Journal of Hazardous Materials Advances 10 (2023) 100281

Contents lists available at ScienceDirect

Journal of Hazardous Materials Advances

journal homepage: www.elsevier.com/locate/hazadv

Light-responsive biodegradation of wastewater pollutants: New developments and potential perspectives

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ARTICLE INFO

Keywords:
Biodegradation
Intimate coupling
Microbial fuel cells
Photocatalysis
Photobiocatalysis
Wastewater treatment

ABSTRACT

The existence of micropollutants in wastewater is one of the most challenging environmental issues in the world today. Due to their high stability and resistance to physicochemical and biological degradation, pollutants like hormonally active substances, pesticides, industrial chemicals, pharmaceuticals, personal care products, doping substances, and narcotics among others are difficult to remove in wastewater treatment plants (WWTPs). A potential technology for treating pollutants is photocatalytic biodegradation. The advancements in light-responsive biodegradation technologies—namely, intimately coupled photocatalysis and biodegradation (ICPB), microbial fuel cells (MFCs), and photobiocatalysis are highlighted in this work. The article identifies opportunities for refining current methodologies. It aims to provide a perspective for future research devoted to assessing and improving pollutant removal.

1. Introduction

The increasing contamination of aquatic environments with recalcitrant contaminants like pharmaceutically active compounds, personal care products, industrial chemicals, polycyclic aromatic hydrocarbons, illicit drugs, pesticides, herbicides, and surfactants, has become a new challenge to global water quality. It is difficult to remove these pollutants from wastewater treatment plants (WWTPs). Prolonged and uncontrolled contact with these micropollutants could contaminate surface and groundwater, wreaking havoc on ecosystems and human health. Biocatalysts or enzymes have the potential to degrade environmental pollutants with high substrate specificity and selectivity under mild conditions. However, their large-scale usage is limited by the high expenses of purification and challenges with recycling and renaturation under practical conditions. Photocatalytic processes cannot treat insufficiently transparent water (Al-Nuaim et al., 2022; Deng et al., 2021; Zhang et al., 2021). Also, incomplete photocatalytic reaction products can be toxic, and mineralization can be expensive. Coupling photocatalysis with biotransformation presents an efficient methodology for the economic and sustainable degradation of recalcitrant pollutants (Özgen et al., 2021; Rittmann, 2018; Zhang et al., 2021). These disadvantages motivate the development of photobiocatalytic systems that combine photocatalytic systems with biocatalytic reactions (Harrison et al., 2022; Maciá-Agulló et al., 2015; Schmermund et al., 2019). They combine new reactivity, high enantioselectivity, and better yields with simple and sustainable syntheses as well as recyclability. Till date, only a few known enzymatic reactions require light, and only a selected few exhibit a light-driven promiscuous activity. In most cases, light either provides the co-substrate or cofactor in a suitable redox state for the biotransformation. Other techniques have combined, light-induced chemical reactions with a biocatalytic step, or light-induced biocatalytic reactions with chemical reactions in a linear cascade. This work provides an overview of the approaches that use light for the biodegradation of wastewater pollutants, namely Intimate Coupling of Photocatalysis with Biodegradation (ICPB), Photo Microbial Fuel Cells (PMFCs), and photobiocatalysts.

Under optimal light intensity, the degradation of recalcitrant organic pollutants can be catalyzed by Intimate Coupling of Photocatalysis with Biodegradation (ICPB) (Liu et al., 2022; Zuo et al., 2021). The protected biofilm harbors a substantial capacity to degrade pollutants and lower the levels of chemical oxygen demand (COD) or dissolved organic carbon (DOC). The macroporous substratum, which anchors the photocatalyst on its exterior shelters the biofilm in its interior from Ultraviolet (UV) light and free radicals. The recalcitrant contaminants transformed by photocatalysis are degraded by the bacterial biofilm. Several recalcitrant contaminants, like phenol (Ma et al., 2015; Zhou et al., 2015), pyridine (Shi et al., 2020), phenanthrene (PHE) (Cai et al., 2019), and tetracycline hydrochloride (TCH) (Xiong et al., 2017) have been proven to be effectively degraded by the ICPB system. Considering that most microbes cannot bear UV light irradiation, visible light has been used to induce Intimate coupling of visible light-induced photocatalysis and biodegradation (VPCB) to treat pollutants (Yu et al., 2020; Zhou et al., 2015). A significantly higher removal efficiency of organic pollutants can be achieved in the VPCB than that induced by UV lights. The enhanced activity is attributed to visible light irradiation and enhanced extracellular polymeric substance secretion that protects the microbial communities in the carriers. The VPCB method also has a better potential energy-saving capability.

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https://doi.org/10.1016/j.hazadv.2023.100281
Received 15 January 2023; Received in revised form 8 March 2023; Accepted 20 March 2023

Microbial fuel cells (MFCs) oxidize organic and inorganic compounds while producing electricity (Antolini, 2019). The main strength of MFCs is the use of wastewater as fuel that combines wastewater treatment with power generation. In fact, power generation when MFCs are light-supported is significantly higher than that obtained in dark conditions. For example, Anode-respiring bacteria (ARB) coupled with a nitrogen-doped Titanium dioxide (TiO_2) photocatalyst on a porous carbon foam electrode (the ICPB-anode) when simulated by visible-light illuminated results in an increased current due to photocatalytically generated electrons (photo-electrons) (Zhou et al., 2018). The Coulombic efficiency produced with the ICPB-anode is significantly higher than that generated by a non-photocatalytic biofilm-anode. An abiotic photocatalyst anode cannot transfer photo-electrons. Electrochemical technology has also been utilized to decompose p-nitrophenol (PNP) by MFCs (Zhao and Kong, 2018). In addition, even after long-term functioning, the anodic biofilm was capable of degrading various aromatic compounds, including flubendiamide, chloramphenicol, benzofluorfen, and fluoxastrobin. The microbial genera *Rhodococcus Corynebacterium, Chryseobacterium,* and *Comamonas* dominated the MFC anode biofilm.

