

Review

Designing truly sustainable polymers through an integrated three-pillar framework

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SUMMARY

Most commodity polymers today are petroleum based, developed nearly a century ago with priorities on performance, durability, scalability, profitability, and disposability. This traditional linear economic model neglects crucial sustainability factors, leading to profound societal, economic, and environmental issues, such as resource depletion, environmental pollution, and loss of energy and material value. Although much progress has been made in developing polymers that utilize bio-based feedstocks, deliver useful performance, and show recyclability or biodegradability, these efforts are often pursued in isolation. In this review, we explore the three core pillars of sustainable polymers—renewable resources, built-in functions, and circular pathways—through the integrated lens of “produce-performance-regenerate.” We uncover the inherent disconnect between intrinsic material advantages and real-world implementation benefits. Employing a SWOT (strengths, weaknesses, opportunities, and threats) analysis, we identify molecular design strategies essential for developing scalable, high-performance, and truly circular polymers, which aligns polymer innovation with the goals of a sustainable materials economy.

INTRODUCTION

Global polymer production, including plastics, fibers, elastomers, adhesives, and coatings, has increased exponentially over the past few decades, exceeding 400 million tons annually. This surge has led to mounting environmental challenges, including resource depletion, persistent plastic waste accumulation, and rising greenhouse gas emissions.^{1–4} While plastics remain the most visible contributor due to their large-scale single-use applications, other polymer-based materials, such as textiles, rubbers, and coatings, also contribute significantly to carbon emissions and waste generation throughout their life cycles. Projections suggest that without systemic intervention, plastic leakage into terrestrial and marine ecosystems could triple by 2040.¹ This escalating crisis underscores the urgent need for transformative material innovation to decouple polymer functionality from environmental degradation.^{5–7} Despite growing awareness of these impacts, petrochemical-based plastics continue to dominate the global market, owing to their desired mechanical performance, ease of processing, and low cost. Their continued reliance, however, is fundamentally constrained by intractable end-of-life (EoL) challenges, calling for a paradigm shift in polymer design centered on full-life-cycle sustainability.

Addressing these challenges requires embedding polymer innovation within the broader principles of sustainability. Sustainability, at its core, demands that resource utilization remain within nature's regenerative capacity, which can be expressed

in relative rates as $k_{(\text{resource regeneration})} \geq k_{(\text{resource utilization})}$. Applied to polymers, this principle requires designs that minimize environmental burdens across their entire life cycle while maintaining or even surpassing the performance of less sustainable, conventional plastics. Critically, such sustainability must operate within realistic regenerative timescales to ensure lasting materials security. For clarity and focus, sustainable polymers are referred to as those that embody three integrated elements: renewable resources to produce, built-in functions to perform, and circular pathways to regenerate.

To date, efforts toward sustainable polymers have largely advanced along three major axes⁸: first, the use of renewable carbon sources, such as lignocellulose, fatty acids, and CO₂, to produce polymers or their precursors (monomers) while reducing dependence on fossil feedstocks (renewability)^{9–12}; second, the molecular engineering of built-in functionalities to ensure mechanical strength, thermal stability, and processability on par with or exceeding that of commodity plastics (functionality)^{13–16}; and third, the development of circular regeneration strategies, including chemical recycling and controlled degradation, to facilitate efficient monomer recovery and minimize long-term environmental persistence (circularity).^{17,18} Yet, these approaches often operate in isolation and fall short of delivering fully integrated solutions for full-life-cycle sustainability. For instance, many biodegradable polymers require highly controlled industrial composting conditions to degrade effectively, which significantly limits their environmental utility in



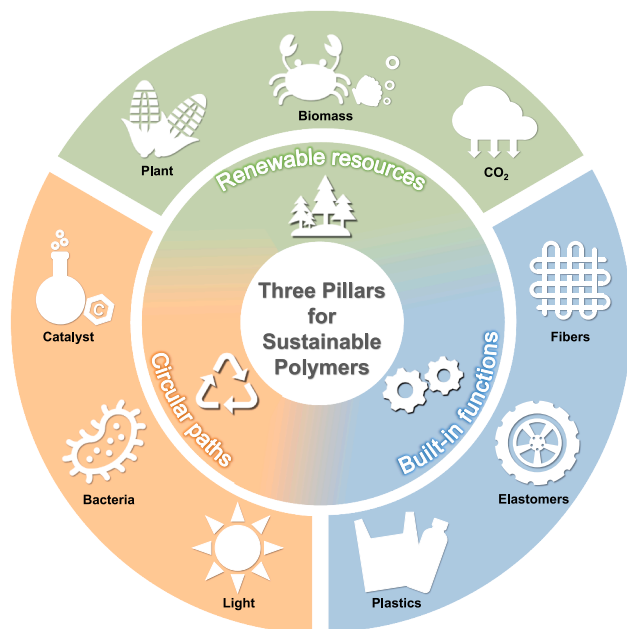


Figure 1. Three pillars for sustainable polymers

Synergistic integration of three pillars—renewable resources, built-in functions, and circular paths—embodied in the produce-perform-regenerate paradigm of next-generation sustainable polymers.

natural or uncontrolled environments.¹⁹ Bio-based polymers, though renewable in origin, often face persistent challenges related to mechanical performance and recyclability.²⁰ And chemically recyclable polymers hold compelling promise for closed-loop circularity without compromising material performance, yet their widespread adoption is still challenged by high energy intensity, limited scalability, and cost ineffectiveness.⁵ Beyond the laboratory, the broader transition to sustainable polymers is further constrained by systemic barriers, including high production costs, underdeveloped manufacturing infrastructure, inconsistent material performance across diverse applications, and a lack of standardized sustainability metrics. In addition, the dominance of low-cost, well-established fossil-based plastics and the complexity of sorting and recycling heterogeneous waste streams continue to impede widespread adoption.

Despite extensive reviews covering bio-based polymers,^{9–12,20,21} high-performance materials,^{22,23} and recyclable or degradable polymers,^{17–19,24–32} there remains a lack of unified perspectives that integrate polymer molecular design with life cycle sustainability principles (Figure 1). In particular, few efforts have synthesized recent advances across these domains to illuminate how renewability, functionality, and circularity can be holistically balanced at the molecular and macromolecular levels.

