

From Waste to Resource: Recent Trends in Plastic Recycling and Upcycling

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Abstract: Only an estimated 9% of all plastic ever manufactured has been recycled, and the remainder persists in landfills, oceans, and, increasingly, in the human body as micro- and nanoplastics. Closing this loop demands chemistry that goes beyond shredding and remelting. This review surveys the mechanistically distinct strategies now converging on the plastic waste problem: classical end-of-life treatments and their limits; selective depolymerization, catalytic bond-breaking, and direct polymer functionalization; photochemical and electrochemical upcycling, including established semiconductor and molecular photocatalytic systems, together with a prospective metal-free, defect-engineered carbon nitride (O-VN-CN) platform developed by our group for ambient-condition processing of mixed plastic waste; enzymatic and chemo-biological recycling, exemplified by industrial PET biorecycling; and, on the front end, the molecular design of plastics whose recyclability is programmed into the monomer itself. Ten reversible and cleavable bond chemistries are organized into five design principles—reversible dynamic bonds, cleavable weak links, bidirectional controlled bonds, self-immolative cascades, and weak-C-C strategies—including a shuttle-controlled, bidirectional disulfide metathesis platform that enables clean, catalyst-free recovery of high-performance thermosets such as disulfide-doped polydicyclopentadiene. Together, these advances mark a decisive shift from managing plastic waste after disposal to engineering circularity by design, and this review closes by outlining the catalytic, biological, and materials-design frontiers most likely to determine whether that shift can be achieved at industrial scale.

Keywords: plastic recycling; chemical upcycling; photocatalysis; enzymatic depolymerization; covalent adaptable networks; vitrimers; disulfide metathesis; circular polymers

1. Introduction

Plastics have become indispensable to modern civilization. From food packaging to medical devices, from aerospace composites to consumer electronics, polymeric materials underpin virtually every sector of the global economy, prized for their low cost, durability, light weight, and processability. These same properties, however, have made plastic waste one of the defining environmental challenges of our time. Global plastic production now exceeds 400 million tonnes annually, with projections suggesting this figure could double by 2050 if current trends continue, yet only an estimated 9% of all plastic ever produced has been recycled;¹ the remainder has been land-filled, incinerated, or released into the environment, where it persists for decades to centuries. Once dispersed, plastics

fragment into microplastics and nanoplastics now detected in oceans, soils, drinking water, food chains, and human blood, breast milk, and placentas, raising growing concern over ecological and health impacts that are still not fully understood. Environmentally, plastic debris suppresses marine biodiversity, entangles megafauna, and leaches toxic additives such as phthalates and bisphenols into food chains; economically, it is estimated to cost the global economy over USD 13 billion annually through environmental damage alone. Compounding these harms, most commodity plastics are derived from finite fossil feedstocks, so their linear take–make–dispose life cycle represents both a materials loss and a continuing source of greenhouse gas emissions, from extraction and polymerization through to end-of-life incineration.

Addressing this problem cannot rely on waste management alone; it requires solutions at both ends of the plastic life cycle. On the front end, there is a pressing need to design new generations of plastics—bio-based, chemically recyclable, or genuinely biodegradable polymers—that retain the performance of conventional materials while being engineered for facile end-of-life deconstruction. On the back end, and of equal urgency, existing plastic waste already in circulation must be dealt with: the challenge is no longer simply how to destroy or dispose of it, but how to convert it, efficiently and sustainably, into higher-value materials, chemicals, fuels, or energy carriers.

Conventional mechanical recycling—shredding, melting, and remolding—suffers from severe limitations. Repeated thermomechanical processing degrades polymer chain length, introduces contaminants, and yields materials of inferior quality relative to virgin plastics; many high-value polymers and multi-component composites are entirely incompatible with mechanical recycling altogether. Chemical and biological strategies therefore offer transformative pathways not merely to recycle—recovering monomers or fuels—but to upcycle, converting waste plastics into materials of higher value than

the original feedstock. This has catalyzed an explosion of research into technologies that can deconstruct or transform plastic waste under milder, cleaner, and more energy-efficient conditions than traditional approaches, ideally using renewable inputs such as light, electricity, or biological catalysis rather than fossil-derived heat and pressure. This Perspective focuses on modern chemical and molecular approaches to plastic recycling and upcycling, organizing them within a coherent framework spanning classical end-of-life treatment, molecularly precise upcycling, and emerging front-end design strategies in which recyclability is programmed into the polymer architecture from the outset. Accordingly, detailed techno-economic, life-cycle, and policy analyses are beyond the scope of this review, although they remain essential for evaluating the broader sustainability and scalability of these technologies.

2. Classification of emerging recycling and upcycling strategies

The primary back-end management strategies discussed across Sections 2.1 through 2.5 are mapped systematically in Figure 1.

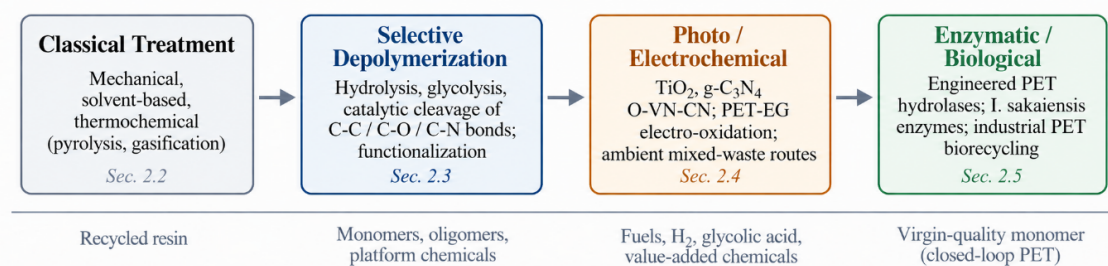


Figure 1. Back-end recycling and upcycling strategies organized by increasing molecular precision, from classical end-of-life treatment (Section 2.2) through selective depolymerization and catalytic upcycling (Section 2.3), photochemical/electrochemical upcycling including the metal-free O-VN-CN platform (Section 2.4), to enzymatic and chemo-biological recycling (Section 2.5).

2.1. End-of-life treatment

The accumulation of end-of-life plastics represents one of the defining environmental challenges of the modern era, driven by low recycling rates, material heterogeneity, and the loss of value each time conventional processing degrades polymer properties. This challenge spans an enormously diverse waste stream—from resin codes 1–7, multilayer and mixed plastics, to more specialized items such as gloves, wind turbine blades, and car tyres—each posing distinct barriers to recovery due to differences in composition, additives, and structural complexity. Addressing this requires strategies that not only manage plastic waste after disposal but increasingly aim to recover value—as materials, monomers, chemicals, or fuels—through molecularly precise transformations.

Traditional approaches such as mechanical, solvent-based, and thermochemical recycling primarily focus on recovering materials or chemical feedstocks after disposal, whereas modern strategies seek greater molecular control through the devel-

opment of new catalytic systems for selective bond breaking of plastics into fuels, high-value platform chemicals, oligomers, and monomers, as well as through selective depolymerization and catalytic upcycling. More recently, this concept has been extended to the deliberate design of monomers and polymers containing cleavable, dynamic, or self-immolative units, enabling materials to retain their required performance during use while undergoing controlled deconstruction at end of life. Together, these approaches reflect a transition from managing plastic waste after it is generated to engineering recyclability directly into the molecular architecture of plastics.

2.2. Established methods and their limits

End-of-life plastic management aims to recover material, chemical feedstocks, or energy from plastics after use. Classical approaches remain important because of their technological maturity and large-scale applicability, but each has significant limitations, particularly in terms of material quality, energy

demand, emissions, and environmental sustainability.

2.2.1. Mechanical recycling

Plastics are sorted, washed, shredded, melted, and reprocessed into new products. It is relatively simple, low-cost, and less energy-intensive than chemical or thermochemical methods. However, repeated processing can degrade polymer properties, and contamination or mixed-plastic streams reduce recyclability; poor-quality recycled products may also shorten material lifetimes and eventually contribute to further waste generation.²

2.2.2. Solvent-based recycling

Selected polymers are dissolved and separated from contaminants or other plastics before being recovered by precipitation. This can produce relatively high-purity polymers and preserve much of the original polymer structure. Its limitations include high solvent consumption, energy requirements for solvent recovery, potential solvent toxicity, and the environmental burden associated with solvent production, loss, and disposal.³

2.2.3. Thermochemical recycling

High temperatures are used to convert plastics into smaller molecules or energy-rich products. Pyrolysis converts plastics into oils, gases, waxes, and char and can process mixed or contaminated plastic streams; however, it is energy-intensive, often produces complex product mixtures, and may generate significant greenhouse-gas emissions if fossil-derived energy is used or the products are ultimately burned as fuels.⁴ Gasification converts plastics into syngas, mainly carbon monoxide and hydrogen, which can be used for fuels or chemical production. Although it can handle heterogeneous waste, it requires very high temperatures, substantial energy input, complex gas-cleaning systems, and can result in considerable carbon emissions.²

2.3. Selective depolymerization, catalytic upcycling, and polymer functionalization

The limitations of classical recycling methods have created a need for strategies that retain more of the chemical and economic value embedded in plastics while reducing energy consumption, emissions, and waste generation. Modern approaches therefore focus on molecular-level control—selective bond cleavage, catalytic depolymerization, chemical upcycling, and polymer functionalization—with the objective of recovering defined monomers, oligomers, platform chemicals, or other high-value products rather than generating low-value mixed streams.

Selective depolymerization uses hydrolysis, glycolysis, methanolysis, aminolysis, hydrogenolysis, and related reactions to break polymers into their original monomers or useful intermediates. These approaches can enable closed-loop

recycling and retain much of the chemical value embedded in the polymer; however, they are generally most effective for polymers containing readily cleavable functional groups and may require catalysts, solvents, elevated temperatures, and extensive product purification.⁵ Notable contributions in this area include the organocatalytic glycolysis, methanolysis, and aminolysis chemistry developed by Sardon, Dove, and co-workers, including their collaborative work with Hedrick on metal-free catalytic systems for polyester depolymerization, as well as the engineered PETase and related hydrolase chemistry advanced by McGeehan and co-workers and the industrial enzymatic PET depolymerization platform developed by Marty and co-workers at Carbios.

For more chemically resistant plastics, particularly polyethylene and polypropylene, catalytic bond-breaking and upcycling strategies have been developed to selectively cleave strong C–C, C–O, or C–N bonds through hydrogenolysis, oxidation, metathesis, aromatization, and related catalytic transformations. These processes can convert waste plastics into fuels, lubricants, waxes, aromatic compounds, platform chemicals, oligomers, and other value-added products with greater selectivity than conventional pyrolysis. Their major advantage is the potential to transform otherwise difficult-to-recycle plastics into useful chemical feedstocks; however, controlling the position and extent of bond cleavage remains challenging, and many catalytic systems rely on expensive metals, hydrogen, high temperatures, or energy-intensive reaction conditions. Leading contributions in this area include the Ru- and Pt-catalyzed hydrogenolysis chemistry developed by Beckham, Román-Leshkov, and co-workers,⁶ the tungstated-zirconia-supported ruthenium systems of Vlachos and co-workers,⁷ and the base-metal, ethylene-mediated deconstruction of polyethylene reported by Hartwig and co-workers.⁸

Recent work from Jiao and co-workers further illustrates how oxidative and nitrogenative chemistry can move polyolefin recycling toward selective, value-added products. Their aerobic catalytic treatment of mixed polyolefins operates under atmospheric pressure with O₂ or air and is compatible with HDPE, LDPE, PP, PS, and mixed streams, producing carboxylic-acid products under comparatively mild conditions.⁹ The group subsequently developed selective nitrogenative upcycling of polyolefins to high-value nitriles and amides, demonstrating that degradation can be coupled directly to product functionalization.¹⁰ Their recent perspective on directed degradation to functional oligomers further highlights the emerging goal of controlling chain length and end-group identity rather than simply maximizing conversion to small molecules.¹¹

