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Review

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**Abstract:** Ranging from traditional food packaging, clothing, and furniture to the current small and large electronic devices and automobiles, plastics serve to fulfill diverse demands in our daily lives. However, the global plastic waste generation is dramatically escalating, currently standing at approximately 150 million metric tonnes annually. While some of regenerated plastics recycled by mechanical methods can be used as their parent plastics, cost and energy savings are limited by multiple preliminary processes such as plastic sorting, shredding, washing, and drying. Moreover, the continuous mechanical recycling process degrades the physical properties of the materials. In this context, chemical recycling is emerging as a promising alternative method due to its high efficiency, simple preliminary steps, reducing reliance on fossil resources, and conversion of plastic waste into value-added chemicals. This review provides a state-of-the-art overview of contemporary chemical recycling of polymeric materials via i) depolymerization: "polymers to small valuable molecules" and ii) closed-loop cycles: "polymers to monomers, and/or to polymers", by encompassing both traditional/advanced depolymerization chemistries and the remaining challenges. These recycling approaches are contextualized within the present industrial technologies, key design principles, and specific recycling case studies related to distinct polymeric materials.

**Keywords:** recycling; plastic wastes; chemical recycling; depolymerization; closed-loop recycling

## 1. Introduction

Plastics have become an essential part of our daily lives, ranging from food packaging, clothing, and housing to the current state-of-the-art technologies found in smart phones, vehicles, and medical equipment. The durability, versatile properties, cost-effectiveness, and light weight of plastics have made them ubiquitous in our daily lives. However, the unintended consequence of this ubiquity is the unprecedented surge in global plastic waste generation[1]. Between 1950 and 2015, cumulative waste generation of primary and secondary (i.e., recycled) plastic waste reached 6300 Mt; with estimates indicating that 2500 Mt of them are currently in use (**Figure 1**)[2,3]. Within this extensive volume, the bulk of discarded plastics, approximately 4900 Mt (equivalent to 60% of plastics produced during the period), reside in landfills or persist in the natural environment. Among the other portions, 800 Mt (12%) of plastics have been ended in incineration, and only 600 Mt (9%) of them have undergone recycling processes. It is worth to note that only a fraction of this recycled plastic has met more than a single round of recycling[4]. This extensive plastic waste generation has increased worldwide plastic-related environmental concerns and the need for more sustainable plastic recycling approaches[2,5–12].
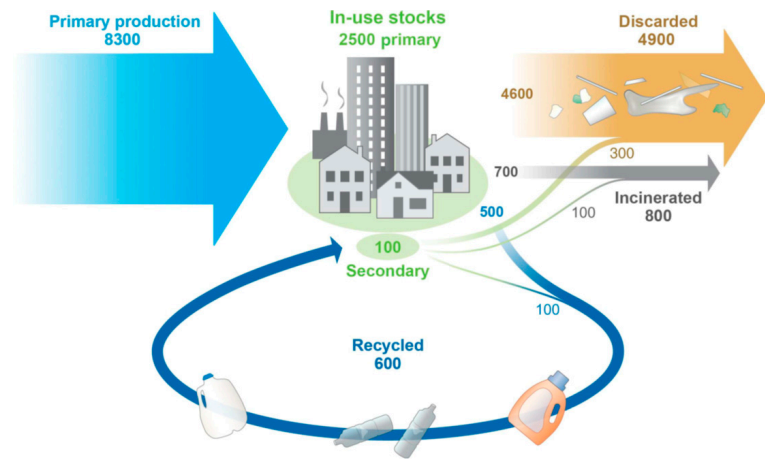
**Figure 1.** Global production, use, recycling, and end-up of plastics (1950 to 2015; in million metric tons).[2] Reproduced with permission. Copyright 2017, American Association for the Advancement of Science (AAAS).

There are four types of plastic recycling methods based on ASTM and ISO standards (**Table 1**)[7], offering distinct approaches to dealing with plastic wastes. Mechanical recycling, namely primary or secondary recycling, is the process of recovering plastic waste by mechanical processes such as sorting, washing, drying, grinding, re-granulating, and compounding[7,13–15]. Given that this process does not change the chemical structure of the material, multiple re-use/re-cycling of polymeric materials could be possible. In addition, other advantages such as efficiency in terms of time, carbon footprint, and relatively low environmental impact make the mechanical recycling attractive. However, the mechanical process within an extruder applies thermal and physical stresses to polymer melts, leading to oxidative radical formation, chain scission, chain branching, and/or crosslinking, thereby inducing degradation of the materials and/or processing facilities. In this regard, some of recycled materials that exhibit worse physical properties compared to the original materials are used in other fields such as textiles, construction, and low grade packaging (2nd recycling)[16]. The 3rd recycling, i.e., chemical recycling, is defined as "the process of recovering petrochemicals and/or chemical building blocks by changing the chemical structures". In general, this has been applied to plastic wastes, where mechanical recycling is no longer possible and the resultant products are valuable enough. The last recycling method, quaternary recycling or energy recovery, is applied to plastics that the recycling methods above is not applicable, and is typically used for energy recovery through pyrolysis[17–23]. The major advantage of this method is a reduction in volume of the plastic wastes, therefore, it retains little value in terms of environmental impact. Furthermore, pyrolysis of some plastics may cause negative environmental impacts, such as greenhouse gas generation and toxic gas emissions.

**Table 1.** Common definitions of plastic recycling methods and their examples.

| ASTM D7209 | ISO 15270:2008 | Example |
|---|---|---|
| Primary (1st) recycling | Mechanical recycling | PET bottle to PET bottle |
| Secondary (2nd) recycling | Mechanical recycling | PET bottle to PET fiber |
| Tertiary (3rd) recycling | Chemical recycling | Glycolysis of PET |
| Quaternary (4th) recycling | Energy recovery | Pyrolysis of PET |

While industrial chemical recycling has been a minor process compared to mechanical recycling, advanced chemical recycling has emerged as a compelling alternative in response to the inherent limitations of mechanical recycling[6,9,24]. Advanced chemical recycling processes are distinguished by their high efficiency, streamlined preliminary steps, reduced reliance on finite fossil resources, and, in some cases, the achievement of physical properties comparable to those of the original

materials. Impartant appealing feature of chemical recycling is to use plastic waste as a carbon source and convert the waste into value-added chemicals or monomers. This approach provides an upcycling strategy for generating value-added chemicals primarily through depolymerization. Furthermore, the recovered monomers obtained by depolymerization can be polymerized again to produce a new polymer that exhibits comparable mechanical properties to the virgin polymer, which is known as closed-loop chemical recycling. This transformative approach holds the potential to redefine the recycling landscape, offering a pathway to not only manage plastic waste more sustainably but also reduce our dependence on virgin fossil resources. However, there remain critical challenges in balancing the polymerizability, depolymerizability, recyclability, and material performance, because there are complicated trade-offs between the factors. Furthermore, achieving mild reaction conditions during chemical recycling is another critical factor to consider, as depolymerization processes often require harsh reaction conditions, including high temperatures, expensive catalysts, and strongly acidic or basic conditions.

