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Review

Upcycling of Waste Plastics into Value-Added Chemicals

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Abstract: The rapid increase in plastic production has led to a severe plastic waste crisis, driving the development of various recycling technologies to mitigate this growing issue. However, these technologies often encounter substantial economic and environmental challenges in their implementation. An increasingly attractive alternative is chemical upcycling, which can transform waste plastics into value-added chemicals. This review systematically examines upcycling technologies applicable to major commercial plastics, including polyethylene terephthalate (PET), polyolefins, polystyrene (PS), and polyvinyl chloride (PVC). We focus on key strategies such as solvolysis, catalytic pyrolysis, hydrocracking and hydrogenolysis, along with some emerging approaches such as electrocatalysis and photooxidation, aiming to summarize emerging trends in the catalytic chemical upcycling of waste plastics.

Keywords: plastics; chemical upcycling; added value; chemicals

1. Introduction

Plastics are extensively utilized in packaging, construction, and electronics due to their low cost, durability and convenience. According to statistics, global plastic production reached 390.7 Mt in 2021, with projections indicating it will exceed 500 Mt by 2050 [1,2]. This surge in production has led to significant challenges regarding the disposal of post-consumer plastic products [3]. A report from the Organization for Economic Co-operation and Development (OECD) indicates that the global waste plastics reached 389 Mt in 2023. However, as illustrated in Figure 1, only 10% of waste plastics were recycled [4].

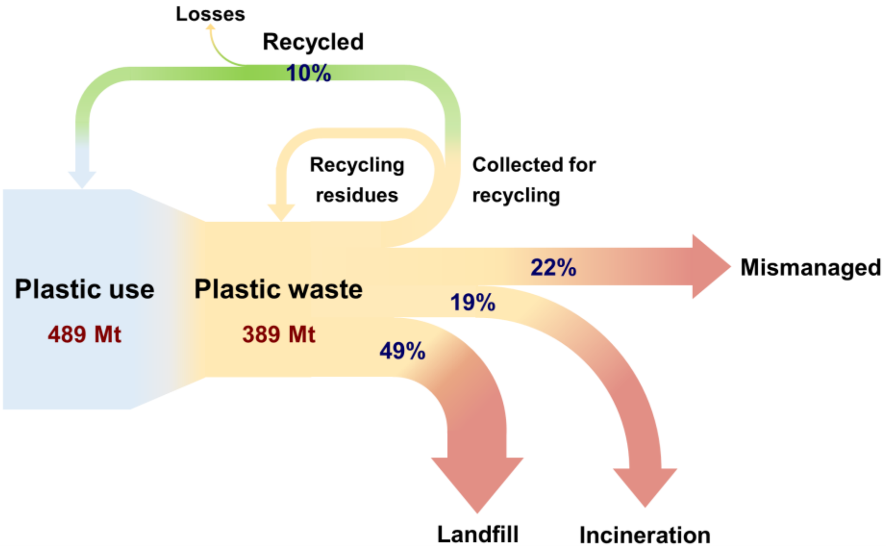


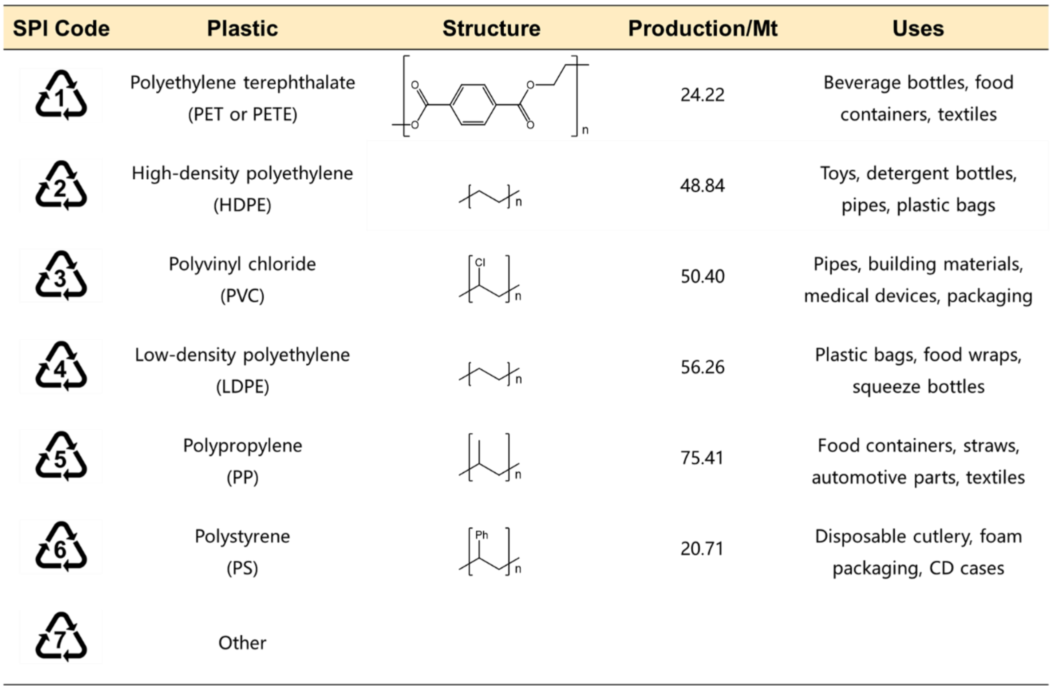
Figure 1. The plastics lifecycle in 2023 [a]. Note: [a] Data were based on OECD ENV-Linkages model from ref. [4].

Waste plastics have emerged as a significant global environmental challenge [5]. Approximately 70% of waste plastics are either landfilled or leaked into the environment, posing serious threats to ecosystems and human health [4,6]. The low degradability means they require extensive time to decompose, during which toxic chemicals are released and contaminate air, soil, and groundwater [7,8]. Some of the rest are incinerated, generating carbon dioxide and dioxins that contribute to global warming and air pollution [9–11]. Poorly managed waste plastics can also result in significant economic losses. Considering that the plastic industry consumes approximately 6% of the world’s annual oil production, effectively recycling plastic waste could save around 3.5 billion barrels of oil, resulting in an economic benefit of approximately $176 billion [12,13]. Consequently, there is an urgent need to develop efficient and economically viable methods for plastic recycling.

Against the backdrop of the circular economy and rapid advancements in plastic recycling technologies, many excellent reviews on plastic recycling have already been published. These existing reviews predominantly focus on chemical recycling, with systematic discussions mainly centered around recovery strategies or target products [14–24]. Categorized analyses of chemical upcycling strategies and their application potential for commercial plastics, particularly from the perspective of different plastic types, remain limited. This review, focusing on six major commercial plastics (PET, HDPE, PVC, LDPE, PP, and PS) and their respective upcycling strategies for producing value-added products, aims to systematically summarize the most recent advances in plastics upcycling. We begin by providing an overview of the current state of plastics and recycling, followed by an assessment of the feasibility of different recycling pathways. Next, we systematically introduce upcycling technologies for these types of plastics, exploring key strategies such as solvolysis, catalytic pyrolysis, hydrocracking, and hydrogenolysis, along with their latest advancements in the production of high-value-added products. Finally, we present an outlook on catalytic upcycling of plastics, emphasizing the future advancements in catalytic technologies and the critical consideration of chlorine resistance in practical plastic recycling processes.

2. An Overview of Plastics and Recycling

The Society of Plastics Industry assigns different types of commercial plastics their own codes, called SPI codes, for plastic type identification and recycling (Figure 2). Globally, polyethylene terephthalate (PET), polyolefins, polystyrene (PS), and polyvinyl chloride (PVC) collectively account for over 70% of total plastic production. However, among these plastics, only PET and polyethylene (PE, including HDPE and LDPE), have seen significant recycling efforts, while the recycling rates for the other plastic types are markedly low or nearly nonexistent [25,26].



SPI Code	Plastic	Structure	Production/Mt	Uses
1	Polyethylene terephthalate (PET or PETE)		24.22	Beverage bottles, food containers, textiles
2	High-density polyethylene (HDPE)		48.84	Toys, detergent bottles, pipes, plastic bags
3	Polyvinyl chloride (PVC)		50.40	Pipes, building materials, medical devices, packaging
4	Low-density polyethylene (LDPE)		56.26	Plastic bags, food wraps, squeeze bottles
5	Polypropylene (PP)		75.41	Food containers, straws, automotive parts, textiles
6	Polystyrene (PS)		20.71	Disposable cutlery, foam packaging, CD cases
7	Other			

Figure 2. The SPI Codes of plastics and their details [a]. Note: [a] Production data were taken from ref. [2].

