



Recent progress on non-metallic carbon nitride for the photosynthesis of H₂O₂: Mechanism, modification and *in-situ* applications

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ABSTRACT

Photocatalytic hydrogen peroxide (H₂O₂) production has been considered as a promising strategy for H₂O₂ synthesis due to its environmentally friendly. Among various photocatalysts, carbon nitride-based materials are excellent candidates for H₂O₂ production because of their excellent visible-light response, low cost and high stability. In this review, we summarize in detail the research progress on the photocatalytic production of H₂O₂ by carbon nitride. First, we summarize the basic principles of photocatalysis and photocatalytic H₂O₂ production. Second, the classification and modification methods of carbon-nitride-based materials are discussed, including morphology modulation, noble metal loading, defect control, heterojunction regulation, molecular structure engineering and elemental doping. Finally, the different *in-situ* applications of H₂O₂ via photosynthesis were discussed, including disinfection and antibiotic resistant genes degradation, organic pollutants degradation, medical applications and fine chemical synthesis. This review brings great promise for *in-situ* H₂O₂ photosynthesis, which is expected to serve as a key component in future applications.

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1. Introduction

Hydrogen peroxide (H₂O₂) serves as an environmentally friendly oxidizing agent and one of the most important chemicals for industrial application [1,2]. Presently, the industrial method to synthesis H₂O₂ always involves the anthraquinone process, which always depends on organic solvents and noble metal catalysts, bringing high energy consumption [3]. Another traditional method is the direct synthesis of H₂O₂ from H₂ and O₂ on Pd-based catalysts [4]. Nevertheless, this approach also exhibits numerous challenges, including the explosive properties of H₂/O₂ mixture and the decomposability of H₂O₂ in alkaline environment. Therefore, it is imperative to develop cost-effective, green and safe methods for H₂O₂ synthesis to tackle the energy and environmental challenges.

Photocatalysis has been considered as the most promising method to synthesize H₂O₂. Since Baur *et al.* reported that H₂O₂ was produced from glycerol and glucose using ZnO under light in 1927 [5], a large number of researchers [6–14] have been working on the photocatalytic H₂O₂ production. This process of photocatalytic H₂O₂ production involves utilizing light energy to ac-

tivate a semiconductor material, which facilitates the reaction between O₂ and H₂O to generate H₂O₂. This method offers several advantages over traditional synthesis methods, including minimizing waste generation and reducing energy consumption.

Currently, among various photocatalytic materials for H₂O₂ production, non-metallic carbon nitride has garnered significant attention. In 2009, Wang *et al.* first reported that carbon nitride was capable of photocatalytic H₂ production [15]. This has sparked widespread exploration of carbon nitride within the field of photocatalysis [16–22]. Carbon nitride does not require any addition of metal ions, and demonstrates excellent chemical stability and large specific surface area, facilitating catalytic reactions. Furthermore, carbon nitride has special band positions, which are well-suited for oxygen reduction reaction (ORR) and water oxidation reaction (WOR) to produce H₂O₂.

Although there are many reviews on photocatalysis by carbon nitride, they always focused on H₂ production and CO₂ reduction. There is still a lack of a separate and detailed review on photocatalytic H₂O₂ production, especially with *in-situ* applications. Based on this, we herein summarize the relevant research on photocatalytic synthesis of H₂O₂ by carbon nitride. As shown in Fig. 1, it begins with the fundamental mechanisms of photocatalysis. Secondly, different modification methods for carbon nitride-based materials, including morphology tuning, noble metal loading, defect

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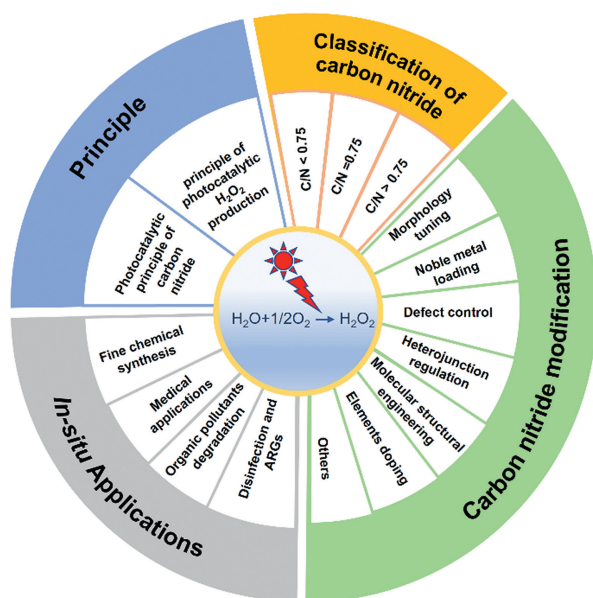


Fig. 1. Principle, classification, modification of carbon nitride-based catalysts for H_2O_2 photosynthesis and their *in-situ* applications.

control, heterojunction regulation, molecular structure engineering, and elements doping, are summarized. Finally, various *in-situ* applications of H_2O_2 photosynthesis are discussed, including disinfection and antibiotic resistant genes (ARGs) degradation, organic pollutants degradation, medical applications and fine chemical synthesis.

2. Principle

2.1. Photocatalytic principle of carbon nitride

Photocatalysis refers to the process of converting light energy into chemical energy for catalyzing chemical reactions under light irradiation [23,24]. With light irradiation, photocatalysts could absorb sufficient light energy, and then the photogenerated electrons (e^-) could be excited from valence band (VB) to conduction band (CB), while positively charged holes (h^+) remain in the VB. Then, e^- and h^+ migrate to the surface of the catalyst and participate in redox reactions. At the same time, photoinduced e^- and h^+ also undergoes bulk recombination and surface recombination, resulting the quenching of e^- and h^+ (Fig. 2a).

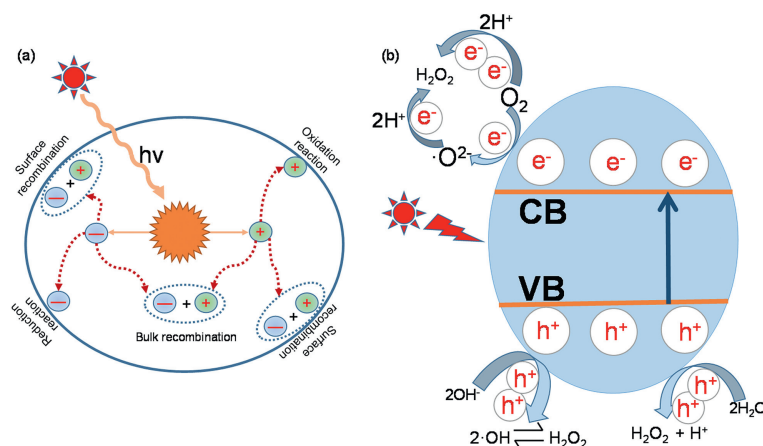
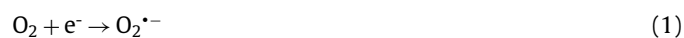


Fig. 2. (a) Photoexcitation and charge decay pathways in photocatalysts. (b) Schematic diagram of photocatalytic system for producing H_2O_2 .

