

# Synergistic Photocatalysis-Self-Fenton Systems for Environmental Remediation: A Mini-Review

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**Abstract:** Environmental contaminants contained in various wastewater can pose a severe danger to ecological environment and public safety if not properly treated. Photocatalysis-self-Fenton (PSF) technology, integrating the advantages of photocatalysis and Fenton methods, can achieve highly efficient removal performance towards multiple pollutants due to “self-production and self-utilization” of H<sub>2</sub>O<sub>2</sub> and increased generation of oxidizing species. Organic photocatalysts (g-C<sub>3</sub>N<sub>4</sub>, porphyrin, etc.) can be appropriate candidates for constructing PSF system by virtue of their characteristic surface structures and adequate band positions for *in-situ* H<sub>2</sub>O<sub>2</sub> production. Meanwhile, several inorganic photocatalysts (Bi<sub>2</sub>WO<sub>6</sub>, Zn<sub>2</sub>In<sub>2</sub>S<sub>5</sub>, etc.) after modification have also been proven to have the potential for H<sub>2</sub>O<sub>2</sub> generation and constructing PSF system. Therefore, in this mini-review, the superiority, synergistic reaction mechanism, and essential requirement for the catalyst of PSF system were precisely described. Moreover, various photocatalysts with excellent H<sub>2</sub>O<sub>2</sub> production activities enhanced by varieties of modification strategies were concretely discussed, while the applications of related PSF systems with high oxidation and mineralization abilities in contaminants elimination were also emphasized. Finally, the challenges and outlooks for the development of PSF technology in future studies were put forward.

**Keywords:** photocatalysis; self-Fenton; H<sub>2</sub>O<sub>2</sub> production; pollutants removal; materials modification

## 1. Introduction

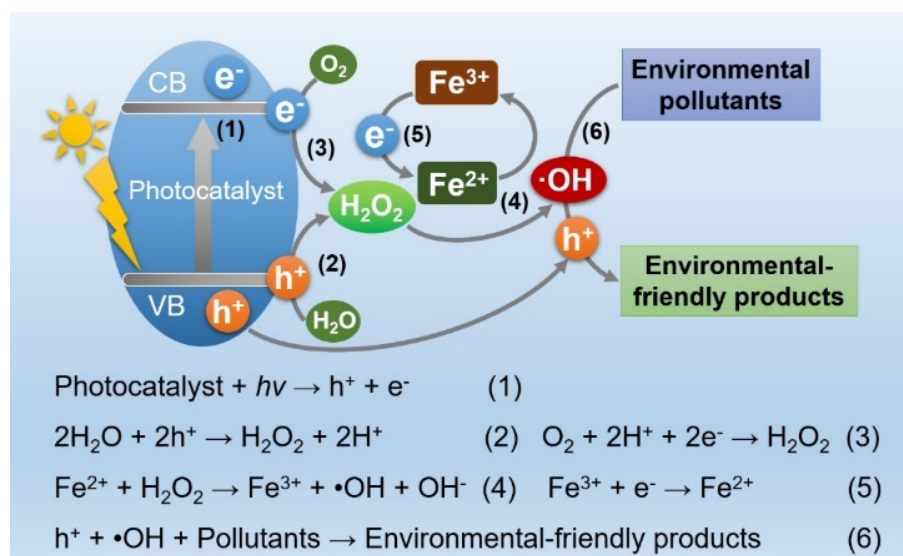
Industrial, medical and aquaculture wastewater usually contains high concentrations of organic pollutants and pathogenic microorganisms, which could pose a severe danger to ecological environment and public safety if not properly treated [1]. Phenolic organics have been included in the precedence-controlled contaminant list because of their genotoxicity, acute toxicity, and endocrine-disruption. Antibiotics can cause potential/immediate harm to health as well as induce drug-resistance problem after long-period accumulation in water. Pathogenic bacteria can lead to aquatic system disturbance and cause the epidemics. Besides, antibiotic resistant bacteria (ARB) and resistance genes (ARGs) can promote the



spread of drug-resistance and decrease the cure rate of bacterial disease. Advanced oxidation processes (AOPs), such as photocatalysis and Fenton methods, are considered as one of the most promising technologies for water treatment to control the above contaminants. As AOPs can produce a variety of oxidizing species including hydroxyl radicals ( $\bullet\text{OH}$ ), holes ( $\text{h}^+$ ), singlet oxygen ( $^1\text{O}_2$ ), and superoxide radicals ( $\bullet\text{O}_2^-$ ), they have attracted the attention of researchers in various countries and gradually replaced the traditional chemical oxidation methods for environmental pollutant removal [2].

Photocatalysis oxidation method has the superiorities of sunlight driven available, mild reaction condition, and low operating cost [3]. However, there are still some problems, such as low utilization of solar energy, high charge combination rate, and unsatisfactory oxidation capacity, which restrict its application prospect in effluent treatment [4]. Therefore, how to increase solar utilization, expedite charge migration, and boost the quantity of oxidizing species are critical to enhancing the application value of photocatalysis technology. Fenton oxidation method has the advantages of simple operation and rapid response [5]. However, there are still some drawbacks, such as increased processing cost of  $\text{H}_2\text{O}_2$ , slow regeneration of  $\text{Fe}^{3+}$ , low aromatic ring opening ability, and narrow pH range ( $\text{pH} \leq 3$ ), which make it difficult to meet the technical and economic requirements of wastewater treatment [6]. Therefore, how to reduce the dosage of  $\text{H}_2\text{O}_2$ , accelerate the cyclic conversion of  $\text{Fe}^{3+}/\text{Fe}^{2+}$ , increase mineralization ability, and expand pH range are crucial to improve the feasibility of Fenton technology for deep treatment of pollutants. Photocatalysis-Fenton method can use the reductiveness of photogenerated electrons ( $\text{e}^-$ ) to improve the regeneration rate of  $\text{Fe}^{3+}$  and the conversion efficiency of  $\text{H}_2\text{O}_2$ , as well as promote the mineralization efficiency via generating more active species ( $\text{h}^+$ ,  $\bullet\text{O}_2^-$ , etc.) via photocatalytic reaction [7]. However, this collaborative system still needs the extra additional of  $\text{H}_2\text{O}_2$ , which increases processing cost and reduces application value.

Photocatalysis-self-Fenton (PSF) oxidation method is a kind of new emerging coupling AOPs, which can take the advantages of Fenton method to solve the problems of photocatalysis reaction, as well as use the superiorities of photocatalysis method to remedy the drawbacks of Fenton reaction, thus achieving more prominent deep treatment effect and higher application potential than single AOPs [8]. As shown in Figure 1, PSF technology primarily uses the photocatalysts to *in-situ* generate  $\text{H}_2\text{O}_2$  effectively, and then promotes the occurrence of self-Fenton reaction via the mediation of metal species ( $\text{Fe}$ ,  $\text{Co}$ , etc.) or  $\text{e}^-$  ( $\text{H}_2\text{O}_2 + \text{e}^- \rightarrow \bullet\text{OH} + \text{OH}^-$ ) [9]. “Self-production and self-utilization” of  $\text{H}_2\text{O}_2$  can realize the generation of abundant  $\bullet\text{OH}$ , thus greatly improved the oxidation capacity of coupling system [10]. Moreover,  $\text{e}^-$  can accelerate the cyclic transformation of metal ions ( $\text{Fe}^{3+}/\text{Fe}^{2+}$ ,  $\text{Co}^{3+}/\text{Co}^{2+}$ , etc.) and facilitate the conversion from  $\text{H}_2\text{O}_2$  to  $\bullet\text{OH}$  [11]. Furthermore, the timely consumption of  $\text{e}^-$  can restrain the combination of  $\text{e}^- \cdot \text{h}^+$  to release a larger number of free  $\text{h}^+$ , thus further boosting the mineralization efficiency of coupling system [12]. Therefore, the synergy between photocatalysis and Fenton can endow PSF system with high oxidation and mineralization abilities without extra addition of  $\text{H}_2\text{O}_2$ , finally achieving high-efficiency removal performance towards multiple environmental pollutants. Besides, in the PSF system, the main Fenton reaction ( $\text{Fe}^{2+} + \text{H}_2\text{O}_2 \rightarrow \text{Fe}^{3+} + \bullet\text{OH} + \text{OH}^-$ ) can smoothly proceed via continuously reducing  $\text{Fe}^{3+}$  concentration by  $\text{e}^-$  ( $\text{Fe}^{3+} + \text{e}^- \rightarrow \text{Fe}^{2+}$ ). Thus, the PSF reaction can be effectively conducted over a relatively wider pH range, which overcomes the limitation of acid pH requirement for Fenton method.



**Figure 1.** The schematic diagram of a representative PSF system for the removal of environmental pollutants.



In this mini-review, the superiority, synergistic reaction mechanism, and essential requirement for the catalyst of PSF system were precisely described. Moreover, the progress of developing various photocatalysts with high H<sub>2</sub>O<sub>2</sub> yield by varieties of modification strategies to construct PSF synergistic system were concretely discussed. Furthermore, the applications of PSF system for the high-efficiency removal of multiple environmental contaminants were also emphasized. Finally, the challenges and outlooks for the development of PSF technology in future studies were put forward.