Direct polymer functionalization provides an alternative to complete depolymerization by chemically modifying the existing polymer through oxidation, hydroxylation, amination, carboxylation, or grafting. Introducing new functional groups can improve polarity, adhesion, compatibility, and

other properties, allowing low-value waste polymers to be converted directly into higher-performance materials. Key advances in this space include the Kayser group's development of a chemical reaction converting polystyrene waste into the conducting polymer PEDOT:PSS for use in electronic devices such as solar cells and organic electrochemical transistors, as well as work by Ciccia and co-workers on direct catalytic functionalization of polyolefins, and the conversion of commercial HDPE into high-performance vitrimers through grafting of maleimides bearing dioxaborolane functionalities followed by cross-linking with a bis-dioxaborolane, yielding materials with markedly improved melt strength and dimensional stability relative to the parent HDPE. This approach preserves much of the original polymer backbone and therefore avoids the energy required for complete deconstruction and repolymerization; nevertheless, achieving uniform functionalization while avoiding uncontrolled chain degradation can be difficult, particularly for heterogeneous post-consumer plastics.¹²

2.4. Photochemical and electrochemical upcycling

Emerging photochemical and electrochemical approaches use light or renewable electricity to drive selective bond cleavage and functionalization under potentially milder conditions, offering the key advantages of ambient-temperature operation, direct compatibility with intermittent renewable power (solar and wind), and the ability to decouple plastic degradation from the fossil-fuel-derived heat that classical thermochemical methods require. Photocatalytic processes can generate reactive intermediates for polymer degradation or photoreforming, and a broad range of catalyst systems has been reported for this purpose, including semiconductor-based systems such as metal oxides, metal sulfides, and metal-organic framework (MOF) composites, as well as solution-based molecular systems that enable selective activation of C–C and C–H bonds to transform plastic waste into valuable fuels and chemicals. Widely studied platforms include TiO₂ (anatase, rutile, and brookite polymorphs) and other metal oxides such as BiVO₄, Fe₂O₃, ZnO, and Nb₂O₅, bismuth-based oxyhalides and vanadates, and graphitic carbon nitride (g-C₃N₄) and its heterojunction composites—with the Reisner group notably demonstrating carbon-nitride-based photoreforming of nonrecyclable plastic waste.¹³

Jiang and co-workers have developed a complementary sequence of low-energy and photochemical plastic-upcycling methods. Visible-light degradation was shown to convert several plastic wastes into commercial chemicals and recoverable monomers,¹⁴ while a subsequent low-energy strategy used swelling and decrystallization to facilitate depolymerization of polyester plastics, enabling diverse monomer recovery from both individual and blended polyester waste streams.¹⁵ More recently, copper-photocatalyzed dual desulfonylative chlorination enabled the ambient-condition upcycling of chemically ro-

bust polysulfones into value-added dichlorinated diaryl ethers, extending photocatalytic deconstruction to super-engineering plastics.¹⁶

Among recent developments in this area, our group has reported a metal-free, defect-engineered carbon nitride (O-VN-CN) photocatalyst for the upcycling of chemically diverse and mixed plastics under ambient conditions. The developed system enables the conversion of individual plastics, including PS, PP, and PVC, as well as ternary mixture of these plastics, into value-added chemicals under visible light irradiation. This work expands the scope of metal-free photocatalytic plastic upcycling and complements the broader range of metal-oxide, molecular, and hybrid photocatalytic systems described above.¹⁷

Electrochemical oxidation and reduction can similarly transform plastic-derived molecules into useful chemicals, sometimes alongside hydrogen production, harnessing renewable electricity to drive selective C–C bond cleavage under ambient conditions rather than relying on high-temperature or high-pressure processing. This field has centered largely on the electro-oxidation of PET-derived ethylene glycol (EG): early work demonstrated the electrocatalytic upcycling of PET to valuable commodity chemicals such as potassium diformate and terephthalic acid alongside H₂ fuel, with EG selectively electrooxidized to formate at high current density using a CoNi_{0.25}P catalyst. Building on this, subsequent studies reported electrocatalytic upcycling of PET-derived EG over an Au/Ni(OH)₂ catalyst into glycolic acid, a key monomer for the biodegradable polymer polyglycolic acid, alongside H₂ and terephthalic acid production, while Chen and co-workers achieved similar EG-to-glycolic-acid conversion using a Pd/Ni(OH)₂ catalyst, and Wang and co-workers integrated PET electro-upcycling with electrochemical nitrate wastewater reformation to co-produce glycolic acid and ammonia at both electrodes. Other groups, such as Zhang and co-workers, have used PdFe electrocatalysts loaded on nitrogen-doped carbon nanotubes for the electrocatalytic upcycling of PET waste, achieving high Faradaic efficiency toward formic acid, illustrating the broader push toward doped and bimetallic catalyst design in this area. These approaches offer the prospect of reducing dependence on high-temperature processing and integrating recycling with renewable energy; however, low reaction rates, limited penetration into solid polymers, catalyst stability, energy efficiency, and scale-up currently remain important challenges.

Despite these advances, most electrochemical upcycling studies currently rely on relatively well-defined, soluble intermediates such as PET-derived ethylene glycol rather than untreated mixed plastic waste. Scale-up will therefore require maintaining high current density and selectivity under industrially relevant conditions while overcoming mass-transfer limitations, catalyst deactivation, feedstock heterogeneity, and the

need for effective pretreatment or depolymerization of solid waste streams.

2.5. Enzymatic and chemo-biological recycling

In nature, certain bacteria and microorganisms have evolved the ability to slowly break down plastic polymers, using secreted enzymes to attack ester, amide, or other hydrolyzable linkages as a carbon source—a process of natural biodegradation that, while typically far too slow to address the scale of plastic waste generated today, has provided the biological blueprint for engineered recycling technologies. Enzymatic and chemo-biological recycling builds directly on this biology, offering a complementary strategy based on the high molecular selectivity of biological catalysts. This field was catalyzed by the discovery of the PET-degrading bacterium *Ideonella sakaiensis* by Yoshida and co-workers,¹⁸ and has since been advanced through the engineered, industrially productive PET hydrolase reported by Tournier, Marty, and co-workers,¹⁹ which enables enzymes to depolymerize susceptible plastics such as PET into well-defined monomers under relatively mild aqueous conditions. Beyond depolymerization, microorganisms can subsequently convert these plastic-derived molecules into platform chemicals, biopolymers, and other valuable products, while combining chemical pretreatment with enzymatic or microbial conversion can further extend these approaches to more resistant, crystalline plastics.

The key advantages of this approach are its exceptional substrate selectivity, which avoids the side reactions and mixed product streams typical of harsh chemical or thermal processes; its mild, aqueous operating conditions, which lower energy input and carbon footprint relative to classical recycling; and its ability to fully depolymerize even complex, coloured, or contaminated PET waste into monomers pure enough to be repolymerized into virgin-quality material. This potential has already been realized at commercial scale: Carbios, founded by Truffle Capital, has commissioned the world's first PET biorecycling plant in Longlaville, France, using its enzymatic depolymerization technology—built on the engineered PET hydrolase from Tournier, Marty, and co-workers—to process PET waste back into virgin-quality monomers at industrial capacity, in partnership with companies including Michelin, PepsiCo, L'Oréal, and Novozymes. This demonstrates that enzymatic recycling can achieve the selectivity and product purity needed for closed-loop, food-grade PET regeneration at a scale that classical methods struggle to match.

Nevertheless, important limitations remain: reaction rates are generally slow, substrate scope is restricted largely to hydrolyzable polymers such as PET, highly crystalline or hydrophobic plastics remain difficult to access, and substantial improvements in enzyme stability, cost, and large-scale processing are still needed before these methods can be extended broadly across the plastics waste stream.

3. Front-end strategies: Designing plastics for circularity from the outset

Conventional end-of-life recycling begins only after a plastic has become waste. Molecularly programmed recyclability takes the opposite approach: it introduces chemically addressable linkages or reaction pathways into the material at the design stage, so that durability during use can be decoupled from controlled deconstruction at end of life. Self-immolative motifs can translate a single trigger into a depolymerization cascade, whereas dynamic or reversible covalent bonds permit exchange, network rearrangement, or, in favorable cases, recovery of defined molecular building blocks under a specific stimulus.

The central design problem is therefore not simply to weaken a polymer, but to create an orthogonal reactivity window: the material must remain stable toward heat, moisture, oxygen, solvents, and mechanical stress during manufacture and service, yet respond rapidly and selectively to a recycling condition that it would not normally encounter. This requirement motivates the mechanistic classification below and the search for bond-forming reactions whose forward and reverse pathways can be controlled with comparable fidelity.

A particularly influential front-end design framework has been established by Eugene Y.-X. Chen and co-workers, who have demonstrated that monomer thermodynamics and catalyst control can be engineered to combine practical material performance with selective chemical recycling to monomer. A 2018 Science study established a synthetic polymer system capable of repeatable chemical recyclability,²⁰ and later work produced chemically circular polyhydroxyalkanoates that are simultaneously melt-processable and mechanically tough.²¹ The same group has also demonstrated orthogonal polymerization/depolymerization of a lactone/olefin hybrid, illustrating how a single monomer platform can be directed into structurally distinct polymers and subsequently returned to the common monomer.²² Together, these studies exemplify the bidirectionally controlled polymerization equilibria highlighted below.

3.1. Overall classification and design principles

Within this framework, polymer deconstruction and rearrangement can be organized into five principal classes according to how reactivity is encoded into the material and how that reactivity is activated at end of life (Figure 2).

- **Reversible dynamic bonds**—including ester,²³ imine,²⁴ acylhydrazone,²⁵ Diels–Alder,²⁶ vinylogous urethane,²⁷ boronic ester,^{28,29} and related exchangeable linkages^{30,31}—undergo reversible cleavage or exchange under appropriate stimuli. Consequently, these dynamic motifs primarily enable material repair, reshaping, reprocessing, welding, and network recycling.^{32–34}

- **Cleavable weak links**—including ester, thioester, carbonate, acetal, disulfide, silyl ether, and other selectively labile bonds deliberately incorporated into otherwise robust polymer backbones. Rather than continuously exchanging during use, these linkages provide predetermined points of cleavage that enable controlled fragmentation into oligomers, functional molecules, or other recoverable products at end of life.^{35–38}
- **Bidirectional controlled bonds**—bonds or polymerization equilibria whose direction can be deliberately switched between bond formation and bond cleavage by changing temperature, catalyst, concentration, redox state, pH, or chemical environment. Because the same underlying chemistry can support both polymer construction and deconstruction, these systems provide a particularly attractive basis for chemical recycling and, where possible, recovery of the original monomers.^{39–42}
- **Self-immolative or cascade-cleavage units**—molecular architectures in which activation of a single trigger initiates a sequence of bond-cleavage events along the polymer chain. By converting one molecular stimulus into multiple subsequent reactions, these systems enable amplified, programmed, and potentially rapid depolymerization under relatively mild conditions.^{43,44}
- **Weak-C–C and cleavable-comonomer strategies**—emerging approaches designed to introduce predetermined points of deconstruction into otherwise highly persistent, often predominantly all-carbon polymer backbones. Their particular significance lies in the possibility of imparting controlled degradability to high-performance or commodity-like polymers while retaining the mechanical strength, crystallinity, chemical resistance, and processability associated with conventional hydrocarbon materials.^{45,46}

These five classes are not completely mutually exclusive. A single bond motif may perform more than one function depending on its molecular environment and the stimulus applied. Disulfides, for example, can behave as reversible dynamic bonds during network reprocessing, as selectively cleavable weak links under reducing conditions, or as bidirectionally controlled bonds when their exchange is driven selectively through an appropriately designed reaction pathway. The classification should therefore be viewed as mechanistic rather than purely structural: what distinguishes the classes is not simply the identity of the bond, but how that bond is used to control polymer construction, rearrangement, fragmentation, or depolymerization.

