

Review

Porphyrin-Conjugated Hybrid Nanomaterials for Photocatalytic Wastewater Remediation

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Abstract

Advanced oxidation processes using porphyrin-based heterogeneous catalysts hold promise for removing hazardous pollutants from wastewater. Their high visible-light absorption coefficients enable absorption of light from the solar spectrum. Moreover, their conjugated aromatic skeletons and intrinsic electronic properties facilitate the delocalization of photogenerated electrons during photodegradation. Delaying the recombination of photogenerated electron-hole pairs by introducing specific materials increases efficiency, as separated charges have more time to participate in redox reactions, boosting photocatalytic activities. However, applying these photocatalysts for wastewater treatment is challenging owing to facile agglomeration, deactivation, and recovery of the photocatalyst for reuse, which can significantly increase the overall cost. Therefore, new photocatalytic systems comprising porphyrin molecules must be developed. For this purpose, porphyrins can be conjugated to nanomaterials to create hybrid materials with photocatalytic efficiencies superior to those of free-standing starting porphyrins. Various transition metal oxides (TiO_2 , ZnO , and Fe_3O_4) nanoparticles, main-group-element oxides (Al_2O_3 and SiO_2) nanoparticles, metal plasmons (silver nanoparticles), carbon-based platforms (graphene, graphene oxide, and $\text{g-C}_3\text{N}_4$), and polymer matrices have been used as nanostructured solid supports for the successful fabrication of porphyrin-conjugated hybrid materials. The conjugation of porphyrin molecules to solid supports improves the photocatalytic degradation activity in terms of visible-light conversion ability, recyclability, active porous sites, substrate mobility, separation of photogenerated charge species, recovery for reuse, and chemical stability, along with preventing the generation of secondary pollution. This review discusses the ongoing development of porphyrin-conjugated hybrid nanomaterials for the heterogeneous photocatalytic degradation of organic dyes, pharmaceutical pollutants, heavy metals, pesticides, and human care in water. Several important results and advancements in the field allow for a more efficient wastewater remediation process.



Academic Editors: Jason G. Parsons
and Matas Alcoutlabi

Received: 27 November 2025

Revised: 22 January 2026

Accepted: 2 February 2026

Published: 4 February 2026

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Keywords: porphyrins; solid supports; hybrid nanomaterials; wastewater remediation; heterogeneous photocatalysis

1. Introduction

Environmental pollution resulting from the rapid growth of industrialization and civilization is increasing. Every year, large quantities of toxic compounds such as synthetic dyes, pesticides, biphenyls, phenols, plasticizers, pigments, herbicides, and nitro/amino molecules are released from leather, textile, paper printing, and pharmaceutical industries into water bodies. These hazardous contaminants not only harm water potability but also

pose a major threat to the entire ecosystem and negatively impact aquatic homeostasis [1,2]. This has prompted researchers and environmentalists to develop advanced approaches for wastewater treatment [3–6]. Several physicochemical and biological processes for removing hazardous compounds from wastewater have been reported, including advanced oxidation processes (AOPs) [7], adsorption [8], chemical coagulation [9], precipitation [10], and bacterial treatments [11].

Among different methods used in wastewater treatment, the advanced oxidative processes (AOPs) are an increasingly adopted process for wastewater treatment owing to their simplicity, economic affordability, and high degradation activity. Moreover, this technique oxidizes toxic contaminants to less toxic CO_2 and H_2O , without causing additional pollution. In these techniques, reactive oxygen species (ROS) are generated after light absorption by an ideal photocatalyst and oxidize toxic molecules [12–17]. Heterogeneous photodegradation reaction has received significant attention owing to the ecological and economic advantages associated with the use of freely available solar light. The degradation mechanism consists of several simple steps. Light harvesting and exciton diffusion occur first, followed by charge separation and transport. Therefore, light absorption and subsequent electron transfer play crucial roles in the degradation of toxic compounds in water. Microscale or nanoscale architectures generally exhibit interesting electronic and optical characteristics, depending on their morphologies. Consequently, the design and construction of micro and nanoscale frameworks are fundamental for fabricating photocatalytic systems with special functions and properties [18–23].

Owing to the benefits of catalytic photodegradation of pollutants in water, numerous organic and inorganic micro- and nanomaterials have been used as building blocks to develop photocatalytic systems. Several inorganic nano and microstructures such as metal-oxide nanoparticles (TiO_2 [24], ZnO [25], SnO_2 [26], Fe_3O_4 [27], CuO [28]), MgO [29], V_2O_5 [30], CeO_2 [31], silicates [32], CdS [33], zeolites [34], and bismuth-based catalysts [35] have been employed to remove hazardous contaminants from sewerage. Among these nanomaterials, TiO_2 and ZnO have emerged as the most useful photocatalysts owing to their large permanent porosity, low cost, eco-friendliness, low toxicity, high stability, and high degree of degradation efficiency for the catalytic photodegradation of toxic compounds in H_2O . However, the large band gap energies (E_g) of ZnO (approximately 3.37 eV) and TiO_2 (approximately 3.20 eV) restrict the absorption of solar light under the visible-light region ($\lambda < 370$ nm). Moreover, a high degree of recombination between the photoinduced electrons and holes limits its practical application. This lowers their visible-light photodegradation activity. Additionally, a high amount of TiO_2 or ZnO is required for initial loading in the catalytic photodegradation reaction to attain fruitful decomposition rates [36–39].

Compared with inorganic photocatalysts, organic-based micro- and nanostructures have received ample attention. This is because of the considerable flexibility in molecular design, outstanding solution-phase characteristics, and excellent tunability of the optoelectronic properties, which make them unique for catalytic photodegradation systems. Various organic building blocks such as graphene oxide (GO) or reduced graphene oxide (rGO) [40], fullerene [41], carbon quantum dots (CQDs) [42], graphitic carbon nitride (g- C_3N_4) [43], and porphyrins [44] have been used to fabricate organic photocatalytic systems for the removal of toxic pollutants from wastewater. Distinguished π -conjugated planar porphyrins are of great interest for their variable coordination geometries and excellent optoelectronic properties [45–50]. Owing to their high absorption coefficients, porphyrins can absorb light in the UV–Vis range and initiate ROS production via a spin-forbidden intersystem crossing route. Moreover, their conjugated aromatic skeleton and inherent electronic properties facilitate self-assembly in solution as well as in the solid phase. Owing

to these properties, porphyrin-based compounds exhibit outstanding photocatalytic activity and have applications in photocatalytic degradation activities towards organic dyes (rhodamine B: RhB, methylene blue: MB, methyl orange: MO), pharmaceutical pollutants (Tetracycline: TC), and many more pollutants. However, their application is restricted because they easily agglomerate in water, are prone to deactivation, have poor recyclability, and are difficult to recover after catalyzing the photodegradation reaction [51–56].

Therefore, new porphyrin-based photocatalytic systems that overcome the aforementioned limitations must be developed. For this aim, porphyrins can be conjugated to solid supports to fabricate hybrid materials with outstanding photocatalytic efficiencies compared with those of free-standing starting porphyrins. Numerous nanoparticles, hydrogels, and zeolites have been used as solid supports for the successful formation of porphyrin-integrated hybrid materials. Additionally, the conjugation of porphyrins to solid supports not only enhances chemical stability but also increases their use several times. Fabrication of solid-supported porphyrin hybrid materials not only uses solar or artificial light efficiently but also lowers the recombination of photoinduced ion-pairs compared to their starting porphyrin. This type of hybridization facilitates complete recovery from the reaction medium and prevents the generation of secondary pollution [57–60].

Recently, several reports have described the recent advancements in porphyrin-based materials and their utilization in energy-linked sectors [61–64]. Owing to their vast array of functional properties and considerable morphology, porphyrin-containing materials have been investigated for the removal of toxic pollutants from wastewater. Porphyrin-based materials can be easily fabricated, and their morphology can be regulated. The morphology of photoactive materials is closely related to their properties, such as photon absorption and conversion, permanent porosity, separation and transportation of photogenerated pairs, active and defect sites, and the interaction of pollutants. However, a substantial understanding of the correlation between porphyrin-conjugated hybrid materials and their catalytic photodegradation efficiencies for wastewater treatment is lacking. In this report, we discuss the recent developments in the fabrication of porphyrin-conjugated materials, their applications in wastewater treatment, and the future directions of these hybrid materials as prospective candidates for other applications. The authors hope that this systematic review can serve as a guide for new researchers for selecting appropriate porphyrin-conjugated hybrid materials and assist in the future development in energy-related applications as well as water remediation.

2. Fundamentals of the Photocatalytic Degradation Process

As depicted in Figure 1, a suitable photocatalyst is energized by the absorption of a photon ($h\nu$) with adequate energy (equal to or higher than the bandgap energy (E_g) of the semiconducting materials). This process will generate a pair of holes (e^- in the highest energy conduction band (CB) and h^+ in the lowest energy valence band (VB)). E_g determines the light-harvesting properties of a photocatalyst: a photocatalyst with a low band gap energy can convert more visible-light photons than a material with a high band gap energy. The next step involves the separation and recombination of the photogenerated pairs (holes and electrons). A photocatalytic reaction can proceed smoothly if the recombination rate of the hole pairs is suppressed. Otherwise, recombination reduces the number of excited charge carriers. The photogenerated holes react with the substrate to produce an oxidized product, and the excited electrons react with the substrate to produce a reduced product. Simply, the photogenerated excited electrons can react with dissolved oxygen (O_2) in water and reduce it to superoxide radical anion ($O_2^{\cdot-}$). The reaction of H_2O with photogenerated reactive holes leads to the formation of highly reactive hydroxyl radicals ($OH^\bullet$). In some cases, peroxide radicals are generated during photodegradation.

The resulting strong oxidizing species $\text{OH}^\bullet$ and $\text{O}_2^{\bullet-}$ can degrade pollutants to smaller fragments and finally mineralize to CO_2 and H_2O . It should be noted that the generation of reactive oxygen species such as $\text{OH}^\bullet$, $\text{O}_2^{\bullet-}$, H_2O_2 , $^\bullet\text{OOH}$, and singlet oxygen $^1\text{O}_2$ depends on the reaction conditions (pH of the solution, catalytic materials, and the presence of any scavenger) [7,12].

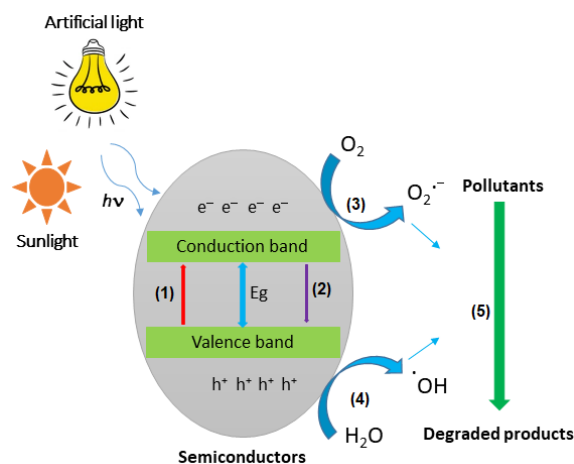
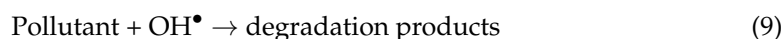
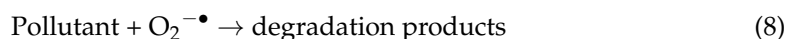
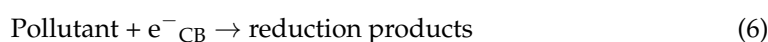
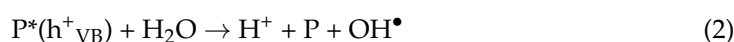


Figure 1. General mechanism for photocatalytic processes over a photocatalyst in H_2O . Excitation and recombination of photogenerated pairs (1) and (2); generation of reactive oxygen species (3) and (4); and photodegradation (5).

The relevant steps for the photocatalytic decomposition of contaminants in H_2O at the catalyst surface are illustrated in Equations (1)–(9).



where $h\nu$ is the photon energy essential for the promotion of electrons from the VB to the CB.

The catalytic performance of an ideal catalyst depends on various crucial features, such as the band gap energy of semiconducting materials, recombination time of electrons and holes, surface area and surface morphology, phase composition, crystallinity, active sites, absorption of light through catalysts, and surface interaction of the substrate with the catalyst. Thus far, photogenerated holes and electrons have been shown to engage in several surface chemical reactions. These pairs of charged carriers can also combine at the surface. Pollutants are first adsorbed on the active sites of the photocatalyst, which enhances charge mobility and redox capacity. Subsequently, a series of chemical reactions with the photogenerated active species occurs, forming degradation products. A higher band gap energy limits the light-harvesting properties. The rapid recombination of the

photogenerated reactive species remarkably suppresses the photocatalytic efficiency. Enhancing photocatalytic performance by lowering the band gap energy and delaying the recombination of photogenerated pairs has received increasing attention from the materials chemistry community [59–61].

Several modifications have been developed to overcome these limitations. The introduction of heterogeneous photocatalytic systems consisting of various porphyrins with solid supports (inorganic oxides, metal plasmons, carbonaceous compounds, and polymer substrates) has contributed to outstanding results in the photocatalytic degradation of hazardous compounds in water compared with the starting porphyrin. The incorporation of porphyrin molecules onto the surface of solid supports not only controls the electronic properties but also significantly enhances the visible-light harvesting capacity, which provides interesting results for the design of excellent photocatalysts for the degradation of pollutants in water. The formation of heterojunctions between porphyrins and semiconductors can directly convert photon energy to generate radicals, resist the destruction of active species, and hence improve recyclability. The recombination time of the photogenerated active pairs is enhanced owing to extensive electronic delocalization over the surface of heterojunctions composed of two semiconducting materials. Based on the electron transport method and the type of semiconductor, heterojunctions can be classified into three categories: type I, type II, and Z-scheme (Figure 2). Owing to their intrinsic benefits, such as high redox capacity and effective separation of photogenerated holes and electrons, Z-scheme heterojunction photocatalysts prominently improve the photodegradation performance of catalysts [65,66]. Z-scheme heterojunction photocatalysts can be further divided into three subgroups depending on the nature of the semiconductor materials used to fabricate the heterojunctions (Figure 2a–c). Furthermore, the Z-scheme hybrid junction can increase the separation of photogenerated reactive pairs, delay the recombination time, and improve the transportation of charged species; hence, more reactive species are produced for the photodegradation of toxic materials in water.

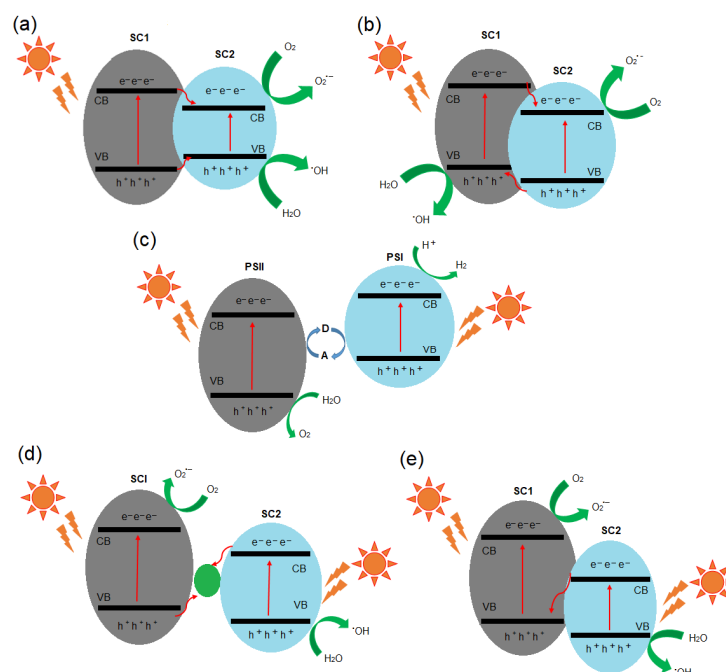


Figure 2. Schematic of various heterojunctions in water: (a) type-I heterojunctions, (b) type-II heterojunctions, and (c) Z-scheme heterojunctions (c–e). Reproduced from [65].

3. Porphyrin-Conjugated Metal Oxide Hybrid Nanomaterials

This section describes the conjugation of porphyrin molecules to the surfaces of various metal oxides, fabricating heterogeneous photocatalytic systems. The synthesis procedure and photocatalytic efficiency for the photodegradation of toxic pollutants are discussed, along with the advantages and disadvantages of these solid-supported porphyrin-conjugated heterojunctions.

3.1. Porphyrin–TiO₂ Hybrid Nanomaterials

Considering the importance of photodegradation reactions, titanium dioxide (TiO₂) and TiO₂-supported materials are widely utilized as catalysts for the photodegradation of hazardous compounds in wastewater [67]. This is primarily owing to its significant catalytic activity, low cost, high UV adsorption, biocompatibility, and excellent stability under extreme reaction conditions. However, the application of TiO₂-supported materials in photocatalytic processes faces many drawbacks. The main limitations are the relatively high degree of recombination of photogenerated electron–hole pairs, high fabrication costs, the need for a large amount of catalyst to achieve significant photodegradation rates, and low light-harvesting properties under cheap visible light. To overcome these drawbacks, porphyrin molecules can be integrated onto the surface of TiO₂ to construct heterogeneous photocatalytic systems for the photodegradation of toxic contaminants in water. The incorporation of porphyrin molecules on the TiO₂ surface lowers the band gap energy (E_g) due to the band alignment between the two materials. This also improves the visible-light harvesting capacity, stabilizes the photogenerated excited electrons, and delays the recombination of the photoinduced active species. This improved the photocatalytic photodegradation performance of the hybrid materials compared with that of either the free porphyrin or bare TiO₂ nanoparticles. Moreover, the entrapment of porphyrin on TiO₂ improves the stability of the catalyst during photodegradation to avoid secondary pollution and ensure complete recovery from the medium for reuse [68,69].

In 2010, Wang et al. constructed porphyrin-sensitized visible-light photocatalysts for the degradation of 4-nitrophenol [70]. Several porphyrin derivatives were sensitized to TiO₂ nanoparticles using a simple absorption method in chloroform. The integration of porphyrin onto the surface of TiO₂ nanoparticles lowered the band gap energy and improved the visible-light harvesting properties. Sensitized porphyrin-integrated TiO₂ photocatalysts exhibited higher catalytic photodegradation performance than TiO₂ nanoparticles. The photocatalytic performance depends not only on the amount of porphyrin incorporated in the hybrid materials but also on the substituent on the porphyrin core (Figure 3).

Langford et al. (Figure 4) observed a substantial improvement in the photocatalytic reaction of CuTCPP/TiO₂/cotton only in 60 min [71]. Using an adsorption method, CuTCPP (meso-tetra(4-carboxyphenyl)porphyrinato copper(II)) was linked to anatase TiO₂ nanoparticles. Within 180 min of visible-light irradiation, CuTCPP/TiO₂-coated cotton exhibited higher photodegradation performance of MB dye in water than free porphyrin CuTCPP or TiO₂-coated samples. The degradation ratios were 99% for CuTCPP/TiO₂-coated cotton, 32% for the TiO₂-coated sample, and 28% for CuTCPP. A simple mechanism for the enhanced photodegradation reaction was proposed by the authors (Figure 4). After irradiation with visible light, the porphyrin molecules are excited to generate a porphyrin singlet state [Pp]*. This excited porphyrin single-state [Pp]* is converted into the porphyrin triplet-excited state 3[Pp]* via intersystem crossing. Subsequently, the excited electrons from both the singlet and triplet states move to the CB of the TiO₂ NPs. Afterwards, the excited electrons react with adsorbed O₂ to form a highly reactive superoxide radical anion (O₂^{•−}) and facilitate the decomposition of MB dye.

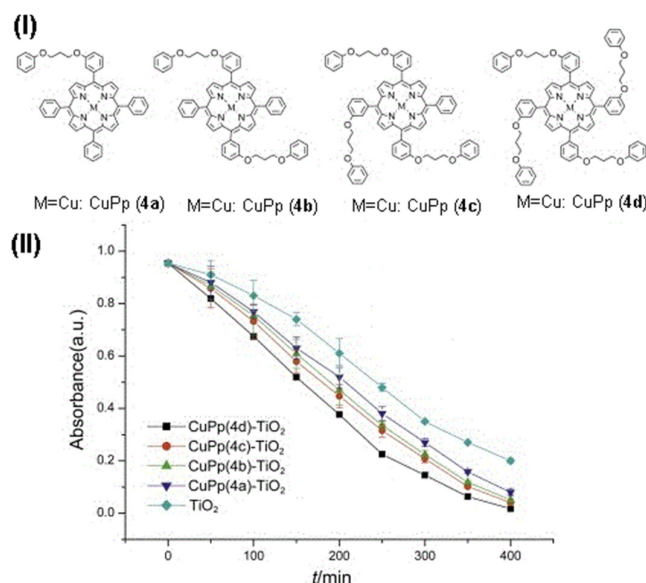


Figure 3. Molecular structure of porphyrin derivatives for the construction of hybrid photocatalysts (I). Photocatalytic degradation of 4-nitrophenol (II). Adapted from [70].

