

## Review

# Sustainable Treatment of Plastic Wastes with Photocatalytic Technologies: A Review

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## Abstract

Plastic waste pollution has been widely recognized as one of the most severe and pressing environmental challenges of our time, posing significant threats to ecosystem stability and human health. The transformation of plastic waste into high-value chemicals and clean energy via photocatalytic reforming technology is increasingly regarded as a promising and sustainable alternative pathway, offering dual benefits of resource recovery and environmental remediation. This review first provides an overview of the current state of research on plastic waste management. It then systematically summarizes recent representative advances in the coupling of plastic upcycling with photocatalytic technologies, with a particular focus on the potential of plastics as carbon sources in both photodegradation and photosynthetic transformation pathways, highlighting their value and future prospects. Finally, this review outlines the key scientific challenges that urgently need to be addressed in the field of photocatalytic conversion of plastic waste, and, in light of emerging research trends, proposes several promising directions for future investigation along with the authors' perspectives. It is hoped that these insights will provide useful guidance and inspiration for the continued advancement of this field.

**Keywords:** conversion of plastic wastes; photocatalytic upcycling; controlled product selectivity; reaction pathway; high-value chemical products



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## 1. Introduction

Plastic products derived from petroleum refining have become indispensable in daily life due to their lightweight nature, low production cost, and excellent durability [1]. According to relevant statistics, global plastic consumption has surged from 335 million tons in 2016 to 400 million tons in 2021, showing a continuous upward trend [2]. During the COVID-19 pandemic, the demand for single-use personal protective equipment (PPE) such as masks and gloves, along with packaging materials, surged dramatically. It is estimated that over 450 billion plastic-based PPE items were used globally in 2020 alone [3]. Moreover, the widespread adoption of fast-paced lifestyles has driven the rapid growth in the food delivery and e-commerce sectors, significantly increasing the consumption of plastic packaging materials. However, despite the convenience plastics bring to modern life, the growing dependence on them has led to increasingly severe challenges in waste management. Owing to the large-scale production and short service life of most plastic

products, post-use plastics continue to accumulate in the environment, leading to mounting pollution and increasingly complex waste management issues.

Currently, the scientific community is actively addressing the challenges of plastic recycling and resource recovery by exploring a range of efficient and viable chemical transformation pathways, including hydrogenolysis, alcoholysis, glycolysis, and pyrolysis [4–6]. Under ideal conditions, chemical conversion pathways should enable the efficient recovery of high-purity monomers from plastic waste, with the recovered products exhibiting quality and reactivity comparable to those of virgin feedstocks, allowing for direct reutilization in the synthesis of new polymers. This approach not only significantly improves resource utilization efficiency but also paves the way toward genuine “closed-loop recycling” [7,8]. However, plastics—particularly thermosets and highly cross-linked polymers—exhibit exceptional chemical stability due to their dense three-dimensional network structures and robust carbon–carbon backbones, which severely hinder their degradation under mild conditions. As a result, current chemical recycling technologies typically require harsh operating conditions—such as elevated temperatures ( $>180\text{ }^{\circ}\text{C}$ ), high pressures (20–40 atm), and concentrated alkali (1–10 M) or strong acids (e.g., concentrated  $\text{H}_2\text{SO}_4$  or  $\text{HNO}_3$ ) as reaction media. While these conditions facilitate polymer-chain scission and improve reaction rates, they also pose significant challenges, including high energy consumption, severe equipment corrosion, secondary environmental pollution, and limited economic feasibility [9–11]. Therefore, the development of mild and green plastic conversion technologies operable under ambient or near-ambient conditions is a critical breakthrough toward achieving sustainable plastic recycling.

Photo-driven photocatalytic technology is regarded as a highly promising strategy for sustainable plastic waste management. In general, photocatalytic plastic conversion systems can be broadly categorized into the following two types: photodegradation and photosynthetic transformation (Figure 1) [12]. The ability to selectively generate value-added products is one of the key criteria distinguishing photodegradation from selective photo-reforming processes. As shown in Figure 1a, conventional photodegradation typically aims at mineralization, relying on highly oxidative radicals (e.g.,  $\bullet\text{OH}$ ) to indiscriminately attack polymer backbones and intermediates. This results in complex reaction pathways and broad product distributions, with final products dominated by  $\text{CO}_2$ , water, and trace organic acids, offering limited value [12,13]. In contrast, photosynthetic conversion emphasizes precise regulation of reaction pathways to efficiently convert hydrocarbon content in plastics into chemically valuable target molecules (e.g.,  $\text{H}_2$ ,  $\text{C}_1\text{--C}_{2+}$  fuels, and aromatic compounds) (Figure 1b) [14]. Therefore, the transition to photosynthetic systems hinges on the modulation of radical type, concentration, and spatial reactivity to prevent uncontrolled mineralization by overly oxidative species.

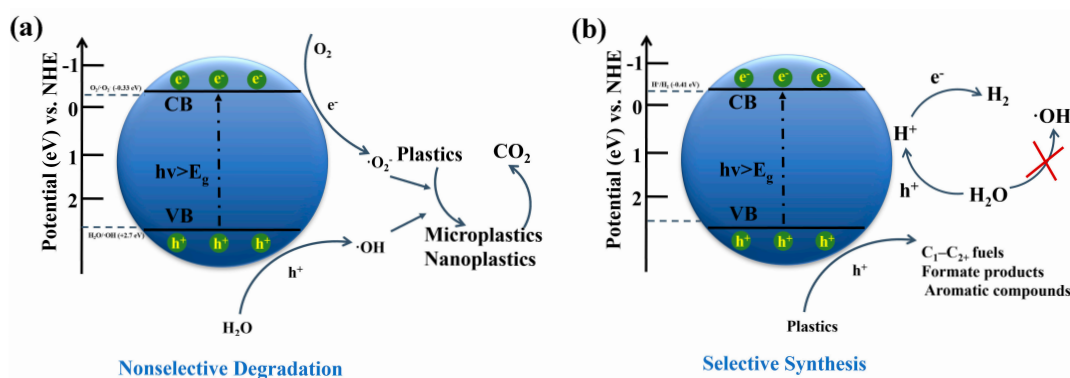


**Figure 1.** Schematic diagram of (a) photodegradation and (b) photosynthesis of plastics.

In this review, we first provide a brief overview of the current status of plastic waste treatment with respect to the basic reaction mechanisms involved in the photocatalytic process for plastics, with special emphasis on the redox pathways and the roles of key reactants in the photocatalytic process. We then systematically summarize representative recent advances in photocatalytic plastic waste conversion, categorized by plastic type (e.g., polyolefins and polyesters) (Figure 2), and critically analyze the performance and applicability of different catalytic systems. Finally, we discuss the key challenges in achieving efficient and selective transformation of plastic waste, including catalyst stability, compatibility with real-world plastics, product selectivity control, and system-level economic feasibility. We also outline future directions in catalyst design, mechanistic investigation, reaction engineering scale-up, and industrial deployment. We hope this review offers theoretical insights and technical references that contribute to the green transition of plastic pollution management and resource recovery.

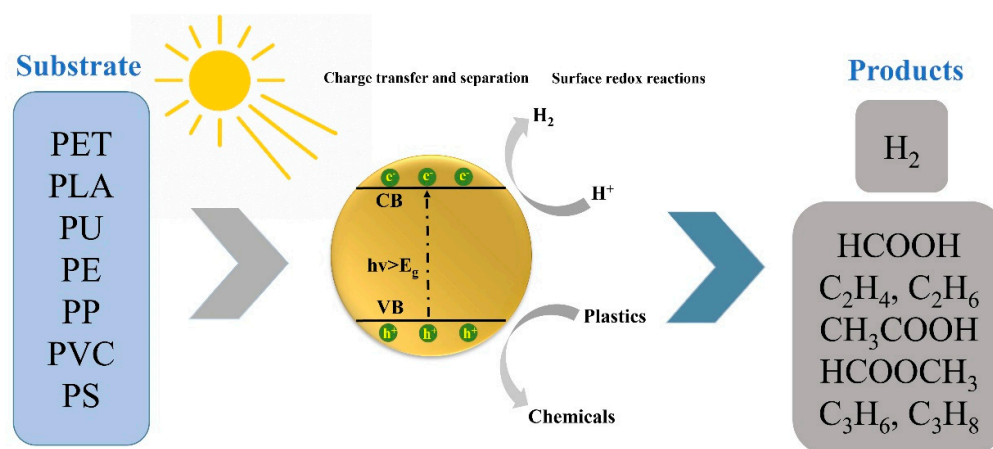


**Figure 2.** Diagram illustrating the photoconversion of plastics into fuels and chemicals.

## 2. Current Status of Plastic Waste Management

The management of plastic waste has attracted widespread global attention due to its significant impact on the sustainability of human life. A variety of techniques are employed in managing plastic waste, including incineration, landfill disposal, microbial degradation, and mechanical shredding with subsequent recycling, as well as transformation methods driven by electricity, heat, or light. These approaches each offer distinct advantages and limitations in the effort to reduce environmental impact and recover value from discarded plastic materials (Figure 3) [15–21]. Currently, plastic waste is primarily managed through landfilling and incineration, while a substantial fraction is still discharged directly into natural environments [22]. Among various plastic products, packaging materials are particularly problematic, as they are prone to entering rivers, lakes, and oceans, posing significant threats to aquatic ecosystems [23]. With the continuous growth in global plastic production and the diminishing availability of landfill space, the economic burden of plastic waste management has increased markedly. More critically, landfilling not only leads to a considerable waste of petroleum-derived resources but also gives rise to severe environmental concerns, including the leaching of chemical additives and long-term land occupation [24]. Similarly, although plastic incineration allows partial energy recovery, it is far less efficient compared to recycling and entails high environmental costs [25]. The life-cycle carbon footprint of incinerated polymers is substantial, with an estimated 5 to 10 tons of CO<sub>2</sub> emitted per ton of plastic processed [17]. Furthermore, incineration can produce toxic byproducts [26], rendering it a suboptimal strategy from both ecological and energy standpoints. Mechanical recycling typically follows a downcycling approach, wherein waste plastics are physically processed through shredding, washing, and remelting [27].