The waste management hierarchy outlines a prioritized approach to waste reduction and management, serving as a foundational framework for addressing the plastic waste crisis (Figure 3) [27]. This policy emphasizes the importance of preventing or minimizing waste generation at the outset. One promising strategy is the adoption of bioplastics—both bio-based and biodegradable alternatives to conventional plastics, which often pose significant degradation challenges [28]. Despite their many advantages, bioplastics still have cost and performance limitations that perpetuate the recycling challenges inherent in traditional plastics [29,30].

Currently, there are four main methods of post-consumer plastics recycling: primary (closed-loop) recycling, secondary (mechanical) recycling, tertiary (chemical) recycling, and tertiary (energy) recycling [31]. Primary recycling involves the recovery and processing of a single type of uncontaminated plastic to produce a product of equal or similar quality to the original material, exemplified by PET bottle-to-bottle recycling [32]. In contrast, secondary recycling mechanically converts plastics into various products. However, this process often degrades the properties of the plastics and may result in the formation of microplastics [33,34]. Tertiary recycling encompasses the conversion of polymers into small molecular chemicals through chemical processes such as pyrolysis, gasification, and depolymerization. Quaternary recycling involves the recovery of energy through heat or electricity. However, as previously noted, this method has significant environmental impacts that must not be overlooked.

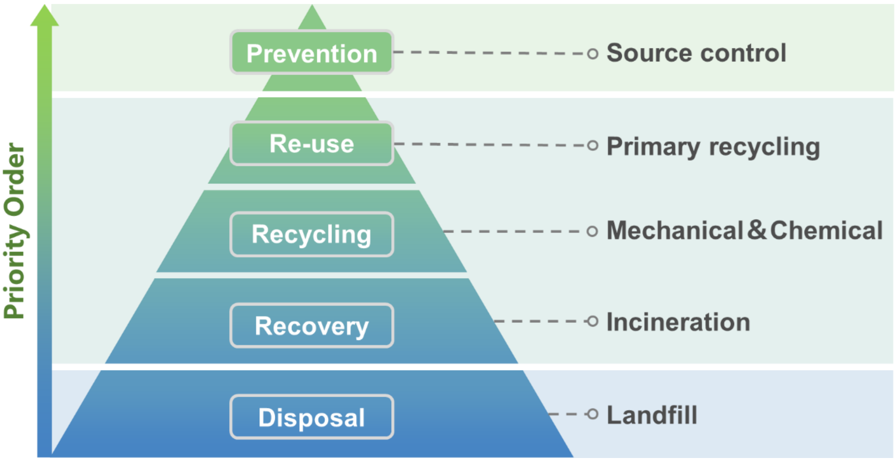


Figure 3. The waste management hierarchy and corresponding plastics recycling strategies.

The waste plastic recycling methods discussed above have played a crucial role in mitigating environmental pollution. However, aside from chemical recycling, most of these approaches are classified as downcycling, where the recovered products exhibit lower quality and economic value compared to the original plastic [35]. This inherent limitation has driven the emergence of chemical upcycling as a transformative concept. Unlike chemical recycling, which focuses on a closed-loop process converting waste plastics into intermediates and new plastics, chemical upcycling aims to transform waste plastics into value-added chemicals or functional materials. This innovative method offers promising solutions for more sustainable management and utilization of plastics. This review aims to summarize the recent advancements in the upcycling of major commercial plastics, with a particular focus on emerging technologies under mild conditions, such as the production of high-value chemicals and fuels through photochemical, electrochemical, and low-temperature catalytic pyrolysis. Other relevant topics, such as mechanical recycling, biodegradation, and the treatment of other types of thermoplastics and thermosetting plastics [36–38], are beyond the scope of this review and are not discussed in detail.

3. Polyethylene Terephthalate (PET, SPI Code 1#)

PET is widely utilized in beverage packaging, textiles, and various other applications due to its exceptional physical and chemical properties [39]. In the context of chemical recycling, PET is primarily depolymerized back into high value monomer chemicals, such as terephthalic acid (TPA), dimethyl terephthalate (DMT), bis(2-hydroxyethyl)terephthalate (BHET), and ethylene glycol (EG), through processes that reverse the synthesis reactions (Figure 4). This involves the use of nucleophilic reagents to attack the ester bonds within the PET polymer, leading to their cleavage and ultimately resulting in complete depolymerization into monomers. Although these solvolysis processes have been demonstrated to be economically viable and may offer certain cost advantages over conventional production routes, their feasibility remains constrained by the cost of feedstock PET and the complexity of product purification [40–43]. As a result, these processes are typically classified as closed-loop chemical recycling, rather than true upcycling in the strictest sense. However, recent advances have introduced a range of monomer-derived upcycling strategies, wherein the depolymerized intermediates are not only repurposed for PET re-synthesis but also explored for the production of value-added materials. These include further conversion into high-value fine chemicals [44–46], repolymerization with modified components to develop high-performance polyesters [47] and synthesis of metal-organic frameworks (MOFs) [48]. Although PET depolymerization itself does not constitute upcycling, it serves as a critical precursor step in many value-added transformation pathways. In this section, we will first examine the depolymerization process, focusing on key factors such as maximizing monomer yield, enhancing reaction kinetics, and achieving mild reaction conditions. Additionally, we will explore cascade reaction strategies based on depolymerized monomers, demonstrating how these intermediates can be further upcycled into high-value fine chemicals. Beyond PET closed-loop recycling and monomer-based cascade upcycling, we will also discuss alternative innovative approaches that directly convert low-value PET waste into high-value products, providing a comprehensive overview of PET waste upcycling potential.

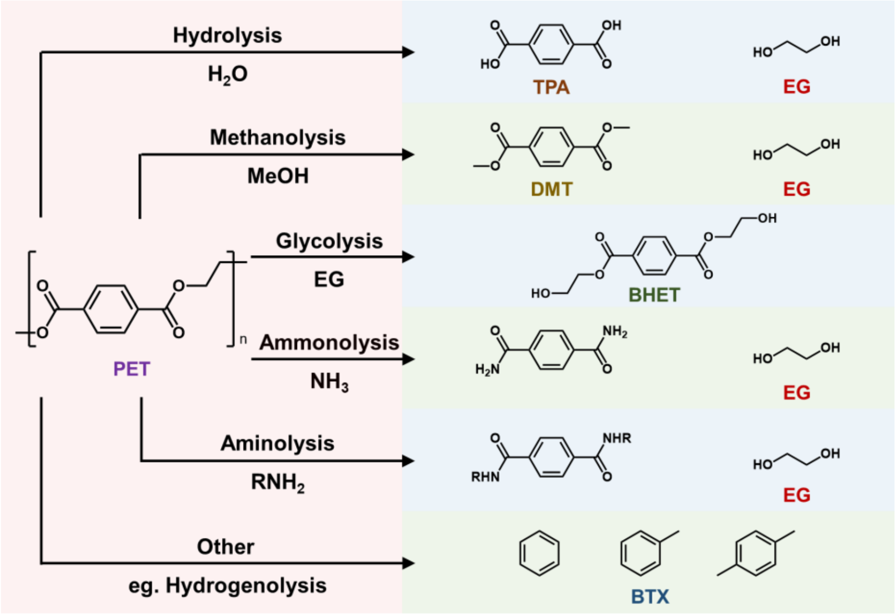


Figure 4. PET chemical recycling and upcycling strategies.

3.1. Chemical Recycling to Monomers and Tandem Upcycling Strategies

3.1.1. Hydrolysis

The hydrolysis of PET typically occurs under high temperatures and pressures, during which water molecules attack and break the ester bonds in the PET polymer chain. This reaction generates carboxyl and hydroxyl functional groups, ultimately leading to the depolymerization of PET into TPA and EG. Due to the relatively weak nucleophilicity of water, catalysts such as acids and bases are often employed to enhance the reaction rate [14].

