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Current perspective on heterogeneous thermal catalytic approaches for closed-loop polyolefin plastic recycling

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Abstract
The escalating accumulation of polyolefin (PO) waste presents one of the most formidable challenges in sustainable catalysis. Heterogeneous catalytic upcycling has emerged as a promising route toward chemical circularity, yet genuine closed-loop recycling—where polymers are converted back into monomers for repolymerization—remains challenging. This perspective critically examines three catalytic pathways: ethylene metathesis, hydrocracking coupled with steam cracking, and high-temperature or tandem catalytic cracking. While each offers distinct advantages in selectivity, scalability, or process simplicity, none independently satisfy the combined criteria of low energy demand, carbon neutrality, and economic viability. Future progress requires low-carbon, renewable-powered catalytic pathways, realistic PO waste investigation, highly selective and adaptive catalysts, and integration of techno-economic and life-cycle assessments. By bridging molecular-level catalyst design with systems-level process engineering, heterogeneous catalysis can transform PO waste from a persistent pollutant into a renewable carbon resource, forming the cornerstone of a sustainable, circular plastic economy.

1 Introduction

Since their large-scale commercialization in the 1950 s, synthetic plastics have become integral to modern civilization, with global production surpassing 430 million tons in 2024 [1, 2]. Their exceptional versatility, durability, lightweight character, and low production cost have enabled widespread use across nearly every sector of daily life, including packaging, construction, transportation, electronics, agriculture, medical devices, textiles, and household goods [2–5]. However, these very attributes—particularly their remarkable chemical and thermal stability—that once symbolized progress and convenience now underpin one of the most persistent environmental challenges of our time [6–8]. As estimated, less than 10% plastics have been effectively recycled, while the majority are landfilled, incinerated, or dispersed into terrestrial and marine ecosystems [1]. Traditional plastic waste management methods, such as landfilling, incineration, mechanical recycling, and pyrolysis, are typically energy-intensive and yield low-value products, often accompanied by greenhouse gas emissions or toxic pollutants [4–7]. Consequently, these strategies are fundamentally unsustainable, driving the urgent search for more circular and resource-efficient solutions.

In recent years, chemical recycling, particularly catalytic approaches, has emerged as a transformative alternative to mechanical recycling. Unlike physical reprocessing, chemical recycling allows for molecular deconstruction of polymers into their monomeric or small-molecule building blocks, thereby enabling true carbon circularity [9, 10]. A broad range of heterogeneously catalyzed transformations, including catalytic cracking, hydrocracking, hydrogenolysis, alkylation, oxidation, nitrogenation, metathesis, and tandem catalysis, have been developed to convert waste plastics into naphtha, gasoline, diesel, lubricants, diacids, amides, nitriles, and olefins, often under milder conditions than those required by traditional thermal pyrolysis or gasification [4–7, 10–21]. Importantly, many of these catalytic processes can be seamlessly integrated into existing petrochemical infrastructures, offering a realistic pathway for large-scale implementation.

Despite these advances, true closed-loop recycling–where polymers are chemically upcycled into monomers or intermediates that can be repolymerized into materials with properties equivalent to the virgin resins–remains challenging. Among the various plastic classes, polyolefins (POs), such as polyethylene (PE) and polypropylene (PP), present the greatest difficulty. These polymers account for approximately half of global plastic production and over 60% of post-consumer plastic waste, yet their strong C–C bonds and chemical inertness render them highly resistant to depolymerization [1, 2, 22, 23]. Furthermore, the complexity of real plastic waste—additives, fillers, pigments, and mixed polymer streams—exacerbates the challenge of selective conversion [24].

In this perspective, we critically examine the current state of heterogeneous catalysis for closed-loop plastic waste recycling, with particular emphasis on PO upcycling. We will discuss fundamental mechanistic insights into catalytic C-C bond activation and practical process strategies for achieving chemical circularity. We also evaluate their strengths and limitations, and discuss emerging trends in catalyst design, reaction engineering, and process integration. Finally, we will propose how next-generation heterogeneous catalysts—capable of controlling selective bond activation, integrating tandem functionalities—may reshape the field and accelerate the transition toward sustainable, closed-loop plastic recycling. By bridging molecular-level catalyst design with systems-level recycling strategies, heterogeneous catalysis can ultimately transform plastic waste from a global pollutant into a renewable carbon resource, paving the way toward a sustainable, circular plastic economy.

2 Current routes for Closed-Loop polyolefin plastic recycling

Despite substantial advances in heterogeneous catalysis for PO upcycling, achieving a true closed-loop recycling process—where plastics are upcycled back to monomers suitable for repolymerization—remains challenging. To date, three primary catalytic routes have been reported in the literature, as summarized schematically in Fig. 1.

