INTERNATIONAL JOURNAL OF NOVEL RESEARCH AND DEVELOPMENT (IJNRD)
© 2026 IJNRD | Volume 11, Issue 5, May 2026 | ISSN: 2456-4184 | IJNRD.ORG

Recent Advances in Photocatalytic Degradation of Chlorpyrifos and Mancozeb Pesticides: Mechanisms, Catalysts, and Environmental Applications

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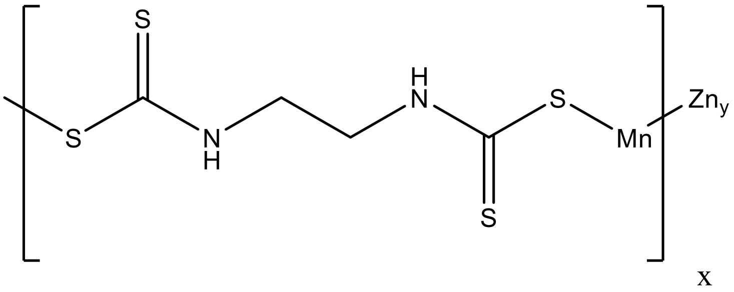
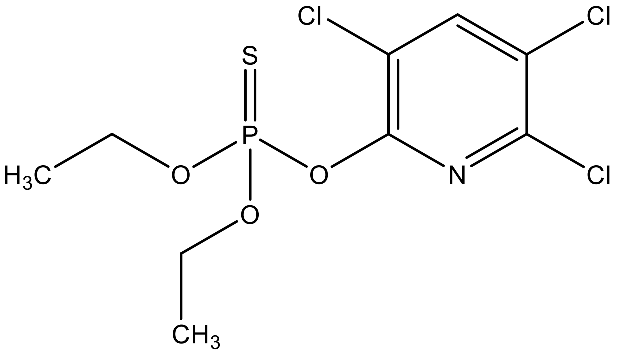
Introduction