## 2. Developing Photocatalysts with High H<sub>2</sub>O<sub>2</sub> Yield to Construct Photocatalysis-Self-Fenton Synergistic System

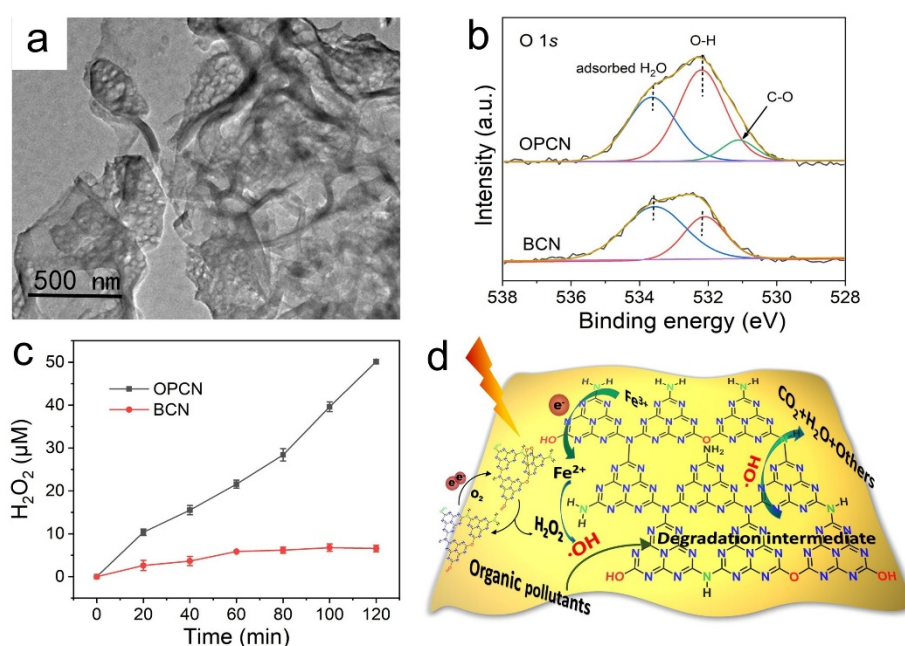
Since the reaction efficiency of PSF oxidation method mainly depends on the amount of *in-situ* generated H<sub>2</sub>O<sub>2</sub>, choosing an appropriate photocatalyst with excellent H<sub>2</sub>O<sub>2</sub> production activity is very crucial to the construction of PSF synergistic system. The photocatalytic synthesis of H<sub>2</sub>O<sub>2</sub> can mainly be achieved through water oxidation reaction (WOR) and oxygen reduction reaction (ORR) [13,14]. In contrast, the latter is more prone to happen via one-step two-electron (2e<sup>-</sup>) ORR (O<sub>2</sub> + 2H<sup>+</sup> + 2e<sup>-</sup> → H<sub>2</sub>O<sub>2</sub>) or two-step 2e<sup>-</sup> ORR (O<sub>2</sub> + e<sup>-</sup> → •O<sub>2</sub><sup>-</sup>, •O<sub>2</sub><sup>-</sup> + e<sup>-</sup> + 2H<sup>+</sup> → H<sub>2</sub>O<sub>2</sub>). Although the direct 2e<sup>-</sup> ORR is regarded as a more selective pathway for H<sub>2</sub>O<sub>2</sub> production, indirect 2e<sup>-</sup> ORR is dominant in major photocatalytic systems [15]. Protons in the above process are generally came from two pathways: (1) h<sup>+</sup> oxidize sacrificial agent to provide protons; (2) h<sup>+</sup> directly oxidize H<sub>2</sub>O to provide protons (H<sub>2</sub>O + 2h<sup>+</sup> → 2H<sup>+</sup> + 1/2O<sub>2</sub>) [16]. The second route has higher requirement on the photocatalyst, which needs to have a suitable surface structure and sufficient valence band (VB) potential for oxidizing H<sub>2</sub>O [17,18]. Conventional inorganic photocatalysts (TiO<sub>2</sub>, ZnO, etc.) show limited H<sub>2</sub>O<sub>2</sub> yields, which might be owing to weak capability to oxidize H<sub>2</sub>O, low selectivity to perform 2e<sup>-</sup> ORR, and high self-decomposition of H<sub>2</sub>O<sub>2</sub> via self-disproportionation/photoreaction on the surface [19]. Thus, it is of great significance to develop suitable photocatalyst to synthesize H<sub>2</sub>O<sub>2</sub> efficiently in PSF reaction process without the addition of sacrificial agent (isopropanol (IPA), ethanol (EtOH), etc.). Besides, various modification methods have been applied to improve photocatalytic H<sub>2</sub>O<sub>2</sub> production activity in recent years. Morphology optimizing can shorten charge transmission distance and facilitate surface activity. Heteroelement doping can improve light response range, redox capacity, and O<sub>2</sub> adsorption ability. Defect engineering can accelerate charge separation as well as promote O<sub>2</sub> activation. Heterogeneous structure construction can induce an internal electric field (IEF) to drive interfacial charge transfer [20]. After modification, the photocatalyst with increased H<sub>2</sub>O<sub>2</sub> yield can be qualified to construct PSF system (Table 1).

**Table 1.** H<sub>2</sub>O<sub>2</sub> generation activities of various photocatalysts for constructing PSF systems.

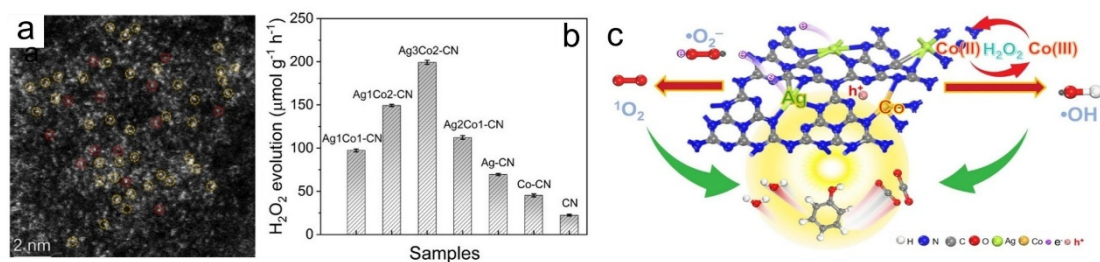
| Photocatalyst                                                                        | Dosage (mg·mL <sup>-1</sup> ) | Light Source                             | Sacrificial Agent      | H <sub>2</sub> O <sub>2</sub> Yield (mM·h <sup>-1</sup> ·g <sup>-1</sup> ) | Ref. |
|--------------------------------------------------------------------------------------|-------------------------------|------------------------------------------|------------------------|----------------------------------------------------------------------------|------|
| P-g-C <sub>3</sub> N <sub>4</sub>                                                    | 1.0                           | λ > 420 nm                               | 2,4-DCP                | 0.78                                                                       | [21] |
| GCN                                                                                  | 0.5                           | λ > 420 nm<br>(700 mW·cm <sup>-2</sup> ) | -                      | 0.51                                                                       | [22] |
| Resin                                                                                | 0.6                           | λ > 420 nm<br>(70 mW·cm <sup>-2</sup> )  | -                      | 3.23                                                                       | [23] |
| OPCN                                                                                 | 0.5                           | λ > 400 nm<br>(100 mW·cm <sup>-2</sup> ) | 2,4-DCP                | 1.00                                                                       | [24] |
| P-C <sub>3</sub> N <sub>4</sub> /O-C <sub>3</sub> N <sub>4</sub>                     | 1.0                           | λ > 420 nm                               | 10 vol% EtOH           | 3.58                                                                       | [25] |
| Defective twin CN                                                                    | 0.5                           | λ = 420 nm                               | -                      | 8.56                                                                       | [26] |
| Cv-PCNNS                                                                             | 1.0                           | λ > 420 nm<br>(140 mW·cm <sup>-2</sup> ) | 10 vol% IPA            | 19.70                                                                      | [27] |
| Cu <sup>0</sup> @CuO <sub>x</sub> -NC/Zn <sub>2</sub> In <sub>2</sub> S <sub>5</sub> | 0.5                           | Visible light                            | -                      | 5.20                                                                       | [28] |
| CPBA-CN                                                                              | 1.0                           | Simulated sunlight                       | 4-CP                   | 7.82                                                                       | [10] |
| CoFeO/PDIsm                                                                          | 1.0                           | λ > 420 nm                               | CIP                    | 5.63                                                                       | [29] |
| Coral-B-CN                                                                           | 0.4                           | Simulated sunlight                       | 4-CP                   | 0.31                                                                       | [30] |
| Cyano-g-C <sub>3</sub> N <sub>4</sub> /In <sub>2</sub> S <sub>3</sub> /Ppy           | 1.0                           | λ > 420 nm<br>(140 mW·cm <sup>-2</sup> ) | 10 vol% IPA            | 17.90                                                                      | [31] |
| C <sub>3</sub> N <sub>4</sub> -Urea                                                  | 0.5                           | λ > 400 nm                               | 10 vol% EtOH           | 79.00                                                                      | [32] |
| rGO/SA-TCPP                                                                          | 0.3                           | λ > 400 nm<br>(100 mW·cm <sup>-2</sup> ) | Sulfadimethoxine (SDM) | 0.92                                                                       | [33] |
| SKCN                                                                                 | 1.0                           | λ > 400 nm<br>(100 mW·cm <sup>-2</sup> ) | 10 vol% IPA            | 0.63                                                                       | [34] |