The photobiocatalysts utilize light to drive biocatalytic reactions. Combining with chemical reactions, they are used to explore solutions for degrading environmental pollutants. For instance, HRP@g-C_3N_4 nanohybrids formed of horseradish peroxidase (HRP) coupled with Graphitic carbon nitride (g-C_3N_4) benefitted from the photoactivation and exhibited significantly increased activity for the synergistic oxidation of organic pollutants in contaminated water (Bian et al., 2021; Gu et al., 2023). Laccase enzyme supported on copper-doped Hollow titanium dioxide or Lac@Cu-H-TiO_2 degraded almost 95% of 2,4-dichlorophenol (2,4-DCP) in 12 h while maintaining functional stability for up to 10 cycles under visible light (Cao et al., 2022). A photocatalyst/enzyme heterojunction (PEH) manufactured by immobilizing horseradish peroxidase (HRP) on mesoporous graphitic carbon nitride (MCN) enhanced visible light absorption, charge carriers separation efficiency, cycle stability, and bisphenol A (BPA) degradation capability (Zhang et al., 2020). Horseradish peroxidase (HRP) immobilization on Bi_2WO_6 hollow nanospheres via electrostatic self-assembly method enhances visible light harvesting and enables high-efficiency charge carrier separation. The photoenzyme synergic catalytic effect is attributed to the photogenerated electrons and H_2O_2 created on Bi_2WO_6 that directly participate in HRP redox cycle reactions (Dong et al., 2021). Indigo carmin can be efficiently degraded by glucose oxidase immobilized on tannin-coated $NiFe_2O_4$ magnetic nanoparticles in presence of the both UV-light (98.6%) and iron(II)chloride (37.6%) (Atacan et al., 2019). A novel photon-enzyme cascade catalytic system was prepared by hybrid HRP-CN/$Cu_3(PO_4)_2$ nanoflowers (Wu et al., 2022). The immobilized enzyme not only showed efficient degradation of BPA and recyclability, but also was tolerant to extreme pH and thermal changes as compared to the free HRP.

There has been significant progress in the study of light-driven biodegradation of wastewater pollutants in the last five years. However, a systematic and critical perspective of the existing knowledge is still lacking. This work provides a comprehensive summary of the current research in PMFCs, ICPB, and photobiocatalysts for removing wastewater pollutants. In addition, it highlights recent works on the use of Quantum Dots (QDs) in remediation of environmental monitoring. An outlook on future challenges and opportunities in this field is also discussed.

2. Methodologies integrating photocatalysis with biodegradation of wastewater pollutants

2.1. Intimately coupled photocatalysis and biodegradation (ICPB)

ICPB is the most widely studied light-driven biodegradation technique employed for wastewater purification (Yu et al., 2020; Zhang et al., 2022). It can overcome the difficulties of sequential photocatalysis and biodegradation many bio-recalcitrant and bio-inhibitory compounds, including phenol (Dong et al., 2016; Ma et al., 2015; Zhang et al., 2010), trichlorophenol (Li et al., 2011; Liang et al., 2022; Marsolek et al., 2014; Zhang et al., 2012), nitrobenzene (Yang et al., 2015), quinolone (Yan et al., 2013), and dyes (Li et al., 2012). In ICPB, the photocatalyst is attached to the outer surface of macroporous carriers that contain a biofilm inside the macropores (Lu et al., 2022). Photocatalysis yields products on the exterior while the biodegradable products are consumed rapidly by the interior microbes. Inside the carriers, the microbes are protected from UV light, free radicals, and environmental pollutants while decomposing the recalcitrant organic pollutants, increasing the mineralization efficiency and lowering operating expenses (Fig. 1).

The ICPB technology has undergone modifications to increase its effectiveness. For instance, incorporating a biodegradable co-substrate that can act as a source of energy and electron can improve both biotransformation and mineralization. Co-substrate acetate significantly enhanced the treatment of the antibiotic tetracycline (TCH) by ICPB (Xiong et al., 2018). It also increased active biomass, the respiratory activity of biomass, and the selectively enriched bacteria capable of photocatalysis and biotransforming intermediates. The co-substrate sodium acetate also decreased the photo-H_2O_2 damage of the biofilm in ICPB (Zhao et al., 2018). During the degradation of 4-chlorophenol (4CP), the system selected for *Thauera* and *Dechloromonas*. While *Dechloromonas* is efficient in dechlorination, enzymes monooxygenase or double oxygenase enable *Thauera* to break the aromatic ring of 4-CP.

As a component of photocatalyst, TiO_2 is often preferred due to its chemical stability. However, photocatalysts manufactured with nano-TiO_2 are activated by UV light. The wide band gap of TiO_2 limits the use of solar energy (Zhou et al., 2017). It is energy-expensive and has high operational costs. It also affects the microorganisms and diminishes the biodegradation efficiency of the ICPB system. Thus, modified photocatalytic systems capable of induction by visible light are now preferred. For example, the direct band-gap energy of nanoscale Molybdenum disulfide (MoS_2) is 1.96 eV which enables photo-excitation with visible light (Pan et al., 2022; Zhang et al., 2015). Ultrathin CoS_2 nanosheets, an effective visible-light photocatalyst, has the potential to catalyze the induction of quicker interfacial charge separation (Meng et al., 2014). The combined effect of MoS_2 and CoS_2 overcomes the limitations of photocatalysts like TiO_2, $BiVO_4$, and TiO_2/WO_3 that are only photo-excited by UV irradiation (Pan et al., 2022). Other visible-light-activated photocatalysts, like CdS, CuO, and Fe_2O_3 are catalytically efficient, but are prone to photo-corrosion and low reuse abilities. In contrast, the MoS_2/CoS_2 composite with superior catalytic stability accelerates the separation of electrons and holes. $\bullet O_2^-$ and h^+ target sulfamethoxazole (SMX) while e^- reduces Cr(VI), offering an efficient method for improving the removal and mineralization of pollutants via the ICPB technique (Pan et al., 2022). The toxicity of the SMX intermediates and Cr (VI) significantly decreased. The bacteria that are conducive to pollutant removal were enriched by the acclimation and ICPB operation processes, thus significantly improving the performance of the ICPB system.

