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Photocatalytic Upcycling of Plastic Waste

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Plastics are indispensable in modern life due to their mechanical strength, chemical resistance, and processing flexibility. Global production exceeds 400 million metric tons annually and is projected to surpass 800 million metric tons by 2050, while less than 10% of post-consumer plastic waste is recovered through closed-loop recycling streams [1]. Traditional processing techniques—particularly mechanical methods—have fundamental limitations, often producing downcycled materials with diminished properties and high life-cycle emissions. In contrast, chemical degradation has gained increasing attention as a means of closing the plastic loop by converting waste into monomers or high-value chemicals [2]. Among innovative chemical strategies, photocatalytic upcycling offers distinctive advantages, including operation under ambient conditions, utilization of solar energy as a renewable input, and high molecular selectivity, making it a strategically promising approach to advance plastic circularity while minimizing environmental impact [3].

Building upon this conceptual foundation, recent advances have focused on designing photocatalytic frameworks capable of facilitating selective bond cleavage in plastic macromolecules under light irradiation [4]. Plastics, which are categorized into seven classes under the Resin Identification Code (RIC) system, present a range of chemical motifs such as C–C, C–O, and C–S bonds that dictate their stability and degradation pathways. By leveraging these structural features, researchers have developed photocatalytic systems that enable selective bond cleavage within complex polymer matrices, thereby facilitating controlled depolymerization into valuable chemical intermediates. Such intermediates include specialty monomers, bioactive compounds, and functional oligomers, which may serve as terminal products or synthetic precursors for constructing next-generation materials. Collectively, these advances establish a transformative platform for precision photochemical conversion in closed-loop plastic utilization (Fig. 1).

1. Photoinduced electron-transfer-driven depolymerization

Photoinduced electron-transfer strategies offer a powerful mechanism for polymer degradation, especially when targeting chemically robust thermoplastics and thermosets. Jiang et al. [5] demonstrated this principle through photoexcited uranyl cations, which mediate a multi-step mechanism involving single-electron transfer (SET), hydrogen atom transfer (HAT), and oxygen atom transfer (OAT). Leveraging scalable continuous-flow photoreactors, this approach enabled the depolymerization of nine types of plastics, including polystyrene (PS), polyethylene terephthalate (PET), and polyethersulfone (PES), into monomers and high-value chemicals (up to 88% yield).

While such strategies excel in linear thermoplastic recycling, the chemical recycling of thermosetting plastics remains a major challenge due to their permanently crosslinked architectures. Knowles et al. [6] developed a photocatalytic strategy leveraging proton-coupled electron transfer (PCET) to achieve the complete depolymerization of thiol–epoxy thermosets and the quantitative recovery of monomeric building blocks. In their system, hydroxyl groups embedded in the thermoset matrix undergo visible-light-induced PCET, generating alkoxy radicals that initiate β-scission at adjacent C–C bonds. This mechanism enables the ambient-condition depolymerization of intractable thermoset matrices through synergistic cascades of photoinduced PCET and β-scission, converting crosslinked networks into small-molecule products. These intermediates are subsequently subjected to dealkylation, achieving the high-yield regeneration of bisphenol A and other native monomers. The process is precision-engineered via the judicious selection of organic acridinium photocatalyst, thiyl radical mediators, and aprotic reaction media to maximize both bond-cleavage fidelity and architectural selectivity.

2. Photooxidative depolymerization via chlorine radicals

Beyond photoinduced electron-transfer strategies, radical pathways in photocatalytic oxidation have emerged as a pivotal route for the selective transformation of hydrocarbon-based polymers, enabling the conversion of inert plastic chains into high-value aromatic compounds. This strategy typically relies on the generation of chlorine radicals or metal-centered oxidants under visible light, which initiate site-specific C–H bond scission via HAT or initiate C–C bond cleavage through homolytic pathways. Compared with thermal oxidation, this approach offers lower activation-energy requirements and enhanced chemoselectivity, making it a promising route for depolymerizing otherwise recalcitrant polymers under mild conditions.