The escalating global demand for food production has led to a pervasive reliance on pesticides, such as chlorpyrifos and mancozeb, to safeguard agricultural yields from pests and diseases. However, the extensive application and persistent nature of these agrochemicals pose significant environmental and health risks, necessitating effective remediation strategies [1]. In response to this critical challenge, photocatalytic degradation has emerged as a promising and sustainable technological approach for detoxifying pesticide-contaminated ecosystems, leveraging the intrinsic properties of semiconductor photocatalysts [2]. This method leverages advanced oxidation processes to convert harmful organic pollutants into simpler, less toxic compounds [3]. Specifically, metal oxide nanoparticles and their composites, characterized by their unique physicochemical properties, crystalline structures, and high surface areas, have garnered considerable attention for their efficacy in degrading such organic contaminants [4]. For instance, titanium dioxide and zinc oxide nanoparticles have demonstrated considerable potential as photocatalytic agents for the remediation of pesticides like chlorpyrifos and atrazine, with TiO2 often exhibiting superior degradation efficiencies and kinetic rates [5]. A wide range of metal oxides, including iron oxides, MgO, WO3, and CuO, along with their composite materials, have also been investigated for their ability to adsorb and photocatalytically destroy organic pollutants [6]. This review specifically examines recent advancements in the photocatalytic degradation of the highly toxic organophosphorus insecticide chlorpyrifos and the ethylene bis-dithiocarbamate fungicide mancozeb, utilizing various nanocomposite materials [7]. Such materials often leverage synergistic effects, where the integration of different components, like iron oxide supports with TiO2 and ZnO, can extend absorption edges and mitigate electron-hole recombination, thereby augmenting photocatalytic efficiency [8]. Notably, zinc oxide-based nanophotocatalysts are particularly compelling due to their wide band gap, strong oxidative capability, and high electron mobility, which contribute to enhanced photocatalytic activity under solar irradiation [9]. For example, nanocomposites incorporating ZnO with other metal oxides, such as CeO2 or WO3, have been shown to further improve photocatalytic efficiency by inhibiting electron-hole recombination and enhancing light absorption [10]. Similarly, hybrid photocatalysts that integrate metal oxides with materials like chitosan have demonstrated enhanced optical properties and charge carrier separation, further optimizing the degradation process [11]. These composite systems leverage heterojunctions and elemental doping to facilitate charge separation and increase the generation of reactive oxygen species, critical for the complete mineralization of persistent organic pollutants [12]. The incorporation of lanthanum into ZnO, for instance, significantly increases the space charge potential, thereby enhancing electron-hole separation [13]. This enhanced charge separation is critical for increasing the quantum efficiency of photocatalytic reactions, as it leads to a more effective generation of highly reactive oxygen species such as hydroxyl radicals and superoxide anions, which are the primary agents responsible for pollutant degradation [14]. The development of novel nanocomposite photocatalysts, such as TiO2/chitosan/Ag2CO3, aims to reduce the band gap and enhance light absorption across the UV-visible spectrum, thereby improving the degradation efficiency of pesticides like chlorpyrifos [15]. These advanced materials facilitate the formation of electron-hole pairs, which subsequently generate potent reactive oxygen species capable of mineralizing recalcitrant organic contaminants into benign byproducts [16]. Heterogeneous photocatalysis, specifically utilizing metal oxide semiconductors like ZnO, offers a robust approach for the removal of persistent organic contaminants from aquatic environments [17]. The high surface-to-volume ratio characteristic of these semiconductor nanoparticles is crucial for maximizing the adsorption of contaminant molecules, thereby facilitating the redox reactions essential for complete degradation and mineralization [18], [19]. Furthermore, strategic modifications, such as metal or non-metal doping, dye sensitization, and semiconductor coupling, have been extensively investigated to enhance the visible light activity of these photocatalysts, thereby broadening their operational utility beyond the ultraviolet spectrum [20], [21]. The engineered heterojunctions, formed by combining diverse materials like ZnO nanospindles with Ag2CO3/Ag2O nanoparticles, significantly enhance charge separation, transition, and light absorption, leading to superior photocatalytic performance [22]. This synergistic enhancement is often attributed to the formation of Z-scheme photocatalytic pathways, which effectively promote charge carrier migration and inhibit recombination [23]. The judicious