



Advanced Catalytic Strategies for Plastic Depolymerization: Mechanistic Insights, Catalyst Engineering, and Sustainable Upcycling Pathways

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Abstract

The exponential rise in plastic production and consumption has resulted in the accumulation of persistent polymeric waste, posing severe environmental and ecological challenges. Global plastic generation exceeds 400 million tonnes annually, with dominant materials including polyethylene terephthalate (PET), polyethylene (PE), polypropylene (PP), and polystyrene (PS). Advanced catalytic depolymerization has emerged as a sustainable strategy to convert plastic waste into valuable monomers, fuels, and chemical intermediates.

This review provides a comprehensive and expanded analysis of catalytic depolymerization approaches, including homogeneous, heterogeneous, biocatalytic, photocatalytic, and pyrocatalytic methods. Mechanistic pathways, catalyst design strategies, and reaction optimization are discussed in detail. The role of nanostructured materials, metal–organic frameworks (MOFs), enzyme engineering, and hybrid catalytic systems is critically evaluated.

Emerging technologies such as plasma-assisted catalysis and electrocatalysis are also explored for their potential in energy-efficient plastic valorization. The integration of these catalytic technologies is essential for advancing circular economy models and mitigating global plastic pollution.

Keywords: Plastic Depolymerization, Catalytic Recycling, Homogeneous Catalysis, Heterogeneous Catalysis, Photocatalysis, Pyrocatalysis.

1. Introduction

The widespread use of synthetic polymers has revolutionized modern society due to their durability, versatility, and cost-effectiveness. However, these same properties contribute to their persistence in the environment, leading to large-scale accumulation of plastic waste in terrestrial and marine ecosystems. The concept of the “plastisphere” highlights the complex interactions between plastic debris and microbial communities, further emphasizing the urgency of addressing plastic pollution^{1–3}.

Traditional waste management strategies, including landfilling and incineration, are associated with environmental hazards such as greenhouse gas emissions, toxic by-products, and inefficient resource recovery. Mechanical recycling, although widely practiced, suffers from limitations such as polymer degradation, contamination, and limited applicability to mixed plastic waste streams.

Catalytic depolymerization has emerged as a transformative approach that enables selective cleavage of polymer chains into monomers and value-added products under controlled conditions. This approach not only reduces environmental impact but also contributes to resource recovery and circular economy development^{4–6}.

Recent advancements in catalysis have focused on improving reaction efficiency, selectivity, and sustainability through the design of advanced catalysts, including nanomaterials, metal–organic frameworks (MOFs), and enzyme-based systems. Furthermore, interdisciplinary approaches integrating materials science, chemical engineering, and biotechnology are accelerating progress in this field^{5–8}.

This review provides an in-depth and expanded discussion of advanced catalytic strategies for plastic depolymerization, highlighting their mechanisms, advantages, limitations, and future prospects.



Scheme 1: General Depolymerization pathway of Polymers into oligomers and monomers

2. Homogeneous Catalysis

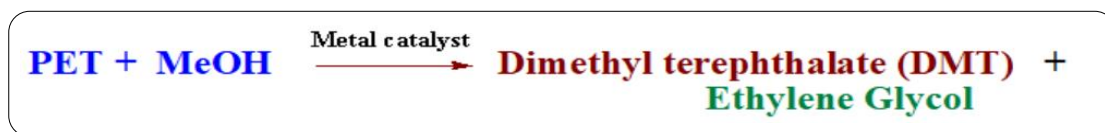
Homogeneous catalysis involves the use of soluble catalysts that form a single phase with the polymer substrate, enabling uniform interaction and efficient reaction kinetics. This approach allows precise control over reaction conditions, resulting in high selectivity and conversion efficiency^{5–7}.

Transition metal complexes, including ruthenium, iridium, and cobalt-based catalysts, have demonstrated significant potential in depolymerizing polyesters, polycarbonates, and polyamides through mechanisms such as hydrogenation, hydrolysis, and alcoholysis^{6,7}. Acid-catalyzed depolymerization processes have also been extensively studied for breaking ester and amide bonds in polymers.

Homogeneous catalytic systems are particularly effective for the degradation of thermosetting plastics and fiber-reinforced composites, which are challenging to recycle using conventional methods⁴. Additionally, computational studies and density functional theory (DFT) analyses have provided valuable insights into reaction mechanisms and catalyst–substrate interactions, enabling rational catalyst design⁷.