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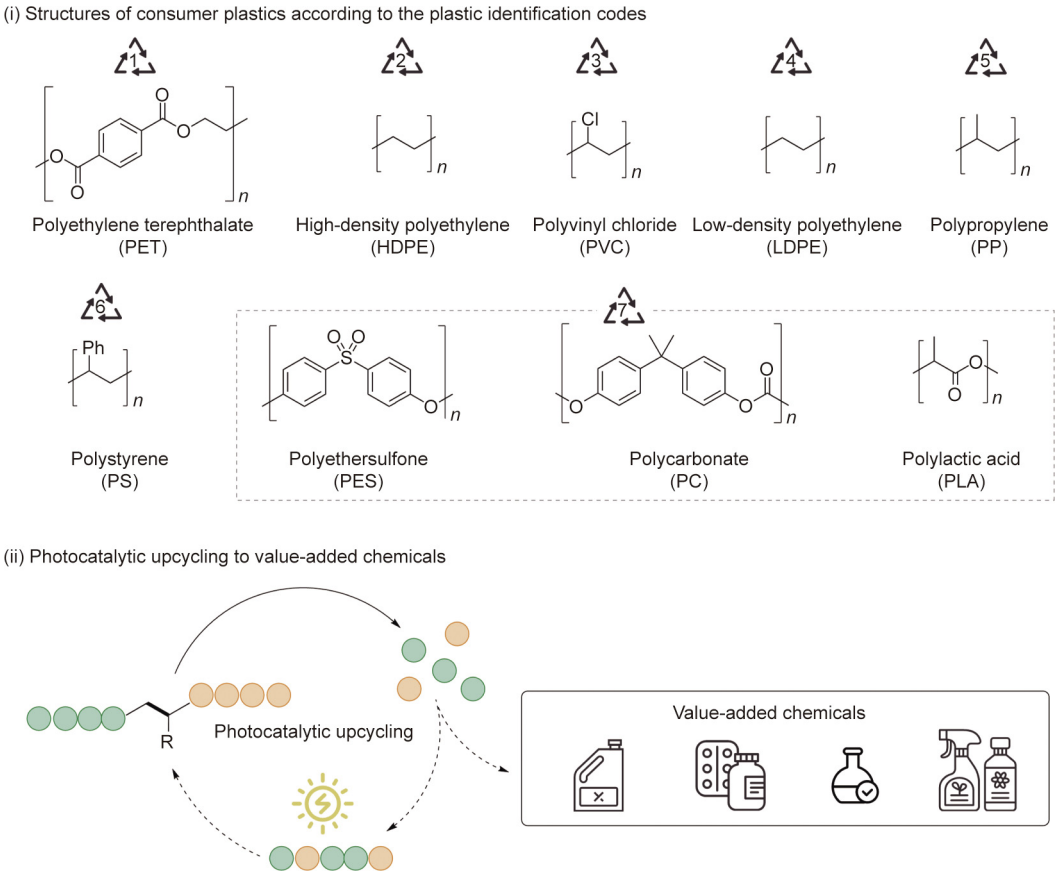

Fig. 1. Photocatalytic platforms for selective plastic degradation and upcycling. Ph: phenyl.

In this context, Stache et al. [7] developed an iron(III) chloride ($FeCl_3$)-catalyzed photooxidative system for PS, in which chlorine radicals (Cl·) generated from $FeCl_3$ under visible light initiate HAT to activate styrene units. High-molecular-weight PS (weight-averaged molar mass (M_w) > 90 kg·mol^{-1}) is efficiently depolymerized in aerobic environments, producing low-molecular-weight fragments (M_w < 1 kg·mol^{-1}) and achieving a benzoic acid yield of 23 mol percent (mol%). Crucially, the protocol exhibits strong tolerance toward commercial PS waste, including expanded foams and coffee cup lids, demonstrating robust material compatibility and process resilience.