In this review, we intend to critically analyze the three key qualities that define sustainable polymers: renewable resources to produce, built-in functions to perform, and circular paths to regenerate. We reveal the persistent misalignment between material functionality and broader sustainability performance. Building upon this assessment, we outline a unified three-pillar framework consolidated through a strengths, weaknesses, opportunities, and threats (SWOT) analysis, embodied in the

“produce-perform-regenerate” paradigm, offering a strategic perspective for integrative perspectives that bridge molecular design with life cycle principles toward sustainable polymers.

THREE ESSENTIAL PILLARS FOR SUSTAINABLE POLYMERS

Polymers are macromolecules comprising many, typically covalently linked, repeating units of the same or different small molecules (monomers) arranged in specific manners through 1D, 2D, or 3D architectures. Because of their length scales far beyond typical small-molecule dimensions (i.e., nanometers to centimeters), as well as functions and interactions that are compounded to demonstrate macroscopic thermal, mechanical, and rheological properties, they represent one of the most versatile classes of synthetic materials encompassing plastics, fibers, elastomers, adhesives, and coatings, underpinning nearly every aspect of modern life. This macroscopic performance stems from precisely engineered molecular connectivity and chain structure. Yet, the same durability and stability that make polymers useful, long lasting, and indispensable also result in persistent waste and environmental accumulation—defining the central sustainability dilemma of polymeric materials.^{20,22,31–34}

In the broader context of sustainability, human activities must align with nature’s regenerative capacity (i.e., $k_{(\text{resource regeneration})} \geq k_{(\text{resource depletion})}$), which means that resource renewal should at least match consumption. In an ideal scenario, a truly sustainable polymer would combine renewable carbon origin, designer performance functionality, and intrinsic capacity for regeneration at EoL: while minimizing life cycle environmental impacts, matching or exceeding the performance of their conventional counterparts is equally vital. Crucially, these processes must occur within practical regenerative timescales, ensuring long-term sustainability. Achieving this balance requires a holistic design principle that integrates sustainable chemistry, polymer science, process engineering, and life cycle analysis.^{26,35}

The three essential pillars that collectively define sustainable polymers are renewability, functionality, and circularity (Figure 1).^{8,36} Each represents a fundamental dimension—feedstock origin, structural performance, and life cycle reversibility—that together determine the environmental and technological viability of polymeric materials. These pillars form the conceptual foundation of this review and provide a unified lens through which we analyze progress and challenges in sustainable polymer design.

Renewable feedstocks for renewability and scalability

Transitioning from fossil-based inputs to renewable carbon sources is a cornerstone of sustainable polymer design.³⁷ A wide range of renewable feedstocks has emerged to support this shift, including plant biomass, microbial products, and carbon dioxide (CO₂). These sources span varying levels of chemical complexity—from small molecules such as sugars, fatty acids, terpenes, and lignin-derived aromatics to macromolecules such as polysaccharides, proteins, chitin—and can be converted into monomers or polymer precursors via chemical, enzymatic, or hybrid processing routes (Figure 2).^{9,10,12,38}

Plant biomass remains the most widely exploited renewable source. Starch, sugars, and lipids derived from crops such as