However, the resulting recycled materials are often utilized in the production of low-performance products, such as flower pots or non-food-contact containers [28]. This degradation in material quality leads to a reduction in overall polymer value, limiting its potential for reuse in high-end applications.

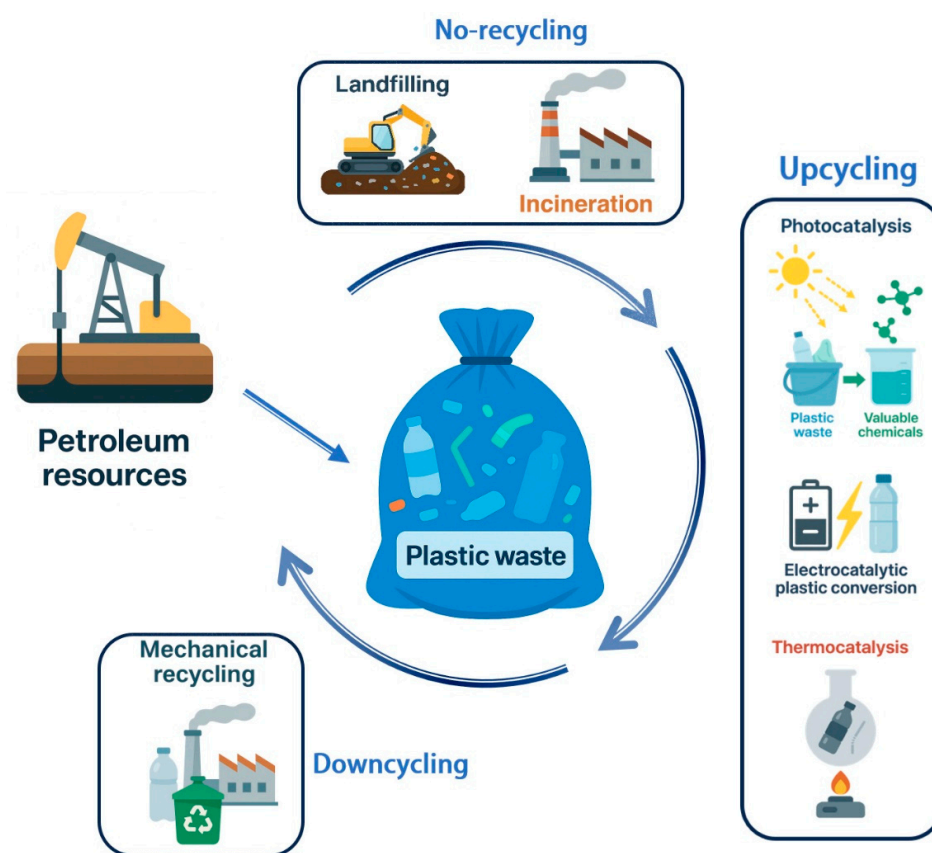


**Figure 3.** Plastic waste disposal strategy.

To address the inherent limitations of conventional recycling approaches, catalytic upcycling has emerged as a highly attractive alternative. This strategy enables the efficient conversion of polymeric waste into high-value chemicals, fuels, and other functional materials with significant economic potential, offering a promising complement to existing recycling methods [29]. Electrocatalysis has demonstrated promising conversion efficiency in the treatment of polymeric waste, offering advantages such as mild operating conditions and product selectivity. However, its reliance on external electrical energy input and the current limitations in electrode material selection, reactor design, and long-term operational stability pose significant technical challenges that must be addressed to enable practical application [30]. Thermocatalytic conversion is highly efficient and offers excellent reaction stability, enabling the rapid cleavage of C–C bonds to produce value-added chemicals. However, the requirement for elevated temperatures and substantial energy input imposes constraints on its scalability and practical implementation [31]. Enzymatic biotechnology relies on highly specific plastic-degrading enzymes, which have a limited substrate scope and face challenges in processing complex or mixed plastic waste. Moreover, these enzymes often exhibit poor stability under harsh reaction conditions. Fermentation, which converts plastic-derived intermediates into valuable chemicals through microbial metabolism, typically requires tedious pretreatment steps to depolymerize plastics into fermentable substrates. This complicates the process workflow and significantly increases downstream separation and purification costs [32]. Photocatalytic conversion has emerged as a promising strategy for plastic waste treatment due to its mild reaction conditions, high energy

efficiency, and potential for selective transformation. As shown in Figure 4, operating under ambient temperature and pressure and utilizing solar or ultraviolet light as the energy source, photocatalysis presents a more economically viable and environmentally friendly alternative compared with conventional thermal and chemical degradation methods [33]. Unlike pyrolysis or incineration, which require high energy input and often generate toxic byproducts, photocatalytic processes can proceed under mild conditions with minimal environmental risk. Furthermore, many photocatalytic systems leverage solar energy, enhancing their long-term sustainability and cost-effectiveness. Certain photocatalysts have demonstrated the ability to selectively cleave polymer chains or oxidize specific moieties, enabling the conversion of plastic waste into valuable monomers or fine chemicals. Compared with enzymatic biotechnology, which relies on highly specific enzymes and suffers from limited substrate scope and low stability under harsh conditions, photocatalytic systems exhibit broader applicability. They can effectively process both homochain and heterochain polymers by modulating reactive oxygen species ( $\bullet\text{OH}$ ,  $\bullet\text{O}_2^-$ ) and optimizing catalyst active sites. Similarly, microbial fermentation—despite its potential for converting plastic-derived intermediates into chemicals—often requires tedious and costly pretreatment to break down polymers into fermentable substrates, complicating process workflows and increasing separation burdens. In contrast, photocatalysis offers an integrated platform for both degradation and upcycling, enabling direct, selective conversion of plastic waste without extensive preprocessing. Its ability to control radical activity also helps suppress over-oxidation, thereby favoring the formation of high-value products. Taken together, the synergistic advantages of photocatalytic technology, including mild operational conditions, solar energy utilization, broad substrate compatibility, and direct value-added conversion, position it as a highly efficient and sustainable core strategy for addressing plastic waste. In comparison, other emerging alternatives remain constrained by energy intensity, substrate specificity, or process complexity.

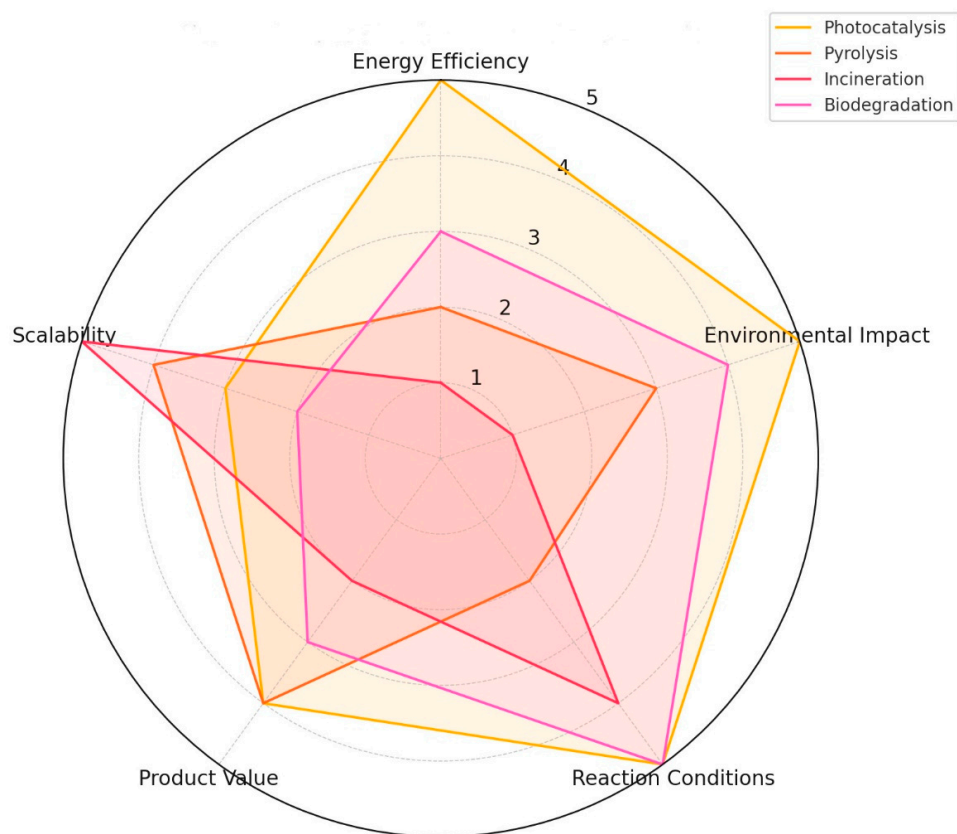


**Figure 4.** Comparison of plastic degradation methods.

### 3. Mechanistic Insights and Design Principles of Photo-Reforming

The photocatalytic process utilizes suitable semiconductor catalysts to convert incident light energy into chemical energy, initiating redox reactions on the catalyst surface. When the energy of incident light equals or exceeds the bandgap of a semiconductor photocatalyst, electrons in the valence band are excited to the conduction band, generating photogenerated electron ( $e^-$ )–hole ( $h^+$ ) pairs. These charge carriers migrate to the surface of the catalyst and undergo redox reactions with adsorbed reactants to sustain the photocatalytic process [34,35]. In conventional photocatalytic reactions (e.g., overall water splitting for hydrogen evolution or  $\text{CO}_2$  reduction), the VB level of the catalyst is typically more positive than the oxidation potential of water, allowing photogenerated holes in the VB to oxidize  $\text{H}_2\text{O}$  to produce  $\text{O}_2$  (Figure 5a). However, the overall efficiency of this process is often limited by sluggish water oxidation kinetics and the recombination of photogenerated holes [36,37]. To enhance the reaction rate, sacrificial agents (e.g., triethanolamine (TEOA), ascorbic acid, or ethanol) are often introduced to act as hole scavengers in place of water (Figure 5b) [38]. Plastic waste has been recognized as an effective sacrificial electron donor in photocatalytic reactions, capable of driving  $\text{H}_2$  evolution or  $\text{CO}_2$  reduction reactions (Figure 5c).