2.2. Principle of photocatalytic H_2O_2 production

The currently recognized mechanisms of photocatalytic H_2O_2 production mainly include ORR and WOR, where ORR occurs at CB, while WOR occurs at VB (Fig. 2b). In details, there are two main ways for ORR, including two-step one-electron ($1e^-$) process Eqs. 1–4, with a reduction potential of $E^0 = -0.33\text{ V}$ and a one-step two-electron ($2e^-$) process (Eq. 5), with a reduction potential of $E^0 = +0.68\text{ V}$. Firstly, $1e^-$ ORR produces superoxide radical ($\text{O}_2^{\bullet-}$) (Eq. 1), which further reacts with H^+ to produce $\text{HO}_2^{\bullet-}$ (Eq. 2). Then, $\text{HO}_2^{\bullet-}$ radical can be readily followed by another $1e^-$ reduction to produce HO_2^- anion (Eq. 3). Finally, HO_2^- reacts with H^+ to produce H_2O_2 (Eq. 4). In addition, a direct one-step $2e^-$ reduction to produce H_2O_2 involves the combination of O_2 and 2H^+ and $2e^-$ (Eq. 5) [25,26].



The above reaction processes are main paths of H_2O_2 production. Besides, there are also some pathways by WOR [27–29]. WOR is achieved through that photogenerated h^+ reacts with H_2O to generate H_2O_2 , including direct $2e^-$ WOR and $1e^-$ WOR. In detail, photoinduced h^+ directly oxidizes H_2O to H_2O_2 ($2e^-$ WOR) (Eq. 6) with an oxidation potential of $E^0 = +1.76\text{ V}$. In addition, h^+ first oxidizes hydroxide ions (OH^-) to generate hydroxyl radicals ($\bullet\text{OH}$) (Eq. 7), and then indirectly generates H_2O_2 through the combination of $2\bullet\text{OH}$ ($1e^-$ WOR) (Eq. 8). The oxidation potential require for this process is $E^0 = +1.99\text{ V}$ [30].



3. Classification of carbon nitride

Since C_3N_4 -based carbon nitride was reported by Wang in 2009 [15], many researchers have devoted their attention to this kind of material. In addition to C_3N_4 with a carbon to nitrogen (C/N) ratio of 0.75, other carbon nitrides with different C/N ratios have been synthesized, including a class of carbon nitride with $C/N < 0.75$ (C_3N_5 , C_3N_6 , C_3N_7 , etc.), and that with $C/N > 0.75$ (C_3N_3 , $C_{3.6}N$, C_5N_2 , etc.) (Fig. 3).

3.1. $C/N=0.75$

The most common carbon nitride is material with $C/N=0.75$, corresponding to C_3N_4 . C_3N_4 structure has several isomers, such as α - C_3N_4 , β - C_3N_4 , cubic- C_3N_4 , pseudo- C_3N_4 and graphite- C_3N_4 (g- C_3N_4). Among these, g- C_3N_4 structure is the most stable isomer under environmental conditions and serves as the most commonly used photocatalyst [17,19]. The group of g- C_3N_4 mainly include triazine-based g- C_3N_4 and tri-s-triazine (heptazine)-based g- C_3N_4 [21].

Nowadays, lots of synthetic methods were developed to prepare g- C_3N_4 [18]. Wang *et al.* proposed a method for synthesizing g- C_3N_4 via putting cyanamide to temperatures ranging from 400 °C to 600 °C (ramp rate: 2.2 °C/min) for 4 h [15]. Besides, C_3N_4 could also be synthesized using melamine as a precursor [31–33]. Firstly, melamine was formed through polymerization and condensation, followed by rearrangement into tri-s-triazine based compounds at around 390 °C, further condensation into polymers, networks and potentially the final polymeric g- C_3N_4 at around 520 °C [33]. Other prevalent precursors included urea and dicyandiamide (DCD). For example, Li *et al.* synthesized g- C_3N_4 by thermal polymerization using these two precursors [34]. In detail, 10.0 g of urea and DCD powders were put in an Al_2O_3 crucible with a half-cover state. The mixture underwent heating in a muffle oven at a rate of 10 °C/min until reaching a certain temperature, which was maintained for 3 h to obtain g- C_3N_4 .

3.2. $C/N < 0.75$

Carbon nitrogen with $C/N < 0.75$ are called N-rich carbon nitrides. This kind of carbon nitride usually exhibits higher photocatalytic activity due to its nitrogen functional groups, which provide many active sites [35–40]. The N-rich carbon nitrogen mainly include C_3N_5 , C_3N_6 and C_3N_7 .

For C_3N_5 , Gillan *et al.* proposed a method for synthesizing this material via azide-based molecular precursors in 2000 [41]. Azide and triazine were selected as the precursors. The triazido-triazine was decomposed in a high-pressure steel reactor under 6 atm N_2

with a gradual heating to 185 °C and then it was maintained at this temperature for 24 h to produce C_3N_5 . For C_3N_6 , Talapaneni and co-workers reported a method to synthesize C_3N_6 at a low temperature to avoid traditional high-temperature process by using amino guanidine [39]. Besides, other C_3N_5 materials (MCN-4-x) were synthesized using SBA-15 as a hard template [39]. The N/C ratios of MCN-4-x samples gradually increased with the increase of initial mixing temperature. The synthesis of C_3N_7 was achieved by pyrolysis of 5-amino-1H-tetrazole (5-ATTZ) at 250 °C via nanotemplating approach [40]. The detailed steps were as follows: 3.14 g of 5-ATTZ was dispersed in 11.0 g of deionized H_2O at 50 °C, and then 1.0 g of KIT-6 was added. The suspension was mixed and heated in an oven at 100 °C for 6 h. The obtained sample was calcined at 250 °C under N_2 for 4 h. Then, KIT-6 crystals were removed with 5 wt% hydrofluoric acid and ethanol. Finally, C_3N_7 was obtained by drying at 100 °C for 12 h.

3.3. $C/N > 0.75$

Carbon nitrogen with $C/N > 0.75$ is called C-rich carbon nitride, which always displays better conductivity and mechanical properties [42–45]. There are several techniques for synthesizing this kind of carbon nitride. For example, Kim *et al.* prepared the synthesis of N-deficient C_3N_3 and C_3N -based carbon nitrides from various single-molecule precursors [42]. They impregnated three precursors, including phosphomolybdic acid hydrate (PMA), dithioamide (DTO) and 5-ATTZ, into the KIT-6 porous hard template and then calcined them at high temperature to finally obtain C_3N_3 hybrid. Similarly, Feng and Li *et al.* synthesized C_3N_3 by using Ullmann reaction to achieve homocoupling of aryl halides [43]. They adopted cyanuric chloride ($C_3N_3Cl_3$) for polymerization within a sealed tube, utilizing copper foil as the substrate to obtain a layered C_3N_3 structure. Xu *et al.* also explored hexamethylenetetramine as a molecular precursor for the synthesis of $C_{3.6}N$ by thermal cleavage within a hard template (SBA-15) [44]. Ma *et al.* synthesized C_5N_2 through Schiff reaction using melamine and *p*-phthalaldehyde dyes as monomers, with an optimal molar ratio of 3:2 [45]. With the increase of synthesis time, the yield of insoluble solid product gradually increased, and C_5N_2 could be obtained after 5 days.

4. Carbon nitride modification

Carbon nitride has been demonstrated as a promising photocatalytic material for H_2O_2 production. However, the efficiency of carbon nitride in photocatalytic H_2O_2 production always depends on various factors. Therefore, we provide a comprehensive summary of modifications to improve the performance of carbon nitride, including morphology tuning, noble metal loading, defect control, heterojunction regulation, molecular structure engineering and elements doping, and some other modifications (Fig. 4).