## 2.1. g-C<sub>3</sub>N<sub>4</sub>-Based Photocatalysis-Self-Fenton System

Over the past decade, graphitic carbon nitride (g-C<sub>3</sub>N<sub>4</sub>) has developed rapidly and been extensively utilized in pollutants removal due to visible-light response, abundant element source, adjustable structure, and high biocompatibility [35,36]. Moreover, the characteristic surface structure and adequate band position of g-C<sub>3</sub>N<sub>4</sub> are beneficial to promote H<sub>2</sub>O<sub>2</sub> production [37]. It has been reported 1,4-endoperoxide species can be formed on the surface of photoactivated g-C<sub>3</sub>N<sub>4</sub> to enhance the selectivity for 2e<sup>-</sup> ORR [17]. Thus, g-C<sub>3</sub>N<sub>4</sub> can be an appropriate photocatalyst for constructing PSF collaborative system. Ao's group developed a PSF system based on garland g-C<sub>3</sub>N<sub>4</sub> with carbon defects (GCN) [22]. Garland morphology modulation and carbon defect regulation accelerated charge separation, increased O<sub>2</sub> adsorption, and improved 2e<sup>-</sup> ORR for H<sub>2</sub>O<sub>2</sub> production of GCN. The H<sub>2</sub>O<sub>2</sub> yield (0.51 mM·h<sup>-1</sup>·g<sup>-1</sup>) of GCN in water was about 5.0 folds as that of bulk g-C<sub>3</sub>N<sub>4</sub> (BCN). This work combines morphology optimizing and defect engineering to synthesize g-C<sub>3</sub>N<sub>4</sub> with high H<sub>2</sub>O<sub>2</sub> yield to realize PSF coupling effect. Xu's group designed a PSF system based on O-doped g-C<sub>3</sub>N<sub>4</sub> porous nanosheets (OPCN) (Figure 2) [24]. The unique morphology and oxygen-containing groups introduced in the structure of OPCN accelerated charge migration, created more reactive sites, strengthened the oxidation ability for H<sub>2</sub>O, and reduced H<sub>2</sub>O<sub>2</sub> self-decomposition rate, thus promoting the selectivity of 2e<sup>-</sup> ORR and greatly increasing H<sub>2</sub>O<sub>2</sub> generation. The H<sub>2</sub>O<sub>2</sub> yield (1.00 mM·h<sup>-1</sup>·g<sup>-1</sup>) of OPCN in 2,4-dichlorophenol (2,4-DCP) solution was 7.6 folds as that of BCN. This study combines morphology modulation and heteroelement doping to develop g-C<sub>3</sub>N<sub>4</sub> catalyst with high H<sub>2</sub>O<sub>2</sub> production activity. Wu's group developed a P-doped carbon nitride (P-C<sub>3</sub>N<sub>4</sub>)/O-doped carbon nitride (O-C<sub>3</sub>N<sub>4</sub>) photocatalyst for PSF reaction [25]. The interfacial IEF in the heterojunction effectively increased the e<sup>-</sup>-h<sup>+</sup> separation efficiency and preserved the reducibility of e<sup>-</sup> for 2e<sup>-</sup> ORR, resulting in the excellent catalytic performance of P-C<sub>3</sub>N<sub>4</sub>/O-C<sub>3</sub>N<sub>4</sub>. The H<sub>2</sub>O<sub>2</sub> production rate in an air-saturated 10 vol% EtOH solution (3.58 mM·h<sup>-1</sup>·g<sup>-1</sup>) was about 7.2, 2.1, 2.5 and 2.5 folds as that of BCN, P,O-co-doped C<sub>3</sub>N<sub>4</sub>, P-C<sub>3</sub>N<sub>4</sub> and O-C<sub>3</sub>N<sub>4</sub>, respectively. This study verifies heterojunction construction is a feasible way for designing g-C<sub>3</sub>N<sub>4</sub>-based PSF system via enhancing the H<sub>2</sub>O<sub>2</sub> production activity. Lian et al. proposed an atomically dispersed Ag,Co-embedded g-C<sub>3</sub>N<sub>4</sub> (AgCo-CN) (Figure 3) [38]. The Ag,Co dual sites in AgCo-CN could both be regarded as electron transfer sites and self-Fenton redox centers. The H<sub>2</sub>O<sub>2</sub> yield of optimum AgCo-CN catalyst in water under visible light reached 0.20 mmol·h<sup>-1</sup>·g<sup>-1</sup>, which was about 9.3, 2.9 and 4.8 folds as that of BCN, Ag-CN and Co-CN, respectively. As the molar ratio of H<sub>2</sub>O<sub>2</sub>/H<sub>2</sub> production was close to 1, the *in-situ* generated H<sub>2</sub>O<sub>2</sub> should be mainly originated from WOR rather than ORR process. This work offers a route of designing high-efficiency single-atom catalysts for achieving PSF reaction.



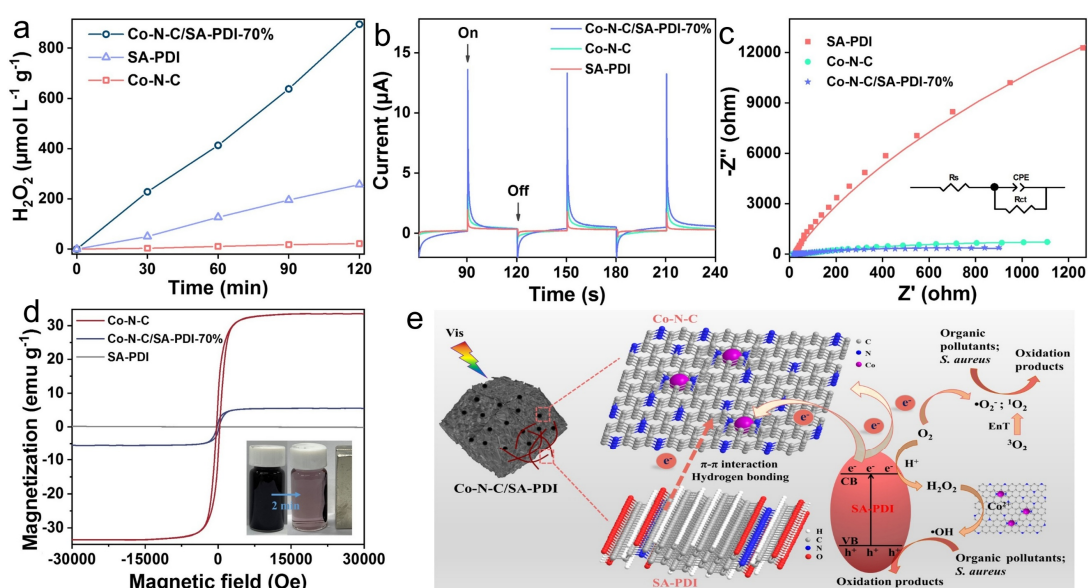
**Figure 2.** (a) TEM image of OPCN. (b) O 1s XPS spectra of BCN and OPCN. (c) Photocatalytic H<sub>2</sub>O<sub>2</sub> evolution of BCN and OPCN without adding specific sacrificial agent under visible light. (d) The reaction mechanism diagram of OPCN-PSF system [24]. Copyright © 2022 Elsevier.



**Figure 3.** (a) The HAADF-STEM image of AgCo-CN. (b) The *in-situ*  $\text{H}_2\text{O}_2$  generation rate of different samples. (c) The reaction mechanism diagram of AgCo-CN under visible light [38]. Copyright © 2024 Wiley-VCH GmbH.