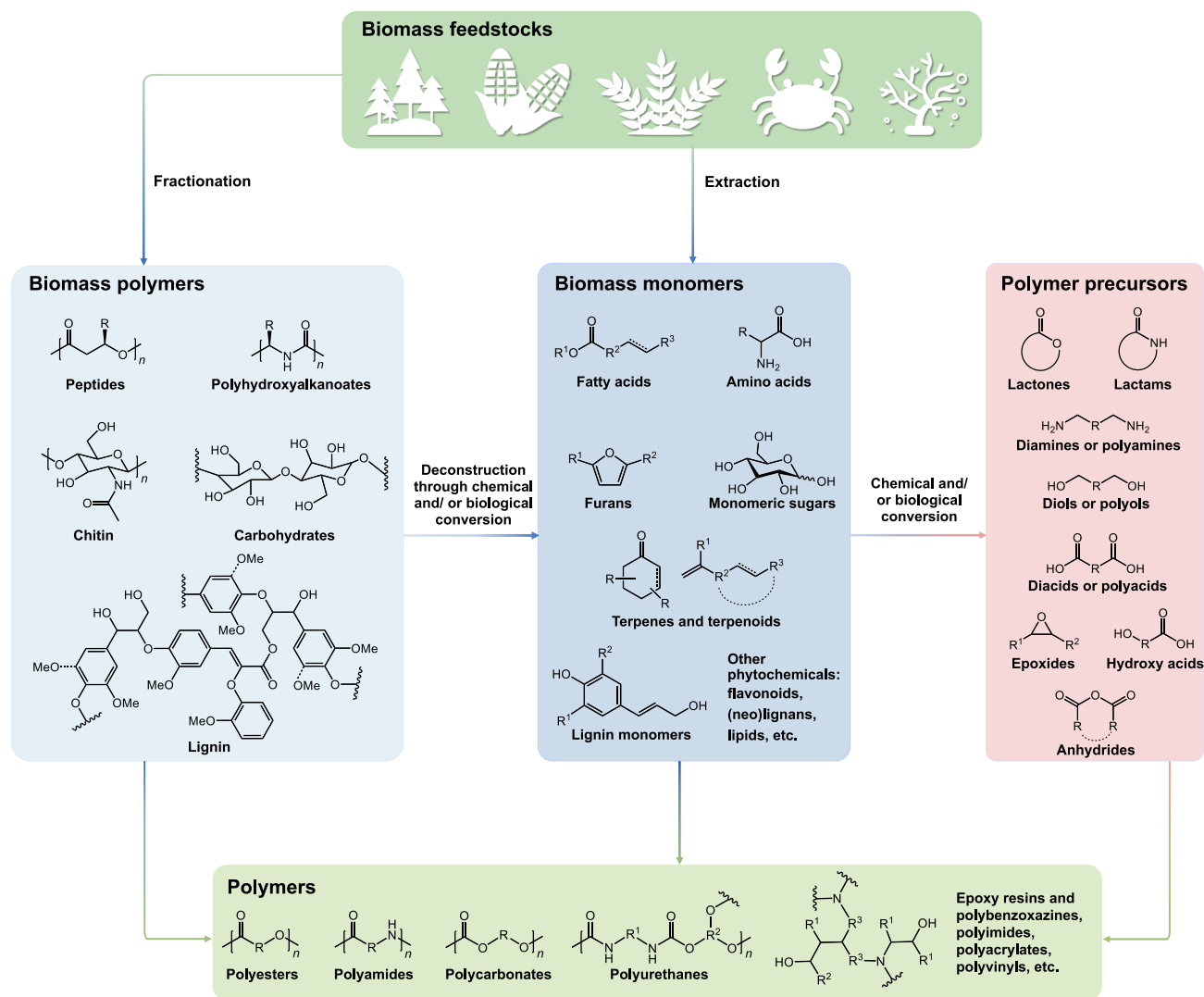


Figure 2. Representative polymers produced from renewable feedstocks

Biomass-derived polymers originate from a variety of renewable feedstocks that can be either directly incorporated into polymer structures or first broken down into valuable intermediates through chemical or biological transformations. In some cases, small molecules can be isolated directly from biomass; these bio-based compounds, if not inherently polymerizable, can serve as precursors after suitable chemical modification. The molecular structures depicted are representative examples and do not capture the full structural diversity found in nature. This diversity can be significantly expanded via synthetic modifications, and the actual repeating units in the resulting polymers may vary depending on the polymerization pathway employed.

corn and sugarcane have enabled scalable synthesis of polyesters, polyamides, and other polymer classes.²⁰ However, concerns over land use and food competition have prompted a transition toward lignocellulosic biomass, non-food feedstocks rich in cellulose, hemicellulose, and lignin.³⁹ While C5–C6 sugars from cellulose and hemicellulose are readily utilizable, lignin valorization remains challenging due to its structural complexity, often requiring oxidative or catalytic cleavage to access aromatic monomers.⁴⁰ In addition, specialized plant components—such as cutin, suberin, and phytochemicals, such as terpenoids—offer access to functionalized building blocks, including long-chain fatty acids and aromatics.⁴¹

A representative example of leveraging bio-based resources for high-performance polymer synthesis is the work by Mecking and co-workers, who developed a series of semi-crystalline

polyesters via step-growth polymerization of diacids and diols derived from plant oils containing 18 methylene units.^{11,24,42,43} These long-chain aliphatic building blocks yield polyesters with crystallinity and thermal properties comparable to those of polyethylene (PE), yet they remain inaccessible via conventional chain-growth polymerization routes.⁴⁴ This approach not only bridges the gap between renewable feedstocks and high-performance materials but also underscores the unique synthetic versatility offered by step-growth polymerization methods in constructing chemically recyclable PE-like materials from bio-derived monomers.

Beyond terrestrial biomass, marine and microbial sources present additional potential. Chitin, a nitrogen-rich polysaccharide found in crustaceans and fungi, offers opportunities for producing nitrogen-containing monomers.⁴⁵ Meanwhile,

microbial fermentation enables the direct biosynthesis of value-added monomers or polymers.^{46,47} Lactic acid, derived from carbohydrate fermentation, serves as a key building block for polylactic acid (PLA),^{9,48} whereas polyhydroxyalkanoates (PHAs) are directly accumulated as intracellular polyesters by various bacterial species. These microbial products are not only biodegradable but also tunable in mechanical and thermal performance.⁴⁹

Owing to its abundance, low cost, and inherent role in the global carbon cycle, CO₂ represents an attractive feedstock for sustainable polymer synthesis.⁵⁰ Owing to its thermodynamic inertness and fully oxidized state, CO₂ has proven challenging to directly incorporate into polymer transformations, with successful applications remaining relatively limited.⁵¹ However, CO₂ offers a fundamentally different and potentially carbon-negative feedstock route. Established strategies include the direct copolymerization of CO₂ with epoxides to produce aliphatic polycarbonates,^{52,53} as well as its conversion into C1–C6 platform molecules—such as methanol, formic acid, or succinic acid—via electrochemical, thermocatalytic, and biocatalytic routes, enabling further transformation into polymerizable monomers.^{54–58} A particularly promising example is 3-ethyl-6-vinyltetrahydro-2H-pyran-2-one (EVP), a six-membered lactone synthesized via palladium-catalyzed coupling of CO₂ and butadiene.^{59,60} EVP incorporates 28.9 wt % CO₂ and features multiple polymerizable moieties—including a vinyl group, an α,β -unsaturated ester, and a lactone ring—making it amenable to radical, coordination, and ring-opening polymerizations (ROPs).^{61,62} This structural versatility enables the design of CO₂-based polymers with tunable architectures and high carbon utilization efficiency.

Despite this broadening spectrum of renewable carbon sources, several key technical and economic challenges persist.³³ Many renewable feedstocks, particularly those derived from biomass, contain low concentrations of desired compounds and are embedded within complex mixtures, making selective extraction and purification inherently difficult.⁶³ As a result, biomass valorization often involves multistep or energy-intensive pretreatment processes, but achieving consistent monomer purity across heterogeneous sources remains a formidable challenge.^{64,65} CO₂-based pathways, while attractive from a carbon-utilization perspective, often suffer from low conversion efficiencies and yield polymers with undesirable material performance.^{58,59,66} Addressing these limitations will require not only continued innovation but also more targeted strategies across catalysis and process engineering. In catalysis design, challenges remain in achieving high activity and selectivity under mild, energy-efficient conditions, particularly for transforming low-reactivity biomass intermediates and CO₂ into polymerization-grade monomers. Rational catalyst development that couples multi-site cooperativity, redox control, and tunable acid-base balance could significantly improve conversion efficiency and monomer quality. In parallel, integrated reaction-separation process intensifications—such as reactive distillation, membrane-assisted catalysis, and continuous-flow bioreactors—offer promising routes to overcome equilibrium limitations and minimize downstream purification. However, scaling these systems while maintaining catalyst recyclability, process safety, and cost effectiveness remains a critical engineering challenge. Progress in these areas will be indispensable to enable sustain-

able, high-yield monomer production that supports both high-performance and circular polymer systems.