4.1. Morphology tuning

Morphology tuning is a crucial technique that enables the manipulation of material morphology and structure, thereby influencing the performance of photocatalytic H_2O_2 production [25,46,47]. By controlling the synthesis conditions and incorporating template agents, surface modifiers, and other additives, it can be employed to achieve different morphology of carbon nitride [48]. For example, different carbon nitride with desired shapes, such as nanorods, nanosheets, and porous structures, can be obtained by adjusting the solution concentration or employing specific template agents.

Xu *et al.* synthesized uniformly porous carbon nitride nanotubes using a supramolecular self-assembly method [47]. They obtained uniform carbon nitride nanotubes (TCN-520) with 1–3 μm

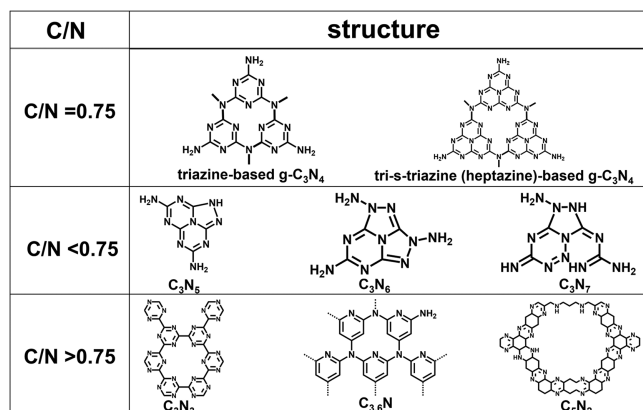


Fig. 3. Types of main carbon nitride.

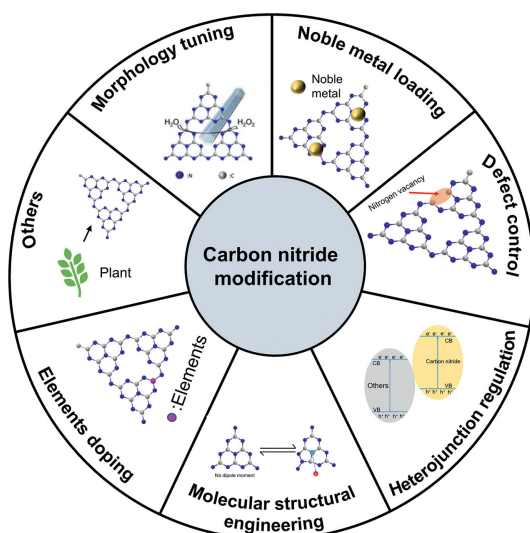


Fig. 4. Different modification methods of carbon nitride.

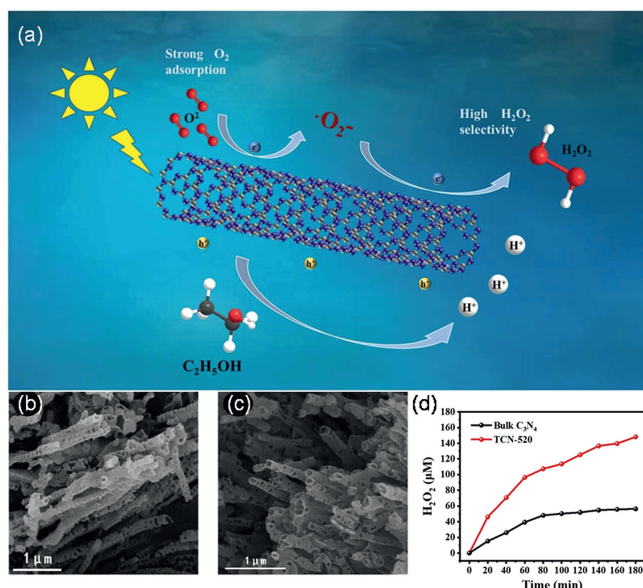


Fig. 5. (a) Mechanism of porous carbon nitride nanotube for H₂O₂ production; (b, c) TCN-520 transmission electron microscope (TEM) images of TCN-520. (d) Performance of TCN-520 and bulk C₃N₄ for H₂O₂ generation. Copied with permission [47]. Copyright 2023, Elsevier.

in length and a diameter of 10–20 nm *via* a calcination temperature of 520 °C. TCN-520 can generate H₂O₂ by the two-step 1e⁻ process (Fig. 5a). Additionally, the nanotube displayed a porous structure due to the gas released during the calcination process (Figs. 5b and c). TCN-520 exhibited a higher photocatalytic H₂O₂ yield than pristine bulk carbon nitride (Fig. 5d). The reasons could be attributed to that the nanotube structure had a large specific surface area, outstanding electron transport rate, O₂ adsorbing capability and high selectivity of 2e⁻ ORR.

In addition, three-dimensional (3D) structures also can enhance the performance of carbon nitride by regulating the surface area. For example, Li *et al.* synthesized ultrathin graphitic carbon nitride nanoplates with thickness of 1–3 nm for facilitating H₂O₂ production [25]. The ultrathin structure displays a larger surface area with a large number of active sites, and enhances redox capability compared to the pristine counterpart. The H₂O₂ production rate of ob-

tained nanoplates was 43.07 μmol g⁻¹ h⁻¹, which is about 4 times that of pristine g-C₃N₄.

4.2. Noble metal loading

The loading of noble metal mainly includes adding noble metal nanoparticles to the surface or interior of carbon nitride, aiming to boost the charge separation and enhance their photocatalytic performance. Noble metal nanoparticles generally serve as active sites in photocatalytic reactions to promote photogenerated carrier separation and selective reactions, thereby increasing the yield of H₂O₂ [49–52]. The selection of appropriate noble metal nanoparticles is crucial for increasing catalytic activity, as different metals show distinct electronic structures. By controlling the properties and interactions of noble metal nanoparticles, enhanced light absorption and carrier separation can be achieved, thereby increasing the yield of H₂O₂.

For example, Song *et al.* reported a method for loading and stabilizing gold nanoparticles on C₃N₄ substrates (Fig. 6a), which simultaneously enhanced the stability of the catalyst and the performance of H₂O₂ photosynthesis. The precursor was annealed for 2 h at 500 °C with N₂, exhibiting the best performance of 1320 μmol/L, which is 2.3 times that of pure C₃N₄. Among the various samples synthesized, Au/CN-100 had the largest production amount (Fig. 6b) [50].

Cai *et al.* prepared silver-decorated ultrathin g-C₃N₄ nanosheets (Ag@U-g-C₃N₄-NS) through post gas etching (PGE) technology (Fig. 6c). As shown in Fig. 6d, U-g-C₃N₄-NS produced the higher yield of H₂O₂ (0.414 × 10⁻⁶ mol L⁻¹ min⁻¹) than that of B-g-C₃N₄ (<0.03 × 10⁻⁶ mol L⁻¹ min⁻¹). The improved performance was due to that the large specific surface area of ultrathin g-C₃N₄ nanosheets (U-g-C₃N₄-NS) provided a large number of active sites [51], and Ag nanoparticles in Ag@U-g-C₃N₄-NS catalyst acted as an electronic bridge of photogenerated electron to form Schottky barriers, thereby reducing the charge recombination and improving hole utilization [52].