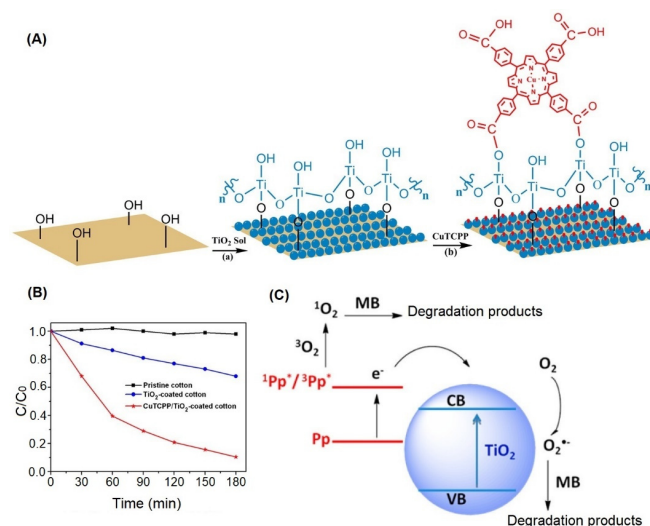


Figure 4. Fabrication of CuTCPP/TiO₂-coated cotton (A). Visible-light catalytic degradation of MB dye in H₂O (B). Photodegradation mechanism (C). Adapted from [71].

In 2015, Yao et al. (Figure 5) showed a significant degradation of MB dye using FeTCPP-SSA-TiO₂ in 60 min compared to the porphyrin-TiO₂ [72]. In this study, salicylic acid (SA) or 2-hydroxy-5-sulfosalicylic acid (SSA) was used as an anchoring ligand between Fe(III)porphyrin and TiO₂ particles (Figure 5). The photocatalytic MB dye degradation performance was found to be 99.3% for FeTCPP-SSA-TiO₂, 85.2% for FeTCPP-SA-TiO₂, and 30.7% for bare TiO₂. Therefore, bridging the Fe(III)porphyrin with TiO₂ not only increases the catalytic degradation activity but also improves the surface area and prevents the dissociation of the constituent molecules during degradation experiments. Photocatalytic activity is directly linked to the nature of the bridging ligands between the porphyrin and TiO₂ particles.

Chemically modified tetra-(para-amino)-phenylporphyrin (TPAPP)/TiO₂ hybrid nanocomposite materials were fabricated for the photocatalytic decomposition of Acid Black 1 dye under direct sunlight [73]. Glycidoxypyrroltrimethoxysilane was used as the coupling agent between 5,10,15,20-meso-TPAPP and TiO₂ nanoparticles. From the diffuse reflectance spectra (DRS), the observed band gap energy of bare TiO₂ was 3.33 eV, and that

of 0.004 wt% TPAPP/TiO₂ was 3.32 eV. The integration of porphyrin molecules onto the surface of TiO₂ in the presence of coupling agents not only increased the photocatalytic efficiency for the removal of Acid Black 1 dye but also improved the stability of the composite nanomaterials during the photodegradation experiments. Within 90 min of direct sunlight irradiation, the TPAPP/TiO₂ nanocomposite degraded 91.3% of the Acid Black 1 dye in water (Figure 6).

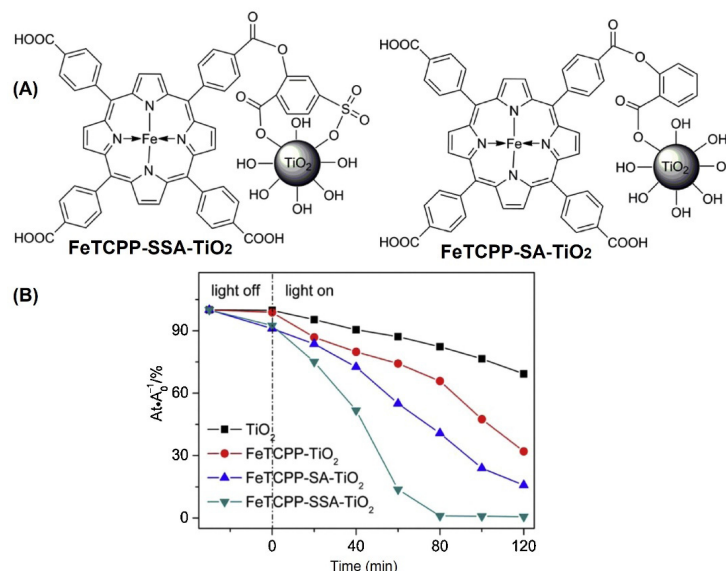


Figure 5. Molecular structure of porphyrin-based hybrid materials (A). Catalytic decomposition of MB dye in H₂O (B). Adapted from [72].

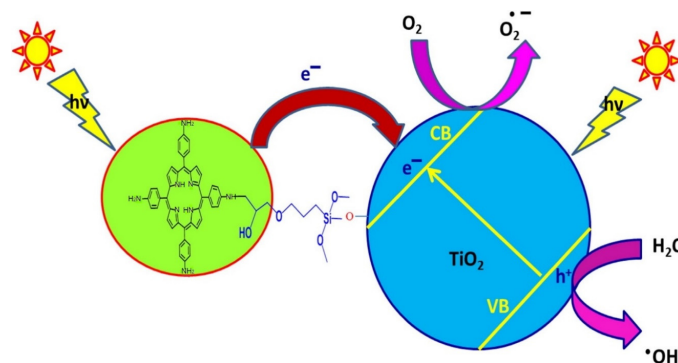


Figure 6. Photocatalytic photodegradation of Acid Black 1 dye in water using TPAPP/TiO₂ hybrid nanocomposite materials. Adapted from [73].

Hu et al. studied an organic–inorganic hybrid material for the catalytic photodegradation of 4-nitrophenol (4-NP) in water [74]. Several Cu(II)porphyrin derivatives (CuPp 1a–1c) were successfully immobilized on the synthesized TiO₂ (p-TiO₂) and commercial TiO₂ (c-TiO₂) (Figure 7). Within 60 min, the photodegradation ratio of 4-NP was 85% for c-TiO₂ compared with p-TiO₂ under metal halide lamp irradiation. Interestingly, cTiO₂@CuPp (1c) exhibited better results than pTiO₂@CuPp (1c). The hydroxylated surface of c-TiO₂ likely affects the interaction of the sensitizer with TiO₂, unlike the other CuPp derivatives.

A composite photocatalyst, TP-222(Zn), was constructed for the photocatalytic photodegradation of RhB dye in water under visible-light irradiation [75]. In a typical procedure, TiO₂ nanoparticles were axially coupled to the Zn²⁺ metal center in PCN-222(Zn) using 4-mercaptopyridine (4-PySH). From the DRS analysis, E_g was found to be 3.2 eV for the bare TiO₂ nanoparticles and 1.74 eV for PCN-222(Zn). The conjugation of the porphyrin framework to TP-222(Zn) significantly lowered the bandgap energy and enhanced

the visible-light harvesting capacity. The aromatic conjugated frameworks in TP-222(Zn) facilitated electron transfer from PCN-222(Zn) to the TiO₂ nanoparticles, stabilized the photogenerated charge carriers, enhanced the recombination time, and boosted the photocatalytic degradation activity. Within 270 min, the first-order degradation rate constant was found to be 0.01239 min⁻¹. Additionally, the incorporation of TiO₂ nanoparticles into TP-222(Zn) produced a large number of active sites, improving the interaction between the substrate molecules and enhancing the photodegradation performance (Figure 8).

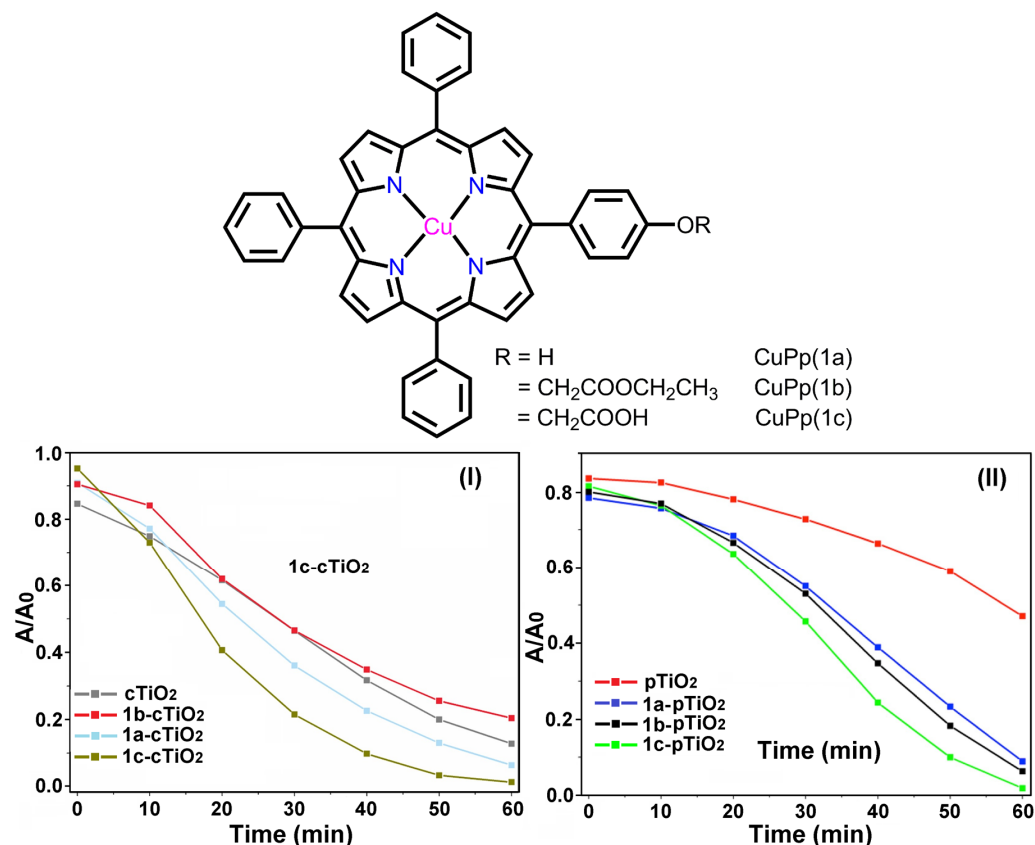


Figure 7. TiO₂@CuPp composite material. Chemical structure of CuPp (1a–1c). Photocatalytic degradation of 4-NP by using cTiO₂ series (I) and p- cTiO₂ series (II). Adapted from [74].

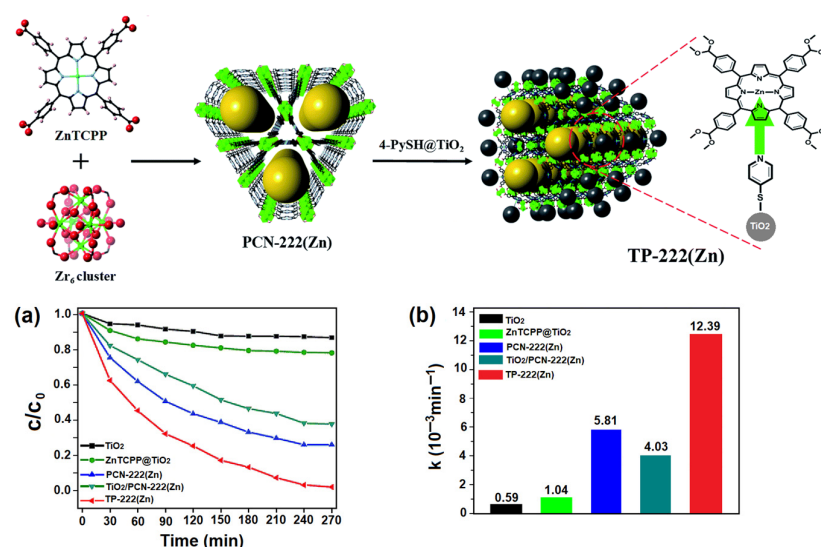


Figure 8. Schematic of TP-222(Zn) fabrication. (a) Visible-light photodegradation of RhB dye. (b) Rate constant of RhB dye. Adapted from [75].

In 2019, Min et al. investigated various porphyrin-integrated TiO_2 composite photocatalysts for MB degradation in water. Various hybrid materials have been fabricated by reacting anatase TiO_2 with carboxyphenyl derivatives of porphyrins [76]. Effective binding between TiO_2 nanoparticles and the carboxyphenyl derivatives of porphyrins reduced the band gap energy and improved the visible-light harvesting activity. All composite catalysts showed higher catalytic photodegradation activity than pure anatase TiO_2 under visible-light irradiation. Within 60 min of irradiation, the photodegradation efficiency of the MB dye was 95%, which was better than that of pure TiO_2 (55%). Moreover, the authors observed that Zn(II)-metalated porphyrin-based hybrid materials exhibited better photocatalytic activity than the metal-free porphyrin-based hybrid materials. The high electronic coupling between the Zn(II)porphyrins and the CB of TiO_2 leads to the crucial separation of the photogenerated charged species, thereby delaying recombination of the photogenerated reactive species. Surface modification via strong chemical bonding between TiO_2 nanoparticles and carboxyphenyl porphyrin not only improved the photocatalytic activity but also prevented the separation of porphyrin molecules from the TiO_2 surface (Figure 9).

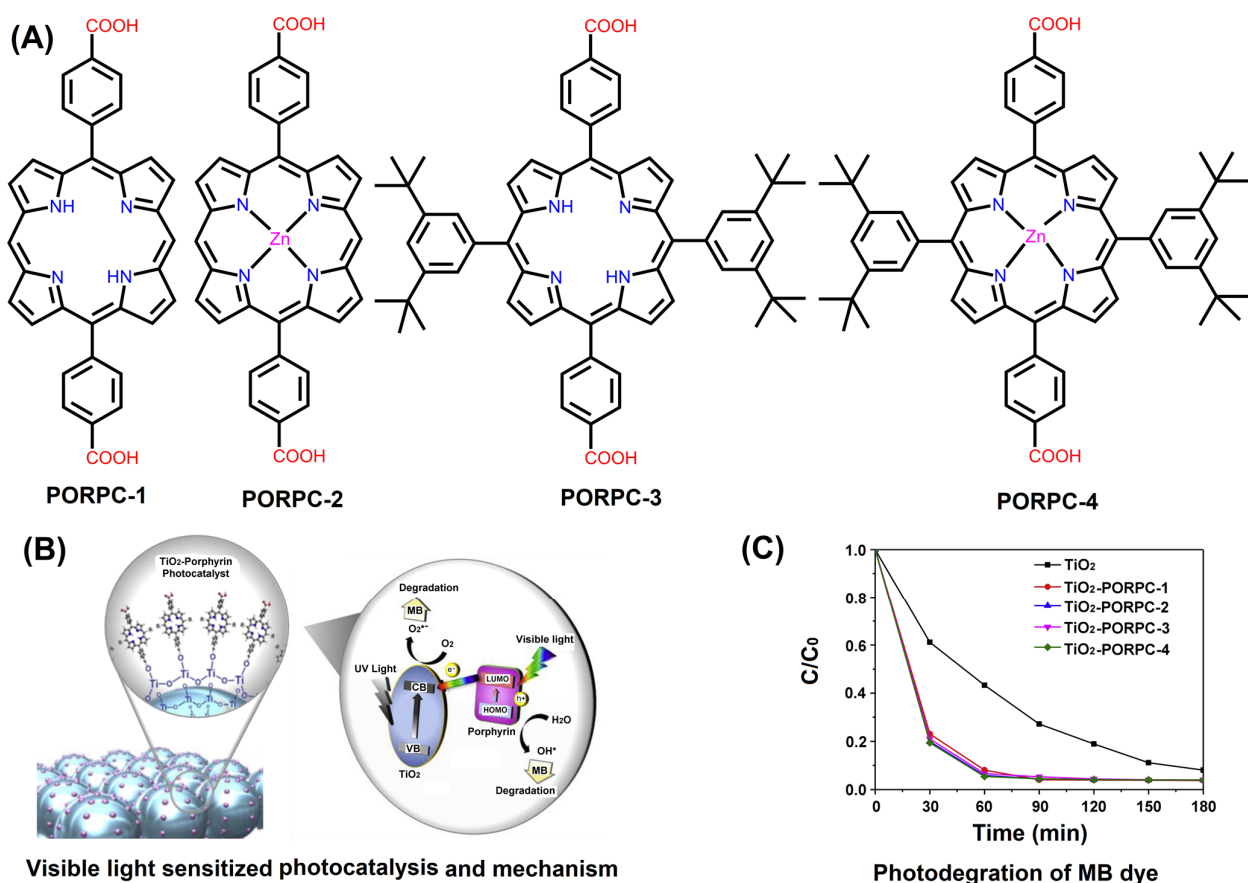


Figure 9. Molecular structure of several porphyrins (A). Visible-light photodegradation mechanism (B). Photodegradation of MB dye in water (C). Adapted from [76].

A Zr-porphyrin-based metal–organic framework (Zr-MOF) containing TiO_2 (PCN-224@ TiO_2) hybrid composite was fabricated for the catalytic photodecomposition of MB dye in water [77]. The solvothermal reaction of ZrCl_4 , H_2TCPP , and the TiO_2 NPs led to the construction of a hybrid material, PCN-224@ TiO_2 . The incorporation of porphyrin frameworks onto the surface of TiO_2 reduces the band gap energy and improves the solar-light harvesting properties. Aromatic conjugated systems stabilize the photogenerated charged species, increase the recombination time, and improve the photocatalytic properties.

Under Xe lamp irradiation, the MB dye degradation ratio was found to be 93.2% for PCN-224@TiO₂. Moreover, the construction of a robust porphyrin framework onto the surface of TiO₂ not only improved the photodegradation activities compared with those of PCN-224 or TiO₂ but also prevented hydrolysis during the photodegradation reactions (Figure 10).

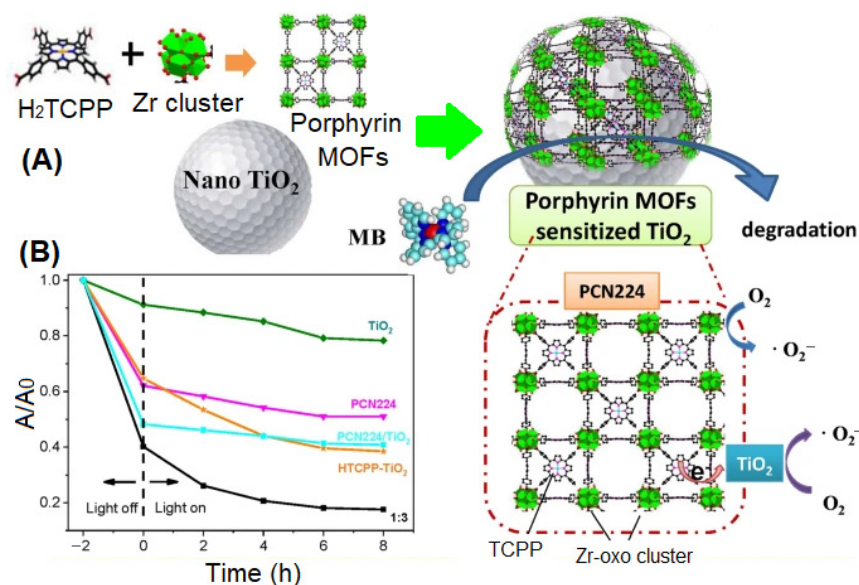


Figure 10. Construction of Zr-MOF-sensitized TiO₂ (PCN-224@TiO₂) hybrid materials (A). Photocatalytic photodegradation of MB dye (B). Adapted from [77].