Built-in functions designed for functionality

Achieving true sustainability in polymer materials requires more than just renewable sourcing; it demands intrinsic functional design that reconciles high-performance properties with circularity principles.^{23,26} As sustainable materials transition from concept to real-world applications, the ability to meet, or even exceed, in some cases, the performance benchmarks of incumbent fossil-based plastics becomes essential. Recent efforts in molecular engineering have focused on constructing polymers that combine mechanical robustness, thermal and chemical stability, and controlled degradability or recyclability. These attributes can be achieved by tailoring monomer structure and macromolecular architecture using strategies such as incorporating rigid or flexible domains for tunable mechanical profiles^{67–70}; designing distinct (linear, branched, cyclic, or networked) topologies to modulate entanglement and processability^{70–73}; stereoregular backbone control to enhance crystallinity and mechanical strength^{14,70,74,75}; and including post-functionalizable sites to enable property tuning or orthogonal reactivity without compromising the material's EoL recyclability or degradability.^{69,76,77} Notably, many of these functionalities are accessible from bio-derived or platform monomers, underscoring the synthetic versatility of renewable resources in delivering performance-driven sustainable materials.^{38,46–49,78}

Several recent studies exemplify how precision molecular design can deliver both advanced properties and circularity. For instance, by controlling the stereochemistry of poly(3-hydroxybutyrate) (P3HB), Zhang et al. developed biodegradable adhesives with adhesion strengths surpassing cyanoacrylate glues while maintaining rapid degradability under environmental conditions (Figure 3A).¹⁴ Dreiling et al. introduced a modular thermoset system where a single monomer, 2,3-dihydrofuran (DHF), undergoes orthogonal polymerizations to form high-performance crosslinked networks that can be selectively deconstructed, thus addressing a key limitation in thermoset EoL management.⁷⁹ Pal et al. reported poly(α,ω -lipoic acid)-based adhesives with broad substrate compatibility and complete monomer recovery via dynamic disulfide exchange.¹⁵ Machado et al. demonstrated that bio-derived resins used in additive manufacturing can combine mechanical integrity with closed-loop recyclability, providing a sustainable path for 3D printing applications.⁸⁰ Leveraging a hybrid monomer design strategy,^{23,67} Shi et al. reported several advanced high-performance circular systems, including topology- and stereochemistry-controlled polythioesters that exhibit tacticity-independent crystallinity (Figure 3B)^{70,71}; architecturally complex polyesters capable of topology-accelerated and selective cascade depolymerization (Figure 3C)⁷³; and “one monomer-two polymers-one monomer” closed-loop systems enabled through orthogonal (de)polymerization of hybrid monomers, such as a lactone/olefin or lactone/epoxide hybrid (Figure 3D).^{69,77} Innovation in the pursuit of mono-material design focuses on the development of circular and biodegradable polymers derived from a single monomer, enabling the tuning of material properties through precise molecular and macromolecular engineering—such as controlling stereochemistry, topology, or

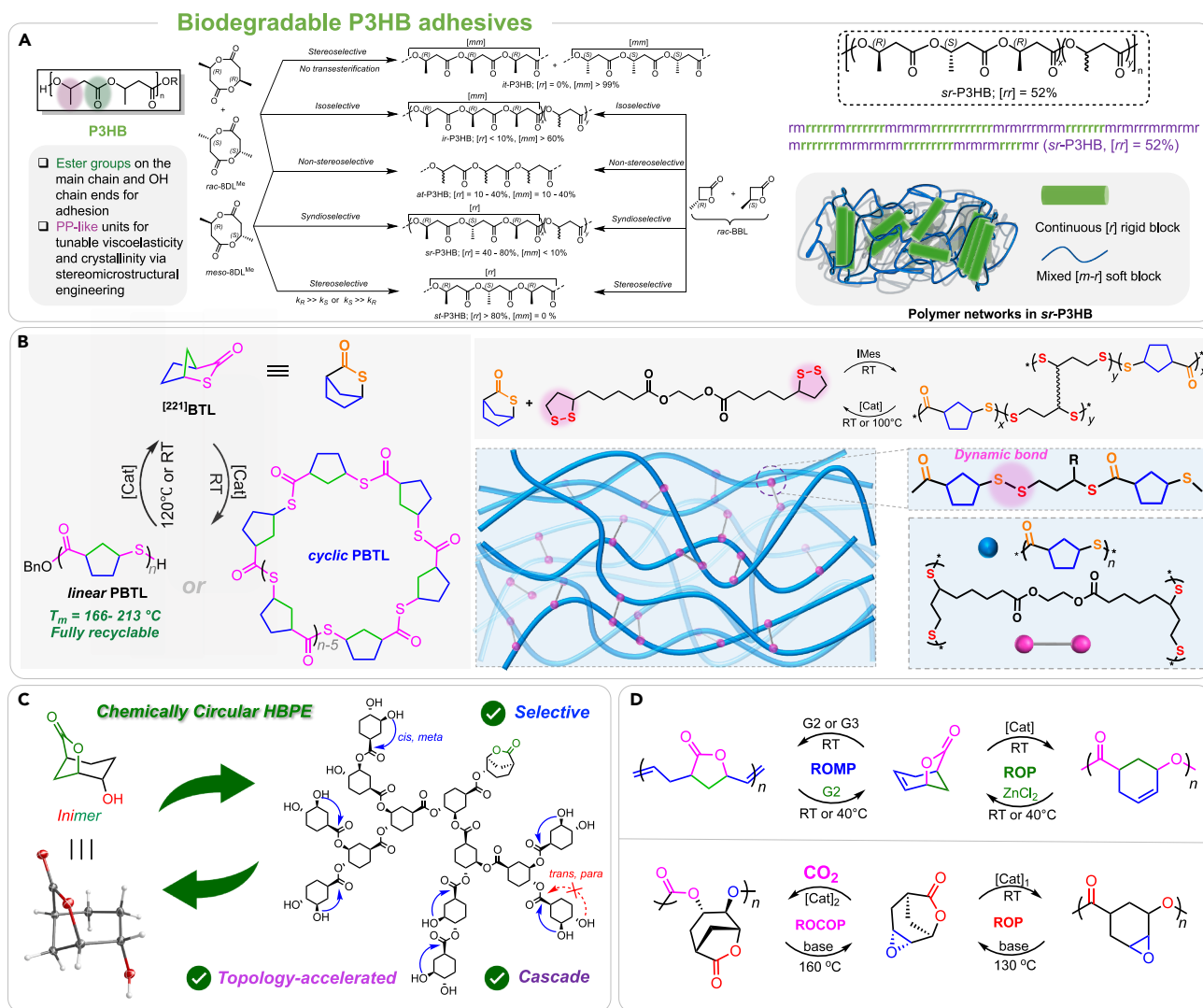


Figure 3. Selected recent examples of sustainable polymers with built-in functions

(A) P3HB-based high-performance biodegradable adhesives.