A TiO_2-coated sponge biofilm carrier can degrade elemental chlorine-free (ECF) bleaching wastewater under visible light (Liang et al., 2021). The removal rate of absorbable organic halogen (AOX), COD, and DOC in wastewater treatment by protocol ICPB also enhanced significantly. Under visible light, upconversion luminescent agent (ULA)-doped photocatalysts, such as Er^{3+}:$YAlO_3$ and Er^{3+}:$YFeO_3$, can coat carriers and achieve TiO_2 photocatalysis (Dong et al., 2016; Ma et al., 2015). Er^{3+}: $YAlO_3/TiO_2$-coated sponge carriers manufactured by low-temperature self-assembly showed improved coating evenness. As a result, the visible-light-responsive photocatalytic circulating-bed bioreactor (VPCBBR) showed enhanced photocatalytic performance (Dong et al., 2016). VPCB conducted with Er^{3+}:$YAlO_3/TiO_2$ photocatalysts prepared at heat-treatment temperatures [HT] yielded more efficient phenol removal capabilities than VPC alone (Ma et al., 2015). The VPCB protocols yielded more efficient phenol removal capabilities than VPC alone. The removal of phenol and dissolved organic carbon

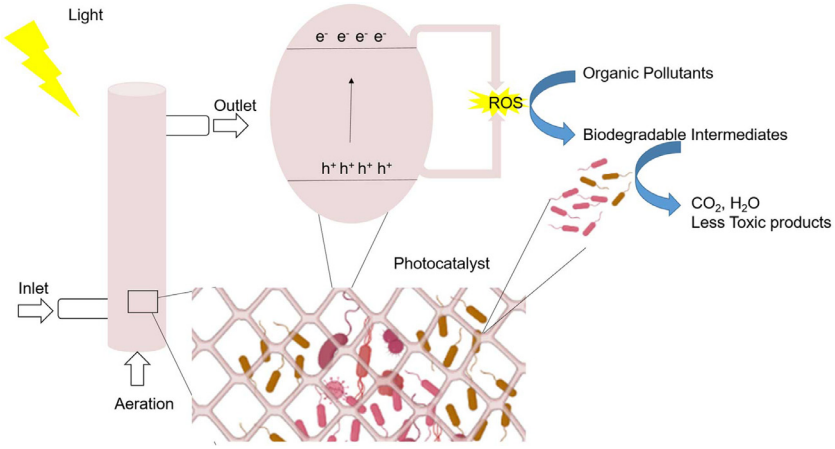

Fig. 1. The synergistic effect of photocatalysis and biodegradation in an intimately coupled photocatalysis and biodegradation (ICPB) system.

from VPCB was found to be the most effective at 130 °C. Because of the lower phenol inhibition, extracellular polymeric substances were not stimulated in VPCB at 130 °C. At this temperature, Dehydrogenase activity, a widely used indicator of microbial activity exhibited the highest decrease. Low levels of Reactive Oxygen Species (ROS) in the interior protected the biofilm while microbes on the carrier surface detached because of high ROS levels. Thus, microbes could self-adjust depending on the exterior environment while enhancing photocatalytic capability and degradation efficiency. The phenol and DOC removal efficiencies using VPCB were higher than those achieved by catalyst-coated carriers without the biofilm in dark, visible-light photocatalysis, biofilm, and catalyst-coated carriers in the dark. An Ag–TiO_{2-x} nanosheet photocatalyst can be combined with the exterior of a macroporous support, and a biofilm within in a bifunctional nest-like self-floating microreactor (Chi et al., 2019). The nanosheets raise the reactor's internal temperature, consequently increasing the activity of the microorganisms. During photocatalytic biodegradation of cephalosporin at low temperatures under solar light irradiation, the photocatalyst biomicroreactor exhibits recyclability and good cycle stability. Among visible-light-driven photocatalysts, graphitic carbon nitride (g-C_3N_4) is an inexpensive, nontoxic, metal-free photocatalyst with a visible-light-driven bandgap that is stable in aqueous solutions and is simple to manufacture. However, g-C_3N_4 has a low photocatalytic efficiency. It can be enhanced by ferric ion doping. Since Fe is affordable and environment-friendly, an ICPB system can be manufactured by coupling Fe^{3+} doped g-C_3N_4 powder (Fe^{3+}/ g-C_3N_4) with the biofilm carrier (Liu et al., 2022). The system functions better than single photocatalysis of Fe^{3+}/ g-C_3N_4 or single biodegradation of sulfamethoxazole (SMX) and COD. In a similar study, visible light-responsive g-C_3N_4 were used to modify cube polyurethane sponges. In presence of mixed culture microbes acquired from swine wastewater, the wastewater treatment system increased the removal efficiency of ciprofloxacin and total organic carbon (TOC) (Li et al., 2021). When illuminated by visible light, the Mn_3O_4/MnO_2-cubic Ag_3PO_4 with exposed {100} facets outperformed Ag_3PO_4 in terms of the photocatalytic degradation of phenanthrene (PHE) (Cai et al., 2019). The genera *Shewanella, Sedimentibacter, Comamonas, Acinetobacter*, and *Pseudomonas,* commonly found in the bioremediation of organic pollutants acclimatized themselves in the biofilms of the interior of VPCB sponge. ICPB systems have successfully used photocatalytic optical hollow-fibers (POHFs) coated with Ag-loaded GeO_2, N-doped TiO_2 photocatalyst, and a biofilm harboring phototrophic and heterotrophic microbes (Zhong et al., 2021). Under UV–visible irradiation, the system exhibited stability while catalyzing rapid photocatalytic degradation of phenolic compounds and generating less toxic, readily biodegradable products. It also emitted visible light that supported biofilm growth and enabled phototrophs to produce oxygen. The transfer of O_2 from the phototrophs to the POHFs assisted in the production of hydroxyl free radicals needed for photocatalysis. Enrichment of *Pseudomonas* and *Rhodococcus* in the biofilm assisted in the fast biodegradation of photocatalytic products.