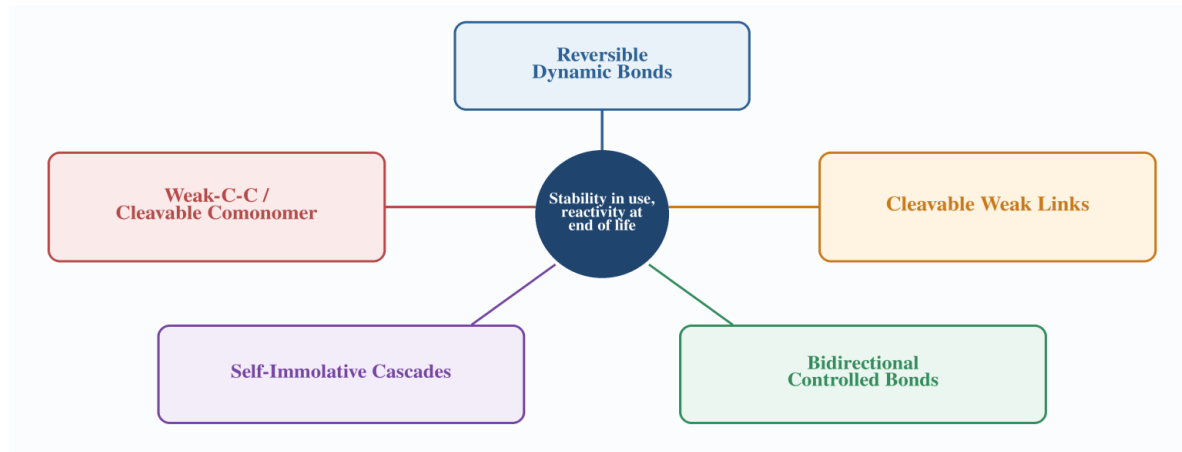


Figure 2. The five mechanistic design classes organizing molecularly programmed recyclability (Section 3.1), each reconciling stability during use with reactivity at end of life.

The overarching objective is consequently not to make a polymer intrinsically weaker. An effective recyclable-by-design material must instead reconcile two apparently competing requirements: stability during use and reactivity at end of life. Ideally, the programmed linkage should remain robust toward heat, moisture, oxygen, solvents, and mechanical stress under manufacturing and service conditions, yet become rapidly and selectively cleavable or exchangeable when exposed to a recycling stimulus that the material would not normally encounter. In this sense, the most successful systems decouple durability from degradability rather than treating

them as opposing properties. This principle is evident across very different recycling technologies, from the engineered PET hydrolases developed by Tournier and Marty to the shuttle-controlled disulfide metathesis developed by Li and Loh: in each case, high in-service stability is preserved while a specific molecular pathway provides selective reactivity when recycling is required.

Achieving this balance requires advances in reaction design on two complementary fronts. First, reversible bond-forming reactions must possess sufficiently tunable thermodynamics and kinetics to allow the material to remain kinetically stable

during use while undergoing rapid exchange or cleavage under defined recycling conditions. This requires moving beyond established disulfide, imine, boronate ester, Diels–Alder, and transesterification chemistry toward a broader range of reactions whose behavior can be adjusted across both associative and dissociative covalent-adaptable-network (CAN) regimes. Second, trigger-responsive and self-immolative motifs must be designed so that their activation threshold and subsequent cascade efficiency can be controlled independently of the bulk mechanical properties of the polymer. Together, these developments are necessary to move molecularly programmed recyclability from isolated proof-of-concept systems toward a

general monomer-level strategy for designing next-generation circular materials.

Tables 1 and 2 provide complementary molecular-level views of the five design classes summarized in Figure 2. Table 1 focuses on reversible and bidirectionally controlled chemistries, in which polymer topology or the polymerization equilibrium can be reorganized without necessarily destroying molecular value. Table 2 focuses on programmed deconstruction, spanning discrete weak links, self-immolative cascades, and emerging weak–C–C or cleavable-comonomer strategies that introduce predetermined fragmentation pathways into otherwise persistent polymer architectures.

Table 1. Reversible dynamic bonds and bidirectionally controlled chemistries used to rearrange, reprocess, or chemically recycle polymers. Dynamic covalent bonds primarily enable topology rearrangement, whereas bidirectionally controlled systems allow reaction conditions to switch deliberately between polymer construction and deconstruction. Reversible ring-opening polymerization represents the limiting case in which the equilibrium can be driven back to the original cyclic monomer.

Design class	Representative chemistry	Control/Trigger	Representative polymer systems/process	Recycling outcome/key function
Reversible dynamic bonds	Ester/transesterification	Heat; alcohol; Lewis acid/base catalyst	Polyester vitrimers; epoxy–ester networks	Stress relaxation, welding, reshaping, thermal reprocessing
Reversible dynamic bonds	Imine/Schiff base	pH; water; heat; amine exchange	Polyimines; epoxy and PU-like networks	Self-healing, reshaping, chemical recyclability
Reversible dynamic bonds	Acyldiazide	Acid/base; heat; hydrazine exchange	Hydrogels; elastomers; adaptive thermosets	Self-healing and adaptive network rearrangement
Reversible dynamic bonds	Disulfide	Heat; light; redox; thiol exchange	PU elastomers; epoxy networks; polyureas	Repair, reprocessing, redox-triggered deconstruction
Reversible dynamic bonds	Boronic ester/boroxine	Diol exchange; water; pH; heat	Dynamic elastomers; hydrogels; vitrimers	Rapid self-healing and reprocessing
Reversible dynamic bonds	Diels–Alder adduct	Heat	Furan–maleimide thermosets and networks	Thermally reversible crosslinking, repair, remolding
Reversible dynamic bonds	Vinyllogous urethane/enamine	Heat; transamination	Epoxy-, acrylate-, PEG- and bio-based vitrimers	Associative exchange and reprocessing
Reversible dynamic bonds	Siloxane/silyl ether	Heat; acid/base; fluoride (motif-dependent)	Silicone elastomers; dynamic siloxane networks	Self-healing, remolding, network rearrangement
Bidirectional controlled bonds	Diels–Alder $\rightleftharpoons$ retro-Diels–Alder	Temperature	Reversible thermosets	Directionally controlled network formation/cleavage
Bidirectional controlled bonds	Imine formation $\rightleftharpoons$ hydrolysis/transimination	pH; water; amine concentration	Polyimines	Construction, rearrangement, or deconstruction
Bidirectional controlled bonds	Disulfide formation $\rightleftharpoons$ exchange/cleavage	Redox state; shuttle; environment	Disulfide-linked polymers and networks	Controlled assembly, reprocessing, chemical recovery
Bidirectional controlled bonds	Olefin metathesis	Catalyst; olefin environment	Unsaturated polymer networks	Backbone reconfiguration or catalytic fragmentation
Bidirectional controlled bonds	Ring-opening polymerization $\rightleftharpoons$ depolymerization	Temperature; catalyst; concentration	Lactone, lactide, cyclic carbonate, cyclic acetal, selected ROMP polymers	Closed-loop monomer recovery and repolymerization

Table 2. Programmed cleavage motifs for controlled deconstruction of recyclable-by-design polymers. Cleavable weak links provide predetermined sites for fragmentation, self-immolative architectures amplify a single triggering event into a depolymerization cascade, and weak–C–C or low-level cleavable-comonomer strategies seek to impart recyclability to otherwise persistent carbon-rich polymers while retaining commodity-like properties.

Design class	Representative motif/strategy	Typical trigger	Representative polymer architecture	Principal deconstruction outcome
Cleavable weak links	Ester	Hydrolysis; chemical cleavage	Polyethylene-like copolymers; vinyl polymers; acrylates	Site-specific backbone cleavage
Cleavable weak links	Thioester/thionolactone-derived units	Nucleophile; base; chemical treatment	Acrylates, acrylamides, styrenics	Controlled fragmentation of otherwise persistent vinyl polymers
Cleavable weak links	Carbonate	Hydrolysis/chemical trigger	Polycarbonates; carbonate-containing copolymers	Backbone cleavage/depolymerization
Cleavable weak links	Acetal/ketal	Acid	Polyacetals; vinyl copolymers; degradable networks	Stable-in-use materials with on-demand cleavage
Cleavable weak links	Disulfide/1,2-dithiolane	Reduction; light; thiol exchange	Radical polymers; acrylates; vinyl polymers	Triggered fragmentation under mild redox conditions
Cleavable weak links	Silyl ether	Fluoride; acid/base; chemical trigger	Vinyl polymers; ROMP polymers; thermoset networks	Selective Si–O–C cleavage
Cleavable weak links	Photocleavable linker (e.g., o-nitrobenzyl)	Light	Crosslinked polymers and functional networks	Spatially and temporally controlled cleavage

Table 2. Cont.

Design class	Representative motif/strategy	Typical trigger	Representative polymer architecture	Principal deconstruction outcome
Self-immolative/cascade units	Polybenzyl carbamate/carbonate	End-cap cleavage; acid/base; light; redox	Self-immolative linear polymers	Single trigger initiates amplified end-to-end depolymerization
Self-immolative/cascade units	Polyacetal/poly(phthalaldehyde)-type architectures	Acid; light; chemical trigger	Depolymerizable polymers	Cascade cleavage to defined small-molecule products
Self-immolative/cascade units	Quinone-methide/1,4- or 1,6-elimination motifs	Chemical or photochemical trigger	Self-immolative oligomers and polymers	Sequential elimination cascade/programmed deconstruction
Weak-C-C/cleavable-comonomer strategies	Intrinsically weakened C-C bonds	Heat/radical generation	Polybutadiene-derived or polymuconate-type carbon-rich backbones	Selective backbone cleavage while retaining carbon-rich character
Weak-C-C/cleavable-comonomer strategies	Low-level cleavable comonomer	Linkage-specific chemical trigger	Polyolefin-like backbones with sparse ester, thioester, carbonate, disulfide, silyl ether or acetal units	Commodity-like properties plus controlled fragmentation
Weak-C-C/cleavable-comonomer strategies	Cleavable comonomers in thermosets	Chemical/redox/catalytic trigger	Crosslinked high-performance networks	Network fragmentation at sparse programmed sites

Taken together, Tables 1 and 2 establish a continuum of molecular control that mirrors Figure 2: reversible dynamic bonds and bidirectionally controlled chemistries enable network rearrangement or closed-loop polymer–monomer equilibria, whereas programmed cleavage strategies introduce predetermined weak links, self-immolative cascades, or selectively weakened backbone sites for controlled deconstruction. Some motifs can span more than one mechanistic class depending

on molecular context and trigger; disulfides, for example, may function as dynamic exchange bonds, selectively cleavable weak links, or bidirectionally controlled bonds.

3.2. Ten reaction platforms for molecularly programmed recyclability

To contextualize the subsequent discussion, Figure 3 outlines the classification of these ten representative chemistry platforms.

SECTION	REACTION PLATFORM	DESIGN CLASS
Sec. 3.2.1	DA / retro-DA	<i>Reversible dynamic</i>
Sec. 3.2.2	Transesterification vitrimers	<i>Reversible dynamic</i>
Sec. 3.2.3	Disulfide exchange / metathesis	<i>Bidirectional control</i>
Sec. 3.2.4	Boronic ester exchange	<i>Reversible dynamic</i>
Sec. 3.2.5	Imine / diketoenamine	<i>Bidirectional control</i>
Sec. 3.2.6	Olefin metathesis	<i>Weak-C-C</i>
Sec. 3.2.7	Transcarbamoylation	<i>Reversible dynamic</i>
Sec. 3.2.8	Siloxane equilibration	<i>Reversible dynamic</i>
Sec. 3.2.9	Alkoxyamine / nitroxide exchange	<i>Self-immolative</i>
Sec. 3.2.10	AFCT chemistry	<i>Self-immolative</i>

Figure 3. The ten reaction platforms of Section 3.2, color-coded by the mechanistic design class each exemplifies.

3.2.1. Diels–Alder/Retro-Diels–Alder (DA/rDA) chemistry

The earliest and still most widely cited example of a thermally reversible bond deliberately engineered into a structural polymer is the furan–maleimide Diels–Alder adduct. Wudl and co-workers, in a landmark 2002 Science paper, copolymerized a tetrafunctional furan monomer with tris(maleimide) to build a highly crosslinked network held together entirely by DA adducts (Figure 4).²⁶

The resulting material was a tough solid at room temperature with mechanical properties comparable to commercial epoxy resins, and above roughly 120 °C, a substantial fraction of the crosslinks underwent retro-Diels–Alder scission, allowing the network to be repeatedly re-mended by simple thermal treatment. The strategy has since been extended to furan-

functionalized biobased polymers, foams, and fiber-reinforced composites, all sharing the same design logic: heat drives retro-DA ring-opening and network loosening, and cooling reforms the DA adduct and restores mechanical integrity.