Hydrolysis in a neutral environment is typically conducted without catalysts at high temperatures (145–400 °C) and high pressures (1–35 MPa) [49,50]. Pereira et al. studied the neutral hydrolysis of PET in different phases under a rapid heating process of approximately 5–10 °C/s (Figure 5a), and the yield of TPA could exceed 90% in 75–95 s [50,51]. Metal salts have been explored as catalysts for neutral hydrolysis of PET, such as $Zn(Ac)_2$, $ZnCl_2$ [52,53]. The mechanism of the catalysis is thought to involve electrolytic destabilization of the PET-water interface, resulting in a greater inter-facial area exposed to the hydrolysis reaction (Figure 5b) [54]. Additionally, pseudohomogeneous ZnO nanoparticles have been identified as highly effective catalysts for enhancing the neutral hydrolysis of PET. This process achieves complete PET depolymerization at 200 °C within 60 min, yielding 95.6% TPA[55].

Acid hydrolysis typically utilizes inorganic acids, with sulfuric acid being the most commonly used. This process does not necessitate high temperatures or pressures; however, low temperatures and acid concentrations can slow hydrolysis [56]. For instance, Islam et al. achieved nearly 100% PET depolymerization using an 80% *v*/*v* sulfuric acid solution at 100 °C for 30 min, resulting in TPA purity exceeding 95% (Figure 5c) [57]. When nitric acid is used, the EG produced by depolymerization is simultaneously oxidized to oxalic acid, creating a valuable recycling pathway for PET [58]. Recent research has sought more environmentally friendly and efficient alternatives to traditional inorganic acids. For example, while TPA can catalyze PET hydrolysis, it requires higher temperatures and longer reaction times due to its weak acidity [59]. Additionally, the use of p-toluenesulfonic acid and adipic acid has been explored [60,61].

Alkaline hydrolysis generally utilizes strong bases like NaOH and KOH, resulting in EG and alkali metal salts of TPA, which require additional acidification. The introduction of phase transfer agents (PTAs) improves the interaction between the alkaline catalyst and the PET surface within the reaction medium, enabling the alkaline hydrolysis of PET under milder conditions, alkaline hydrolysis of PET under milder conditions, typically at 70–95 °C and atmospheric pressure [62]. Commonly used PTAs include tetrabutylammonium iodide (TBAI) and tributylhexadecylphosphonium bromide (TBHDPB) (Figure 5d) [63,64].

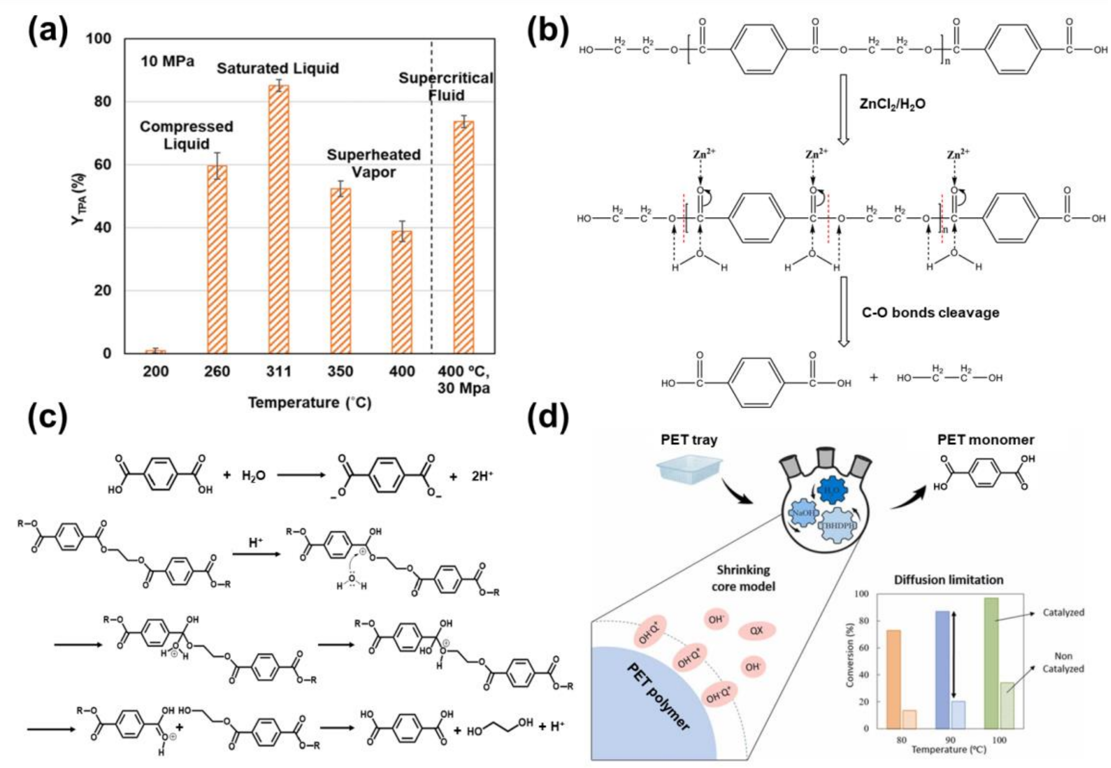


Figure 5. Different types of PET hydrolysis. (**a**) TPA yield neutral hydrolysis of PET chips in different phases. Reproduced from ref. [50]. Copyright 2023 American Chemical Society. (**b**) Possible neutral hydrolysis mechanism of PET using $ZnCl_2$ as a catalyst. Reproduced from ref. [53]. Copyright 2019 Elsevier. (**c**) Acid hydrolysis mechanism of PET using TPA as an acid catalyst. Reproduced from ref. [59]. Copyright 2021 Elsevier. (**d**) Alkaline hydrolysis of PET assisted by TBHDPB. Reproduced from ref. [64]. Copyright 2023 Elsevier.

The hydrolysis of PET yields monomers (TPA and EG), both of which have been extensively explored for value-added applications. For instance, TPA serves as a crucial organic linker in the synthesis of MOFs, a rapidly expanding field in chemical recycling. Zhang et al. successfully synthesized lanthanide-based TPA-derived MOFs from waste PET bottles [65]. Initially, PET was subjected to alkaline hydrolysis to obtain TPA, followed by a solvothermal reaction between the extracted TPA and $Tb(NO_3)_3 \cdot 6H_2O$, yielding a flower-like lanthanide-based MOF. However, this two-step approach is hindered by complex processing steps and high energy consumption, which limit its scalability. To streamline the process and enhance efficiency, researchers have explored a one-pot strategy, enabling direct PET conversion into TPA while simultaneously synthesizing MOFs [66]. By employing a one-pot strategy in which MOF metal ions are directly introduced into the PET neutral hydrolysis slurry, the high-yield direct synthesis of value-added MOFs can be achieved. This approach not only eliminates additional separation steps but also reduces the consumption of strong acids and bases during the depolymerization and purification processes.

Beyond TPA valorization, the upcycling of EG has also garnered significant attention, enabling its efficient conversion into high-value products such as glycolic acid (GA) and hydrogen [67,68]. A one-step tandem catalytic strategy has been developed to achieve thermocatalytic oxidation of PET, yielding TPA and high-value GA [46]. Using a Au/NiO catalyst with abundant oxygen vacancies, this system not only facilitates accelerated PET hydrolysis but also achieves remarkable yields of 99% TPA and 87.6% GA, demonstrating superior catalytic efficiency and resource valorization potential. Electrocatalytic strategies have also been employed to upcycle EG, a PET depolymerization product, into valuable commodity chemicals and H_2 fuel [69]. Zhou et al. first hydrolyzed PET in an alkaline solution to produce TPA and EG, which were then selectively cleaved via C-C bond cleavage to generate potassium dicarboxylate (KDF) over an anodic $CoNi_{0.25}P$ electrocatalyst, with a cathode facilitating hydrogen generation [70].

3.1.2. Methanolysis

Alcoholysis of PET primarily employs methanol, referred to as methanolysis, in which methanol engages in an ester exchange reaction. This process breaks the ester bonds in the polymer chain, yielding DMT and EG. Ethanol, butanol, and iso-octanol are also used to generate different monomers [71–73].

To improve the efficiency of PET methanolysis, various methods have been employed, including supercritical conditions, microwave assistance, and the use of cosolvents [74,75]. Additionally, the development of effective catalysts is also essential for facilitating rapid methanolysis, with common choices including metal acetates, oxides, and hydroxides. For example, zinc acetate can catalyze PET methanolysis to achieve 95% DMT yield within 20 min at 160 °C, and the resulting EG can be further converted into various diesters and diols through hydrogenation [76]. Considering the reversibility of the methanolysis reaction, Tanaka et al. captured EG by adding dimethyl carbonate (DMC), resulting in the formation of ethylene carbonate (EC) and methanol. This approach shifted the equilibrium of the methanol depolymerization reaction towards increased DMT production [77].