Coupled microbial extracellular electron transfer (EET) and photoelectrochemical technique involves synchronous oxidation of extracellular electron donors by microbes for respiration and reduction of extracellular electron acceptors to form an integrated respiratory chain (Dong et al., 2020). Hybrid microbial-photoelectrochemical processes can be promoted by Cadmium sulfide nanoparticles (CdS NPs) that are potential biocompatible visible-light-driven photocatalysts with efficient electrical conductivity and a large specific surface area. Under solar light irradiation, the CdS nanoparticles (NPs)-assisted periphyton bioelectrochemical system (PCdS-BES) are capable of reducing nitrate to gaseous nitrogen while avoiding nitrous oxide emission (Zhu et al., 2018). In addition, CdS nanoparticles selectively enriched electroactive bacteria strains in the periphyton. Increased extracellular polymeric substance (EPS) production aided CdS stability in the periphyton matrix. Also, it enabled electroactive bacteria to acquire electrons from CdS NPs, and catalyze nitrate reduction. Interestingly, nitrate can function as an electron acceptor that has the potential to enhance the anoxic biodegradation of recalcitrant contaminants. Under anoxic conditions, nitrate accepts electrons at the semiconductor/biofilm interface, and promotes tetracycline (TC) biodegradation (Hou et al., 2022). The reduction in the toxicity of intermediate products promotes microbial activity. In particular, it selectively enriches species mediating electron transfer (*Delftia*), denitrification (*Thauera*), and TC biodegradation (*Rhodopseudomonas,* Stenotrophomonas*,* and *Phreatobacter*) at Ag-TiO_2/biofilm/nitrate interface. Combining chemical pre-oxidation with biological post-treatment can produce easily biodegradable intermediates. A combined photocatalytic pre-oxidation reactor (PPOR) and sequencing batch bioreactor (SBBR) have proven to be efficient for wastewater treatment (He et al., 2020). The system included a $K_2S_2O_8$-doped-TiO_2 photocatalyst that showed high photoactivity and was used to oxidize the pollutants in the photocatalytic step. As a post-biological treatment SBBR ensured COD levels of the treated water met emission standards. Compared with seed sludge, bacterial genera capable of degrading recalcitrant pollutants, such as *Rhodobacter, Defluviimonas, Dokdonella*, and *Thauera* were enriched in active sludge from the SBBR.

A novel coupling system, Simultaneous coupling of memory photocatalysts and microbial communities (SCMPMC) can present an efficient opportunity for the synergistic removal of microbial nitrate and CO_2 fixation in darkness (Zhou et al., 2022). In SCMPMC, g-C_3N_4 has been applied to the surface of WO_3 nanoparticles to create a novel core/shell-structured g-C_3N_4@WO_3 memory photocatalyst. The memory photocatalyst releases stored electrons in the dark and accelerated key enzyme activities such as nitrate reductases (NAR), nitrite reductase (NIR), catalase (CAT), and electron transport system activity (ETSA).

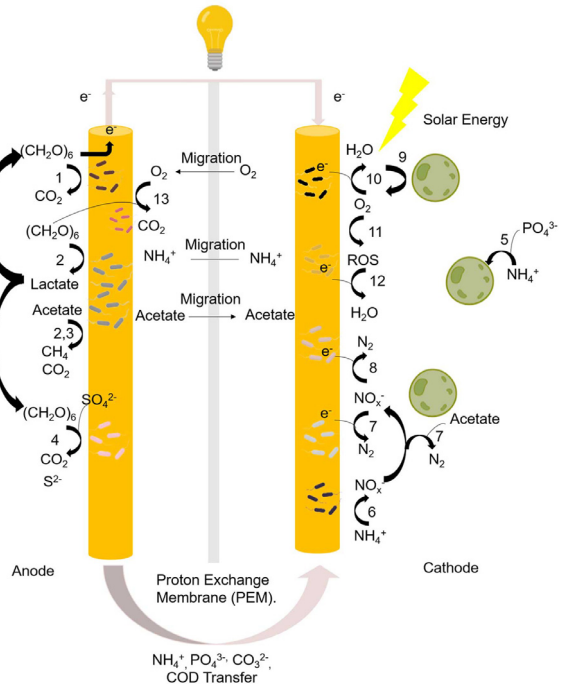

Fig. 2. Diagram showing the mechanism of PMFC for degradation of pollutants (after Wang et al., 2019). 1: Electric generation, 2: Fermentation, 3: Methanogenesis, 4: Sulfate reduction, 5: Algal uptake, 6: Nitrification, 7: Aerobic denitrification, 8: Autotrophic denitrification, 9: Photosynthetic Oxygen, 10: Oxygen biocatalysis, 11: ROS generation, 12: ROS reduction, 13: COD removal by aerobic microbes
Removal of C: 1: Electric generation, 2: Fermentation, 3: Methanogenesis, 4: Sulfate reduction, 13: COD removal by aerobic microbes
Removal of Nitrogen: 5: Algal uptake, 6: Nitrification, 7″ Aerobic denitrification, 8: Autotrophic denitrification
Removal of P: 5: Algal uptake.

The photocatalyst was efficient in alternately removing dimethyl phthalate (DMP) and nitrate under (12/12 h) light/dark cycles. Within one cycle, photogenerated electrons stored in the memory photocatalysts enhanced nitrate reduction by microbes during the dark phase. DMP was efficiently removed via robust photocatalytic oxidation during the light phase. WO_3/g-C_3N_4 increased enzyme activities as well as the relative abundance of denitrifying bacteria like Proteobacteria and Bacteroidetes.

2.2. *Photocatalytic microbial fuel cells*

Fig. 2 depicts the working mechanism of a photocatalytic microbial fuel cell. As an example, a two-chambered model shown here comprises an anode, a cathode, and an ion-exchange membrane that separates the two compartments. The photogenerated electrons flow from the anode to the cathode through the connecting wire to generate electrical energy. Reduction and oxidation reactions occur at the cathode and anode respectively. As a result, both pollution control and energy generation are managed simultaneously (Tong et al., 2022). As an example, heavy metals with high redox potential can be used as effective electron acceptors instead of oxygen in the cathodic chamber of microbial fuel cells. Photocatalysts with a large band gap can capture only 5% of the solar irradiation. The use of visible light as an alternative to UV light saves energy and prevents damage to the biofilm. Their efficiency can be enhanced by doping photocatalysts with suitable components like Cd and Se, which have a type II band alignment with TiO_2. The preparation of TiO_2 with CdSe targeted changing the band-gap of TiO_2 while remaining in the UV region and increasing the catalytic activity stable photocurrent density (Caglar et al., 2022c). The addition of CdS into TiO_2 by a wetness impregnation (WI) method demonstrates that it can significantly increase the absorption of UV light for catalytic activity (Caglar et al., 2022b). In Methylene blue oxidation tests, 0.5% Cd/TiO_2 demonstrated the highest catalytic activity and stability in the dark and under UV illumination (Kivrak et al., 2021). 0.1% Cd/TiO_2 catalyst exhibited the highest stability and the lowest resistance while demonstrating the best photocatalytic glucose electro-oxidation activity (Caglar et al., 2021). In a similar work, the photocatalytic glucose electrooxidation results indicated that the 0.1% CdTe(50–50)/TiO_2 catalyst demonstrated significantly greater photocatalytic activity, stability, and resistance than other catalysts both in the dark and under UV illumination (Caglar et al., 2022a). TiO_2 can also be sensitized with nanocrystal quantum dots for photovoltaic applications (Markna and Rathod, 2022). These structures are beneficial in having thermal and photo stability, and large absorption cross-sections. White commercial TiO_2 (w-TiO_2) can be modified by introducing defects to a light gray color (g-TiO_2), thereby increasing absorbance in the visible and near-infrared regions and improving visible-light-induced photocatalytic and photoelectrochemical performances (Khan et al., 2018).

