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Review

Graphitic Carbon Nitride-Based Composite in Advanced Oxidation Processes for Aqueous Organic Pollutants Removal: A Review

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Abstract: In recent decades, a growing number of organic pollutants released have raised worldwide concern. Graphitic carbon nitride ($g\text{-C}_3\text{N}_4$) has drawn increasing attention in environmental pollutants removal thanks to its unique electronic band structure and excellent physicochemical stability. This paper reviews the recent progress of $g\text{-C}_3\text{N}_4$ -based composites as catalysts in various advanced oxidation processes (AOPs), including chemical, photochemical, and electrochemical AOPs. Strategies for enhancing catalytic performance such as element-doping, nanostructure design, and heterojunction construction are summarized in detail. The catalytic degradation mechanisms are also discussed briefly.

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1. Introduction

In recent years, environmental pollution, especially water pollution, is increasingly becoming a major concern worldwide. Many organic pollutions, such as pharmaceuticals and personal care products (PPCPs), pesticides, and organic dyes are toxic and refractory [1–4]. Various techniques have been developed to eliminate aqueous organic pollutants (e.g., extraction, adsorption, biological treatment, and advanced oxidation processes) [5–9]. Advanced oxidation processes (AOPs) are regarded as effective techniques for organic contaminants removal from water and wastewater [10–14].

The AOPs utilize highly reactive species (mainly hydroxyl radical, $\bullet\text{OH}$) to oxidize the organic pollutants into less toxic or no-toxic products such as CO_2 and H_2O [15,16]. According to supplied energies and reactive species, AOPs can be categorized as photocatalysis, electrocatalysis, sonolysis, ozonation, Fenton/Fenton-like reactions, and sulfate radical-based AOPs (SR-AOPs), among others [17,18]. In recent decades, numerous studies have been conducted to develop novel AOPs. Emerging energy sources (e.g., ionizing radiation with electron beams and γ -radiolysis, pulsed plasma, etc.) were applied, and different reactive species (such as periodate or ferrate reagent) were introduced [19–22]. Considering the merits of different AOPs, combinations of various processes are more common approaches to enhance degradation efficiency [21]. Although some AOPs (e.g., UV/ H_2O_2 , UV/peroxymonosulfate (PMS)) work properly without catalysts, employing catalysts can significantly reduce energy and reagent (source of reactive species) consumption [23]. Therefore, designing an effective and stable catalyst is a crucial strategy for the development of AOPs.

Graphitic carbon nitride ($g\text{-C}_3\text{N}_4$), known as a metal-free polymer semiconductor, has attracted increasing attention due to its unique electronic band structure, anti-photocorrosion, excellent physicochemical stability, and easy availability [24,25]. The bandgap of $g\text{-C}_3\text{N}_4$ is about 2.7 eV, which enables it to absorb all viewing range of solar irradiation. The valance band (VB) and conduction band (CB) mainly encompass nitrogen and carbon p_z orbitals

while VB top and CB bottom are located at about +1.4 and -1.3 eV, respectively [26–28]. The study of g-C₃N₄ can be traced back to 1834, when Berzelius first synthesized a polymeric derivative of g-C₃N₄, and Liebig named it melon [29]. In 2006, Goettmann and his co-workers investigated Friedel–Crafts reactions that can be catalyzed by g-C₃N₄, which is its first application in the catalytic field [30]. In 2009, g-C₃N₄ was demonstrated as a good metal-free photocatalyst for water splitting by Wang et al. [26]. Up to now, g-C₃N₄ has been in-depth studied and extensively applied in photocatalysis. The g-C₃N₄ preparation relies on (solvo)thermal polymerization of nitrogen-rich precursors such as melamine, dicyandiamide, and urea [31]. In addition, hard/soft template-assisted methods and sol-gel methods are frequently used to modify the synthesis approaches. The reaction parameters such as precursors and temperature could significantly affect the physicochemical property, including specific surface area, bandgap, etc. [32]. However, the pure g-C₃N₄ encounters several drawbacks, including tiny surface area, inefficient use of visible light, low electric conductivity, and fast recombination of photo-induced carriers, which are not beneficial to its catalytic activity [33,34]. To address these issues, a lot of efforts such as (1) engineering the nanostructure of g-C₃N₄ [27,35–37], (2) introducing heteroatoms (metals [38–44] or non-metals [45–50]), (3) coupling with other semiconductors [51–57] and (4) co-polymerization [58–61] were made. g-C₃N₄ based composites hold unique advantages for organic pollutants removal from groundwater and wastewater due to the good adsorption capacity of g-C₃N₄ for organic molecules, which could be attributed to strong intermolecular forces like hydrogen bonding, π - π interactions between pollutant molecules and residual amino groups in the g-C₃N₄ fragment [62,63]. On the other hand, introducing extra sources of reactive species such as H₂O₂ or PMS in photocatalysis can significantly increase degradation efficiency [64,65]. Furthermore, some studies have explored g-C₃N₄ based composites for organic pollutants removal without light irradiation in the presence of PMS or H₂O₂ [66–69].