Two organic–inorganic hybrid photocatalysts (SnP/AA@TiO₂ or SnP@TiO₂) were developed for the visible-light photocatalytic decomposition of RhB dye [78]. The reaction of (trans-dihydroxo)(5,10,15,20-tetraphenylporphyrinato)tin(IV) with anatase TiO₂, with and without adipic acid (AA), led to the formation of SnP/AA@TiO₂ and SnP@TiO₂, respectively. From the Tauc plot, E_g was calculated to be 2.46 eV for SnP/AA@TiO₂ and 3.12 eV for bare TiO₂. The integration of tin porphyrin onto the surface of TiO₂ not only altered E_g but also enhanced the visible-light harvesting capacity (Figure 11). Within 80 min of irradiation, the RhB dye photodegradation rate constants were found to be 0.0366, 0.0078, 0.0060, and 0.0016 min^{−1} for SnP/AA@TiO₂ (0.0366 min^{−1}), SnP@TiO₂ (0.0078 min^{−1}), TiO₂ (0.0060 min^{−1}), and SnP (0.0016 min^{−1}). Mechanistic investigation indicated that the synergistic effect between TiO₂ and SnP via bridging adipic acid was responsible for the significant increase in the photocatalytic performance of SnP/AA@TiO₂ compared with that of SnP@TiO₂. Additionally, the anchoring ligands firmly prevented the isolation of tin porphyrin from the surface of TiO₂ during the catalytic photodegradation reactions, thereby improving catalytic stability. After 10 consecutive cycles, SnP/AA@TiO₂ still maintained a high degradation activity, with only a 3% reduction, indicating that the SnP/AA@TiO₂ possesses remarkable stability. The structure of SnP/AA@TiO₂ was examined after the degradation reaction to confirm its stability. FE-SEM (Field Emission Scanning Electron Microscopy), PXRD (Powder X-Ray Diffraction), and FT-IR (Fourier Transform Infrared Spectroscopy) spectra of the used SnP/AA@TiO₂ were similar to the fresh sample, indicating that the framework of this catalyst remained intact during the degradation reaction.

Zhao et al. also investigated two porphyrin-sensitized TiO₂ materials (Cb-Ph-ZnP/TiO₂ and TPA-BiPh-ZnP/TiO₂) for the solar-light photodegradation of AB1 [79]. The integration of porphyrin onto the surface of TiO₂ lowered the band gap energy (approximately 1.94 eV for composite catalysts and 3.28 eV for bare TiO₂) and also improved the light-harvesting efficiency in the visible-light range. Within 60 min, the AB1 degradation

ratio was 90% for TPA-BiPh-ZnP/TiO₂ and 80% for Cb-Ph-ZnP/TiO₂. The interfacial charge transfer, separation, and electron delocalization are likely more facile in TPA-BiPh-ZnP/TiO₂ than in Cb-Ph-ZnP/TiO₂. Mechanistic investigations indicated that the superoxide holes and radicals were the most reactive components in the catalytic photoreduction of AB1 in water (Figure 12).

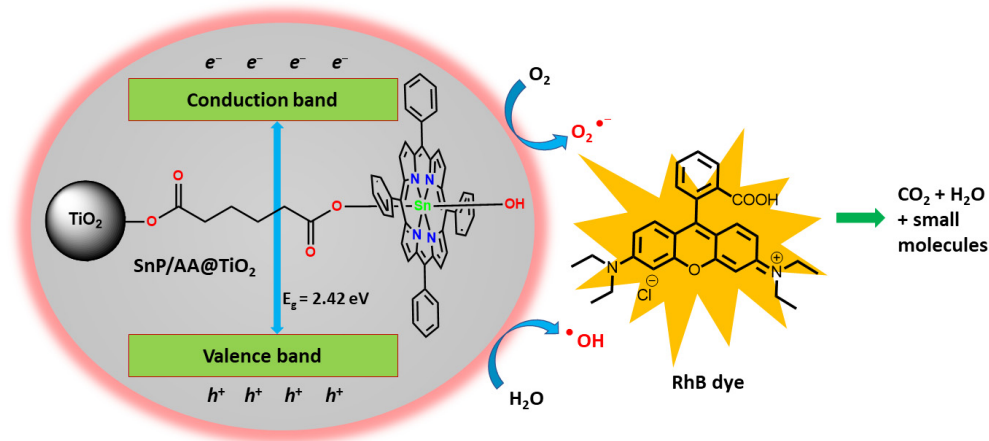


Figure 11. Schematic of visible-light catalytic photodegradation of RhB using SnP/AA@TiO₂ hybrid nanostructures. Adapted from [78].

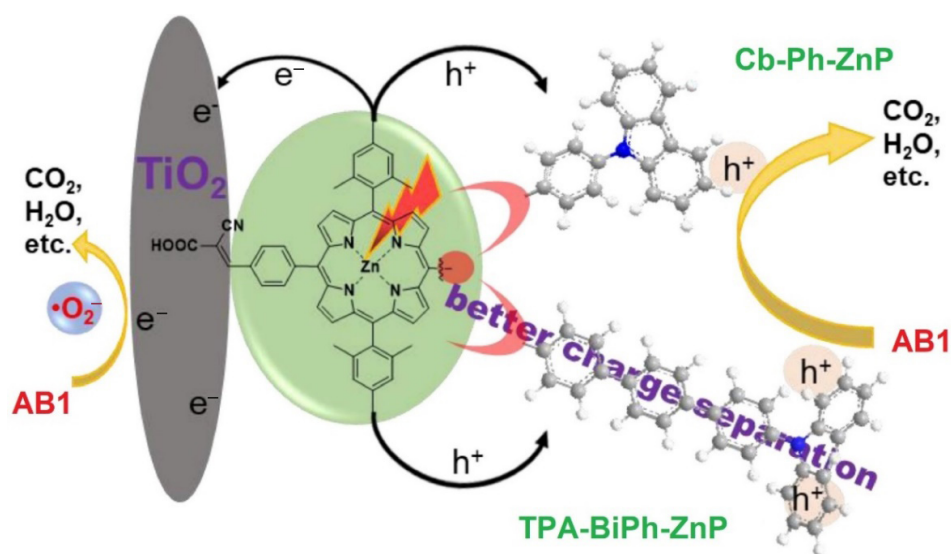


Figure 12. Diagram showing two porphyrin-sensitized TiO₂ photocatalysts for the photodegradation of pollutant AB1 in water. Adapted from [79].

An organic–inorganic composite catalyst (2D CuMOF-Ti) was studied for the decomposition of hazardous pollutants in wastewater [80]. The solvothermal reaction between H₂TCPP and Cu(NO₃)₂ in the presence of a polyvinylpyrrolidone surfactant led to the fabrication of 2D CuTCPP MOF nanosheets. Subsequently, a solvothermal reaction between tetrabutyl titanate and 2D CuTCPP MOFs led to the fabrication of the desired hybrid photocatalyst (2D CuMOF-Ti). The bandgap energy for the 2D CuTCPP MOFs was found to be 2.81 eV and is lower than 3.5 eV for TiO₂. The immobilization of TiO₂ on the core of the CuTCPP MOFs reduced the band gap energy and improved the light-harvesting properties in the visible range. Moreover, the conjugated porphyrin system facilitates the separation, transportation, and stabilization of photogenerated electron–hole pairs. This delays the recombination time and hence increases the photocatalytic decomposition activity. The

composite catalyst 2D CuMOF-Ti exhibited significant photodegradation of toxic Cr(IV) and RhB dyes in water under solar-light exposure. The photodegradation rate of RhB dye for 2D CuMOF-Ti is found to be 12.6 and 2.8 times better than that of 2D CuTCPP MOFs and TiO_2 , respectively. Additionally, the robust framework resists hydrolysis of the catalyst during the photodegradation reaction (Figure 13).

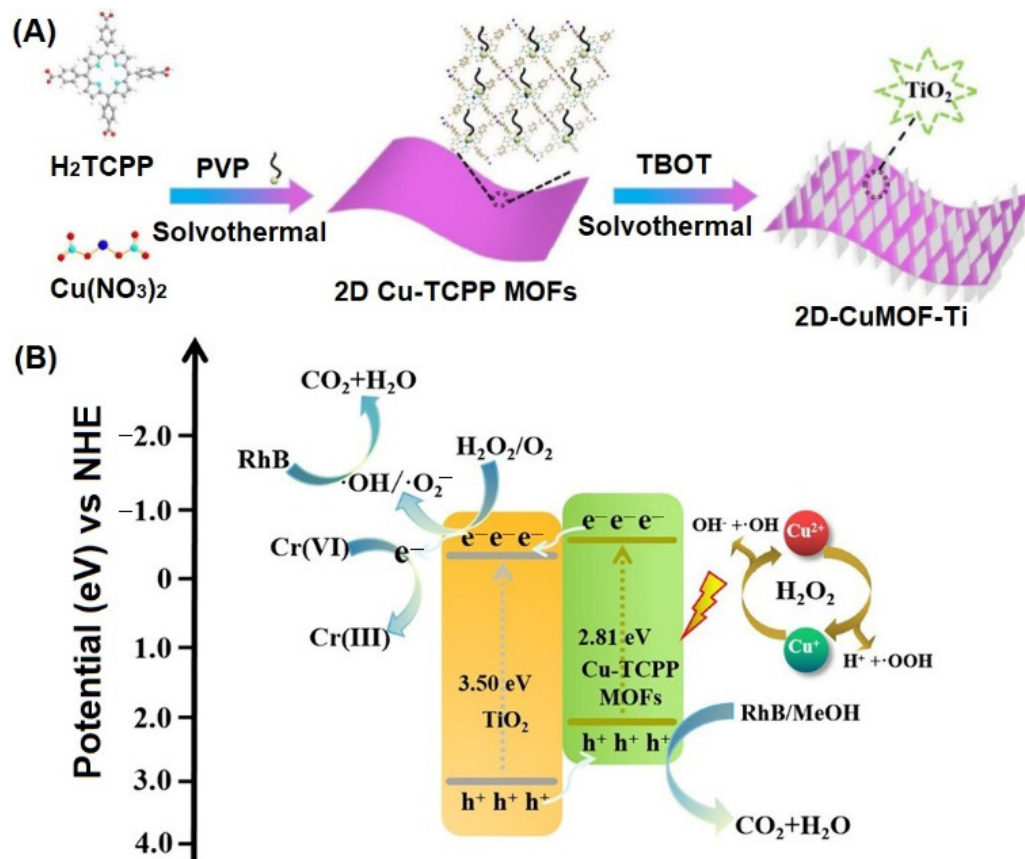


Figure 13. Fabrication of 2D CuMOF-Ti hybrid photocatalyst (A). Photodegradation mechanism of RhB dye and Cr(VI) ions in water (B). Adapted from [80].

Several porphyrin-sensitized TiO_2 materials were recently fabricated for the photocatalytic decomposition of MB [81]. The reaction of numerous metal salts of the tetra(4-carboxylphenyl)porphyrin MTCPP ($M = \text{Fe(III)}$, Zn(II) , and Co(III)) with TiO_2 (P25) in ethanol led to the formation of hybrid photocatalysts MTCPP@TiO_2 . From the DRS plot, E_g for FeTCPP@TiO_2 was found to be lower than that of TiO_2 (2.43 eV vs. 3.2 eV). The integration of metal porphyrins onto the surface of TiO_2 not only alters E_g but also improves the visible-light harvesting capacity. Moreover, electronic delocalization over the conjugated framework assists charge transport and separation, thereby delaying the recombination of excited holes and electrons. This enhanced the photodegradation performance of MTCPP@TiO_2 compared with that of either MTCPP or TiO_2 . Among the MTCPP@TiO_2 photocatalysts, FeTCPP@TiO_2 exhibited the highest photodegradation performance for MB dye degradation in water. Therefore, the nature of the metals in the metalloporphyrin controls the photocatalytic performance of MTCPP@TiO_2 (Figure 14).

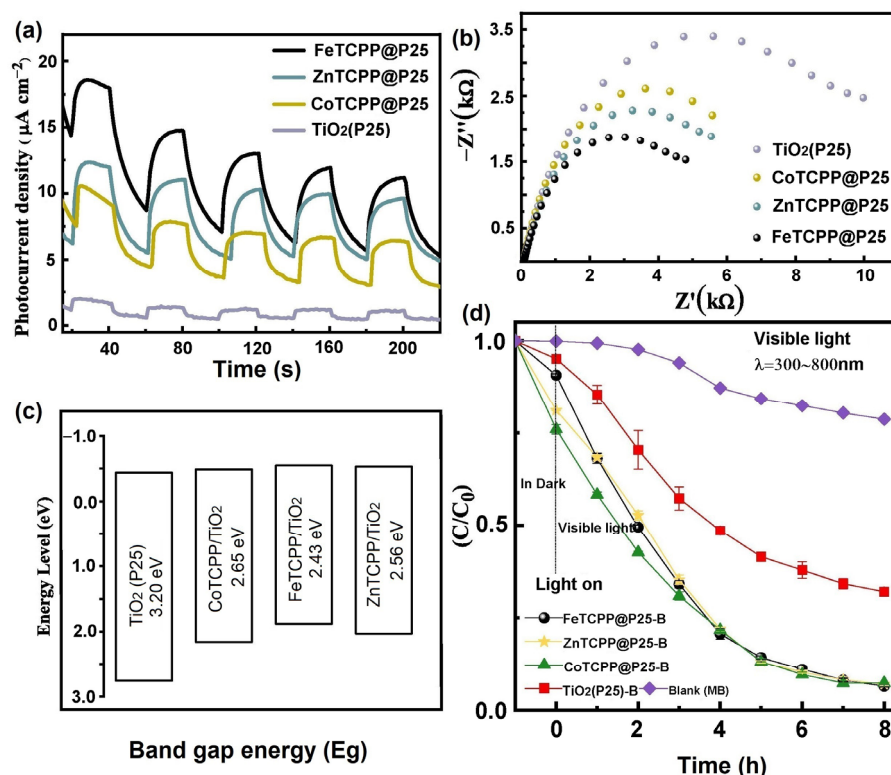


Figure 14. MTCPP@TiO₂ photocatalysts (where M = Zn(II), Co(III), and Fe(III)). (a) Transient photocurrent response, (b) EIS Nyquist plots, (c) band energy diagram, and (d) photodegradation of MB dye. Adapted from [81].

3.2. Porphyrin–ZnO Hybrid Nanomaterials

Aside from TiO₂, ZnO and ZnO-based catalysts have been extensively utilized for the catalytic decomposition of toxic contaminants in wastewater, owing to their high chemical stability, low cost, good photosensitivity, low toxicity, and significant photocatalytic activity [82]. However, their high bandgap energies prevent them from participating in photocatalytic reactions under visible-light irradiation. Moreover, the quick recombination rate of the photogenerated pairs and low recovery are disadvantages. To overcome these obstacles, researchers have developed strategies for incorporating porphyrins onto the surface of ZnO nanostructures.

In 2013, Zhang et al. constructed an organic–inorganic composite photocatalytic system for the catalytic photodegradation of RhB dye in H₂O under solar-light exposure [83]. The reaction of mono-[4-(2-ethyl-p-hydroxybenzoate)ethoxyl]-10,15,20-tri(phenyl)porphyrin copper(II) (CuPp) with ZnO via a mixing method led to the formation of a CuPp–ZnO hybrid material. Scanning electron microscopy (SEM) images of CuPp–ZnO confirmed that CuPp was successfully incorporated onto the surface of the ZnO NPs. Within 210 min, the RhB degradation ratios were 63% and 23% for CuPp–ZnO and ZnO, respectively. The incorporation of porphyrin onto the surface of ZnO enhanced the visible-light harvesting activity and increased the recombination time by stabilizing the excited electrons over the aromatic conjugated metalloporphyrin CuPp. However, the authors observed a reduction in photocatalytic activity during the recyclability test of CuPp–ZnO (Figure 15).

TPAPP/SiO₂–ZnO hybrid materials were investigated for the photodegradation of toxic compounds in H₂O under visible-light irradiation [84]. The authors demonstrated the chemical modification of mixed inorganic oxides (70% SiO₂ and 30% ZnO) with TPAPP, using 3-glycidoxypolytrimethoxysilane as the coupling agent. The DRS plot indicated E_g

values of 3.23 eV and 3.30 eV for TPAPP/SiO₂-ZnO and ZnO, respectively. Within 180 min, the photodecomposition ratio of naphthol blue black in H₂O reached 60% under visible-light exposure. Chemical modification not only prevents catalyst hydrolysis during the photodegradation reaction but also allows the catalyst to be used multiple times without degrading its performance (Figure 16).

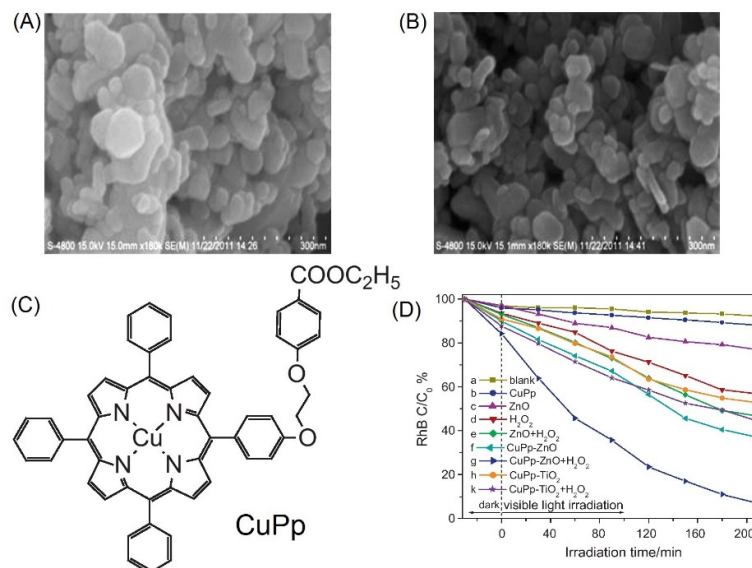


Figure 15. Cu(II)porphyrin@ZnO hybrid nanocomposite materials. SEM images of ZnO (A) and CuPp-ZnO (B). Chemical structure of CuPp (C). Visible-light catalytic degradation of RhB dye (D). Adapted from [83].

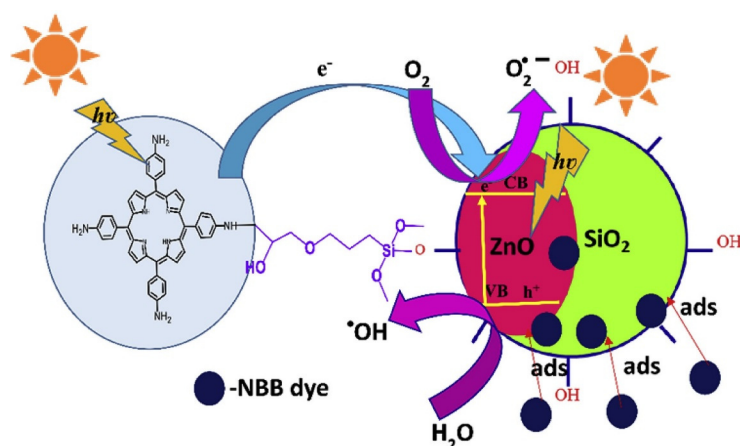


Figure 16. Visible-light decomposition of naphthol blue black dye using porphyrin/SiO₂-ZnO hybrid nanocomposite materials in H₂O. Adapted from [84].

In 2021, Vo et al. reported enhanced photocatalytic degradation performance of H₂TCPP@ZnO nanofibers [85]. A self-assembly approach was used to fabricate the H₂TCPP@ZnO nanofibers. The SEM images of H₂TCPP@ZnO reveal that the ZnO NPs were uniformly dispersed on the self-assembled H₂TCPP nanofiber matrix. From the DRS plot, E_g for H₂TCPP@ZnO and ZnO were found to be 2.43 eV and 3.21 eV, respectively. The incorporation of porphyrin onto the surface of ZnO not only lowered the band gap energy of H₂TCPP@ZnO (3.12 and 2.43 eV) compared with the free-standing H₂TCPP nanofibers (2.6 eV) and ZnO nanostructures (3.21 eV), but also improved the visible-light harvesting activity. Within 180 min of visible-light irradiation, the ZnO@H₂TCPP nanofibers showed significant catalytic activity for the photodecomposition of the RhB dye. The high pho-

todegradation rate constant (0.02 min^{-1}) of the $\text{ZnO}@H_2\text{TCPP}$ nanofiber is attributed to the synergistic activity between ZnO NPs and $H_2\text{TCPP}$ nanofibers (Figure 17).