1. **Route I**: Ethylene-assisted metathesis of POs to yield propylene monomers, followed by re-polymerization to regenerate PP.
2. **Route II**: Hydrocracking of POs to naphtha-range hydrocarbons, which are subsequently steam-cracked to C_2–C_3 olefins and re-polymerized into PE or PP.

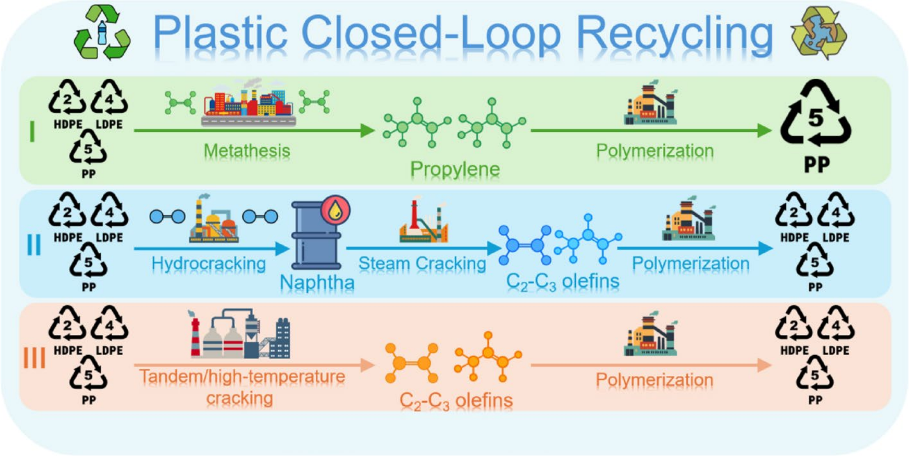

Fig. 1 Three routes for polyolefin plastic waste closed-loop recycling

3. **Route III**: Direct high-temperature or tandem cracking of POs into C_2–C_3 olefins, bypassing the naphtha intermediate.

Each route has distinct mechanistic pathways, catalytic requirements, and techno-economic trade-offs, which are discussed below:

Route I – Ethylene-Assisted Metathesis

The metathesis pathway represents one of the most conceptually elegant closed-loop recycling strategies because it directly converts PE/PP into propylene monomers using ethylene as a co-reactant. Catalysts such as $MeReO_3/Cl–Al_2O_3$ and WO_3/SiO_2 have been employed, operating at relatively low temperatures (even ~ 100 °C) [20, 25]. In this reaction, ethenolysis cleaves internal C = C bonds in the polymer chain, yielding shorter olefins. A subsequent isomerization step shifts terminal olefins to internal positions, enhancing propylene selectivity. Fully or partially unsaturated polymers (containing C = C bonds) can participate directly, whereas saturated POs require a preliminary dehydrogenation or thermal cracking step to introduce olefinic sites. For instance, $PtSn/Al_2O_3$ catalysts can perform selective dehydrogenation, while Na/Al_2O_3 can induce low-temperature chain activation. The resulting propylene can then be re-polymerized to regenerate PP, thus completing a closed-loop cycle.

Although metathesis can operate under mild conditions and achieve high propylene selectivity, several economic and thermodynamic constraints limit its large-scale applicability. Due to the fact that the reaction equilibrium favors longer-chain olefins, a large excess of ethylene is required to continuously drive the reaction toward propylene formation. In practice, significant ethylene is also lost to side reactions such as transfer dehydrogenation. Given that propylene's large-scale market price (≈ USD 700–900 t^{-1}) is only about twice that of ethylene (≈ USD 500 t^{-1}) [26, 27], the economic incentive for converting ethylene into propylene via polymer intermediates is marginal. Therefore, although this route achieves the chemical ideal of "plastic-to-plastic" conversion, the process economics and ethylene demand remain substantial barriers to commercialization.

Route II – Hydrocracking to Naphtha Followed by Steam Cracking

In Route II, POs are first hydrocracked into naphtha-range hydrocarbons (C_5–C_{12}), which are subsequently subjected to steam cracking to yield a mixture of ethylene and propylene. These olefins can subsequently be re-polymerized into PE or PP, thereby establishing a closed-loop route. This approach typically employs bifunctional catalysts comprising metal sites (Pt, Ni) for (de)hydrogenation and acidic supports such as β-zeolite for C–C bond scission [13, 24, 28]. Hydrocracking can proceed at moderate temperatures (200–300 °C) under hydrogen atmosphere, producing narrow-range liquid fractions with high selectivity. However, the secondary steam-cracking step represents the major energy bottleneck. Steam cracking requires extremely high temperatures (≈ 850 °C) and substantial steam input to produce ethylene/propylene mixtures, followed by energy-intensive separation of these olefins [29, 30]. Although this process avoids the use of costly co-reactants such as ethylene (as in Route I), the high thermal demand of steam cracking and the separation costs of mixed olefin streams significantly diminish the net energy and economic efficiency.