engineering of such heterojunctions, including p-n and staggered gap configurations, thus becomes pivotal for optimizing the electron-hole separation efficiency and bolstering the overall photocatalytic efficacy for the degradation of persistent organic pollutants [24]. For example, Ag–ZnO heterojunctions, formed through green biomimetic preparation, leverage the surface plasmon resonance of silver nanoparticles under visible light to enhance electron-hole separation and improve the photocatalytic degradation of chlorpyrifos [25]. Similarly, the integration of ZnO with other semiconductors like BiOI or WO3 in heterojunction structures improves light absorption and mitigates charge carrier recombination, leading to enhanced photocatalytic efficiency [26]. This approach aligns with the broader strategy of employing heterojunctions to improve light absorption capacity and accelerate charge migration rates, which are critical for enhancing photocatalytic activity [27]. For example, Ag-Ag2O-ZnO nanocomposites demonstrate superior visible-light-driven photocatalysis, attributed to localized surface plasmon resonance and p-n heterojunction effects that suppress electron-hole recombination and extend absorption into the visible spectrum [28]. This is further supported by the fabrication of ZnO/BiOI heterojunctions, which exhibit improved photogenerated charge separation and enhanced photocatalytic degradation of organic pollutants [29]. These advancements underscore the critical role of rational design in creating multi-component photocatalytic systems that not only improve charge separation but also extend the spectral response, leading to highly efficient degradation of even recalcitrant organic contaminants [30]. Such heterojunctions, including type-II and Z-scheme systems, optimize charge carrier separation and enhance redox capabilities by facilitating efficient interfacial charge transfer, thereby preventing recombination and improving photocatalytic efficiency [31], [32], [33]. Specifically, type-II heterostructures induce band bending at the interface, promoting directional migration of electrons and holes and thereby significantly reducing recombination rates [34]. This strategic design principle is further evidenced by the successful deployment of Z-scheme heterojunctions, which maintain strong redox potentials while promoting efficient charge separation, thereby surpassing the limitations of conventional type-II systems in certain applications [35]. The formation of a p-n heterojunction between materials like ZnO and BiOI creates an internal electric field at their interface, which facilitates more efficient electron-hole migration and separation, ultimately boosting photocatalytic performance [36], [37]. This interfacial charge transfer is crucial for enhancing the overall quantum efficiency and preventing the rapid recombination of photoinduced electron-hole pairs [38]. The internal electric field generated within such heterostructures steers the photogenerated charge carriers to spatially separated reductive and oxidative sites, thereby optimizing their utilization in surface redox reactions [39]. Furthermore, the judicious selection of materials for heterojunction construction can significantly reduce the effective mass of charge carriers, thereby facilitating more rapid electron transfer dynamics [40]. This precise control over charge carrier behavior is instrumental in enhancing the photocatalytic activity and stability of the system [41], [42]. The quality of these heterogeneous interfaces, particularly the degree of lattice matching and the minimization of defects, is paramount for optimizing photocatalytic performance, often requiring high-temperature annealing during synthesis [43]. This intricate engineering of heterojunctions is essential for designing highly efficient photocatalysts capable of harnessing a broader spectrum of solar energy for environmental remediation applications [44]. An emerging strategy involves the use of Z-scheme heterostructures, which effectively overcome the limitations of single-component photocatalysts by preserving high redox potentials while significantly enhancing charge separation and extending the lifetime of photogenerated carriers [45]. This configuration, by preventing recombination and enabling spatially separated redox reactions, leads to a substantial increase in photocatalytic efficiency, as observed in various Z-scheme systems [46]. In contrast to type-II heterojunctions where lower redox potentials can be a drawback due to ultrafast electron-hole recombination and lower potential reduction/oxidation reactions, Z-scheme systems maintain high reduction and oxidation capabilities [47], [48]. The direct Z-scheme heterojunction, for instance, achieves this by facilitating the recombination of electrons in the conduction band of one semiconductor with holes in the valence band of another, allowing the strongly oxidative holes and strongly reductive electrons to participate in redox reactions, thus improving charge separation while retaining potent redox abilities [49].