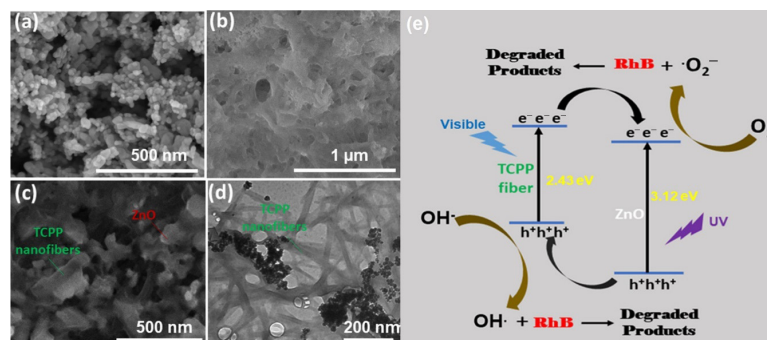


Figure 17. SEM images of (a) ZnO NPs, (b) $H_2\text{TCPP}$ nanofiber, and (c) $\text{ZnO}/H_2\text{TCPP}$ nanofiber. (d) TEM images of $\text{ZnO}/H_2\text{TCPP}$ nanofiber. (e) Degradation mechanism of RhB dye. Adapted from [85].

Organic–inorganic hybrid photocatalysts ($\text{SnP}@ZnO$ and $\text{SnP}/AA@ZnO$) were investigated for the solar-light-induced photodegradation of amaranth dye [86]. The reaction of trans-dihydroxo[5,10,15,20-tetrakis(4-pyridyl)porphyrinato]tin(IV) with ZnO, with and without AA pretreatment, led to the production of $\text{SnP}/AA@ZnO$ and $\text{SnP}@ZnO$, respectively. From the Tauc plot, the E_g values of $\text{SnP}/AA@ZnO$ and the ZnO NPs were calculated to be 2.45 eV and 3.24 eV, respectively. The integration of SnP onto the surface of the ZnO NPs not only altered the morphology but also enhanced the visible-light photocatalytic activity (Figure 18). Within 90 min of irradiation, the degradation rate constants were found to be 0.048 min^{-1} ($\text{SnP}/AA@ZnO$), 0.008 min^{-1} ($\text{SnP}@ZnO$), 0.006 min^{-1} (ZnO), and 0.001 min^{-1} (SnP). The synergistic effect between the ZnO NPs and tin porphyrin via the bridging AA group was responsible for the significantly enhanced catalytic photodegradation activity of $\text{SnP}/AA@ZnO$ over that of $\text{SnP}@ZnO$. Moreover, the anchoring effect of the bridging ligands promptly restrained ZnO from interacting with SnP during photocatalytic reactions. After 10 consecutive cycles, $\text{SnP}/AA@ZnO$ maintained a high efficiency for the degradation of AM dye with a reduction of only 5%, which indicates that the $\text{SnP}/AA@ZnO$ exhibited a remarkable stability.

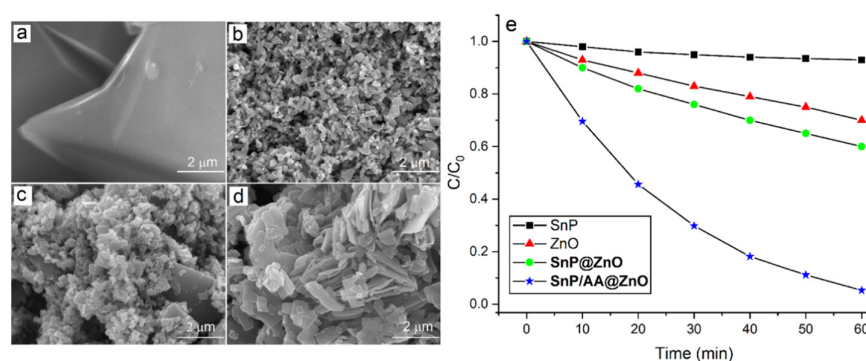


Figure 18. Field-emission scanning electron microscope images of (a) SnP; (b) ZnO; (c) $\text{SnP}@ZnO$; and (d) $\text{SnP}/AA@ZnO$. Catalytic photodegradation of AM dye (e). Adapted from [86].

3.3. Other Porphyrin-Conjugated Metal Oxide Hybrid Nanomaterials

In addition to TiO_2 and ZnO, porphyrin molecules can be immobilized on other inorganic oxides to fabricate hybrid solar-light photocatalysts for the photodegradation of toxic compounds in H_2O .

Two Zr porphyrin-based magnetic visible-light photocatalysts, $\text{Fe}_3\text{O}_4@\text{SiO}_2@\text{PCN-222}(\text{Fe})$ and $\text{Fe}_3\text{O}_4@\text{SiO}_2@\text{PCN-222}$, were synthesized and investigated for their photodegradation of toxic dyes in H_2O [87]. E_g for Fe_3O_4 was found to be 1.53 eV and 1.9 eV for $\text{PCN-222}(\text{Fe})$. The incorporation of porphyrin ligands into these photocatalysts reduced the bandgap energy and improved the solar-light harvesting properties. Moreover, the aromatic conjugated porphyrin framework stabilizes the photogenerated species and increases the recombination time. This improved the photocatalytic activity compared with that of $\text{Fe}_3\text{O}_4@\text{SiO}_2$. Under visible-light irradiation, this photocatalyst exhibited significant photocatalytic activity for the degradation of EryB, RhB, and Rose Bengal dyes. Mechanistic experiments confirmed that the highly reactive superoxide radical anion ($\text{O}_2^{\cdot-}$) is the principal reactive species involved in dye photodegradation. Owing to the magnetic properties of these photocatalysts, separating them after the degradation reactions is simple, requiring only a normal magnet (Figure 19).

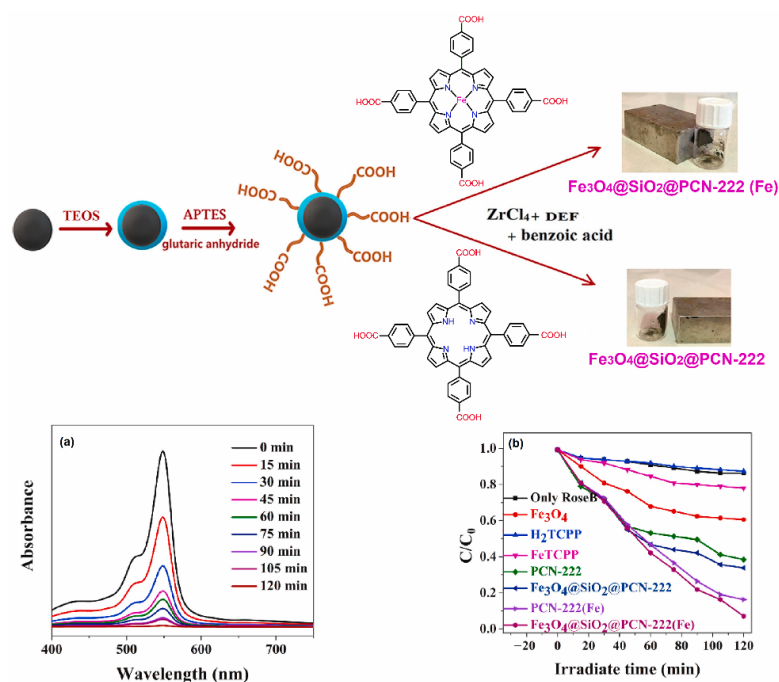


Figure 19. Fabrication of hybrid composite $\text{Fe}_3\text{O}_4@\text{SiO}_2@\text{PCN-222}(\text{Fe})$. Photodegradation of RB dye under solar-light exposure: (a) UV–Vis spectra and (b) photodegradation kinetics. Adapted from [87].

In 2021, Castro et al. reported the synthesis of a porphyrin-based nanomagnetic hybrid photocatalyst for methyl orange (MO) degradation in water [88]. The reaction of $\text{Zn}(\text{II})$ porphyrin with silica-coated Fe_3O_4 led to the formation of a nanomagnetic porphyrin hybrid. The incorporation of porphyrin into the core of the silica nanoparticles not only enhanced the photodegradation activity but also prevented the separation of porphyrin from the surface of the hybrid catalyst during the degradation reaction. Within 270 min of visible-light irradiation, the MO dye degradation ratio of the hybrid photocatalyst reached 99% in the presence of H_2O_2 (hydrogen peroxide). Additionally, the catalyst could be recovered from the reaction chamber using a simple magnetic bar (Figure 20).

Nguyen et al. also demonstrated the construction of porphyrin-based hybrid materials for visible-light photodegradation of pollutant dyes [89]. The acid–base neutralization self-assembly of H_2TCPP in the presence of the $\text{graphene}@\text{Fe}_2\text{O}_3\text{-TiO}_2$ nanohybrid led to the production of $\text{graphene}@\text{Fe}_2\text{O}_3\text{-TiO}_2@\text{porphyrin}$ composite materials. From the DRS plot, the E_g values for $\text{Fe}_2\text{O}_3\text{-TiO}_2$ and TiO_2 were determined to be 2.83 eV and 3.2 eV, respectively. The integration of H_2TCPP onto the core of the $\text{graphene}@\text{Fe}_2\text{O}_3\text{-TiO}_2$ nanohybrid

increased the visible-light harvesting properties and enhanced the photocatalytic degradation activity. Within 100 min of visible-light irradiation, the photodegradation ratio of RhB was 90% for the graphene@Fe₂O₃-TiO₂@porphyrin composite in water (Figure 21).

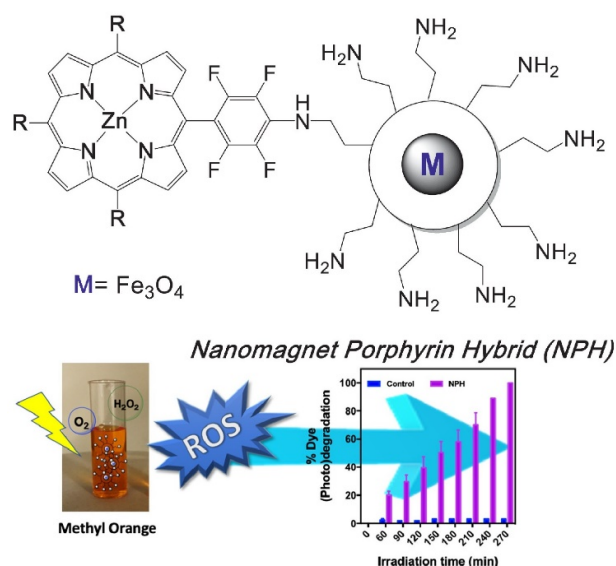


Figure 20. Schematic of silica-coated nanomagnet porphyrin hybrid (NPH) for MO dye degradation. Adapted from [88].

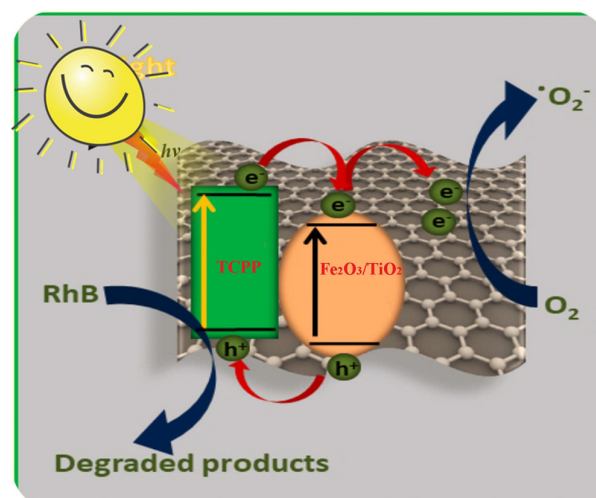


Figure 21. Diagram of graphene@Fe₂O₃-TiO₂@porphyrin composite materials for RhB dye degradation. Adapted from [89].

Hybrid nanocomposites (SnP@SiO₂ and SnP@MCM-41) were fabricated by the chemisorption of (*trans*-dihydroxo)(5,10,15,20-tetraphenylporphyrinato)tin(IV) (SnP) on SiO₂ and mesoporous MCM-41 (Mobil Composition of Matter No. 41) powder [90]. As determined from the Tauc plot, the E_g values for SnP, SnP@SiO₂, and SnP@MCM-41 were 2.85, 2.64, and 2.53 eV, respectively. The integration of tin porphyrin onto the surfaces of SiO₂ and MCM-41 not only altered the surface morphology but also enhanced the catalytic performance for the photodegradation of organic contaminants in water. Under solar-light exposure, these two hybrid nanocomposites exhibited significant catalytic photodegradation activities for cationic RhB, anionic erioglaucine, and neutral m-cresol purple in H₂O. The degradation activity of SnP@MCM-41 was higher than that of SnP@SiO₂. Moreover, SnP@MCM-41 exhibits excellent photostability compared to SnP@SiO₂ (Figure 22). After 10 consecutive cycles, SnP@MCM-41 still maintains a high performance for the degrada-

tion of the anionic erioglaucine dye with a removal of only 8%, which shows the remarkable stability of SnP@MCM-41. The FTIR spectrum and FE-SEM image of the used sample SnP@MCM-41 were closely resemble that of the fresh sample, which indicates that the mesoporous framework of this photocatalyst was intact during the photocatalytic reaction. Moreover, the process for the recovery of these catalysts from the reaction vessel was very simple through successive filtration, washing, and drying procedures.

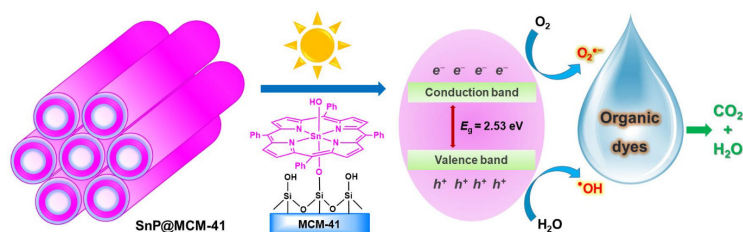


Figure 22. Schematic of the composite material SnP@MCM-41 for catalytic degradation of organic dyes in H₂O. Adapted from [90].

Recently, Zhu et al. reported the formation of several inorganic–organic hybrid photocatalysts for the photodegradation of 4-chlorophenol under visible-light irradiation [91]. Several porphyrins with various polar groups (–NO₂, –NH₂, –OH, and –COOH) have been integrated into BiOI to construct hybrid materials via impregnation. The incorporation of porphyrin onto the surface of BiOI not only increased the light-harvesting properties of the hybrid materials but also improved the photocatalytic activity. Within 60 min, all hybrid catalysts accomplished the degradation of >90% 4-chlorophenol. Interestingly, the photodegradation performance varies with the polarity and dipole moment of the porphyrin cores. The photodegradation performances of all the hybrid catalysts followed the order H₂TNPP/BiOI < H₂TAPP/BiOI < H₂THPP/BiOI < H₂TCPP/BiOI (Figure 23).

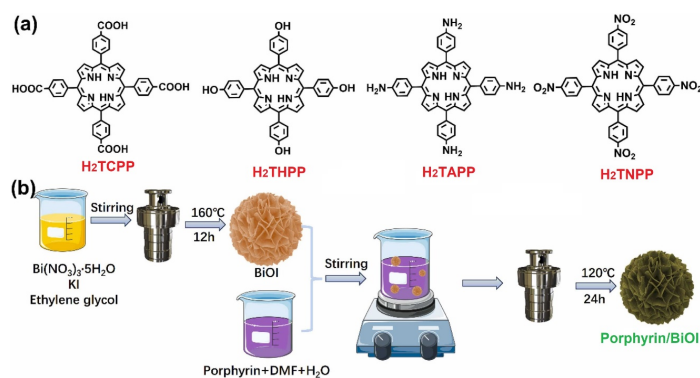


Figure 23. Chemical structures of several porphyrins (a); schematic of the synthesis of porphyrin-sensitized photocatalysts (b). Adapted from [91].

Alumina-supported porphyrin nanocomposite (SnTTP/SA@Al₂O₃) was utilized for MB degradation under exposure to solar light [92]. The reaction of SnTTP {trans-dihydroxo-[5,10,15,20-tetrakis(p-tolyl)porphyrinato]tin(IV)} with Al₂O₃ in the presence of dibasic succinic acid (SA) guided the fabrication of the hybrid nanocomposite SnTTP/SA@Al₂O₃. From the Tauc plot, the E_g values were found to be 3.95 eV for Al₂O₃, 2.96 eV for SnTTP, 2.85 eV for SnTTP@Al₂O₃, and 2.48 eV for SnTTP/SA@Al₂O₃. The incorporation of SnTTP onto the surface of alumina via the bridging anchoring ligand (SA) in SnTTP/SA@Al₂O₃ lowered E_g but also increased the light-harvesting properties compared with SnTTP. Within 90 min, the MB dye degradation rate constant was found to be 0.0406 min^{−1} for SnTTP/SA@Al₂O₃ and 0.0024 min^{−1} for SnTTP. Moreover, the strong attachment between SnTTP and Al₂O₃ via SA not only prevented SnTTP hydrolysis during the photodegrada-

tion reaction in water but also increased the amount of SnTTP on the surface of alumina, facilitating the transfer of electrons from the excited SnTTP molecules to the CB of alumina (Figure 24). Radical trapping experiments have been performed to determine the details of the photogenerated reactive species during the photodegradation process. Various scavengers (tert-butanol, sodium azide, para-benzoquinone, ethylenediaminetetraacetic acid disodium: Na₂-EDTA) were employed during the degradation reaction of MB. It was observed that the decay rate of MB was significantly reduced in the presence of either Na₂-EDTA, *tert*-butanol, or *para*-benzoquinone. The degree of reduction rate is higher in the presence of Na₂-EDTA compared to *tert*-butanol or *para*-benzoquinone. These results confirmed that photogenerated h⁺ is the main reactive species compared to either OH[•] or O₂^{•-}. However, the MB decay was unaffected by the presence of sodium azide. This indicated that singlet oxygen had no effect on the degradation of MB in water in this system [92].

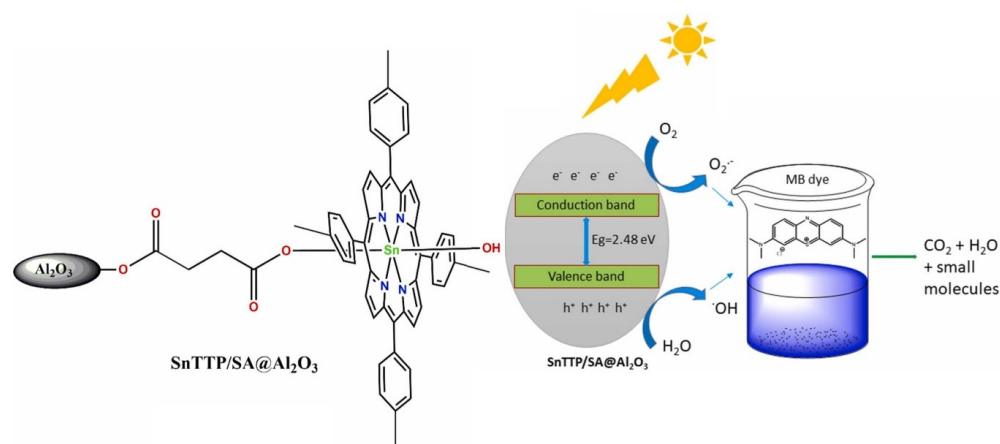


Figure 24. SnTTP/SA@Al₂O₃ hybrid materials for catalytic photodegradation of MB dye. Adapted from [92].

4. Porphyrin-Conjugated Metal Plasmon Nanoparticles

In addition to metal oxides or inorganic oxides, the fabrication of metal plasmons on porphyrin or porphyrin-based frameworks improves the photocatalytic activity.

Yang et al. fabricated a porphyrin-containing hybrid material (Ag@MOF-525) for the catalytic photodecomposition of dyes [93]. Under visible-light exposure, the photoreduction of AgNO₃ on the surface of a Zr-based porphyrin metal–organic framework (MOF-525) led to the fabrication of the hybrid photocatalyst Ag@MOF-525. The integration of Ag plasmon (Ag NPs) onto the core of MOF-525 improved not only the visible-light harvesting efficiency but also electron transportation, separation, and delay of the recombination rate of the photogenerated reactive species. The RhB photodegradation rate of Ag@MOF-525 was 91% within 60 min. By contrast, the photodegradation ratio of TC for Ag@MOF-525 was 200 min. The degradation efficiency of Ag@MOF-525 was higher than that of either H₂TCPP or MOF-525 (Figure 25). Radical trapping experiments advocated that h⁺ and O₂^{•-} are the dominant reactive species produced during the degradation reactions.