From an industrial integration standpoint, Route II is compatible with existing refinery operations and may be viable in regions with low-cost renewable hydrogen. Nevertheless, its overall carbon intensity remains high, suggesting that this route is better suited as a transitional bridge rather than a long-term circular solution.

Route III – Direct Cracking or Tandem Catalytic Cracking to C_2-C_3 Olefins

Route III bypasses the naphtha intermediate entirely by directly converting POs into light olefins through either high-temperature thermal cracking or tandem catalytic processes. This route offers higher atom efficiency but poses greater challenges in energy management and selectivity control. Recent advances illustrate how multi-stage catalytic systems can decouple polymer chain scission and olefin formation. For example, Bi et al. (2025) reported a kinetic decoupling–recoupling strategy using a dual-reactor tandem system with dual zeolite catalysts [31]. In the first reactor, modified ZSM-5 at 260 °C converts long polymer chains into shorter intermediates (C_4, C_5 and above). In the second stage, these intermediates are transformed into C_2–C_3 olefins at 540 °C over P-modified zeolites, achieving high overall olefin yields. In parallel, Joule heating–based processes have demonstrated rapid polymer decomposition at temperatures exceeding 1000 °C within seconds, producing high C_2-C_3 selectivity even without external catalysts [32, 33]. Such processes exploit resistive or plasma heating for ultrafast temperature ramping, minimizing over-cracking and coke formation. While Route III avoids the need for co-reactants or secondary steam cracking, both high-temperature tandem catalysis and pyrolysis-based systems remain energy-intensive. The use of Joule heating to achieve the ultrafast heating is still mainly in lab-scale and hard to control, making it different to further commercialization. Moreover, the ethylene/propylene mixtures produced still require downstream separations, adding to process costs.

In summary, the three catalytic routes towards closed-loop PO recycling represent distinct yet complementary strategies, each associated with characteristic techno-economic and life-cycle trade-offs that collectively define the emerging landscape of catalytic plastic circularity. To facilitate comparison, Table 1 summarizes these routes from the perspectives of technology readiness, process efficiency, economic feasibility, and sustainability. Route I, based on ethylene-assisted metathesis, offers a conceptually elegant, low-temperature pathway with high propylene selectivity and favorable intrinsic

Table 1 Systematic comparison of closed-loop PO recycling routes from technological maturity, process performance, economic feasibility, and sustainability perspectives

Metric	Route I: Ethylene-Assisted Metathesis	Route II: Hydrocracking + Steam Cracking	Route III: Direct/Tandem Cracking
Primary products	Propylene	C_2–C_3 olefins	C_2–C_3 olefins
Operating temperature	Low (≈ 100–200 °C)	Moderate (hydrocracking: 200–300 °C) + high (steam cracking: ≈850 °C)	High (≈ 500–1000 °C, depending on process)
Energy input	Low	High (steam cracking and olefin separation)	High (thermal/Joule heating and olefin separation)
Carbon emission	Moderate–high (ethylene co-feed penalty)	High (energy-intensive steam cracking and olefin separation)	Moderate–high (severe thermal input and olefin separation)
Process efficiency	High (single-step reaction with high selectivity)	Low (two-step process with extensive separation)	Moderate–high (tandem reactions with separation)
Economic potential	Low (strongly dependent on ethylene cost)	Moderate-high	Moderate-high
Catalyst lifetime	Generally robust and regenerable	Generally robust and regenerable	Limited by coking and thermal stability
Scalability	Moderate	High (refinery-compatible)	Moderate for tandem cracking; limited for Joule heating