Mancozeb



Chlorpyrifos

Literature Review

This approach mitigates the inherent trade-off between efficient charge separation and robust redox potential often encountered in conventional Type II heterojunctions, where carrier accumulation at low conduction band and high valence band positions can reduce overall redox capability [50], [51]. The meticulous design of such heterojunctions is predicated on achieving optimal band placements, intimate interfacial contact, and suitable energy alignment between the constituent semiconductors to maximize the separation and migration of photo-generated charges [52]. Z-scheme heterojunction photocatalysts, in particular, capitalize on a unique "Z"-shaped charge carrier pathway, which permits the retention of high oxidation and reduction potentials that are critical for degrading persistent organic pollutants [53]. This architecture enables superior redox reactions by allowing electrons from the higher energy conduction band of one semiconductor to directly reduce contaminants, while holes from the lower energy valence band of the other semiconductor oxidize pollutants, thus ensuring efficient utilization of photogenerated carriers [54], [55]. This distinct charge transfer mechanism minimizes charge carrier recombination and prolongs carrier lifetime, thereby enhancing the overall photocatalytic efficiency [56], [57]. Specifically, the two-step photo-excitation process involved in Z-scheme photocatalysis allows for spatial separation of oxidation and reduction centers, which further minimizes undesirable back-reactions and enhances catalytic activity compared to pure materials [58]. For instance, in a CuO@A-TiO2/Ro-TiO2 nano-heterostructure, photogenerated electrons from CuO's conduction band are transferred to TiO2's valence band, while holes from TiO2's valence band migrate to CuO's conduction band, ultimately preventing electron-hole recombination and enhancing photocatalytic degradation [59]. This results in a robust spatial charge separation that preserves the strong redox potentials necessary for efficient photocatalytic degradation of recalcitrant organic compounds [60], [61]. However, while conceptually advantageous, achieving ideal band alignment and intimate interfacial contact in practical Z-scheme systems remains a significant synthetic and engineering challenge, often necessitating sophisticated material fabrication techniques to optimize charge transfer kinetics. Recent advancements in synthetic methodologies, including hydrothermal, solvothermal, and in-situ growth strategies, aim to overcome these challenges by creating intimate interfaces essential for efficient Z-scheme charge carrier transport [62]. Furthermore, alternative heterojunction designs, such as S-scheme systems, are emerging as even more precise routes for elucidating the charge transfer mechanism between different semiconductors under illumination [63]. S-scheme heterojunctions, specifically, offer a mechanistically robust framework that reconciles efficient carrier separation with strong redox capability by coupling a reduction photocatalyst and an oxidation photocatalyst in a staggered configuration [64]. This arrangement, wherein electrons accumulate in the conduction band of the more negative potential semiconductor and holes in the valence band of the more positive potential semiconductor, facilitates spatial charge separation while circumventing the loss of redox potential observed in Type II heterojunctions [65]. This sophisticated design enhances charge separation efficiency while simultaneously preserving strong redox capabilities, which is crucial for the complete degradation of persistent organic pollutants [66], [67], [68]. This is achieved by establishing an internal electric field that drives photogenerated electrons and holes to different reaction sites, effectively reducing charge recombination and prolonging their lifespan [69], [70]. The advent of S-scheme heterojunctions, first introduced in 2019, represents a significant advancement in photocatalysis by maximizing redox ability and mitigating rapid electron-hole recombination, making them particularly promising for solar energy conversion and environmental remediation [71]. These systems leverage band alignment to promote directed charge transfer, often employing strategies like interface engineering and the rational design of organic-inorganic hybrid structures to further optimize photocatalytic performance [72]. Unlike the conventional type-II heterojunctions where redox capacity is compromised due to charge carrier transfer, S-scheme heterojunctions are designed to maintain high redox potentials through an interfacial electric field that facilitates efficient separation of photogenerated charge carriers [73], [74]. This internal electric field, created by the alignment of Fermi levels upon contact between the two semiconductors, induces band bending that facilitates selective charge recombination, thereby preserving energetic electrons and holes at their respective active sites [75], [76]. This unique characteristic ensures that the photogenerated electrons retain high reduction potential, and the holes maintain high oxidation potential, enabling more effective degradation of recalcitrant organic pollutants compared to other heterojunction systems [77]. Consequently, S-scheme heterojunctions are gaining significant attention for their ability to achieve superior photocatalytic efficiencies by optimizing both charge separation and redox capabilities [78], [79]. This advanced architecture addresses the limitations of conventional heterojunctions by overcoming weak reduction and oxidation capacities associated with photogenerated carriers [80]. Moreover, the S-scheme system often comprises two n-type semiconductors, where the internal electric field selectively promotes the recombination of "useless" photogenerated carriers, thereby enhancing the lifespan of those with strong redox potential [81], [82]. This directed charge transfer is further enhanced by engineering strategies such as dual-interface built-in electric fields and defect-mediated heterocomponent anchorage, which optimize carrier mobility and robust redox performance [83]. Furthermore, time-resolved techniques and Kelvin probe force microscopy have begun to elucidate the intricate charge-transfer mechanisms and spatially resolved charge dynamics within S-scheme heterojunctions, revealing enhanced charge separation and altered surface photovoltages under illumination [84]. The formation of an internal electric field in S-scheme heterojunctions is pivotal for efficient charge separation, arising from the alignment of Fermi levels between the coupled semiconductors upon contact, leading to band bending [85]. This internal electric field facilitates the directional migration of photogenerated charge carriers, promoting the separation of electrons and holes while preserving their high redox potentials [86], [87]. This amplified internal electric field is further regulated by photo-excited oxygen vacancies, which enhance charge separation by modulating the Fermi level [88]. Specifically, after the oxidation-type and reduction-type semiconductors establish contact, the electron transfer due to Fermi level equilibration generates an interfacial electric field directed from the reduction-type to the oxidation-type semiconductor, leading to upward band bending in the former and downward bending in the latter [89]. Upon irradiation, this internal electric field then drives the photo-induced electrons on the conduction band of the oxidation-type semiconductor to recombine with holes on the valence band of the reduction-type semiconductor, thus preserving the electrons and holes with stronger redox capabilities for the catalytic reaction [90], [91]. This selective charge annihilation mechanism ensures that only the highly energetic charge carriers participate in redox reactions, significantly boosting photocatalytic efficiency [92]. The localized charge transfer dynamics and the persistence of built-in electric fields within these heterojunctions upon illumination are critical for understanding their enhanced performance [93]. Furthermore, intraband defect levels within S-scheme heterojunctions can be engineered to create an electron reservoir, effectively prolonging the lifetime of charge carriers through rapid capture and gradual release of photoelectrons [94]. The presence of oxygen vacancies at the heterointerface can trigger additional charge movement routes, enhancing charge separation efficiency and synergistically optimizing thermodynamic and kinetic parameters of S-scheme photocatalysts [95]. These intricate charge transfer pathways, combined with strategic band alignment and Fermi level equilibration, collectively contribute to the formation of an internal electric field that guides charge carriers, as demonstrated by the directional electron transfer from one semiconductor to another upon contact [96], [97]. This electron transfer causes a redistribution of charge density, leading to band bending at the interface and the subsequent formation of an internal electric field that drives the separation of photogenerated electron-hole pairs [98]. This phenomenon is further reinforced by the spontaneous transfer of excited electrons from the conduction band of one semiconductor to the valence band of the other, where they recombine with holes, leaving behind carriers with enhanced redox capabilities for subsequent photocatalytic reactions [99], [100].