(B) Topology- and stereochemistry-controlled polythioesters exhibiting tacticity-independent crystallinity.

(C) Architecturally complex polyesters with topology-accelerated and selective cascade depolymerization.

(D) One monomer-two polymers-one monomer closed loop through orthogonal (de)polymerization of hybrid monomers. These examples illustrate how molecular and macromolecular engineering strategies render or enhance material properties, ensuring both high functional performance and circularity in sustainable polymers.

bifunctional monomers—without altering the chemical composition of the monomer.^{36,81} Such an approach offers a promising pathway to achieving functional versatility while maintaining closed-loop recyclability and material simplicity.

Collectively, these examples highlight how rational design to exert control over segmental flexibility, macromolecular topology, stereochemistry, and chemical functionality enables the simultaneous enhancement of both material performance and EoL options. Approaches such as backbone stereomicrostructural engineering, dynamic covalent crosslinking, or the modulation of thermal and chemical stability provide complementary pathways to develop polymers that combine high mechanical and thermal robustness with the capacity for degradation or chemical recycling under appropriate conditions.

To translate these advances into practice, a comprehensive evaluation of structure-property-function relationships is essential. Mechanical and thermal parameters, including modulus, tensile strength, elongation at break, toughness, glass transition temperature (T_g), melting transition temperature (T_m), and thermal/oxidative stability, should be benchmarked using standardized techniques. Standardized characterization techniques, such as tensile and impact testing, differential scanning calorimetry, thermogravimetric analysis, and dynamic mechanical analysis, provide foundational data to benchmark these sustainable polymers against incumbent materials: PE, polyethylene terephthalate, and poly(methyl methacrylate). Beyond intrinsic material performance, life cycle assessment (LCA) and techno-economic analysis (TEA) are also critical for quantifying

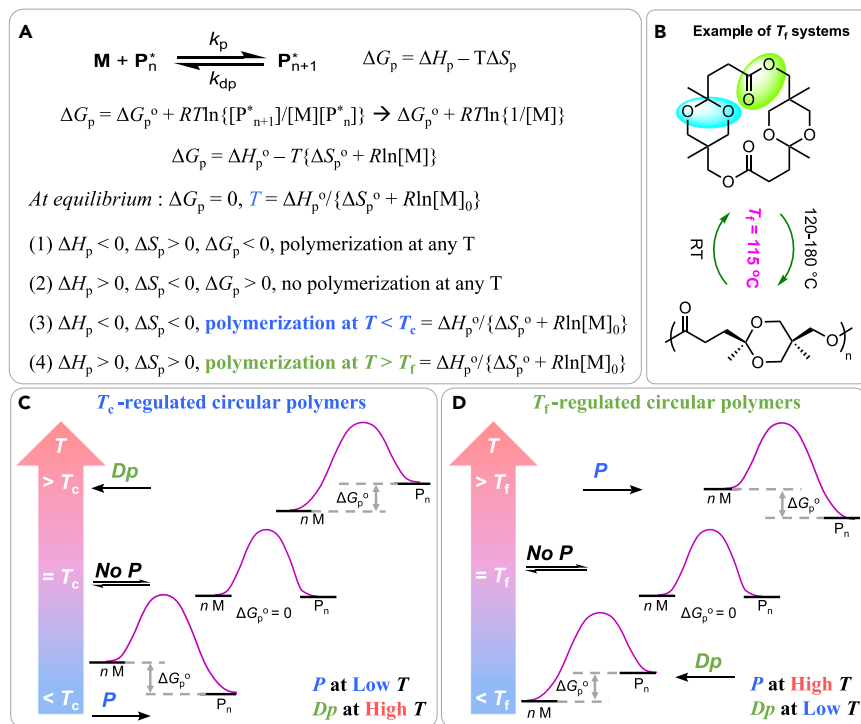


Figure 4. Circular pathways for polymer regeneration

(A) Thermodynamics analysis of polymerization (P)/depolymerization (Dp) equilibria.

(B) Representative example of T_f -regulated circular polymer systems.

(C) Energy profile illustrating the thermodynamic landscape of T_c -regulated circular polymers.

(D) Energy profile illustrating the thermodynamic landscape of T_f -regulated circular polymers.

tion requires $\Delta G_{\text{p}} < 0$. Based on the signs and magnitudes of the enthalpy (ΔH_{p}) and entropy (ΔS_{p}) of polymerization, four archetypal thermodynamic scenarios emerge (Figure 4A). When $\Delta H_{\text{p}} < 0$ and $\Delta S_{\text{p}} > 0$, polymerization is spontaneous across all temperatures. In contrast, systems with $\Delta H_{\text{p}} > 0$ and $\Delta S_{\text{p}} < 0$ are thermodynamically unfavorable for polymerization under any condition. For cases where both ΔH_{p} and ΔS_{p} are positive, polymerization becomes feasible only above a critical floor temperature (T_f), characterized as entropy-driven systems, commonly observed for macrolactones. More typically, ROP of small- to medium-sized rings involves scenarios where $\Delta H_{\text{p}} < 0$ (due to ring strain release) and $\Delta S_{\text{p}} < 0$ (due to loss of translational freedom), resulting in a defined ceiling temperature (T_c). Above T_c , entropy dominates and favors depolymerization ($\Delta G_{\text{p}} > 0$); below T_c , polymerization is thermodynamically favored. ΔG can be further expressed as $\Delta G_{\text{p}} = \Delta H_{\text{p}}^{\circ} - T(\Delta S_{\text{p}}^{\circ} + R \ln[\text{M}])$, linking it to standard enthalpy ($\Delta H_{\text{p}}^{\circ}$), entropy ($\Delta S_{\text{p}}^{\circ}$), temperature (T), and monomer concentration ($[\text{M}]$). This relationship implies that monomer-polymer equilibria can be tuned by adjusting the temperature and concentration to favor either polymerization or depolymerization. A low- T_c ($< 200^{\circ}\text{C}$) system is especially desirable for efficient chemical recycling to monomers. Intrinsically circular polymers (ICPs) exemplify materials engineered with low T_c values that enable closed-loop recycling with essentially quantitative selectivity for monomer reformation or recovery. While such ICPs are thermodynamically biased toward monomer formation, they remain stable under ambient or elevated conditions due to kinetic trapping. Efficient depolymerization can be triggered by employing a suitable catalyst that lowers the activation energy barrier of depolymerization.