energy efficiency. However, from a techno-economic assessment (TEA) and life-cycle assessment (LCA) perspective, its reliance on a large excess of ethylene—currently predominantly fossil-derived—introduces a substantial carbon footprint and economic penalty. The narrow price margin between ethylene feedstock and propylene product further limits its scalability and greenhouse-gas (GHG) reduction potential unless low-carbon ethylene sources become widely available. Route II, combining hydrocracking with steam cracking, benefits from high technological maturity, robust catalyst lifetime, and direct compatibility with existing refinery infrastructure, which favors near-term scalability. Nevertheless, its overall sustainability is constrained by the very high energy input associated with steam cracking, significant CO_2 emissions, and energy-intensive downstream separation of C_2-C_3 olefins, resulting in modest net GHG reduction despite high process robustness. Route III, involving direct or tandem catalytic cracking and emerging Joule-heating-assisted processes, offers improved atom efficiency by bypassing intermediate steps and can achieve high olefin selectivity. From a life-cycle standpoint, this route holds promise for reduced process complexity and potentially lower separation burdens. However, its current reliance on severe thermal conditions, catalyst deactivation via coking or sintering, and the limited technological maturity of Joule-heating reactors pose challenges for long-term catalyst stability, capital cost, and large-scale continuous operation.

Taken together, no single route presently satisfies the combined TEA–LCA criteria of low energy consumption, minimal carbon footprint, high selectivity, long catalyst lifetime, and economic scalability required for genuine circularity. Future progress will therefore depend on the co-optimization of catalyst durability, low-carbon energy integration, feedstock sourcing, and process intensification, guided by early-stage TEA and LCA frameworks to identify pathways with truly sustainable closed-loop potential.

3 Conclusions and further perspectives

The closed-loop recycling of PO plastics remains one of the foremost challenges in sustainable catalysis. While remarkable progress has been achieved in heterogeneous catalytic upcycling, the development of polymer-to-monomer conversion pathways that deliver true material circularity is still in its infancy. From a comparative standpoint, Route I excels in selectivity and mild conditions, Route II in industrial maturity and refinery compatibility, and Route III in process simplicity and direct olefin production. However, none of these approaches independently meet the combined criteria of low energy demand, carbon neutrality, and economic competitiveness required for real-world closed-loop recycling.

Consequently, the next generation of catalytic plastic recycling must evolve beyond incremental advances toward rational catalysis design and integrated system engineering. Several promising directions are envisioned:

3.1 Low-Carbon catalytic pathways and renewable integration

Replacing traditional, hydrogen-intensive hydrocracking processes with low-carbon catalytic pathways offers a viable route to reduce dependence on fossil-derived hydrogen and lower GHG emissions. Emerging catalytic strategies–such as acid-catalyzed cracking, and alkylation–can yield similar carbon-range products to hydroconversion without requiring external H_2 input [14, 34, 35]. Furthermore, coupling these reactions with renewable energy inputs–for example, utilizing sunlight to drive the thermal catalytic conversion, or driving hydrogenolysis using green hydrogen from water electrolysis–can substantially decrease the carbon footprint of the overall process [36, 37].

Beyond energy supply, innovations in reactor engineering will be crucial. Tandem reactors or membrane-assisted catalytic modules capable of combining cracking and separation steps within integrated systems could improve heat management, hydrogen utilization, and process intensification. Together, these developments point toward electrified and renewable-powered catalytic processes, transforming plastic recycling from an energy-intensive operation into a carbon-efficient, circular technology.

3.2 From model PO feeds to realistic waste streams

Although substantial progress has been achieved in developing catalytic strategies for closed-loop PO recycling, the majority of studies still rely on idealized model feeds, such as pure PE or PP, which often exhibit high conversion and apparent catalyst stability. In contrast, realistic post-consumer plastic waste streams are compositionally complex, typically containing 1–10 wt% organic additives (e.g., antioxidants, plasticizers, stabilizers), inorganic fillers (e.g., $CaCO_3$, TiO_2), residual catalysts, and mixed polymer fractions such as PE/PP, PE/ethylene vinyl acetate (EVA), or PE/polyvinyl chloride (PVC) blends. Bridging the gap between model systems and realistic waste feeds is therefore essential for assessing the practical viability of catalytic upcycling technologies.

Recent studies have begun to provide quantitative insights into catalyst performance under realistic waste conditions. Ngu et al. systematically investigated the impact of representative organic additives, including hindered amine light stabilizers (HALS) and phenolic antioxidants, at concentrations relevant to post-consumer plastics (≈ 1–5 wt%), and demonstrated severe deactivation of metal and bifunctional catalysts due to strong adsorption on metal sites and neutralization of Brønsted acid sites [24]. Similarly, PO blends containing EVA with oxygen contents of 5–15 wt% were shown to accelerate coke formation and catalyst deactivation during hydrogenolysis and hydrocracking. To address these challenges, Zhao et al. demonstrated a tandem hydrodeoxygenation–hydrogenolysis catalyst could selectively remove oxygenated fragments derived from EVA prior to C–C bond scission, enabling stable conversion of PE/EVA mixtures with significantly reduced coke formation [38]. In another study, sulfide-based catalysts combined with in-situ sorbents were shown to sustain activity in mixed plastic feeds containing PVC and nitrogen-containing stabilizers by selectively capturing HCl and NH_3, thereby protecting active sites and preserving product selectivity [39]. These case studies highlight clear differences in catalyst stability, selectivity, and regeneration behavior when transitioning from model feeds to additive-laden waste streams.