To efficiently utilize waste resources and further enhance the value of PET methanolysis products, researchers have developed various tandem catalytic strategies for the upcycling of PET methanolysis-derived intermediates. One key pathway involves the selective hydrogenation of DMT to form dimethyl 1,4-cyclohexanedicarboxylate (DMCD), which can subsequently undergo catalytic hydrogenation to yield 1,4-cyclohexanedimethanol (CHDM) [78–80]. Compared to the former, CHDM is generally more attractive, as it serves as a key precursor for high-performance functional polyesters and exhibits a significantly enhanced value along with a rapidly expanding market demand [45]. Huang et al. have reported an innovative and compact two-step catalytic process for PET upcycling [81]. In the first step, a bifunctional Ru/MnO_2 catalyst is employed to integrate PET methanolysis and the hydrogenative cyclization of the DMT intermediate into a one-pot tandem reaction system, enabling the high-yield synthesis of DMCD (84% yield). Additionally, the liquid-phase product from the tandem conversion can be directly subjected to hydrogenation over a CuZnZr mixed oxide catalyst to produce CHDM, eliminating the need for intermediate separation and achieving a final CHDM yield of 78%.

Beyond CHDM synthesis, DMT can also undergo further hydrogenolysis to produce p-xylene (PX). Li et al. have developed a one-pot catalytic system utilizing a CuFeCr catalyst to facilitate the synergistic coupling of CO_2 hydrogenation, PET methanolysis and DMT hydrogenation [82]. This integrated process not only yields DMT (3.7%) and DMCD (28.6%) but also achieves a high-yield conversion of PX (49.3%). Further improvements in PX yield have been demonstrated via catalytic transfer hydrogenolysis of DMT over $Cu/ZnZrO_x$ catalyst, achieving ~63% PX yield at 240 °C over 16 h [83].

3.1.3. Glycolysis

The glycolysis process employs glycols such as ethylene glycol (EG), diethylene glycol (DEG), and propylene glycol as degradation agents for transesterification reactions with PET. Among these, EG is the most commonly reported glycol, with BHET being the primary product of the reaction, which can be further utilized to synthesize polyesters or to create high-performance materials such as unsaturated polyesters and polyurethanes. Numerous studies have investigated the effects of reaction parameters on conversion and selectivity during PET glycolysis, focusing on the screening of suitable catalysts [84,85].

Catalysts used in PET glycolysis can be either homogeneous or heterogeneous, including metal salts, metal oxides, organic catalysts, and ionic liquids (ILs). Homogeneous catalysts typically exhibit high catalytic performance. Metal acetates were among the first homogeneous catalysts employed in PET glycolysis. For example, $Zn(OAc)_2$ has been shown to achieve high conversion rates and BHET yields in this process [86]. Due to the environmental concerns associated with heavy metal salts, researchers have increasingly turned to eco-friendly catalysts, such as sodium salts, ionic liquids, and organocatalysts [86–88]. Javed et al. reported sodium ethoxide as an environmentally friendly and cost-effective catalyst for the glycolysis of post-consumer PET waste, achieving a conversion rate of 98% and a BHET separation yield of 76% [89]. Ionic liquids are considered ideal green solvents for the glycolysis of PET due to their excellent environmental profile, and their positive impact on enhancing reaction activity has been widely confirmed (Figure 6a) [90–92].

Recent studies have predominantly focused on heterogeneous catalysts for the glycolysis of PET, primarily employing metal oxides. These catalysts include alkaline options, such as those based on Mg and Ca, as well as Lewis acid catalysts, which encompass Mn, Fe, Co, and Zn [93–95]. Heterogeneous catalysts present several advantages over homogeneous catalysts, notably their ease of separation from reaction products and their minimal impact on the purity of BHET. However, they also exhibit limitations in terms of catalytic activity and selectivity. To overcome these challenges, researchers are increasingly utilizing nanoscale catalysts and those with optimized pore structures. These methods can significantly enhance catalytic performance by increasing the surface area and the availability of active sites (Figure 6b) [96,97]. Veregue et al. demonstrated that ultra-small Co nanoparticles (~3 nm), could achieve a 77% yield of BHET within 3 h at a temperature of 180 ℃ [98]. Additionally, Lechuga-Islas et al. utilized thermo-responsive (co)polymer catalysts ([PIL]Zn_mCl_n) that enable a transition from a homogeneous to a heterogeneous state. This innovative approach enables controlled catalyst recovery and reusability as well as product purification [99].

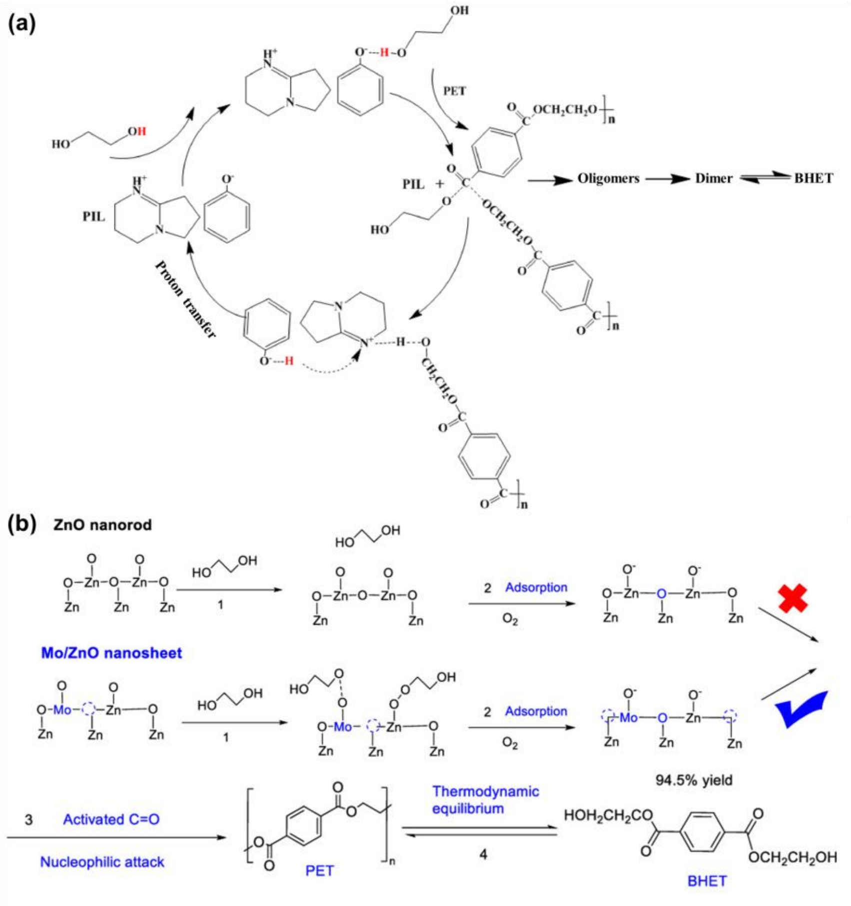


Figure 6. Homogeneous and heterogeneous catalytic mechanisms of PET glycolysis. (**a**) Reaction pathway of PET glycolysis catalyzed by protic ionic liquid. Reproduced from ref. [90]. Copyright 2022 Elsevier. (**b**) Mechanisms of PET glycolysis catalyzed by heterogeneous ZnO nanorods and Mo/ZnO nanosheets. Reproduced from ref. [96]. Copyright 2022 American Chemical Society.

3.2. Other Upcycling Strategies for Fine Chemicals

In addition to repolymerizing PET-derived monomers for PET regeneration or further high-value applications, alternative upcycling strategies have been explored to convert PET into various high-value fine chemicals. One promising pathway involves aminolysis and ammonolysis, as nitrogen-containing nucleophiles such as amines and ammonia can effectively degrade PET, yielding corresponding amination products. Compared to alcohols, amines exhibit higher nucleophilicity, enabling faster reaction rates under milder conditions in both catalytic and non-catalytic systems [100,101]. Ethanolamine (EA) is commonly employed, yielding bis(2-hydroxy ethylene)terephthalamide (BHETA) [102]. To enhance the efficiency of this process, Demarteau et al. used thermally stable acid-base mixture catalysts, achieving rapid and complete PET depolymerization within 10 min at 140-180 ℃ in the presence of EA, with a high yield of BHETA [103]. BHETA can be readily modified through further chemical transformations, such as ester bond formation via its amide and -OH functional groups when reacted with various dicarboxylic acids. This approach has been employed to design and synthesize novel polyester amides with enhanced bone regeneration properties and controlled drug release capabilities [104]. Aminolysis and ammonolysis of PET have not yet been widely adopted as commercial recycling technologies within the realm of chemical recycling. However, despite the inability of their reaction products to be repolymerized into PET, these intermediates can undergo further chemical transformations for value-added applications, including the synthesis of adhesives, hydrogel adsorbents, and fluorescent materials, demonstrating promising potential for upcycling [105–107].