4.3. Defect control

Recently, defect control has been considered as an important way to improve photocatalytic efficiency [53–56]. The formation of defects can influence the electronic structure, energy band structure, and surface active sites of material, thereby promoting carriers separation and prolonging the lifetime of carriers. Moreover, the crystal structure and morphology of carbon nitride can also be changed by defect control. Lots of studies have clearly demonstrated the advantages of defect control in carbon nitride for boosting photocatalytic production of H₂O₂ [57–61].

Zhu's group synthesized reduced g-C₃N₄ with N vacancies by heat treatment with NaBH₄ under N₂ [58]. The N vacancies in C₃N₄ were formed by reduction treatment involving a cleavage reaction with the pyridine nitride of s-triazine-C₃N₄ (Fig. 7a). This led to the narrowing of energy bandgap to boost visible light-driven WOR capability [62,63]. In particular, N vacancies can promote the spatial separation of photogenerated e⁻ and h⁺. As shown in Fig. 7b, R₃₇₀-CN could achieve photocatalytic production of H₂O₂ from pure H₂O and ambient atmosphere under visible light, reaching 170 μmol/L in 1 h. Zheng *et al.* prepared N-vacancy CN spheres (NVCNS) in H₂ plasma by low-temperature heating (Fig. 7c). Under visible light, NVCNS containing N vacancies could produce 250 μmol H₂O₂ in 2 h (Fig. 7d) [59]. In another investigation, Han *et al.* achieved the incorporation of two distinct N vacancy types into g-C₃N₄ through the pyrolysis of melamine under both Ar and NH₃, coupled with subsequent HNO₃ oxidation. It was worth noting that the pyrolysis atmosphere had a significant effect for producing N vacancies. The N vacancies in N groups (N₃C) and sp² hybrid N

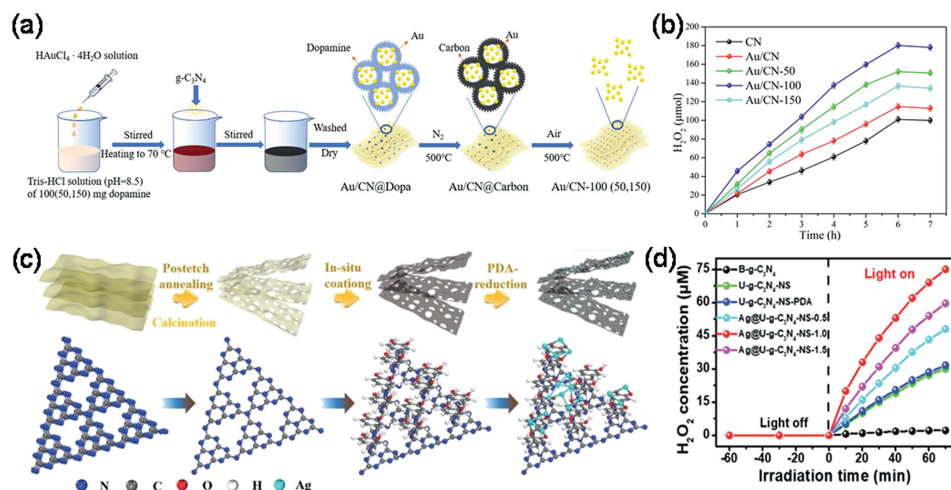


Fig. 6. (a) The preparation process for samples (the Au load is 2 wt% of carbon nitride). (b) Photocatalytic yield of H_2O_2 for different samples. Copied with permission [50]. Copyright 2022, Royal Society of Chemistry. (c) Stepwise representation of route to Ag NP-decorated ultrathin $\text{g-C}_3\text{N}_4$ nanosheets. (d) H_2O_2 production is accomplished by photocatalysis. Copied with permission [51]. Copyright 2019 Wiley-VCH.

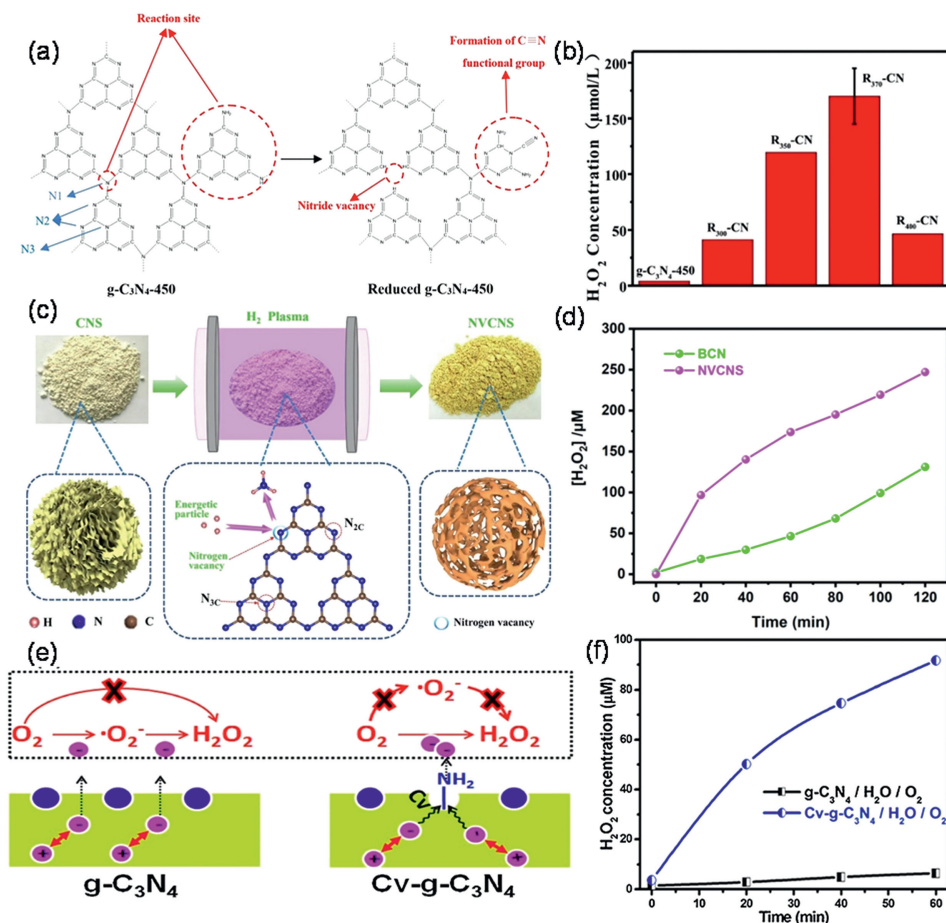


Fig. 7. (a) Possible N vacancies synthetic process. (b) Performance of different photocatalysts for H_2O_2 generation. Copied with permission [58]. Copyright 2018, Elsevier. (c) The schematics of H_2 plasma process of NVCNS. (d) Yield of photocatalytic production of H_2O_2 ($>420\text{ nm}$). Copied with permission [59]. Copyright 2022, Elsevier. (e) Mechanisms of H_2O_2 production with $\text{Cv-g-C}_3\text{N}_4$. (f) The concentration of H_2O_2 generated via $\text{g-C}_3\text{N}_4$ and $\text{Cv-g-C}_3\text{N}_4$. Copied with permission [61]. Copyright 2016, Elsevier.

atoms (N_2C) could be formed under NH_3 pyrolysis and Ar pyrolysis, respectively [60]. For carbon nitride, C vacancies building is another way for defect control. For example, Li *et al.* proposed a thermal annealing method under Ar atmosphere to generate C vacancies in $\text{g-C}_3\text{N}_4$ ($\text{Cv-g-C}_3\text{N}_4$) [61]. The results showed that $\text{g-C}_3\text{N}_4$ with C vacancies exhibited higher photocatalytic production

of H_2O_2 compared to pure $\text{g-C}_3\text{N}_4$. The increased photocatalytic performances were attributed to extending visible light absorption and increasing charge separation with $\text{Cv-g-C}_3\text{N}_4$. Furthermore, the NH_3 groups surrounding carbon vacancies changed the H_2O_2 production pathway from two-step indirect reaction to one-step direct reaction, thereby accelerating the synthesis of H_2O_2 (Fig. 7e).