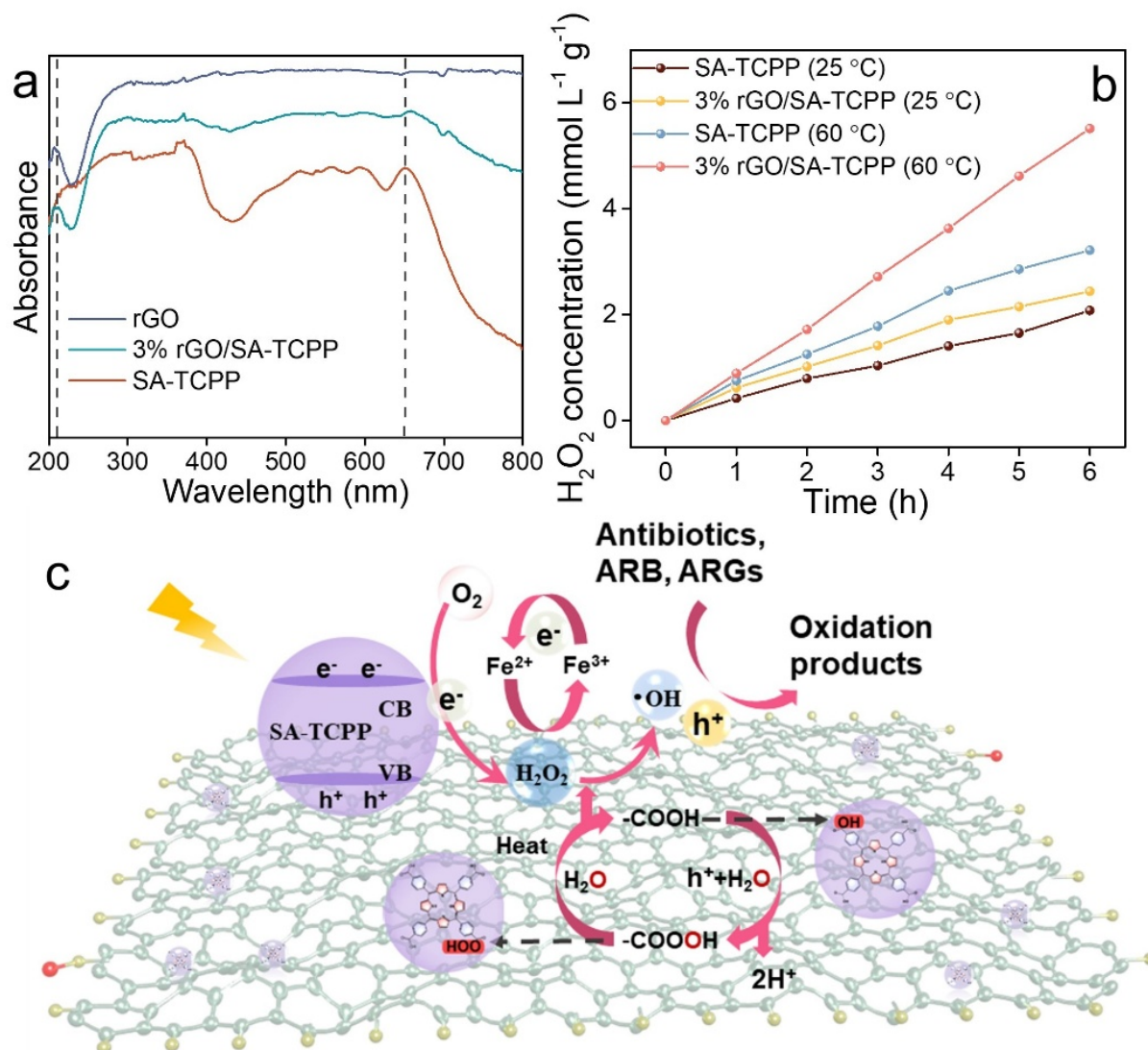
## 2.2. Other Organic Photocatalyst-Based Photocatalysis-Self-Fenton System

Except for g- $\text{C}_3\text{N}_4$ , other organic photocatalysts (perylene imide (PDI), porphyrin (TCPP), metal organic frameworks (MOFs), resin, etc.) have also become a hotspot in recent years because of their high redox capacity and excellent photocatalytic activity, which show an extraordinary potential in catalytically producing  $\text{H}_2\text{O}_2$  and constructing PSF system. PDI and TCPP aggregates self-assembled by single molecules via  $\pi$ - $\pi$  stacking and hydrogen-bonding have been extensively investigated as supramolecular photocatalysts with adequate band structures [39,40]. Xu et al. designed a Co-imbedded N-doped carbonaceous framework (Co-N-C)/supramolecular PDI (SA-PDI) photocatalyst for high-efficiency pollutant degradation and disinfection via PSF reaction (Figure 4) [4]. The large surface area, 3D porous framework, high conductivity and strong magnetism of Co-N-C endowed SA-PDI with increased adsorption capacity, extended spectral-response range, accelerated charge separation rate, and improved recycling ability. Thus, Co-N-C/SA-PDI photocatalyst showed greatly improved  $\text{H}_2\text{O}_2$  production rate ( $0.45 \text{ mM}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$ ) in phenol solution under visible light, which was about 3.5 folds as that of SA-PDI. The study offers a new route to construct SA-PDI-based PSF system for environmental remediation. Supramolecular TCPP (SA-TCPP) photocatalyst has been reported to possess high quantum efficiency of  $\text{H}_2\text{O}_2$  synthesis from  $\text{H}_2\text{O}$  and  $\text{O}_2$  under visible light (420 nm, 14.9%) and near-infrared light (940 nm, 1.1%) [41]. Besides the ORR pathway, SA-TCPP showed another pathway for  $\text{H}_2\text{O}_2$  generation via oxidizing its -COOH groups by  $h^+$  into -COOOH groups with thermal instability and then hydrolyzing into  $\text{H}_2\text{O}_2$  by heating. Xu et al. constructed a PSF system based on reduced graphene oxide/SA-TCPP (rGO/SA-TCPP) (Figure 5) [33]. As the modification of rGO greatly broadened the light absorption and improved the charge transfer of SA-TCPP, the  $\text{H}_2\text{O}_2$  yield of rGO/SA-TCPP catalyst ( $60^\circ\text{C}$ ) increased to  $0.92 \text{ mM}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$ , which was 1.7 and 2.3 folds as that of SA-TCPP ( $60^\circ\text{C}$ ) ( $0.53 \text{ mM}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$ ) and rGO/SA-TCPP ( $25^\circ\text{C}$ ) ( $0.40 \text{ mM}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$ ), respectively. This work provides a novel strategy to design a PSF system based on SA-TCPP catalyst.



**Figure 4.** (a) Photocatalytic  $\text{H}_2\text{O}_2$  generation curves, (b) photocurrent response curves and (c) EIS Nyquist plots under visible light of Co-N-C, SA-PDI and Co-N-C/SA-PDI. (d) The magnetic hysteresis loops of Co-N-C, SA-PDI and Co-N-C/SA-PDI. (e) The reaction mechanism diagram of Co-N-C/SA-PDI [4]. Copyright © 2023 Elsevier.





**Figure 5.** (a) DRS analysis of different samples. (b) H<sub>2</sub>O<sub>2</sub> generation curves of SA-TCPP and rGO/SA-TCPP at different temperatures. (c) The schematic diagram of rGO/SA-TCPP-PSF system [33]. Copyright © 2024 Elsevier.

The tunable porous framework and highly-dispersed unsaturated metal sites of Fe-MOFs offer an adsorption-catalysis collaborative platform for PSF reaction. Zhao et al. proposed an integrated Ti<sub>3</sub>C<sub>2</sub> MXene/MIL-100(Fe) PSF catalyst via Fe-protoporphyrin bridging for PSF synergistic degradation [42]. The Fe-protoporphyrin structure imitated biomimetic heme behavior for O<sub>2</sub>-carrying, while the Schottky junction between MXene and MIL-100(Fe) facilitated the charge separation, thus greatly accelerating the conversion from dissolved O<sub>2</sub> into H<sub>2</sub>O<sub>2</sub>. MXene/MIL-100(Fe) showed 12.0 folds higher of H<sub>2</sub>O<sub>2</sub> yield (117.51 mM·g<sup>-1</sup>) under air-bubbles than MIL-100(Fe), which was beneficial to PSF degradation reaction. This work provides a promising MOFs-based contender for constructing PSF system via *in-situ* H<sub>2</sub>O<sub>2</sub> generation. Wang et al. discovered a PSF system based on resorcinol formaldehyde (RF) resin and Fe<sup>3+</sup> [23]. Due to its broad spectral response, RF resin efficiently generated 3.23 mM·h<sup>-1</sup>·g<sup>-1</sup> of H<sub>2</sub>O<sub>2</sub> in water under visible light, which was 34.5 and 3.4 folds as that of P-g-C<sub>3</sub>N<sub>4</sub> and CdS/rGO, respectively. The high H<sub>2</sub>O<sub>2</sub> evolution rate greatly facilitated the production of •OH and improved the removal of pollutants. Because of the approximately 100% reduction of Fe<sup>3+</sup> by e<sup>-</sup>, H<sub>2</sub>O<sub>2</sub> utilization efficiency in RF resin-PSF reaction was promoted to 82.1%. Meanwhile, the strong oxidizing h<sup>+</sup> (2.15 eV) of RF resin reduced the production of macromolecular polymer intermediates, thus resulting in the direct opening of aromatic rings. This study successfully develops resin as a novel catalyst of PSF system for eco-friendly and low-cost water treatment.