In addition, Yang et al. published a porphyrin-based composite visible-light catalyst (AZTx) for the reduction in toxic Cr(VI) ions in water [94]. In a typical procedure, Zr–TCPP was synthesized via a one-pot reaction between H₂TCPP, UiO-66-NH₂, and Zr₆ precursors. Under UV-light irradiation, the photoreduction of AgNO₃ on the core of Zr–TCPP led to the fabrication of the desired hybrid catalyst, AZTx (Ag@Zr–TCPP). From the DRS plot, E_g was determined to be 2.35 eV for AZTx (x = 5), 2.69 eV for Zr–TCPP, and 2.83 eV for UiO-66-NH₂. The integration of H₂TCPP into Zr–TCPP increased its visible-light harvesting capacity. The integration of silver plasmons onto the surface of Zr–TCPP further

intensified its photocatalytic activity for the reduction in an aqueous solution of toxic Cr(VI) ions to less toxic Cr(III). Under 30 min of visible-light irradiation, AZT5 (5 wt% ratio of Ag nanoparticles to Zr–TCPP) converted 94.1% of the toxic Cr(VI) ions into less toxic Cr(III) under acidic conditions. The degradation rate constant of Ag@Zr–TCPP was 3.6–5.4 times superior to that of Zr–TCPP. The integration of Ag nanoparticles onto the core of Zr–TCPP improves the light-harvesting activity, enhances the separation of electron–hole pairs, delays the recombination time, and improves the photocatalytic activity (Figure 26). Interestingly, bimetallic MOFs increase the active site of the compound, encourage internal electron transfer, and change the activation path via the synergistic effect of bimetals compared to monometallic MOFs [95,96].

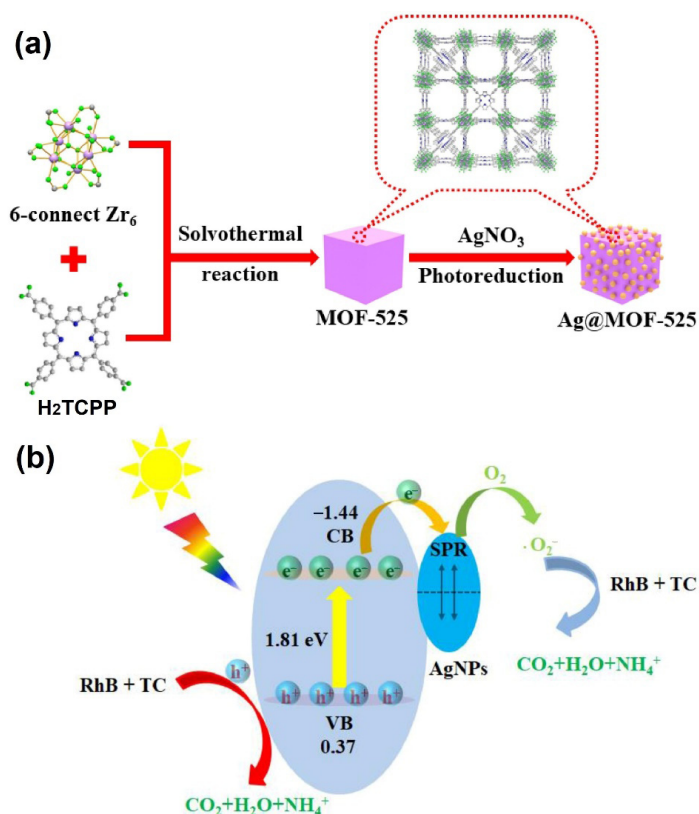


Figure 25. Hybrid photocatalyst Ag@MOF-525: (a) synthesis and (b) photodegradation mechanism. Adapted from [93].

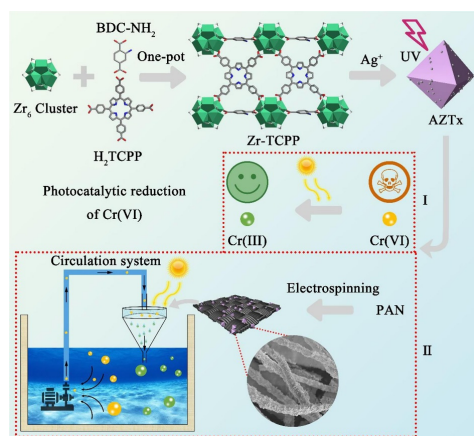


Figure 26. Mechanistic procedure for the visible-light catalytic reduction in Cr(VI) in H₂O. Adapted from [94].

In 2024, Kim et al. constructed SA-T@Ag nanocomposite materials for catalytic MB dye degradation under visible-light exposure [97]. The reaction of Zn(II)porphyrin with tin(IV)porphyrin led to the formation of the Zn(II)–Sn(IV)–Zn(II) triad (SA-T). Silver NPs were then successfully decorated onto the surface of the porphyrin nanofibers (SA-T) via photoreduction to construct the nanocomposite SA-T@Ag. The E_g values of SA-T and SA-T@Ag calculated from the Tauc plot were calculated to be 2.55 eV and 2.44 eV, respectively. The formation of SA-T@Ag reduced the E_g but enhanced the visible-light harvesting properties compared with those of SA-T. This delays the recombination of the photogenerated reactive pairs and hence improves the photodegradation rate. Within 45 min, the first-order MB dye photodecomposition rate constant was found to be 0.081 min^{-1} for the SA-T@Ag nanocomposite and 0.028 min^{-1} for SA-T only (Figure 27). To know the degradation products of the MB dye, mass spectra of the reaction mixture were taken after 30 min for each photodegradation experiment. New peaks appeared in the mass spectra that confirm the degradation of the MB dye into new fractions. Initially, the base peak ($m/z = 284.1$; $[\text{MB-Cl}]^+$) corresponds to the MB dye. It can undergo oxidative ring opening with an m/z of 304.1. This species can be further fragmented into lower-molecular-weight molecules with m/z of 175.0 or 95.0. These low-molecular-weight aromatic compounds can then undergo successive ring cleavage and hydrolysis, giving rise to compounds with an m/z of 119.0. In contrast, MB underwent *N*-de-ethylation to form a chromophoric species with an m/z of 228.0. After successive ring cleavages, it was fragmented into low-molecular-weight molecules with an m/z of 119.0 or 111.0. Finally, all low-molecular-weight molecules were mineralized into nontoxic H_2O and CO_2 [97].

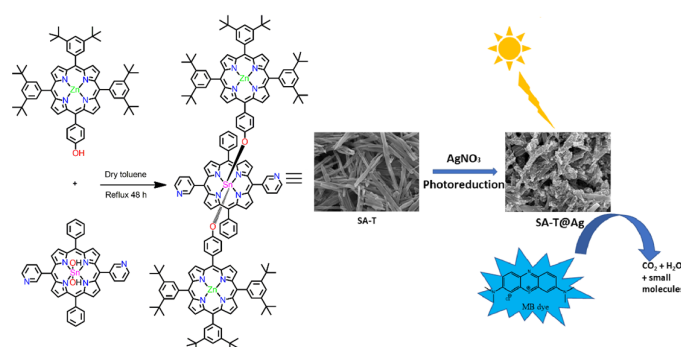


Figure 27. SA-T@Ag composite material for catalytic photodecomposition of MB dye. Adapted from [97].

5. Conjugation of Porphyrin to Carbon-Based Nanomaterials

Various carbon-based materials (graphene/g- C_3N_4 -based) have emerged as excellent catalytic systems for the photodegradation of hazardous pollutants owing to their low toxicity, narrow bandgap, and high stability [98,99]. However, the utilization of these photoactive compounds is limited by their low dispersibility in organic solvents and water. The integration of porphyrins into these materials can improve their photodegradation efficiency.

5.1. Porphyrin- C_3N_4 Hybrid Nanomaterials

A hybrid CuTCPP/g- C_3N_4 composite was fabricated using the ethanol interspersed approach [100]. CuTCPP compounds were easily incorporated onto the surface of the g- C_3N_4 nanosheets via covalent bonds. However, various intermolecular interactions (π - π stacking and electrostatic interactions) between CuTCPP and the g- C_3N_4 nanosheets were responsible for the association of these two materials. Within 10 h of white-light irradiation, the first-order photocatalytic phenol degradation rate constants were determined

to be 0.024 h^{-1} for the CuTCPP/g- C_3N_4 composite, 0.011 h^{-1} for g- C_3N_4 , and 0.0012 h^{-1} for CuTCPP. The improved photocatalytic performance arises from the significant electron transfer to g- C_3N_4 originating from the photoexcited CuTCPP. The incorporation of CuTCPP onto the surface of g- C_3N_4 not only increased the photodegradation performance but also enhanced the visible-light harvesting activity (Figure 28).

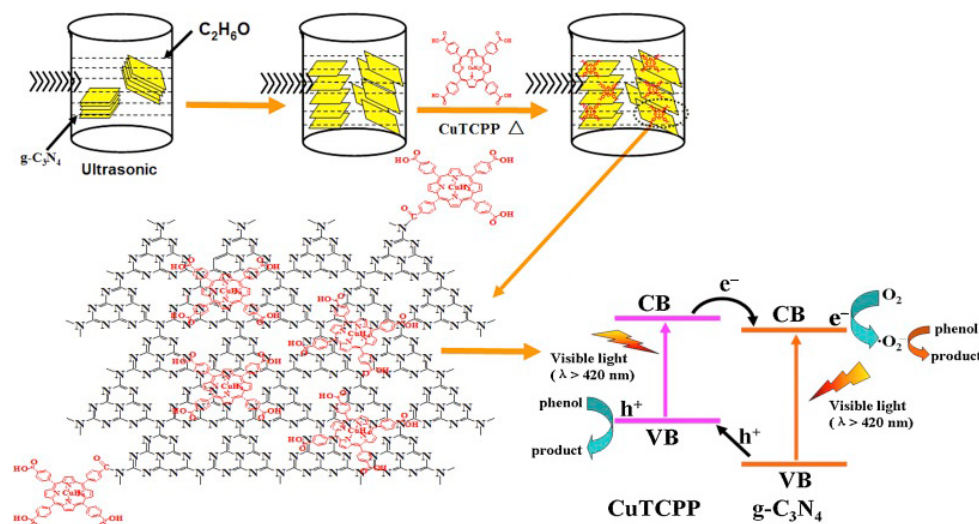


Figure 28. CuTCPP@g- C_3N_4 composite material for photocatalytic photodegradation of phenol. Adapted from [100].

In 2018, Zhang et al. synthesized hybrid nanomaterials with several metalloporphyrins and g- C_3N_4 [101]. The solvothermal reaction between g- C_3N_4 and the metalloporphyrin leads to the formation of these composite nanomaterials. The integration of porphyrin onto the surface of g- C_3N_4 not only intensifies the visible-light harvesting property but also lowers the band gap energy of the hybrid materials compared with either metalloporphyrin or g- C_3N_4 . From the DRS, the E_g values were found to be 2.37 eV for g- C_3N_4 , 2.22 eV for CoTPP/g- C_3N_4 , 2.25 eV for NiTPP/g- C_3N_4 , and 2.27 eV for CuTPP/g- C_3N_4 . Under visible-light irradiation, all these hybrid nanomaterials exhibited an amplified catalytic photodegradation activity for RhB dye in water. The CoTPP/g- C_3N_4 composite was much more efficient than CuTPP/g- C_3N_4 , NiTPP/g- C_3N_4 , and pristine g- C_3N_4 . This enhanced photodegradation performance was attributed to the more effective interfacial charge transfer between g- C_3N_4 and CoTPP (Figure 29).

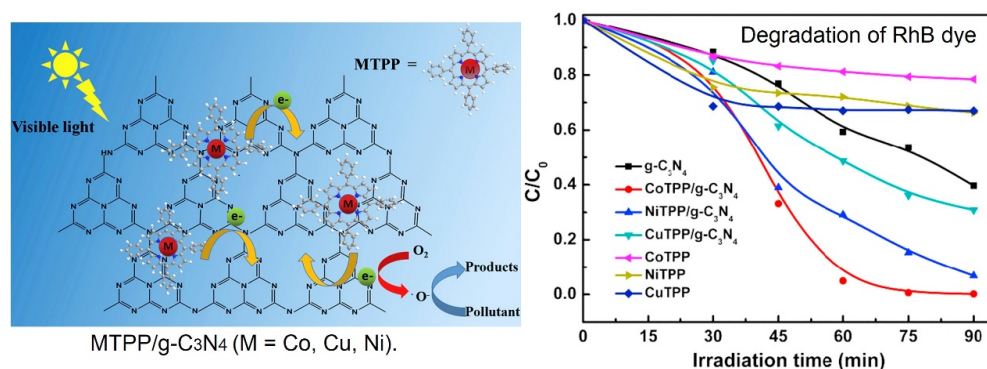


Figure 29. Decomposition of RhB dye in H_2O using porphyrin/g- C_3N_4 -based hybrid materials under visible light. Adapted from [101].

An all-organic hybrid nanomaterial, SA-TCPP/O-CN, was synthesized for photocatalytic application under visible-light irradiation [102]. Oxygen-doped g- C_3N_4 (O-CN)

nanosheets and self-assembled porphyrin (SA-TCPP) nanostructures were integrated to fabricate 0D/2D hybrid nanostructures via in situ electrostatic interactions. From the DRS plot, the E_g values for O-CN and SA-TCPP were found to be 2.31 eV and 1.78 eV, respectively. The integration of SA-TCPP with O-CN improved the visible-light harvesting properties of SA-TCPP/O-CN. Interfacial carrier transfer and electron delocalization between SA-TCPP and O-CN occurred readily owing to highly similar π - π interactions and band structures of these two materials. This improved the visible-light catalytic activity of the hybrid material. Compared with SA-TCPP or O-CN, SA-TCPP/O-CN showed efficient catalytic performance for O_2 evolution, toxic pollutant decomposition, and disinfection (Figure 30).

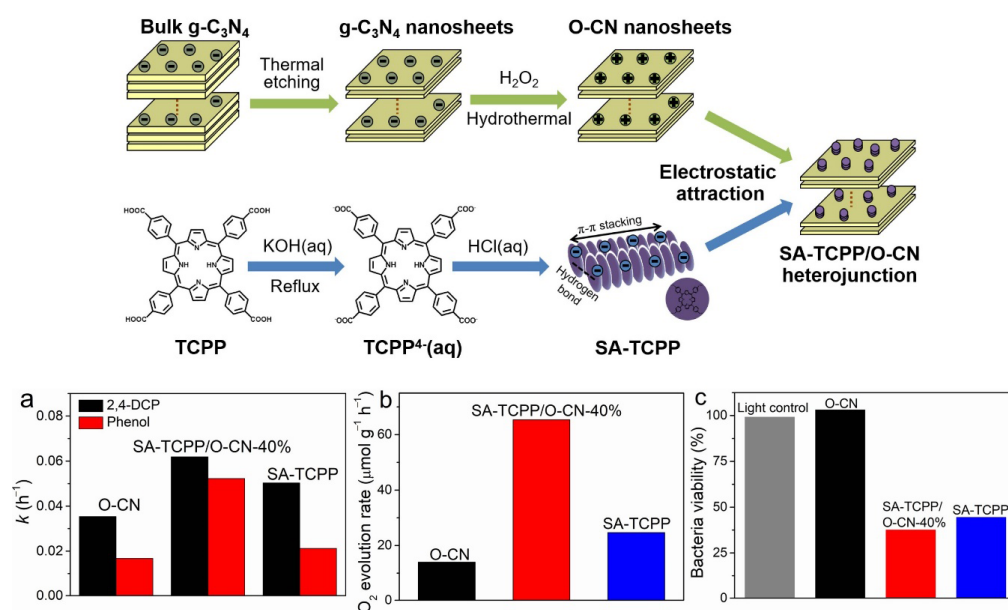


Figure 30. Schematic of the fabrication of the SA-TCPP/O-CN hybrid photocatalyst. Photocatalytic degradation rate constants of phenol and 2,4-dichlorophenol (2,4-DCP) (a). O_2 evolution reaction (b). Bacterial viabilities of *S. aureus* cells under visible light (c). Adapted from [102].

Recently, a Pd(II)-mediated multiporphyrin-sensitized g-C₃N₄ hybrid catalyst was constructed for Cr(VI) reduction in water [103]. The reaction of oxidized graphitic carbon nitride (O-g-C₃N₄) with Pd(II)-mediated multiporphyrin arrays of tetrakis(4-aminophenyl)porphyrin (TAPP) led to the fabrication of hybrid photocatalyst O-g-C₃N₄@(Pd-TAPP)_n. From the DRS, the E_g values for O-g-C₃N₄ were found to be 2.69 eV and 1.98 eV for O-g-C₃N₄@(Pd-TAPP). The integration of porphyrinic materials onto the surface of O-g-C₃N₄ improved the visible-light harvesting ability and enhanced the transportation and separation of photogenerated carriers, thereby increasing the catalytic photodegradation activity. Under solar-light exposure, >90% Cr(VI) was reduced using the hybrid photocatalyst O-g-C₃N₄@(Pd-TAPP)₃ ($K = 0.107 \text{ min}^{-1}$) within 60 min (Figure 31).

Zhao et al. also reported the formation of a porphyrin-based heterojunction NVCN/TCPP for the visible-light photodecomposition of RhB dye in H₂O [104]. The reaction of tetra(4-carboxyphenyl) porphyrin (TCPP) with nitrogen-vacancy-enriched g-C₃N₄ (NVCN) led to the fabrication of an NVCN/TCPP heterojunction. A mechanistic investigation indicated that π - π interactions between g-C₃N₄ and TCPP were responsible for the formation of this heterojunction. From the DRS, the E_g values were 1.67 eV and 2.60 eV for TCPP and NVCN, respectively. The integration of TCPP molecules onto the surface of nitrogen-vacancy-enriched g-C₃N₄ in the NVCN/TCPP heterojunction reduced the bandgap energy and boosted the photocatalytic performance via the Z-scheme strategy. Within 60 min, the degradation rate constant was 0.034 min^{-1} for the NVCN/TCPP heterojunction vs. 0.012 min^{-1} for NVCN (Figure 32).

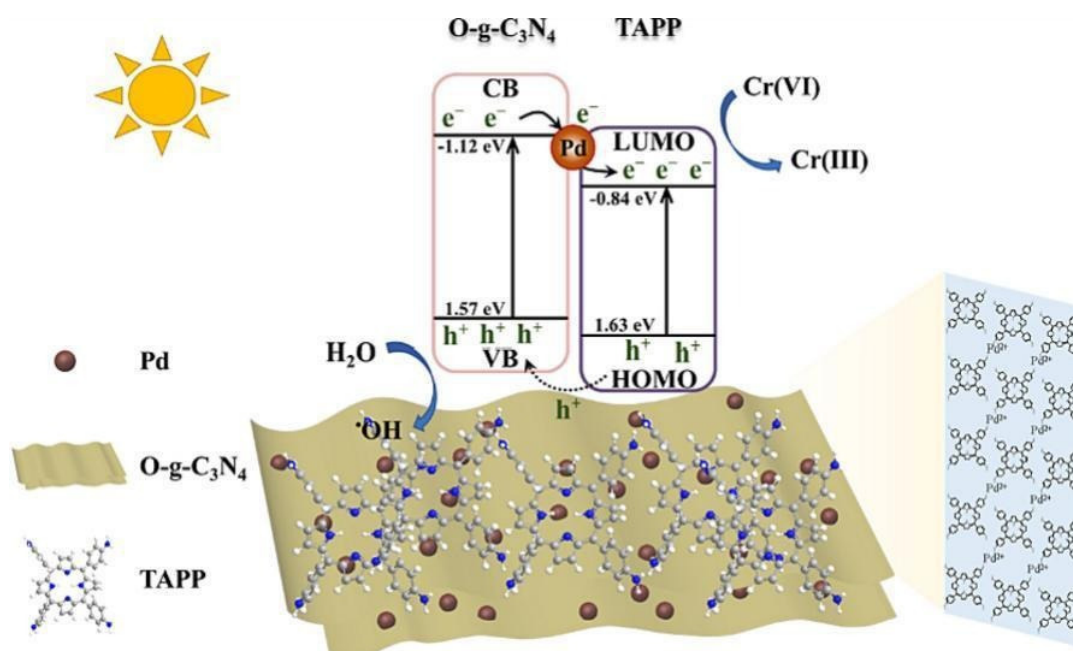


Figure 31. Pd(II)-mediated multiporphyrin-sensitized g-C₃N₄ hybrid photocatalyst for Cr(VI) reduction in water. Adapted from [103].