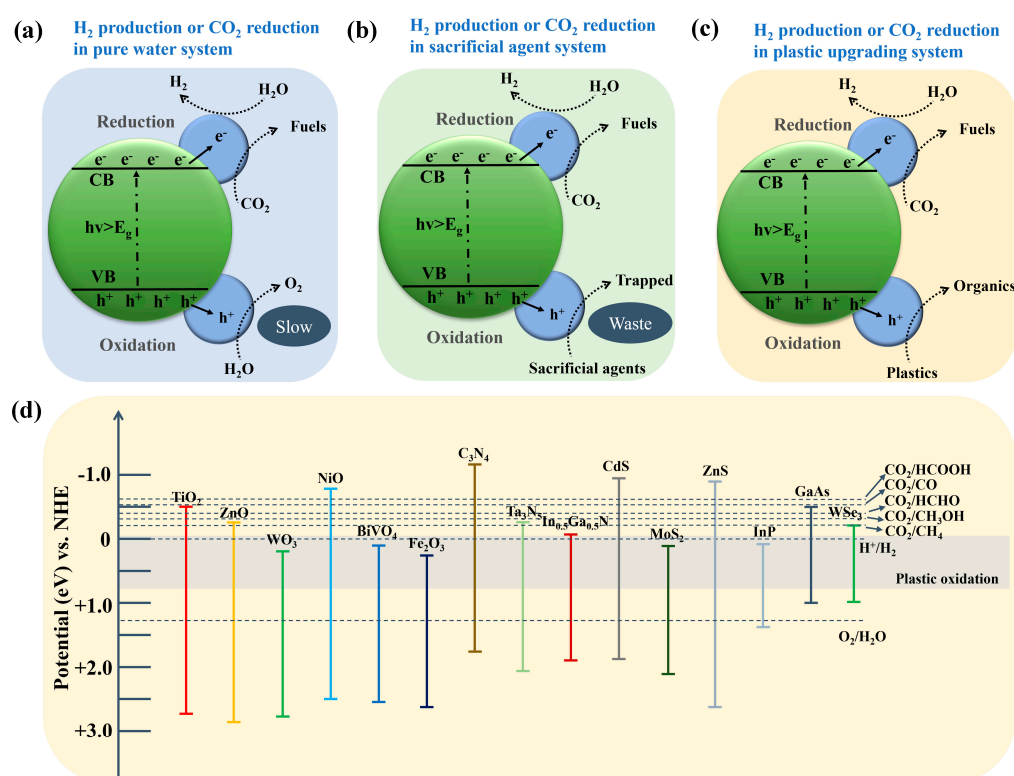


**Figure 5.** Schematic diagrams of photocatalytic  $\text{H}_2$  production and  $\text{CO}_2$  reduction: (a) pure water system, (b) sacrificial reagent-assisted system, and (c) plastic-integrated system; (d) band gaps and band edge positions of representative semiconductors relative to the NHE.

As shown in Figure 5d, the integration of plastics into photocatalytic coupling systems allows for the utilization of semiconductors with narrower bandgaps, offering greater flexibility and freedom in catalyst design and optimization. In contrast, conventional water oxidation generally requires wide-bandgap metal oxides to provide sufficiently positive valence band potentials for  $\text{H}_2\text{O}$  oxidation. Narrow-bandgap materials, such as sulfides, phosphides, and graphitic compounds (e.g., g- $\text{C}_3\text{N}_4$ ), generally possess lower valence band positions and are, thus, inadequate for direct water oxidation. However, in photocatalytic plastic reforming, the oxidation potentials of polymers are significantly lower than that of water, making these narrow-bandgap materials more suitable as photocatalysts. They

can not only absorb visible light efficiently but also oxidize plastic molecules at lower hole energy levels.

In addition, one of the major environmental concerns in plastic degradation is the formation of microplastics and nanoplastics as intermediate or end products of incomplete breakdown (Table 1). These small particles can evade filtration, accumulate in ecosystems, and pose serious health risks to humans and marine organisms. Photocatalysis offers a distinct advantage in addressing this issue, as it does not rely on mechanical fragmentation but instead utilizes photo-induced reactive oxygen species (ROS)—such as hydroxyl radicals and superoxide ions—to oxidatively cleave polymer chains. This mechanism allows for progressive oxidation, ultimately leading to complete mineralization under optimal conditions (e.g., prolonged irradiation, sufficient catalyst loading, and proper surface area). In recent studies,  $\text{TiO}_2$ -based and doped  $\text{g-C}_3\text{N}_4$  photocatalysts have demonstrated the ability to degrade microplastic particles ( $<5\text{ mm}$ ) to  $\text{CO}_2$  and small organic acids, thereby preventing their persistence in the environment. Moreover, the photoinduced hydrophilization of microplastics enhances their dispersion and accessibility to catalytic sites. Nevertheless, complete mineralization may still require extended reaction times, and future designs should focus on enhancing catalyst–particle contact and light absorption efficiency for more effective micro/nano plastic removal.

**Table 1.** Photocatalytic control of microplastic formation.

| Particle Size                                           | Typical Source                                          | Risk Level | Can Photocatalysis Address?  | Notes                                   |
|---------------------------------------------------------|---------------------------------------------------------|------------|------------------------------|-----------------------------------------|
| Macroplastic ( $>5\text{ mm}$ )                         | Packaging, bottles                                      | Medium     | Yes                          | Physical degradation starts the process |
| Microplastic ( $1\text{ }\mu\text{m}$ – $5\text{ mm}$ ) | Microplastic ( $1\text{ }\mu\text{m}$ – $5\text{ mm}$ ) | High       | Yes (slow mineralization)    | Needs prolonged irradiation             |
| Nanoplastic ( $<1\text{ }\mu\text{m}$ )                 | Weathering, fragmentation                               | Very High  | Partially (advanced systems) | Harder to capture and identify          |

Based on the type of catalyst employed, the photocatalytic conversion of plastic waste can be categorized into the following two main systems: homogeneous and heterogeneous. In homogeneous systems, light-responsive photosensitizers are typically dissolved in the reaction medium, allowing for molecular-level interactions with the substrates. The catalyst absorbs photons and is promoted to an excited state, wherein an electron is transferred from the highest occupied molecular orbital (HOMO) to the lowest unoccupied molecular orbital (LUMO). This excited species then interacts with the plastic substrate via mechanisms such as hydrogen atom transfer (HAT) or electron transfer, generating reactive intermediates that drive the selective transformation of polymer waste [39]. In contrast, heterogeneous systems utilize solid semiconductor catalysts, where reactions occur on the catalyst surface [40]. These systems offer advantages such as facile separation and catalyst recyclability. Both approaches exhibit distinct merits in terms of reaction mechanisms, energy utilization efficiency, and product selectivity, providing diverse strategies for the sustainable valorization of plastic waste.

#### 4. Recent Advances in Photocatalytic Conversion of Plastics

To date, extensive research has been conducted on the photodegradation of plastics, encompassing both heterochain polymers (e.g., PLA, PET, and PU) and homochain polymers (e.g., PE, PS, PP, and PVC), as summarized in Figure 6. The complete photodegradation of such plastics typically requires several days to weeks, depending on the type of photocatalyst used, the specific conditions of the reaction system, and the structural characteristics of

the plastic itself. Among various types of plastics, polyolefin materials—such as PE and PP—account for approximately 57% of the total global plastic production. As a result, they have been the most extensively investigated representatives in photodegradation-related studies, as summarized in Table 2.