Some excellent reviews on g-C₃N₄ based composites involving pollution remediation have been published [25,31,70,71]. g-C₃N₄ based composites as photocatalysts for water purification have been summarized in these reviews, while no reviews involve other AOPs such as chemical AOPs and electrochemical AOPs. The dramatically increasing amounts of g-C₃N₄ based composites in the range of AOPs fields requires a broader, thorough, and up-date assessment. This review is mainly focused on the application of g-C₃N₄ based composites in chemical AOPs and electrochemical AOPs. The challenge and strategies for enhancing photodegradation performance using g-C₃N₄ based composites as photocatalyst were also included.

2. Chemical AOPs

The chemical AOPs started as early as the application of the Fenton reaction to water treatment, in which \bullet OH can be generated from the catalytic decomposition of H₂O₂ by Fe²⁺ for the destruction of various organic pollutants (Equation (1)) [10].



With the increasing demand for water treatment, various oxidants such as O₃, PMS, peroxydisulfate (PDS) were applied in chemical AOPs. The PMS and PDS could be heterogeneous activated, and reactive species such as $\bullet\text{SO}_4^-$ are subsequently generated to degrade organic pollutants [72–74]. Table 1 summarizes the part of a representative study using g-C₃N₄ based composites as a catalyst in chemical AOPs.

Table 1. Graphitic carbon nitride (g-C₃N₄) based composites for chemical advanced oxidation processes (AOPs).

Catalyst	Target Contaminants	Oxidant	Reaction Conditions	Performance	Ref.
Cu(II)/CuAlO ₂ /g-C ₃ N ₄	Bisphenol A (BPA)	H ₂ O ₂	BPA, 25 mg/L; catalyst, 1 g/L; H ₂ O ₂ , 10 mM; T, 35 °C; pH, 7	95.5% in 120 min	[75]
Cu/Al ₂ O ₃ /g-C ₃ N ₄	Rhodamine B (RhB)	H ₂ O ₂	RhB, 20 mg/L; catalyst, 1 g/L; H ₂ O ₂ , 10 mM; T, 25 °C; pH, 4.9	96.4% in 100 min	[76]
Iron oxide/g-C ₃ N ₄	Ciprofloxacin	H ₂ O ₂	ciprofloxacin, 20 mg/L; catalyst, 1 g/L; H ₂ O ₂ , 5.6 mM; pH, 3	100% in 60 min	[68]
g-C ₃ N ₄ /carbon nanotubes/Fe(II)	Methylene blue	H ₂ O ₂	Methylene Blue, 90 mg/L; catalyst, 0.5 g/L; H ₂ O ₂ , 1 mM; T, 25 °C; pH, 4.9	66.8% in 1 h	[69]
Fe ₃ O ₄ @C/g-C ₃ N ₄	Acid orange 7 (AO 7)	PMS	AO 7, 20 mg/L; catalyst, 0.6 g/L; PMS, 0.1 g/L; T, 25 °C; pH, 4	97% in 20 min	[77]
CoFeO ₂ /g-C ₃ N ₄	Levofloxacin	PMS	levofloxacin, 10 mg/L; catalyst, 0.15 g/L; PMS, 0.5 mM; T, Room temperature; pH, 3	100% in 60 min	[78]
Co-doped g-C ₃ N ₄	4-chlorophenol	PMS	4-chlorophenol, 50 mg/L; catalyst, 1 g/L; PMS, 2.5 mM	100% in 30 min	[79]
Mn-doped g-C ₃ N ₄	Acetaminophen	PMS	acetaminophen, 20 mg/L; catalyst, 0.05 g/L; PMS, 0.8 g/L; pH, 6.5	100% in 15 min	[80]
Cu ⁺ -g-C ₃ N ₄	Rhodamine B	H ₂ O ₂	Rhodamine B, 50 mg/L; catalyst, 0.8 g/L; H ₂ O ₂ , 40 mM; pH, neutral	99.2% in 1 h	[81]
Pd/g-C ₃ N ₄	BPA	PMS	BPA, 20 mg/L; catalyst, 0.1 g/L; PMS, 1 mM; T, 25 °C; pH, 9	91% in 60 min	[82]
FeO _y /S-g-C ₃ N ₄	Sulfamethoxazole	PMS	sulfamethoxazole, 10 mg/L; catalyst, 0.5 g/L; PMS, 0.8 mM; T, 25 °C; pH, 3.54	100% in 60 min	[83]
Fe(III)-doped g-C ₃ N ₄	AO 7	PMS	AO 7, 8.5 mg/L; catalyst, 0.1 g/L; PMS, 0.1 g/L; pH, 3–4	97% in 30 min	[84]
cryptomelane-type manganese oxide/g-C ₃ N ₄	AO 7	PMS	AO 7, 0.13 mM; catalyst, 0.2 g/L; PMS, 0.65 mM; T, 8 °C; pH, 7.25	88% in 30 min	[85]
carbon and oxygen dual-doped g-C ₃ N ₄	BPA	PMS	BPA, 0.1 mM; catalyst, 0.5 g/L; PMS, 5 mM; T, 30 °C; pH, 6.7	100% in 60 min	[86]
Active carbon/g-C ₃ N ₄	AO 7	PMS	AO 7, 50 mg/L; catalyst, 0.2 g/L; PMS, 0.4 g/L; T, 27 °C; pH, 3.82	100% in 20 min	[87]
Fe-doped g-C ₃ N ₄ /graphite	4-chlorophenol	PMS	4-chlorophenol, 0.1 mM; catalyst, 0.1 g/L; PMS, 0.1 mM; pH, 3	100% in 10 min	[88]
Oxygen-doped g-C ₃ N ₄	BPA	PMS	BPA, 0.05 mM; catalyst, 1 g/L; PMS, 10 mM; T, 30 °C; pH, 3–9	100% in 60 min	[66]
Fe(II)-doped g-C ₃ N ₄	Phenol	PMS	phenol, 0.1 mM; catalyst, 1 g/L; PMS, 5 mM; T, 23 °C; pH, 2.6	100% in 20 min	[89]
Mn ₃ O ₄ /g-C ₃ N ₄	4-chlorophenol	PMS	4-chlorophenol, 50 mg/L; catalyst, 0.3 g/L; PMS, 1 mM; T, 25 °C; pH, 4	100% in 40 min	[90]