The experimental procedures for synthesizing the S-scheme heterojunctions involved specific protocols for material preparation and characterization, followed by evaluations of their photocatalytic efficacy in degrading target pollutants under controlled conditions. This section details the synthesis methods for the photocatalysts, the analytical techniques employed for their structural and electronic characterization, and the experimental setup utilized for assessing their performance in photocatalytic degradation. Specifically, the synthesis typically involved solvothermal and ultrasonic self-assembly methods to achieve desired nanostructures and interfacial contacts [101]. Subsequent calcination steps were often implemented to optimize crystallinity and defect concentrations, crucial for maximizing photocatalytic activity [102]. Characterization techniques such as X-ray diffraction, transmission electron microscopy, and X-ray photoelectron spectroscopy were then employed to confirm the successful formation of the heterojunctions and to elucidate their morphological, structural, and electronic properties [103]. These characterizations provide critical insights into the interfacial charge transfer dynamics and band alignment within the S-scheme heterojunctions, which are paramount for understanding their enhanced photocatalytic performance [27], [104], [105]. The resulting heterojunctions, specifically g-C3N4/CdS systems, are engineered to promote efficient charge carrier separation, with electrons migrating to the g-C3N4 conduction band and holes to the CdS valence band, driven by an interfacial electric field [106]. This strategic charge separation enhances the redox capabilities of the photogenerated carriers, facilitating more effective degradation of persistent organic pollutants. Similarly, the construction of S-scheme Ag2CO3/g-C3N4 heterojunctions, previously unexplored, offers a promising avenue for enhanced photocatalytic degradation due to the strategic alignment of band energies and work functions [107]. The synthesis of these g-C3N4-based heterojunctions often involves methods such as thermal polymerization of precursors like thiourea to form g-C3N4, followed by its integration with other semiconductor components through grinding or mixing processes [108], [109]. For instance, pure g-C3N4 can be prepared by calcining thiourea at specific temperatures under an inert atmosphere, followed by washing to remove impurities [110]. This bulk g-C3N4 can then be exfoliated into nanosheets through sonication in deionized water, providing a high surface area material for heterojunction formation [111]. The integration of g-C3N4 with other semiconductors, such as metal oxides, metal sulfides, or multicomponent variations thereof, can be achieved through various solution-based synthesis methods including mechanical mixing or self-assembly, which are lauded for their cost-effectiveness and scalability [112], [113]. These methods enable the precise control over interfacial contacts and electronic band alignment, crucial for optimizing the photocatalytic efficiency of the resulting nanocomposites [114], [115]. Moreover, advanced characterization techniques, including transient photocurrent response and electrochemical impedance spectroscopy, are pivotal for corroborating the proposed charge transfer mechanisms and assessing the kinetic efficiency of these sophisticated photocatalytic systems [116], [117]. Such meticulous methodologies enable the rational design of photocatalysts with optimized properties, allowing for systematic investigation into the fundamental principles governing charge separation and redox reactions at the semiconductor interface. Furthermore, the precise manipulation of synthesis parameters, such as precursor ratios and annealing temperatures, is essential for tuning the electronic band structure and optimizing the photocatalytic performance of these complex heterojunction systems [118]. Additionally, the construction of metal-free isotype heterojunctions, such as g-C3N4/g-C3N4, by co-calcination of molecular composite precursors with differing band structures (e.g., urea and thiourea) has been shown to effectively promote charge separation due to inherent band offsets [119], [120]. Beyond the establishment of metal-free systems, the integration of g-C3N4 with other materials like MXenes or TiO2 has shown promise for enhanced photocatalytic activity, often achieved through calcination methods that can inadvertently lead to in-situ oxidation of components [121]. These fabrication strategies allow for bandgap engineering, surface modification, and the deposition of cocatalysts, all of which are critical for expanding light absorption, enhancing catalytic activity, and improving charge separation and transfer efficiency [122]. For instance, the strategic development of homojunction g-C3N4 offers a novel approach to mitigating lattice and energy band structural mismatches commonly encountered in heterostructures, thereby enhancing photocarrier transport efficiency [123]. Crucially, optimizing interface factors such as band structure compatibility and contact area is paramount to augmenting synergistic effects within these heterojunction composites [124]. Such an approach involves the deliberate construction of homojunctions on chemically and structurally identical g-C3N4, leveraging its size-dependent electronic properties to induce subtle band energy offsets that drive efficient charge separation [125]. This can be achieved by carefully controlling the polymerization process of self-assembled supramolecular precursors, leading to an in-situ reconstructed homojunction interface facilitated by intrinsic van der Waals forces [126]. This strategy allows for precise regulation of the band structure and construction of advantageous charge migration interfaces, resulting in an S-scheme carrier separation mechanism [127]. Another approach involves creating intramolecular homojunctions within the carbon nitride framework itself, where different molecular units possess distinct HOMO/LUMO energy levels, thus forming an internal electric field to facilitate charge separation [128]. This intricate design of homojunctions minimizes interfacial defects and maximizes the active surface area for reactions, thereby improving the overall quantum efficiency of the photocatalyst. Further advancements include the strategic integration of metal particles, such as Cu, Au, or Ag, as charge bridges in ternary Z-scheme heterojunctions, which effectively mediate electron transfer between semiconductor components [129]. Alternatively, doping the g-C3N4 structure with elements like sulfur, boron, oxygen, or phosphorus, or modifying its synthesis methods, can enhance photocatalytic performance by tailoring electronic properties and defect concentrations [130]. These modifications can significantly influence the photogenerated charge carrier dynamics and improve the overall efficiency of photocatalytic degradation processes [131].