In contrast to depolymerization processes based on hydrolysis or ester exchange, which primarily yield monomers suitable for purification and reuse in PET manufacturing, catalytic hydrogenolysis provides a valuable route to convert PET into valuable aromatic compounds. The carbon-supported single-site Mo-dioxo complex catalyst efficiently and selectively catalyzes the solvent-free depolymerization of PET, yielding terephthalic acid and ethylene [108]. Mechanistic studies revealed that the reaction proceeds through an initial retro-hydroalkoxylation/β-C-O scission, followed by the hydrogenolysis of the vinyl benzoate intermediate. Further research has concentrated on upcycling PET into aromatic hydrocarbons, such as benzene, toluene, and xylene (BTX) [109,110]. In a prior study, our group identified a hydrogenolysis reaction pathway influenced by the ruthenium coordination environment, suitable for converting plastics containing aromatic monomers [111]. Moreover, using renewable low-carbon alcohols as hydrogen donors for PET hydrodeoxygenation to BTX circumvents the need for external hydrogen sources. Gao et al. demonstrated this approach by employing low-cost $CuNa/SiO_2$ to integrate methanol dehydrogenation, PET methanolysis, and subsequent DMT hydrodeoxygenation to PX, facilitating in situ hydrogen production [112].

4. Polyolefin (HPDE, SPI Code 2; LDPE, SPI Code 4; PP, SPI Code 5)

Polyolefins, including polyethylene (LLDPE, LDPE, HDPE) and polypropylene (PP), represent the most widely demanded plastics, accounting for nearly 50% of global plastic production [2]. These materials are predominantly utilized in the manufacturing of nondurable products, such as short-lived containers and packaging, which leads to their prevalence in municipal plastic waste streams [113]. Their molecular structure, consisting solely of C-C and C-H bonds, makes them chemically inert. Therefore, traditional chemical recycling strategies for polyolefins (such as pyrolysis and gasification) are typically carried out under high temperatures [114,115], leading to complex product distributions upon degradation. As such, these approaches are not discussed in depth in this review. To tackle these challenges, researchers are actively exploring more efficient catalysts and advanced strategies to upcycle polyolefins into high-value products, light olefins, aromatics, and fuels, under mild conditions (Figure 7).

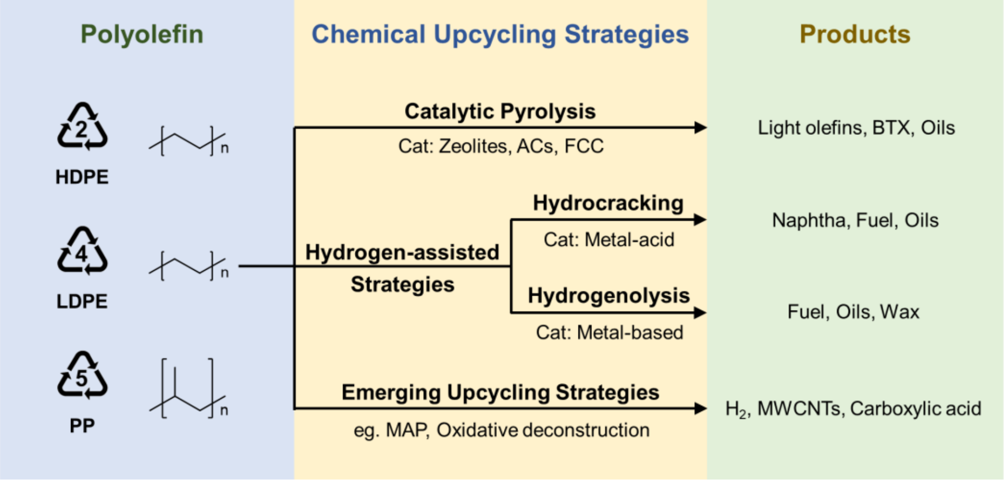


Figure 7. Chemical upcycling strategies of polyolefins and corresponding value-added products.

4.1. Catalytic Pyrolysis

Pyrolysis is a thermochemical process in which polyolefins are heated in an oxygen-free environment, breaking C-C bonds to produce smaller molecular hydrocarbons. Traditional pyrolysis methods are typically conducted in non-catalytic settings at temperatures exceeding 500 °C. In these conditions, random chain breaking occurs mainly via a free radical mechanism, leading to low selectivity, high energy consumption, and the formation of coke deposits [116,117].

In contrast, catalytic pyrolysis of polyolefins primarily relies on zeolites and other catalysts with acidic sites and porous structures [118,119]. The use of catalysts enhances both the selectivity and controllability of product distribution while significantly reducing the overall energy consumption. This catalytic reaction primarily follows the carbenium ion mechanism, wherein the polyolefin is protonated at a Brønsted acid site to form a carbenium intermediate. This intermediate undergoes various backbone rearrangements and β-scissions, ultimately yielding lower molecular weight products [120]. Due to various secondary reactions, such as isomerization, aromatization, cyclization, dehydrogenation, and oligomerization (Figure 8a), this process can generate a diverse array of valuable chemical, including olefins, aromatics, and paraffins [121,122]. Furthermore, in-situ and ex-situ processes affect the reaction pathways and product distributions (Figure 8b). In-situ catalytic pyrolysis facilitates the aromatization of olefins within the catalyst's pores, with the released hydrogen saturating the olefins. Conversely, ex-situ catalytic pyrolysis predominantly involves catalytic cracking, resulting in notably high olefin yields [123].

Different types of zeolite catalysts are extensively used in polyolefin pyrolysis, including HZSM-5, HUSY, Hβ and HMOR, where parameters such as acidity, pore structure, and morphology play a crucial role in dictating catalytic activity and product selectivity [120]. As mentioned above, catalytic pyrolysis primarily occurs at Brønsted acid sites, and therefore, factors such as the acidity and its distribution have a profound impact on catalytic performance [124]. Further research focuses on the influence of pore structure and zeolite morphology on catalyst activity and selectivity, as they determine the accessibility of acid sites and the diffusion rate of reactants [125,126]. To further enhance the activity of catalytic pyrolysis and improve target product selectivity, researchers have employed metal/zeolite bifunctional catalysts. The introduction of metals alters the acidity of the original zeolite catalysts [127], with different metals influencing product selectivity: Fe and Cu have been shown to increase yields of cycloalkanes and olefins [128]; active metals such as Pt, Zn, and Ga enhance the selectivity for aromatics [129–131]. Our previous work demonstrated that incorporating Pt increased the aromatization rate of light alkenes generated from cracking at the acid sites of ZSM-5, leading to a significant increase in BTX yields (Figure 8c,d) [131]. However, during the catalytic pyrolysis of plastics, the excessive Brønsted acid sites in zeolite-based catalysts often promote coke formation. Coke precursors, such as olefins and aromatics, initially adsorb onto the acid sites of the catalyst and undergo further transformation, eventually leading to coke deposition and a reduction in catalytic stability [132]. Therefore, developing coke-resistant catalysts, for instance, by enhancing intermediate diffusion to mitigate coke accumulation and improve catalyst stability, has become a critical research focus in the field [125].