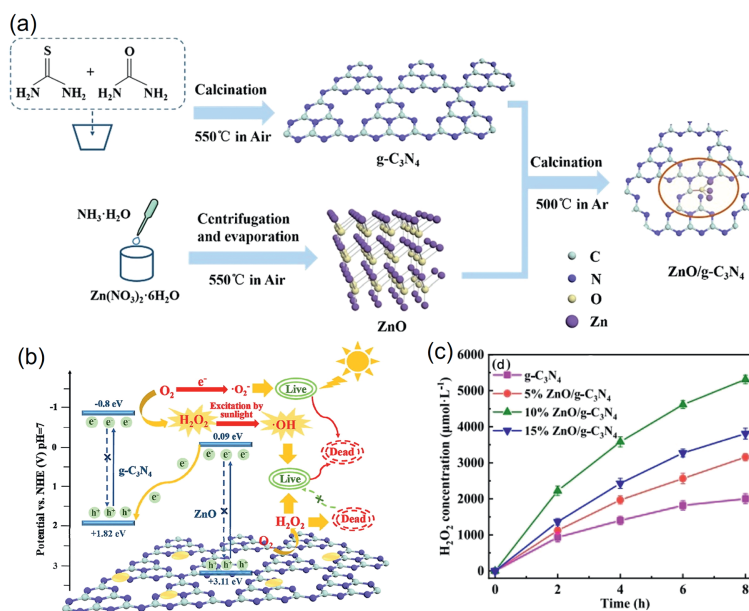


Fig. 8. (a) Synthetic process of ZnO/g-C₃N₄ heterojunction photocatalyst. (b) Mechanisms of H₂O₂ production and photocatalytic sterilization. (c) Performance of different catalysts for H₂O₂ generation. Copied with permission [66]. Copyright 2021, Elsevier.

As shown in Fig. 7f, the yield of Cv-g-C₃N₄ was 14 times that of g-C₃N₄.

4.4. Heterojunction regulation

Heterojunction regulation is an effective approach for regulating the production of H₂O₂ from carbon nitride. A built-in electric field can be generated in the interface between both different materials, thereby enhancing the catalytic activity and selectivity. Common control strategies include semiconductor heterojunctions [64], metal-semiconductor heterojunctions [65,66] and two-dimensional material composite heterojunctions [67]. These approaches can tune electron transport and interfacial interactions to enhance the efficiency of H₂O₂ generation [68].

For example, Geng *et al.* used a hydrothermal method to prepare ZnO/g-C₃N₄ heterojunction catalyst (Fig. 8a), which significantly improved the production of H₂O₂ [66]. The reaction mechanisms were shown in Fig. 8b. 10 wt% ZnO/g-C₃N₄ exhibited a H₂O₂ yield of 5312.45 μmol/L after 8 h, which was 2.65 times higher than that of pure g-C₃N₄ (Fig. 8c). The high performance was due to: (i) Enhancement of light-absorbing capacity by constructing a heterojunction between ZnO and g-C₃N₄ from ultraviolet to visible light; (ii) Separation of photogenerated e⁻ and h⁺ at the interface between ZnO and g-C₃N₄; (iii) Facilitating electron transfer between ZnO and g-C₃N₄.

4.5. Molecular structural engineering

Charge recombination in the photocatalysts includes bulk recombination (BR) and surface recombination (SR). SR behavior usually takes tens of nanoseconds to occur, whereas BR often occurs within picoseconds. Therefore, the reduction of BR plays a vital role in improving the photocatalytic activity of carbon nitride. Utilizing structural engineering to enhance dipole field effects is an important strategy to promote directional bulk electron separation [69,70].

Li *et al.* reported a novel process with dipole field-driven spontaneous polarization in N-rich triazole-based carbon nitride (C₃N₅) to boost the photocatalytic synthesis of H₂O₂ [71]. The existence of dipole field facilitated redox reactions by influencing its elec-

tron cloud distribution. Fig. 9a illustrated the dipole field and its variations under external force. Density functional theory (DFT) revealed that C₃N₅ displayed a dipole moment, while C₃N₄ did not show any dipole moment (Fig. 9b). The significant dipole moment in C₃N₅ enabled the utilization of dipole field-induced spontaneous polarization to leverage photogenerated charge separation dynamics. The results showed that the H₂O₂ production rate of C₃N₅ was 3809.5 μmol g⁻¹ h⁻¹ (Fig. 9c), which was superior to the performance of pure g-C₃N₄. The reaction mechanism was shown in Fig. 9d. This straightforward technique address a critical need in facilitating the transfer of photoinduced carriers through the introduction of structural engineering induced dipole field.

4.6. Elements doping

Doping elements can determine the electronic structure, lattice configuration, and surface characteristics of carbon nitride, thereby realizing the regulation of catalytic performance [72,73]. By introducing elements, the electronic structure of material, additional active sites, and the electron transfer rate are improved. The metal doping including Cu or Fe, *etc.* can highly improve the catalytic activity of H₂O₂ [74–76]. In addition, non-metallic doping elements such as nitrogen, phosphorus, sulfur, and selenium also tune the selectivity of carbon nitride to synthesis H₂O₂ [27,77,78].

For example, Li *et al.* doped Cu or Fe into C₃N₄ by using thermal polymerization. The H₂O₂ yield of Cu- and Fe-doped C₃N₄ photocatalysts was 24.5 and 13.7 μmol/L, respectively, which was much higher than the original C₃N₄ [74]. There are also corresponding examples for non-metal doped carbon nitride. Cao *et al.* obtained phosphorus-doped porous carbon nitride (CPN) as a metal-free photocatalyst by uniformly mixing and calcining urea and phosphorus chloride trimer through hydrothermal method. The morphology of CPN were shown in Figs. 10a and b, with a porous structure. The elemental mappings of CPN confirmed the presence of C, N and P (Figs. 10c-e). A dual-channel pathway involving both WOR and ORR was observed, achieving a remarkable H₂O₂ yield of 1968 μmol g⁻¹ h⁻¹ without any sacrificial agents and co-catalysts, under ambient conditions (Fig. 10f) [27].