This review organizes the latest research efforts on the chemical recycling of the polymeric materials from the polymer life cycle viewpoint: *i*) polymer to small molecules and *ii*) polymers to monomers, and monomers to polymers. With this, our objective is to provide a detailed examination of both traditional and state-of-the-art depolymerization chemistries, elucidating their underlying principles, technological implications, and potential advantages. We believe this review provides insight into developing an advanced circular economy for polymeric materials through chemical recycling.

## 2. Chemical Recycling via Depolymerization: Polymers to Small Valuable Molecules

In a typical chemical recycling process, the major objective is to break the chemical bonds of the polymers (i.e., depolymerization) to produce small organic molecules or oligomers[5,6]. In this process, the reactivity of the depolymerization reaction, as well as the purity and yield of the products, are key parameters to consider. Therefore, polymers that can control reaction selectivity within the polymer backbone have been a primary consideration. The most representative polymer that used for chemical recycling via depolymerization is poly(ethylene terephthalate) (PET) given the regular ester-linkage distributions over the polymer backbone[25–27]. Depending on the type of the nucleophiles, PET totally depolymerizes to terephthalic acid derivatives[28–30], for example, terephthalic acid (TA), bis(hydroxylethylene) terephthalamide (BHETA), dimethyl terephthalate (DMT), and bis(hydroxylethylene) terephthalate (BHET) (**Figure 2a**). Among them, glycolysis is the most prevalent chemical recycling approach for PET, first patented in 1965[26,28]. While there are a number of candidates for glycolysis including propylene glycol, 1,4-butanediol, and pentaerythritol, ethylene glycol (EG) has been the most widely used[31,32]. This is because the depolymerization product, BHET, can undergo self-polymerization to PET under the specific heat and low pressure[33,34]. In contrast, the other glycolysis products need additional monomeric part, for example, ethylene glycol[30]. Therefore, extensive researches for the glycolysis of PET to produce BHET have focused on lowering temperature, improving reaction rate, and increasing purity of BHET. Examples include solvent/supercritical fluid assisted-, microwave assisted-, and catalysts assisted- glycolysis[26,34–39]. However, many of these examples require a large amount of wasteful/expensive solvents, damand high-energy facilities, use toxic/heavy metal catalysts, require post-purification steps, and/or would likely experience significant technological hurdles with respect to sustainability and scalability. Hydrolysis, aminolysis, and methanolysis of PET are the other well-recognized chemical recycling methods for PET[29,32,40–44]. However, problems associated with use of strong acids or bases, production of many byproducts, and slow reaction rate remain challenges for practical applications.
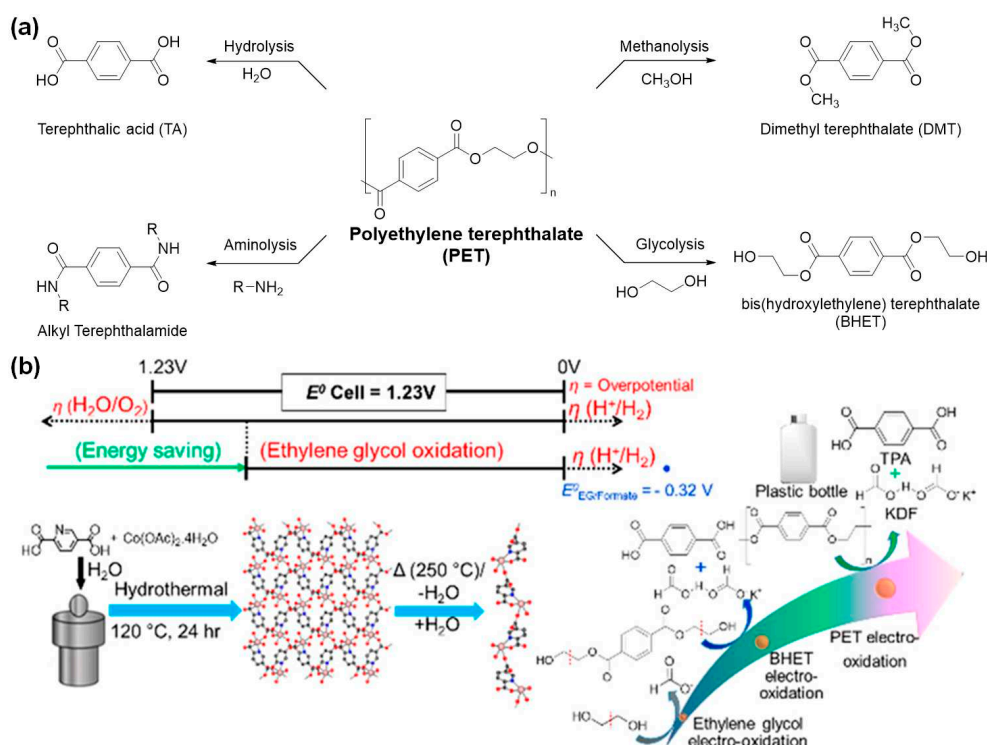
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**Figure 2. Chemical recycling methods of PET.** (a) Traditional depolymerization pathways of PET and depolymerized products. (b) depolymerization of PET by electro-catalytic reaction[45]. Reproduced with permission. Copyright 2023, American Chemical Society (ACS).