Photocatalytic microbial fuel cells are being studied for their effective use to remove pollutants (He et al., 2018; Li et al., 2019, 2009; Long et al., 2017; Ren et al., 2018; Wang et al., 2022, 2018, 2019a, 2019b; Zhang et al., 2019; Zhu et al., 2021). Du et al., 2014 investigated the degradation of methyl orange within the anode chamber using a photoelectric anode-biocathode (Du et al., 2014). They suggested the use of a biocathode-photocatalytic fuel cell with a nitrifying biocathode. The terminal electron acceptor in the cathode was oxygen, which was reduced to H_2O. The C-type cytochrome of electrochemically active bacteria in biocathodes can receive electrons from the electrode and transfer them to electron acceptors with greater affinity for electrons, such as O_2, in the periplasm and inner membrane. Ammonia nitrification simultaneously occurred in the cathode chamber. Nitrification reaction and O_2 reduction showed synergistic and competitive interaction in the cathode chamber. O_2 reduction was dependent on microbial activity that resulted in higher catalytic efficiency. Reduction of O_2 helped to maintain a proper pH essential for nitrification. The competition for O_2 also existed in the biocathode. However, the concentration of dissolved oxygen was enough for both the cathodic reaction and nitrification. Biocathode microbes can derive electrons from NH_4^+ oxidation as well as from the external circuit. A passive cathode limits the efficiency of a Photocatalytic Fuel Cell as the competence depends entirely on the photoanode. Here, biocathode was not a passive electron consumer, but also participated in nitrification. Thus, the active biocathode functioned as a central player to beat the efficiency bottleneck (Du et al., 2014; Li et al., 2019).

Rutile can function as a cathodic catalyst instead of noble metals. It can also generate considerable power output and reduce Cr(VI) (Li et al., 2009). Li et al., 2019 investigated Cr^{6+}removal by a synergism between a bioanode and the rutile-coated graphite photocathode in PMFCs. It can not only promote Cr(VI) reduction in the cathode chamber but also power output. He et al., 2018 investigated the efficiency of coupled photoelectro-catalysis and microbial fuel cell on the removal of contaminants in wastewater. Photo-electro-catalytic electrode integrated with the bioanode, effectively reduced Cr(VI) in the cathode chamber and rhodamine B (RhB) concentration in the sand (He et al., 2018). Thus, PMFCs can be adapted for decontaminating polluted sands near riverside, coastal areas, and seashore wetland sites. At present, studies related to the use of PMFCs in remediating complex wastewater is limiting (Table 1). One of the studies highlights the use of PMFCs with the intimately coupled photocatalytic-electrogenic anode in degrading recalcitrant contaminants like 2,4,6-trichlorophenol (2,4,6-TCP) in municipal sewage (Wang et al., 2019). The photocatalytic-electrogenic anode, coated with visible-light-induced photocatalyst mpg-C_3N_4 on a carbon-felt anode degrades TCP. TCP degradation is higher when exposed to visible light in PMFC compared to unilluminated MFC and the photocatalytic-only process (Wang et al., 2019). In future, studies related to the understanding of the dynamics of com-

Table 1
Effect of photo catalytically coupled microbial fuel cell systems (PMFC) used for pollutant removal from wastewater.

Setup	Remediation	Refs.
Electrodes: Photocatalytic air-cathode made from gas diffusion layer (PDMS layer), cathode electron collector (stainless steel mesh), catalyst layer (carbon materials) and photocatalytic layer (N-TiO_2), Carbon cloth anode A single chamber air-cathode MFC Light: visible range Wastewater: MFCs inoculated with municipal wastewater from Tangxun Lake wastewater treatment plant (Wuhan, China)	Pollutants removal: 50 mg L^{-1} TCP completely degraded within 72 h, rate constant higher than MFC with air cathode and without N-TiO_2 photocatalyst. Different degradation pathways compared to Wang et al. (2019) due to unique intermediates (2-CHQ and 2,6-DCQ), thus simultaneously improving the electricity generation. 6-CHQ and 2-CHQ can be oxidized to easily biodegradable intermediates (2-CMA and hexanoic acid) and eventually to CO_2 and H_2O. Bacterial composition: Enriched exoelectrogen (55.2% of *Geobacter*), and TCP-degrading microbe (7.1% of *Thauera*) on the cathode biofilm. 61.8% of *Pseudomonas* in the culture solution. Power Generation: Stable electricity generation of 350 mV obtained during 130 days of operation.	Zhu et al. (2021)
Electrodes: mpg-C_3N_4 photocatalytic-electrogenic anode air-cathode, single chamber MFC Light: 420–780 nm. Intensity of the incident light controlled at 1500 lx Wastewater: Municipal wastewater from Tangxun Lake wastewater treatment plant (Wuhan, China) was used for MFC inoculation	Pollutants removal: 79.3% of TCP removed within 10 h with an original concentration of 200 mg L^{-1}. Removal percentage higher than that obtained by unilluminated MFC (66.0%) or photocatalytic-only systems (56.1%). Bacterial composition: *Pseudomonas* (not *Geobacter*) in the unilluminated MFC bioanode, *Pseudomonas* in the photocatalytic-electrogenic anode MFC biofilm. Considered responsible for greater current generation in the coupled system. *Rhodococcus*- rich biofilm on air-cathode, responsible for better TCP removal. *Pseudomonas* enhanced TCP degradation in presence of sodium acetate Power Generation: Higher electrochemical performance mediated by the coupled system compared to non- photocatalysis MFCs. Future Goals: Long cycle for complete TCP degradation. The process would be practical with shorter cycles.	Wang et al. (2019)

plex wastewater treatment using suitable electrodes open up the avenue for the large scale use of PMFCs for preventing environmental pollution.