### 2.3. Inorganic Photocatalyst-based Photocatalysis-Self-Fenton System

Currently, several inorganic photocatalysts (Bi<sub>2</sub>WO<sub>6</sub>, CdS, Zn<sub>2</sub>In<sub>2</sub>S<sub>5</sub>, etc.) have been proven to have the potential for *in-situ* H<sub>2</sub>O<sub>2</sub> generation and constructing PSF systems via various modification strategies. Bi<sub>2</sub>WO<sub>6</sub>



is a famous photocatalyst with visible-light response ( $\lambda < 450$  nm) and high oxidation ability. Xie et al. fabricated an adsorption-enhanced PSF synergy system using  $\text{Bi}_2\text{WO}_6$  (BWO) as the photocatalytic  $\text{H}_2\text{O}_2$  generator, DMP5 polymer (DMPBP[5]) as the adsorption center and  $\text{Fe}_3\text{O}_4$  as the Fenton center [43]. In this DMPBP[5]-BWO- $\text{Fe}_3\text{O}_4$  system, the decoration of DMPBP[5] with rich cavities and  $\text{Fe}_3\text{O}_4$  ( $\text{Fe}^{2+}/\text{Fe}^{3+}$ ) on BWO promoted the charge separation, improved the selectivity of  $2\text{e}^-$  ORR, accelerated the anchoring of active species, and increased the adsorption effect towards  $\text{H}_2\text{O}_2$ , thus reducing the  $\text{H}_2\text{O}_2$  self-decomposition rate and achieving a high  $\text{H}_2\text{O}_2$  yield under visible light. The  $\text{H}_2\text{O}_2$  concentration in DMPBP[5]-BWO- $\text{Fe}_3\text{O}_4$  system was accumulated to  $0.06 \text{ mM}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$  under visible light, which was about 90.3 folds higher than that of BWO photocatalyst. This work provides a new insight into the adsorption-enhanced PSF synergy system for *in-situ*  $\text{H}_2\text{O}_2$  formation and transformation. As CdS photocatalyst has been reported to show an acceptable capacity for *in-situ*  $\text{H}_2\text{O}_2$  generation, Guo et al. constructed a PSF system based on red mud (RM) particles decorated CdS nanospheres (RM/CdS) for pollutant degradation [44]. The formed IEF at the interface of this S-scheme heterojunction promoted the separation and distribution of  $\text{e}^-$ - $\text{h}^+$  pairs. The  $\text{H}_2\text{O}_2$  yield of 2%RM/CdS ( $1.05 \text{ mg}\cdot\text{L}^{-1}\cdot\text{h}^{-1}$ ) in water under visible light was slightly lower than that of CdS ( $1.37 \text{ mg}\cdot\text{L}^{-1}\cdot\text{h}^{-1}$ ), suggesting  $\text{H}_2\text{O}_2$  *in-situ* generated by CdS could simultaneously be transformed into  $\bullet\text{OH}$  species by the activation of  $\text{Fe}^{2+}$  in RM. This research presents a novel idea of constructing integrated PSF system via the formation of S-scheme heterojunction. The modification of  $\text{Zn}_2\text{In}_2\text{S}_5$  has also been reported to result in enhanced  $\text{H}_2\text{O}_2$  production efficiency. Cao et al. designed a PSF system based on  $\text{Cu}^0/\text{CuO}_x$ @nitrogen-doped graphitic carbon ( $\text{Cu}^0/\text{CuO}_x$ -NC) decorated  $\text{Zn}_2\text{In}_2\text{S}_5$  (CuZS) for pollutant degradation [28]. The combination between  $\text{Cu}^0/\text{CuO}_x$ -NC and  $\text{Zn}_2\text{In}_2\text{S}_5$  improved solar utilization and reduced charge recombination. Thus, CuZS catalyst showed much higher  $\text{H}_2\text{O}_2$  yield in water under visible light ( $5.20 \text{ mM}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$ ), which was 5.0 folds higher than  $\text{Zn}_2\text{In}_2\text{S}_5$ . Besides, because of the complexation between  $\text{H}_2\text{O}_2$  and Cu species with different valence states on the surface of  $\text{Cu}^0/\text{CuO}_x$ -NC, the  $\text{H}_2\text{O}_2$  decomposition rate of CuZS was greatly enhanced compared with  $\text{Zn}_2\text{In}_2\text{S}_5$ , which was conducive to the activation of *in-situ* generated  $\text{H}_2\text{O}_2$ . This study offers a delicate strategy for the combination of *in-situ* production and consumption of  $\text{H}_2\text{O}_2$  to trigger the PSF reaction.

### 3. Applying Photocatalysis-Self-Fenton System for the High-Efficiency Removal of Environmental Contaminants

As PSF coupling technology conquers the limitations of single AOPs via sufficiently taking the advantages of high throughput feature of Fenton method and strong mineralization characteristic of photocatalysis method, it can achieve boosted oxidation capacity and mineralization level to promote the elimination performance towards hazardous substances [45]. In recent years, PSF synergetic systems have been widely applied for the high-efficiency removal of different kinds of environmental contaminants, including phenolic compounds, antibiotics, ARB and ARGs (Table 2) [24,33,46]. Under the combined effect of multiple reactive species ( $\bullet\text{OH}$ ,  $\text{h}^+$ ,  $\bullet\text{O}_2^-$ , etc.), PSF system can not only efficaciously mineralize the molecular structures of organic pollutants through various pathways (chemical bond breakage, aromatic ring opening, etc.) to decompose them into harmless small molecules, but also effectively damage the cell wall/membrane of ARB and the double helix structure of ARGs via varieties of routes (deoxyribose phosphate backbone cleavage, base opening, etc.) to hinder their repair and regeneration [47]. The removal performances of PSF systems were quantitatively evaluated via measuring the degradation rate constant ( $k$ ) value and total organic carbon (TOC) removal ratio towards organic pollutants as well as calculating the survival ratio and inactivation ratio towards ARB and ARGs. Besides, the pollutant removal efficiency of PSF system was constantly compared with those of the corresponding photocatalysis and homogeneous Fenton systems to highlight its superiority in environmental remediation. Moreover, the application potential of PSF system was mainly estimated via conducting the pollutant removal experiments in different conditions (pH, ion, natural organic matter (NOM), etc.) and various water matrices (lake water, aquaculture water, secondary effluent, etc.), cycling tests, and continuous-flow experiments to verify its practical feasibility in water treatment.

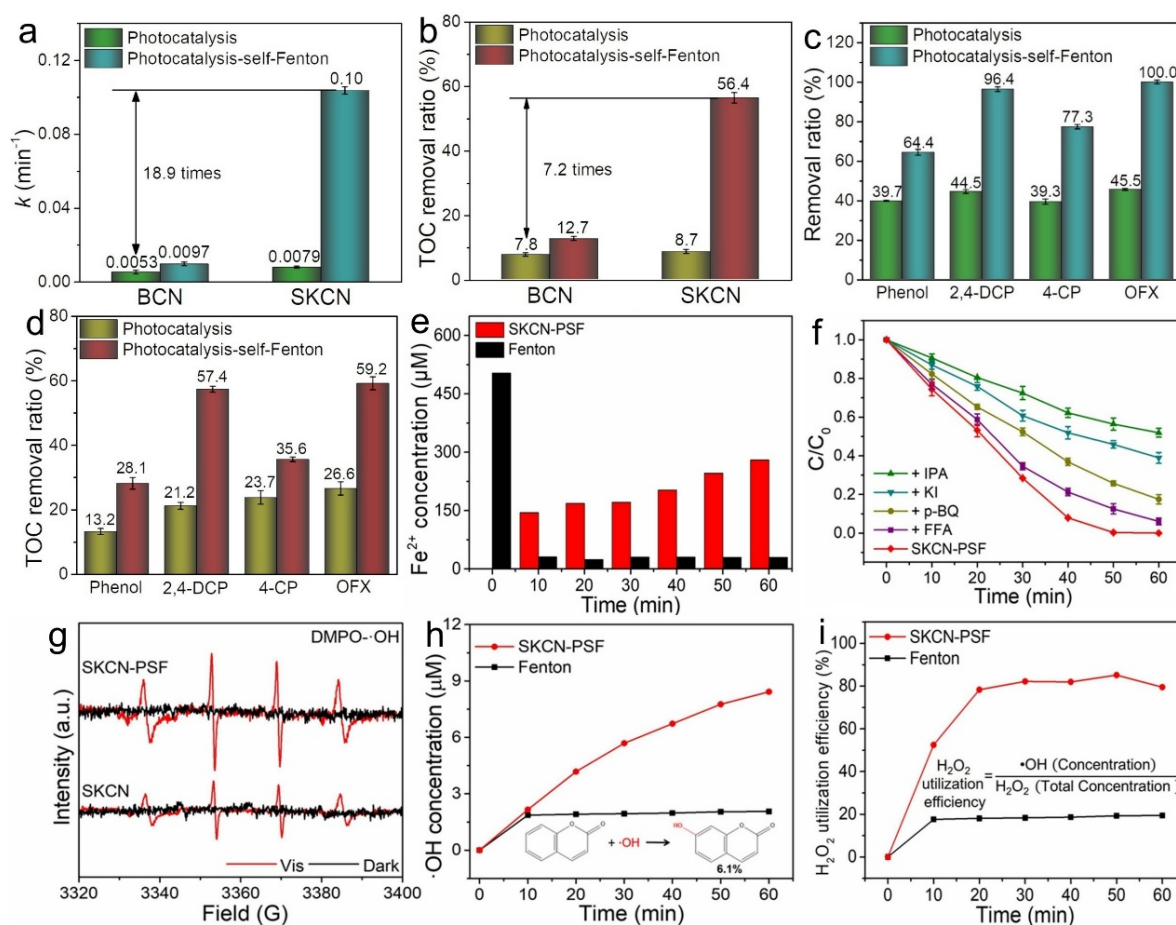
**Table 2.** Contaminant removal performances of different PSF systems.