Besides, dual elements co-doping is also developed to modify carbon nitride [79–81]. Liu *et al.* designed oxypotassium double

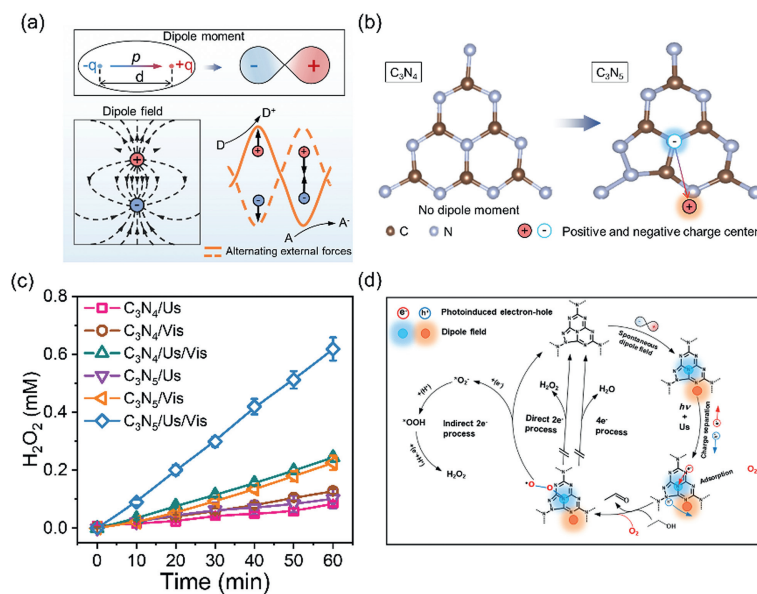


Fig. 9. (a) Dipole moment and the distribution of electron clouds. (b) The structural units of C_3N_4 and C_3N_5 . (c) Time profiles of H_2O_2 production by photocatalysis of C_3N_3 and C_3N_4 under different reaction conditions. (d) Possible mechanisms of H_2O_2 generation. Copied with permission [71]. Copyright 2023, Springer Nature.

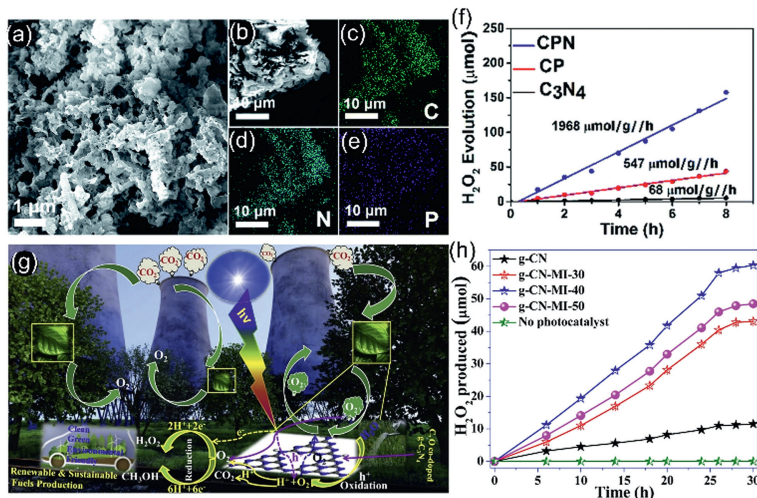


Fig. 10. (a, b) scanning electron microscope (SEM) images of CPN. (c–e) The corresponding energy dispersive spectrometer (EDS) elemental mappings of constituent elements for CPN. (f) Time profiles of photocatalytic H_2O_2 production with different catalysts. Copied with permission [27]. Copyright 2020, Royal Society of Chemistry. (g) Mechanisms of CO_2 reduction and H_2O_2 production process. (h) Photocatalytic H_2O_2 production of various catalyst. Copied with permission [81]. Copyright 2019, Elsevier.

heteroatom polymerized carbon nitride (O/K-CN) [79]. The addition of heteroatoms not only improved the adsorption of O_2 and the carrier separation, but also promoted the production of $\cdot\text{OOH}$. The rate of H_2O_2 generation was as high as $309.44 \mu\text{mol L}^{-1} \text{h}^{-1} \text{mg}^{-1}$, surpassing the other C_3N_4 -based photocatalysts. Che *et al.* prepared K and I co-doped polymer $g\text{-}C_3N_5$ ($g\text{-}C_3N_5\text{-K, I}$) by calcining a mixture of 3-amino-1,2,4-triazole and KI [80]. The photocatalytic H_2O_2 production rate over $g\text{-}C_3N_5\text{-K, I}$ ($2933.4 \mu\text{mol L}^{-1} \text{h}^{-1}$) exceeded that of $g\text{-}C_3N_5$. This high activity was attributed to the introduction of co-doping of K and I, which notably enhanced O_2 adsorption capacity and $2e^-$ ORR selectivity. Subhajyoti *et al.* synthesized O and C co-doped $g\text{-}C_3N_4$ by thermal polymerization [81]. The obtained photocatalyst could be used for both CO_2 photoreduction and production of H_2O_2 from dissolved O_2 in H_2O (Fig. 10g). As shown in Fig. 10h, all samples could produce H_2O_2 for a long time, and the performance of $g\text{-}CN\text{-MI-40}$ was much higher than that of pure C_3N_4 . After 30 h of reaction, the yield of $CN\text{-MI-40}$ was approximately 6 times that of pure C_3N_4 [81].

4.7. Others

The surface characteristics and energy band structure of $g\text{-}C_3N_4$ can be regulated by integrating natural dead leaves into urea polymerization process. For example, Li *et al.* utilized natural dead leaves as both a morphology regulator and a carbon source to prepare porous carbon-doped $g\text{-}C_3N_4$ (PCCN) to obtain C_3N_4 nanopores (Fig. 11a) [82]. The yield of H_2O_2 was $37.44 \mu\text{mol/L}$ under visible light, which was higher than pure C_3N_4 (Fig. 11b).

Besides, single-atom doping is also an effective mean to regulate the properties of materials [30,83]. It is mainly realized through the combination of selected atoms and other functional groups. These can influence the electronic structure, chemical activity and optical properties of catalyst. For example, Ren *et al.* prepared a Mn single atom anchored graphitic carbon nitride ($g\text{-}C_3N_4$) with arylamino group (Fig. 11c). It could directly produce H_2O_2 from seawater (Fig. 11d), and the yield of H_2O_2 was $2230 \mu\text{mol/L}$ (Fig. 11e) [30].

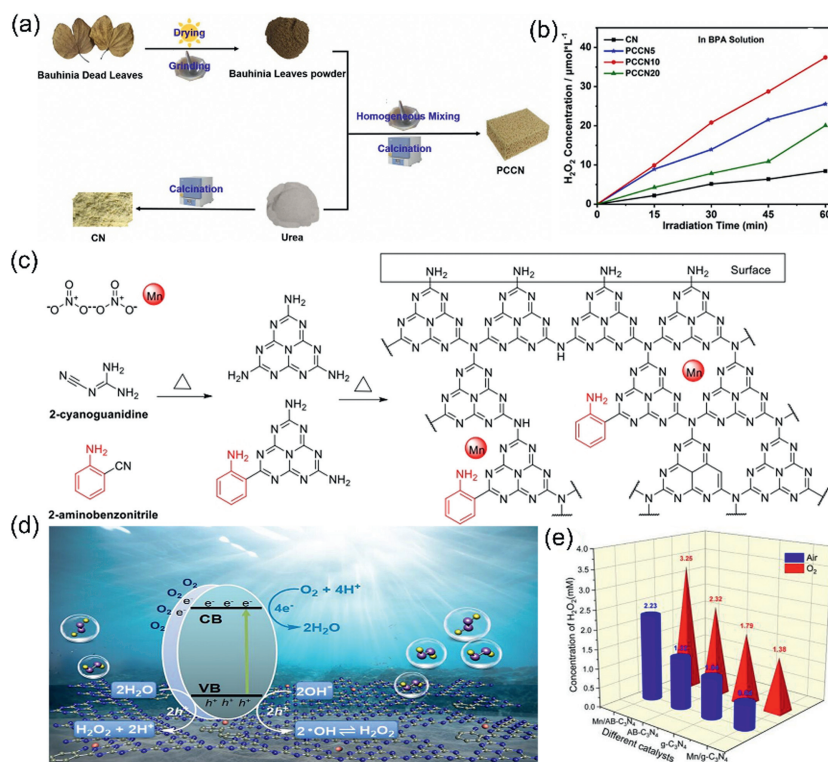


Fig. 11. (a) The synthesis process of PCCN sample. (b) Time profiles of photocatalytic H₂O₂ production with different catalysts. Copied with permission [82]. Copyright 2022, Elsevier. (c) The synthesis process of Mn/AB-C₃N₄. (d) Possible reaction mechanisms of H₂O₂ production. (e) Photocatalytic H₂O₂ production using different modified g-C₃N₄ catalysts. Copied with permission [30]. Copyright 2023, American Chemical Society.