Pure g-C₃N₄ holds inert activation performance of oxidants such as H₂O₂ and PMS. Considering that g-C₃N₄ has excellent affinity to entrap transition metal ions, metal doping is the main strategy for improving the catalytic activity. Oh et al. investigated the catalytic activities of Me-doped g-C₃N₄ (Me = Cu, Co, and Fe) as PMS activator for sulfathiazole degradation. Among the prepared catalyst, Co-doped g-C₃N₄ (0.59 wt% Co) exhibited the highest degradation efficiency for sulfathiazole, while excessive metal

doping and surface defects ($-C\equiv N$) had a scavenging effect for $\bullet SO_4^-$ [91]. The authors further studied Fe-doped $g-C_3N_4$ for acid orange 7 degradation, and the non-radical pathway was proposed [84]. Li et al. prepared Fe doped $g-C_3N_4$ as PMS activator for phenolic compounds degradation. $(Fe(V)=O)$ generated from the oxidation of $Fe(III)-N$ was proposed as dominant reactive species [67]. In another work, Fe doped $g-C_3N_4$ was also employed in PMS activation for phenol degradation. Authors investigated the ratio of 46% and 54% of Fe(III) and Fe(II) via Mössbauer spectra, while the XPS survey spectra suggested the primary Fe on the surface of the catalyst was in the 3+ state. It was proposed that the Fe(II) complex heterolyzed at the O-O bond of activated PMS to form $Fe(IV)=O$, which was the primary active species [89]. In PMS/Mn-doped $g-C_3N_4$ system, superoxide radical was firstly generated due to the PMS bounding to the Mn-N site, and singlet oxygen produced by superoxide radical was proposed as the responsible reactive species for acetaminophen degradation [80]. Ma et al. synthesized Cu(I)-doped $g-C_3N_4$ for the removal of rhodamine B in a Fenton-like reaction. Cu(I) could be firmly embedded in $g-C_3N_4$ and reactive species produced by the interaction of H_2O_2 and Cu(I) [81]. The unique adsorption capacity of $g-C_3N_4$ for some organic pollutants also leads to superior degradation performance. Xie et al. investigated that different monochlorophenols isomers (2-chlorophenol, 3-chlorophenol, and 4-chlorophenol) could be degraded efficiently using Co-doped $g-C_3N_4$ as a catalyst in the presence of PMS. It was confirmed that the degradation rate was in the same order as the adsorption quantity [79]. This was attributed to the strong intermolecular forces between pollutant molecules and residual amino groups in the $g-C_3N_4$ fragment [92]. Pd-doped $g-C_3N_4$ was successfully synthesized by anchoring Pd nanoparticles on $g-C_3N_4$ using KBH_4 reduction method, which was regularly active for PMS activation toward bisphenol A removal [82]. Metal oxide such as manganese oxide [85,90] and iron oxide [68] decorated on $g-C_3N_4$ are also employed for organic pollutions degradation via activating H_2O_2 or PMS (Figure 1). Lyu et al. prepared Cu(II)/ $CuAlO_2/g-C_3N_4$ composite as a Fenton-like catalyst. The Cu and C were investigated as dual reaction centers, and C-O-Cu acts as bridges to accelerating electrons transfer [75]. Nonmetal doping is also considered to be an efficient approach to improve electron transfer capability. Electronic structure modulation was achieved in oxygen-doped $g-C_3N_4$ for PMS activation, which was fabricated using urea and oxalic acid dihydrate [66]. The authors further investigated carbon and oxygen doped $g-C_3N_4$ exhibited better PMS activity due to its dual active sites-electron-poor C atoms and electron-rich O atoms [86]. Co-doping of iron and sulfur was found to be an approach to charge distribution and density of $g-C_3N_4$ for PMS activation [83]. To improve its chemical activity and electron transportation ability, Coupling nanocarbon materials $g-C_3N_4$ was developed to realize efficient PMS or H_2O_2 activation [87]. Moreover, combining nanocarbon materials and metal doping was frequently fabricated with $g-C_3N_4$ to exploit both materials' synergistic effect [69,77,88].