| PSF System                                                          | Dosage (mg·mL <sup>-1</sup> ) | Light Source                                     | Reaction Time (h) | Contaminant                                                                   | Removal Rate         | TOC Removal Ratio | Ref. |
|---------------------------------------------------------------------|-------------------------------|--------------------------------------------------|-------------------|-------------------------------------------------------------------------------|----------------------|-------------------|------|
| P-g-C <sub>3</sub> N <sub>4</sub>                                   | 1.00                          | $\lambda > 420$ nm                               | 3.00              | 2,4-DCP (20 ppm)                                                              | 0.82 h <sup>-1</sup> | 42.7%             | [21] |
| GCN                                                                 | 0.50                          | $\lambda > 420$ nm<br>(700 mW·cm <sup>-2</sup> ) | 0.50              | 2,4-DCP (5 ppm)                                                               | 4.20 h <sup>-1</sup> | -                 | [22] |
| PSF System                                                          | Dosage (mg·mL <sup>-1</sup> ) | Light Source                                     | Reaction Time (h) | Contaminant                                                                   | Removal Rate         | TOC Removal Ratio | Ref. |
| Resin                                                               | 0.60                          | $\lambda > 420$ nm<br>(70 mW·cm <sup>-2</sup> )  | 2.00              | BPA (20 ppm)                                                                  | 1.84 h <sup>-1</sup> | 67.0%             | [23] |
| OPCN                                                                | 0.50                          | $\lambda > 400$ nm<br>(100 mW·cm <sup>-2</sup> ) | 1.00              | 2,4-DCP (5 ppm)                                                               | 4.14 h <sup>-1</sup> | 42.2%             | [24] |
| Ti <sub>3</sub> C <sub>2</sub> MXene/MIL-100(Fe)                    | 0.20                          | UV-vis light                                     | 2.00              | Thiacloprid (80 ppm)                                                          | 3.06 h <sup>-1</sup> | -                 | [42] |
| Graphitic carbon interface-modified g-C <sub>3</sub> N <sub>4</sub> | 1.00                          | $\lambda > 420$ nm                               | 3.00              | Rhodamine B                                                                   | 1.07 h <sup>-1</sup> | 63.8%             | [11] |
| MoS <sub>2</sub> @carbon nitride nanotube                           | 1.00                          | $\lambda < 420$ nm                               | 2.00              | Tetracycline (10 ppm)                                                         | 1.40 h <sup>-1</sup> | -                 | [48] |
| CPBA-CN                                                             | 1.00                          | Simulated sunlight                               | 1.00              | 4-CP (20 ppm)                                                                 | 6.00 h <sup>-1</sup> | 74.5%             | [10] |
| CoFeO/PDIsm                                                         | 1.00                          | $\lambda > 420$ nm                               | 1.00              | CIP (10 ppm)                                                                  | 2.67 h <sup>-1</sup> | 63.7%             | [29] |
| Fe/N-CQDs/MCN1                                                      | 0.50                          | $\lambda < 420$ nm                               | 2.00              | CIP (15 ppm)                                                                  | 0.54 h <sup>-1</sup> | -                 | [49] |
| Coral-B-CN                                                          | 0.40                          | Simulated sunlight                               | 0.83              | 4-CP (20 ppm)                                                                 | 5.51 h <sup>-1</sup> | 70.3%             | [30] |
| RM/CdS                                                              | 0.50                          | 400 nm < $\lambda$ < 760 nm                      | 2.00              | Amoxicillin (20 ppm)                                                          | 0.46 h <sup>-1</sup> | -                 | [44] |
| rGO/SA-TCPP                                                         | 0.30                          | $\lambda > 400$ nm<br>(100 mW·cm <sup>-2</sup> ) | 6.00              | SDM (10 ppm)                                                                  | 0.53 h <sup>-1</sup> | 67.9%             | [33] |
| SKCN                                                                | 1.00                          | $\lambda > 400$ nm<br>(100 mW·cm <sup>-2</sup> ) | 1.00              | BPA (10 ppm)                                                                  | 6.00 h <sup>-1</sup> | 56.4%             | [34] |
| GO/SA-PDI                                                           | 0.20                          | $\lambda > 400$ nm<br>(100 mW·cm <sup>-2</sup> ) | 0.25              | Sulfonamide ARB<br>(1×10 <sup>7</sup> CFU·mL <sup>-1</sup> )                  | 100.0%               | -                 | [50] |
| GO/SA-PDI                                                           | 0.25                          | $\lambda > 400$ nm<br>(100 mW·cm <sup>-2</sup> ) | 2.00              | Sulfonamide ARGs<br>(10 <sup>7.19</sup> copies·μL <sup>-1</sup> <i>su11</i> ) | 99.9%                | -                 | [50] |
| ST-NS/NC                                                            | 0.20                          | $\lambda > 400$ nm<br>(100 mW·cm <sup>-2</sup> ) | 2.00              | Sulfonamide ARB<br>(1×10 <sup>8</sup> CFU·mL <sup>-1</sup> )                  | 100.0%               | -                 | [47] |
| ST-NS/NC                                                            | 0.25                          | $\lambda > 400$ nm<br>(100 mW·cm <sup>-2</sup> ) | 2.00              | Sulfonamide ARGs<br>(10 <sup>7.63</sup> copies·μL <sup>-1</sup> <i>su11</i> ) | 99.6%                | -                 | [47] |

### 3.1. Phenolic Compounds

Phenolic compounds have been listed as one type of priority-controlled pollutants due to their acute toxicity, genotoxicity, and endocrine-disruption, thus becoming the main target of PSF systems [51]. Zhu's group constructed a PSF system based on P-doped g-C<sub>3</sub>N<sub>4</sub> (P-g-C<sub>3</sub>N<sub>4</sub>) and Fe<sup>2+</sup> [21]. P doping enhanced visible-light absorption and oxidation abilities of g-C<sub>3</sub>N<sub>4</sub>, which was favorable for H<sub>2</sub>O<sub>2</sub> generation (783 μM·h<sup>-1</sup>·g<sup>-1</sup>). H<sub>2</sub>O<sub>2</sub> *in-situ* generated by P-g-C<sub>3</sub>N<sub>4</sub> was efficiently utilized by Fe<sup>2+</sup> to produce abundant •OH, which obtained high-fluent phenols removal efficiency under visible light. The degradation and mineralization activities of P-g-C<sub>3</sub>N<sub>4</sub>-PSF for 2,4-dichlorophenol (2,4-DCP) were 20.5 and 22.5 times as those of bulk g-C<sub>3</sub>N<sub>4</sub> (BCN)-photocatalysis, 3.7 and 3.8 times as those of Fenton, respectively. Besides, the degradation reactions were effectively proceeded at acidic, neutral and weakly alkaline conditions, indicating P-g-C<sub>3</sub>N<sub>4</sub>-PSF system can be applied over a wide pH range. This work proposes a new collaborative strategy to address the bottlenecks of traditional Fenton reaction for controlling phenolic pollution. Dong's group reported a PSF system based on graphite carbon rings (Cg)-doped g-C<sub>3</sub>N<sub>4</sub> (Cg-C<sub>3</sub>N<sub>4</sub>) and Fe<sup>2+</sup> [52]. The introduction of Cg in the conjugated network of g-C<sub>3</sub>N<sub>4</sub> led to a more positive VB position (2.20 eV) and wider spectral response range, thus enhancing the mineralization ability and formation amount of h<sup>+</sup>. Meanwhile, IEF established by Cg-doping enhanced carriers separation of Cg-C<sub>3</sub>N<sub>4</sub>, thus raising H<sub>2</sub>O<sub>2</sub> generation (217 μM·h<sup>-1</sup>·g<sup>-1</sup>) and facilitating the cycle of Fe<sup>3+</sup>/Fe<sup>2+</sup> to sustainably produce abundant •OH. In contrast, Cg-C<sub>3</sub>N<sub>4</sub>-PSF system showed remarkably

boosted phenols removal efficiency. The corresponding  $k$  value towards 4-chlorophenol (4-CP) degradation was 32.1 folds as that of g-C<sub>3</sub>N<sub>4</sub>-photocatalysis, while TOC removal ratio was 11.4 folds as that of Fenton. This study paves a new avenue for developing PSF system to eliminate phenolic pollutants. Wang's group designed a PSF system based on S, K co-doped g-C<sub>3</sub>N<sub>4</sub> nanosheets (SKCN) and Fe<sup>3+</sup> (Figure 6) [34]. The unique structure of SKCN accelerated charge migration and created more reactive sites, thus increasing H<sub>2</sub>O<sub>2</sub> yield (625  $\mu\text{M}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$ ) to trigger the self-Fenton reaction. Moreover, Fe<sup>3+</sup> was rapidly turned into Fe<sup>2+</sup> via reductive e<sup>-</sup>, which facilitated continuous transformation from H<sub>2</sub>O<sub>2</sub> to  $\bullet\text{OH}$ . Additionally, the timely consumption of e<sup>-</sup> suppressed charge recombination to release more h<sup>+</sup>. Hence, SKCN-PSF system achieved enhanced removal efficiency towards phenolic compounds. The  $k$  value of SKCN-PSF system (0.10 min<sup>-1</sup>) towards bisphenol A (BPA) degradation was 43.5 and 18.9 times as those of Fenton and g-C<sub>3</sub>N<sub>4</sub>-photocatalysis systems, respectively, while the corresponding TOC removal ratio (56.4%) was improved by 12.8 and 7.2 times, respectively. This study shows a promising feasibility of PSF system for removing phenolic compounds.