2.3. Photobiocatalysts

Different photo-enzyme-coupled catalysts have been reported to participate in degradation of pollutants like Aniline blue, crystal violet, 2,4-DCP, reactive red X-3B dye, bisphenol A (BPA), phenol, p-nitrophenol, 2-naphthol, and methyl orange (MO) (Bian et al., 2021; Cao et al., 2022; Cheng et al., 2018; Dong et al., 2021; Gu et al., 2023; Ji et al., 2016; Li et al., 2018a, 2017, 2018b; Wu et al., 2022; Zhang et al., 2020a, 2020b). The primary goals of designing the photobiocatalysts is to overcome photo-corrosion while promoting enhanced charge migration and transfer, stability, activity, recyclability along with thermal resistance. One of the reasons for the increased performance efficiency is direct participation of the photogenerated electrons in the redox cycle reactions (Fig. 3). Gu et al. (2023) summarize the latest progress in graphite carbon nitride (g-C_3N_4)-based photobiocatalysts for decomposing organic pollutants.

Recent reports show that exfoliating pristine bulk carbon nitride (g-C_3N_4) into ultrathin structures improves photoactivity by active site exposure and optimizing mass transfer. Ultrathin carbon nitride nanosheets with nitrogen vacancies function as metal-free and robust host support for enzyme immobilization. It also provides an *in-situ* technique for horseradish peroxidase (HRP) activation by forming peroxides and radicals (Bian et al., 2021). HRP@g-C_3N_4_HN photobiocatalysts are effective at removing methyl orange (MO), methylene blue (MB), and bisphenol A (BPA) from water. The synergy between bioenzymes and polymeric photocatalysts resulted in enzymatic stability and catalytic activity. Peak intensity of g-C_3N_4_HN is stronger in the ultrathin nanosheets than in bulk g-C_3N_4 due to structural defects in the former. This defect causes enhanced charge migration and transfer and is beneficial for the activating the biohybrid interface. The photo-enzyme cascade consisting of HRP and g-C_3N_4 efficiently uses solar energy, does not require additional H_2O_2 or yield toxic intermediates. In the

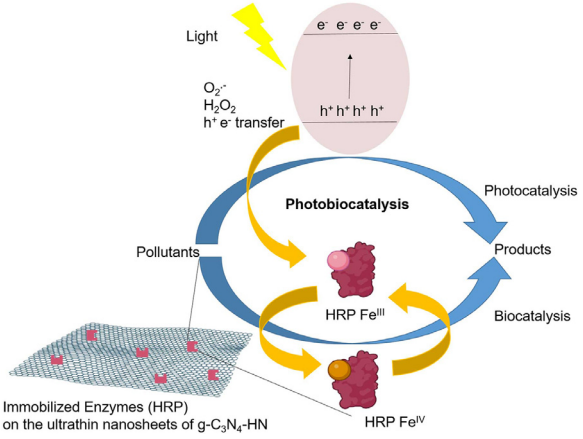

Fig. 3. Diagram showing the mechanism of photobiocatalysts for synergistic water decontamination. (after Bian et al., 2021).

synergistic system, g-C_3N_4 increased the loading of HRP, accelerated the electron transport of g-C_3N_4, and enhanced its enzymatic activity (Bian et al., 2021). The g-C_3N_4-based photocatalyst ACN_{45} was synthesized such that the microenvironment supplied by ACN_{45} channels reduced the leaching of the immobilized enzyme and enhanced its stability and activity (Wu et al., 2023). Internal immobilization of HRP did not impair the absorption of visible light by the photocatalyst. A high loading amount of the system significantly improved the photocatalytic degradation of BPA than that of pure g-C_3N_4. Encapsulated HRP inside nanochambers of TiO_2-doped hollow nanofibers avoided UV-induced deactivation of the enzymes (Ji et al., 2016). The efficacy of 2,4-DCP degradation was greatly enhanced when compared to free HRP or TiO_2/UV either individually or simultaneously. The hybrid catalysts system also demonstrated excellent recycling and thermal resistance. Polydopamine (PDA) tethered chloroperoxidase (CPO)/HRP-TiO_2 nanocomposites had high catalytic activity, stability under diverse severe re-

action conditions, and good recyclability in the decolorization of soluble dyes (Cheng et al., 2018). TiO_2 functioned both as a solid carrier and a photo-catalyst. Organophosphorus hydrolase (OPH) immobilized on BiOBr (OPH@BiOBr) was synthesized to degrade the methyl parathion under visible light (Jiang et al., 2020). OPH@BiOBr could hydrolyze the P-S bond in methyl parathion into p-nitrophenol (p-NP), which could then be photocatalyzed into a less toxic molecule. The nanocatalyst's unique flower-like structure could not only enhance photocatalytic activity but also prevent enzyme degradation. The OPH@BiOBr could be recycled five times while retaining 83% of the original activity. Studies related to the degradation of pollutants in presence of natural organic matter (NOM) are very rare. Gong et al., 2020 studied acetaminophen (AAP) degradation in presence of NOM by HRP in sunlit water. Depending on their aromaticity and average molecular weights, NOM can successfully accelerate the degradation rate of AAP. NOMs with low average molecular weight and minimal aromaticity are more efficient. The inhibition rate constants are positively correlated with the amount of H_2O_2 photogenerated by NOM. The generation of H_2O_2 by NOM under simulated sunlight has been suggested to be accompanied by a change in the structure of NOM itself (Gong et al., 2020).