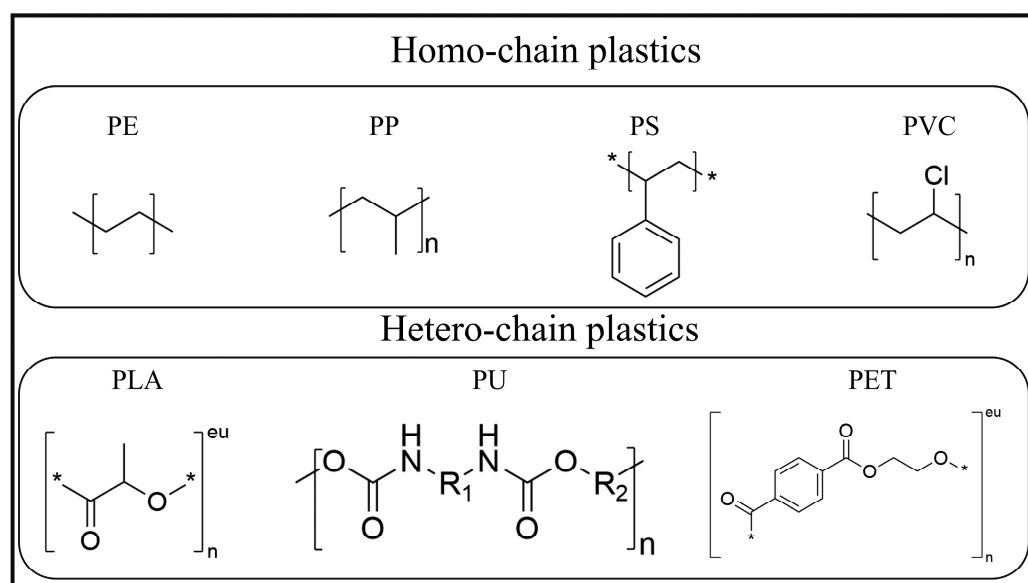


**Figure 6.** Classification of plastics and associated chemical structures.

**Table 2.** Summary of representative studies on photocatalytic conversion of PE and PP.

| Plastic | Photocatalyst                                        | Light Source                  | Main Product                                                           | Conversion | Time | Ref. |
|---------|------------------------------------------------------|-------------------------------|------------------------------------------------------------------------|------------|------|------|
| PE      | Bi <sup>0</sup> @Bi <sup>3+</sup> -KNbO <sub>3</sub> | 300 W Xe lamp                 | CO, CO <sub>2</sub>                                                    | 100%       | 12 h | [41] |
| PE      | CeO <sub>2</sub> -nPO <sub>x</sub>                   | 300 W Xe lamp                 | CO <sub>2</sub>                                                        | 94%        | 24 h | [42] |
| PE      | BiOI/BiVO <sub>4</sub>                               | 300 W LED                     | C <sub>4</sub> -C <sub>30</sub>                                        | 100%       | 6 h  | [43] |
| PE      | Pd1-TiO <sub>2</sub>                                 | 365 nm LED                    | CH <sub>3</sub> CH <sub>2</sub> COOH,<br>C <sub>2</sub> H <sub>4</sub> | 100%       | 3 h  | [44] |
| PE      | NF@CoNiFe(VZn-Al)-LDHs                               | 300 W Xe lamp                 | CH <sub>3</sub> COOH, CO <sub>2</sub>                                  | 100%       | 48 h | [45] |
| PE      | ZnIn <sub>2</sub> S <sub>4</sub>                     | 300 W Xe lamp                 | CO, CO <sub>2</sub>                                                    | 84.5%      | 60 h | [46] |
| PP      | MoS <sub>2</sub> /g-C <sub>3</sub> N <sub>4</sub>    | 300 W Xe lamp                 | ethanolic                                                              | 100%       | 24 h | [47] |
| PE      | ZrCoFe <sub>2</sub> O <sub>4</sub> QDs               | 300 W Xe lamp                 | CH <sub>3</sub> COOH                                                   | 85.4%      | 2 h  | [48] |
| UHMWPE  | FeSA-hCN                                             | 300 W Xe lamp                 | carboxylic acids,<br>ethers, alkanes,<br>furanone,<br>carboxylic acids | 100%       | 12 h | [49] |
| PE      | VPOM/CNNS                                            | Visible light<br>(λ > 420 nm) | HCOOH                                                                  | 100%       | 36 h | [50] |
| PP      | NaAlO <sub>4</sub>                                   | 350 W metal halide<br>lamp    | CO <sub>2</sub> , H <sub>2</sub> O                                     | 12.5%      | 5 h  | [51] |

#### 4.1. Overview and Conversion Strategies for Homochain Polymer Waste

Taking PE photo-reforming as an example, recent research has primarily focused on employing metal sulfides and their modified forms as photocatalysts. Recently, Yang et al. developed and reported a ZnIn<sub>2</sub>S<sub>4</sub>-based photocatalyst featuring hydroxyl functional groups and sulfur vacancies, which significantly enhanced its photocatalytic performance for the photodegradation of PE [41]. In this system, PE undergoes chain scission via the synergistic attack of ROS, such as •OH and •O<sub>2</sub><sup>−</sup>. After 60 h of light irradiation, the mass loss of PE reached 15.5% (Figure 7a). Microscopic images captured before and

after the reaction clearly reveal that the continuous structure of PE was disrupted and fragmented into smaller, irregular pieces (Figure 7b). This morphological transformation provides direct evidence of the effective degradation of PE during the photocatalytic process. Concurrently, the release rates of CO<sub>2</sub> and CO reached 1438.1 and 9.4  $\mu\text{mol}^{-1}\text{g}^{-1}$ , respectively (Figure 7c,d). Furthermore, by coupling this photocatalytic process with electrocatalytic techniques, the generated CO<sub>2</sub>/CO can be in situ converted into higher-value products such as formic acid (HCOOH), enabling efficient resource utilization and a closed-loop carbon cycle.

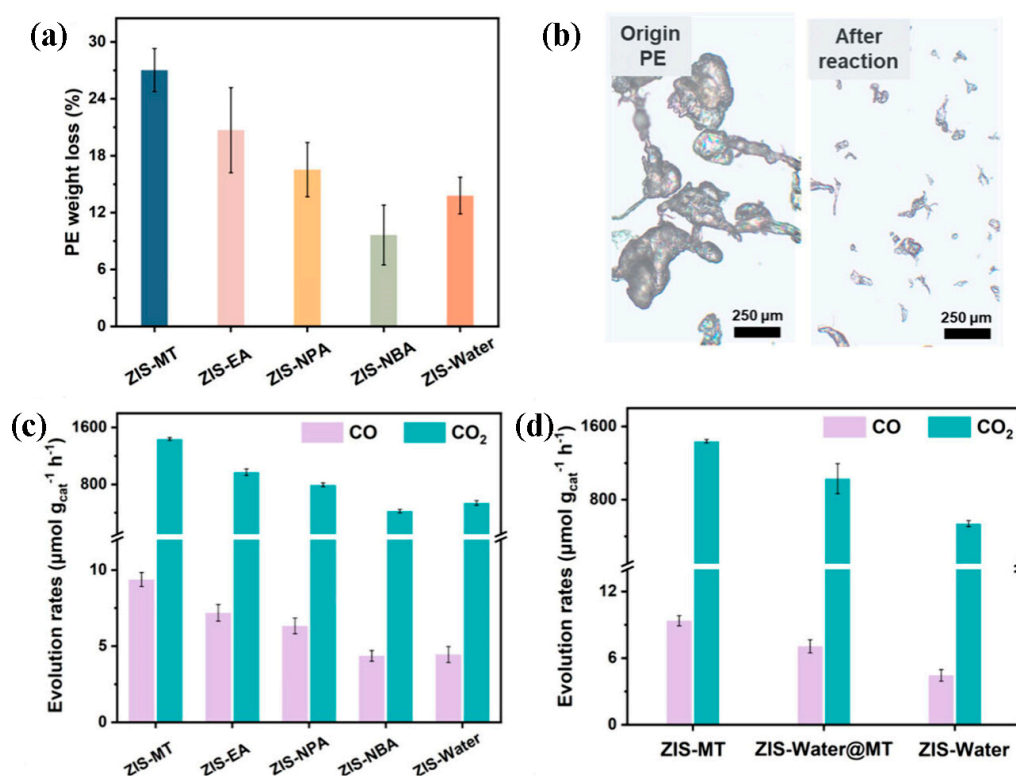


**Figure 7.** (a) Weight loss of PE after irradiation for 12 h; (b) microscopy image of PE before and after reaction for 12 h; (c) photoconversion of PE into CO<sub>2</sub> and CO over different photocatalysts; (d) photoconversion of PE via ZIS-MT, ZIS-Water@MT, and ZIS-Water [41].

In addition to metal sulfide-based photocatalysts, various other high-performance catalysts have also been reported for the photo-reforming of plastic waste. These include metal oxides, such as TiO<sub>2</sub> [45,52] and CeO<sub>2</sub> [53], as well as g-C<sub>3</sub>N<sub>4</sub> [41,50], rGO [54], and zeolite-based materials [55]. For example, Dai et al. developed a biomass-derived three-dimensional MoS<sub>2</sub>/rGO/cotton composite catalyst, which was effectively employed for the degradation of PE and further utilized for sustainable freshwater production driven by solar energy [54]. The three-dimensional rGO/cotton scaffold served as an ideal support, offering a large specific surface area and efficient electron transport pathways to uniformly disperse MoS<sub>2</sub> (Figure 8a). This structural configuration not only enhanced the exposure and utilization of active sites for effective PE degradation but also facilitated the integration of solar-driven sustainable freshwater production. As shown in Figure 8b, the mass of PE decreased by 12% after 1 h of light-driven degradation. Concurrently, optical imaging of the post-reaction sample (Figure 8c) revealed extensive surface cracking and the formation of pores, providing clear morphological evidence of significant PE degradation within this photocatalytic system. Specifically, during the photocatalytic process, PE is initially attacked by  $\bullet\text{OH}$ , leading to the cleavage of C–C bonds within its polymer chains. Simultaneously, O<sub>2</sub> participates in a series of reduction reactions, sequentially forming  $\bullet\text{O}_2^-$ . The ROS

generated in this process continuously act on PE microfibrers, further breaking down their carbon skeletons and, ultimately, mineralizing the polymer into carbon dioxide ( $\text{CO}_2$ ). However, FT-IR spectroscopy analysis revealed that characteristic functional groups of PE remained detectable in the residual materials after the reaction, indicating that the non-selective degradation process did not completely convert all PE into low-molecular-weight products (Figure 8d). During degradation, polyethylene is incompletely decomposed into carbon dioxide, microplastics, and complex mixtures of organic compounds. Selective synthesis is, to a certain degree, capable of mitigating the aforementioned problems.

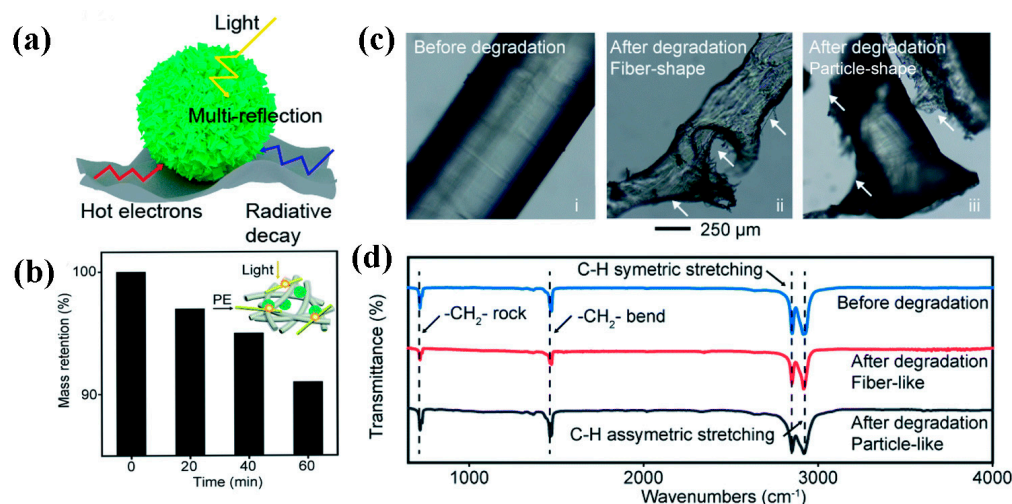


**Figure 8.** (a) A schematic illustration of the enhanced light absorption and photothermal conversion of MoS<sub>2</sub> anchored on wrinkled RGO sheets; (b) mass retention of PE with reaction time; (c) optical images; (d) corresponding FTIR spectra of the collected PE microplastics before and after degradation, confirming the component of microplastics and showing the shape-evolution [54].