Recent research has focused on developing sustainable catalytic systems using earth-abundant metals and green solvents to minimize environmental impact⁸. However, challenges such as catalyst recovery, separation, and recyclability remain significant barriers to industrial application. Strategies such as biphasic systems, immobilization, and ligand design are being

explored to address these limitations^{5–8}.



Scheme 2: Homogeneous catalytic methanolysis of PET

3. Heterogeneous Catalysis

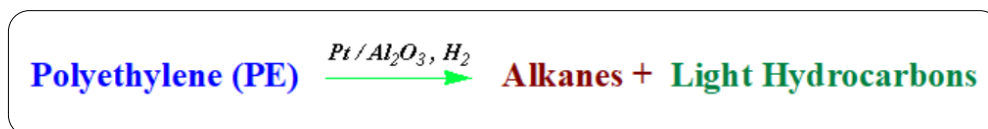
Heterogeneous catalysis has gained significant attention due to its operational advantages, including ease of catalyst separation, recyclability, and compatibility with continuous industrial processes^{9–11}.

Solid catalysts such as zeolites, metal oxides, and supported noble metals play a crucial role in facilitating polymer degradation. These catalysts enable C–C bond cleavage through mechanisms such as hydrogenolysis, hydrocracking, and olefin metathesis, producing hydrocarbons with high selectivity^{9,10}.

Zeolite-based catalysts (HZSM-5, HY, USY) exhibit strong acidity and well-defined pore structures, which enhance hydrocarbon formation and aromatization^{11,12}. Supported metal catalysts, including Pt, Ru, and Ni-based systems, have demonstrated high activity in polyolefin depolymerization under moderate conditions¹⁰.

Biochar-based catalysts represent a sustainable alternative, offering high surface area, tunable porosity, and environmental compatibility. These catalysts have shown promising results in converting plastic waste into liquid fuels and valuable chemicals¹³.

Despite these advantages, heterogeneous catalysis faces challenges such as catalyst deactivation due to coke formation, diffusion limitations, and pore blockage. Advanced catalyst design, including hierarchical structures and bifunctional catalysts, is being explored to overcome these limitations^{11–13}.



Scheme 3: Heterogeneous catalytic hydrogenolysis of polyolefins

4. Biocatalysis

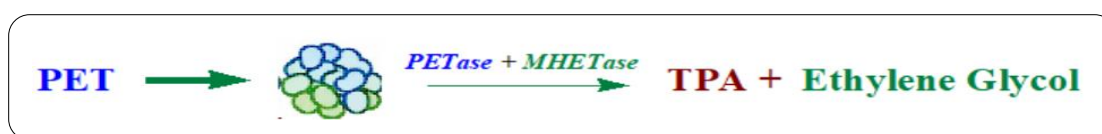
Biocatalytic depolymerization utilizes enzymes and microorganisms to achieve selective polymer degradation under mild conditions, offering an environmentally friendly alternative to conventional methods^{14–17}. Page | 99

Enzymes such as PETase, MHETase, cutinases, and laccases play a crucial role in breaking down polymer chains into monomeric units. Engineered enzymes have demonstrated significantly improved catalytic efficiency and thermal stability, enabling rapid hydrolysis of crystalline PET into terephthalic acid and ethylene glycol¹⁴.

Advances in protein engineering, including directed evolution and computational design, have enabled the development of robust biocatalysts with enhanced substrate specificity and activity¹⁵. Microbial consortia capable of expressing multiple enzymes have been developed to address the complexity of mixed plastic waste streams¹⁶.

Immobilization techniques further enhance enzyme stability and recyclability, making biocatalytic processes more suitable for industrial applications. Integration of biocatalysis with chemical depolymerization processes has also been explored to achieve complete conversion of polymers into valuable products^{17,18}.

However, challenges such as limited substrate accessibility, slow reaction kinetics, and scalability issues remain significant barriers. Ongoing research focuses on improving enzyme performance and developing hybrid catalytic systems to overcome these limitations^{15–18}.



Scheme 4: Enzymatic hydrolysis of PET using engineered enzymes

5. Photocatalytic Methods

Photocatalysis has emerged as a promising approach for plastic depolymerization, utilizing light energy to drive chemical reactions and reduce energy consumption^{19–22}.