More sustainable and cost-effective process for plastic wastes including PET have been developed to address problems associated with traditional depolymerization processes[46–49]. The most representative example is the depolymerization via electrochemical reaction[45,50–53]. Recently, Behera et al. have demonstrated an electrochemical upcycling process for PET waste (**Figure 2b**)[45]. Hybrid water electrolysis was adopted to drive the water splitting to generate hydrogen gas at the cathode, while the water oxidation at the anode was replaced by oxidation of PET hydrolysate that contains EG monomer and BHET oligomer. During the electrolysis process, value-added products such as TA and potassium diformate (KDF) are generated along with hydrogen gas under the presence of a cobalt-based one-dimensional (1D) coordination polymer as an electrocatalyst. The polymer catalyst can efficiently perform the oxidation of PET hydrolysate at a low onset potential of 1.27 V (*vs*. reversible hydrogen electrode (RHE)), which is 160 mV less than the alkaline oxygen evolution reaction under the same conditions. Furthermore, the resulting TA and KDF products can be easily separated with a high yield from the electrolyte solution by simple pH adjustment. Thus, the electrochemical PET upcycling process allows full recyclability of the PET waste with a less energy-intensive strategy. Zhou et al. reported that $CoNi_{0.25}P$ electrocatalyst in KOH electrolyte enhances the selectivity and reactivity upon electrochemical PET depolymerization, which is attributed to easier EG oxidation with low-crystalline CoNi oxyhydroxide, thereby producing highly pure $H_2$, TA, and KDF. Another benefits of the electro-catalyst are found to be high current density (~ 350mA cm$^{-2}$ at 1.7 V *vs*. RHE) and high Faradic efficiency and selectivity (> 80%)[52].

Beside PET, depolymerization of polyolefins including polyethylene (PE) and polypropylene (PP) has received considerable attentions given that they constitute the largest portion of the plastic waste, approximately over 40% of all plastic wastes[54–58]. Since they composed of only hydrogen and carbon, common depolymerization chemistries (i.e., addition of nucleophile, strong acid or base) are not applicable. Therefore, developing efficient and highly selective depolymerization chemistries has been the major challenge. Duan et al. have proposed an efficient strategy for low-temperature and noble-metal-free catalytic depolymerization of PE into $C_3$–$C_6$ olefins (**Figure 3a**)[58]. By mixing PE with rationally designed ZSM-5 zeolite nanosheets at 280 °C in flowing hydrogen as a carrier gas, light hydrocarbons ($C_1$–$C_7$) were produced with a yield of up to 74.6%, where 83.9% of these products

were $C_3$–$C_6$ olefins with almost undetectable coke formation. The key technique is to use the ZSM-5 zeolite with a controlled *b*-axis direction as a catalyst in PE depolymerization, where the ZSM-5 kinetically matches the cascade cracking steps on its external surface and within the micropores by boosting the intermediate diffusion. Compared with the mechanical recycling of thermoplastics, which is mostly considered as downcycling from the resulting material property perspective, the catalytic PE depolymerization modulated by ZSM-5 zeolite provides new opportunities for industrially viable treatment of PE wastes to produce value-added materials.
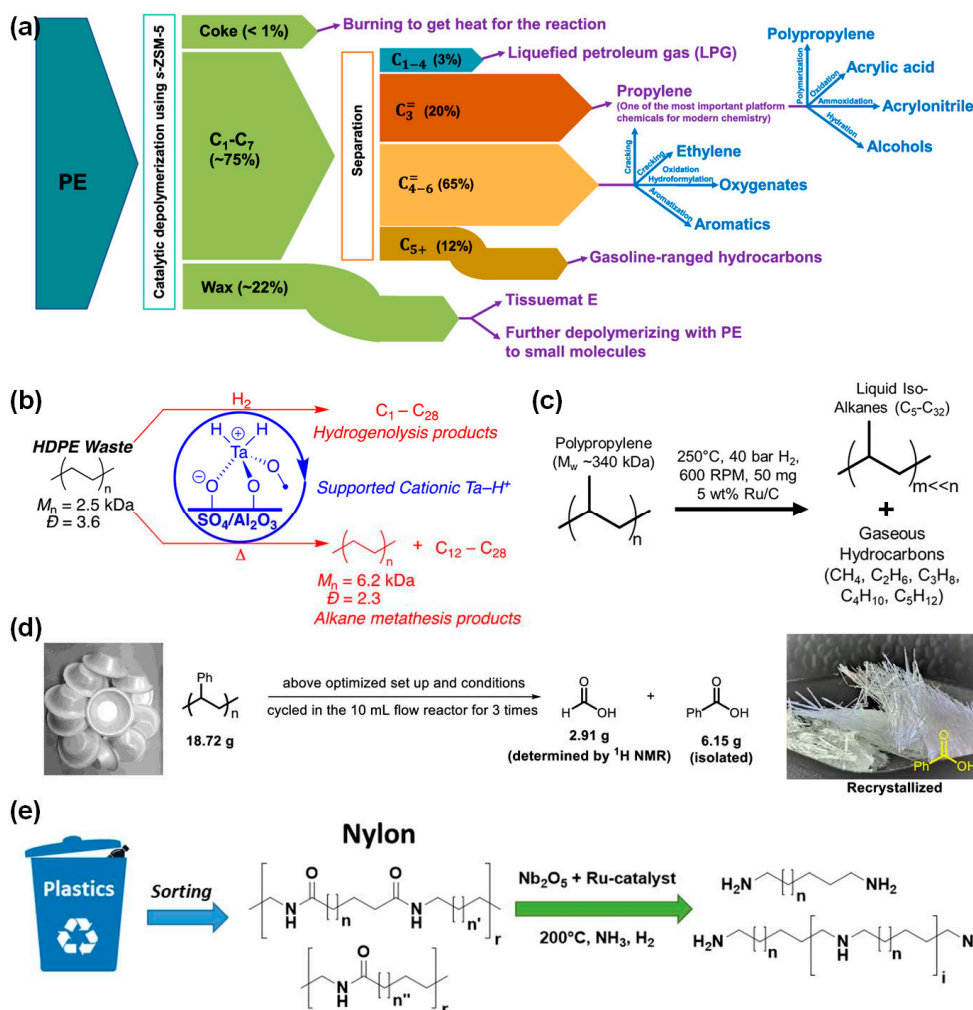


**Figure 3. Chemical recycling via depolymerization of commodity plastics to produce valuable small organic molecules.** (a) Schematic illustration of the carbon flow upon the s-ZSM-5 catalyzed depolymerization of PE and possible utilization of the products[58]. Copyright 2022, American Chemical Society (ACS). (b) Hydrogenolysis of a generic alkane Ta–H$^+$ supported on sulfated aluminum oxide to convert high density PE (HDPE) to low molecular weight hydrocarbons[55]. Copyright 2023, American Chemical Society (ACS). (c) Hydrogenolysis of high molecular weight PP ($M_w$ ~ 340 kDa) over ruthenium nanoparticles (5 wt %) supported on carbon.[54] Copyright 2021, American Chemical Society (ACS). (d) Depolymerization of PS in photo-catalyzed flow process yielding highly pure formic acid and benzoic acid[59]. Copyright 2022, American Chemical Society (ACS). (e) Chemical recycling of polyamide via catalyzed ammonolytic hydrogenation[60]. Copyright 2022, American Chemical Society (ACS). All the figures are reproduced with permission.