Cao et al. developed a heterojunction photocatalyst composed of MoS<sub>2</sub> and C<sub>3</sub>N<sub>4</sub> (MoS<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub>), which was effectively applied to the photocatalytic transformation of PP waste [47]. As illustrated in Figure 9a, compared with conventional g-C<sub>3</sub>N<sub>4</sub> photocatalysts, the MoS<sub>2</sub>/C<sub>3</sub>N<sub>4</sub> heterojunction demonstrates superior performance in modulating the reaction pathway. This catalyst regulates the generation of ROS, particularly favoring •OH radicals, while simultaneously suppressing excessive oxidation reactions. Consequently, it facilitates the selective transformation of PP into methanol with high stability. Under the optimized conditions, the methanol yield reaches 1358.8 μmol<sup>-1</sup>g<sup>-1</sup>, with an alcohol selectivity as high as 80.3% (Figure 9b). As shown in Figure 9c, the comparison of PL spectra between C<sub>3</sub>N<sub>4</sub> and MoS<sub>2</sub>/C<sub>3</sub>N<sub>4</sub> reveals that the hybrid composite exhibits significantly enhanced emission intensity. This result suggests that the generation of •OH radicals is more efficient in the composite system. In summary, the selective conversion mechanism of PP follows the pathway depicted in Figure 9e,f: O<sub>2</sub> undergoes electron acceptance and is reduced to H<sub>2</sub>O<sub>2</sub> on the MoS<sub>2</sub> surface, H<sub>2</sub>O<sub>2</sub> then decomposes to generate •OH. The •CH<sub>3</sub> intermediate is subsequently transformed into •CH<sub>3</sub>OH under the influence of •OH radicals, and overoxidation is avoided due to the relatively low concentration of other strong oxidizing radicals in the system. Finally, •CH<sub>3</sub>OH desorbs from the catalyst surface, yielding the target product CH<sub>3</sub>OH.

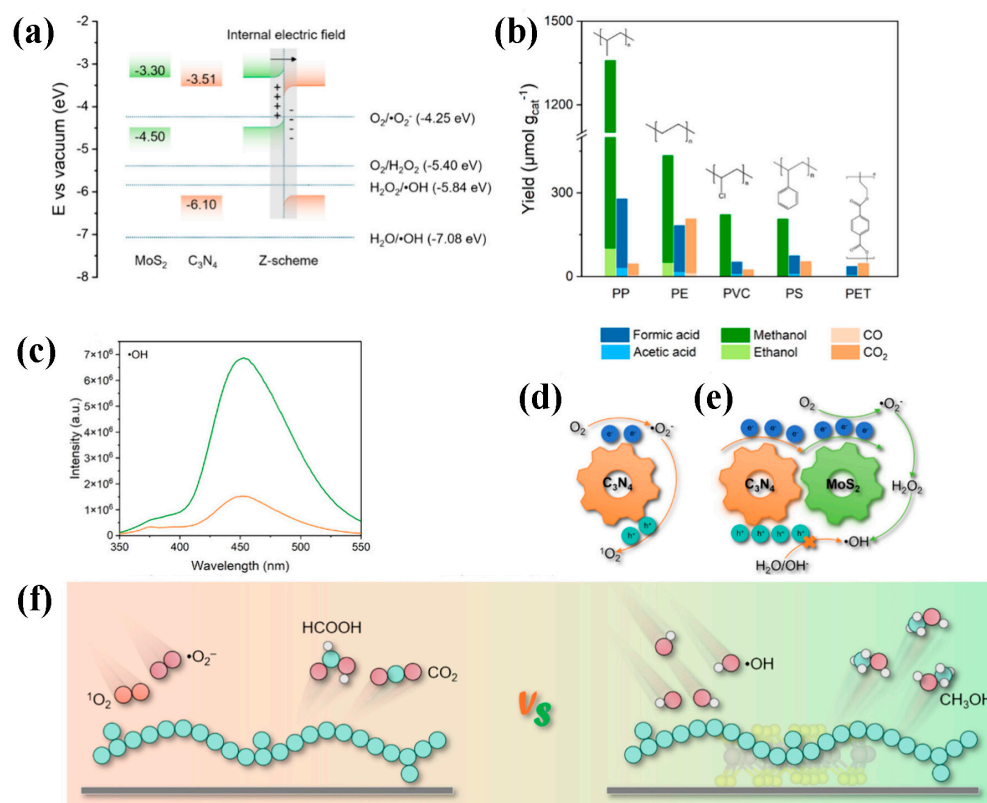


**Figure 9.** (a) Band structures of MoS<sub>2</sub> and C<sub>3</sub>N<sub>4</sub> with the redox potential of typical radicals; (b) conversion of different plastics over MoS<sub>2</sub>/C<sub>3</sub>N<sub>4</sub>; (c) PL spectra of coumarin solutions with C<sub>3</sub>N<sub>4</sub> and MoS<sub>2</sub>/C<sub>3</sub>N<sub>4</sub> photocatalysts. Schematic illustration of ROS formation in the presence of (d) C<sub>3</sub>N<sub>4</sub> and (e) MoS<sub>2</sub>/C<sub>3</sub>N<sub>4</sub>. (f) Schematic of the conversion of PP to HCOOH/CO<sub>2</sub> and CH<sub>3</sub>OH on C<sub>3</sub>N<sub>4</sub> and MoS<sub>2</sub>/C<sub>3</sub>N<sub>4</sub>, respectively [47].

In addition to PE and PP, the photoreforming of other non-polyolefin plastics, such as PS and PVC, has also been extensively studied (Table 3). As an example, Zhang and co-workers developed a composite material (CDs/Zr-MOF) by encapsulating carbon dots within a zirconium-based metal–organic framework, intended for the photocatalytic decomposition of PVC (Figure 10a) [56]. Under light irradiation, the CDs/Zr-MOF catalyst achieved a 76.5% conversion rate of PVC, with acetic acid being produced at a yield of 14% (Figure 10b,c). A series of comparative experiments further highlighted the superior recyclability and catalytic efficiency of the CDs/Zr-MOF system for PVC transformation, as illustrated in (Figure 10d,e). Quenching experiments combined with ESR analysis revealed that •O<sub>2</sub><sup>-</sup> and •OH act as key ROS in the photocatalytic conversion of PVC.

**Table 3.** Summary of the photocatalytic conversion of PS and PVC in recent studies.

| Plastic | Photocatalyst                   | Light Source  | Main Product                       | Ref. |
|---------|---------------------------------|---------------|------------------------------------|------|
| PVC     | CDs/Zr-MOF                      | AM 1.5        | CH <sub>3</sub> COOH               | [56] |
| PVC     | Ni-TCPP                         | AM 1.5        | diesel olefin products             | [57] |
| PS      | g-C <sub>3</sub> N <sub>4</sub> | 300 W Xe lamp | benzoic acid                       | [58] |
| PS      | Triton X-100 TiO <sub>2</sub>   | 32 W lamp     | CO <sub>2</sub> , H <sub>2</sub> O | [59] |

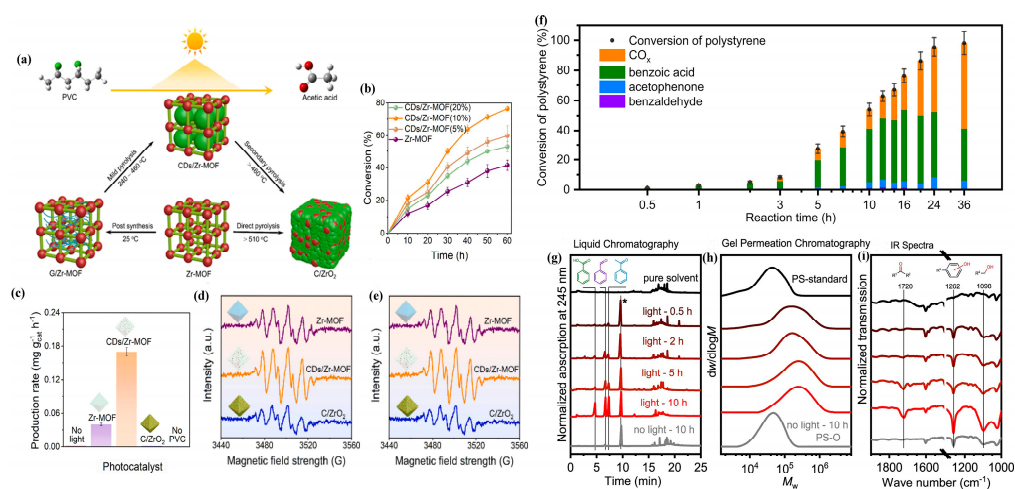


**Figure 10.** (a) The structural transformation of Zr-MOF into G/Zr-MOF, CD/Zr-MOF, and C/ZrO<sub>2</sub> by post-synthesis, direct pyrolysis, mild pyrolysis, and subsequent secondary pyrolysis treatment, respectively. The resultant CD/Zr-MOF displays high activity in the upcycling of PVC plastics toward acetic acid; (b) PVC conversion activity over Zr-MOF and CD/Zr-MOF (X); and (c) PVC conversion rates under C/ZrO<sub>2</sub>, Zr-MOF, and CD/Zr-MOF, with no light and no catalyst reaction systems, respectively. ESR spectra of (d) DMPO-•O<sub>2</sub><sup>−</sup> and (e) DMPO-•OH for C/ZrO<sub>2</sub>, Zr-MOF and CD/Zr-MOF [56]. (f) Time-evolution of different products for polystyrene photocatalytic oxidation reaction at 150 °C under 10 bar O<sub>2</sub>, 50 mg g-C<sub>3</sub>N<sub>4</sub>, 10 mg polystyrene (*M<sub>w</sub>* ~ 50 kDa), 30 mL acetonitrile, and 300 W Xenon lamp. Distribution of different products are shown by column with different colors. The error bars represent the standard deviation of conversion in 3 parallel experiments. (g) Liquid chromatograms of reaction solutions with different time and conditions. The first line pure solvent refers to the signal of pure solvent. The peaks marked with an asterisk represent the nitrobenzene internal standard. (h) Molecular weight distribution measured by gel permeation chromatography (GPC) and (i) the infrared (IR) transmission spectra of the polystyrene reactant and the recovered polystyrene after reactions with light irradiation for 0.5 h, 2 h, 5 h, and 10 h and without light irradiation for 10 h [58].