DA/rDA networks combine epoxy-like stiffness and toughness at service temperature with genuine thermal remendability and reprocessability, a combination that conventional thermosets, permanently locked by irreversible crosslinks, cannot offer. Because the rDA reaction proceeds without any added catalyst, damaged parts can be healed in place by localized heating, and whole components can be reshaped by hot pressing; in fiber-reinforced composites, driving the equilibrium fully toward retro-DA allows the polymer matrix to be de-crosslinked so that the often more valuable reinforcing fibers can be recovered largely undamaged. Its main limitations are that the DA/rDA equilibrium competes with

irreversible side reactions at elevated temperature—especially maleimide homopolymerization—which progressively reduce recyclability over repeated cycles; the rDA temperature must

also remain well above the intended service temperature, and furan/maleimide monomers are generally more synthetically elaborate than commodity thermoset feedstocks.

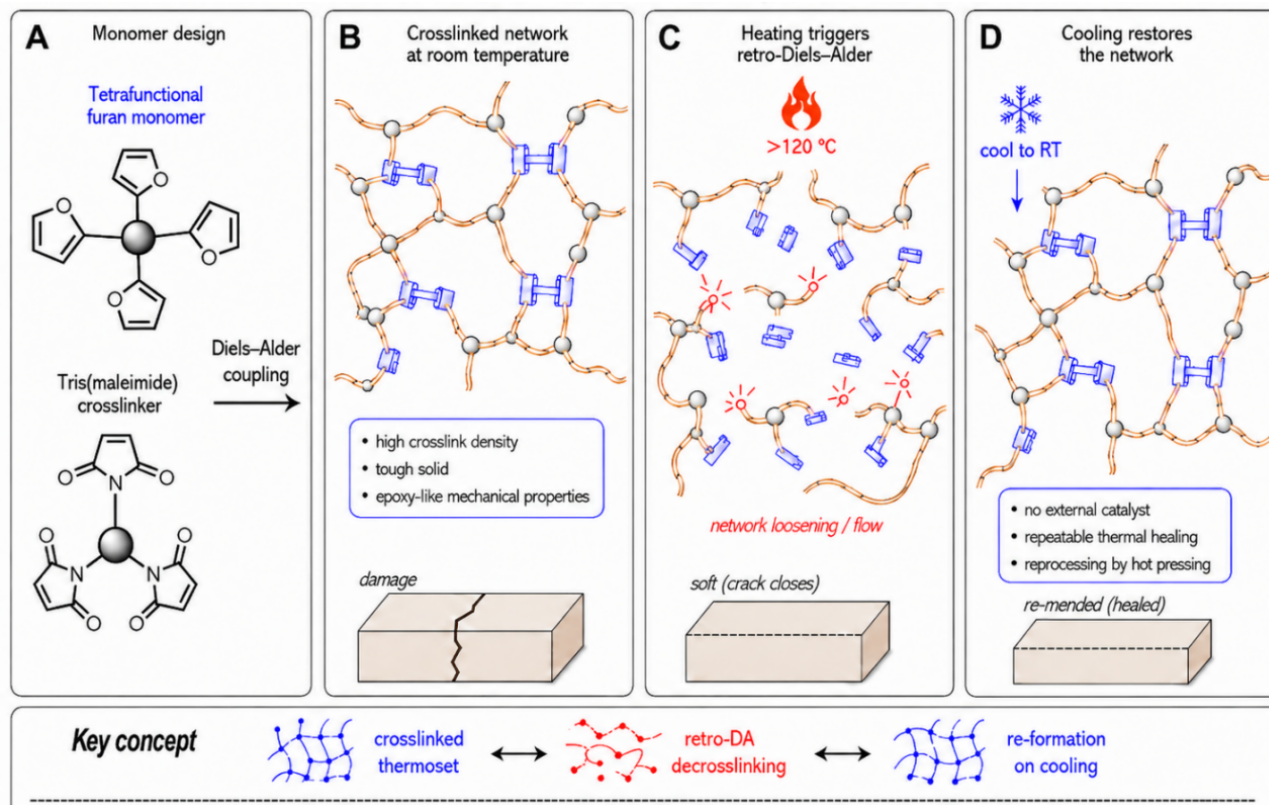


Figure 4. Thermally reversible Diels–Alder network chemistry (Wudl et al., Science 2002): furan–maleimide crosslinking gives a tough network at service temperature that undergoes retro-DA network loosening above $\sim 120\text{ }^{\circ}\text{C}$ and re-forms on cooling, enabling repeated thermal healing and reprocessing.²⁶

3.2.2. Transesterification-based vitrimers

In 2011, Leibler and co-workers (Montarnal, Capelot, Tournilhac, and Leibler) introduced the vitrimer concept in Science, showing that a conventional epoxy network—built from a diglycidyl ether of bisphenol A cured with a mixture of di- and tricarboxylic acids—could be made to flow reversibly if a small amount of zinc acetate catalyst was included (Figure 5).²³

In the temperature range where the transesterification reaction takes place, the crosslinked epoxy network is able to interchange fragments of its structure through production and recombination of glycol and diester units, without the total number of crosslinks ever changing. This associative exchange mechanism is what gives vitrimers their defining behavior: they are solid, insoluble, and creep-resistant like a conventional thermoset below a ‘topology-freezing transition’ (T_v), yet flow like a viscous liquid above it, enabling thermoforming, welding, and recycling without ever losing network integrity.

Vitrimers retain the solvent resistance, dimensional stability, and mechanical strength of classical thermosets while gaining

the reprocessability normally reserved for thermoplastics, a combination the field had lacked for nearly three decades since Dušek’s earlier, largely overlooked observations of related exchange behavior in epoxy networks. Because the network never fully depolymerizes, vitrimer parts can be reshaped, welded to one another, or ground and hot-pressed into new articles repeatedly, and fiber-reinforced vitrimer composites can be reprocessed without dissolving the matrix away from the fibers. This route offers clear practical advantages: existing epoxy/polyester curing chemistry can largely be reused, the exchange rate and T_v are tunable through catalyst identity and loading, and the resulting materials retain classical thermoset-grade performance. Its limitations center on the catalyst itself—it must remain active and non-leaching over the material’s service life, or exchange slows and reprocessability is lost; at low catalyst loading the exchange reaction can be prohibitively slow, making reprocessing energy- and time-intensive; and catalyst residues (commonly zinc- or amine-based) can affect color, long-term hydrolytic stability, or regulatory acceptability in some applications.

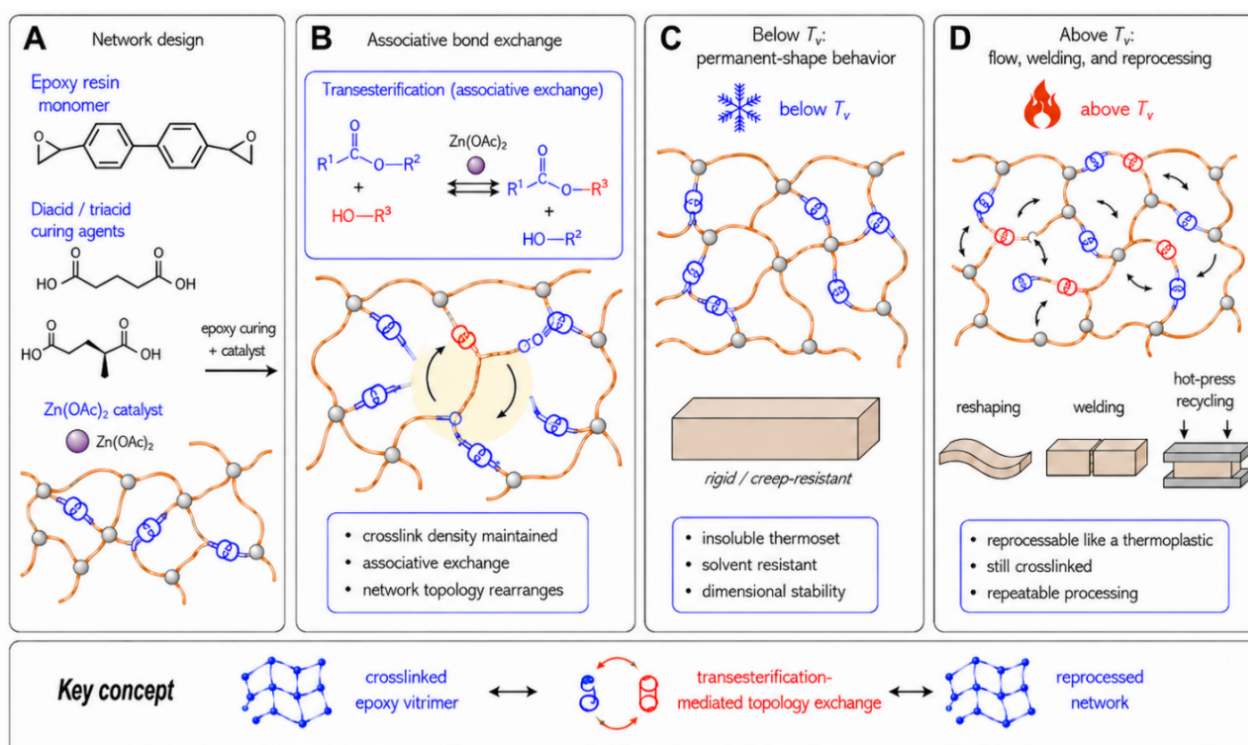


Figure 5. Transesterification-based vitrimer chemistry (Leibler and co-workers, Science 2011): $\text{Zn}(\text{OAc})_2$ -catalyzed associative bond exchange preserves crosslink density while allowing network topology to rearrange, giving permanent-shape behavior below T_v and reprocessable flow above it.²³

3.2.3. Disulfide exchange/metathesis

A parallel and, in some respects, gentler route to dynamic covalent networks exploits the sulfur–sulfur bond. Odriozola and co-workers (Rekondo, Martin, Ruiz de Luzuriaga, Cabañero, and Grande) showed in 2014 that incorporating bis(4-aminophenyl) disulfide as a crosslinker into a poly(urea-urethane) elastomer gave a material capable of essentially complete self-healing at room temperature. Aromatic disulfide metathesis is one of the very few dynamic covalent chemistries that proceeds at room temperature, and the resulting poly(urea-urethane) elastomers showed quantitative healing efficiency without the need for any catalyst or external intervention. This aromatic disulfide platform was subsequently extended to fully reprocessable and even fiber-composite thermosets by the same group, and, closer to the present authors' own work on bidirectional disulfide metathesis in disulfide-doped pDCPD, to tougher structural network architectures where disulfide exchange is combined with a stiffer backbone to give self-healing, weldable structural polymers rather than soft elastomers alone (Figure 6).

Depending on the disulfide substitution pattern (aromatic versus aliphatic) and backbone rigidity, this chemistry spans very soft, rapidly self-healing elastomers through to tough, dimensionally stable structural thermosets that can still be hot-pressed or reprocessed. Thermally accelerated disulfide exchange allows damaged pieces to be welded, reshaped, or hot-pressed into new articles, and aromatic-disulfide epoxy

networks have also enabled repairable and recyclable fiber-reinforced composites. Among its advantages, aromatic disulfide exchange can proceed without an added catalyst and, in suitably designed networks, at or near ambient temperature; the chemistry is compatible with polyurethane and epoxy backbones; and exchange rates can be accelerated thermally when faster reprocessing is required. Its limitations stem from the same sensitivity that gives it room-temperature activity: exchange kinetics are highly sensitive to the electronic environment and network mobility, so formulations that heal rapidly may also show unwanted stress relaxation or creep at service temperature, and raising backbone rigidity improves dimensional stability but generally increases the temperature required for practical reprocessing.