**Figure 6.** (a) The  $k$  values and (b) TOC removal ratios of photocatalysis and PSF systems based on SKCN and BCN towards BPA degradation. (c) The removal ratios and (d) TOC removal ratios of photocatalysis and PSF systems based on SKCN towards other phenolic pollutants. (e) Fe<sup>2+</sup> concentration changes over time in SKCN-PSF and Fenton systems. (f) BPA degradation curves of SKCN-PSF system with the addition of different scavengers. (g) EPR spectra for the detection of  $\bullet\text{OH}$ . The changes of (h)  $\bullet\text{OH}$  concentration and (i) H<sub>2</sub>O<sub>2</sub> utilization efficiency over time in SKCN-PSF and Fenton systems [34]. Copyright © 2024 Elsevier.

### 3.2. Antibiotics

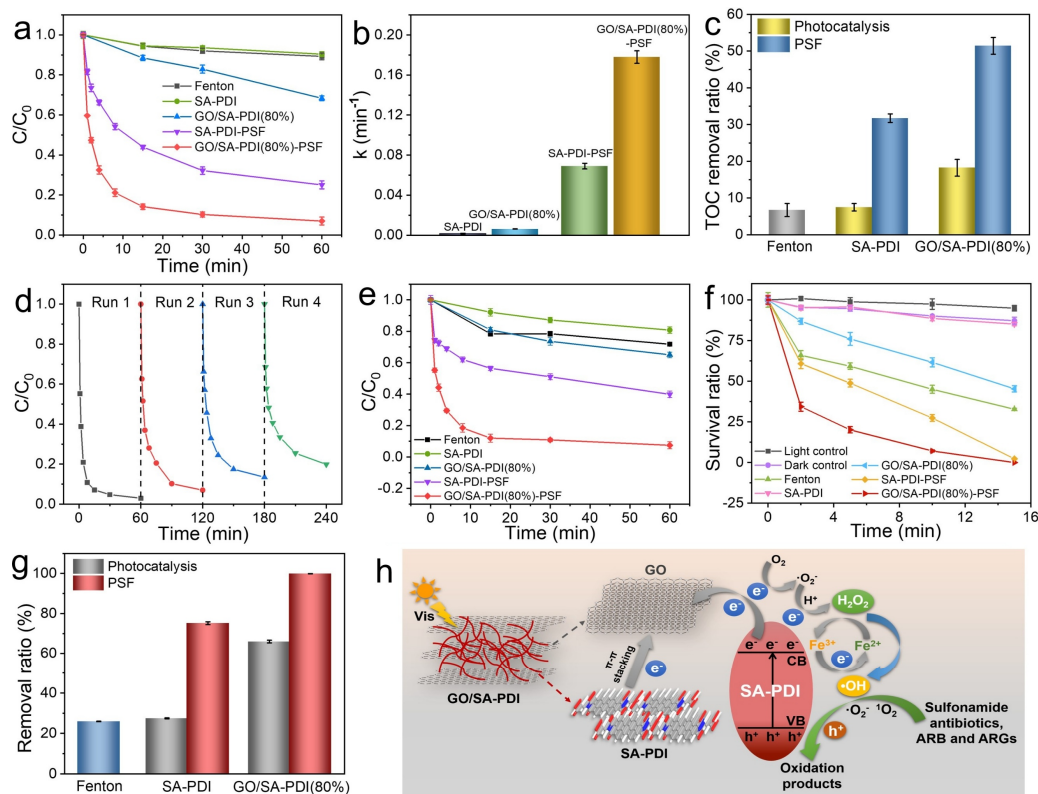
Antibiotics have been classified as one kind of new pollutants under key control due to their environmental persistence, biotoxicity and bioaccumulation [53,54]. Thus, PSF systems were also applied for the elimination of antibiotics. Yang's group designed a PSF system based on g-C<sub>3</sub>N<sub>4</sub> nanosheets with carbon vacancies (Cv-PCNNS) and Fe<sup>2+</sup> for efficient metronidazole (MTZ) degradation [27]. Carbon vacancies and nanostructure improved carriers transfer and adsorbed O<sub>2</sub> activation. Besides, -NH<sub>x</sub> groups around carbon vacancies provided protons for one-step 2e<sup>-</sup> ORR to boost H<sub>2</sub>O<sub>2</sub> production rate (19.7 mM·h<sup>-1</sup>·g<sup>-1</sup>), which was 10.0 folds higher than BCN. Benefiting from the cascade mode of PSF reaction, the MTZ degradation efficiency and TOC removal ratio of Cv-PCNNS-PSF reached 90.7% and 62.0% within 100 min, about 5.0 and 2.8 folds

higher than those of Cv-PCNNS-photocatalysis and Fenton, respectively. This work shows the advantage of highly active g-C<sub>3</sub>N<sub>4</sub>-PSF system for antibiotics elimination. Dong's group designed a PSF system based on Co-Fe oxide/PDI supermolecules (CoFeO/PDIsm) and Fe<sup>2+</sup> [29]. Z-scheme heterojunction structure endowed CoFeO/PDIsm with faster charge transfer rate and more superior redox ability. e<sup>-</sup> accumulated in the conduction band (CB) of PDIsm were used for H<sub>2</sub>O<sub>2</sub> generation and Fe<sup>3+</sup> regeneration, while h<sup>+</sup> cumulated in the VB of CoFeO cooperated with •OH were used for ciprofloxacin (CIP) mineralization. Thus, CoFeO/PDIsm showed extremely enhanced H<sub>2</sub>O<sub>2</sub> yield of 5.6 mM·h<sup>-1</sup>·g<sup>-1</sup> in CIP solution. Moreover, the *k* value (0.045 min<sup>-1</sup>) and TOC removal ratio (63.7%) of CoFeO/PDIsm-PSF towards CIP degradation were 9.7 and 10.4 folds as those of PDIsm-photocatalysis, respectively. This study designs a novel Z-scheme heterojunction photocatalyst to construct PSF synergetic system for high-efficiency antibiotics removal. Wang's group developed a PSF system based on Fe-doped resorcinol formaldehyde (FRF) resin, which could degrade and mineralize different kinds of antibiotics (sulfonamides, quinolones, tetracyclines and macrolides) effectively at neutral condition without additional reagents [55]. Compared to RF-photocatalysis, the degradation activity towards oxytetracycline (OTC) raised by 34.2 folds. The improved removal performance of FRF resin-PSF system was owing to two main factors as follows: (i) Fe-doping can facilitate electron transfer, thus promoting the recycling of Fe<sup>3+</sup>/Fe<sup>2+</sup> and facilitating the release of more free h<sup>+</sup>. (ii) *In-situ* produced H<sub>2</sub>O<sub>2</sub> can be efficiently utilized via heterogeneous self-Fenton reaction to generate abundant •OH. This study provides a new pathway to construct heterogeneous PSF system for removing antibiotics.

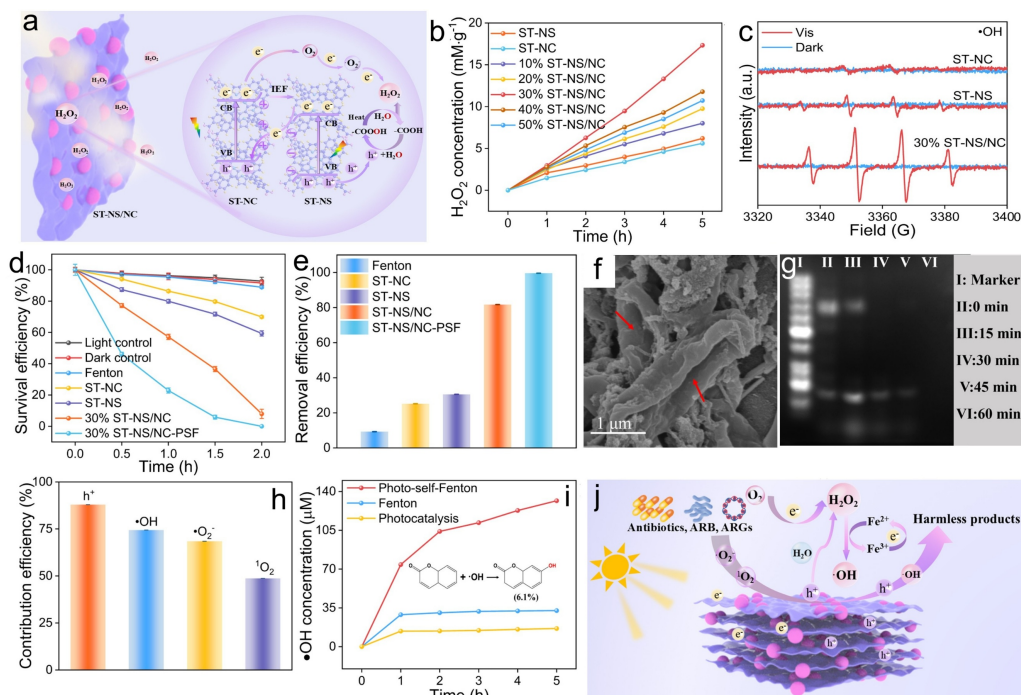
### 3.3. Antibiotic Resistant Bacteria and Resistance Genes