Compared to T_c -based strategies, T_f -regulated closed-loop recycling systems are relatively underexplored. Elemental sulfur polymerization represents a prototypical example of a T_f -governed polymerization/depolymerization process. Specifically, at ambient temperature, elemental sulfur exists predominantly as S_8 rings. When heated above its T_f (159°C), S_8 undergoes ROP to form polymeric sulfur (S_n); upon cooling below T_f , S_n rapidly depolymerizes back to S_8 molecules.⁸⁴ This reversibility is enabled by the eight-membered ring structure, which allows the sulfur atoms to adopt a bond angle close to the ideal 108° , minimizing electron repulsion and system energy. This mechanism demonstrates the potential feasibility of T_f -governed

environmental impact and economic feasibility, enabling informed decisions for industrial adoption and policy alignment.^{82,83} However, most current studies remain fragmented—strategic control over stereochemistry, dynamic covalent bonding, and backbone architectures is not effectively coupled with scalable synthetic methodologies or comprehensive structure-property-function relationships that can guide molecular innovations toward practical materials combining high performance with circularity.²² Such disconnection continues to hinder the translation of high-performance polymer designs into practical, recyclable materials that can operate within a circular economy framework.

Circular pathways engineered for polymer regeneration

Realizing a sustainable polymer life cycle requires not only renewable sourcing and functional performance but also judicious molecular and/or macromolecular designs that enable efficient regeneration or degradation at their EoL. Recent advances have focused on two complementary pathways: closed-loop recycling and environmentally benign degradation. The former is rooted in precise control over the thermodynamics and kinetics of polymer/monomer (de)polymerization to facilitate selective depolymerization and monomer recovery.^{27,28,30} The latter relies on the incorporation of labile chemical linkages or bio-assimilable motifs into polymer backbones that facilitate degradation under composting or natural environmental conditions.^{19,31}

A key design principle in closed-loop chemical recycling is the rational engineering of polymer backbones to enable selective and efficient depolymerization under mild, accessible conditions.²⁶ Central to this approach is a comprehensive understanding of both the thermodynamics, governed by the Gibbs free energy change ($\Delta G_{\text{p}} = \Delta H_{\text{p}} - T\Delta S_{\text{p}}$), where favorable polymeriza-

closed-loop recycling systems (i.e., polymerization at elevated temperatures and monomer recovery at lower temperatures), but the rational design of such systems remains a significant challenge. Meng et al. recently reported a rare example of a T_f -governed closed-loop recyclable polymer system (Figure 4B).⁸⁵ In this study, a macrocyclic ketone ester was efficiently synthesized from inexpensive, biomass-derived resources and subjected to ROP under industrially accessible high-temperature conditions ($\sim 120^\circ\text{C}$ – 180°C). The resulting polyester exhibits enhanced thermodynamic stability relative to its monomer at elevated temperatures, delivering excellent thermal stability. Moreover, the polymer demonstrates strong tolerance to residual polymerization catalysts and can be processed thermally without degradation. Notably, under mild conditions (room temperature), the polymer undergoes rapid and efficient chemical depolymerization to regenerate the pristine macrocyclic monomer within 1 min upon the addition of a trace amount of depolymerization catalyst. This work not only introduces a highly efficient strategy for the synthesis of biomass-derived macrocyclic ketone esters but also establishes a chemically recyclable platform that combines high polymerization activity with controllable depolymerization capability. Importantly, this system addresses three long-standing challenges associated with T_c -regulated recyclable polymers: (1) polymerization can be carried out under industrially relevant high-temperature conditions, avoiding the high energy consumption and technical complexity of low-temperature processes; (2) the resulting polymers remain thermodynamically stable even in the presence of catalysts at elevated temperatures, enabling direct melt processing; and (3) efficient monomer recovery can be achieved under mild conditions with minimal energy input, simply by introducing an appropriate depolymerization catalyst. Though limited in number, subsequent studies have highlighted the distinct efficacy of T_f -regulated systems in facilitating closed-loop polymer recycling.^{86–88}

From a kinetic standpoint, the activation energy barrier, which is largely dependent on catalyst efficacy, dictates the rate and feasibility of polymerization or depolymerization at a practical timescale.⁸⁹ Efficient catalysts can significantly lower this barrier, enabling equilibrium to be reached under milder conditions and shorter times. Recent advances in catalytic depolymerization, coupled with concurrent monomer removal, have markedly improved the kinetic efficiency and selectivity of polymer closed-loop recycling, especially for systems with less favorable depolymerization thermodynamics. Catalysts such as Lewis and Brønsted acids or bases effectively lower the activation energy barriers, enabling depolymerization at reduced temperatures. When integrated with reduced-pressure operation, these systems allow for the continuous removal of monomer products—via distillation for volatile liquids or sublimation for solid species—thereby driving the equilibrium toward monomer formation.²⁶ This synergy between catalytic acceleration and dynamic product separation enables the depolymerization of polymers with relatively high T_c under milder conditions, expanding the applicability of closed-loop recycling strategies.

Highlighted here as an example is a ZnCl_2 /poly(ethylene glycol) catalyst system that enables efficient ring-closing depolymerization of high- T_c polyesters and polycarbonates under mild reactive distillation conditions (160°C), affording high mono-

mer yields and selectivity.⁹⁰ Building on this progress, Zhang and co-workers reported a solvent-free depolymerization strategy employing a simple magnesium salt (MgCl_2).⁹¹ This system demonstrated exceptional catalytic performance across eleven aliphatic polyesters and polycarbonates—including poly(ϵ -caprolactone) and poly(trimethylene carbonate)—achieving monomer yields of up to 98% at temperatures significantly below their T_c values. Furthermore, it exhibited excellent selectivity and broad compatibility with (co)polyesters and polyester-polycarbonate blends, even in the presence of commodity plastics, while maintaining catalytic activity over multiple recycling cycles.

However, in scenarios where chemical recycling is impractical—such as dispersed single-use plastics, agricultural films, or biomedical devices—biodegradable polymers serve as a critical complement.^{19,31} Beyond these specific applications, integrating biodegradability into polymers designed for chemical recyclability offers a vital form of environmental insurance. While chemical recycling is typically conducted under controlled conditions, unintended plastic leakage into natural environments is often unavoidable during use and disposal. Embedding inherent biodegradability ensures that such escaped materials can break down naturally, thereby mitigating long-term ecological impact. This dual-functionality paradigm demands more sophisticated molecular design, balancing structural stability for closed-loop recycling with susceptibility to microbial or enzymatic degradation when released into the environment. The design of such materials often incorporates labile linkages or selectively cleavable motifs that enable performance during service life while ensuring responsible degradation upon environmental exposure.