Despite these advances, systematic datasets correlating catalyst performance with additive type, concentration, and polymer blend composition remain limited. Future studies should therefore prioritize controlled evaluations using realistic waste formulations, including defined additive loadings and mixed polymer ratios, coupled with long-term stability and regeneration analyses. Such efforts will be critical for translating laboratory-scale catalytic insights into robust technologies capable of processing post-consumer PO waste.

3.3 Selective and adaptive catalyst design

Among the catalytic routes discussed, ethylene-assisted metathesis (Route I) uniquely enables the selective production of propylene monomers without requiring extensive downstream separation. However, achieving such high selectivity across compositionally diverse PO waste streams demands catalysts with precise active-site control and tailored surface environments. Future catalyst design efforts should therefore focus on rationally tuning pore architecture to simultaneously facilitate polymer chain activation and regulate product distribution. In this context, advances in hierarchically porous materials, which alleviate macromolecular diffusion limitations while retaining shape-selective microporous domains, represent an important step toward improved performance [40]. Beyond single-function catalysts, the development of bimetallic and bifunctional systems has enabled efficient tandem catalytic pathways that enhance process intensification. Representative examples include Sn–W-based catalysts for tandem dehydrogenation–metathesis reactions, as well as Pt–Ru-based catalysts that couple dehydrogenation of liquid organic hydrogen carriers (e.g., methanol and formic acid) with subsequent plastic hydrogenolysis [41–43]. In parallel, metal–organic frameworks (MOFs) and MOF-derived materials have emerged as promising platforms for plastic upcycling due to their highly tunable metal coordination environments and pore structures [44, 45]. Continued development of these materials is expected to further expand the design space for selective and robust catalysts.

In addition, the integration of artificial intelligence (AI) and machine learning (ML) into catalyst discovery workflows offers powerful opportunities to accelerate identification of structure–performance relationships, enabling rapid screening of catalyst compositions and predictive optimization of reaction conditions. Looking beyond static materials, the development of adaptive catalysts capable of dynamically adjusting their electronic and structural states in response to changing polymer feedstocks will be critical for processing realistic waste streams. Such “intelligent” catalytic systems, when combined with in-situ and operando characterization techniques, can provide unprecedented insight into active-site dynamics and guide the rational design of next-generation heterogeneous catalysts for closed-loop PO recycling.

3.4 Sustainability metrics and lifecycle integration

The advancement of catalytic plastic recycling must be guided by rigorous TEA and LCA to ensure that promising laboratory processes translate into environmentally and economically viable industrial operations. These tools can quantify trade-offs between energy input, carbon footprint, and resource utilization, enabling the prioritization of catalytic routes that offer the highest overall sustainability benefit.

Equally important is cross-sector collaboration among academia, industry, and policymakers to align catalyst innovation with the broader circular-economy framework. Coordinated efforts in regulation, infrastructure development, and market incentives—for example, carbon pricing or recycled-content mandates—will be essential to accelerate commercialization and adoption.

The frontier of heterogeneous catalysis is shifting from the simple conversion of waste plastics to the design of catalytic circularity—a paradigm in which catalyst structure, polymer chemistry, and process engineering are co-optimized to achieve complete carbon reutilization. Future breakthroughs will rely on integrating mechanistic insight, materials innovation, and systems engineering to bridge the gap between molecular-level transformations and industrial-scale circularity. By coupling the selectivity and precision of molecular catalysis with the robustness and scalability of heterogeneous systems, it is conceivable to transform plastics from persistent pollutants into renewable carbon resources. In this emerging vision, heterogeneous catalysis serves not merely as a depolymerization tool but as a foundational platform for circular chemistry, enabling the sustainable regeneration of polymers and redefining the material economy for a carbon-neutral future.

Author contributions

H.W. received the invitation to contribute this perspective and was responsible for the conceptualization, writing, and revision of the manuscript. D.O'H. reviewed and provided critical feedback on the manuscript. Both authors approved the final version of the work.

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Data availability

No datasets were generated or analysed during the current study.

Author information

Competing interests

The authors declare no competing interests.

Ethical approval and Consent to participate

Not applicable.

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