, 3, and 5%) with weight of *g*-C_3N_4. They used as-prepared nanocomposites for degrading phenol moieties. It was revealed that enhanced photocatalytic degradation of phenol was there with GN-based sulfonated s-PANI@*g*-C_3N_4/$GN_{(1–5\%)}$. An increase in photocatalytic activity was attributed to the involvement of the sp^2 carbon atoms, the interaction of the O C carbon atoms, and the high migration efficiency of charge carriers.

Rana et al.[54] synthesized metal-free graphitic carbon nitride and used for photocatalytic degradation of phenol using visible light-emitting diodes (LEDs). It was reported that exfoliated g-C_3N_4 completely degraded phenol in 90 min, while only 25% phenol could be degraded with bulk g-C_3N_4 in 3 h, because the exfoliated g-C_3N_4 has more availability of active sites, which promoted the degradation of phenol.

Kavitha and Kumar[55] prepared polymeric g-C_3N_4 (GCN) and used it as photocatalytic material for degradation of phenolic compounds (reduction of 4-nitrophenol (4-NP) to respective 4-aminophenol (4-AP)) in aqueous medium.

Photocatalysis has a potential to degrade majority of organic contaminants present in polluted water. But mostly, the photocatalysts used so far contain some metals. There is an urgent demand to search for some metal free semiconductor as a photocatalyst. In this direction, graphitic carbon nitride can be considered a suitable candidate for treatment of waste water as it is metal free as well as has a desired band gap in the range of semiconductor (Eg = 2.7 eV). Therefore, it was planned to carry out some systematic investigations for the degradation of some commonly used dyes such as; Azure A, Evans blue, Rose Bengal, Methylene blue, Rhodamine B, and Alizarin red-S in presence of graphitic carbon nitride and light.

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