Building on the disulfide-exchange platform described above, the Loh group has developed a dual-mode recycling strategy for polydicyclopentadiene (pDCPD), a high-performance thermoset whose permanent carbon–carbon crosslinked network has historically resisted recycling despite its outstanding toughness, chemical resistance, and thermal stability. Loh's group embedded a cyclic aromatic disulfide comonomer into the pDCPD network through ring-opening metathesis polymerization (ROMP), giving a disulfide-doped pDCPD that supports two complementary recovery pathways within the same material: thermal reprocessing through associative disulfide exchange, and a separate chemical fragmentation–reconstitution route built around a small-molecule disulfide shuttle, dimethyl disulfide (DMDs).⁴⁰

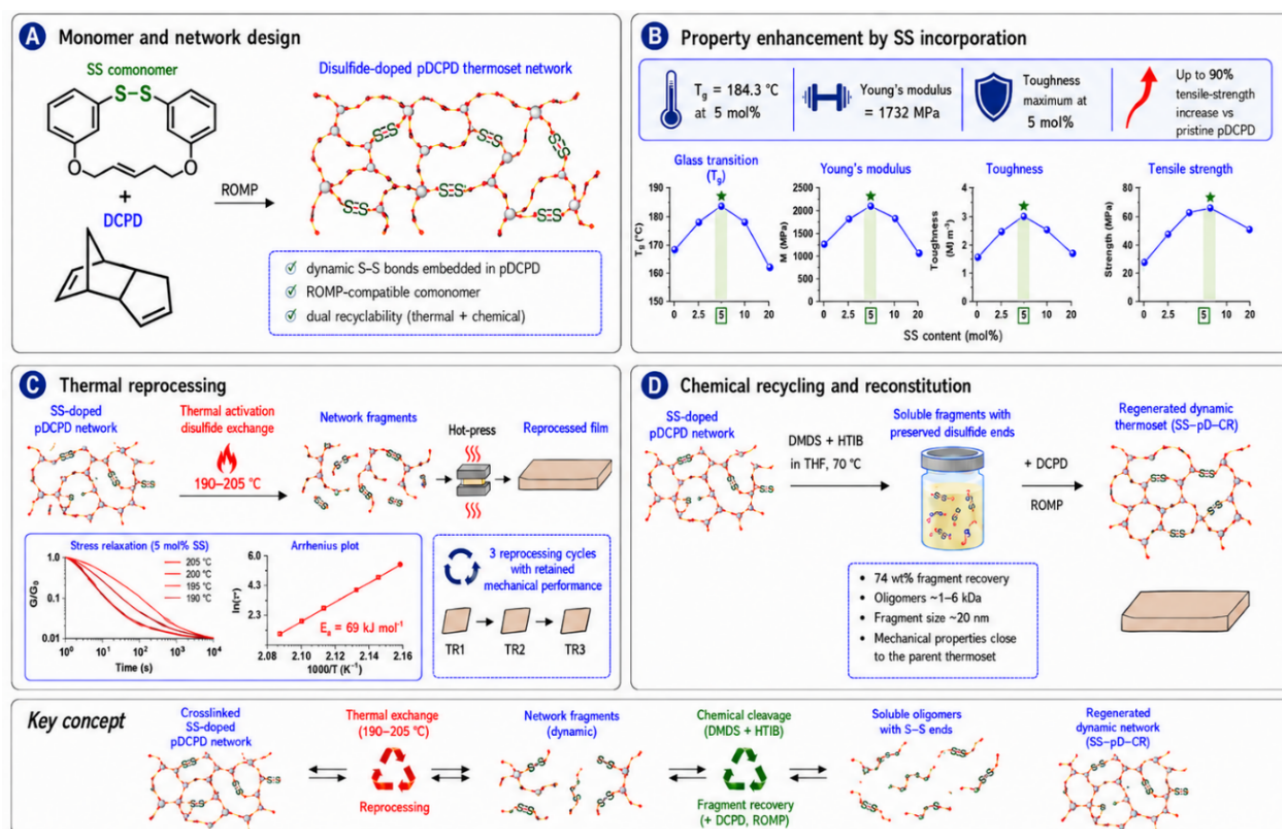


Figure 6. Bidirectional disulfide metathesis in disulfide-doped pDCPD (Li, Loh, and co-workers): a cyclic aromatic disulfide comonomer enables both thermal reprocessing via disulfide exchange and chemical fragment recovery/reconstitution via a DMDS shuttle, while enhancing T_g , modulus, and toughness relative to pristine pDCPD.

In this second pathway, DMDS undergoes bidirectional disulfide metathesis with the network's S–S bonds, progressively cleaving the crosslinked thermoset into soluble oligomeric fragments that retain disulfide termini and pendant cyclopentene/alkene functionality from the original ROMP backbone. These reactive fragments are then re-incorporated into a regenerated network by ring-opening copolymerization with fresh dicyclopentadiene monomer, with residual DMDS detectable by GC–MS in the reconstituted film—direct evidence that disulfide exchange continues through the reconstitution step.⁴⁰ Beyond enabling recovery, the aromatic disulfide unit reinforces the network itself: the optimal formulation (5 mol% disulfide comonomer) reached a glass transition temperature of 184.3 $^\circ\text{C}$ and a Young's modulus of 1,732 MPa, both exceeding pristine, non-recyclable pDCPD. Materials recovered through either pathway retained stiffness and tensile behavior close to the original.

It is important to be precise about what this strategy does and does not achieve, since the distinction matters for how it should be described in the literature. Because the degradation step (disulfide metathesis) and the reconstitution step (olefin metathesis with fresh DCPD) proceed through different bonds, the recovered material is not architecturally identical to the parent thermoset, and full depolymerization to a single, well-

defined monomer is not possible for a highly crosslinked hydrocarbon network of this kind—nor is it the goal of the strategy. This work therefore describes controlled network fragmentation and reconstitution rather than closed-loop recycling: the DMDS shuttle enables a genuinely tough, high-performance thermoset to be chemically recovered and rebuilt using fresh monomer, but the term 'closed-loop' is reserved in this review for chemistries—such as the diketoenamine systems of Section 3.2.5—that regenerate the original monomers themselves.

3.2.4. Boronic ester (boroxine/diol–boronate) exchange

Boron-centered dynamic bonds offer some of the fastest exchange kinetics of any covalent adaptable network, often operating at or near room temperature without any catalyst. Guan and co-workers (Cromwell, Chung, and Guan) demonstrated in 2015 that networks crosslinked through boronic ester bonds between boronic acid- and diol-functionalized polymer segments could be made both malleable and self-healing, with the rate of bond exchange tunable simply by adjusting the electronic and steric character of the boronic ester (Figure 7).²⁸

Related work from the same and other groups extended this to fully boroxine-crosslinked thermosets that are recyclable, strong, and highly malleable, exploiting the same underlying diol/boronate exchange equilibrium.²⁹

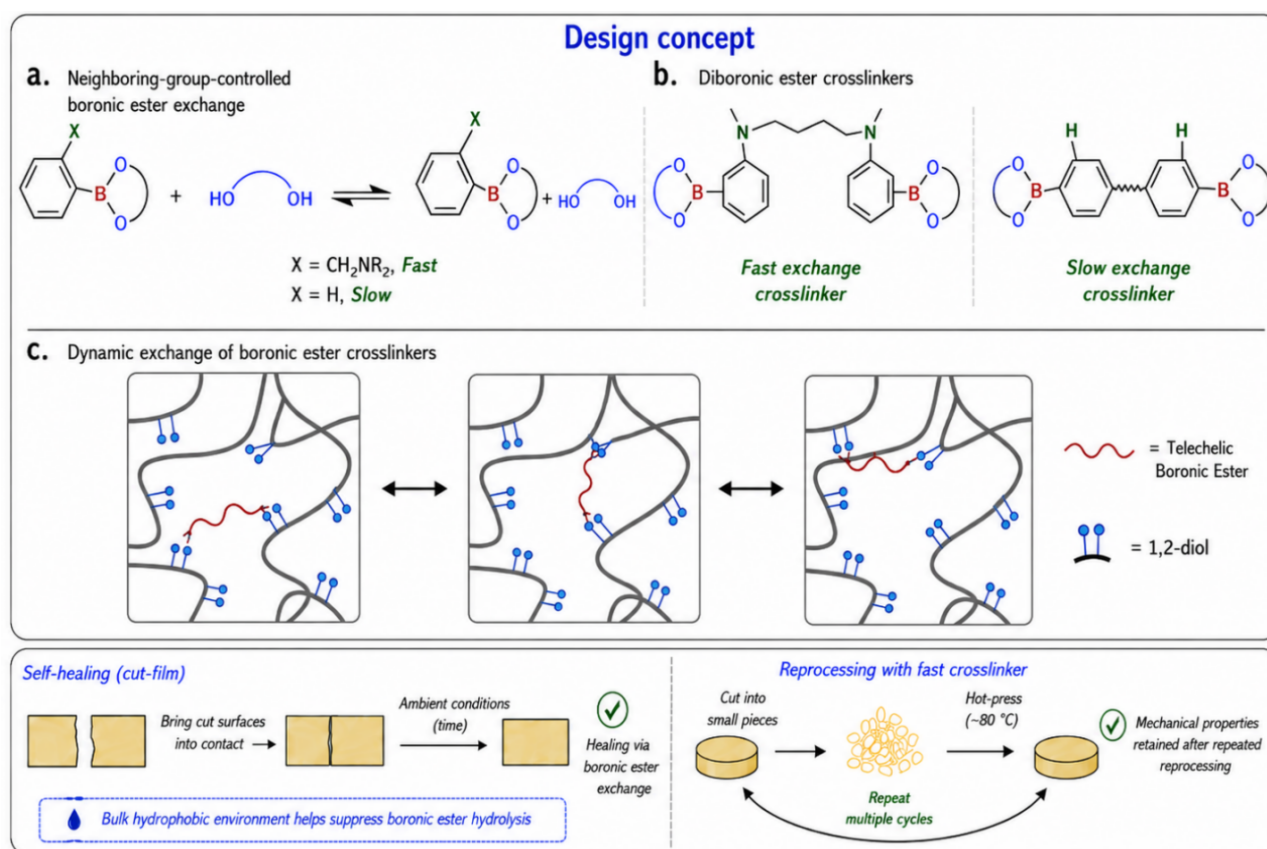


Figure 7. Neighboring-group-controlled boronic ester exchange (Guan and co-workers): electronic tuning of the boronic ester crosslinker sets fast- versus slow-exchange behavior, enabling self-healing and repeated hot-press reprocessing of the network.

These networks can be engineered from very soft, rapidly self-healing materials through to mechanically robust, malleable thermosets, depending on the density, rigidity, and exchange kinetics of the boronic ester crosslinks. Dynamic boronic ester networks can be reprocessed by mild heating as exchange redistributes crosslinks; competing diols can also shift boronate equilibria, while boroxine networks can be chemically deconstructed by water back to boronic-acid-containing building blocks. Among their advantages, boronic ester exchange can be extremely fast and can often proceed without an added catalyst; neighboring-group effects provide a powerful way to tune exchange kinetics over orders of magnitude; and hydrolytic reversibility can be deliberately exploited when degradability is desired. The corresponding limitations largely mirror this same reactivity: hydrolytic sensitivity can become a liability for long-term service in humid or aqueous environments, and water content must therefore be controlled during formulation and processing, while the same rapid exchange that improves malleability can also increase creep if it remains too active at the intended service temperature.

3.2.5. Imine and diketoenamine exchange (dynamic covalent nitrogen chemistry)

One of the most complete demonstrations of chemical circularity among reversible-bond strategies comes from imine-type

chemistry, and specifically from the diketoenamine bond introduced by Helms and co-workers (Christensen, Scheuermann, Loeffler, and Helms) in 2019 (Figure 8). Poly(diketoenamine)s ‘click’ together from a wide variety of triketones and aromatic or aliphatic amines, yielding only water as a by-product, and, critically, these networks can be depolymerized in strong aqueous acid to recover the triketone and amine monomers, even from plastics compounded with colourants, fillers, and flame retardants. The recovered monomers can then be re-manufactured into the same polymer formulation without loss of performance, or into other polymer formulations with different, differentiated properties—a genuine closed loop rather than mere reprocessing. Notably, the click-condensation itself proceeds at room temperature without catalyst, additive, or solvent, requiring only a few minutes of ball milling.