Ma and his team employed heterogeneous graphitic carbon nitride (g-C<sub>3</sub>N<sub>4</sub>) as a catalyst and successfully developed a novel strategy for the oxidation of PS into oxygenated aromatic compounds under visible light irradiation [58]. At a reaction temperature of 150 °C, PS conversion surpassed 90%, yielding primarily benzoic acid, acetophenone, and benzaldehyde in the liquid phase, with an overall product selectivity above 80%. As illustrated in Figure 10f, the reaction exhibited a pronounced induction period during the initial 3 h, followed by a rapid accumulation of products between 3 and 24 h. During the induction phase, only trace amounts of benzoic acid, acetophenone, and benzaldehyde were detected. Liquid chromatography analysis revealed no substantial formation of polystyrene-derived oligomers, dimers, or trimers (Figure 10g). Concurrently, analysis of the solid fraction showed that the molecular weight of the residual polystyrene increased within the first 30 min and continued to rise throughout the reaction, rather than undergoing cleavage into lower-molecular-weight (*M<sub>w</sub>*) short-chain intermediates (Figure 10h). Furthermore, during the main product formation window (3–10 h), neither a notable number of oligomers, dimers, or related derivatives were detected (Figure 10g), nor was any decrease observed in the molecular weight of the recovered polystyrene (Figure 10h). These results collectively suggest that during the induction period, polystyrene does not follow a depolymerization pathway involving the formation of soluble oligomeric intermediates. Infrared (IR) spectral analysis (Figure 10i) indicates that the characteristic C–O stretching bands associated with phenolic and alcoholic hydroxyl groups had already emerged during the induction stage.

#### 4.2. Photocatalytic Conversion of Heterochain Plastics

In contrast to carbon-chain-only polymers, plastics with heteroatom-containing backbones (such as PET, PU, and PLA) are characterized by strong polar bonds and enhanced structural robustness, which render them less susceptible to efficient photocatalytic breakdown. As summarized in Table 4, recent advances in photocatalytic reforming of polyester-based plastics coupled with H<sub>2</sub>O reduction reveal that many systems rely on harsh pretreatment conditions. Typical strategies include alkaline hydrolysis using NaOH concentrations ranging from 0.1 to 10 M, or hydrothermal treatments at elevated temperatures. Consequently, their degradation proceeds more slowly and imposes greater demands on both catalyst performance and reaction conditions [20,60,61]. This section focuses on recent studies and advancements concerning the selective photo-reforming of heteroatom-containing plastics into H<sub>2</sub> and value-added organic products.

**Table 4.** Summary of recent studies on plastic pretreatment coupled with photocatalytic H<sub>2</sub> evolution: key materials and process conditions.

| Plastic    | Photocatalyst                                         | Pretreatment Conditions                             | Light Source          | H <sub>2</sub> Yield<br>(mmol H <sub>2</sub> g <sub>sub</sub> <sup>−1</sup> h <sup>−1</sup> ) | Main Product                                        | Ref. |
|------------|-------------------------------------------------------|-----------------------------------------------------|-----------------------|-----------------------------------------------------------------------------------------------|-----------------------------------------------------|------|
| PET<br>PLA | CdS/CdO <sub>x</sub>                                  | NaOH 10 M                                           | AM 1.5 G              | 12.4 ± 2.0<br>64.3 ± 14.7                                                                     | glycolate, ethanol,<br>acetate, lactate<br>pyruvate | [62] |
| PET<br>PLA | CN <sub>x</sub>   Ni <sub>2</sub> P                   | NaOH 2 M                                            | AM 1.5                | 0.14<br>0.13                                                                                  | acetate, formate,<br>glycolate<br>formate, acetate  | [63] |
| PET<br>PLA | MoS <sub>2</sub> /CdS                                 | NaOH 10 M                                           | AM 1.5 G              | 3.90 ± 0.07<br>6.68 ± 0.10                                                                    | formate, acetate,<br>glycolate<br>formate           | [64] |
| PET        | Pt@N-TiO <sub>2-x</sub>                               | hydrothermal<br>pretreatment at<br>180 °C for 12 h. | AM 1.5 G              |                                                                                               |                                                     | [65] |
| PET        | MoS <sub>2</sub> /Cd <sub>x</sub> Zn <sub>1-x</sub> S | NaOH 10 M                                           | AM 1.5 G              | 15.90                                                                                         | formate, methanol,<br>acetate, ethanol              | [66] |
| PET        | MoS <sub>2</sub> /g-C <sub>3</sub> N <sub>4</sub>     | NaOH 2 M                                            | AM 1.5 G              | 0.0498                                                                                        | formate, CH <sub>4</sub>                            | [67] |
| PET        | CuIn <sub>5</sub> S <sub>8</sub>                      | KOH 1.5 M                                           | AM 1.5 G              | 2.57 ± 0.02                                                                                   | formate, glycolate,<br>acetate                      | [68] |
| PET        | Pt/g-C <sub>3</sub> N <sub>4</sub>                    | NaOH 0.1M                                           | AM 1.5 G              | 2.00                                                                                          | formate, methanol,<br>acetate                       | [69] |
| PET        | B-doped<br>g-C <sub>3</sub> N <sub>4</sub>            | NaOH 0.1M                                           | Xe lamp<br>λ > 420 nm | 3.24                                                                                          | formic acid, acetic<br>acid, glycolic acid          | [70] |
| PET        | BiVO <sub>4</sub> /MoO <sub>x</sub>                   | KOH 1 M                                             | Xe lamp               | 1.60 ± 0.09                                                                                   | formic acid, acetic<br>acid                         | [71] |

The Reisner group reported a cost-effective CdS/CdO<sub>x</sub> quantum dot catalyst capable of driving plastic photoreforming reactions under alkaline conditions, enabling efficient H<sub>2</sub> production [62,72]. As shown in Figure 11a, under visible light irradiation, heteroatom-containing plastics, such as PET, PLA, and PU, can undergo oxidation via photogenerated holes, leading to the formation of value-added organic products, such as acetate, pyruvate, and formate. Simultaneously, the photogenerated electrons reduce protons in the solution to produce H<sub>2</sub>. The long-term photo-reforming of PET bottles from real-life sources was performed to assess the performance and durability of the catalyst under realistic conditions (Figure 11b). However, due to the toxicity of Cd, which limits its compliance with environmental and sustainability standards for practical applications, it has been replaced by a more affordable, non-toxic, and environmentally benign carbon nitride/nickel phosphide (CN<sub>x</sub> | Ni<sub>2</sub>P) photocatalyst [63]. Under alkaline aqueous conditions, this catalytic system enables the efficient conversion of PET and PLA into value-added organic chemicals and

hydrogen fuel. TEM analysis revealed that  $\text{Ni}_2\text{P}$  nanoparticles, with an average diameter of approximately 9.4 nm, are uniformly dispersed on the surface of carbon nitride ( $\text{CN}_x$ ) (Figure 11c). To enhance the reaction activity during plastic photo-reforming, researchers systematically optimized several key parameters, including the loading ratio of  $\text{Ni}_2\text{P}$  to  $\text{CN}_x$ , the pH of the reaction system, and the pretreatment conditions of the plastic substrate. The  $\text{CN}_x/\text{Ni}_2\text{P}$  photocatalyst enables efficient photo-reforming of PET and PLA when pretreated plastics (stirred in 2 M KOH aqueous solution at 40 °C for 24 h) are used as the substrate. In this process,  $\text{CN}_x$  absorbs light energy and excites electrons, which then migrate to the  $\text{Ni}_2\text{P}$  co-catalytic sites, where they drive proton reduction to generate  $\text{H}_2$ . Simultaneously, the plastic serves as an electron donor and undergoes oxidation, yielding corresponding small-molecule organic products (Figure 11d,e). To evaluate the practical applicability of the  $\text{CN}_x/\text{Ni}_2\text{P}$  catalytic system under real-world conditions, five-day photo-reforming experiments were conducted on various real plastic samples, including polyester microfibers, commonly used PET bottles, and oil-contaminated PET. The results demonstrated hydrogen evolution rates of  $104.22 \pm 10$ ,  $22 \pm 1.3$ , and  $11.4 \pm 1.2 \mu\text{mol}_{\text{H}_2} \cdot \text{g}^{-1}$  for the three types of plastics, respectively (Figure 11f,g). These findings indicate that  $\text{CN}_x/\text{Ni}_2\text{P}$  possesses strong reactivity and practical potential for the treatment of real-world plastic waste.