5. In-situ applications

H₂O₂ is a green and clean resource, which displays wide application in energy and environmental processes, such as disinfection and antibiotic resistant genes degradation, organic pollutants degradation, medical applications, and fine chemical synthesis.

5.1. Disinfection and antibiotic resistant genes degradation

Compared to traditional disinfection methods, H₂O₂ presents excellent environmental friendliness. In a photocatalytic H₂O₂ system, H₂O₂ can be produced on-site and on-demand, presenting a more convenient and safer alternative to traditional disinfection methods. Consequently, the *in-situ* utilization of H₂O₂ generated through carbon nitride photocatalysis for disinfection and sterilization has garnered significant attention for researchers [34,66,71].

It is commonly believed that photocatalytic disinfection is carried out by reactive oxygen species (ROS), which can attack bacteria, thereby disrupting their physiological structure, antioxidant enzyme system, and respiratory system [84]. Geng *et al.* prepared ZnO/g-C₃N₄ catalyst for H₂O₂ generation and *in-situ* sterilization of natural water under sunlight irradiation [66]. The sterilization rate reached 97.4% after 60 min with 10% ZnO/g-C₃N₄. Similarly, Li *et al.* used C₃N₅ to produce H₂O₂ to disinfect *E. coli* at different initial concentrations and achieved 100% inhibition rate at 60 min (Fig. 12a) [71]. In another work, they also constructed an *in-situ* self-photo-Fenton reaction system for the removal of oxytetracycline (OTC) contaminants and antibiotic resistance genes (ARGs) using C₃N₄. The results showed good antimicrobial properties (Figs. 12b and c) [34].

5.2. Organic pollutants degradation

The degradation of various organic pollutants by H₂O₂ has been recognized as an effective strategy, and many related works have

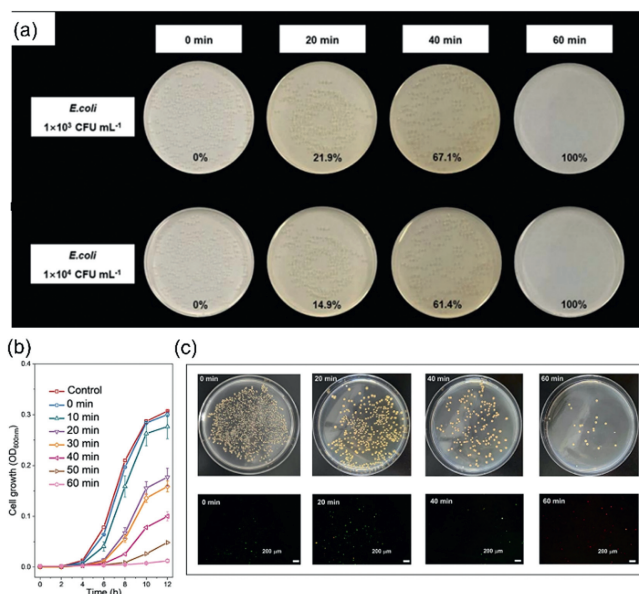


Fig. 12. (a) The disinfection rate of *E. coli*. Copied with permission [71]. Copyright 2023, Springer Nature. (b) MRSA growth curves. (c) Plate incubated with LB medium and live/dead staining (merge). Copied with permission [34]. Copyright 2024, Elsevier.

been carried out [29,82,85–89]. However, the degradation, particularly mineralization, of organic contaminants requires a higher oxidative capacity. The *in-situ* Fenton reaction can address the above challenges. Consequently, many researchers have explored *in-situ* Fenton or *in-situ* Fenton-like reactions to enhance degradation efficiency.

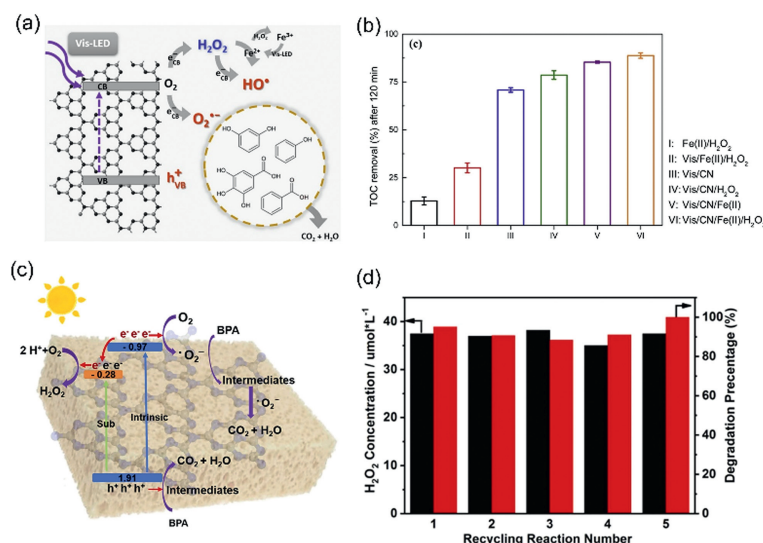


Fig. 13. (a) *In-situ* self-Fenton mechanism diagram. (b) Removal of TOC in different reaction environments. Copied with permission [89]. Copyright 2020, Elsevier. (c) Photogenerated electron-hole pair transfer pathway and the mechanism of simultaneous degradation of BPA by H₂O₂ generation. (d) Photocatalytic generation of H₂O₂ and simultaneous degradation of BPA in a cyclic experiment. Copied with permission [82]. Copyright 2022, Elsevier.

For example, Torres-Pinto *et al.* constructed an *in-situ* self-Fenton system using prepared *g*-C₃N₄ (Fig. 13a). This system showed the high mineralization rate with pure photocatalytic system, which was five times higher than that of conventional Fenton system. After the addition of Fe²⁺, the mineralization rate of *in-situ* self-Fenton system increased by 20% (Fig. 13b) [89]. This result demonstrated the potential of carbon nitride used for *in-situ* self-Fenton systems for the degradation of organic pollutants. Li *et al.* also attempted to degrade BPA via an *in-situ* self-Fenton system (Fig. 13c). These results showed a good degradation efficiency for five cycles, demonstrating the excellent stability of prepared *g*-C₃N₄ (Fig. 13d) [82].

5.3. Medical applications

Due to H₂O₂ is a natural metabolite of many organisms, it has a wide range of applications in biomedicine [90]. H₂O₂ decomposes directly into H₂O and O₂, leaving no toxic residues or harmful by-products, posing harmlessness to human health and the environment. Thus, researchers begin to extend photocatalytic H₂O₂ production with carbon nitride to medicine field including sensing and cancer therapy [45,91–94].