Building on the versatile reactivity of chlorine radicals, Jiang et al. [8] recently developed a complementary upcycling strategy targeting polysulfones (PSFs), a class of engineering plastics known for their thermal and chemical stability. Through a visible-light-induced copper-catalyzed dual desulfonylative chlorination, PSFs were efficiently converted under mild conditions into value-added dichlorinated diaryl ethers, with representative products achieving yields of up to 85%. The key mechanistic feature of this transformation involves a ligand-to-metal charge transfer (LMCT) process that facilitates the visible-light-driven *in situ* generation of Cl· from copper(II) chloride ($CuCl_2$), which subsequently mediates the selective cleavage of aryl C(sp^2)–SO_2 bonds via homolytic *ipso*-aromatic substitution. Despite the high bond dissociation energy (~70–82 kcal·mol^{-1}, 1 kcal = 4184 J), this strategy achieves efficient conversion through the directional selectivity of chlorine radicals and catalyst design. The reaction system exhibits broad substrate tolerance, effectively processing glass-fiber-reinforced PSF composites and commercial-grade plastic articles without noticeable loss of efficiency. Notably, the gram-scale continuous-flow photoreactor employed to validate the synthetic scalability of the method maintained high product selectivity in the presence of mixed polyolefin and polyester contaminants.

At this stage, it is worth briefly contrasting the two most intensively studied mechanisms. While both photoinduced electron transfer and chlorine-radical-mediated oxidation involve radical intermediates, their mechanistic nuances differ significantly. Electron-transfer pathways typically initiate via the excitation of photoactive centers (e.g., uranyl and acridinium) followed by SET and subsequent radical propagation steps, favoring the depolymerization of oxygen-rich plastics under ambient conditions. In contrast, chlorine-radical-mediated mechanisms rely on the homolytic cleavage of C–H or C–C bonds via hydrogen abstraction, often facilitated by LMCT processes. While electron-transfer pathways offer greater substrate tunability and lower overoxidation, chlorine-radical pathways exhibit higher oxidative power and better performance on hydrocarbon polymers such as PS. However, photoinduced electron-transfer mechanisms are more amenable to integration with C–N or C–C coupling, offering further product diversification.

3. Photocatalytic upcycling with nitrogen incorporation

For oxygen-containing biodegradable plastics, the coupling of photodegradation with vacancy defect modulation has opened new avenues for generating value-added organic chemicals. Qiao et al. [9] developed a defect-rich nickel phosphorus trisulfide (d-$NiPS_3$)/cadmium sulfide (CdS) photocatalyst for the light-driven conversion of polylactic acid (PLA) and PET into hydrogen gas and organic acids under ambient conditions. Sulfur vacancy defects were introduced via argon plasma etching, significantly increasing charge separation and surface reactivity. The system achieved a

remarkable hydrogen (H_2) evolution rate of 40 mmol·g^{-1}·h^{-1}, generated 78 μmol of organic acids over 9 h, and maintained a stable performance over 100 h of continuous operation. Ultrafast spectroscopy revealed sub-5 ps electron transfer from CdS to d-$NiPS_3$, which effectively suppressed recombination and promoted hydrogen evolution at electron-rich active sites. Meanwhile, the hole-mediated oxidation of plastic chains limited carbon dioxide (CO_2) byproduct formation. This integrated approach demonstrates the potential for concurrent plastic depolymerization and hydrogen generation under light irradiation.

Complementing this nitrogen-insertion strategy, Chen et al. [10] recently reported a photocatalytic route to convert PLA into alanine under mild conditions using a cobalt phosphide (CoP)–CdS heterojunction catalyst. Visible-light irradiation induced spatially resolved charge carrier dynamics, in which photogenerated electrons were directed to CoP sites for hydrogen evolution via proton reduction, while holes accumulated on CdS drove oxidative cleavage of the PLA backbone to generate carbon-centered radicals. Upon the introduction of ammonium reagent, the catalytic C–N coupling reaction achieved high selectivity for alanine (75%) and afforded a product yield of 2.13 g from 3 g of PLA, corresponding to a yield of 56.6%. The CoP–CdS heterojunction retained 73% of its initial activity after 20 h, demonstrating excellent operational stability. Mechanistic studies revealed that the reaction relies on the synergistic effect of hole-induced polymer backbone cleavage and CoP-assisted proton reduction, enabling the one-pot nitrogen functionalization of plastic substrates.

In addition to photocatalytic systems, recent work has demonstrated the potential of photothermal catalytic strategies for PLA upcycling. For example, a photothermal platinum (Pt)-catalyst enabled the selective conversion of PLA into small-molecule acids under dual photothermal input, highlighting the synergistic role of heat in increasing depolymerization efficiency and overcoming kinetic barriers [11].