Gao et al. have developed a cationic d$^0$ metal hydride (M–H$^+$) species supported on sulfated aluminum oxide (SAO) that exhibits increased catalytic activity in hydrogenolysis and alkane metathesis reactions (**Figure 3b**)[55]. It was found that the cationic tantalum hydrides (Ta(=CH$^t$Bu)(CH$_2$$^t$Bu)$_3$–H$^+$) sites supported on the SAO are more active in hydrogenolysis and alkane

metathesis reactions than the neutral Ta–H sites supported on silica. This reaction chemistry extends to melts of high-density PE (HDPE), where Ta–H$^+$ converts 30% of a low molecular weight HDPE ($M_n$ = 2.5 kg mol$^{-1}$, $Đ$ = 3.6) to low molecular weight paraffins at moderate temperature, 150 °C, under hydrogenolysis conditions. Under alkane metathesis conditions, Ta–H$^+$ converts the HDPE to a high molecular weight fraction ($M_n$ = 6.2 kg mol$^{-1}$, $Đ$ = 2.3) and low molecular weight alkane products ($C_{13}$–$C_{32}$). This strategy demonstrates that incorporating charge as a design element in supported d$^0$ metal hydrides is viable to catalyze the reactions involving the reorganization of C–C bonds in PE wastes. In their follow-up study, they expanded the Ta-H$^+$ sites supported on SAO catalytic system for the degradation of PP[61]. In this case, low molecular weight branched alkanes with a narrower distribution ($C_{11}$–$C_{30}$) were produced in good yield (~70%).

While PP has similar chemical structure with PE, hydrogenolysis activity of PP is unclear. This is because PP has methine carbon in the repeating unit, higher $T_m$ than PE, and different crystallinity from PE[7]. Therefore, finding an optimized condition for hydrogenolysis of PP is a critical issue. Rorrer et al. have demonstrated a catalytic depolymerization of high molecular weight PP ($M_w$ ~ 340,000 Da) to liquid and gaseous hydrocarbons over ruthenium nano-particles supported on carbon (Ru/C) catalyst under mild conditions (200–250 °C, 20–50 bar H$_2$)[54]. Unlike PE hydrogenolysis, which produces linear *n*-alkanes, PP hydrogenolysis produces a range of iso-alkanes in both the liquid ($C_5$–$C_{32}$) and gas ($C_1$–$C_5$) with yields above 68% in the absence of solvent. Moreover, the Ru/C catalyst was effective in depolymerizing a mixture of HDPE and PP to produce a mixture of linear and branched liquid alkanes, demonstrating feasibility for the depolymerization of streams of mixed polyolefin waste.

In case of polystyrene (PS), all atoms are connected by strong C–C and C–H bonds, PS is remarkably inert and difficult to degrade under mild conditions[59,62–64]. Generally, thermal and catalytic pyrolysis of PS requires high temperature (typically > 300 °C) and expensive catalysts, which limits its practical applications[64–66]. To address the issues of the chemical recycling of PS wastes, Huang et al. have proposed light-driven and acid-catalyzed aerobic oxidation for the chemical recycling of PS to valuable chemicals under ambient temperature and 1 bar of O$_2$ (**Figure 3d**)[59]. Electron paramagnetic resonance (EPR) and density functional theory (DFT) calculations indicate that singlet oxygen ($^1O_2$) is involved as the reactive oxygen species in the degradation process, which abstracts a hydrogen atom from a tertiary benzylic C–H bond in PS, leading to hydroperoxidation and subsequent C–C bond cracking via a radical process. Furthermore, an adduct of PS-acid catalyst is formed *in situ*, acting as a photosensitizer to initiate the formation of singlet oxygen. This simple selective degradation method enables the oxidative cleavage of PS to yield valuable chemicals including benzoic acid, formic acid, and acetophenone under mild conditions. More recently, Rabot et al. converted PS to fungal secondary metabolites (SMs) such as ergothioneine, pleuromutilin, and mutilin by coupling catalytic oxidation with genetic engineering[67]. An oxidative catalytic method degraded PS, rapidly producing distributions of diacids, and these acids are suitable for fungal metabolism. Then, pharmacologically active SMs were produced through the engineering strains of fungi. This result implies a possible pathway for consuming upcycled plastics on an agrochemical scale.

Although polyamides (PAs) can be recycled both chemically and physically, physical recycling generally leads to plastics of lower quality due to impurities and reduced mechanical strength[60,68]. Chemical recycling, on the other hand, enables the valorization of waste into high-value building blocks for the production of virgin PA[69,70]. However, the resonance in the amide group makes PAs much more stable than polyesters, and the strong internal hydrogen bonds in PAs impede their dissolution in common organic solvents. Thus, most PA depolymerization processes often require harsh reaction conditions such as high temperatures (> 250 °C) and strong acidic or basic conditions. Coeck et al. have developed a strategy using ammonolytic hydrogenation of secondary amides for efficient chemical recycling of long-chain PAs (LCPAs) (**Figure 3e**)[60]. They have demonstrated a heterogeneous catalytic system based on Nb$_2$O$_5$, where the reactions were performed at a relatively low temperature of 200 °C in cyclopentyl methyl ether (CPME) as a green solvent and with limited addition of NH$_3$ and H$_2$. Since ammonolysis is an equilibrium reaction, it was eventually coupled to

a hydrogenation process, with the addition of a RuWO$_x$/MgAl$_2$O$_4$ hydrogenation catalyst, in order to achieve complete depolymerization. Industrial LCPA samples were successfully and completely depolymerized to $\alpha,\omega$-diamine monomers, and oligomers, resulting in product distributions of 62% primary amines and 36% secondary amines. Furthermore, the catalytic system was proven to be very robust against a variety of contaminants, e.g., fillers, other plastics, and additives.

Bis-phenol A based polycarbonate (BPA-PC) has been a widely used polymer with environmental concerns given that its traditional synthesis relies on toxic reagents (i.e., BPA, phosgene, or carbon monoxide)[71–73]. Therefore, preventing the release of BPA-PC to the environment after disposal is one of the critical concerns. Jehanno et al. proposed organo-catalytic depolymerization approach using the thermally stable and recyclable triazabicyclodecene(TBD)/methanesulfonic acid(MSA) co-catalysts[71]. Relatively low temperature (90 – 160 °C) transesterification produced high-quality six-membered cyclic carbonates with excellent yields due to the thermodynamic stability of the cyclic carbonate structures. This catalytic system is applicable to the complete depolymerization for a large amount of polymers, and can be regenerated and reused for multiple cycles. Upon the depolymerization cycles, they found that steric size of substituents on the six-membered cyclic carbonate determines their ability to ring-open as a consequence of the polymer chain torsion rather than the cyclic monomer conformation.