ARB and ARGs are two other kinds of new pollutants, which can make the spread of antibiotic resistance more widespread and reduce the cure rate of bacterial diseases [56]. Recently, several PSF systems were developed to remove ARB and ARGs. Wang's group constructed a PSF system based on graphene oxide/SA-PDI (GO/SA-PDI) and Fe<sup>3+</sup> (Figure 7) [50]. The *k* value (0.18 min<sup>-1</sup>) towards sulfamethoxazole (SMX) degradation of GO/SA-PDI-PSF was 120.0 times as that of SA-PDI-photocatalysis system, while the TOC removal ratio within 1 h (57.2%) towards sulfadimethoxine (SDM) degradation was 9.7 times as that of Fenton system. The inactivation ratio of GO/SA-PDI-PSF system toward sulfonamide ARB (100.0%) within 15 min was 6.8 folds as that of SA-PDI-photocatalysis, while the corresponding inactivation ratio toward sulfonamide ARGs (99.9%) within 2 h was 3.9 folds as that of Fenton. The higher oxidation capacity and mineralization efficiency of GO/SA-PDI-PSF system were due to the interaction between GO and SA-PDI for increasing the H<sub>2</sub>O<sub>2</sub> production (1.50 mM·h<sup>-1</sup>·g<sup>-1</sup>), as well as the synergic action between photocatalysis and Fenton reactions for enhancing the formation of •OH and free h<sup>+</sup>. This work offers a new route to develop efficient PSF system to control antibiotic resistance pollution in water. Xu's group developed a PSF system based on SA-TCPP nanosheets/SA-TCPP nanocrystals (ST-NS/NC) homojunction and Fe<sup>2+</sup> (Figure 8) [47]. The formed IEF promoted the interfacial charge separation and thus improved the H<sub>2</sub>O<sub>2</sub> production activity of ST-NS/NC homojunction (3.46 mM·g<sup>-1</sup>·h<sup>-1</sup>), which was 2.8 and 3.1 times as that of SA-TCPP nanosheets (ST-NS) and SA-TCPP nanocrystals (ST-NC), respectively. The trigger of PSF reaction led to the formation of multiple oxidizing species (h<sup>+</sup>, •OH), endowing ST-NS/NC-PSF system more excellent elimination performance towards sulfonamide ARB and ARGs. The inactivation efficiencies of ST-NS/NC-PSF system within 2 h were 100.0% for sulfonamide ARB and 99.6% for sulfonamide ARGs, which were significantly higher than those of ST-NC (30.1%/25.2%) and ST-NS (40.7%/30.2%) photocatalysis. Furthermore, ST-NS/NC-PSF system had prevented ARB and ARGs from regeneration by disrupting their genomic DNA. This study shows a promising feasibility of PSF system for removing biological pollutants. Xu's group designed a PSF-photothermal antibacterial nanoplatform based on Co-N-C/OPCN catalyst [57]. Co-N-C/OPCN showed boosted antimicrobial ratio (100.0%) towards sulfonamide-resistant *Staphylococcus aureus* under visible light within 9 min, which was outperforming OPCN (59.1%) and Co-N-C (31.7%). Meanwhile, the removal ratio of Co-N-C/OPCN (94.5%) towards quinolone ARGs within 30 min was also superior to that of Co-N-C (21.9%). The considerably improved disinfection performance could be primarily attributed to the PSF effect between Co<sup>2+</sup> from Co-N-C and H<sub>2</sub>O<sub>2</sub> *in-situ* produced by OPCN as well as the photothermal conversion ability of Co-N-C. The study offers a new strategy of integrating PSF technology with photothermal approach for controlling antibiotic resistance pollution.





**Figure 7.** (a) The SMX degradation curves, (b)  $k$  values, and (c) TOC removal ratios of Fenton, photocatalysis and PSF systems based on SA-PDI and GO/SA-PDI. (d) The cycle tests of SMX degradation over GO/SA-PDI-PSF system. (e) The SDM degradation curves of Fenton, photocatalysis and PSF systems based on SA-PDI and GO/SA-PDI. (f) The inactivation activities towards (f) sulfonamide ARB and (g) ARGs (*su1*). (h) The reaction mechanism diagram of GO/SA-PDI-PSF system [50]. Copyright © 2025 Elsevier.



**Figure 8.** (a) The schematic diagram of  $\text{H}_2\text{O}_2$  synthesis mechanism over ST-NS/NC homojunction. (b) The  $\text{H}_2\text{O}_2$  production curves of ST-NC, ST-NS and ST-NS/NC. (c) ESR spectra for the detection of  $\cdot\text{OH}$ . The comparison of (d) the survival ratio of sulfonamide ARB and (e) the removal ratio of sulfonamide ARGs (*su1*) over Fenton, photocatalysis and ST-NS/NC-PSF systems. (f) SEM image of sulfonamide ARB after treated by ST-NS/NC-PSF system for 2 h. (g) The agarose gel electrophoresis image of the PCR products extracted from sulfonamide ARB cells treated by ST-NS/NC-PSF system. (h) The contribution ratios of different active species in ST-NS/NC-PSF system. (i)  $\cdot\text{OH}$  concentration variation trends in different systems. (j) The proposed mechanism of ST-NS/NC-PSF system for the elimination of sulfonamide ARB and ARGs [47]. Copyright © 2025 Elsevier.

#### 4. Summary, Challenge and Outlook

In conclusion, this mini-review summarized varieties of PSF collaborative systems based on photocatalysts in previous literatures, and discussed their reaction mechanisms and practical applications in environmental pollutants removal. Moreover, the strategies of various modification methods to improve  $\text{H}_2\text{O}_2$  production activity of photocatalysts were also detailed explored including morphology optimizing, heteroelement doping, defect engineering and heterogeneous structure construction. The current researches have made significant progress to solve the problems of photocatalysis reaction via increasing solar utilization, expediting charge migration, and boosting the quantity of oxidizing specie, as well as remedy the drawbacks of Fenton reaction via avoiding extra dosage of  $\text{H}_2\text{O}_2$ , accelerating the cyclic conversion of  $\text{Fe}^{3+}/\text{Fe}^{2+}$ , and expanding pH range, thus enhancing the oxidation ability and mineralization rate of PSF system.

Although PSF collaborative system demonstrates excellent pollutant removal performance on a laboratory scale, its practical application still faces several key operational challenges. Firstly, the fouling problem: NOM, metal ions, and suspended solids in real wastewater can deposit on the photocatalyst surface, blocking active sites and reducing light penetration. Secondly, the catalyst deactivation problem: iron leaching from iron-based materials, the photocorrosion of semiconductors (e.g., CdS,  $\text{BiVO}_4$ ), and the occupation of surface active sites by reaction intermediates or anions (e.g.,  $\text{Cl}^-$ ,  $\text{HCO}_3^-$ ), all prone to cause the catalyst deactivation. Thirdly, the energy consumption problem: the electricity requirement of high-power light sources, low energy conversion efficiency, and the need for external aeration or mechanical stirring, increasing the operating energy consumption. Fourthly, the environmental safety concerns: the difficulty of recycling the catalyst from the reaction system, the potential toxicity of nanocatalyst on aquatic organisms (e.g., oxidative stress, membrane damage, bioaccumulation), and the risk of metal leaching (e.g.,  $\text{Fe}^{2+}$ ,  $\text{Co}^{2+}$ ) into the treated water (e.g., exceeding discharge limits, requiring post-treatment), raising the environmental safety concerns.

To further promote the application potential of PSF technology in water treatment, there are some outlooks to be addressed in future studies as follows: (1) Iron species can be anchored on the photocatalyst to trigger self-Fenton reaction rather than dosed additionally, thus constructing an integrated PSF catalyst. (2) Future research should not only focus on broadening the spectral response range of photocatalyst to near-infrared region, but also concentrate on designing “low-loss, low-cost” composite interfaces, such as adopting earth-abundant plasmonic alternatives and exploring triplet-triplet annihilation-based up-conversion systems that operate at low light intensity. (3) The recycling ability of PSF system can be improved by combining catalyst with magnetic substance or modifying catalyst into hydrogel, which is beneficial for recovering catalyst from reaction system and reducing environmental safety risk. (4) Except for phenolic compounds, antibiotics, ARB and ARGs, PSF system can also be utilized to remove other contaminants (pesticides, microplastics, etc.). (5) PSF technology can be embedded into some advanced processing equipments (fluidized bed, tower reactor, etc.) to achieve large-scale industrial application in wastewater treatment. (6) The nanomaterial ecotoxicity and metal leaching tests can be routinely conducted, while life cycle assessment can be combined to evaluate the environmental benefits and potential costs from a holistic perspective.

We hope this mini-review will provide a better understanding of PSF synergistic mechanism during pollutants removal process and further promote the development of PSF technology in water treatment.

#### Author Contributions

J.X.: Conceptualization, literature investigation, data curation, visualization, writing—original draft preparation, fund acquiring; H.Z.: Supervision, validation, writing—reviewing and editing. All authors have read and agreed to the published version of the manuscript.

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## Data Availability Statement

Not applicable.

## Conflicts of Interest

The authors declare no conflict of interest.

## Use of AI and AI-Assisted Technologies

No AI tools were utilized for this paper.

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