For example, the degradation of H₂O₂ can alleviate O₂ deprivation in the hypoxic tumor microenvironment, thus improving the effectiveness of photodynamic therapy (PDT) [94]. Based on this, Ma *et al.* reported C₅N₂ photocatalyst for the selective and non-sacrificial generation of H₂O₂ in both normoxic and anoxic systems [45]. Due to the reinforcement of delocalization of π -electrons by linkers in C₅N₂, the side H₂ evolution was thermodynamically eliminated, while WOR was kinetically promoted (Fig. 14a). The results showed that C₅N₂ had excellent performance of photocatalytic H₂O₂ production (Figs. 14b and c), and also led to fairly significant cell death in both normoxic and hypoxic cellular environments (Fig. 14d). Based on this, they then developed a C₅N₂-based •OH generator and fluorescent agent for photodynamic hypoxic tumor therapy (Fig. 14e).

5.4. Fine chemical synthesis

Photocatalytic H₂O₂ production can also be used for organic chemical synthesis. Moreover, this approach is of great significance to the current energy and environmental challenges, as it allows

the oxidation reaction to be carried out under mild conditions, avoiding the environmental pollution and waste treatment problems associated with strong oxidizing agents.

Up to now, researchers are directing their attention towards *in-situ* photocatalytic H₂O₂ production for fine chemical synthesis [3,95–98]. For example, a tandem system for *in-situ* photocatalytic H₂O₂ generation and propylene epoxidation was constructed by Zhang *et al.* (Fig. 15a). Carbon nitride with N₃C vacancies was first synthesized with the synergistic effect of Ar pyrolysis and precursors self-assembled at the supramolecular level. The results showed that prepared catalyst had excellent photocatalytic H₂O₂ performance (5775 $\mu\text{mol g}^{-1} \text{h}^{-1}$) (Fig. 15b), and the efficiency of synthesized propylene oxide (PO) was 5515 $\mu\text{mol g}^{-1} \text{h}^{-1}$ with 99.1% selectivity (Fig. 15c) [98].

6. Summary and outlook

Photocatalytic H₂O₂ production has attracted widespread attention and has become a hot research topic. In this review, we firstly discuss the basic principles of photocatalysis and photocatalytic H₂O₂ production. Second, we briefly introduce the main current types of carbon nitrides and highlight different modification methods of carbon nitride, including morphology tuning, noble metal loading, defect control, heterojunction regulation, molecular structure engineering, and elements doping, *etc.* Finally, a wide range of *in-situ* applications for photocatalytic H₂O₂ production are displayed, including disinfection and antibiotic resistant genes degradation, organic pollutants degradation, medical applications, and fine chemical synthesis. It is important to emphasize here that the concentration of H₂O₂ by photocatalytic production is hard to meet industrial need. However, the miniaturization and rapidity of photocatalytic system easily make it a key component *in-situ* practical applications with small-dose.

Although some promising results have been achieved, the photocatalytic production of H₂O₂ from carbon nitride is still in its infancy. At present, the H₂O₂ yield of carbon nitride with photocatalysis is still relatively low, and there are still a lot of gaps to meet the requirements of industrial applications. Current main difficulties mainly include narrow light absorption range, low photoelectric conversion efficiency and design of new photocatalytic systems. Therefore, future effort should focus on the following aspects:

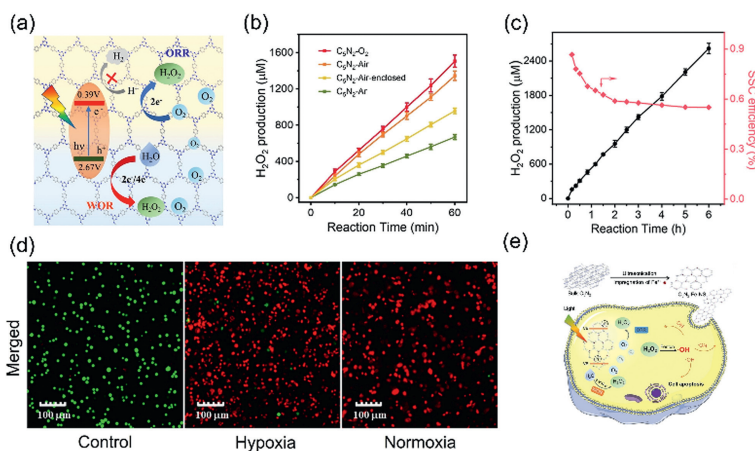


Fig. 14. (a) Mechanism of photocatalytic production of H₂O₂ by C₅N₂. (b) Time profiles of photocatalytic H₂O₂ production with different catalysts. (c) H₂O₂ production for long time and solar-to-chemical conversion (SCC) efficiency. (d) Live/dead double staining of C₅N₂-Fe-NS-treated 4T1 cells under light with FDA (green, live cells) and PI (red, dead cells). (e) Tumor therapy processes. Copied with permission [45]. Copyright 2022, Wiley-VCH.

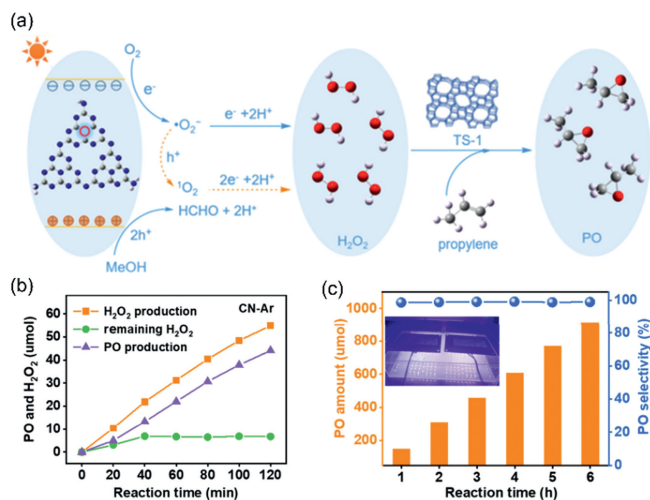


Fig. 15. (a) Mechanism diagram of *in-situ* photocatalytic H₂O₂ production and propylene epoxidation. (b) H₂O₂ production, PO production, and remaining H₂O₂ during the reactions. (c) Productivity and selectivity of PO. Copied with permission [98]. Copyright 2023, American Chemical Society.

- (i) To improve the response range of carbon nitride to visible light. We should continue to broaden the absorption of materials into the visible and near-infrared regions through the development of new materials and material modification methods.
- (ii) To design new photocatalyst forms. Currently, the mainstream photocatalyst form is powders. We should optimize these with immobilization methods by preparing membranes, sponges, aerogels and photoelectrodes, etc.
- (iii) To design new photocatalytic systems. The current laboratory catalytic reaction system mainly depends on simple glass instruments. Therefore, we should design suitable *in-situ* photocatalytic reaction system in series and parallel connections under different catalytic scenarios and practical application scenarios.

Declaration of competing interest

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

CRediT authorship contribution statement

Hao Lv: Writing – original draft, Data curation. **Zhi Li:** Writing – review & editing. **Peng Yin:** Writing – review & editing, Conceptualization. **Ping Wan:** Writing – review & editing, Conceptualization. **Mingshan Zhu:** Writing – review & editing, Resources, Project administration, Funding acquisition, Conceptualization.

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