Biodegradable polyesters such as polylactic acid (PLA) are promising alternatives to conventional plastics[74]. However, complete biodegradation usually takes a few months or even longer and their biodegradation leads to CO$_2$ emissions[74–76]. By addressing these issues as well as producing value-added chemicals, Sun et al. proposed a two-step catalytic process to convert polylactic acid waste into methyl methacrylate, a high-value building-block molecule[77]. Polylactic acid is firstly transformed into methyl propionate (MP) with near-quantitative yields (conversion > 99%, selectivity~98%) over the $\alpha$-MoC catalyst in methanol solution, followed by reacting the obtained MP with formaldehyde to produce methyl methacrylate with a conversion of 81% and selectivity of 90%. Most importantly, the entire transformation process does not require any external high-pressure H$_2$, which adds sustainability benefit to this chemical upcycling process.

## 3. Chemical Recycling for Closed-loop Cycles: Polymers to Monomers, and/or to Polymers

Chemical recycling that converts polymers to small organics can address plastic waste concerns, however, economical imbalance between polymer and depolymerization products, needs for continuous fossil feedstock, extra sorting process, and ending up with greenhouse gas emissions remain concerns. The development of a circular plastics economy has emerged as a critical goal in addressing the concerns. A key aspect of achieving this goal is the concept of chemical recycling polymers back into their constituent monomers, a process referred to as chemical recycling to monomer (CRM)[78,79]. Theoretically, all the polymerizations are reversible under a certain condition such as temperature, pressure, and concentration. However, CRM process for the most common polymers such as PE, PP, PS, and PVC is practically hard or impossible. The ceiling temperature ($T_c$) serves as a thermodynamic measure of polymerizability or depolymerizability[80–82]. It represents the temperature where polymerization and depolymerization. Most of common polymers have high $T_c$ values that are above the decomposition temperature, therefore, rendering CRM process is hardly achievable (**Table 2**)[83]. Therefore, designing intrinsically circular polymers is a critical challenge. Balancing the requirements of polymerizability, depolymerizability, recyclability, and material performance often involves complex trade-offs: achieving this balance hinges on meeting thermodynamic, kinetic, and practical capability. Currently, extensive research efforts have focused on developing polymers originating from moderately strained heterocyclic monomer. These monomers exhibit relatively high thermodynamic stability, which in turn moderate their $T_c$ (< 250 °C) (**Figure 4a**).

**Table 2.** Ceiling temperature of common polymers.

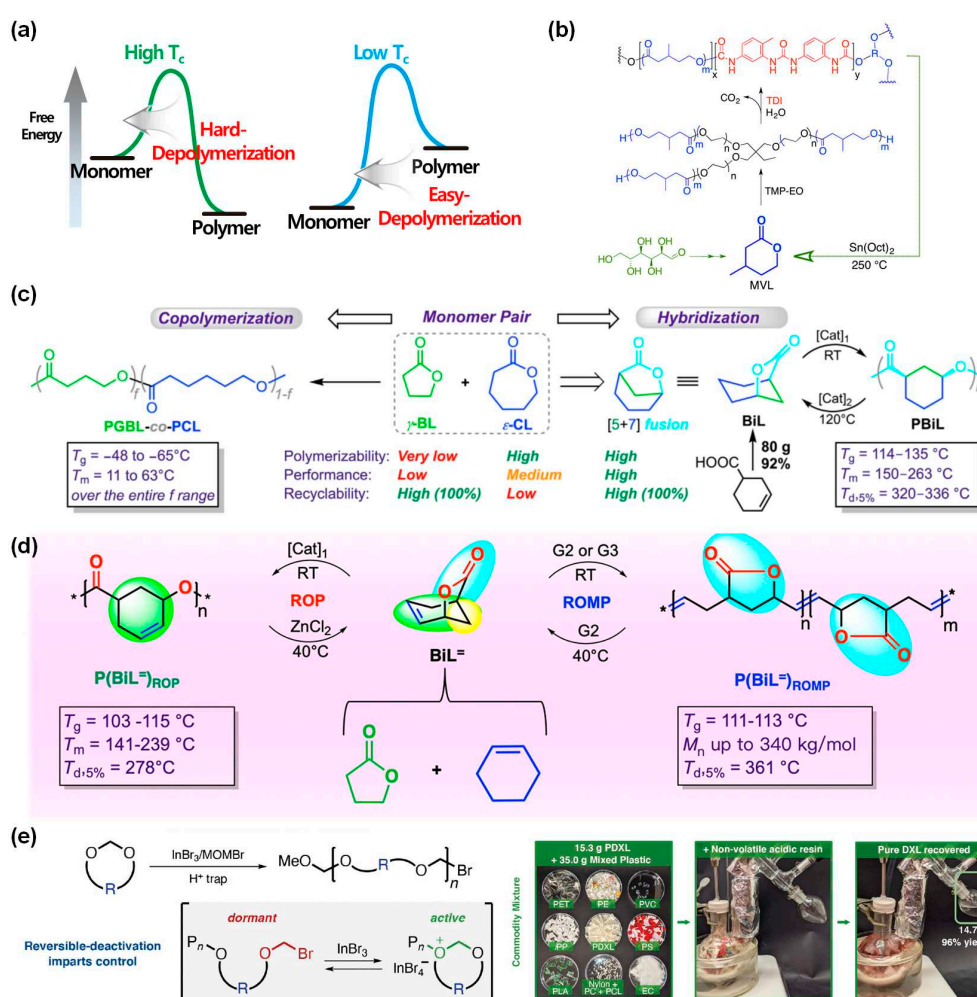| Monomer | Polymer | $T_c$ (°C) |
|---|---|---|
| Ethylene | Polyethylene | 610 |
| Isoprene | Polyisoprene | 466 |
| 1,3-butadiene | Polybutadiene | 585 |
| Methyl methacrylate | Polymethyl methacrylate | 198 |
| Styrene | Polystyrene | 395 |
| Tetrafluoroethylene | Polytetrafluoroethylene (Teflon™) | 1100 |



**Figure 4. Chemical recycling systems for closed-loop circular polymers.** (a) Thermodynamic preferences for depolymerization to monomers based on the free energy differences between monomers and polymers. (b) Synthesis of P$\beta$MVL polyol, PU foam, and chemical recycling to $\beta$MVL from the crosslinked PU foam[84]. Copyright 2016, American Chemical Society (ACS). (c) Hybrid 5-/7- membered lactone system for balancing polymerizability and recyclability[85]. Copyright 2021, Elsevier Inc. (d) "One Monomer–Two Polymers–One Monomer" strategy by hybrid lactone/olefin monomers[86]. Copyright 2022, American Chemical Society (ACS). (e) Closed-loop chemical recycling of cyclic acetals and monomer recovery from a mixed commodity plastics feedstock[87]. Copyright 2021, American Association for the Advancement of Science (AAAS). All the figures are reproduced with permission.