Mechanical properties on par with thermoplastic polyurethanes (materials used in phone cases and shoe soles) have been reported for poly(diketoenamine)s, combined with excellent in-service stability—the network is robust under normal use conditions and only cleaves on demand under strongly acidic aqueous conditions. Immersion in strong acid cleanly hydrolyzes the diketoenamine linkages, separating the polymer’s monomers from compounded additives and fillers; the recovered triketone and amine can then be repolymerized into virgin-quality resin, or diverted toward a different polymer

formulation entirely—true upcycling. Among its advantages, complete depolymerization to pure, reusable monomers is achievable even from filled or colored composite parts, the click-type condensation chemistry requires no catalyst, solvent, or elevated temperature to form the polymer in the first place, and the material is robust in normal service, decoupling ‘stable while in use’ from ‘recyclable on demand.’ The main limi-

tations lie downstream of this same acid-triggered cleavage: full depolymerization requires concentrated aqueous acid, introducing handling, corrosion, and downstream neutralization/separation requirements; monomer recovery from mixed or compounded waste still requires extraction and purification steps; and PDK feedstocks and processing infrastructure are less mature than those of commodity thermoset chemistries.

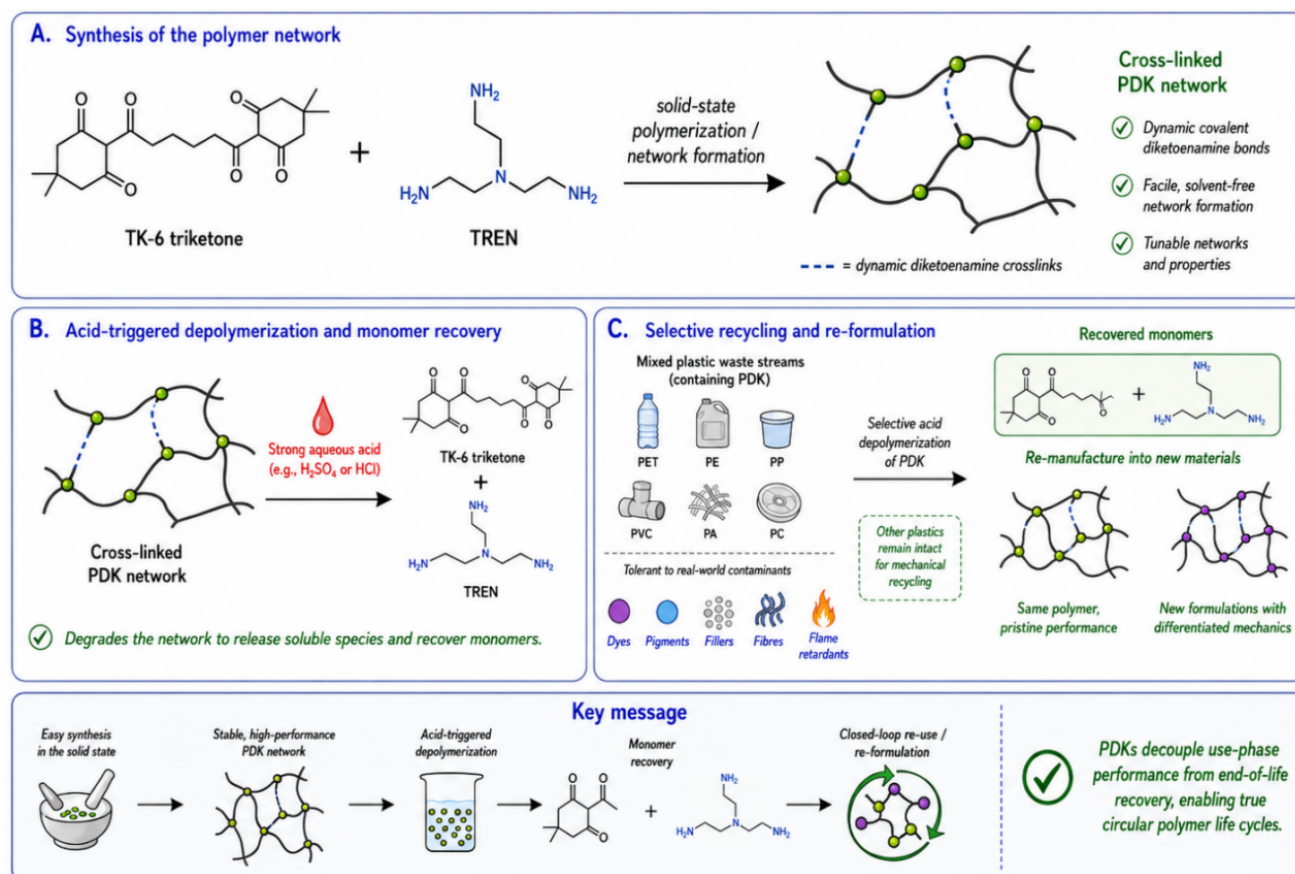


Figure 8. Closed-loop recycling via dynamic covalent diketoenamine bonds (Helms and co-workers): poly(diketoenamine) networks formed from a triketone and a triamine can be depolymerized in strong aqueous acid to recover the original monomers, even from filled or colored composite waste, for re-formulation into new materials.

3.2.6. Olefin metathesis of unsaturated networks

Ring-opening metathesis polymerization of strained cyclic olefins such as dicyclopentadiene, using Grubbs-type ruthenium catalysts, produces some of the toughest structural thermosets available (e.g., pDCPD), and the residual backbone unsaturation these networks retain offers a route back to degradation. Smith, Boothroyd, Thompson, and Khosravi demonstrated in 2016 that first- and second-generation Grubbs ruthenium catalysts break down crosslinked polybutadiene networks at their residual double bonds via cross-metathesis, producing a dramatic reduction in molecular weight—down to around 2000 g mol⁻¹—to give readily soluble molecules; applying the same G2-catalyst approach, together with a diester chain-transfer agent, to a styrene-butadiene rubber sheet (the

material used in vehicle tyres) broke the crosslinked network down into rubber crumb at room temperature within a few hours. This bond-breaking depolymerization route stands apart from the associative exchange strategies above in that it genuinely severs the polymer backbone rather than merely rearranging crosslink topology (Figure 9).

Crosslinked polybutadiene and styrene-butadiene rubber are normally insoluble, difficult-to-process networks; metathesis-triggered bond cleavage provides a catalytic route to much lower-molecular-weight or fragmented products under conditions far milder than pyrolysis. Among the advantages of this approach, olefin-metathesis catalysts can cleave unsaturated polymer backbones under comparatively mild conditions, providing a true bond-breaking pathway rather

than merely rearranging crosslink topology, and the chemistry also builds on a mature mechanistic and catalyst-design framework.⁴¹ Its limitations are largely practical: ruthenium catalyst cost and recovery from the product stream are im-

portant barriers, catalyst deactivation by impurities can complicate treatment of real waste, and controlling the extent of chain scission while avoiding incomplete degradation remains a process-design challenge.

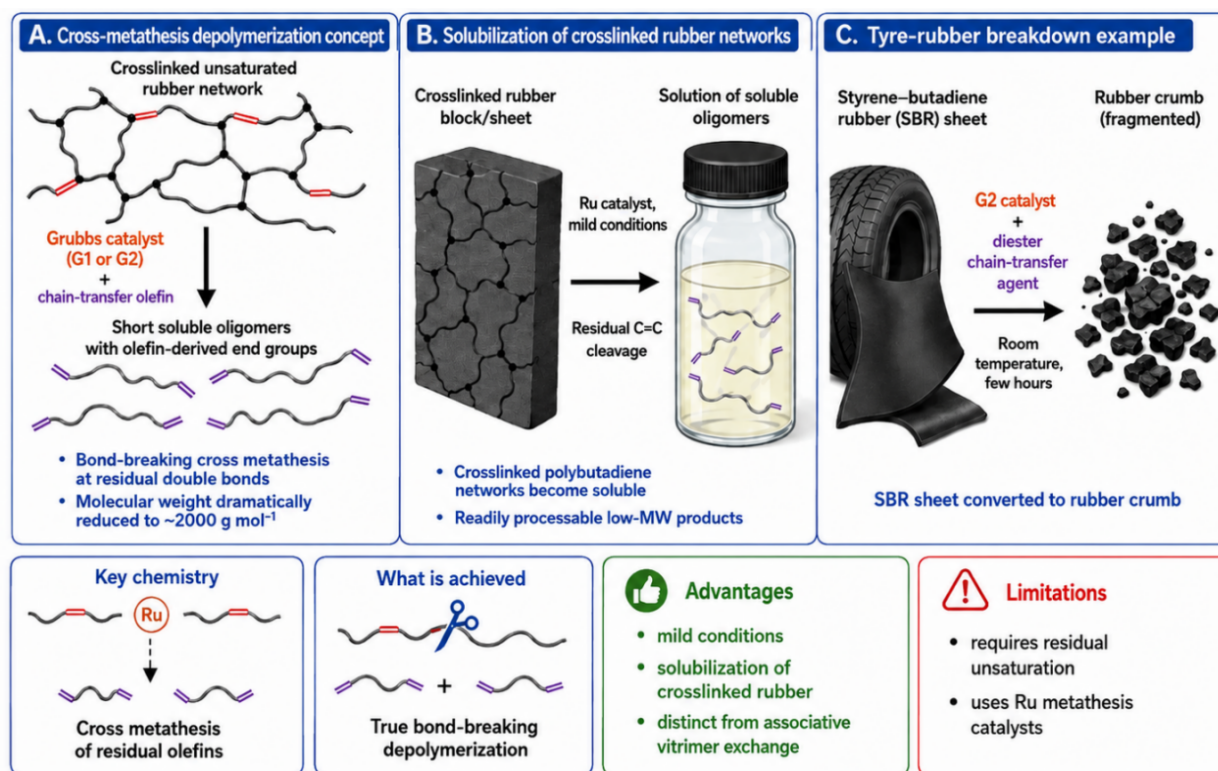


Figure 9. Bond-breaking depolymerization by olefin cross-metathesis: Grubbs ruthenium catalysts cleave residual C=C bonds in crosslinked unsaturated rubber networks, solubilizing crosslinked polybutadiene and converting styrene-butadiene tyre rubber into rubber crumb under mild conditions.

3.2.7. Transcarbamoylation (dynamic polyurethane vitrimers)

Extending the vitrimer concept beyond ester chemistry, Dichtel and co-workers (Fortman, Brutman, Cramer, and Hillmyer) demonstrated in 2015 that polyhydroxyurethane networks—built without any isocyanate monomers, from bis(cyclic carbonate) and triamine precursors—undergo associative transcarbamoylation exchange and can be reprocessed by heat and pressure alone. Notably, this exchange was found to be mechanically activated: applying stress lowers the effective activation energy for bond exchange relative to that measured for small-molecule model compounds, a mechanistic nuance not seen in the Leibler-type ester vitrimers. Follow-up work from the same group showed that carbamate exchange in conventional (isocyanate-derived) crosslinked polyurethanes could also be catalyzed directly, opening a route to reprocessing existing polyurethane thermosets and foams rather than only purpose-built polyhydroxyurethane networks (Figure 10).

Reprocessed polyhydroxyurethane vitrimer parts recovered a substantial majority of their as-synthesized tensile properties

after multiple reprocessing cycles, while avoiding the toxic isocyanate monomers used in conventional polyurethane manufacture. Among its advantages, this chemistry targets a large and historically difficult-to-recycle polymer class; the polyhydroxyurethane route also avoids isocyanate monomers, while mechanically assisted exchange provides a catalyst-free route to reprocessing purpose-built PHU networks. Its limitations are chiefly practical: carbamate exchange generally requires substantial thermal input, and conventional polyurethane systems typically need an exchange catalyst to reach useful reprocessing rates; some effective catalysts, including organotin species, raise toxicity and regulatory concerns, while conventional isocyanate-derived feedstocks retain the upstream sustainability drawbacks of isocyanate chemistry.