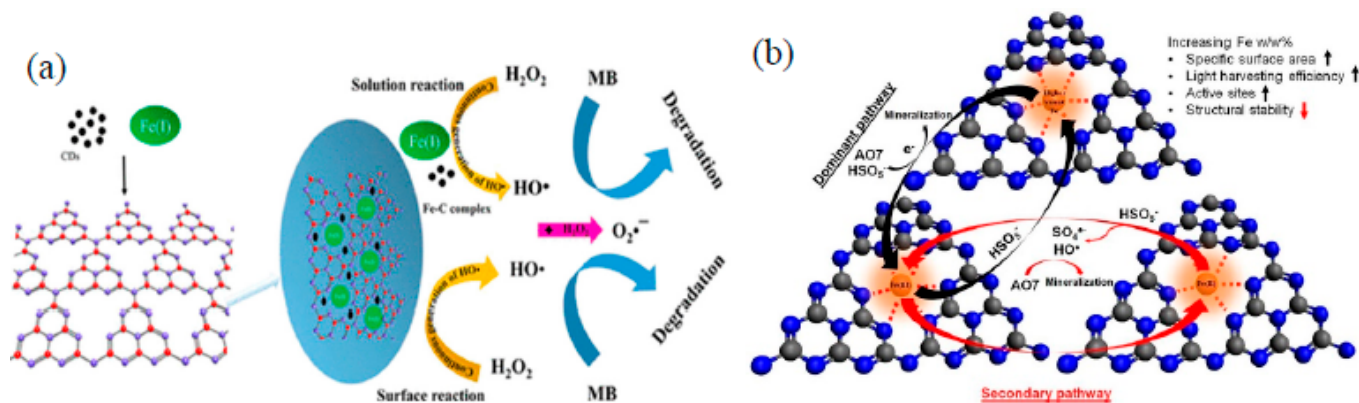


Figure 1. (a) Schematic illustration of the catalytic mechanism of $g-C_3N_4/CDs/Fe(II)$ in the presence of H_2O_2 , reprinted with permission from [68]. Copyright 2019 American Chemical Society. (b) Proposed mechanism of PMS activation by $gCN-Fe_3$ for AO7 removal, reprinted with permission from [83]. Copyright 2018 Elsevier.

3. Photochemical AOPs

Light irradiation is the most widely used method of applying additional energy to assist reactive species generating, which presents the advantages of simple, clean, relatively inexpensive, and efficient. TiO_2 and ZnO were firstly used as photocatalysts for catalytic oxidation of organic contaminants. In this case, photocatalysis induces the formation of h^+ , $\bullet\text{O}_2^-$ and $\bullet\text{OH}$, which act as principle reactive species for pollutants degradation. Consequently, visible light irradiations have been coupled with powerful oxidants such as H_2O_2 and PMS, including catalysis with a modified photocatalyst, resulting in various AOPs. In this section, the applications of these different AOPs as photocatalysis, Photo-Fenton (like) reactions, and photo-assisted sulfate radical based AOPs are summarized. Some representative applications of $\text{g-C}_3\text{N}_4$ based composites as a catalyst in photochemical AOPs are shown in Table 2.