Among several families of chemically recyclable polymers that have recently developed, polyesters derived from the ring-opening polymerization (ROP) of lactones are particularly

attractive, due to their degradability and ROP reversibility[88–91]. These features of polyester derivatives provide an efficient strategy to revert the polyesters back to their monomer lactones by carefully manipulating the thermodynamics (such as temperature, concentration, and pressure) of the (de)polymerization, as well as the kinetics and processing conditions. For example, Schneiderman et al. investigated thermodynamic parameters of $n$-Alkyl-$\delta$-valerolactone monomers[92]. All substituted $\delta$-valactones with arbitrarily long $n$-alkyl substituents showed thermodynamic- and kinetic- feasibility for polymerization. Among them, $\beta$-methyl-$\delta$-valactone ($\beta$MVL) exhibited the lowest $T_c$. In their following study, poly($\beta$-methyl-$\delta$-valactone) (P$\beta$MVL) with trihydroxyl end groups has been synthesized, and used as a polyol for the preparation polyurethane (PU) foam (**Figure 4b**)[84]. In spite of the crosslinked structure of the PU foam, the monomer (i.e., $\beta$-methyl-$\delta$-valactone) was easily recovered through a simple distillation set-up given the high tendency of depolymerization. This is beneficial because it does not require the addition of any solvent, alcohol, or water like glycolysis of PET and PU. Other examples for depolymerizable polyester derivatives include poly(2-thiabicyclo[2.2.1]heptan-3-one), polyhydroxyalkanoates (PHA), cyclic strained poly($\gamma$-butyrolactone), PLA, and poly(norcamphor lactone)[82,93–98].

For more controllable $\delta$-valerolactone system, Eugene and co-workers have presented a synergistic use of gem-$\alpha$,$\alpha$-disubstitution of available at-scale, bio-based $\delta$-valerolactones to yield gem-dialkyl-substituted six-membered lactones[99] . It was found that the $\alpha$,$\alpha$-disubstitution of valerolactones monomers (VL$^{R_2}$) exhibits much lower thermodynamic polymerizability-or higher depolymerizability-relative to the parent $\delta$VL, leading to greatly reduced $T_c$ upon disubstitution. Note that the $T_c$ values of the PVL and PVL$^{Pr2}$ (Pr: propyl) were found to be 298 °C and 67 °C, respectively. This is primarily due to a more negative entropic change of VL$^{R_2}$, primarily ascribed from a more pronounced loss in conformational (rotational and vibrational) freedom ($\Delta S_c$). Thus, the VL$^{R_2}$ allows both the facile polymerization of high-molecular-weight polyesters (PVL$^{R_2}$, $M_n$ up to 333 kg mol$^{-1}$, Đ = 1.16) under ambient conditions and selective depolymerization to fully recover VL$^{R_2}$ monomers under mild conditions (150 °C with a ZnCl$_2$ catalyst). The recovered monomers can be repolymerized to regenerate the same polymers, thereby closing the monomer-polymer-monomer circular loop. Besides their full chemical recyclability, these polyesters also exhibited better tensile properties, high crystallinity ($T_m$ up to 140 °C), thermal stability ($T_d$ up to 330 °C), and oxygen barrier properties compared to the parent PVL, proving that the PVL$^{R_2}$ are good candidates for replacing low density PE (LDPE) in packaging applications. Exploiting the thermodynamic stability of multi-substituted lactones, they synthesized $\alpha$,$\alpha$-disubstited PHA in their following work[100]. The straightforward $\alpha$,$\alpha$-disubstitution in PHAs significantly enhanced the thermal stability, making PHAs amenable to melt processing. Simultaneously, this structural modification imparts mechanical toughness, intrinsic crystallinity, and the potential for closed-loop chemical recycling to PHAs.

Meanwhile, the design of such circular polymers is challenged by unyielding trade-offs between the monomer's polymerizability and the polymers' depolymerizability. For example, the ROP of non-strained five-membered cyclic lactone monomers leads to polylactone that can be completely depolymerized back to pure monomers with low energy input[101–103]. However, due to the high stability of monomers, polymerizability and the mechanical properties are very limited. Eugene and co-workers reported the synthesis of the copolymer, i.e., poly(glycolide-*co*-$\gamma$-butyrolactone), through the transesterification at high temperature (160 °C), however, the maximum $\gamma$-butyrolactone content in the polymer is only ~20%[104]. To address this trade-off issue, they have introduced a hybrid monomer design strategy in their following work[80,85]. They demonstrated synergistical coupling of a high $T_c$ $\varepsilon$-caprolactone sub-structure for high polymerizability and performance properties with a low $T_c$ $\gamma$-butyrolactone sub-structure for high depolymerizability and recyclability within the same monomer structure (**Figure 4c**). Thus, structural hybridization between two distinct sub-structures leads to an offspring [3.2.1]bicyclic lactone, which exhibits both high polymerizability and depolymerizability, otherwise conflicting properties in a single monomer structure. Furthermore, the resulting polymer became a high-performance material, and thermal transition temperatures are 200 °C higher and tensile modulus 10 times higher than its parent polymers. These results demonstrate

that the hybrid monomer design strategy is a powerful approach for designing circular polymers where conflicting properties must be exploited and unified.