Together, chemically recyclable systems and environmentally responsive biodegradation form the comprehensive foundation of next-generation circular polymers that unites performance, sustainability, and adaptability across diverse EoL scenarios. This integrated framework, grounded in precise control over depolymerization thermodynamics and kinetics as well as the strategic incorporation of cleavable linkages, ensures that materials can be efficiently recovered in industrial settings or safely degraded in natural environments. Despite notable advances in monomer and catalyst design, system-level integration remains underdeveloped. Current research often excels in isolated domains—such as molecular innovation or recycling process optimization—but rarely bridges the gap between laboratory-scale materials and industrial implementation. The absence of standardized infrastructure for both chemical recycling and biodegradation further constrains scalability and real-world circularity. This persistent disconnect between molecular design and system-level deployment underscores the pressing need for cohesive strategies that integrate chemistry, engineering, and policy to achieve a truly circular polymer economy.

A UNIFIED FRAMEWORK ADVANCING POLYMER SUSTAINABILITY

To critically evaluate the current progress and limitations of sustainable polymers, we begin with a strategic SWOT analysis (Figure 5), which provides a holistic overview of both intrinsic material attributes and external systemic factors shaping their development and adoption. SWOT contrasts sustainable polymers with conventional petrochemical-based counterparts and

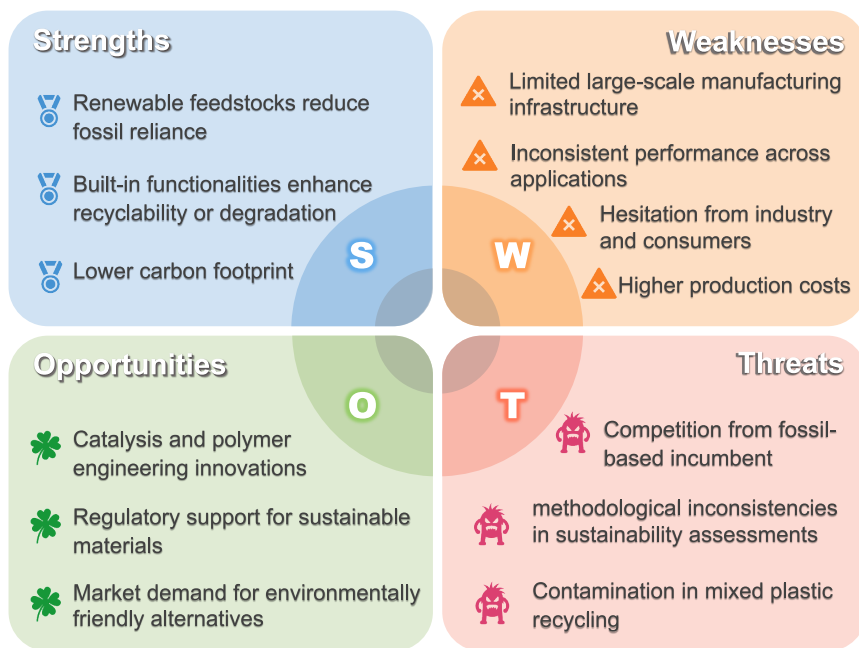


Figure 5. SWOT analysis

SWOT analysis of sustainable polymers compared to conventional plastics: a quadrant-based summary capturing the key internal (strengths and weaknesses) and external (opportunities and threats) factors influencing the development, scalability, and adoption of sustainable polymers.

This SWOT framework not only delineates the current landscape but also underscores the urgency and rationale for a unified molecular design framework that can balance renewability, functionality, and circularity in a coherent and scalable manner.

Building off of this strategic landscape, we propose a unified three-pillar framework to guide the design of next-generation sustainable polymers. This framework emphasizes (1) renewable resources to produce, (2) built-in functions to perform, and (3) circular paths to regenerate—serving as a molecularly and macromolec-

ularly grounded strategy to address the challenges and leverage the opportunities identified above. First, while renewable feedstocks diversify carbon sources and reduce dependence on fossil resources, key challenges persist in terms of scalability, processing compatibility, and economic viability. The effective transformation of biomass- or CO₂-derived intermediates into high-performance polymers requires not only advances in catalytic and biotechnological methodologies but also seamless integration into existing manufacturing infrastructures and supply chains. Without such systems-level alignment, the impact of renewable monomer platforms will remain limited to niche applications rather than enabling broad-based displacement of fossil-derived plastics. Second, recent advances have demonstrated that, to a certain extent, polymers derived from renewable resources can achieve thermal and mechanical performance comparable to that of conventional plastics.²² However, their applicability under the stringent requirements of sectors such as aerospace, electronics, and structural engineering remains largely underexplored. Unlike conventional fossil-based polymers, which have benefited from decades of industrial optimization and performance benchmarking, many bio-based polymers are relatively new and lack long-term validation under harsh operating conditions. A particular challenge lies in the chemical composition of many bio-based polymers. The frequent incorporation of heteroatoms (e.g., oxygen and nitrogen), while beneficial for reactivity or biodegradability, often compromises thermal stability, moisture resistance, and long-term durability compared to hydrocarbon-based analogs. Moving forward, greater emphasis should be placed on systematically evaluating the long-term performance and reliability of bio-based polymers under demanding environmental and operational conditions, to establish their viability in high-end applications beyond current niche uses. Third, the emergence of closed-loop chemical recycling and environmentally benign degradation has provided promising EoL strategies for sustainable polymers. Nonetheless, current recycling technologies face

helps contextualize recent advances within broader techno-economic and environmental frameworks. Specifically, key strengths include the use of renewable feedstocks that reduce reliance on fossil resources, lower life cycle carbon footprints, and the potential for built-in functionalities that enhance performance and enable recyclability or degradation. However, these advantages are countered by weaknesses such as higher production costs, limited industrial infrastructure, and inconsistent material performance across diverse applications. For example, although bio-based terephthalic acid and succinic acid have reached pilot-scale production, full commercial scaling remains constrained by feedstock supply and cost-parity issues.⁹² Moreover, there is hesitation from the industry to adopt functional replacements due to concerns over reliability, regulations, and compatibility with existing systems. Reviews on industry-academia collaborations in polymer research report that industry partners often hold back due to unclear intellectual property terms, mismatched timelines, and uncertain routes to commercialization.⁹³