4. Photoreforming of polyolefins into fuels and chemicals

Polyolefins such as polyethylene (PE), polypropylene (PP), and polyvinyl chloride (PVC) are among the most recalcitrant plastics due to their saturated C–C backbones and hydrophobic nature. Recent advances in photocatalytic reforming have enabled the direct transformation of these polyolefins into liquid fuels and platform molecules via controlled radical-mediated C–C bond cleavage and reconstruction. Xie et al. [12] developed an atomically defined single-unit-cell-thick niobium(V) oxide (Nb_2O_5) catalyst that drives the complete photochemical mineralization and recycling of such polyolefin waste under atmospheric oxygen (O_2), aqueous media, and solar irradiation. This engineered material enables tandem oxidative depolymerization via hydroxyl-radical-mediated C–C bond cleavage followed by CO_2 photoreduction to C_2 oxygenates, achieving a production rate of 47.4 μg·g^{-1}·h^{-1}. Operando spectroscopic analyses and theoretical modeling identified O_2 and ·OH radicals-mediated oxidative processes, in which activated oxygen species regulate charge-carrier dynamics at interfacial Nb–O sites, while the C–C coupling of ·COOH intermediates governs the selectivity-determining C–C bond assembly step.

Separately, Zhan et al. [13] developed a supramolecular nickel/zinc (Ni/Zn) metalloporphyrin catalyst system that activates singlet oxygen generation under visible light, enabling the targeted cleavage of C–C and C–H bonds in PE and PVC through specific reactive oxygen species (ROS)-mediated pathways. This photocatalytic platform achieves deep restructuring of polymer chains into diesel-range α-olefins (C_{15}–C_{28}) with 85% selectivity and production rates of 76.1 μmol·h^{-1}, with gram-scale demonstrations having validated operational stability. A technoeconomic analysis projected an estimated revenue of 247 USD per metric ton of processed plastic waste, with life-cycle assessments indicating a >90% reduction in CO_2-equivalent emissions compared with conventional thermal cracking.

The mechanistic diversity of photocatalytic plastic upcycling extends beyond bond scission to include the selective cleavage of C–C, C–S, and C–O bonds, as well as oxidative transformations and heteroatom-functionalization reactions. These processes are enabled by photoinduced SET, HAT, OAT, and PCET, facilitating the conversion of diverse polymer substrates under mild conditions. Notably, representative strategies also include the oxidative depolymerization of inert polyolefins and nitrogen incorporation through C–N coupling with small-molecule reagents. The key mechanisms of these upcycling pathways are illustrated in Fig. 2.

These recent breakthroughs illustrate how photoreforming expands the mechanistic toolbox of photocatalytic plastic upcycling. Photocatalytic upcycling of plastics currently relies on four mechanistic archetypes—photoinduced electron transfer (SET/PCET), chlorine-radical-mediated oxidation, nitrogen-incorporation pathways, and photoreforming strategies—each with distinct reactivity and application niches. SET- and PCET-based approaches excel at depolymerizing oxygen-rich plastics such as PET, PES, and thermosets under mild conditions, leveraging tunable redox potentials and enabling integration with C–N or C–C coupling. Their moderate oxidative power helps preserve valuable intermediates but limits efficacy toward hydrocarbon polymers. In contrast, chlorine-radical-mediated pathways exhibit far higher oxidative strength, enabling the robust cleavage of inert C–H and C–C bonds in PS and PSFs, yet they often risk overoxidation and demand careful catalyst design to maintain selectivity. Nitrogen (N)-insertion strategies represent a newer paradigm, coupling photocatalytic backbone cleavage with heteroatom functionalization to generate value-added N-containing products such as alanine, but they remain catalyst-sensitive and substrate-limited. Finally, photoreforming platforms offer the broadest substrate scope, even encompassing recalcitrant polyolefins and PVC, but they typically produce less-defined product mixtures and require further optimization for selectivity and energy efficiency. Beyond these four mechanistic archetypes, hybrid strategies are emerging that combine multiple activation modes. For instance, a recent study reported the photothermocatalytic wet reforming of mixed plastic waste under simulated solar irradiation, achieving efficient C–C bond cleavage and gas-phase fuel generation in aqueous media. Such hybrid photothermal approaches illustrate a promising avenue for upgrading low-value plastic streams into syngas or hydrocarbons with high carbon-utilization efficiency [14].