3.2.8. Siloxane (Si–O–Si) equilibration

Polydimethylsiloxane networks are intrinsically prone to Si–O bond redistribution under acid or base catalysis—a phenomenon characterized mechanistically by Kantor, Grubb, and Osthoff in 1954 and directly linked to chemical stress relaxation

of PDMS elastomers by Osthoff, Bueche, and Grubb. Zheng and McCarthy revisited this chemistry in 2012, showing that active silanolate-ended 'living' silicone networks could self-heal and be reversibly reshaped by siloxane equilibration.³¹ More

recent silicone systems exploit either direct dynamic siloxane exchange or dynamic boronic-ester motifs within silicone architectures to enable reprocessing, self-healing, and additive manufacturing (Figure 11).

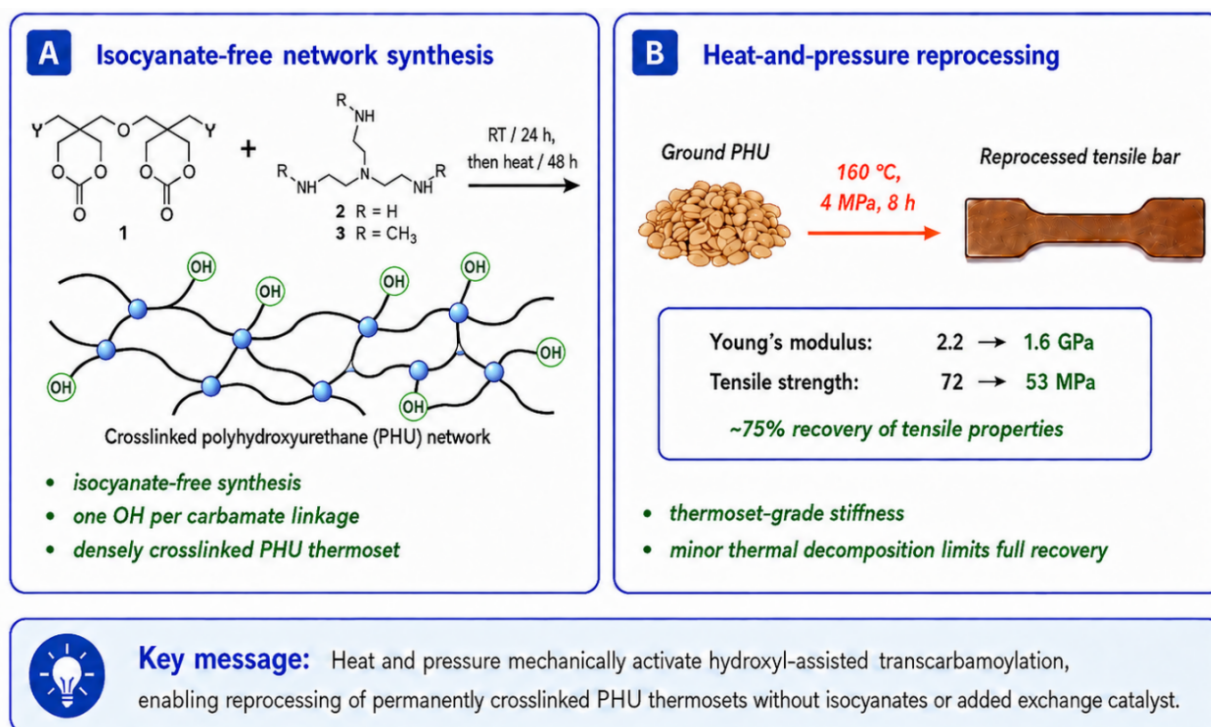


Figure 10. Transcarbamylation-based polyhydroxyurethane vitrimers (Dichtel and co-workers): isocyanate-free networks from bis(cyclic carbonate) and triamine precursors undergo mechanically activated associative transcarbamylation, recovering roughly 75% of tensile properties after heat-and-pressure reprocessing.

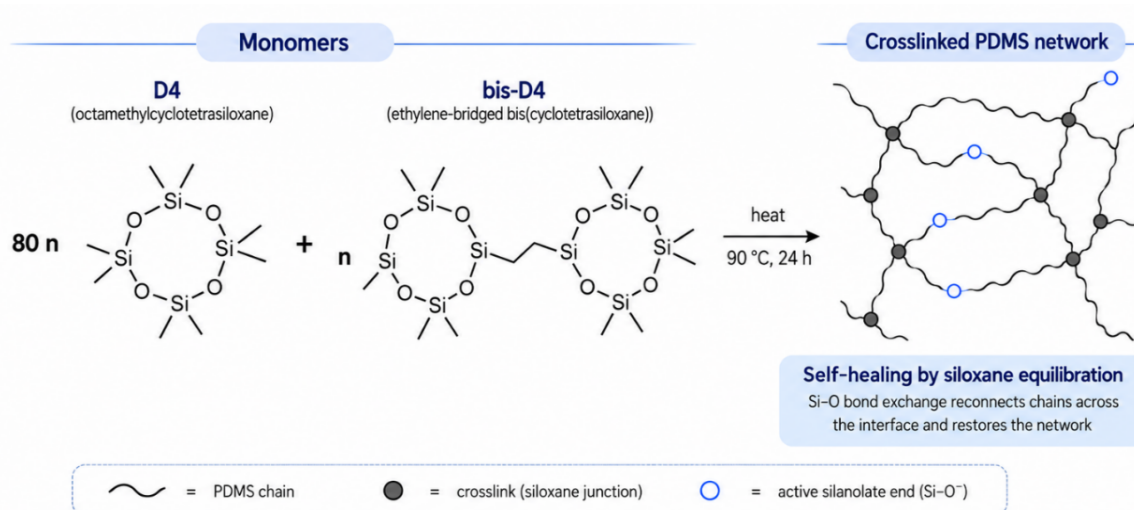


Figure 11. Siloxane equilibration in a living PDMS network (Zheng and McCarthy): Si-O bond redistribution at active silanolate ends reconnects chains across a cut interface, enabling self-healing of the crosslinked silicone network.

Reprocessable silicone elastomers retain the excellent thermal and oxidative stability that silicones are prized for in service, while gaining the capacity for reshaping or self-healing that a conventional, permanently cured silicone network lacks.

Among its advantages, siloxane exchange can be rapid under suitable catalysis, silicone backbones retain their characteristic flexibility and thermal stability, and chemical equilibration can support both self-healing/reprocessing and, under appropriate

conditions, recovery of cyclic siloxane feedstocks. Its limitations center on catalyst control: the exchange catalyst must be controlled or quenched to prevent continued equilibration during service, and filled commercial silicone formulations complicate clean feedstock recovery, while very rapid exchange can also compromise dimensional stability unless catalyst loading and service temperature are carefully matched.

3.2.9. Alkoxyamine/Nitroxide exchange (radical-based dynamic bonds)

Otsuka and co-workers (Otsuka, Aotani, Higaki, and Takahara) reported in 2002 that TEMPO-containing poly(alkoxyamine ester)s undergo a dynamic, reversible radical crossover reaction: the C–ON alkoxyamine bond homolyzes reversibly at elevated temperature to generate a nitroxide radical and a carbon-centered radical that can recom-

bine at a different site. Subsequent work demonstrated macro-molecular ‘polymer scrambling’ between alkoxyamine-based dynamic covalent polymer chains and thermodynamically controlled reversible crosslinking of poly(methacrylic ester)s bearing alkoxyamine side groups (Figure 12).

Alkoxyamine-based dynamic covalent polymers are stable under normal conditions but reorganize on heating, allowing molecular-weight redistribution, polymer scrambling, and thermodynamically controlled network formation without changing the underlying bond chemistry. Among its advantages, exchange is encoded in a single thermally reversible covalent bond, reaction rate and equilibrium can be tuned through alkoxyamine structure, temperature, concentration, and stoichiometry, and the mechanism is closely connected to the well-established chemistry of nitroxide-mediated radical processes.

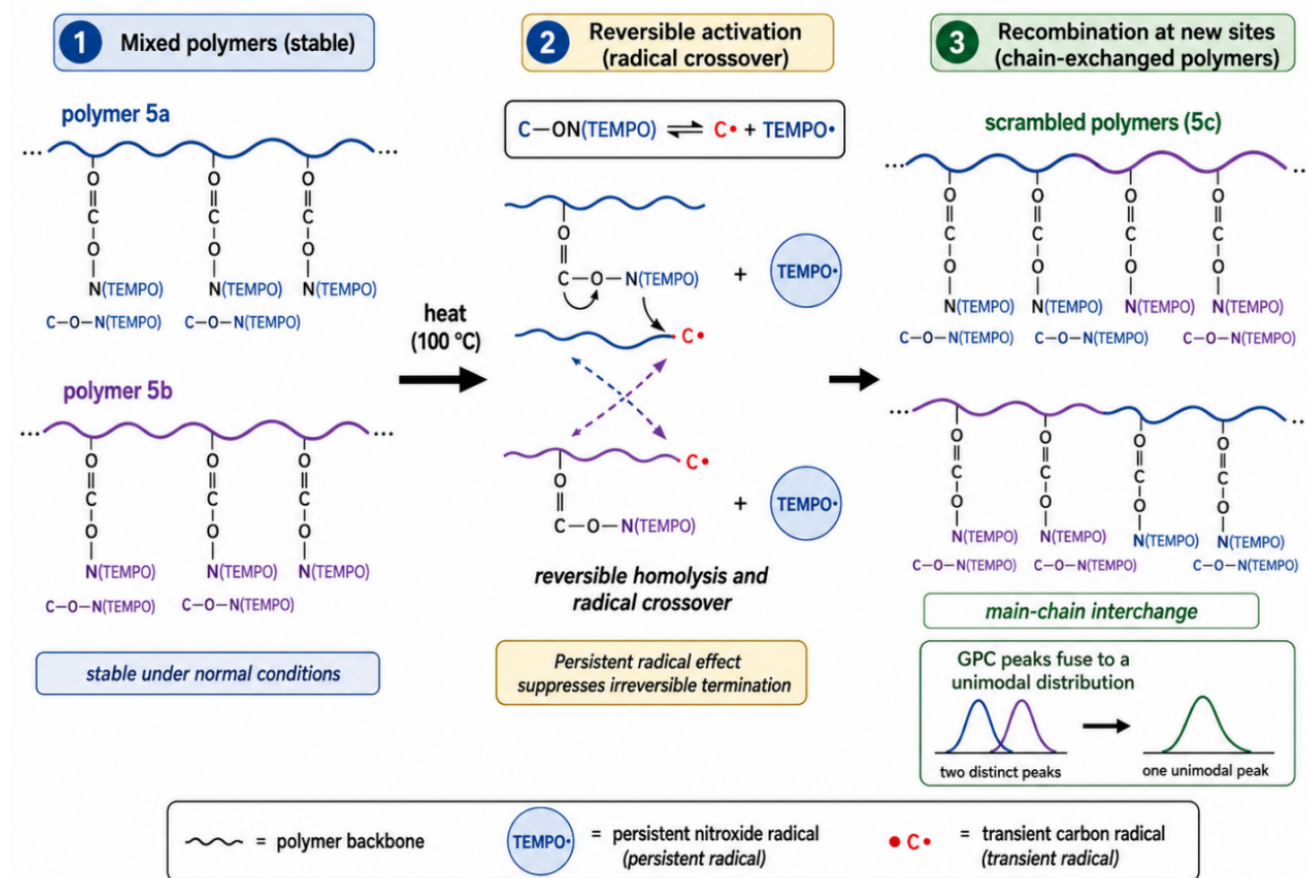


Figure 12. Reversible radical crossover in TEMPO-containing poly(alkoxyamine ester)s (Otsuka and co-workers): thermally reversible C–ON homolysis and recombination scrambles two distinct polymer populations into a single, chain-exchanged distribution.

Its limitations reflect the radical nature of the exchange: useful exchange generally requires elevated temperature, the radical intermediates introduce pathways for irreversible termination or oxidation over repeated cycles, and, compared with ester, disulfide, and boronate covalent adaptable networks, network-level recycling demonstrations remain relatively lim-

ited.