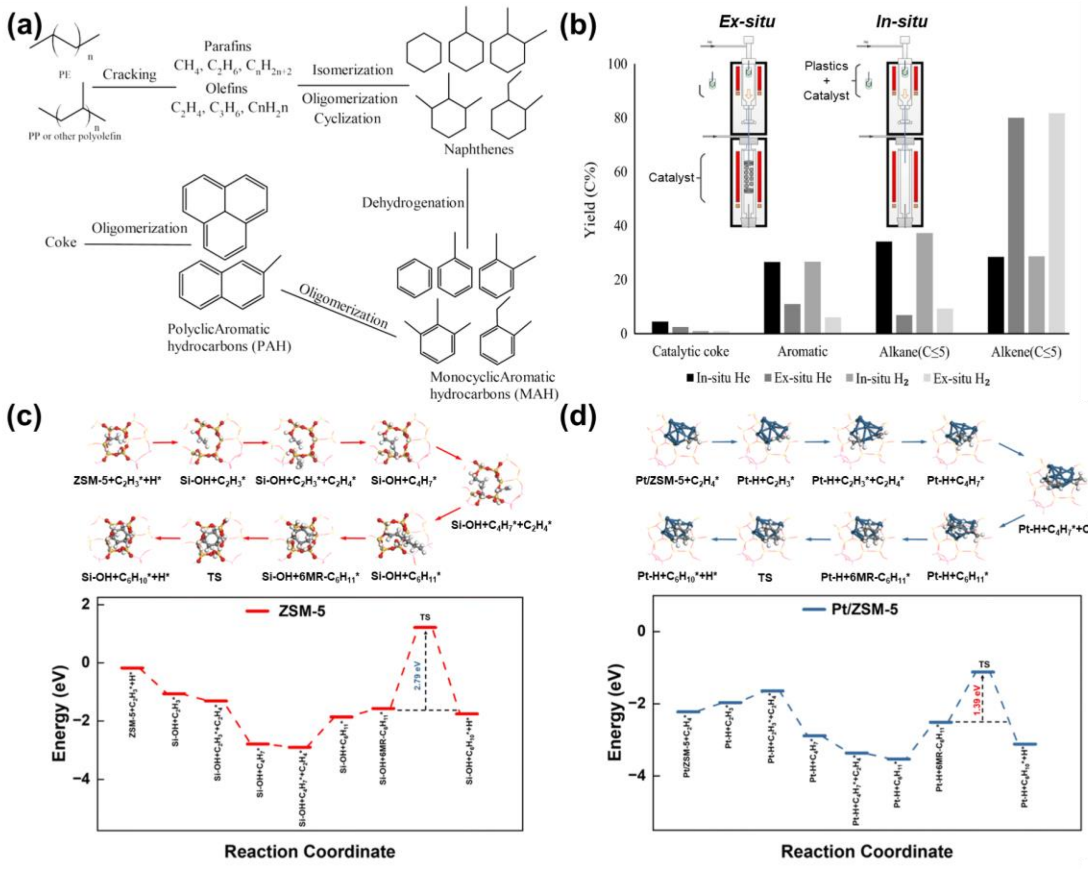


Figure 8. Catalytic pyrolysis of polyolefins. (**a**) Reactions involved in the catalytic pyrolysis process of polyolefins. Reproduced from ref. [121]. Copyright 2024 American Chemical Society. (**b**) Product distribution during catalytic pyrolysis of PE under in situ and ex situ conditions. Reproduced from ref. [123]. Copyright 2017 Elsevier. (**c**,**d**)

Reaction energy diagram of ethylene to the benzene precursor over ZSM-5 and Pt/ZSM-5. Reproduced from ref. [131]. Copyright 2023 Royal Society of Chemistry.

4.2. Hydrocracking and Hydrogenolysis

The catalytic degradation of polyolefins in a hydrogen atmosphere has attracted considerable research attention as an emerging strategy that operates under milder reaction conditions compared to conventional methods like pyrolysis and gasification, which are constrained by high temperature and coke formation. This process utilizes heterogeneous catalysts in a hydrogen atmosphere, resulting in saturated hydrocarbons with varying molecular weight distributions. The products include various fuels, such as gasoline, diesel, and jet fuel, as well as naphtha, waxes, and lubricants [133,134]. Depending on the catalyst type used, this hydrogen-assisted degradation can be further classified into hydrocracking and hydrogenolysis.

Hydrocracking typically involves bifunctional metal-acid catalysts. Long-chain polyolefin molecules are dehydrogenated at the metal sites to form olefins. These olefins then desorb and diffuse to the Brønsted acid sites, where they are converted into carbenium ion intermediates. Subsequently, these intermediates undergo isomerization and β-scissions before diffusing back to the metal sites, where they are hydrogenated to yield alkanes (Figure 9a) [135]. Catalysts featuring metals with (de)hydrogenation activity, such as Pt, Ni, and Ru, along with various acidic carriers, are commonly utilized [136–138]. Notably, Pt-based catalysts have demonstrated exceptional performance in the hydrocracking of polyolefins. For instance, Pt/WO_3/ZrO_2 and HY zeolite composite catalysts have demonstrated high efficiency in converting polyolefins into branched liquid fuels, including diesel, jet fuel, and gasoline-range hydrocarbons, achieving yields of up to 85% [136]. The efficiency and selectivity of this process are strongly influenced by several key factors, including the metal-acid balance, the spatial proximity of active sites, and their accessibility, which collectively determine the rate-limiting steps and product distribution. To further optimize catalyst performance, Li et al. designed β-zeolite and silicalite-1 encapsulated Pt nanoparticles (Pt@S-1) composite catalysts, which effectively hydrocrack LDPE into naphtha, achieving yields of up to 89.5% at 250 °C (Figure 9b) [139]. In this system, hydrogenation over confined metal sites is well-matched with the cracking steps, as it selectively facilitates the transport of appropriately sized olefins, while the enhanced diffusion kinetics promote the formation of narrowly distributed alkanes. Overall, the rational design of high-performance hydrocracking catalysts, leveraging metal-acid synergy and structural shape selectivity, is crucial for achieving high yields of value-added products.

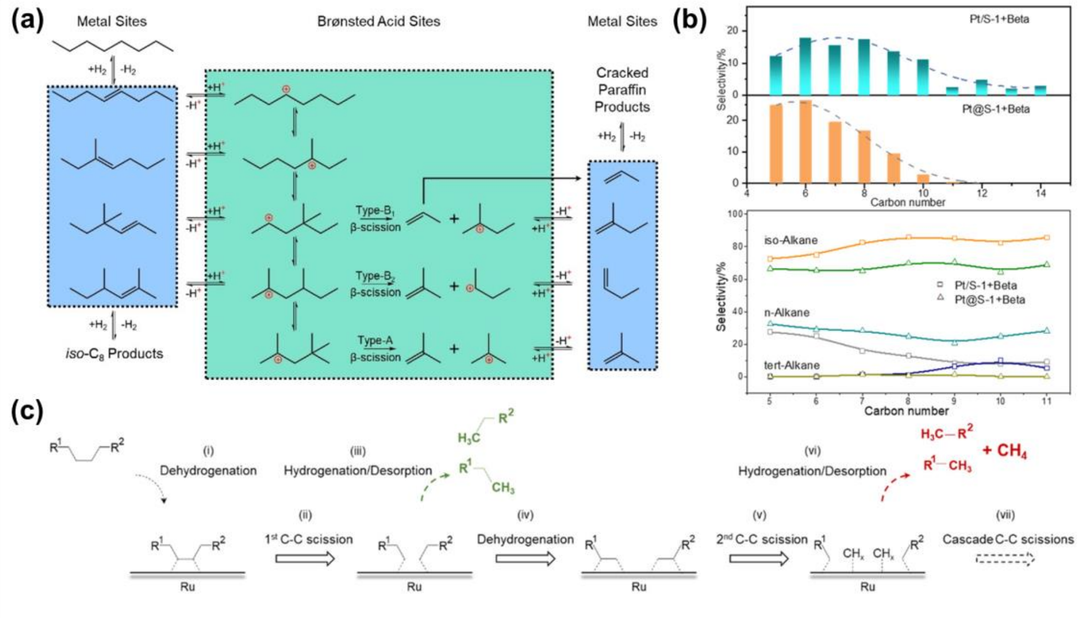


Figure 9. The hydrogen-assisted degradation strategies of polyolefins. (**a**) Mechanism of polyolefins hydrocracking using octane as an example. Reproduced from ref. [140]. Copyright 2021 Elsevier. (**b**) Distribution of hydrocracking products over Pt@S-1 and Pt/S-1 catalysts. Reproduced from ref. [139]. Copyright 2023 American Chemical Society. (**c**) Proposed reaction mechanism for the hydrogenolysis on Ru-based catalysts. Reproduced from ref. [141]. Copyright 2021 American Chemical Society.