In a similar manner, Shi et al. further developed the monomer structural hybridization strategy toward a "one monomer-two polymers-one monomer" loop via orthogonal (de)polymerization of a lactone/olefin hybrid monomer (**Figure 4d**)[86]. Two well-known low $T_c$ polymers, $\gamma$-butyrolactone ($\gamma$-BL) toward ROP to polyester and cyclohexene toward ring-opening metathesis polymerization (ROMP) to poly(cyclic olefin), are notoriously non-polymerizable. The authors have demonstrated a strategy to render not only the polymerizability of both the $\gamma$-BL and cyclohexene sites, orthogonally, but also complete and orthogonal depolymerization, through creating a bicyclic lactone/olefin (BiL$^=$) hybrid. The produced polyester, P-(BiL$^=$)$_{ROP}$, by ROP exhibited a high $T_m$ of 141–239 °C and $T_g$ of 103–115 °C, while the poly(cyclic olefin), P-(BiL$^=$)$_{ROMP}$, by ROMP has a $T_g$ of 113 °C. Despite their excellent thermomechanical properties, both P-(BiL$^=$)$_{ROP}$ and P-(BiL$^=$)$_{ROMP}$ showed full chemical recyclability to regenerate the same monomer under mild conditions (25-40 °C) in the presence of a catalyst. These findings demonstrated that orthogonal polymerization of a bifunctional monomer offers an attractive strategy to produce two different classes of polymers from a single monomer, but a more significant, modern challenge is to additionally enable both types of the orthogonally produced polymers with orthogonal depolymerizability to recover the same monomer.

Besides polylactone derivatives, polyacetals are promising candidates for chemical recycling to monomer, but lack useful tensile strengths due to the low molecular weights produced using current uncontrolled cationic ROP (CROP) methods[70,105,106]. Abel et al. have presented reversible-deactivation CROP (RD-CROP) of cyclic acetals using a commercial halomethyl ether initiator and an indium bromide catalyst (**Figure 4e**)[87]. The RD-CROP method affords molecular weight control and living chain-end retention for the polymerization of cyclic acetal monomers, enabling the synthesis of high molecular weight polymers. By adopting this RD-CROP, they have synthesized poly(1,3-dioxolane) (PDXL), which exhibited tensile strength comparable to some commodity polyolefins. Furthermore, depolymerization of PDXL using strong acid catalysts returns monomer in near-quantitative yield and even proceeds from a commodity plastic waste mixture. This result demonstrated that the RD-CROP method affords a tough thermoplastic that can undergo selective depolymerization to monomers. The highest molecular weight of PDXL is ~220 kDa high in this study, however, its properties could be further improved by reaching to the ultrahigh molecular weight (UHMW). In the following study, Hester et al. reported the synthesis of UHMW PDXL by employing 2,6-di-*tert*-butylpyridine (DTBP) as a proton trap agent and [Et$_3$O$^+$][BF$_4^-$] as an initiator[107]. In general, protic impurities cause a several unwanted reactions such as initiation of new polymer chain, protonation of an existing chain, reaction with polymerization counter anion. This results in macrocyclization, transacetalization, and degradation of counter anion. A small amount of DTBP scavenges protic impurities upon the polymerization, making the polymerization controllable and predictable. Ultra-low loading of initiator is another key factor to achieve UHMW. Triethyl oxonium salts, also known as Meerwein salts[108], which are composed of an oxonium cation and a noncoordinating counterion such as BF$_4^-$, PF$_6^-$, and SbF$_6^-$ found to be effective and robust. With them, the molecular weight of PDXL was over 1000 kDa, and the resultant polymer exhibited competitive mechanical properties to the commodity polymers such as UHMWPE and HDPE.

As presented above, there are a number of examples that can construct CRM process, however, most examples are limited within polymers synthesized from cyclic monomer containing heteroatoms. Other common polymers such as polyolefins, polyesters from low stability monomers, thermosets are not suitable for CRM process due to the thermodynamic reasons[109,110]. Therefore, there is an additional research requirement to develop chemical recycling system by chain scission/reconstruction process. There have been extensive research efforts for developing the chemical recycling system by adding key materials or chemistries into the reprocessing periods.

The primary reason that depolymerization and/or degradation of conventional polyolefin materials normally happens at high temperatures of 350~800 °C is primarily due to the inherently large bond strength of C–C bonds with standard lengths of 1.53~1.54 Å[111,112]. To address this challenge, Luo et al. demonstrated a topochemical approach for creating reversible elongated C–C

bonds with a bond length of 1.57~1.63 Å between repeating units in the solid state with decreased bond dissociation energies (**Figure 5a**)[113]. In solid-state topochemical polymerization, the condensed-phase interactions of the crystal lattice favorably position monomer molecules to react. In this way, adding extra steric hindrance to fine-tune the C–C bond length is possible without inhibiting polymerization. The resulting polymers exhibited rapid depolymerization via breakage of the elongated bond within a desirable temperature range of 140~260 °C while otherwise remaining remarkably stable under harsh conditions. Furthermore, the topochemical polymerization produces high-quality stereo-regular and ultra-high-molecular-weight crystalline polymers. Thus, this strategy potentially enables polymer systems that perform similarly to traditional hydrocarbon-based polymers while also easily depolymerizing to monomers through thermal cleavage of the relatively weak C–C bonds. In addition, the resulting polymers can be further processed to useful forms using techniques applied to other conventional polymers such as molding, extrusion, and 3D printing.
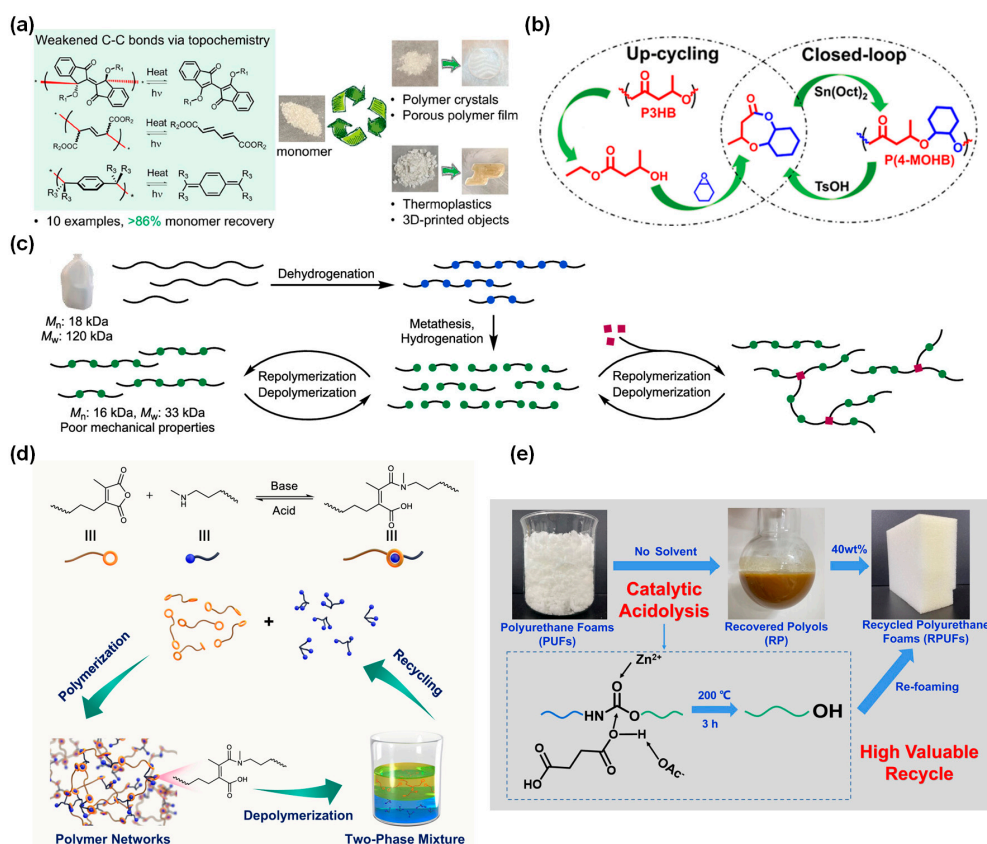