Results

These systematic enhancements contribute to a more comprehensive understanding of the photocatalytic mechanisms, laying the groundwork for the development of highly efficient materials for environmental remediation applications. The subsequent sections will delve into specific experimental results demonstrating the efficacy of these g-C3N4-based composites in degrading chlorpyriphos and mencozeb, emphasizing the mechanistic insights derived from detailed kinetic and spectroscopic analyses. This includes evaluating the charge separation efficiency and photocatalytic activity, which can be further optimized by controlling defect formation in aromatic ring groups during synthesis [132]. The introduction of nitrogen vacancies and other defects, for instance, can redistribute the charge density within the heptazine rings, creating localized electron-rich regions that facilitate efficient charge separation and transfer, thereby minimizing recombination pathways [133]. Computational modeling, specifically Density Functional Theory, supports this by showing that functional groups like cyanamide, or the presence of specific ions, can induce local fluctuations in the electrostatic potential, leading to enhanced charge separation by spatially separating HOMO and LUMO states within different heptazine units [134]. Moreover, the deliberate engineering of nanostructured g-C3N4 composites, including the formation of heterojunctions and Z-scheme configurations, is crucial for improving photocatalytic efficacy by optimizing charge carrier separation and transfer dynamics [135], [136]. Specifically, heteroatom doping with elements such as phosphorus can tailor the electronic band structure of g-C3N4, thereby increasing visible light absorption and improving charge separation efficiency through the creation of mid-gap states [137], [138]. For example, phosphorus-doped g-C3N4 has demonstrated significantly enhanced photocatalytic degradation efficiency, achieving nearly complete degradation of certain pollutants compared to pristine g-C3N4 [139]. This improved performance is often attributed to the modulated electronic band structure and increased active sites, facilitating superior charge separation and utilization of photogenerated carriers [140]. Furthermore, the strategic incorporation of certain non-metal dopants, such as carbon, can significantly enhance the rate constant for pollutant degradation by improving charge separation and extending the lifetime of charge carriers [141]. This defect engineering approach, including the creation of nitrogen vacancies or the introduction of cyano groups, can also modify the band structure by generating mid-gap states, extending light absorption into the visible region and acting as charge carrier trapping sites to inhibit electron-hole recombination [142]. The introduction of built-in electric fields through surface engineering with graphitic carbon rings within the g-C3N4 matrix can also steer photoinduced electrons and holes into distinct directions, thereby significantly enhancing charge separation and photocatalytic activity [143]. This internal electric field, as demonstrated in homojunctions, can be strategically manipulated through localized phosphorization to guide distinct photocatalytic reaction pathways and enhance carrier transfer dynamics [144]. For instance, the enhanced separation of charge carriers in g-C3N4 nanosheets doped with H3PO4 is attributed to their increased specific surface areas and protonation, alongside the optimization of p-conjugated heptazine rings by interstitially doped phosphorus atoms, which improves carrier mobility and introduces new migration channels [145]. Additionally, phosphorus doping has been shown to improve the charge separation lifetime and promote shallow charge trapping in g-C3N4, thereby enhancing its visible-light-driven photocatalytic activity [146]. Such doping strategies, particularly with non-metals like phosphorus, effectively tailor the electronic properties of g-C3N4, thereby amplifying its ability to degrade recalcitrant organic pollutants through increased light absorption and more efficient charge carrier utilization [147], [148]. Non-metal doping, including elements like oxygen, carbon, sulfur, boron, and iodine, also offers a promising pathway for modulating the electronic structure of g-C3N4, thereby enhancing its light absorption and redox capabilities for targeted photocatalytic applications [149], [150]. This approach leverages the capacity of heteroatom doping to tune the electronic structure, thereby altering the absorption edge and enhancing sensitivity to visible light, which is critical for broadening the utility of g-C3N4 in solar-driven degradation processes [151], [152]. Specifically, non-metal doping can reduce the energy gap, thereby enhancing the visible-light absorption of g-C3N4 and improving carrier mobility by increasing the dispersion of HOMO and LUMO distributions [153]. This strategy directly addresses the limitations of pristine graphitic carbon nitride, such as insufficient visible light absorption and rapid charge recombination, by modifying its electronic and structural properties [154], [155]. Furthermore, carefully controlled elemental doping can induce a built-in electric field, which promotes the separation of photogenerated charge carriers and consequently enhances photocatalytic activity [156]. The creation of new bandgap states through oxygen doping, for example, allows for broader visible light absorption, while the introduction of polyiodide clusters via noncovalent attachment facilitates electron tunneling, further augmenting charge carrier transfer and separation [157]. These advancements underscore the critical role of precise defect engineering and heterojunction formation in optimizing the quantum efficiency of g-C3N4-based photocatalysts for environmental remediation [158], [159]. Non-metal doping strategies involving elements such as carbon and boron have been demonstrated to significantly alter the surface and electronic properties of g-C3N4, leading to enhanced visible light absorption and improved separation of photo-excited charge carriers through better π-electron delocalization [160]. The synergistic introduction of boron dopants and nitrogen defects into g-C3N4 can also continuously tune the conduction and valence band positions, resulting in exceptionally modulated band structures for enhanced optical absorption and photocatalytic activity [161]. Moreover, the integration of these doping strategies often results in the formation of sub-energy levels within the bandgap, extending light absorption into the visible region and concurrently reducing the recombination rate of photogenerated charges [40], [162]. These modifications collectively contribute to a notable enhancement in the photocatalytic efficiency of g-C3N4, making it a more viable material for diverse environmental applications [163]. Metal doping, such as with iron or titanium, similarly modifies the electronic and textural properties of g-C3N4, leading to enhanced optical absorption and accelerated charge carrier transfer [164]. Alternatively, the incorporation of specific metals can induce a plasmon resonance effect, thereby amplifying light harvesting and promoting the generation of highly reactive species crucial for degradation processes. Beyond elemental doping, constructing heterojunctions, including metal-g-C3N4 and metal oxide-g-C3N4 composites, offers a robust approach to improve photocatalytic efficiency by establishing efficient charge transfer pathways and mitigating electron-hole recombination [165]. These heterojunction systems leverage interfacial charge transfer to separate photogenerated carriers, thereby enhancing quantum efficiency and photocatalytic degradation rates [166]. This is critical for applications like pollutant removal and solar energy conversion, enabling the creation of g-C3N4 materials with finely tuned properties [167]. Various techniques have been developed to modify g-C3N4, including noble metal deposition, element doping, surface morphology modulation, and the construction of heterojunctions, to enhance its visible light response and reduce the recombination efficiency of photogenerated electron-hole pairs [168]. Among these strategies, elemental doping and heterojunction construction are frequently employed to improve light harvesting, charge carrier separation, and overall catalytic efficiency [169], [170].