Overall, SET-based systems provide high selectivity for oxygenated polymers, chlorine-radical-driven oxidation offers unmatched power for hydrocarbon plastics, N-insertion pathways unlock functionalization potential, and photoreforming grants the broadest substrate scope—albeit at the expense of product specificity. These trade-offs frame the choice of mechanism for different upcycling targets. To aid comparison and the practical selection of strategies, Table 1 summarizes representative photocatalytic systems in terms of plastic type, product yield, energy efficiency, and selectivity. This comparison highlights the diversity of mechanistic strategies and underscores trade-offs between catalyst stability, substrate compatibility, and product profiles.

5. Toward universal, scalable, and integrated photocatalytic systems

Despite significant progress, translating laboratory photocatalytic breakthroughs into industrially viable processes remains challenging; key obstacles include balancing broad-spectrum light absorption with charge separation, mitigating catalyst deactivation, and engineering tolerance to the complexity of real-world

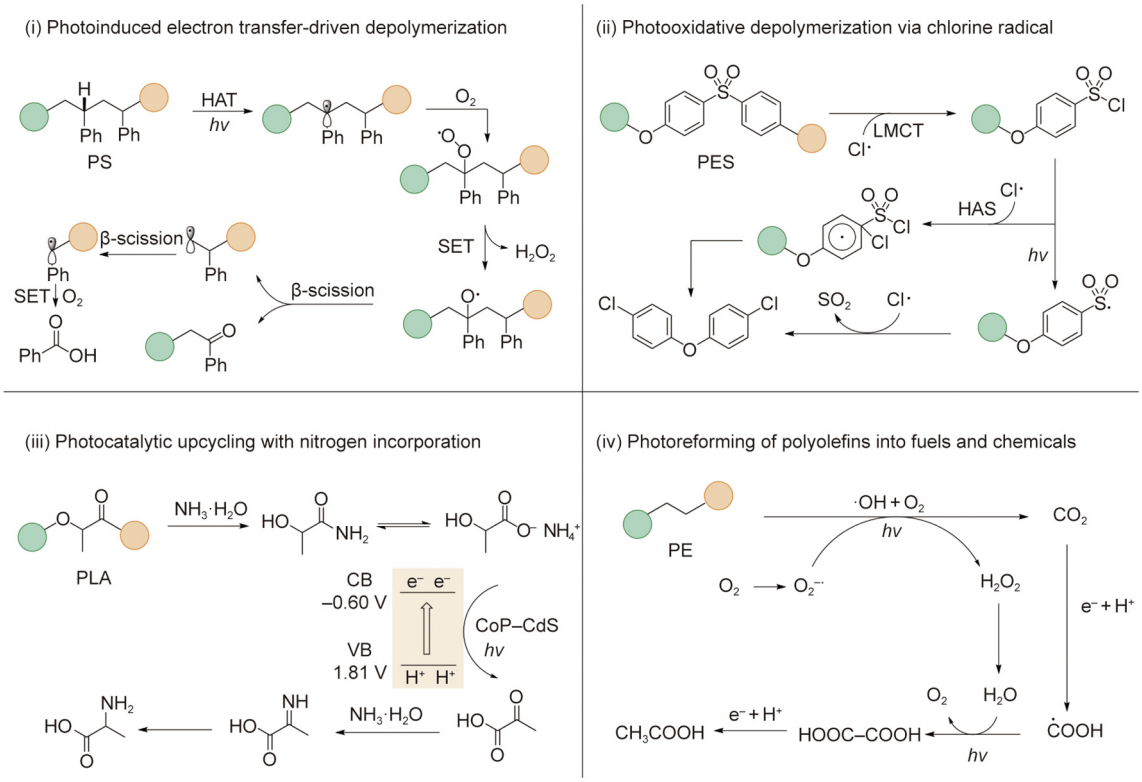

Fig. 2. Representative mechanisms in photocatalytic plastic upcycling. *h*: Planck's constant; *v*: the frequency of light; HAS: homolytic aromatic substitution; CB: conduction band; VB: valence band.

Table 1
Comparison of representative photocatalytic systems for plastic upcycling.