Table 2. $\text{g-C}_3\text{N}_4$ based composites for photochemical AOPs.

Catalyst	Target Contaminants	Light Source	Reaction Conditions	Performance	Ref.
$\text{NiCo}_2\text{O}_4/\text{g-C}_3\text{N}_4$	Carbamazepine	500 W Xenon lamp, Visible light	carbamazepine, 10 mg/L; catalyst, 0.5 g/L; PMS, 1 mM;	100% in 10 min	[93]
$\text{TiO}_2/\text{g-C}_3\text{N}_4$	Acetaminophen	300 W Xenon lamp, Visible light	acetaminophen, 5 mg/L; catalyst, 0.5 g/L; PS, 2 mM; pH, 7	100% in 30 min	[94]
Fe doped $\text{g-C}_3\text{N}_4/\text{graphene}$	Trimethoprim	350 W Xenon lamp, Visible light	Trimethoprim, 0.02 mM; catalyst, 0.5 g/L; PMS, 0.2 mM; pH, 6	100% in 120 min	[95]
$\text{MoS}_2/\text{A g/g-C}_3\text{N}_4$	Tetracycline	300 W Xenon lamp, Visible light	tetracycline, 20 mg/L; catalyst, 0.2 g/L; PMS, 0.1 mM; T, 20 °C; pH, 5.5	98.9% in 50 min	[96]
activated carbon/ $\text{g-C}_3\text{N}_4$	Atrazine	300 W Xenon lamp, Visible light	atrazine, 5 mg/L; catalyst, 1 g/L; PMS, 5 mM; T, 25 °C; pH, 5.56	97.5% in 120 min	[97]
Cobalt-doped $\text{g-C}_3\text{N}_4$	Rhodamine B	500 W halogen tungsten lamp, Visible light	rhodamine B, 10 mg/L; catalyst, 0.4 g/L; PMS, 0.12 mM; T, 25 °C; pH, 4.68	100% in 25 min	[98]
Sulfur-doped/ $\text{g-C}_3\text{N}_4$	Bisphenol A	150 W Visible light lamp	Bisphenol A, 50 mg/L; catalyst, 0.3 g/L; PMS, 0.3 g/L; T, 20 °C; pH, 5	85% in 120 min	[99]
$\text{g-C}_3\text{N}_4$ -imidazole-based ligand- FePcCl_{16}	Carbamazepine	Xenon lamp, Visible light	carbamazepine, 25 μM ; catalyst, 0.1 g/L; PMS, 0.3 mM; pH, 7	95% in 25 min	[100]
Cu-modified alkalized $\text{g-C}_3\text{N}_4$	Rhodamine B	halogen tungsten lamp, Visible light	rhodamine B, 10 mg/L; catalyst, 0.4 g/L; H_2O_2 , 9.8 mM; pH, 4.6	95% in 10 min	[101]

3.1. Photocatalysis

As one typical technique of AOPs, photocatalytic degradation held the advantages of non-toxic, convenient operation, and high efficiency. With the irradiation of UV or visible light with energy larger than the semiconductor's energy gap, the electron-donating and electron-accepting sites are formed in the surface of the semiconducting catalyst. The photo-generated electrons migrate from the valence band (VB) to the corresponding conduction band (CB), leaving holes in the VB, resulting in the electrons and holes occupying the CB and VB, respectively. Holes can directly oxidize pollutants or react with $\text{H}_2\text{O}/\text{OH}^-$ to produce hydroxyl radicals ($E^\theta_{(\bullet\text{OH}/\text{H}_2\text{O})} = 2.8 \text{ eV/NHE}$). Whereas the electrons capture dissolved oxygen (O_2) to yield superoxide radical ($E^\theta_{(\text{O}_2/\bullet\text{O}_2^-)} = -0.3 \text{ eV/NHE}$). The resulting $\bullet\text{O}_2^-$ are subsequently protonated to produce the $\bullet\text{OH}$. Finally, those generated radicals take part in the oxidation of pollutants. In the early seventies, Fujishima and Honda showed the possibility of using the photo-excited semiconductor titanium dioxide (TiO_2) to split water into hydrogen and oxygen in a photo-electrochemical solar cell [102].