3. Conclusion and future perspectives

(a) Challenges in commercialization of the technology:

The commercial viability of photocatalytic materials in biodegradation is still challenging because of difficulties in economic mass production, purification, activity, stability, and identifying transformation products during the light irradiance in complex "real" wastewater. Sufficient knowledge related to the influence of micro- and nanoplastics, plasticizers, and other hazardous compounds in contaminant degradation is still lacking. The majority of studies are focused on synthetic wastewater with a single pollutant to understand the wastewater remediation dynamics. The choice of economic yet stable photocatalysts is crucial to understand the degradation processes in actual WWTPs where complex interactions among photocatalysts, pollutants, and biocatalysts occur. It remains to be deciphered how the co-presence of pollutants modulates the functioning of the light-driven catalysis. For instance, phenol addition increased the efficiency of 4-chlorophenol (4CP) degradation (Zhang et al., 2017). Degradation of toxic products increased the ratio of living/dead cells. However, pyrocatechol decreased 4CP degradation and the ratio of living/dead cells. Photodegraded products of phenol provided extra electron donors to the bacteria and enhanced 4CP degradation. Meanwhile, easily photo-oxidized pyrocatechol competes with 4CP for ROS, thus hindering its degradation (Zhang et al., 2017). In another work, H_2O_2 had a detrimental effect on photocatalysis by ZnO but was beneficial or disadvantageous for TiO_2 depending on the H_2O_2/1,2-dimethoxybenzene ratio (Maciá-Agulló et al., 2015). In contrast, superoxide dismutase inhibited the photocatalytic efficiency of both TiO_2 and ZnO, supporting the essential role of $O_2^{\cdot -}$ as the active ROS promoting the disappearance of 1,2-dimethoxybenzene. Such complex interactions in WWTPs, in addition to factors like the aging effect of catalysts, need to be studied to enhance the efficiency of photobiocatalytic systems.

(b) Standardizing operational parameters:

Various operational parameters including light source and intensity, coating ratio of photocatalysts, carriers, co-substrate or use of any additional matrix for support, immobilization strategies (Zdarta et al., 2022), and reactor structure are still being optimized to increase biocatalyst lifetime and reusability for future work (Chanquia et al., 2022). For example, solar simulators have been developed for use in photocatalytic disinfection applications with simultaneous removal of emerging contaminants (Philippe et al., 2016). In addition, the following technologies can integrated when purifying wastewater in WWTPs.

- A powder spraying method which adheres the photocatalyst to the carrier exterior while reserving the inside pore structures for biofilm accumulation has been developed (Li et al., 2020). The modification technique enhanced the degradation rate of tetracycline hydrochloride and maintained the stability of recycled carrier under visible light illumination even after six cycles of operation. The adhesion of photocatalysts to the carrier can be increased by the coupling agent γ-glycidoxypropyltrimethoxysilane (KH560) (Fu et al., 2021). The novel ICPB system maintained stability even after 5 cycles of operation. The photocatalytic removal competencies of geosmin (GSM) and 2-methylisoborneol (2-MIB) remained high. The enriched functional microbial populations, *Flavobacterium* and *Thauera* effectively biodegraded the photocatalytic intermediates (Fu et al., 2021).
- Often the sequential coupling of photocatalytic and biological system is not ideal due to the difficulty in controlling the formation of specific photocatalytic products (Zhang et al., 2021). In particular, insufficient and excessive photocatalytic processing can produce toxic and less biodegradable intermediates. It can waste photogenerated reactive species and the energy to produce over-oxidized recalcitrant products. Combining photocatalysis and biotransformation simultaneously in a single system can overcome these challenges because microorganisms can quickly digest biodegradable intermediates derived from photocatalysis (Zhang et al., 2021).
- To drive solar-driven catalysis, efforts are being organized to use carbon nanodots (CNDs) as photosensitizers in biological systems as they are photostable and nontoxic, water-soluble compounds with easily modifiable surface chemistry (Hutton et al., 2016). Electron transfer from a photosensitizer can activate biomolecular catalysts via energy transfer, hydrogen abstraction, and electron transfer (Michelin and Hoffmann, 2018). In future, the observations can be extended to remediate wastewater pollutants. A new solar-driven biological wastewater treatment (SDBWT) technology can use photosensitizers to capture energy from light as a driving force for biological wastewater treatment. The synergy of photosensitizers and non-photosynthetic bacteria under visible light, enables accelerated electron transfer to enhance biodegradation efficiency and improvement of the energy recovery efficiency from wastewater treatment process (Du et al., 2020). The spatial connection between non-photosynthetic bacteria and electron acceptors can be improved further by ensuring suitable electrical conductivity of some photosensitizers.
- Higher current density and columbic efficiency can be generated by an electrochemical cell harboring anode-respiring bacteria (ARB) coupled with a nitrogen-doped TiO_2 photocatalyst on an illuminated porous carbon foam ICPB-anode. The efficiency is higher compared to either the bio-anode or the photo-anode cell. The better competence results from the biofilm's ability to conduct photocatalytically generated electrons through the extracellular electron carriers of the biofilm matrix (Zhou et al., 2018). The system can be modified further to catalyze the breakdown of refractory pollutants.
- Semiconductor TiO_2 can be used to manufactures photocatalytic biosensors that can evaluate biochemical oxygen demand (BOD) levels in water and not be affected by free radicals yielded by photocatalysis. Biosensor coupled to the photocatalytic biodegradation system can enable Biosensor-based real-time monitoring of pollutant photocatalytic degradation (Calas-Blanchard et al., 2015).
- Quorum-Quenching (QQ) biofilm can be engineered to senses near-infrared (NIR) light and blue light and adjust its growth by controlling the c-di-GMP level and mitigate biofouling of water purification forward osmosis membranes (Mukherjee et al., 2018). The technology can be extended to design biofilm-based photobiocatalytic applications in wastewater treatment.
- A flower-like S-scheme Bi_2WO_6/BiOCl nano-heterojunction can be manufactured by a one-pot hydrothermal method. Under visible light, it displays an efficient photocatalytic activity and RhB degra-

dation rate (Zheng and Sun, 2022). The degradation activity is higher than those of BiOCl and Bi_2WO_6. Triboelectric nanogenerators (TENGs) have extensive microscale energy harvesting capabilities that can be utilized for photo/electric catalysis and increase the removal efficiency of pollutants (Dong et al., 2022).