Mechanism	Catalyst system	Plastic type	Light source	Key products	Product yield	Reaction time	Energy input	Selectivity/note
SET/HAT/OAT (uranyl)	Uranyl cation	PET, PS, PES	Blue light (460 nm)	Monomers, chemicals	88%	48 h	Low	Broad substrate scope, continuous flow compatible
PCET (acridinium)	Acridinium photocatalyst/thiol radical system	Thermosets (epoxy)	Blue light (456 nm)	Bisphenol A monomers	~90%	48 h	Low	Selective C–C scission in thermosets
Cl· oxidation	$FeCl_3$	PS	White light	Benzoic products	23 mol%	20 h	Low	Works with foam and commercial plastics
LMCT-driven chlorination	$CuCl_2$	PES	Blue light (430 nm)	Dichlorinated diaryl ethers	85%	36 h	Low	Tolerant to additives and composites
Charge transfer/ oxidation	d-$NiPS_3$/CdS	PLA, PET	White light	H_2 + organic acids	H_2: ~40 mmol·g^{-1}·h^{-1}; organic acids: 78 μmol (in 9 h)	9 h	Low	Enhanced by S-vacancy defect engineering
Nitrogen insertion	CoP–CdS	PLA	Xe light	Alanine	56.6%	20 h	Low	C–N coupling for nitrogen-functionalized product
Photoreforming	Nb_2O_5, Ni/Zn porphyrin	PE, PP, PVC	Solar/ visible light	α-olefins (C_{15}–C_{28})	76.1 μmol·h^{-1}	72 h	Low	Converts to α-olefins or C_2 oxygenates

waste [15]. A central obstacle lies in reconciling the trade-off between broad-spectrum light absorption and efficient charge separation in photocatalyst design. Catalysts capable of harvesting visible and near-infrared light often exhibit rapid carrier recombination, which severely reduces photocatalytic efficiency. Additionally, the limited durability of catalysts under continuous operation, reliance on precious metals, and chemically inert nature of most polymers pose significant challenges. Common additives, stabilizers, and pigments in plastic waste can further poison active sites, reducing selectivity and efficiency. Additionally, mass transfer limitations at solid–solid and solid–liquid interfaces, along with energy-intensive scaling dynamics, restrict the throughput and practicality of current approaches (Fig. 3). Overcoming these interconnected challenges requires a dual-track strategy that combines rational catalyst design with systems-level process engineering.

Next-generation photocatalysts should feature precisely engineered defect sites, dopants, and heterojunctions to enhance charge separation, broaden light harvesting, and boost redox reactivity. In particular, defect engineering and nanoscale structuring are synergistic avenues toward the development of robust, highly selective