Semiconductor photocatalysts generate electron–hole pairs upon light irradiation, leading to the formation of reactive radicals such as hydroxyl and superoxide species. These radicals initiate polymer chain scission through oxidative and reductive pathways^{19,20}.

Advanced photocatalytic systems incorporate nanostructured materials, doped semiconductors, and metal–organic frameworks (MOFs) to enhance charge separation and catalytic efficiency^{20,21}. Hybrid systems combining photocatalysis with persulfate oxidation or chemical depolymerization have demonstrated improved reaction kinetics and selectivity^{22,23}.

Photocatalytic depolymerization of vinyl polymers, including polystyrene and polymethyl methacrylate, proceeds via β -scission mechanisms, enabling the recovery of monomers such as styrene and methyl methacrylate¹⁹.

Despite its advantages, photocatalysis faces challenges such as limited light absorption, recombination of charge carriers, and control of reaction selectivity. Future research focuses on developing advanced photocatalysts with improved efficiency and stability^{20–23}.



Scheme 5: Photocatalytic depolymerization via radical scission

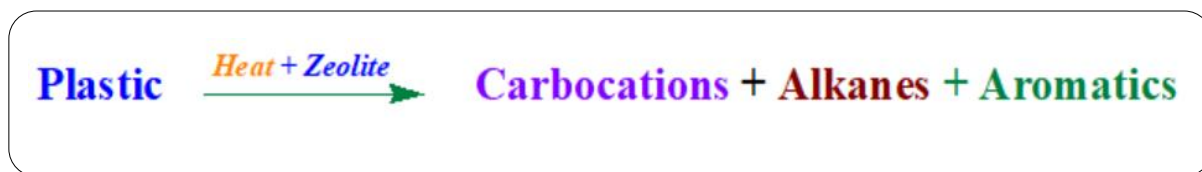
6. Pyrocatalytic Methods

Pyrocatalysis involves the thermochemical degradation of plastics in the presence of catalysts under inert conditions. This process is driven by radical-mediated chain scission and facilitated by catalysts such as zeolites and silica–alumina materials²⁴.

Catalysts promote carbonium ion formation, enabling efficient cleavage of polymer chains into hydrocarbons such as alkanes, olefins, and aromatic compounds. Pyrocatalytic depolymerization is particularly effective for processing mixed plastic waste and achieving high conversion efficiencies²⁴.

Advanced catalyst systems, including hybrid nanoparticles and carbon-based materials, have been developed to enhance catalytic performance and product selectivity. Process optimization, including temperature control and reactor design, plays a critical role in improving efficiency.

However, high energy requirements and catalyst deactivation remain significant challenges. Research efforts are focused on developing energy-efficient processes and improving catalyst stability²⁴.



Scheme 6: Pyrocatalytic Depolymerization via carbocation cracking

7. Emerging Catalytic Approaches

7.1 Non-Thermal Plasma Catalysis

Non-thermal plasma enables plastic depolymerization through the generation of reactive species at near-ambient conditions, offering energy-efficient and environmentally sustainable processing routes. Plasma-assisted catalysis can enhance reaction rates and selectivity by combining plasma activation with catalytic surfaces.

7.2 Electrocatalysis

Electrocatalytic methods utilize electrical energy to drive polymer degradation reactions. These processes enable selective bond cleavage and conversion into valuable chemical

products, supporting circular economy initiatives and sustainable chemical production.

8. Conclusion

Advanced catalytic strategies provide promising solutions for addressing the global plastic waste crisis. Homogeneous and heterogeneous catalysis offer high efficiency and scalability, while biocatalysis provides environmentally sustainable alternatives under mild conditions. Photocatalytic and pyrocatalytic methods contribute to energy-efficient and large-scale recycling processes. Emerging technologies such as plasma and electrocatalysis further expand the scope of plastic valorization.

Future research should focus on improving catalyst durability, enhancing reaction selectivity, and developing scalable industrial processes. Integration of interdisciplinary approaches will be essential for achieving sustainable and economically viable plastic recycling systems.

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Conflict of Interest/Competing Interests

No conflict of interest.

Data Availability

No quantitative data is used as it is a conceptual paper.

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