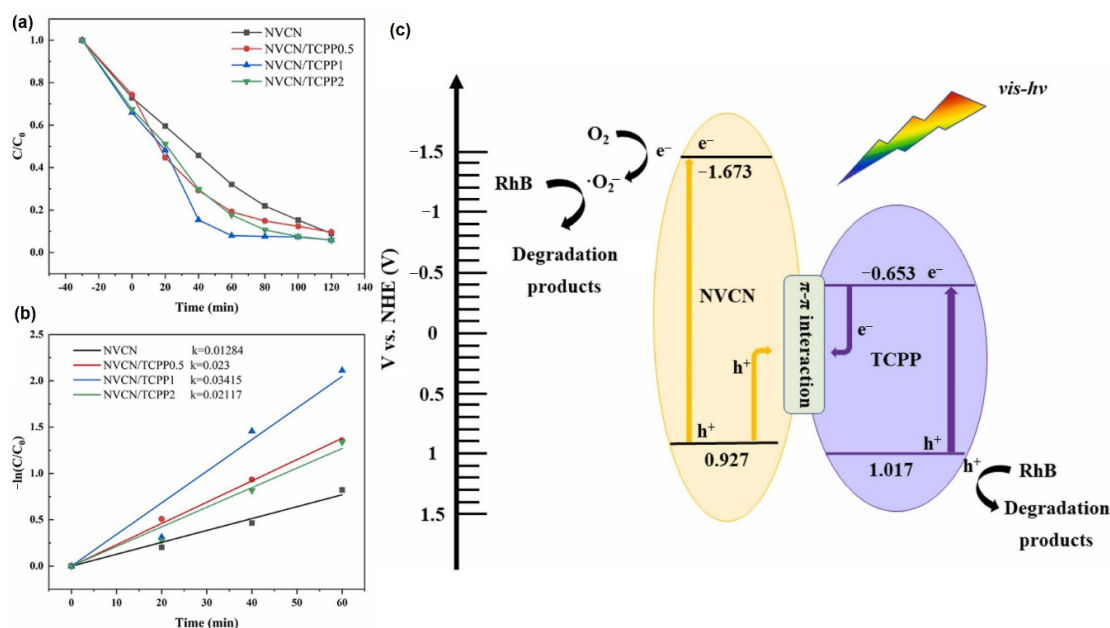


Figure 32. Porphyrin-based heterojunction NVCN/TCPP: (a) photodegradation of RhB dye, (b) rate kinetics, and (c) mechanism. Adapted from [104].

5.2. Porphyrin–Graphene Hybrid Nanomaterials

Graphene, GO, and rGO have been examined as promising photocatalysts owing to their huge active surface area, excellent electron mobility, peculiar two-dimensional honeycomb morphology, and flexible functional groups [105]. Additionally, the fabrication of porphyrin with these compounds leads to the fabrication of hybrid photocatalysts with high conductivity, excellent stability, and photocatalytic performance.

In 2017, La et al. constructed a graphene@porphyrin nanofiber composite material for visible-light photocatalysis [106]. In the presence of D-arginine, the acid–base neutralization reaction of tetrakis(4-carboxyphenyl) porphyrin (H₂TCPP) with graphene nanoplates (GNPs) led to the fabrication of graphene@TCPP nanofibers (Figure 33). The self-assembled

porphyrin nanofibers were consistently spread on the graphene surface. The synthesized composite materials exhibited enhanced catalytic photodegradation of RhB and MO dyes compared with the starting materials (graphene nanoplates or free-standing H₂TCPP). Within 150 min, the RhB photodegradation ratio was 100% for the graphene@porphyrin nanofibers under solar-light irradiation. However, within 180 min of irradiation, the MO degradation ratio was 80% for graphene@porphyrin nanofibers.

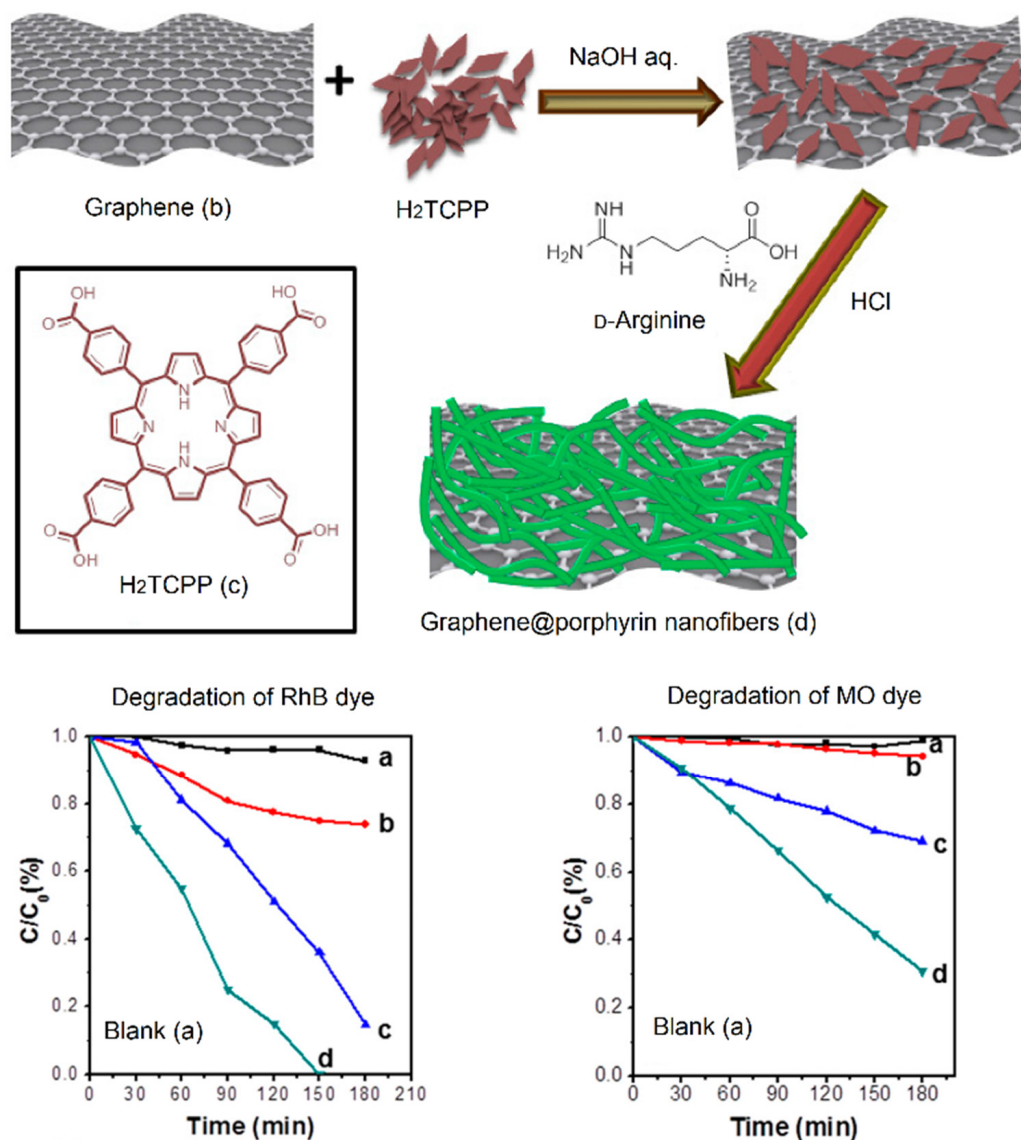


Figure 33. Photocatalytic decomposition of RhB dye in H₂O using graphene@porphyrin nanofiber hybrid material under visible light. Adapted from [106].

Andralojc et al. reported improved photocatalytic decomposition of RhB dye using a porphyrin/graphene oxide-based nanohybrid [107]. Two cationic porphyrins (TMPyP and ZnTMPyP) were immobilized over GO via non-covalent interactions to form TMPyP-GO and ZnTMPyP-GO nanohybrids. The incorporation of GO into porphyrins not only amplifies the surface area and electron conductivity but also enhances the optical properties. Within 120 min, the photodegradation ratio of RhB was 19% for ZnTMPyP-GO, which was higher than that of TMPyP-GO (10%) and GO (3.7%). Various spectroscopic measurements confirmed that ZnTMPyP can enhance the affinity of the metalloporphyrin for the GO surface, facilitate charge separation, and hence increase the photodegradation activity under visible-light irradiation (Figure 34).

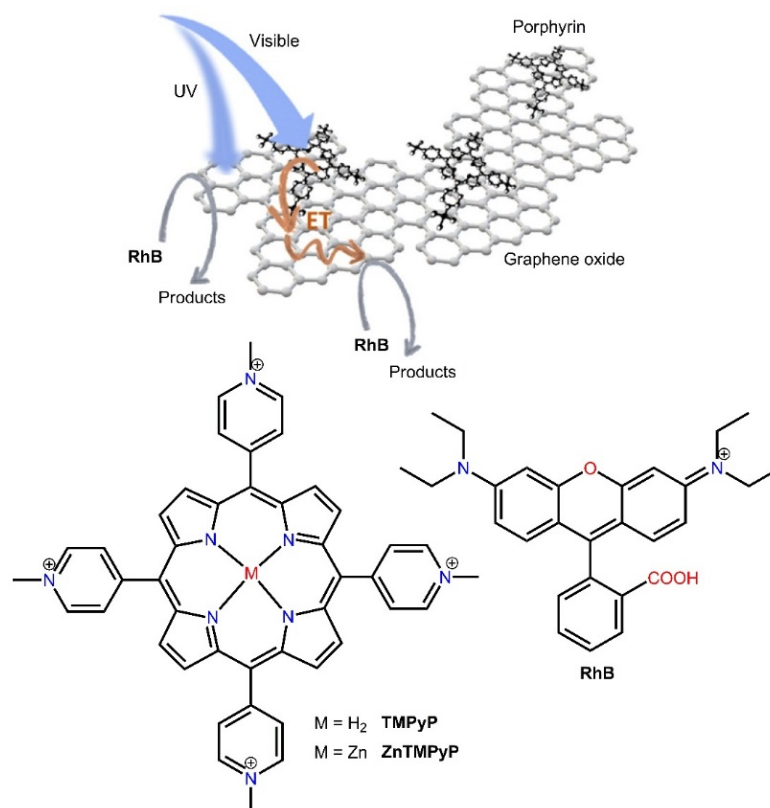


Figure 34. Catalytic photodegradation of RhB dye in water using graphene oxide-based porphyrin under visible-light irradiation. Adapted from [107].

Porphyrin-based reduced graphene oxide composites (rGO-P) were constructed for Congo Red (CR) degradation in water [108]. Sonicating reduced graphene oxide (rGO) with 5,15-bisdodecyl porphyrin led to the fabrication of rGO-P. The incorporation of porphyrin with rGO not only boosts the visible-light harvesting capacity but also generates a large active surface for the interaction and diffusion of substrate molecules. Within 90 min of UV-light irradiation, the CR degradation ratio was 57% for rGO-P and 30% for rGO (Figure 35). The improved degradation activity was attributed to the synergistic effect between rGO and the porphyrin.

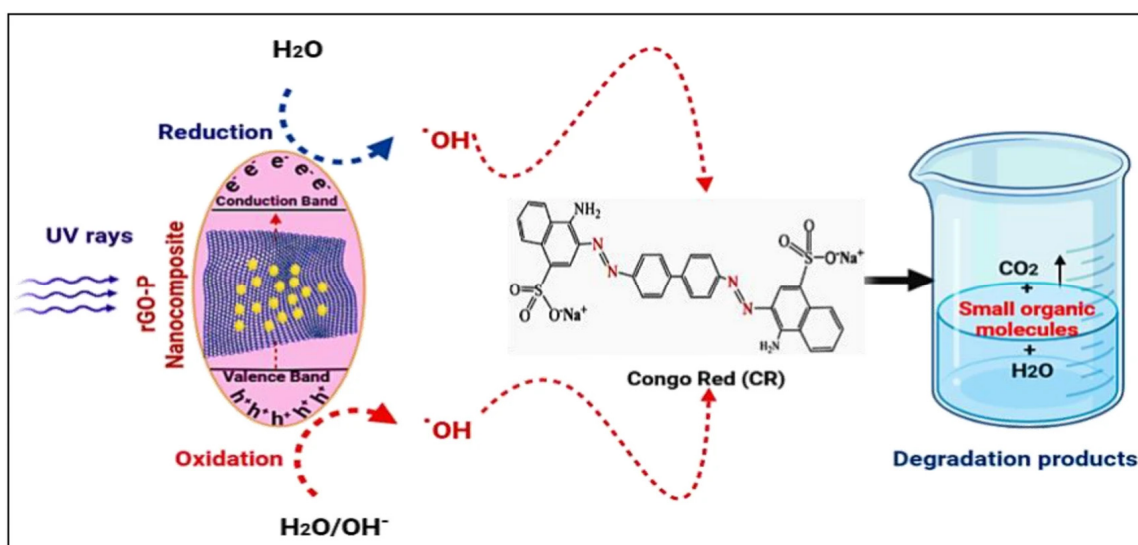


Figure 35. Schematic of rGO-P hybrid material for CR dye degradation. Adapted from [108].

Recently, a porphyrin-sensitized GO nanohybrid [Co(II) TPHPP]-Cs/GO was fabricated for the visible-light catalytic photodegradation of Acid Orange 7 dye in H₂O [109]. Sonication of 5-, 10-, 15-, and 20-tetrakis[4-(hydroxy)phenyl] porphyrin [Co(II) TPHPP] covalently supported chitosan with GO led to the formation of a [Co(II) TPHPP]-Cs/GO nanocomposite. The integration of porphyrin with GO not only enhanced the visible-light harvesting properties but also generated a large active surface area. The substrate dye molecules were speculated to interact with the [Co(II) TPHPP]-Cs/GO nanohybrid, thereby improving the transportation of charged species. The high stability of the nanocomposite prevents hydrolysis during photodegradation and can be used without loss of catalytic activity. Within 60 min, the degradation ratio of Acid Orange 7 dye was found to be 94% for the [Co(II) TPHPP]-Cs/GO/H₂O₂ system. Mechanistic investigations indicated that reactive hydroxyl radicals were the main reactive species during the photodegradation reaction (Figure 36).

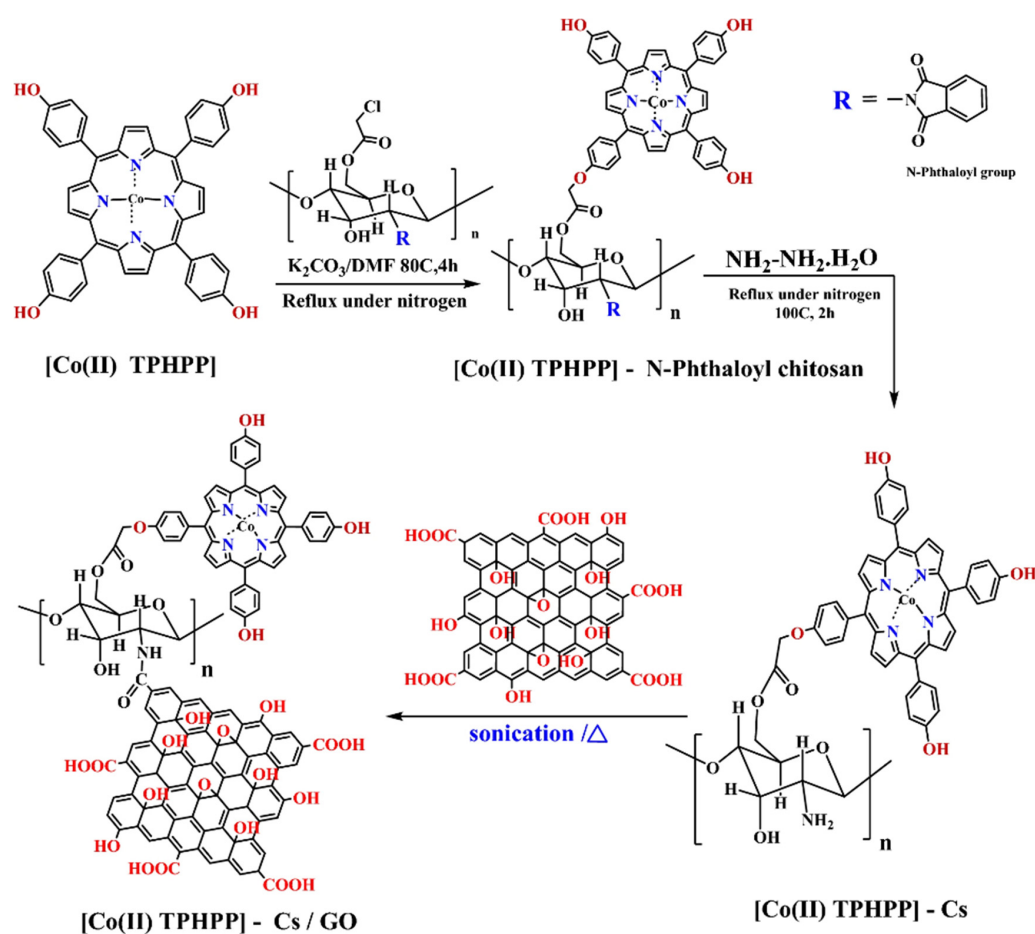


Figure 36. Schematic of the synthesis of [Co(II) TPHPP]-Cs/GO nanohybrid for photocatalytic decomposition of Acid Orange 7 dye in H₂O. Adapted from [109].

6. Porphyrin-Conjugated Organic Polymer Nanomaterials

The conjugation of porphyrin to organic polymeric matrices provided significant results for the photocatalytic decomposition of hazardous contaminants in water with respect to the starting porphyrins. Moreover, the strong covalent backbone prevents the hydrolysis and separation of porphyrins during photodegradation.

In 2020, Mota et al. demonstrated the effective incorporation of a water-soluble [5,10,15,20-tetrakis(N-methyl-4-pyridinium)]zinc(II) porphyrin cation into a polyacrylic acid (PAA) matrix, leading to Zn(II)-porphyrin/poly(acrylic acid) hybrid materials [110].

Under visible-light irradiation, the as-synthesized hybrid microparticles were excellent catalysts for the photodegradation of organic contaminants such as nitrobenzene (NB), MO dye, and MB dye in water, compared with the starting monomeric porphyrin unit. The immobilization of metalloporphyrin not only improved the interaction of porphyrin molecules with the polymeric matrix but also increased the stability of the microparticles for further use (Figure 37).

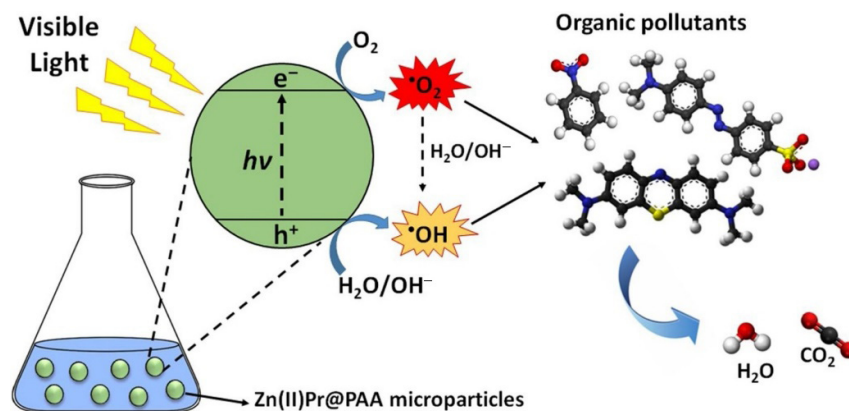


Figure 37. Zn(II)-porphyrin/poly(acrylic acid) hybrid materials for catalytic decomposition of organic contaminants. Adapted from [110].

Several cationic porphyrins (TMPyP, TMPyPZn, and TMPyPMn) were integrated into a polymer aerogel to form hybrid porphyrin-integrated photocatalysts (CPA-TMPyP, CPA-TMPyPZn, and CPA-TMPyPMn) in situ [111]. Polyacrylic acid (PAAc)/polyvinyl alcohol (PVA) was used as the source of the polymer matrix (CPA). These cationic porphyrins were successfully immobilized inside the polymeric aerogels via hydrogen bonding and electrostatic interactions and exhibited excellent photocatalytic activity for the decomposition of various pharmaceuticals (naproxen, caffeine, and amoxicillin) under visible-light irradiation in water. Within 60 min, CPA-TMPyPMn exhibited higher photocatalytic activity than the free porphyrin, TMPyPMn, or the other hybrid materials (CPA-TMPyP and CPA-TMPyPZn). Recycling experiments confirmed that this hybrid-porphyrin-integrated aerogel maintained superior photocatalytic performance, even after 15 cycles (Figure 38).