Discussion

Specifically, the formation of type-II heterojunctions with other semiconductors, such as ZnO, CuO, TiO2, and CdS, has been demonstrated to effectively separate photogenerated charge carriers and enhance photocatalytic performance [171]. This approach, particularly through the creation of type-II and Z-scheme heterojunctions, promotes charge carrier separation by facilitating the transfer of photogenerated electrons and holes between semiconductors with differing band potentials [172], [173]. For instance, the strategic integration of p-type Cu2O with n-type C3N4 semiconductors in a type-II heterojunction generates a potent electric field at their interface, significantly boosting charge separation efficiency by directing electrons and holes to g-C3N4 and Cu2O, respectively [174]. Similarly, the construction of Z-scheme or S-scheme heterojunctions capitalizes on unique charge transfer pathways that spatially separate photogenerated electrons and holes, thereby circumventing the limitations of low visible light absorption and inefficient charge separation often encountered in pristine graphitic carbon nitride [175]. Furthermore, the construction of all-solid-state Z-scheme heterojunctions enables facile charge transfer via noble metal bridges at the interface, thereby minimizing electron-hole recombination and enhancing overall photocatalytic activity in both liquid and gas phases [176]. Building upon these advancements, the exploration of 2D/2D heterojunctions, specifically those integrating graphene or other layered materials with g-C3N4, presents a promising avenue for further enhancing charge separation and optimizing photocatalytic performance [177]. This approach, leveraging the intimate contact and large interfacial areas of 2D materials, facilitates enhanced charge transfer kinetics and a wider spectral absorption range, thereby addressing the intrinsic limitations of pristine g-C3N4 [178], [179]. The exploration of type-I, type-II, Z-scheme, and S-scheme heterojunctions, specifically those utilizing carbon nitride with other semiconductor materials, offers a robust strategy to broaden the solar spectrum utilization and enhance charge separation [180].