Unlike hydrocracking, the hydrogenolysis process typically employs monofunctional transition metal catalysts, with Ru-based catalysts being well-studied for their excellent activity and selectivity [141]. In this process, C-C cleavage of long-chain polyolefin molecules occurs following the activation of C-H bonds and dehydrogenation at the metal sites, leading to the formation of shorter hydrocarbon chains after further hydrogenation and desorption (Figure 9c). This cycle is repeated, resulting in multiple C-C bond cleavages and ultimately yielding small molecule hydrocarbon products [142]. Nakaji et al. utilized Ru/CeO_2 under mild reaction conditions (473 K and 2 MPa H_2 pressure), achieving liquid fuel (C_5–C_{21}) yields of 77% and wax (C_{22}**)**C_{45}) yields of 15% [143]. However, the cleavage of terminal C-C bonds often leads to the excessive production of low-value methane. While increasing hydrogen pressure can mitigate methane formation, it may also reduce the reaction rate due to competitive adsorption between hydrogen and polyolefin [144–146]. Research has shown that the size and structure of the catalyst are crucial factors influencing the activity and product selectivity in polyolefin hydrogenolysis. Ji et al. prepared Ru nanoclusters by finely regulating Ru sizes and achieved a balance between the metal-support interactions and the hydrogen spillover effect [147]. Additionally, strategies such as incorporating transition metals as hydrogen reservoirs [148] or confining catalytic sites within the pores [149,150] can further inhibit excessive hydrogenolysis and enhance selectivity towards liquid products.

4.3. Advanced Upcycling Strategies

Recent studies have explored innovative processes and emerging technologies to produce value-added products from polyolefins, including cross alkane metathesis, tandem dehydrogenation/olefin metathesis, microwave-assisted pyrolysis, and oxidative upcycling.

Cross alkane metathesis (CAM) involves the exchange or rearrangement of alkyl fragments between two alkane molecules and was initially applied to small molecule alkanes [151]. Jia et al. utilized this strategy in polyolefin degradation, employing low-value light alkanes (such as n-hexane) to completely convert various polyolefins with differing molecular weights into useful liquid fuels and waxes [152]. In their process, a molecular pincer-type iridium complex dehydrogenates PE and the light alkane, followed by $Re_2O_7/\gamma\text{-}Al_2O_3$-catalyzed cross metathesis to yield two new olefins. These olefins are then hydrogenated over the iridium catalyst to produce saturated alkanes, completing the catalytic cycle. The researchers further optimized the cost and stability of the catalysts in this tandem system [153,154]. Unlike the CAM process, the tandem dehydrogenation/olefin metathesis process produces highly selective olefinic products. Researchers Conk and Wang et al. achieved propylene yields of over 80% and 94%, respectively, demonstrating the efficacy of this strategy [155,156].

Microwave-assisted pyrolysis (MAP) technology has demonstrated its efficiency in converting polyolefins into fuel oil, while also producing clean hydrogen fuels and value-added multi-walled carbon nanotubes (MWCNTs) [157,158]. Jie et al. reported that microwave-assisted catalytic deconstruction of plastic waste, achieving over 97% of the theoretical mass yield of hydrogen and producing carbon materials with more than 92wt% MWCNTs in approximately 20 s [159].

The introduction of heteroatoms such as oxygen and nitrogen during the depolymerization process can significantly enhance product value. The oxidative deconstruction process can convert polyolefins into high-value chemicals, including aliphatic alcohols and fatty acids [160]. Wang et al. developed a low-temperature, hydrogen-free process utilizing Ru/TiO_2 to upcycle PE, resulting in a liquid product primarily consisting of low-molecular-weight aliphatic dicarboxylic acids with an 85% yield [161]. Oxidative deconstruction can also occur via photo-oxidative C-C bond cleavage over a photocatalyst. As an example, single-unit-cell thick Nb_2O_5 layers enable the complete photodegradation of PE into CO_2 within 40 h and the resulting CO_2 is then further photoreduced to generate acetic acid [162]. Furthermore, Zhao et al. utilized the catalytic properties of manganese dioxide in conjunction with urea to introduce nitrogen atoms into the polyolefin depolymerization process, yielding high-value nitrogen-containing compounds such as nitriles and amides [163].

5. Polystyrene (PS, SPI Code 6)

PS is a widely used thermoplastic produced through the polymerization of styrene monomers, primarily used in packaging and construction applications. While styrene monomers can be recovered from polystyrene via non-catalytic pyrolysis, this method is associated with high energy consumption (approximately 500 °C) and results in complex product distributions [164]. In contrast, catalytic processes can achieve highly selective target products with lower energy consumption, while strategies such as photooxidative degradation further offer an alternative pathway for the production of value-added chemicals (Figure 10).

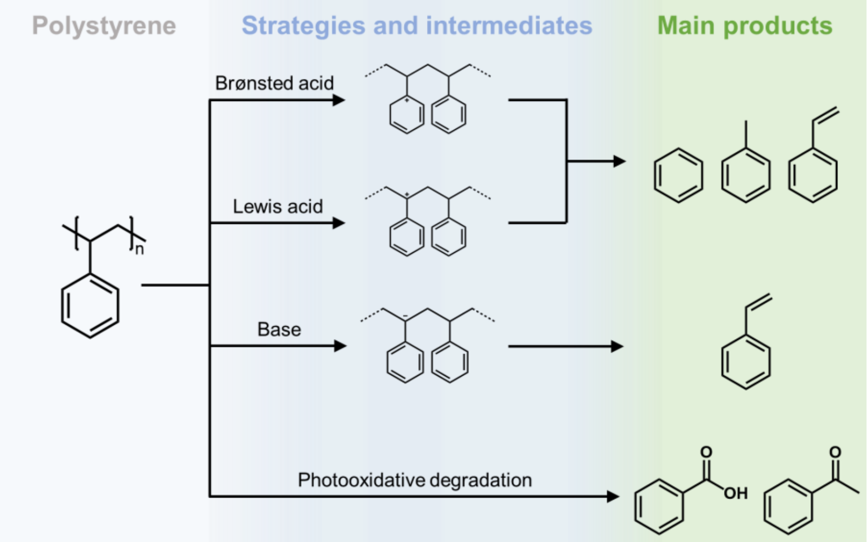


Figure 10. Chemical upcycling strategies of polystyrene and its main products.

During catalytic pyrolysis of PS, the choice of catalyst significantly influences the reaction pathways, thereby altering product distribution and selectivity. Specifically, the use of acidic catalysts (either Brønsted or Lewis) enables the efficient production of benzene and alkyl aromatics [165,166]. In this process, phenyl protonation on PS leads to the formation of Wheland intermediates, from which benzene can be generated through β-scissions of the secondary carbocation intermediates. Additionally, this catalytic pyrolysis can yield various byproducts, including alkylbenzenes, indane, and styrene, with aromatic olefins like styrene being prone to further conversion via hydrogen transfer [167,168]. Alkaline catalysts deprotonate PS, resulting in β-scissions of the aliphatic chain and yielding styrene as the main product [16].

In addition to converting PS to styrene, benzene, and alkyl aromatic hydrocarbons via catalytic pyrolysis, researchers have explored the transformation of PS into oxygenated aromatic compounds, including ketones, aldehydes and carboxylic acids [169–171], and nitrogen-containing aromatic amines [172]. Notably, much of the oxidative deconstruction of PS involves a photocatalytic process. A hydrogen atom transfer (HAT) mechanism can generate a selective carbon-centered radical on the phenyl ring of PS. Subsequent oxidative conditions facilitate the formation of an organic peroxide, which further undergoes C-C bond cleavage via photocatalysis, predominantly yielding benzoic acid [173,174]. Oh et al. further enhanced product selectivity by leveraging mechanistic insights [175]. They found that bromine radicals preferentially target tertiary C-H bonds, and HBr can donate a hydrogen atom to a terminal methyl radical, thereby favoring the formation of acetophenone as the main degradation product when an external bromine source is introduced to enhance HBr concentration.

Given that current PS upcycling methods usually rely on complex catalysts and encounter challenges such as product selectivity and economic viability, some studies have implemented a tandem upcycling strategy, involving two or more consecutive reactions to yield valuable chemicals [176]. For instance, the degradation-upcycling (Deg-Up) strategy was proposed by Xu et al. to convert PS waste into valuable chemicals such as diphenylmethane, benzophenone [177]. Similarly, Qin et al. first generated benzoic acid through photo-oxidation and then employed a one-pot tandem conversion to produce benzene, biphenyl, salicylic acid, toluene, and other important bioactive compounds [173].