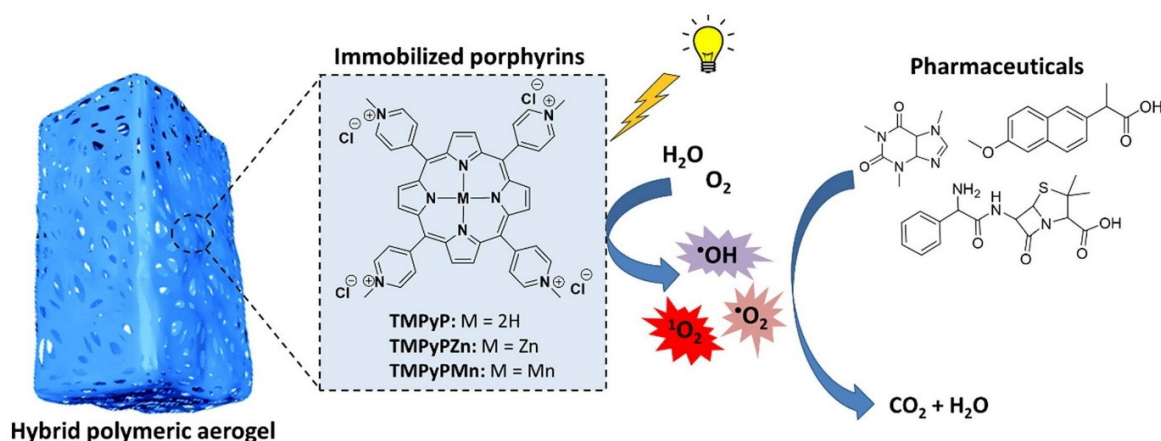


Figure 38. Visible-light catalytic photodegradation of various pharmaceuticals in water using porphyrin-based hybrid polymer aerogels. Adapted from [111].

A mesoporous porphyrin-based Amberlite photocatalytic system was studied for the photodegradation of pollutants under visible-light irradiation [112]. The top-down method between the anion exchange polymer nanoAmbN(Me)₃Cl (Amb = Am-

berlite IRA-410(Cl)) and the BF_3 complex meso-tetrakis(4-sulfonatophenyl)porphyrin (H_2TPPS_4) led to the construction of the highly porous porphyrin-based photocatalyst $\text{nanoAmbN}(\text{Me})_3\text{@H}_2\text{TPPS}_4(\text{BF}_3)_2$. The incorporation of porphyrin into the core of polymeric matrices not only changes the surface morphology but also creates a large active porous site for catalysis. Within 320 min of visible-light irradiation, 99% 1,5-dihydroxynaphthalene (DHN) was degraded using $\text{nanoAmbN}(\text{Me})_3\text{@H}_2\text{TPPS}_4(\text{BF}_3)_2$ in acetonitrile (Figure 39). The high degradation rate was attributed to the synergistic effect between the porphyrin and Amberlite and was better than that of either $\text{nanoAmbN}(\text{Me})_3\text{Cl}$ or $\text{H}_2\text{TPPS}_4(\text{BF}_3)_2$.

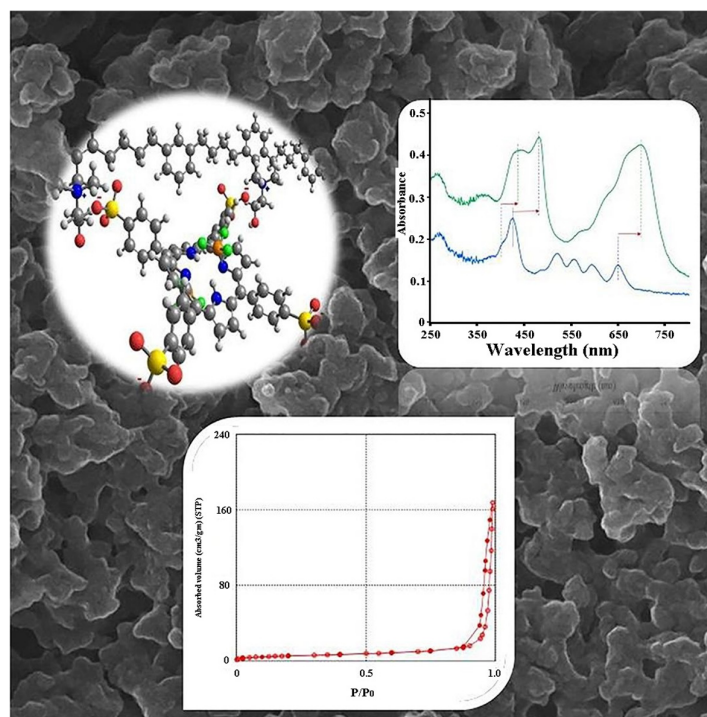


Figure 39. Porphyrin-integrated highly mesoporous Amberlite hybrid photocatalyst for water treatment. Adapted from [112].

In 2025, Kenawy et al. fabricated a porphyrin in polyacrylonitrile (PAN) matrices for crystal violet dye degradation in the presence of H_2O_2 [113]. Electrospinning of Co(II) -porphyrin with chloroacetylated poly(*p*-hydroxystyrene) led to the formation of porphyrin-supported nanofiber materials. The incorporation of porphyrin into PAN matrices not only changes the surface morphology but also enhances the degradation performance in water. Within 60 min, the crystal violet degradation ratio was 99% for porphyrin-supported nanofiber materials in the presence of H_2O_2 . Immobilization of the rigid porphyrin framework onto the polymeric backbone aided the recovery process while maintaining catalyst rigidity (Figure 40).

A porphyrin-based polyelectrolyte composite membrane was constructed for the catalytic photodecomposition of RhB and MB dyes in H_2O [114]. A multistep reaction between tetrakis(4-carboxyphenyl)porphyrin (TCPP) and hyperbranched polyamide-amine (HPAMAM) in the presence of phytic acid (PA) guided the fabrication of $\text{TCPP@HPAMAM@PA/PVDF}$ composite membranes through electrostatic interactions. The integration of porphyrin with the polyamide-amine not only enhanced the catalytic performance but also prevented the separation of the porphyrin from the $\text{TCPP@HPAMAM@PA/PVDF}$ membrane during the photodegradation reactions. The visible-light photodecomposition ratios of RhB and MB

dyes for the TCPP@HPAMAM@PA/PVDF composite membrane reached 93% and 96%, respectively (Figure 41).

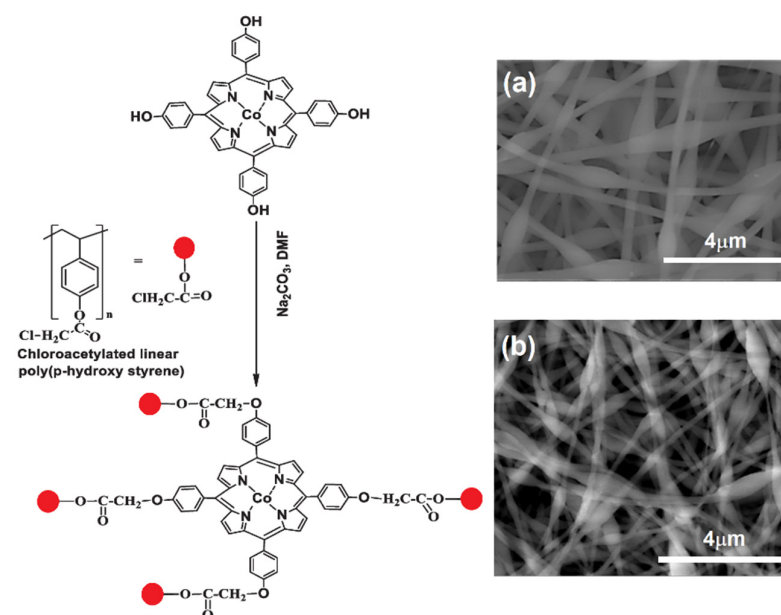


Figure 40. Schematic of the synthesis of porphyrin-based electrospun polyacrylonitrile nanofiber for visible-light photodegradation of crystal violet dye. SEM images of a (a) fresh sample and a (b) used sample. Adapted from [113].

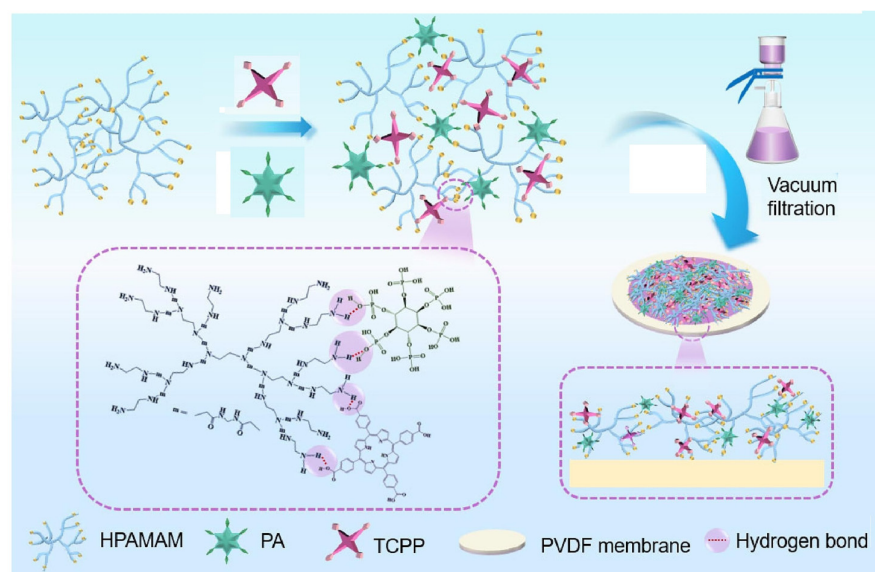


Figure 41. Diagram of the synthesis of the TCPP@HPAMAM@PA/PVDF composite membrane for dye degradation. Adapted from [114].

7. Other Porphyrin-Conjugated Hybrid Nanomaterials

A Co-N-C/SA-TCPP hybrid photocatalyst for water remediation was studied [115]. The self-assembled porphyrin nanostructures of H₂TCPP were integrated over the surface of the N-doped carbon framework materials in situ. In the Co-N-C/SA-TCPP heterojunction, Co-N-C nanocubes were tightly coupled with porphyrin nanostructures to fabricate a 3D/0D hybrid composite via chemical bonding, hydrogen bonding, and π - π interactions. E_g derived from the Tauc plot was found to be 1.79 eV for SA-TCPP and 1.30 eV for Co-N-C/SA-TCPP. The integration of the porphyrin nanostructure onto the surface of

Co-N-C not only lowered the band gap energy in Co-N-C/SA-TCPP but also exhibited photocatalytic degradation activity against organic pollutants as well as oxygen evolution in water under visible-light irradiation. The photocatalytic efficiency of Co-N-C/SA-TCPP was several times higher than that of Co-N-C and SA-TCPP. The synergistic effect between the Co-N-C nanocubes and SA-TCPP nanostructures has been reported to be responsible for the improved photocatalytic oxidation performance. The incorporation of porphyrin nanostructures onto the surface of the Co-N-C nanocubes not only strengthened light utilization but also facilitated charge separation, improved oxidation capacity, and better recyclability for further use (Figure 42).

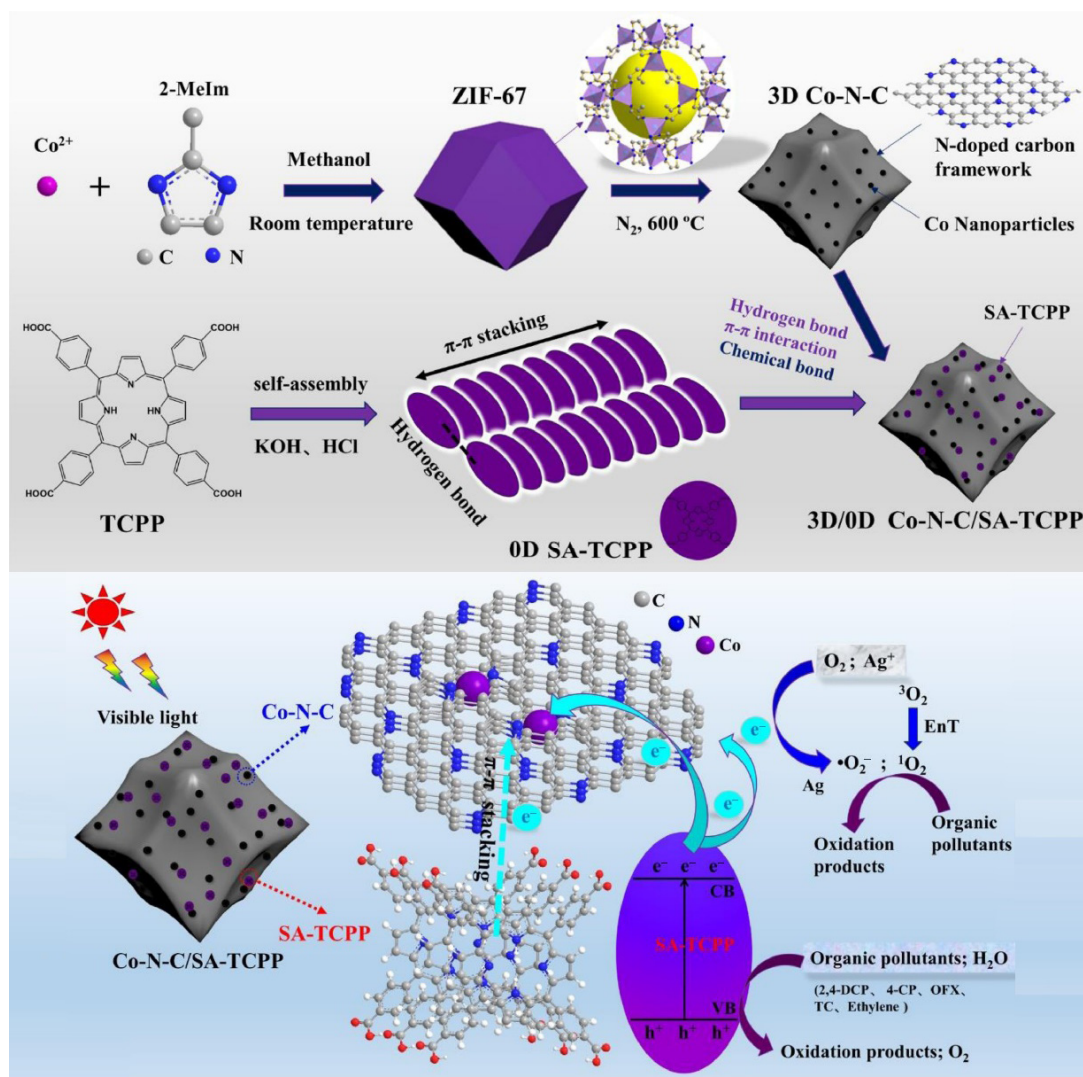


Figure 42. Co-N-C@SA-TCPP composite for catalytic photodecomposition of water contaminants. Adapted from [115].

Bhosale et al. also developed porphyrin-integrated supramolecular architectures for solar-light photodecomposition of the RhB dye [116]. The reaction of gentamicin with meso-tetrakis(4-sulfonatophenyl)porphyrin (TPPS) led to the fabrication of nanorod-like architectures with lengths of several micrometers. Their diameters varied from 50 to 100 nm. Mechanistic investigations indicated that H-bonding interactions between the $-\text{OH}$ and $-\text{NH}_2$ groups of gentamicin and the sulfonate groups of porphyrins were the main driving force for the fabrication of self-assembled nanostructures. Moreover, aromatic π - π interactions between porphyrin and gentamicin contribute to the nanoaggregation.

The integration of TPPS porphyrin with gentamicin not only lowered the band gap energy (E_g varied from 2.7 eV to 3.0 eV) but also enhanced the photodegradation activity. Within 240 min of visible-light irradiation, this supramolecular nanostructure degraded 99% of RhB dye with a rate constant of $2 \times 10^{-2} \text{ min}^{-1}$ (Figure 43).

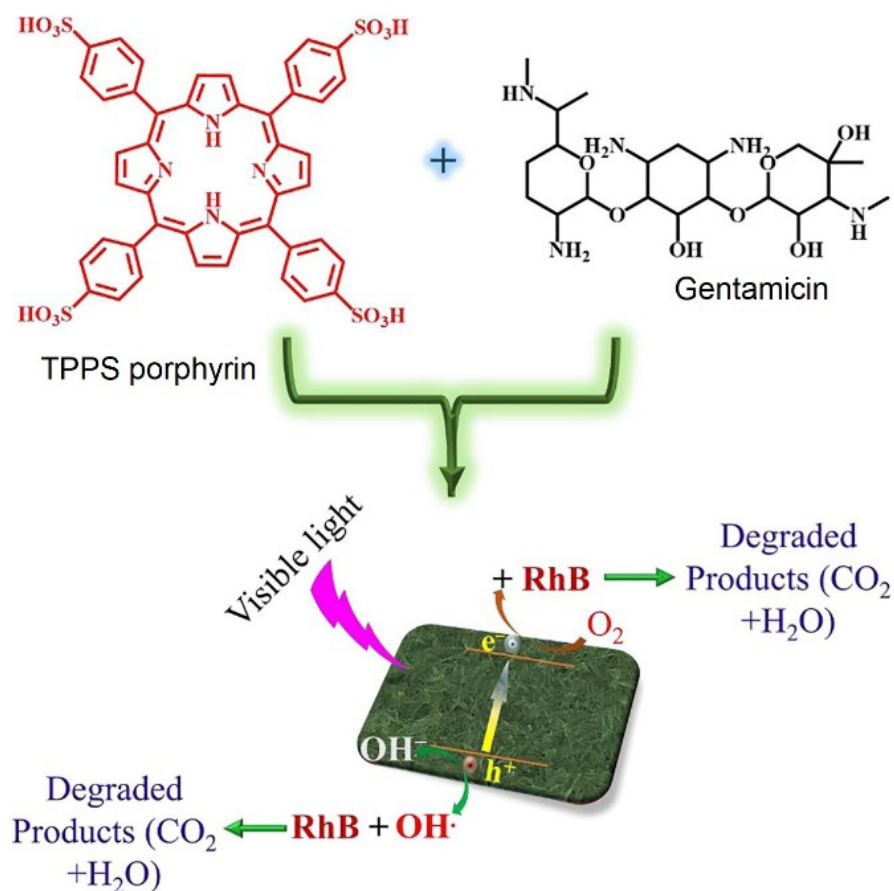


Figure 43. Diagram of the synthesis of TPPS porphyrin-conjugated supramolecular architectonics for catalytic degradation of RhB dye in water. Adapted from [116].

Additional porphyrin-conjugated hybrid nanomaterials used for the photodegradation of toxic contaminants are summarized in Table 1.

Table 1. Reaction parameters for the photodecomposition of water contaminants using porphyrin-conjugated hybrid photocatalysts.

Photocatalysts	Pollutant	Pollutant Concentration (mg L^{-1})	Catalyst Dosage (mg L^{-1})	Energy Source Lamp	Irradiation Time (min)	Degradation Efficiency (%)	Ref.
TiO_2	4-NP	14	200	125 W Iodine–Tungsten	400	9.19	[117]
CuPp-TiO_2	4-NP	14	200	125 W Iodine–Tungsten	400	99.1	[117]
H_2O_2	Atrazine	20	1000	300 W Xenon	60	5	[118]
$\text{TCPPCu/TiO}_2\text{@H}_2\text{O}_2$	Atrazine	20	1000	300 W Xenon	60	82	[118]
SnP-TiO_2	4-NP	14	200	500 W Xe lamp	500	98	[119]
ZnO	RhB	5	2000	1000 W Halogen–Tungsten	60	30	[120]
ZnO/TAPPI-CoTPPS	RhB	5	2000	1000 W Halogen–Tungsten	60	100	[120]

Table 1. Cont.