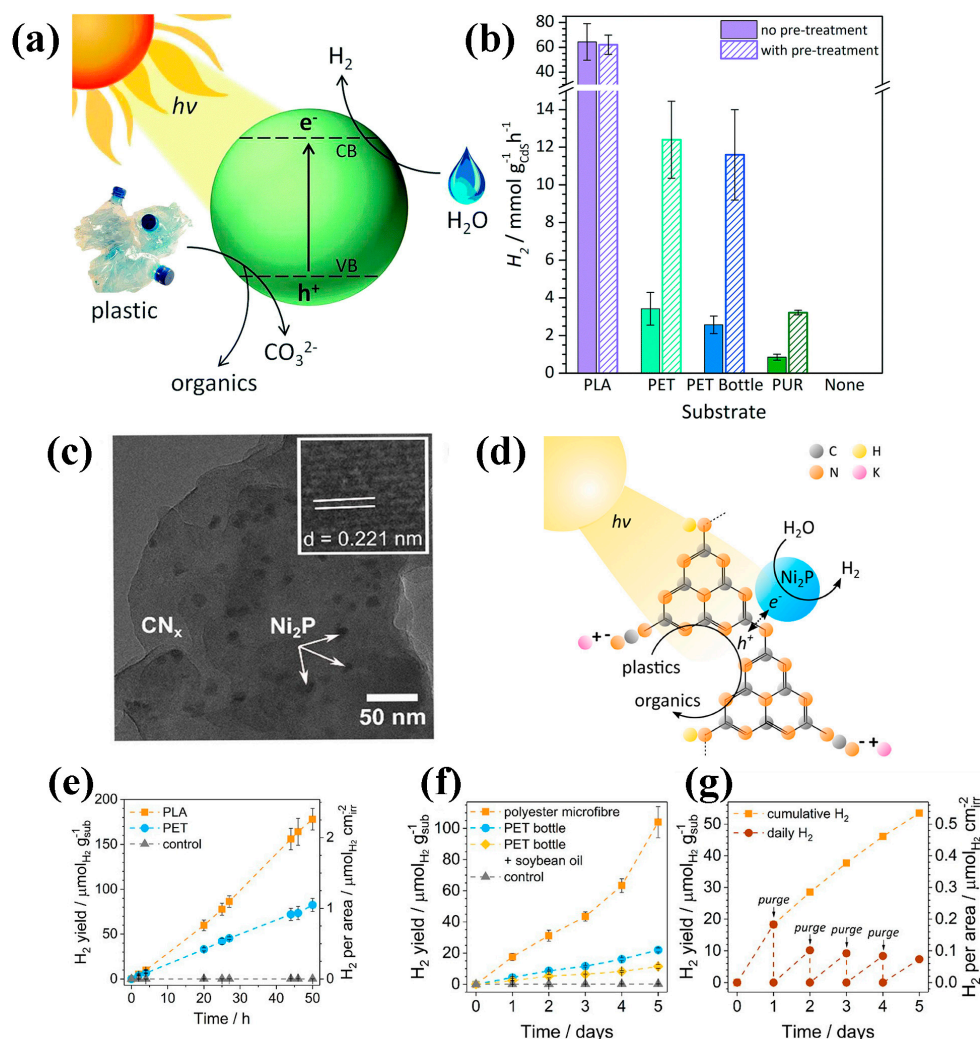


**Figure 11.** (a) Diagram of the polymer photo-reforming process with a  $\text{CdS}/\text{CdO}_x$  quantum dot photocatalyst in alkaline aqueous solution. (b) Photo-reforming of polymers to  $\text{H}_2$  using  $\text{CdS}/\text{CdO}_x$  (1 nmol) under simulated solar light (4 h, AM 1.5G,  $100 \text{ mW cm}^{-2}$ , 25 °C). Conditions: powdered plastic

(50 mg mL<sup>-1</sup> PLA, 25 mg mL<sup>-1</sup> PET, PET bottle or PUR) freshly prepared (no pretreatment) or pretreated in 10 M aq. NaOH (2 mL) [62]. (c) TEM image of the CN<sub>x</sub> | Ni<sub>2</sub>P catalyst, with inset showing the lattice spacing of Ni<sub>2</sub>P. (d) Schematic diagram of the polymer photo-reforming process using a CN<sub>x</sub> | Ni<sub>2</sub>P photocatalyst. (e) Long-term photo-reforming of PET and PLA. Conditions unless stated otherwise in figure: CN<sub>x</sub> | Ni<sub>2</sub>P 2 wt % (1.6 mg mL<sup>-1</sup>), pretreated PET (25 mg mL<sup>-1</sup>), aqueous KOH (1 M, 2 mL), and simulated solar light (AM 1.5 G, 100 mW cm<sup>-2</sup>, 25 °C). Photo-reforming of nonrecyclable plastic waste. (f) Long-term photo-reforming of polyester microfibers, a PET bottle, and an oil-coated PET bottle. (g) Upscaled photo-reforming of polyester microfibers; sample was purged every 24 h. Conditions: CN<sub>x</sub> | Ni<sub>2</sub>P (1.6 mg mL<sup>-1</sup>), 1 M KOH (2 mL for part a and 120 mL for part b), pretreated microfibers (5 mg mL<sup>-1</sup>) or PET bottle (25 mg mL<sup>-1</sup>) without or with soybean oil (5 mg mL<sup>-1</sup>), and simulated solar light (AM 1.5G, 100 mW cm<sup>-2</sup>) [63].

The aforementioned photo-reforming systems typically rely on harsh pretreatment conditions for plastics, which significantly increases the overall processing cost and may compromise the stability of the photocatalyst. Moreover, the generation of highly alkaline waste during pretreatment imposes an additional burden on downstream processing, limiting the feasibility of this technology for large-scale practical applications [33,73–75]. Recently, Zhang and colleagues reported a novel tandem catalytic system comprising a dinuclear zinc molecular catalyst and carbon nitride nanosphere photocatalyst. This system enables efficient chemical depolymerization of PET under mild conditions, followed by photo-reforming of the depolymerized products into value-added chemicals and H<sub>2</sub> (Figure 12a) [69]. As shown in Figure 12b, the Zn<sub>2</sub>-complex serves as an effective catalyst for PET hydrolysis, achieving complete depolymerization within 48 h under reaction conditions of 60 °C and 0.1 M NaOH. Comparative results (Figure 12c) indicate that under identical conditions, the catalytic efficiency of the Zn<sub>2</sub>-complex is approximately ten times higher than that of the uncatalyzed system. The high catalytic efficiency is primarily attributed to the proximity effect between the two metal centers. This effect enhances the local concentration of the substrate and nucleophile near the catalytic centers while reducing the spatial separation between them, accelerating the reaction rate and improving overall catalytic efficiency (Figure 12d). FT-IR spectral analysis (Figure 12e) revealed the formation of a coordination bond (C=O–Zn) between the carbonyl oxygen in EB and the zinc ion in the Zn<sub>2</sub>-complex during the reaction, confirming the critical role of the catalyst in substrate activation. In addition, the dinuclear Zn complex mimics enzymatic behavior in polyester hydrolysis, demonstrating both selective and efficient catalytic performance. The system achieved a H<sub>2</sub> production rate of 100 μmol h<sup>-1</sup> within 4 h, indicating that mild pretreatment of PET enhances photoreforming efficiency.

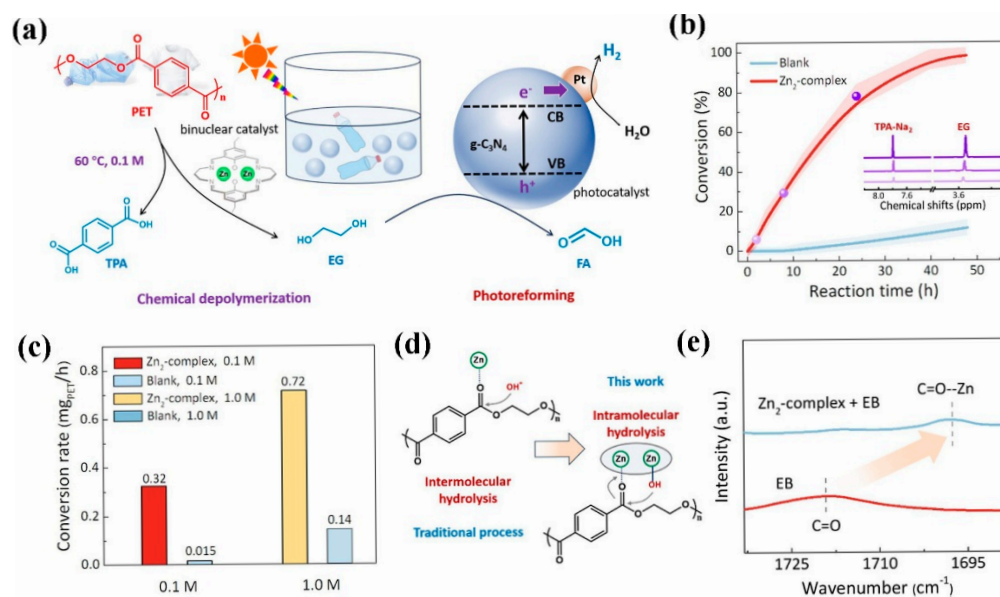


**Figure 12.** (a) Schematic illustration of the tandem chemical depolymerization and photo-reforming of waste PET plastics. (b) Hydrolysis kinetics of the PET granule (10 mg) over the Zn<sub>2</sub>-complex (4 mg) and blank under 0.1 M NaOH and 60 °C. The inset depicts the <sup>1</sup>H NMR evolution at different reaction times over the Zn<sub>2</sub>-complex. (c) Comparison of the conversion rate in different alkali concentration solutions over the Zn<sub>2</sub>-complex and blank at 60 °C. (d) Intramolecular hydrolysis mechanism of PET over the Zn<sub>2</sub>-complex. (e) DRIFTS spectra of ethyl benzoate over the Zn<sub>2</sub>-complex [69].