3.2.10. Addition–fragmentation chain transfer (AFCT) chemistry

Scott, Schneider, Cook, and Bowman reported in 2005 that a crosslinked polymer network containing allyl sulfide groups

could relax stress and change shape upon exposure to light without a measurable loss of bulk material properties. Photocleavage of residual photoinitiator in the polymer matrix generates radicals that diffuse through the network by addition–fragmentation chain transfer at midchain allyl sulfide groups, producing photoinduced plasticity, actuation, and equilibrium shape change without residual stress. Because the addition–

fragmentation step regenerates an exchangeable radical, topology rearrangement can propagate through the network while radical flux persists. Follow-up work from the Bowman group extended this exchange logic to thiol-ene/thiol-yne, methacrylate, and trithiocarbonate-containing networks, particularly to reduce polymerization-induced shrinkage stress and enable post-cure stress relaxation (Figure 13).

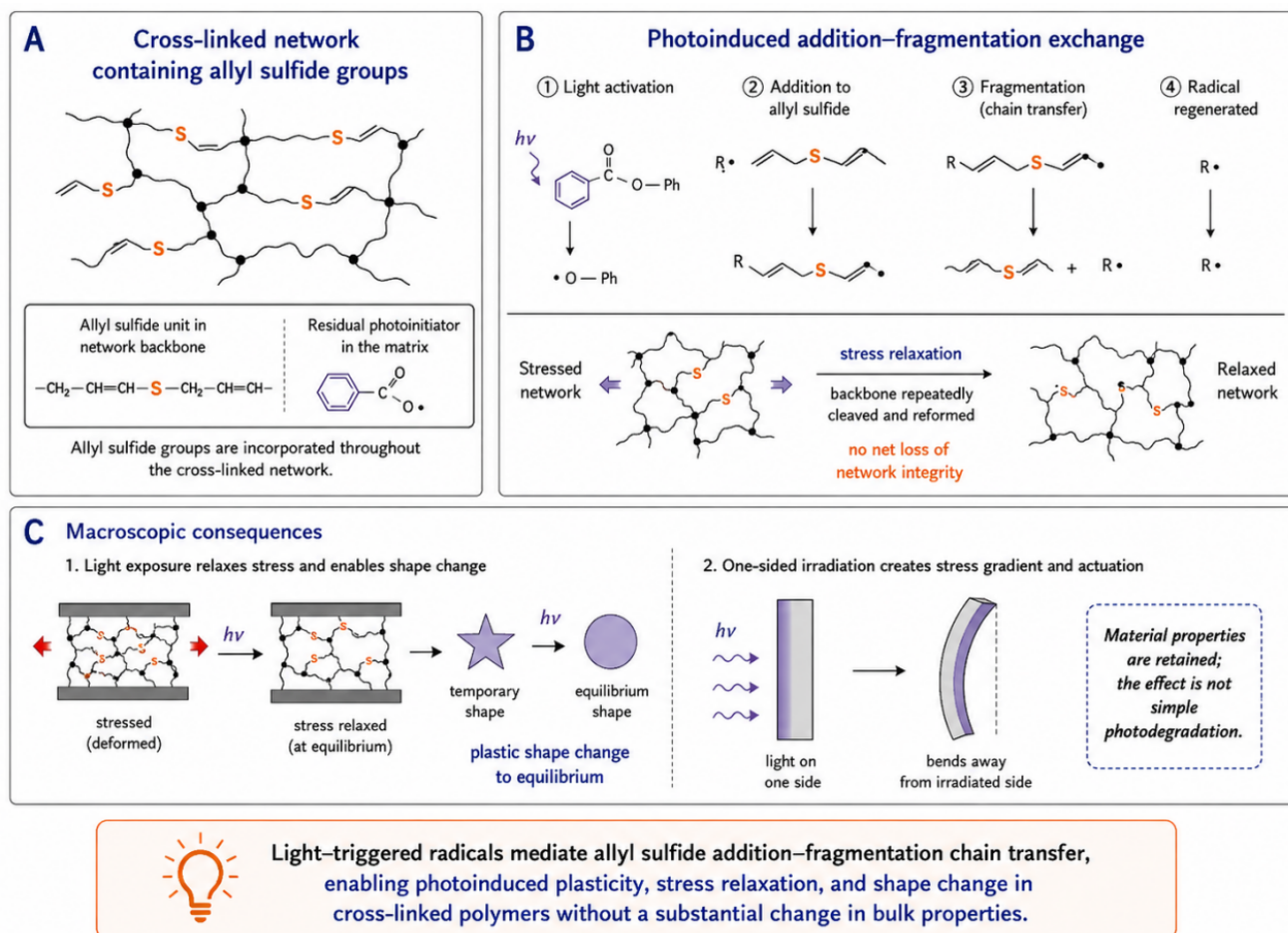


Figure 13. Photoinduced addition–fragmentation chain transfer at allyl sulfide groups (Bowman and co-workers, building on Scott et al., Science 2005): light-triggered radicals repeatedly cleave and reform the network backbone, relaxing stress and enabling plastic shape change without net loss of network integrity.

Networks containing AFCT-active groups can be photochemically stress-relaxed and reshaped while retaining network connectivity and mechanical properties, providing spatial and temporal control that is difficult to achieve with purely thermal exchange chemistries. Among its advantages, the addition–fragmentation event regenerates the exchange functionality and a radical carrier, so repeated topology rearrangement can continue while radicals are supplied; photochemical activation gives strong spatial and temporal control; and the chemistry draws on mature thiol-ene and radical-chain-transfer design principles. Its limitations follow from its radical-mediated mechanism: a radical source is required, useful exchange depends on radical lifetime and the density of

exchangeable groups, oxygen and other radical traps can suppress the process, and sulfur-containing allyl sulfide or trithiocarbonate motifs may complicate color, odor, or formulation in some applications.

4. Future trends and outlook

Several converging trends are likely to define the next decade of research at the interface of polymer chemistry, catalysis, and sustainability science.

Mixed-waste, sorting-free processing. The overwhelming majority of post-consumer plastic waste is not a single, pure resin but a heterogeneous mixture of resin types, additives, and contaminants. Photocatalytic platforms such as the Loh

group's metal-free O-VN-CN system, which operate under ambient conditions and tolerate mixed feedstocks without pre-sorting, point toward a broader shift away from single-polymer depolymerization demonstrations and toward technologies engineered from the outset for the compositional reality of municipal and industrial waste streams. Enzymatic platforms will likely follow a parallel trajectory, with engineered hydrolase cocktails and chemical pretreatment steps extending selectivity beyond PET to polyamides, polyurethanes, and polyolefin blends.

Hybrid photo-, electro-, and biocatalytic systems. The boundaries between photochemical, electrochemical, and enzymatic upcycling are already beginning to blur, as illustrated by coupled enzymatic–photocatalytic photoreforming platforms and by electro-biological hybrids that pair electrochemical PET-derived ethylene glycol oxidation with microbial upgrading. Integrating renewable electricity, light, and biocatalysis within a single reactor, so that H₂ co-production, CO₂ utilization, and monomer recovery are optimized together rather than separately, is a natural and largely unrealized extension of the chemistry reviewed here.

A broader toolbox of bidirectionally controlled reactions. As emphasized in Section 3, the central synthetic bottleneck for molecularly programmed recyclability is not the concept of reversibility itself but the scarcity of exchange chemistries that are simultaneously robust in service and cleanly reversible on demand. Expanding beyond the now well-established disulfide, imine, boronate, and Diels–Alder motifs toward new redox-, photo-, and mechanically triggered bidirectional bonds—building on precedents such as shuttle-controlled disulfide metathesis—will be essential for bringing covalent adaptable networks to genuinely commodity-scale, high-performance thermosets rather than niche specialty materials.

Weak-C–C and cleavable-comonomer strategies for polyolefins. Because polyethylene and polypropylene together account for the largest share of global plastic production, extending controlled degradability to all-carbon backbones without sacrificing the crystallinity, strength, and processability that make these polymers commercially dominant is arguably the single highest-value target in the field. Continued progress on topochemically weakened backbones, low-level cleavable comonomers, and catalytic hydrogenolysis and metathesis chemistries will determine how much of the polyolefin waste stream can realistically be brought into a closed loop.

Data-driven catalyst and monomer design. Machine-learning-guided screening of catalyst composition, defect density, and reaction conditions is beginning to accelerate the discovery of photocatalysts, hydrogenolysis catalysts, and exchange chemistries, complementing the largely empirical structure–property exploration that has characterized the field to date. Coupling high-throughput experimentation with computational screening of bond dissociation energies and ex-

change kinetics could shorten the path from a promising bond motif to a scalable, orthogonally stable polymer design.

Life-cycle assessment, standardized terminology, and policy alignment. As the number of mechanistically distinct ‘recycling’ and ‘upcycling’ technologies grows, so does the risk of terminology outrunning substance. This review has deliberately distinguished controlled network fragmentation and reconstitution from genuine closed-loop monomer recovery; adopting similarly precise, mechanism-based terminology across the field, paired with rigorous life-cycle and techno-economic assessment, will be necessary to direct investment and extended-producer-responsibility policy toward the technologies that deliver real reductions in virgin plastic demand and greenhouse-gas emissions, rather than toward those that are merely described most favorably.

Scale-up and industrial integration. Enzymatic PET biorecycling has already demonstrated that molecularly selective recycling chemistry can be commercialized at plant scale. Continuous-flow reactor engineering offers a promising route to mass-producing these chemistries at scale, improving light and mass transport, mixing, and heat management relative to batch processing, but translating lab-scale photocatalytic, electrocatalytic, and depolymerization chemistries into robust flow platforms will require overcoming challenges such as catalyst fouling and immobilization, reactor clogging from heterogeneous or particulate plastic feedstocks, and maintaining consistent residence time and conversion at industrially relevant throughput. The next test for the field is whether photocatalytic, electrocatalytic, and covalent-adaptable-network technologies can achieve comparable reliability, catalyst lifetime, and cost structure outside the laboratory. Continued collaboration between academic groups, catalyst and materials suppliers, and waste-management operators will be essential to translate the mechanistic diversity surveyed in this Perspective into measurable reductions in landfilled and incinerated plastic waste.

5. Conclusion

This Perspective has surveyed a chemistry landscape that is expanding rapidly in both mechanistic diversity and practical impact: classical end-of-life treatment and its well-characterized limits; selective depolymerization, catalytic bond-breaking, and polymer functionalization; photochemical and electrochemical upcycling, including a metal-free, ambient-condition carbon nitride platform capable of processing mixed plastic waste without pre-sorting; enzymatic and chemo-biological recycling, now operating at industrial scale for PET; and, at the front end, ten reaction platforms that organize the design of molecularly programmed, recyclable-by-design polymers into five coherent principal classes. Across all of these approaches, the same design logic recurs: durability during service and reactivity at end of life are not competing requirements to be

traded off, but properties that can be engineered independently once the chemist controls precisely where, when, and how a bond breaks. From shuttle-controlled, bidirectional disulfide metathesis that recovers high-performance thermosets without sacrificing their mechanical properties, to diketoenamine networks that regenerate pure monomer from filled and colored composite parts, the most powerful systems in this review succeed because they treat recyclability as a molecular design parameter rather than an afterthought. As these mechanistically distinct strategies mature and increasingly converge, the field is moving decisively—and irreversibly—from managing plastic waste after the fact toward closing the loop by design.

Author Contributions

Conceptualization, T.-P.L. and B.L.; methodology, T.-P.L. and B.L.; writing—original draft preparation, T.-P.L. and B.L.; writing—review and editing, X.W. and X.Y.K.; visualization, B.L.; supervision, T.-P.L.; project administration, T.-P.L.; funding acquisition, T.-P.L. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement

No new data were generated in this review.

AI Use Statement

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Conflict of Interest

The Editor-in-Chief (T.P.L.) is an author of this manuscript and therefore had no involvement in its editorial evaluation, peer-review process, or publication decision. The manuscript was handled independently by a designated editor in accordance with the journal's editorial procedures. The authors declare no conflict of interest.

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