6. Polyvinyl Chloride (PVC, SPI Code 3)

PVC accounts for 12.9% of global plastics production and is the third largest contributor just below PE and PP [2]. The presence of chlorine in PVC leads to the generation of corrosive HCl and other highly toxic chlorine-containing compounds, such as dioxins, during its chemical recovery [17,178]. Our group's work also demonstrate that HCl derived from PVC contributes to the poisoning of Pt/ZSM-5, which can be attributed to the removal of framework aluminum [179]. In comparison to the upcycling of various types of plastics discussed in Sections 3 to 5, the upcycling of PVC presents greater difficulties due to the challenges posed by chlorine species. Consequently, we address this topic last in our discussion.

Similar to the chemical recovery of polyolefins, PVC can be recovered through processes such as pyrolysis and gasification. In these processes, chlorine in PVC is recovered primarily in the form of hydrogen chloride or neutralization salts. Pyrolysis involves the thermal decomposition of PVC under anaerobic conditions and occurs in two stages. The first stage entails the removal of chlorine atoms through an autocatalytic dehydrochlorination process, resulting in the formation of π-conjugated polyenes from the remaining polymer [180]. These polyenes can readily undergo cyclic reactions, leading to the formation of cross-linked intermediates and ultimately yielding products such as polycyclic aromatic hydrocarbons (PAHs), non-condensable gases, and coke [181,182]. Gasification processes convert PVC into syngas-based products in the presence of air, oxygen, or steam. Overall, both processes involve the cleavage of C-Cl bonds at elevated temperatures, followed by the further degradation of hydrocarbons into fuels and small molecular compound. However, these processes often release corrosive gases that can damage equipment. Consequently, PVC upcycling research has focused on mild dechlorination methods to produce value-added products, such as fuels, aromatics and carbon materials through tandem processes [183–185]. The chlorine species in PVC can be converted into metal chlorides or organic chlorides (Figure 11) [186–188].

The relatively low binding energy of the C-Cl bond facilitates dechlorination through nucleophilic substitution and elimination reactions (dehydrochlorination), resulting in products that can be directly utilized or further transformed into higher-value chemicals [189]. In the nucleophilic substitution process, reagents containing sulfur, nitrogen, or oxygen attack the C-Cl bond, leading to the replacement of chlorine atoms. Concurrently, additives can be chemically grafted onto the polymer backbone, enabling the chemical modification [190–192]. the elimination process is typically conducted under alkaline conditions, where the C-Cl bond is cleaved, releasing hydrogen chloride and producing olefins [193]. Beyond the use of metal salts as catalysts, ILs have been investigated as alternative catalysts for dehydrochlorination due to their solubility and high alkalinity [194,195]. The interplay between these two reaction pathways can significantly impact the selectivity and efficiency of the dechlorination reaction, ultimately affecting product selectivity. Some studies have optimized this competitive relationship by modulating reaction parameters such as nucleophilicity, solvent temperature, and reaction time to enhance substitution yields [196,197]. Hydrothermal treatment (HTT) technology has been extensively studied in the context of dechlorination, exploring a range of pressures and temperatures from subcritical water to supercritical water, and investigating various enhancers to improve dechlorination efficiency [198–200]. Additionally, electrochemical and non-thermal plasma (NTP)-mediated strategies have been investigated, providing new pathways for the chemical upcycling of PVC [187,201].

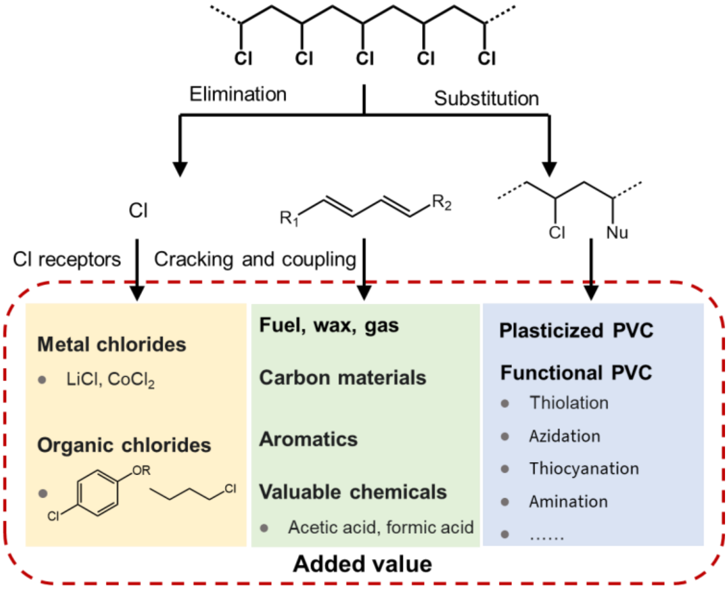


Figure 11. Value-added upcycling strategies for PVC.

7. Summary and Outlook

Upcycling technologies for plastics, which convert waste plastics into high-value monomers, fuels, and chemicals, are increasingly recognized as vital solutions to address plastic pollution and promote a circular economy. However, given the diversity of plastics, a one-size-fits-all recycling solution is not feasible. Instead, tailored upcycling strategies must be developed for each specific type of plastic. Significant progress has been made in various upcycling strategies for mainstream plastics, including solvolysis, catalytic pyrolysis, hydrocracking and hydrogenolysis, largely driven by innovations in catalysts and reaction systems. And this review seeks to provide an overview of the current advancements in upcycling technologies for these widely used plastics.

However, the process of plastics upcycling is fraught with numerous challenges and obstacles. One significant limitation arises from the physical properties of plastics, such as their poor solubility, which can hinder the effectiveness of many catalytic systems. In heterogeneous catalytic systems, the limited contact between the catalyst and the polymer molecules often results in reduced catalytic efficiency. Additionally, the complex reaction networks can lead to low product selectivity and catalyst deactivation due to carbon accumulation. To address these challenges, researchers are focusing on the design of efficient catalysts and the development of advanced catalytic technologies that enhance both reaction efficiency and selectivity. Strategies include fine-tuning the size and structure of active sites and optimizing interactions between polymers and their carriers. A notable example is the use of single-atom catalysts, which maximize metal atom utilization and leverage unique metal-carrier interactions to improve both the activity and stability of the catalyst [202]. Furthermore, enzyme-mimicking catalysts have been studied to achieve highly selective target products through precise design of active site structures [149]. Among the various catalytic upcycling strategies, thermal catalytic technology has received considerable attention due to its relevance to industrial applications. Additionally, emerging methods that operate under mild conditions, such as photocatalysis, electrocatalysis, and biocatalysis, also show significant potential for further development [203]. Tandem catalytic systems also offer promising avenues for enhancing the economic value of the plastic recycling process by enabling the design of successive reaction steps [204].

In current research, the majority of efforts are concentrated on model plastics. However, actual recycled plastic waste typically comprises a mixture of various plastic types and often contains numerous impurities, including additives, inorganic materials, and metals. Notably, the presence of PVC can generate corrosive and toxic by-products, such as HCl, which pose significant risks to catalyst performance and reactor integrity. Consequently, it is essential to effectively remove these contaminants prior to their introduction into the reactor. While some studies have investigated mixed plastics and low-purity plastic systems, there remains a notable gap in research concerning the dechlorination effects and chlorine resistance of catalysts [202,205,206]. Considering the incompatibility of PVC with conventional recycling processes, pyrolytic dehalogenation processes that incorporate various metal oxides and alkaline adsorbents show promising potential for industrial application as a cost-effective pre-treatment strategy [207]. In this context, the optimized modular extruder (MODIX), developed by VTT, presents a compelling solution for pre-treatment. This innovative system integrates preheating, impurity removal, and the feeding of melt-mixed plastic waste into a single, streamlined process [208]. Moreover, the introduction of alkaline earth metal oxides and Brønsted acids into the catalyst can effectively enhance both the deep dechlorination capability and the chlorine resistance of the reaction.

The field of chemical upcycling of plastics is currently experiencing rapid advancement. Progress in various upcycling technologies, particularly in catalyst development and optimization of reaction systems, has created new opportunities for efficient treatment of plastic waste. Future research should prioritize the enhancement of treatment technologies and dechlorination methods applicable to diverse plastic mixtures, as well as the assessment of their industrial feasibility. This focus will further promote the widespread adoption of plastic upcycling technologies and contribute to the overarching objective of transforming waste into valuable resources.

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