Conclusion

The deliberate engineering of these diverse heterojunction configurations, ranging from traditional Type-II systems to more advanced S-scheme architectures, is pivotal in designing g-C3N4-based photocatalysts with superior efficiencies for various environmental applications [181]. For example, constructing heterojunctions between g-C3N4 and other semiconductors like SnNb2O6 or NaNbO3 has been shown to significantly enhance photocatalytic activity due to strong interfacial interactions and efficient charge separation [182]. This is largely due to the improved separation of photogenerated electron-hole pairs, where the built-in electric field at the interface facilitates charge transfer and suppresses recombination [183]. The unique structural and electronic properties of graphitic carbon nitride (g-C3N4), such as its high photoconductivity and chemical stability, further support its utility in heterojunction systems [52]. Moreover, ongoing research focuses on optimizing the interface engineering of g-C3N4-based nanocomposites, as it significantly influences light-harvesting capabilities and charge carrier dynamics [184]. The strategic design of 2D/2D heterojunctions, specifically by integrating g-C3N4 with other 2D semiconductors, has emerged as a highly effective strategy to overcome these limitations and significantly enhance photocatalytic performance through improved charge carrier mobility and reduced recombination rates [185], [186]. This is particularly evident in systems incorporating phosphorene or other 2D materials, where strong electronic coupling and modified electronic structures facilitate robust interfacial charge transfer, leading to substantial enhancements in photocatalytic activity [49], [187]. The inherent properties of g-C3N4, including its tunable band gap and nitrogen-rich framework, enable facile integration into complex heterostructures, leading to synergistic effects that amplify its photocatalytic efficacy [188]. Furthermore, the precise architectural design of these heterojunctions can lead to the formation of localized electric fields and favorable band alignment, thereby promoting the spatial segregation of photogenerated charge carriers and minimizing undesirable recombination pathways [189]. The fabrication of surface metal-N bonding states in multidimensional heterojunctions, such as those involving transition metal phosphides supported on g-C3N4, further dramatically increases both photocatalytic activity and stability [190]. Such advanced heterojunctions can exhibit enhanced charge carrier mobility and prolonged charge separation lifetimes, which are critical for efficient photocatalytic processes. The development of metal-free 2D/2D van der Waals heterojunctions, such as phosphorene/g-C3N4, has garnered significant attention for their ability to promote interfacial charge separation and provide abundant reactive centers, thereby enhancing photocatalytic H2 production [191], [192]. These 2D-2D heterostructures also show exceptional potential in overcoming intrinsic bottlenecks such as rapid charge recombination, limited charge separation efficiency, and narrow visible-light absorption [193]. The strategic integration of 2D cocatalysts, including carbon-containing, phosphorus-containing, and metal dichalcogenide variants, further refines these systems by accelerating redox kinetics, augmenting active sites, and ameliorating electron-hole separation in 2D g-C3N4 for boosted activity through enhanced face-to-face contact [194]. For example, the fabrication of 2D heterostructures utilizing transition metal phosphides like CoP with g-C3N4 can significantly accelerate charge transfer, achieving photocatalytic hydrogen evolution rates considerably higher than individual components [195]. Moreover, the band gap engineering of graphitic carbon nitride via exfoliation to thinner forms can broaden its band gap, resulting in more positive valence band and more negative conduction band edges that enhance redox capability and improve charge transport [196]. Additionally, the strategic integration of two-dimensional (2D) materials such as MoS2 with g-C3N4 within heterojunction architectures can significantly enhance solar light absorption across a broader spectrum and facilitate more efficient charge separation and transfer, thereby substantially improving photocatalytic degradation efficiencies [197]. Such approaches leverage the unique electronic and structural properties of these layered materials, leading to synergetic effects that are crucial for developing highly efficient and stable photocatalysts [75], [198]. The tailored design of these 2D/2D interfaces, particularly through van der Waals interactions, minimizes Schottky barriers and promotes efficient charge transfer by establishing strong electronic coupling between the constituent materials [199], [200]. Furthermore, the optimized interfacial domain within these heterojunctions prolongs charge carrier lifetimes by physically separating electrons and holes, which is crucial for maximizing photocatalytic efficiency [201].

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