Photocatalysts	Pollutant	Pollutant Concentration (mg L ⁻¹)	Catalyst Dosage (mg L ⁻¹)	Energy Source Lamp	Irradiation Time (min)	Degradation Efficiency (%)	Ref.
NiPp-TiO ₂	4-NP	14	200	400 W Halogen lamp	66	78	[121]
SnP-TiO ₂	4-NP	14	1000	1000 W Iodine–Tungsten	180	85	[122]
Ti-700	MB	10	1000	150 W Halogen lamp	120	30	[123]
NiTCPP/Ti-700	MB	10	1000	150 W Halogen lamp	120	64	[123]
TNTs–ZnTPP	MO	15	1000	1000 W Iodine–Tungsten	60	85	[124]
PtPp(1)–TiO ₂	4-NP	14	250	500 W Xe lamp	300	99	[125]
Poly(styrene)@FeTPP/TiO ₂ /H ₂ O ₂	MO	50	666	40 W Incandescent	300	99	[126]
P25-TiO ₂	Famotidine	280	310	500 W Halogen lamp	180	10	[127]
TCPP/P25-TiO ₂	Famotidine	280	310	500 W Halogen lamp	180	100	[127]
CuPp-TiO ₂	MO	10	1000	50 W Fluorescent	240	45	[128]
CuPp-La-TiO ₂	MO	10	1000	50 W Fluorescent	240	50	[128]
TCPP-NH ₂ -V-TiO ₂	MO	6	500	400 W Xe lamp	180	62	[129]
TCPP-V-TiO ₂	MO	6	500	400 W Xe lamp	180	95	[129]
FeTPP/Poly(acrylonitrile)	Reactive orange	5	666	40 W Incandescent	120	83	[130]
ZnPz(COOH) ₄ @Amberlite CG-400 resin	RhB	10	800	500 W Halogen	720	80	[131]
CoTPP/N-TiO ₂	MB	5	1000	150 W Xe lamp	180	94	[132]
CoTHPP/N-TiO ₂	MB	5	1000	150 W Xe lamp	180	92	[132]
ZnTPP/TiO ₂	MO	10	1000	1000 W	180	70	[133]
HTPP/TiO ₂	MO	10	1000	Iodine–Tungsten 1000 W	180	86	[133]
SnHTPP/TiO ₂	MO	10	1000	Iodine–Tungsten	180	50	[133]
H ₂ TCPP-CdS	RhB	4	1000	Sunlight	120	92	[134]
CuAPTPP-TDI-TiO ₂	MB	20	1000	150 W Xe lamp	120	99	[135]
PP-ZnO	MB	NA	NA	8 W UV light	60	25	[136]
(Fe)PP-ZnO	MB	NA	NA	8 W UV light	60	100	[136]
(Cu)PP-ZnO	MB	NA	NA	8 W UV light	60	80	[136]
TiO ₂ -nanowire	MB	9.2	180	200 W Fluorescence Xe	420	51	[137]
TiO ₂ /graphene	MB	9.2	180	200 W Fluorescence Xe	420	72	[137]
TNO ₂ PP-TiO ₂ /graphene	MB	9.2	180	200 W Fluorescence Xe	420	85	[137]
P25- TiO ₂	MB	20	500	250 W Iodine–Tungsten	50	5	[138]
ZnTCPP–TNTS	MB	20	500	250 W Iodine–Tungsten	50	100	[138]
ZnTCPP/Ag-TiO ₂	4-NP	10	1000	500 W Hg lamp	150	100	[139]
Graphene/TiO ₂ /H ₂ TCPP	MO	10	1200	450 W Xe lamp	240	93	[140]
UPC-CMP-1	Congo red	28	NA	300 W Xe lamp	2	88	[141]
TiO ₂ nanotube	RhB	5	330	300 W Xe lamp	240	61	[142]
H ₂ TCIPP-TNTs	RhB	5	330	300 W Xe lamp	240	93	[142]
CuTCPP	Phenol	20	1000	500 W Xe lamp	120	4	[143]

Table 1. Cont.

Photocatalysts	Pollutant	Pollutant Concentration (mg L ⁻¹)	Catalyst Dosage (mg L ⁻¹)	Energy Source Lamp	Irradiation Time (min)	Degradation Efficiency (%)	Ref.
BiPO ₄	Phenol	20	1000	500 W Xe lamp	120	18	[143]
CuTCPP/BiPO ₄	Phenol	20	1000	500 W Xe lamp	120	36	[143]
CoCPpTiO ₂	4-NP	14	200	400 W Halogen	70	100	[144]
FeTCPP/TNT	MB	10	100	500 W Halogen	120	90	[145]
PTCDA-ZnO	Eosin yellow	50	5000	Sunlight	80	85	[146]
TCPP-TiO ₂	RhB	5	5000	15 W UV	300	86	[147]
CuPp-ZnO	RhB	5	1000	300 W Halogen	150	96	[148]
BiVO ₄ /Mn ₃ O ₄ -CuTCPP	MB	10	1000	5 W LED	180	95	[149]
BiVO ₄ /Mn ₃ O ₄ -CoTCPP	MB	10	1000	5 W LED	180	100	[149]
Porphyrin/ZnFe ₂ O ₄ @polythiophene	MO	10	1000	5 W LED	180	94	[150]
TiO ₂ (hollow nanobox)	RhB	10	1000	210 W Xe lamp	180	30	[151]
ZnTCP	RhB	10	1000	210 W Xe lamp	180	35	[151]
ZnTCP-TiO ₂ (hollow nanobox)	RhB	10	1000	210 W Xe lamp	150	99	[151]
TiO ₂ -APTES-P1	MB	10	1000	Solar light	180	92	[152]
ZnF ₂ POH@TiO ₂	Tramadol	10	666	20 J cm ⁻² LED	10	65	[153]
F ₂ POH@TiO ₂	Tramadol	10	666	20 J cm ⁻² LED	10	75	[153]
Porphyrin-polyimide	MO	4	1000	300 W Xenon	480	84	[154]
Si-CIPTMS-FeTHPP	Orange II	25	10,000	15 W UV light	1440	84	[155]
P(PPor-BBO)	RhB	5	250	318 mw cm ⁻² Xe	150	98	[156]
Fe ^{III} -TCPPCl	RhB	30	2500	800 W Xe lamp	60	51	[157]
Fe ₃ O ₄ @SiO ₂ @TiO ₂ -TAPP	RhB	15	NA	300 W Xe lamp	180	95	[158]
TPyP/TiO ₂	RhB	5	NA	Visible light	180	40	[159]
Si/CuTPPS/TiO ₂	RhB	5	NA	Visible light	240	50	[160]
g-C ₃ N ₄	MB	10	1500	Visible light	30	50	[161]
ZnTCPP	MB	10	1500	Visible light	30	77	[161]
ZnTCPP/g-C ₃ N ₄	MB	10	1500	Visible light	30	96	[161]
H ₂ TF5PP-silica	Metoprolol	50	NA	1500 W Xe arc lamp	720	90	[162]
g-C ₃ N ₄	RhB	10	400	300 W Xenon lamp	90	20	[163]
CuPor-Ph-COF	RhB	10	400	300 W Xenon lamp	90	30	[163]
CuPor-Ph-COF/g-C ₃ N ₄	RhB	10	400	300 W Xenon lamp	90	86	[163]
BiOBr/SnTCPP	2,4-Dichlorophenol	10	600	5 W White LED	240	80	[164]
FeTPP/NaY (zeolite)/H ₂ O ₂	4-NP	28	1000	12 W UV	120	85	[165]
TMPyP@SPSf/PES	MO	20	NA	300 W Xenon	120	93	[166]
polymer-supported Zn-porphyrin PSBAZnPP	MO	10	400	5 W LED lamp	18	98	[167]
TPPS@Quaternized polysulfone	MO	10	NA	300 W Xenon	300	92	[168]
BiOBr/BiOCl/PANI@TCPP	MO	10	30	5 W White LED	10	95	[169]
BiOBr/BiOCl/PANI@SnTCPP	MO	10	30	5 W White LED	10	96	[169]
Porphyrin-based porous organic polymer	RhB	10	200	400 W Xenon	320	82	[170]
SnTCPP/g-C ₃ N ₄ /Bi ₂ WO ₆	Levofloxacin	10	NA	250 W Xe lamp	150	86	[171]
TCPP/TiO ₂	MB	20	666	400 W Halogen	40	50	[172]
SP-TBU-TiO ₂	MB	20	666	400 W Halogen	40	75	[172]
FePcCl16-Py-MWCNTs/H ₂ O ₂	4-Chloro-3,5-dimethylphenol	40	150	500 Xe lamp	60	100	[173]

Table 1. Cont.

Photocatalysts	Pollutant	Pollutant Concentration (mg L ⁻¹)	Catalyst Dosage (mg L ⁻¹)	Energy Source Lamp	Irradiation Time (min)	Degradation Efficiency (%)	Ref.
Zn-MOC	Tetracycline	5	1000	500 W Halogen	50	96	[174]
H ₂ TCIPP-H ₂ PVMo	2-chloroethyl ethyl sulfide	5	3	300 W Xe lamp	30	99	[175]
H ₂ TCPP@g-C ₃ N ₄ /Ag	RhB	10	1000	350 W Xenon	90	90	[176]
Porphyrin@lignin	Trimethoprim	30	NA	Hg vapor lamp	240	99	[177]
NiFe-LDH	Tetracycline	30	200	400 W Xe lamp	120	67	[178]
PdTCPP/NiFe-LDH	Tetracycline	30	200	400 W Xe lamp	120	91	[178]
Tb-porphyrin aerogel	RhB	10	1000	250 W Xenon	75	61	[179]
SnO ₂	Tetracycline	20	200	300 W Xe lamp	210	7	[180]
CuTPP-N ₃	Tetracycline	20	200	300 W Xe lamp	210	90	[180]
10%CuTPP-N ₃ /SnO ₂	Tetracycline	20	200	300 W Xe lamp	210	32	[180]
Au-COP-180/H ₂ O ₂	microcystin-LR	NA	500	20 W White LED	120	97	[181]

8. Conclusions and Future Perspectives

Heterogeneous advanced oxidation process using a porphyrin-based catalyst is a promising method for wastewater remediation. This is because the operation is simple, cost-effective, and efficient. Owing to their superior visible-light absorption coefficients, porphyrin-based materials can absorb light from freely available solar light. Moreover, their structural skeletons and intrinsic aromatic electronic properties facilitate the delocalization of photogenerated electrons during photodegradation. This delays the recombination of the photogenerated excited pair, thus increasing the catalytic performance. However, as mentioned earlier, the application of porphyrin-based materials for wastewater treatment is hindered by limitations such as facile agglomeration in solution, deactivation, difficult separation of the photocatalyst after the degradation process, and limited reuse of the photocatalyst, which can significantly increase the overall cost of the method. Therefore, the fabrication of new photocatalytic systems comprising porphyrin molecules is required to overcome these challenges.

For this purpose, porphyrins can be integrated on solid supports to fabricate hybrid materials with photocatalytic efficiencies superior to those of free-standing starting porphyrins. Numerous transition metal oxides (TiO₂, ZnO, and Fe₃O₄), main element oxides (Al₂O₃ and SiO₂), metal plasmons (Ag NPs), carbon-based micro- and nanoparticles (graphene, graphene oxide, and g-C₃N₄), gels, and polymeric matrices have been used as solid supports for the smooth fabrication of porphyrin-conjugated hybrid materials. The incorporation of porphyrins onto the solid supports not only shortens the band gap energy but also amplifies the photocatalytic degradation performance of the hybrid materials compared with the starting components. Hybridization of porphyrins onto solid supports enhances visible-light harvesting activity, recyclability for reuse, permanent porosity, substrate mobility owing to active sites, and charge separation owing to electronic delocalization over the conjugated aromatic porphyrin frameworks. Additionally, the incorporation of porphyrins on solid supports not only enhances the chemical stability but also facilitates the full recovery of the catalyst after the photodegradation experiments and prevents the generation of secondary pollution. Therefore, the fabrication of porphyrins with photoactive functional components can facilitate the generation of visible-light nanohybrid photocatalytic systems. Further experiments with porphyrin-conjugated materials are required to optimize their photocatalytic degradation activities. To date, analyses have been restricted to the photocatalytic degradation of toxic contaminants in H₂O. The construction

of a robust porphyrin-conjugated hybrid heterojunction with good visible-light harvesting properties could also be paramount to future research in many important sectors.

Previously, we reported the development of the photocatalytic degradation activity of single-component porphyrin-based photocatalysts derived from the self-assembly of porphyrin molecules for water treatment [59]. Fabrication of solid-supported porphyrin hybrid nanomaterials not only uses solar or artificial light efficiently but also lowers the recombination of photoinduced ion-pairs compared to their starting bare porphyrin unit. Moreover, the hybrid nano-ensembles prevent the hydrolysis of the catalysis during the reaction. On the other hand, this review focuses on porphyrin-conjugated hybrid photocatalysts for water treatment. In this review, the current progress in the fabrication of porphyrin-conjugated hybrid nanomaterials for the catalytic decomposition of toxic pollutants in water is discussed. Throughout, several important results and progress have emerged that enable the development of an improved and significant process for wastewater remediation. Despite this progress, many pivotal aspects of this field remain to be addressed by researchers in materials chemistry. The evolution and utilization of these porphyrin-conjugated hybrid nanomaterials should be explored in greater depth for the remediation of toxic molecules from wastewater, addressing the following points:

1. The recombination of photogenerated electrons and holes remains an obstacle to achieving efficient photodegradation in water. Several techniques, such as metal oxides, inorganic oxides, metal plasmon doping, heterojunctions, and ternary junctions, have been used to minimize this problem. However, more coherent processes are required to improve the photodegradation activities of porphyrin-based photocatalytic systems.
2. The incorporation of porphyrins into solid-supported materials lowers the band gap energy and increases the visible-light harvesting capacity. This reduces the treatment costs. More than 50% of the solar light is infrared. Therefore, more efficient photocatalysts need to be designed such that they are more suitable for the application of solar light as well as infrared light. The effect of the support material (especially those in micro- and macro-scale) on the ability of the porphyrin to absorb electromagnetic radiation. As noticed, the effect of the support material on the quantum efficiency of the supported porphyrin is barely addressed by the studies published in the literature.
3. The light-harvesting capacities of porphyrin-conjugated hybrid nanomaterials compared with those of bare porphyrin should be examined in future studies. This will provide some insight into how the quantum efficiency of photocatalytic systems controls visible-light photodegradation performance.
4. The fabrication of porphyrin on a solid support not only improves the surface area but also creates a large, accessible active site for the substrate or dye molecules. The functional groups of the contaminants can interact with these active sites. This increases substrate transportation and thus the degradation activity.
5. Most photodegradation experiments have been conducted in the laboratory rather than the actual environment. Other compounds or contaminants can perturb photodegradation experiments. Therefore, interference from other compounds must be investigated and developed on an industrial scale.
6. Most pollutants (dyes, pharmaceuticals, and pesticides) discussed in the literature do not represent the actual compounds present in industrial wastewater. A more reliable photodegradation process should be developed using pollutants procured directly from the pharmaceutical, food, and textile industries.
7. Most photodegradation reactions have been investigated at the laboratory scale. Likely, small-scale experiments do not mimic real-life scenarios. Therefore, pho-

todegradation reactions with high degradation efficiency should be investigated on a large scale. This reduces the time and cost of treatment.

8. The toxicity of these porphyrin-conjugated hybrid nanomaterials, as well as the byproducts or secondary pollutants from the photodegradation process, must be properly examined and understood.
9. The recovery and reuse of these hybrid catalysts are important for industrial applications. Therefore, the separation and reuse of catalysts after photodegradation must be optimized to reduce the treatment costs, and the increase in the sustainable aspect of these catalysts must be conducted more effectively since this area covers lines of multidisciplinary research.
10. The chemical stabilities of the hybrid catalysts during the photodegradation experiments are directly related to their efficiencies. Porphyrin molecules may hydrolyze and separate during photodegradation experiments. Therefore, strongly attaching porphyrins to solid supports may improve their photocatalytic performance. In this regard, polymeric backbones or MOF-like materials may be useful in preventing separation and hydrolysis.
11. The porphyrin synthesis procedure is simple for symmetrical molecules. However, the synthesis cost of unsymmetrical molecules is high, and this process is not easy. This increases the fabrication cost of porphyrin-conjugated catalysts. For industrialization, the cost of photocatalysts should be lower. Recently, cost-effective and sustainable materials, including activated carbon, bentonite, biochar, and biomass, have been integrated to enhance the photodegradation activity of hybrid composites and lower the cost of treatment. Therefore, more attention is required to upgrade catalytic photodegradation activity using cost-effective and sustainable components.
12. Generally, for the fabrication of porphyrin-based photocatalyst, the solvothermal method has been used. Conventional heating was the primary energy source for these reactions. However, the microwave-assisted method has been increasingly used for the construction of high-purity materials compared to the solvothermal method [182,183]. Therefore, more attention is required to alter the synthesis method from solvothermal to microwave-assisted in the future.
13. The fabrication of porphyrin on a solid support not only increases the photodegradation performance but also improves the effectiveness of the photocatalyst under extreme conditions, such as highly acidic or basic media, high temperature, and the presence of various salts. Therefore, more attention is required to improve the photocatalytic degradation activity under extreme conditions.
14. The catalytic photodegradation rate of a photocatalyst is believed to be significantly affected by light source characteristics, pH, pollutant concentration, and catalyst content. Therefore, further attention is needed to identify a universal system whose photocatalytic degradation activity is unaffected by these factors.
15. The interpretation of the photodegradation mechanisms of the selected pollutants can be complex, since the pollutants can be absorbed by the support material. Moreover, depending on the type of compound used in the photocatalytic process, several ROS may be at work, making the reaction less selective.

In addition to these factors, studies on the progress of porphyrins, solid-support photocatalysts, and fabrication processes will be the basis for future research. The photodecomposition of other hazardous chemicals using porphyrin-conjugated materials should also be investigated. The distinctive physio-chemical properties of some selected carrier systems are summarized in Table 2.

Table 2. Comparison of some selected carrier systems for the degradation of pollutants in water.

Carrier Systems	Mechanism of Action	Performance Differences and Advantages	Disadvantages and Limitations	Best Degradation Efficiency/Pollutants Treated
Metal oxides	UV light activation generates h^+ / e^- pairs; $OH^\bullet$ radicals oxidize pollutants.	High surface area, high catalytic efficiency, good mechanical/chemical stability, non-toxic, cost-effective synthesis, excellent for dyes.	Primarily UV-active; poor visible light utilization; rapid electron–hole recombination (limits efficiency).	Effective for many dyes, phenols, organophosphorus pesticides (OPPs).
Porphyrins/metall-oporphyrins	Visible light activation generates h^+ / e^- pairs; $O_2^{-\bullet} / OH^\bullet$ radicals oxidize pollutants.	Efficient use of the solar and artificial light spectrum, tailorable structure and functions, assembly in well-defined nanostructures, elevated molar absorptivity, applicable in oxidative and reductive processes.	Burdensome synthesis; fast recombination of photoinduced electron–hole pairs; potential for agglomeration; deactivation-limited recyclability.	Low efficiency for dyes (e.g., MB, RhB), phenols.
Porphyrins/metall-oporphyrins conjugated metal oxides	Visible light activation generates h^+ / e^- pairs; $O_2^{-\bullet}$, $OH^\bullet$, $\bullet OOH$, and singlet oxygen 1O_2 oxidize pollutants.	Tunable structures, large surface area, controllable porosity for pollutant adsorption and catalysis, low catalyst loading, easy catalyst separation, good visible light response, effective for most organic dyes.	Efficiency depends heavily on extreme conditions (high temperature, low or high pH), stability issues in water, and difficult synthesis for unsymmetrical porphyrin.)	Excellent for dyes, pesticides, pharmaceuticals; <90% for complex organics.

Finally, we believe that this organized discussion of porphyrin-conjugated hybrid nanomaterials will help guide materials chemists and encourage progress in photocatalytic wastewater remediation.

Author Contributions: Conceptualization, validation, and writing—original draft, N.K.S.; funding acquisition, project administration, resources, review, and writing—review and editing, H.-J.K. All authors have read and agreed to the published version of the manuscript.

Funding: This study was supported by the National Research Foundation of Korea (grant no. 2022R1F1A1074420), funded by the Korean government (MSIT).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: No new data were created or analyzed in this study. Data sharing is not applicable to this article.

Acknowledgments: We acknowledge the support by the Regional Innovation System & Education (RISE)-(Regional Growth Innovation LAB unit) program through the Gyeongbuk RISE Center.

Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

The following abbreviations are used in this manuscript:

SEM	scanning electron microscopy
TEM	transmission electron microscopy
AOPs	advanced oxidation processes
PXRD	powder X-ray diffraction
DRS	diffuse reflectance spectra

MO	methyl orange
MB	methylene blue
RhB	rhodamine B
4-NP	4-nitrophenol
TC	tetracycline
VB	valence band
CB	conduction band
E _g	Band gap energy
GO	graphene oxide
AA	adipic acid
rGO	reduced graphene oxide
g-C ₃ N ₄	graphitic carbon nitride
H ₂ TCPP	5,10,15,20-tetrakis(4-carboxyphenyl)porphyrin
CuTCPP	[<i>meso</i> -tetrakis(4-carboxyphenyl)porphyrinato]copper(II)
H ₂ TPyP	5,10,15,20-tetrakis(4-pyridyl)porphyrin
ZnTPyP	[5,10,15,20-tetrakis(4-pyridyl)porphyrinato]zinc(II)
ROS	reactive oxygen species
MOF	metal–organic framework
THF	tetrahydrofuran
MeOH	methanol
DCM	dichloromethane
H ₄ TPPS ₄ ^{2−}	[5,10,15,20-tetrakis(sulfonatophenyl)]porphyrin
SnCl ₂ TPP	Dichloro(5,10,15,20-tetraphenylporphyrinato)tin(IV)

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