(c) Role of microorganisms:

Microorganisms and photocatalysts can interact in diverse ways. Photacatalysts can damage microorganisms and sterilize them; they can generate more energy or degrade pollutants more efficiently in synergy with microorganisms; microorganisms can assist in the manufacture of photocatalytic system (Deng et al., 2020). Thus, it is essential to have an understanding of microbial eco-evolutionary processes driven by the interaction between evolving cells and environmentally driven selection in wastewater. For instance, Xue et al., 2019 studied that microorganisms are the main contributor to SMX degradation in MFCs. *Shewanella* sp. and *Geobacteria* sp. have a key role in power production while *Alcaligenes, Pseudomonas* and *Achromobacter* contributes to the degradation of SMX (Xue et al., 2019). Interestingly, the abundance of antibiotic resistance genes (ARGs) in MFC biofilms and effluent was observed to be lower than that in conventional bioreactors, WWTPs, anaerobic reactors, and natural waters. This suggests that MFCs may be more effective than conventional biological processes to treat wastewater. Integrons aid ARG transmission bacterial evolution by driving new resistant and pathogenic species. Xue et al., 2019 suggested that a decrease in integrons inhibited horizontal transfer and the ARGs evolution in MFC. It will be important to use the functional microbial diversity in WWTPS in ensuring better performing light-responsive biodegradation of pollutants.

(d) Designing novel photobiocatalytic systems

Immobilized Enzymes have great promise for improving wastewater purification either as single copies or as components of a multienzyme cascade. Benefits of immobilizing single enzymes can include increased stability, close proximity and orientation to the substrate, reaction rates and recoverability, bypassed off-target pathways, and toxic byproducts (Ellis et al., 2020). *De novo* cascades can be designed by linking enzymes that do not associate in nature. Factors like Scaffold Size, Scaffold: Enzyme Ratio, and Scaffold Aggregation can be exploited to ensure immobilization benefits in large-scale processes.

Novel Photocatalytic Material–Microbe Hybrid (PMH) systems composed of microbial cells and photocatalysts coated directly on bacterial surfaces are now being designed for minimizing environmental pollution (Yu et al., 2022). Electrochemically active bacteria supply biogenic electrons to eliminate the photogenerated holes on the photocatalysts and drive reductive degradation of oxidative organic pollutants like nitroaromatic compounds and azo dyes under anaerobic conditions. Photocatalysts yield electrons that can be transferred to microbes to promote intracellular enzymatic reductions of heavy metal ions like Se^{4+} and Cr^{6+}. On the other hand, photogenerated holes can oxidize heavy metal ions like Sb^{3+} and As^{3+}.

(e) Decreasing the dependence on mediators:

The efficacy of the photobiocatalytic mechanism is heavily reliant on mediators. However, most of the mediators used to transfer the electrons between the photocatalysts and the cofactor or the enzyme contain noble metals either as coordination or organometallic complexes, thus limiting the practical applicability of the system. This dependence of catalysts on mediators or cofactors can decrease if the enzyme is supported on morphologically defined photocatalysts like TiO_2 NPs, which allows electron migration between the active center and the photocatalyst (Maciá-Agulló et al., 2015). While being improved, transient absorption spectroscopy can be used as a routine technique for photochemical characterization in photocatalytic systems (Friedmann, 2022).

(f) Quantum Dot (QDs) Photocatalysts

QDs have significant potential as remediators of hazardous pollutants (Jouyandeh et al., 2021). Due to a higher surface area-to-volume ratio and lower recombination of the electron–hole pair than their bulk counterparts, QDs can function as efficient photocatalysts. Surface alteration can change their chemical, optical, and photocatalytic characteristics for enhanced pollutant degradation. It is possible to design the catalyst–substrate interface of QDs without compromising their redox potential. In addition, Colloidal QD catalysts also aid their simple removal from the reaction solution system. When a QD is exposed to light, an electron is stimulated to the conduction band, creating a positively charged hole in the original valence band. These photogenerated charge carriers can move to the QD surface and catalyze redox reactions using electrons and holes. Nanocrystal quantum dots can also be used with various metals to sensitize TiO2. They have advantages such as thermal stability, resistance to photodegradation, and large absorption cross-sections. Carbon quantum dots (CQDs) are appealing due to their visible-light absorption, strong photoresponsiveness, redox properties, stability, nontoxic characteristics, tunable photoluminescence (PL), upconverted photoluminescence (UCPL), and efficient photo-excited electron transfer. Thus, QDs that are water soluble and can be modified for further functionalization are good candidates for wastewater purification (Basavaraj et al., 2021; Jung et al., 2022). Thus, QDs have immense potential to function in the production of photobiocatalysis. The CQDs can be produced by eco-friendly methodologies. In this contribution, the two major beer wastes, spent grains and spent yeasts, have been used to yield carbon dots (CDs). The CDs entrapped into a polyvinyl alcohol matrix can adsorb methylene blue and be efficiently removed from water solutions and subsequently fully degraded by UV irradiation (Cailotto et al., 2022). At ambient temperature and neutral pH functionalized nitrogen-doped carbon quantum dots (NCQDs) degraded methyl green (MG) dye solution efficiently under UV-light exposure. In a similar study, empty fruit bunches (EFB) biomass were observed to be a viable option for the production of nitrogen-doped carbon quantum dots (NCQDs). They demonstrated exceptional durability and reactivity even after a year of storage (Abd Rani et al., 2022). They degraded 97% and 98% of methylene blue and malachite green upon sunlight irradiation, respectively.

Carbon dots (CDs) have been found to be efficient photosensitizers for photobiocatalysis in two independent systems (Hutton et al., 2016). Optical spectroscopy monitored the photoreduction of the four Fe^{3+}-heme cofactors in fumarate reductase (FccA) from *Shewanella oneidensis* MR-1. Photoexcited CDs transfered electrons to FccA during the photoconversion of fumarate to succinate. In another study, CDs and a [NiFeSe]-H_2ase from *Desulfomicrobium baculatum* (Dmb) catalyzed the reduction of protons to H_2. Exploring the potential of QDs as drivers of photobiocatalytic degradation of pollutants in real wastewater treatment is a future avenue of work. Additional research is being carried out toxic ions may be emitted from QDs during corrosion (Hardman, 2006; Nikazar et al., 2020). For example, the Cd-core QDs were found to be cytotoxic, particularly when surface oxidation from air or UV exposure causes the release of free Cd^{2+} ions (Wang et al., 2008). Manipulating factors like particle size and charge and the surface coating bioactivity (capping substance and functional groups) can help design less toxic QDs.

Funding sources

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

Declaration of Competing Interest

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

Data Availability

No data was used for the research described in the article.

Acknowledgments

The author acknowledges the support of Sammilani Mahavidylaya, University of Calcutta for creating a productive work environment.

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