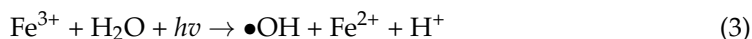
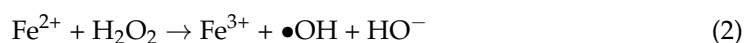
This fundamental work led to developing a new AOP technology, based on semiconductor photocatalysis, for water purification.

g-C₃N₄ compounds have emerged as up-and-coming candidates to replace TiO₂, owing to its graphite-like structure and medium bandgap [103,104]. However, the photocatalytic activity of g-C₃N₄ is still limited by its low electric conductivity and fast recombination of photo-induced carriers [105]. In this regard, modulating the nanostructure of g-C₃N₄ towards enhancing light harvest efficiency and catalytic mass-transfer is highly desirable. Researchers have made great efforts to design g-C₃N₄ with various structures, including 3D porous/nanospheres structure, 2D nanosheet and nanorod, etc. [27,37,106]. Such structures such as 3D porous and 2D nanosheet could provide high surface area, exposing more active sites for catalytic surface reactions. Furthermore, nanostructured g-C₃N₄ could significantly reduce photo-induced carriers' transfer distance, leading to a lower recombination possibility. Moreover, the light quantum efficiencies could be significantly improved by constructing 0D, 1D nanorod, and 2D architectures g-C₃N₄ [107,108].

The VB top of g-C₃N₄ locates at about 1.4 V, leading to a small thermo-dynamic force for organic pollutants oxidation. Moreover, the more positive potential of •OH/H₂O standard redox voltage results the hole cannot directly oxidize the H₂O to generate •OH ($E^{\circ}(\bullet\text{OH}/\text{H}_2\text{O}) = 2.8 \text{ eV/NHE}$). To overcome this shortcoming, several elements of doping have been conducted [44,48,50,108]. Generally, metal doping occurs by inserting into the framework. In contrast, non-metal doping occurs in C or N atoms of g-C₃N₄ replaced by a heteroatom, which could enhance photocatalytic activity via improve the transfer and separation rates of photogenerated carriers and modulate bandgap [41,47]. Constructing heterojunction is another approach to enhance photodegradation performance for g-C₃N₄ [109,110]. Generally, Z-schemed heterojunction could be a good option that possesses higher redox potentials in forming reactive radicals and directly hole oxidation ability [111,112].

3.2. Photo-Fenton Like Processes

The Photo-Fenton process, the combination of ultraviolet or visible light with the conventional Fenton process, can enhance catalysts' catalytic capacity and increase the degradation efficiency of organic pollutants and reduce iron sludge production [113]. The successive and competitive steps reaction mechanism for the photo-Fenton process are shown in Equations (2) and (3).



As shown in Equation (2), Fe²⁺ rapidly reacts with H₂O₂ to generate Fe³⁺. The main form of Fe³⁺ is [Fe(OH)]²⁺ at pH 2.8–3.5, which plays a key role in reactions. Subsequently, the reduction of [Fe(OH)]²⁺ under light irradiation achieves redox recycling (Figure 2). Moreover, •OH can be generated via direct photolysis of H₂O₂ [16]. In the photo-Fenton process, the key is to accelerate the reduction of Fe³⁺ to Fe²⁺ via light irradiation. In the heterogeneous photo-Fenton reactions, the active sites' redox cycle determines the reaction rate [114]. Although g-C₃N₄ cannot act as active sites for H₂O₂ decomposition, unique up conversion property, and substantial nitrogen coordinating sites make it become the ideal support for active sites [115]. In addition, the excellent photocatalytic activities of g-C₃N₄ based composites endue unique advantages as a catalyst for photo-Fenton-like reactions [116]. Metal doping into g-C₃N₄ is an important approach to enhance degradation efficiency in photo-Fenton reactions. Fe-doped g-C₃N₄ has been successfully synthesized by thermal shrinkage polymerization for aqueous organic pollutants degradation in photo-Fenton reactions. Introducing Fe in g-C₃N₄ accelerated the separation of photogenerated electron-holes. The Fe accepts electrons towards rapid reduction from trivalent to divalent, promoting the rapid generation of reactive species [117]. Another report about porous Fe-doped g-C₃N₄ revealed that the porous g-C₃N₄ structures enhance the photo-Fenton

activity, owing to more active sites (Fe-N₄) exposure [118]. An et al. embedded Fe into g-C₃N₄ by pyrolysis of Fe-N-containing precursor and melamine. The high-density Fe-N_x was investigated as a reactive site for H₂O₂ activation [119]. Another strategy used to realize efficient photo-Fenton-based degradation is heterojunction construction, including the Z scheme [120] and type II [121,122]. Zhang et al. prepared MnO₂/Mn-modified alkalized g-C₃N₄ by the calcination-impregnating method. It was proposed that Z-scheme charge transfer accelerated the redox cycle of the Mn⁴⁺/Mn³⁺/Mn²⁺ [123].