## 5. Photocatalytic Product Selectivity

Efficiency and selectivity are two key metrics for evaluating the performance of photocatalytic plastic conversion. However, the field remains in an early exploratory stage, and current catalytic systems still exhibit significant limitations in reaction activity, product specificity, and scalability. Drawing on insights from more established photocatalytic applications such as water splitting and CO<sub>2</sub> reduction, we propose several feasible strategies to simultaneously enhance the efficiency and product selectivity of photocatalytic plastic conversion.

Currently, optimizing the structural design of photocatalysts and precisely tuning the reaction system conditions are widely regarded as key strategies for enhancing the selectivity of photocatalytic reactions [76]. In this context, the regulation of external reaction conditions primarily involves the selection of solvent type, adjustment of pH, and control of the reaction atmosphere (e.g., air, oxygen, or inert gases). The design of photocatalysts typically focuses on tuning their band structure, particularly the positions of the VB and CB, in order to suppress the generation of highly reactive radicals such as •OH, preventing non-selective oxidation and improving both product selectivity and reaction controllability. For example, Yang et al. recently reported a nitrogen and phosphorus (N, P) co-doped porous Mo<sub>2</sub>C photocatalyst. This photocatalytic system activates peroxymonosulfate (HSO<sub>5</sub><sup>−</sup>) on the catalyst surface to generate singlet oxygen (<sup>1</sup>O<sub>2</sub>), enabling selective oxidation of ethylene glycol (EG), a hydrolysis product of PET, under <sup>1</sup>O<sub>2</sub>-mediated mild oxidative conditions. This process efficiently produces formate as the major product [77]. This strategy enhances reaction selectivity while effectively avoiding non-specific mineralization pathways induced by highly oxidative radicals. The integration of different heterojunction types, especially those engineered for band alignment such as Z-scheme heterojunctions, effectively promotes the separation of charge carriers while enhancing redox capacity, thus contributing to improved photocatalytic conversion efficiency. Additionally, the application of external stimuli—including electric bias, magnetic fields, thermal energy, and ultrasonic

irradiation—can facilitate the activation of reactant molecules by accelerating the migration and separation of photogenerated carriers, thereby further boosting photocatalytic performance. Given that photocatalytic reactions primarily occur on semiconductor surfaces, the regulation of surface-active sites has become one of the key focuses in current research. For instance, molecular recognition sites (MRSs) are a class of surface structures capable of selectively adsorbing target molecules, forming surface complexes through spatially defined configurations and tailored chemical environments. This mechanism not only facilitates the enrichment of specific reactants but also directs them to preferentially participate in catalytic reactions, enabling the selective conversion of organic pollutants or plastic degradation intermediates, and significantly improving the reaction precision and product selectivity of photocatalytic processes [78,79]. Defect engineering and crystal structure modulation have been widely employed to optimize the surface properties and crystallinity of photocatalysts, thereby enhancing their interaction with reactants and improving the selectivity toward target product formation. By tuning the type of surface functional groups and the surface charge distribution, the adsorption behavior of both reactants and products on the photocatalyst can be significantly altered, enabling precise control over the reaction pathway [80]. Moreover, the introduction of cocatalysts with suitable bandgap energies, surface chemistries, and structural morphologies, or the construction of composite systems through synergistic integration with co-catalytic components, not only facilitates more efficient utilization of photogenerated electrons but also promotes the generation of hydrogen and the selective synthesis of organic products [14]. For example, metal doping has shown notable effectiveness in tuning Pd catalytic sites and has been demonstrated to substantially improve the selective oxidation of polyol molecules. However, the inherently low affinity of Pd surfaces for oxygen species limits the effective adsorption of  $\text{OH}^-$  and the subsequent formation of adsorbed hydroxyl radicals ( $\cdot\text{OH}_{\text{ad}}$ ), reducing overall catalytic conversion efficiency. Studies have shown that the introduction of oxygen-affinitive moieties into Pd catalytic centers can enhance  $\text{OH}^-$  surface adsorption, which facilitates reactant activation and transformation, ultimately leading to significantly improved oxidative catalytic activity and reaction kinetics [81].

## 6. Future Directions in Photocatalytic Plastic Conversion

In recent years, significant progress has been made in the field of photocatalytic plastic conversion. The research focus has gradually shifted from early-stage efforts aimed at complete mineralization toward more valuable selective synthesis pathways. Through catalyst structural optimization, modulation of reaction pathways, and control over product selectivity, the field has achieved high conversion efficiencies while progressively improving selectivity toward target products, such as hydrogen, organic acids, and alcohols. This marks a critical transition in photocatalytic plastic utilization—from simple degradation to value-added resource recovery. Despite recent breakthroughs, the field of photocatalytic plastic conversion remains in its early stages and faces several significant challenges. Existing catalysts require further optimization in terms of light absorption range, charge carrier separation efficiency, and reaction stability. In addition, poor selectivity in product formation restricts access to value-added chemicals in a controlled manner. Furthermore, transitioning from laboratory research to industrial deployment requires substantial improvements in catalyst scalability, system energy efficiency, and cost-effectiveness. These improvements are essential to enhance the overall economic viability and practicality of the process. Addressing these challenges will be critical for the successful implementation of this technology in real-world settings. The following points outline the key directions and strategies for future development in the field of photocatalytic plastic conversion:

1. Rational design and optimization of photocatalysts. The performance of photocatalysts plays a decisive role in the construction of efficient and stable systems for plastic photo-reforming. To achieve efficient conversion of plastic waste, photocatalysts must be precisely engineered across multiple dimensions, including morphology (e.g., nanoparticles, thin films, and hollow structures), composition (e.g., multicomponent composites and metal/non-metal doping), and crystal structure with surface electronic properties. These strategies aim to enhance solar light harvesting, improve the separation and migration efficiency of photogenerated charge carriers, and increase the number of reactive sites, maximizing the efficient conversion of solar energy into chemical energy. Advanced catalytic systems have opened new avenues for the development of photocatalytic plastic reforming. Systems such as single-atom photocatalysts (SACs), molecularly coordinated heterogeneous catalysts, and covalent organic frameworks (COFs) offer distinct structural and electronic features that show tremendous promise in improving photocatalytic efficiency and fine-tuning product selectivity. The recyclability and reusability of photocatalysts are critical for their practical application, especially in systems involving noble metal cocatalysts. To prolong catalyst lifetime and reduce overall reaction costs, several key design and development strategies should be considered, as follows: 1. development of stable and efficient catalyst supports, 2. construction of recyclable catalyst architectures, 3. prevention of noble metal deactivation and loss, and 4. enhancement of structural resistance to photo-corrosion. Improving structural stability under harsh redox conditions enables long-term operation without significant deactivation, ultimately reducing replacement frequency and operational costs.

2. Regulation of product selectivity. Photo-reforming of plastics is a multi-step process with a complex mechanism, typically involving oxidative cleavage of polymer chains along with the reduction of  $O_2$  and  $H_2O$ . Reactive oxygen species ( $\bullet OH$ ,  $\bullet O_2^-$ , and  $^1O_2$ ) formed in this process are highly oxidative, which facilitates the breakdown of the otherwise stable polymer matrix but also contributes to diverse reaction pathways, leading to a wide range of intermediates and products. This non-selectivity often leads to significant byproduct formation and a convoluted product spectrum, which poses substantial challenges for subsequent analysis, separation, and purification processes. Therefore, a deep mechanistic understanding of the formation pathways of target products and the involvement of side reactions is crucial for accurately elucidating the photo-reforming process and enhancing product selectivity. The active sites of the catalyst play a central role in this process; they govern the adsorption strength and orientation of plastics or their degradation intermediates on the catalyst surface and directly influence electronic structure rearrangements and subsequent reaction pathways. Particularly, active sites with well-defined geometric or electronic configurations can selectively generate specific reactive oxygen species (e.g., favoring  $\bullet OH$  formation or suppressing  $\bullet O_2^-$  generation), which allows for fine control over oxidation intensity and product profiles, favoring the desired output and minimizing undesired byproducts. In addition, reaction media (e.g., water, alcohols, or alkaline aqueous mixtures) and gas atmosphere (e.g., inert gas, air, or pure  $O_2$ ) significantly affect the generation and stability of reactive species, thermodynamic driving forces, and product release behavior. Rational design and control of these external parameters can substantially improve both reaction selectivity and efficiency. In summary, achieving controllable synthesis of desired products in plastic photo-reforming requires synergistic optimization of catalyst design, reaction condition selection, and mechanistic understanding.

3. Mechanistic insights and multiscale analysis. A comprehensive understanding of the reaction mechanisms underlying plastic conversion is a fundamental prerequisite for improving photocatalytic efficiency and product selectivity. Mechanistic elucidation facilitates the identification of rate-determining steps in the catalytic process and provides

theoretical guidance for tailoring surface properties and structural features of photocatalysts. This, in turn, affects the adsorption/desorption behavior of reactants, intermediates, and products, enabling precise control over reaction pathways and product distributions. Plastic degradation typically involves the stepwise transformation of various intermediates, such as aldehydes, ketones, and radical species, whose formation and conversion rates directly determine the type, selectivity, and yield of the final products. To monitor these complex processes, advanced in situ characterization techniques, such as FTIR, XPS, and EPR, along with isotope-labeling experiments, are widely employed to track dynamic reactions and identify transient intermediates for accurate pathway elucidation. In parallel, DFT calculations allow for the in-depth exploration of electronic interactions between catalysts and reactants, including adsorption energetics and reaction barrier profiles, to predict feasible transformation pathways and product formation mechanisms. The synergy between theoretical modeling and experimental observation not only elucidates the origin of product selectivity but also enables feedback-driven design of active sites with tailored reactivity. Future advances will rely on integrating multiscale models (from electrons to interfaces) with state-of-the-art characterization and computation, enabling a leap from passive product formation to tunable plastic upcycling. This approach will lay a robust theoretical foundation for the design of next-generation photocatalysts and provide scientific guidance for the controlled industrialization of photocatalytic plastic conversion.

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