Meanwhile, opportunities arise from innovations in catalysis, polymer engineering, and increasing regulatory and market pressure for more eco-friendly materials. Cross-disciplinary integration—linking catalysis, polymer science, and LCA—also provides a pathway to accelerate both molecular innovation and large-scale implementation. For example, the NSF's MFS-SPEED (Molecular Foundations for Sustainability: Sustainable Polymers Enabled by Emerging Data Analytics) initiative explicitly promotes multi-disciplinary teams uniting polymer chemists, data scientists, and industry partners to bridge discovery and deployment.⁹⁴ On the other hand, persistent threats remain, such as entrenched competition from petrochemical-based incumbents, variability in renewable feedstock availability across regions, methodological inconsistencies in sustainability assessments despite standardized metrics, and the technical complexity of recycling heterogeneous plastic waste streams.

ularly grounded strategy to address the challenges and leverage the opportunities identified above. First, while renewable feedstocks diversify carbon sources and reduce dependence on fossil resources, key challenges persist in terms of scalability, processing compatibility, and economic viability. The effective transformation of biomass- or CO₂-derived intermediates into high-performance polymers requires not only advances in catalytic and biotechnological methodologies but also seamless integration into existing manufacturing infrastructures and supply chains. Without such systems-level alignment, the impact of renewable monomer platforms will remain limited to niche applications rather than enabling broad-based displacement of fossil-derived plastics. Second, recent advances have demonstrated that, to a certain extent, polymers derived from renewable resources can achieve thermal and mechanical performance comparable to that of conventional plastics.²² However, their applicability under the stringent requirements of sectors such as aerospace, electronics, and structural engineering remains largely underexplored. Unlike conventional fossil-based polymers, which have benefited from decades of industrial optimization and performance benchmarking, many bio-based polymers are relatively new and lack long-term validation under harsh operating conditions. A particular challenge lies in the chemical composition of many bio-based polymers. The frequent incorporation of heteroatoms (e.g., oxygen and nitrogen), while beneficial for reactivity or biodegradability, often compromises thermal stability, moisture resistance, and long-term durability compared to hydrocarbon-based analogs. Moving forward, greater emphasis should be placed on systematically evaluating the long-term performance and reliability of bio-based polymers under demanding environmental and operational conditions, to establish their viability in high-end applications beyond current niche uses. Third, the emergence of closed-loop chemical recycling and environmentally benign degradation has provided promising EoL strategies for sustainable polymers. Nonetheless, current recycling technologies face

substantial hurdles, including limited scalability, high energy input, and contamination from mixed waste streams. Embedding recyclability at the molecular level—through rational monomer and backbone design—is a critical step, but its impact will remain constrained without concurrent advancements in recycling infrastructure, integrated processing, and standardized systems for material labeling and sorting. Addressing these system-level barriers is essential to realize the full benefits of molecular innovations in circular polymer design. Similarly, the deployment of environmentally benign degradation pathways—such as industrial composting or enzyme-mediated breakdown—remains limited by the lack of standardized degradation environments, slow degradation rates under real-world conditions, and concerns over microplastic residues or incomplete mineralization. Ensuring that biodegradable polymers fulfill their intended environmental function requires not only precise molecular engineering but also alignment with waste management systems and regulatory frameworks. Addressing these system-level barriers is essential to realize the full benefits of molecular innovations in circular polymer design.

Collectively, this unified framework propels the field of sustainable polymers along three critical dimensions: (1) establishing rigorous sustainability metrics that distinguish genuinely circular polymers from superficial “greenwashing” claims; (2) demonstrating how molecular design can reconcile high-performance requirements with environmental responsibility, thereby challenging the conventional notion of trade-offs; and (3) underscoring the necessity of industrial scalability and systems-level integration to ensure that molecular-level innovations align with real-world manufacturing, logistics, and recycling infrastructures. By addressing both fundamental scientific challenges and practical implementation barriers, this integrated strategy sets the stage for a transformative shift toward the next generation of sustainable polymers—materials that are not only environmentally sustainable but also technologically competitive and economically scalable, enabling meaningful impact across the evolving materials economy.

TOWARD SCALABLE AND CIRCULAR POLYMER SYSTEMS

The coming decade presents a critical opportunity to transition sustainable polymers from academic innovation into the marketplace. Realizing this vision will require coordinated, multi-tiered strategies that address challenges across molecular design, process integration, and infrastructure development. First, catalytic and biotechnological advancements are urgently needed to improve the efficiency, selectivity, and energy footprint of renewable monomer synthesis and polymer depolymerization. Second, the design of future materials must be guided by interdisciplinary collaboration that integrates polymer chemistry and physics, polymer science and engineering, chemical and biological catalysis and engineering, and EoL management. Embedding LCA principles into the earliest stages of polymer design is critical to ensure that sustainability targets are met without unintended trade-offs. In this regard, early-stage methods, such as hotspot analysis and simplified LCA strategies, offer practical tools for researchers who may lack access to expensive databases and software, enabling rapid identification of the most environmentally impactful stages in a material’s life cycle and

guiding molecular innovation toward systems-level sustainability.^{95,96} Third, scalable industrial implementation is essential for the success of sustainable polymers. Key priorities include the development of modular, energy-efficient recycling technologies, standardized material coding for streamlined separation, and the construction of regionally distributed closed-loop recycling infrastructures. Establishing clear economic incentives and lowering logistical barriers will be essential to facilitate widespread industrial adoption. Fourth, the establishment of harmonized, transparent sustainability standards will be critical. These standards should encompass resource origin, production energy, recyclability, biodegradability, and life cycle greenhouse gas emissions, providing stakeholders with objective criteria to evaluate and certify truly circular materials while curbing superficial or misleading sustainability claims. Achieving a future of truly circular, high-performance polymers demands a concerted global effort, integrating scientific discovery, technological development, and industrial collaboration. By aligning breakthroughs in materials science and engineering with regulatory policy and economic frameworks, the plastics economy can be reimaged into a system that is both functionally superior and environmentally sustainable, ensuring material security and ecosystem resilience for future generations.

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DECLARATION OF INTERESTS

The authors declare no competing interests.

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