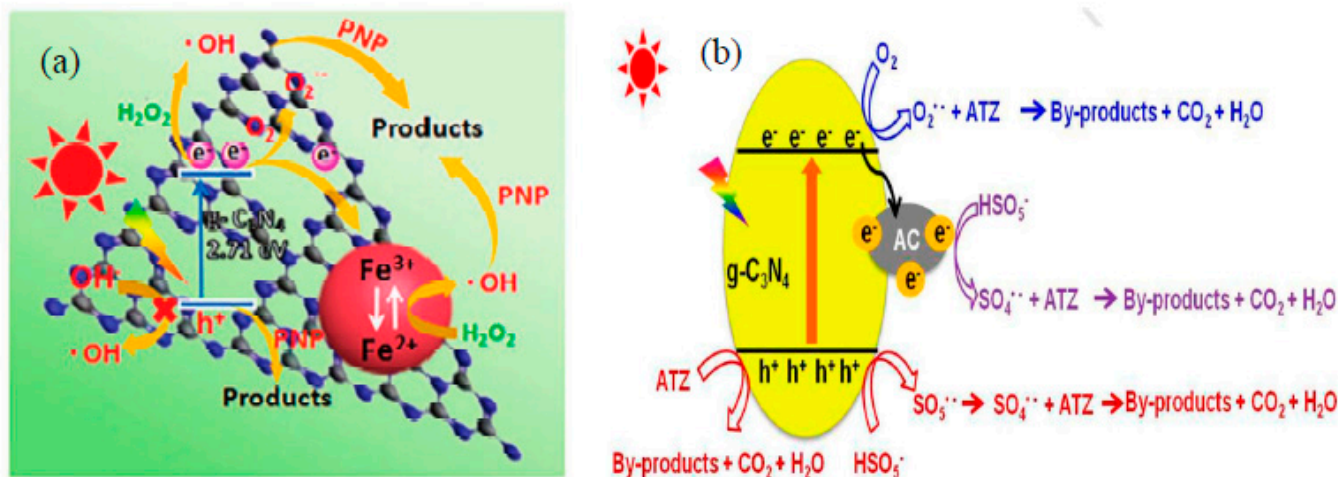
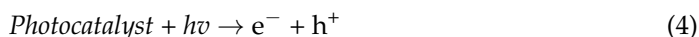


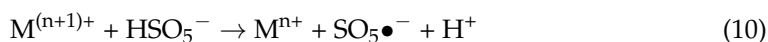
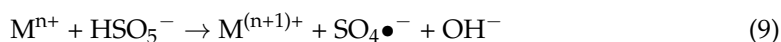
Figure 2. (a) schematic illustration of the catalytic mechanism of Fe₂O₃ QDs/g-C₃N₄-900 in H₂O₂/vis system, reprinted with permission from [113]. Copyright 2019 John Wiley & Sons, Inc. (b) Mechanism of photocatalytic degradation of atrazine with PMS, reprinted with permission from [96]. Copyright 2018 Elsevier.

3.3. Photo-Assisted Sulfate Radical Based AOPs

Sulfate radical-based advanced oxidation processes (SR-AOPs) are increasingly gaining attention as an effective solution to the destruction of recalcitrant organics in water [124]. Among various approaches to generate sulfate radicals via activation of additional sources of reactive species (such as peroxymonosulfate (PMS) and persulfate (PS)), the photo-activation in the presence of a heterogeneous catalyst is worth mentioning [65]. The general mechanism is presented in Equations (4)–(8).



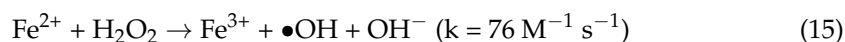
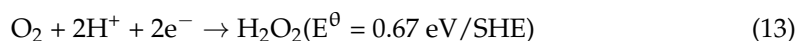
Firstly, photocatalysts are excited under light irradiation to form photo-induced electrons and holes. Then the $\bullet\text{SO}_4^-$ and $\bullet\text{OH}$ are generated through the combination of electrons and PMS or PS. When transition metals are constructed into photocatalysts, they could be potential reactive sites for PMS/PS activation (shown in Equations (9)–(12)).