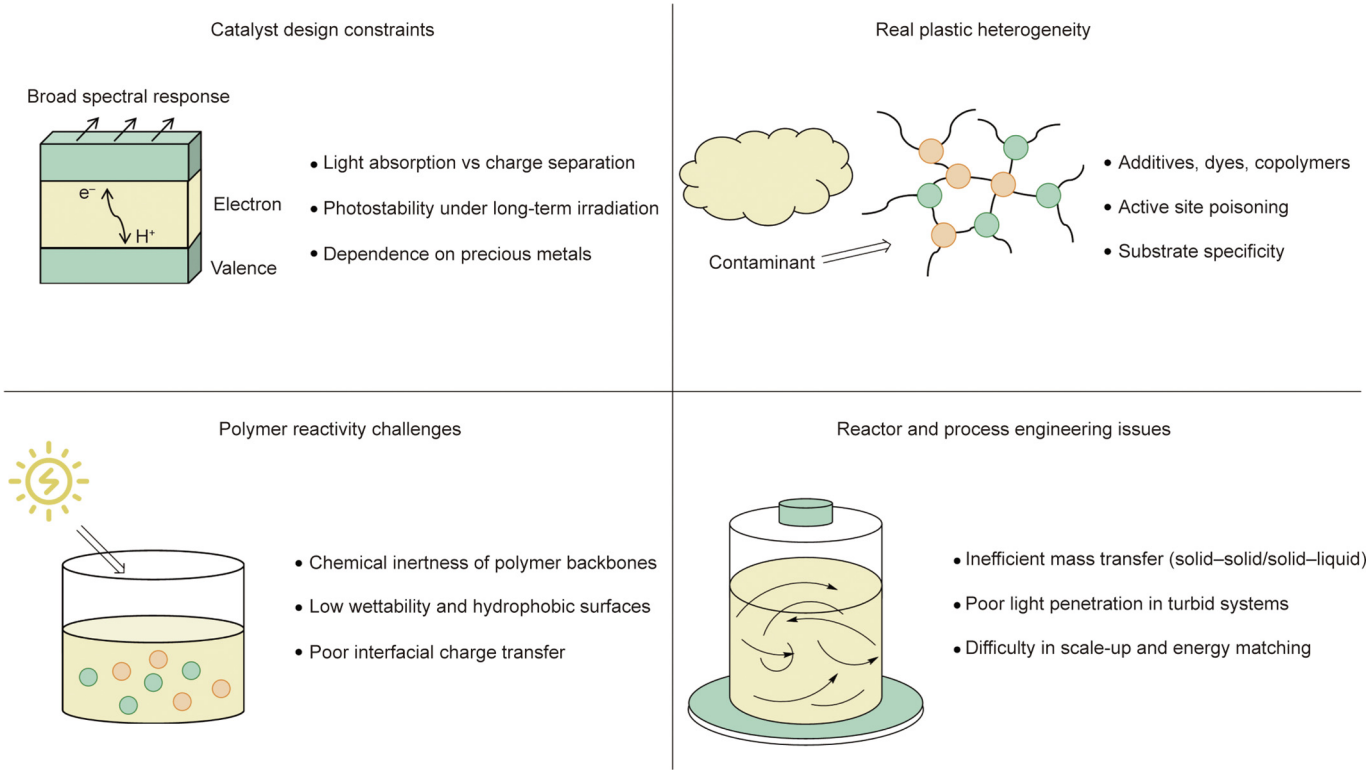

Fig. 3. Four major bottlenecks in photocatalytic plastic upcycling: catalyst design constraints, real plastic heterogeneity, polymer reactivity challenges, and reactor and process engineering issues.

catalysts suitable for complex waste streams. Moreover, integrating photocatalysis with electrocatalysis or thermocatalysis can generate synergistic effects that accelerate reaction kinetics and expand product versatility. The chemical and physical heterogeneity of real-world plastic waste further demands catalytic platforms that exhibit broad substrate scope and high selectivity. Future systems must tolerate mixed polymers, additives, and pigments, while enabling selective bond cleavage via adaptive surfaces, targeted oxidation/reduction pathways, or real-time reaction modulation. Achieving such generality and tunability will be essential for transitioning from idealized substrates to scalable, real-waste applications.

In conclusion, photocatalysis offers a promising and selective strategy for transforming plastic waste into valuable resources, potentially surpassing conventional thermal and mechanical recycling. Unlocking its full potential will require cross-disciplinary collaboration spanning materials chemistry, photophysics, process engineering, and economic analysis. Beyond chemical feedstock production, emerging directions include converting plastic waste into advanced functional materials, such as carbon nanomaterials, porous frameworks, or hybrid composites [16]. For instance, aromatic-rich depolymerization products from PS and PET have been used as precursors for carbon nanodots, while PLA chains modified via photocatalysis have been transformed into porous carbon materials suitable for supercapacitor electrodes [9,17,18]. Furthermore, upcycling routes incorporating dopants or heteroatoms into plastic fragments may yield functional polymers or catalysts with intrinsic redox or optical activity. These emerging applications suggest a future in which waste plastics not only serve as a carbon feedstock but also evolve into high-performance material precursors, unlocking additional circular economy value. These alternative pathways expand the scope of plastic upcycling toward applications in catalysis, energy storage, or structural materials, offering additional value streams and circularity strategies. If fully developed, photocatalytic technologies could redefine plastic waste not merely as a renewable carbon feedstock but also as a versatile raw material for next-generation materials, fundamentally reshaping its role in a sustainable future.

CRediT authorship contribution statement

Rui Huang: Writing – original draft. **Jiaolong Meng:** Writing – original draft. **Xuefeng Jiang:** Writing – review & editing, Validation, Supervision, Conceptualization.

Declaration of competing interest

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

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