Similar to the application in photo-Fenton-like reactions, g-C₃N₄ generally plays as reactive site supporters or participate in heterojunction construction in photo-assisted sulfate radical based AOPs. The TiO₂/g-C₃N₄ composite was fabricated for paracetamol photocatalytic degradation in the presence of visible light and persulfate. As prepared composite held type II heterojunction, which inhibits the electron-hole recombination in photocatalyst and adding persulfate increased 13 times degradation rate [94]. Liang et al. prepared porous 0D/3D NiCo₂O₄/g-C₃N₄ composite for carbamazepine removal. 99% of degradation was achieved in 10 min under visible light irradiation [93]. Jin et al. constructed Z-scheme MoS₂/Ag/g-C₃N₄ via a method of chemical electrostatic adsorption. The deposited Ag further enhances photocatalytic activity via improving light utilization ability and the separation rate of photogenerated e⁻/h⁺ pairs. The results indicated that the presence of PMS dramatically accelerates the photocatalytic reaction [96]. Through metal ions such as Fe and Co doping, enhancing photocatalytic activity and improving PMS activation could synchronize implementation towards an efficient organic pollutant removal [95,98].

4. Electrochemical AOPs

Electrochemical advanced oxidation processes (EAOPs) have gained increasing attention as one of the most potent classes of AOP [125–127]. The direct electrochemistry oxidation known as EAOP is anodic oxidation (AO), which is conveniently operated and widely used [125,128]. Electro-Fenton reaction (EF), also known as EAOPs involving the Fenton process, constitutes a clean and effective way to accelerate contaminants degradation via in situ electrocatalytically generated H₂O₂ from the oxygen reduction reaction (ORR) and regeneration of Fe²⁺ on the cathode [126] (Equations (13)–(15)).



One of the EF technology's key points is to fabricate a cathode with high efficiency towards ORR [129,130]. G-C₃N₄ have been recently considered for cathode modification, which can provide more active sites than other nitrogen-containing materials for electrocatalytic reactions to serve as ORR catalyst. Zhu et al. prepared g-C₃N₄ doped gas diffusion electrode as a cathode in electro-Fenton processes, and the H₂O₂ accumulation reached 2.59 mg h⁻¹ cm⁻², while it is 1.86 mg h⁻¹ cm⁻² using cathode without g-C₃N₄ doping [131]. Loading g-C₃N₄ on activated carbon fiber electrode was investigated as an efficient way to boost electrocatalytic activity for rhodamine B degradation in electro-Fenton processes, which was due to the g-C₃N₄ accelerated the cyclic utilization of ferrous ions on the surface of activated carbon fiber [132]. Furthermore, light irradiation was combined with electro-Fenton processes using g-C₃N₄/activated carbon fiber and g-C₃N₄/carbon nanotube electrodes as cathodes, which achieved higher H₂O₂ production [133,134]. Moreover, g-C₃N₄ based composites such as ZnO/g-C₃N₄ and TiO₂/g-C₃N₄ served as photoanodes were applied in photo-assisted anodic oxidation of organic pollution, which was attributed to the superior photocatalytic activity of g-C₃N₄ [135,136].

5. Conclusions and Perspectives

This review has shown the application of g-C₃N₄ based composites in various AOPs, including chemical AOPs, photochemical AOPs, and electrochemical AOPs. In the past years, g-C₃N₄ has been served as an excellent candidate for pollutants removal from water and wastewater thanks to the tunable electronic structure and excellent physicochemical stability. However, pristine g-C₃N₄ still suffered numerous issues such as smaller surface area, faster recombination of charge, and poor oxidant decomposition activity. It can be summarized the main challenges and strategies in the following aspect:

- (1) For chemicals AOPs, pristine g-C₃N₄ holds inert activation performance of oxidants such as H₂O₂ and PMS. Considering that g-C₃N₄ has excellent affinity to entrap transition metal ions, metal doping is the main strategy to improve the catalytic activity and electrons transfer capability. Combining nanocarbon materials and metal doping was also frequently fabricated with g-C₃N₄ to exploit both materials' synergistic effect.
- (2) In the case of the photochemical technologies, the challenges for enhancing the photodegradation performance could be ascribed as (1) expand the absorption edge of g-C₃N₄ and enhance the light-harvesting capability, (2) make separating charge more efficient and suppress the recombination of photo-induced carriers, and (3) coordinate energy band structures to enhance reduction or oxidative capacity. The recent progress of g-C₃N₄ based composites regarding the photodegradation performance improvement include nanostructure design, element doping, hetero-junction construction, and co-polymerization.
- (3) Cathode modification in electron-Fenton processes is the major application of g-C₃N₄ based composites in electrochemical AOPs. The key to improving EF performance is to fabricate a cathode with superior ORR efficiency towards higher H₂O₂ production.

Although g-C₃N₄ based composite shows promising activity in various AOPs, the synergic effect between different components is still not fully understood. In addition, developing methods towards stable morphology/structure synthesis is highly demanded for practical applications.

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