**Figure 5. Chemical recycling via chain scission/reconstruction by adding key chemicals and/or chemistries.** (a) Circularly recyclable polymers by topochemically weakened C−C bonds[113]. Copyright 2022, American Chemical Society (ACS). (b) Chemical upcycling of P3HB toward the bicyclic monomer 4-MOHB, which can be polymerized to afford P(4-MOHB) with a closed-loop cycle[114]. Copyright 2022, American Chemical Society (ACS). (c) Chemical recycling of post-consumer waste HDPE into a chemically recyclable material[115]. Copyright 2022, American Chemical Society (ACS). (d) Preparation of polymer networks from bifunctional maleic anhydrides and multifunctional secondary amines, their depolymerization in acid aqueous solution, and circularity[116]. Copyright 2022, Springer Nature. (e) Chemical recycling of PUFs by catalytic acidolysis process[117]. Copyright 2023, American Chemical Society (ACS). All the figures are reproduced with permission.

Despite the successful examples of developing chemically recyclable synthetic polymers, studies regarding the upcycling of biopolymers into new degradable polymers are rare. Poly(3-hydroxybutyrate) (P3HB), as one type of PHAs, is a complete biopolymer that is generally produced

via microbial fermentation[118,119]. Although P3HB takes several months to fully degrade into $CO_2$ and $H_2O$, chemical upcycling of P3HB is required given that the annual output of PHAs has been increasing in recent years[120]. However, chemical upcycling of P3HB is not easy because simple thermal pyrolysis usually produces undesired products, such as low molecular weight oligomers with ketene, propylene, acetaldehyde, and $CO_2$. In this context, Li et al. have proposed a chemical upcycling of poly(3-hydroxybutyrate) (P3HB) into bicyclic ether-ester monomers toward value-added, degradable, and recyclable poly(ether ester)[114]. The upcycling of P3HB involved four steps: efficient conversion of P3HB into ethyl 3-hydroxybutyrate (HBEt), selective ring-opening with cyclohexene oxide (CHO), cyclization to form the new bicyclic monomer 4-methyloctahydro-2*H*-benzo[*b*][1,4]dioxepin-2-one (4-MOHB), and its ROP. Thermal analysis revealed that the resulting polymers exhibited an amorphous state with moderate $T_g$ and great thermal stability. Most importantly, the polymers can be efficiently recycled back to monomers through solution depolymerization catalyzed by TsOH or pyrolysis using $Sn(Oct)_2$ or $ZnCl_2$ as a catalyst. This work provides a feasible approach for the chemical upcycling of P3HB bioplastics to produce new recyclable polymers with a closed-loop life cycle.

Arroyave et al. have reported a chemical recycling route for the conversion of post-consumer HDPE into telechelic macromonomers suitable for circular reprocessing (**Figure 5c**)[115]. Unsaturated C-C bond was introduced into HDPE waste by catalytic dehydrogenation using an Ir-POCOP catalyst. The dehydrogenated HDPE waste undergoes cross-metathesis to form telechelic macromonomers with molecular weights dependent on the degree of unsaturation. While the direct re-polymerization of the macromonomers yields a brittle material primarily due to the low molecular weight, aminolysis of telechelic macromonomers with a small amount of diethanolamine increased the overall mechanical properties. After hydrogenation and partial aminolysis, these macromonomers can be polymerized to yield a material with comparable mechanical and thermal properties to HDPE. This strategy demonstrated that the incorporation of main-chain ester functionalities in PE enables the chemical recycling of PE waste and envision that this methodology will offer a pathway to functionalize and recycle PE waste.

Reversible bond reorganization of various covalent bonds has enabled the regeneration of polymers, which are inherently depolymerizable to form pristine monomers under certain conditions[121–125]. However, the breakage of covalent bonds within polymeric networks often requires an energy-intensive process and undergoes side reactions, resulting in mixed products rather than clean reusable monomers. Thus, it remains a major challenge to design dynamic bonds, capable of effective bonding and reversible cleaving, for preparing chemically recyclable cross-linked polymers. To resolve this issue, Qin et al. have developed chemically recyclable cross-linked polyamic acid materials through the incorporation of dynamic maleic acid tertiary amide bonds (**Figure 5d**)[116]. The dynamic maleic acid tertiary amide bond is based upon a reversible amidation reaction between maleic anhydrides and secondary amines, enabling reversible chemical control over depolymerization of the resulting polymer networks. This dynamic bond allows for the construction of polymer networks with tailorable and robust mechanical properties, covering strong elastomers with a tensile strength of 22.3 MPa and rigid plastics with a yield strength of 38.3 MPa. Furthermore, these robust polymeric materials can be completely depolymerized in an acidic aqueous solution at ambient temperature, leading to efficient monomer recovery with >94% separation yields. The recovered monomers can be used to remanufacture cross-linked polymeric materials without losing their original mechanical performance. This reversible amidation chemistry unveils a general approach to designing polymer networks with tunable mechanical performance and closed-loop recyclability.

PU is the sixth most produced polymer worldwide, with polyurethane foams (PUFs) accounting for about 67% of global polyurethane consumption[124,126,127]. However, existing methods of chemical recycling of PUF waste suffer from incomplete degradation of urethane groups and lack of selectivity for degradation of urethane bonds alone, resulting in differently functionalized polyols containing various side products. Grdadolnik et al. have addressed this issue by introducing microwave-assisted aminolysis with amine reagents that contain primary and tertiary amino groups

in the structure[117]. These reagents enable complete degradation of the urethane groups in the structure of the flexible PUFs with a much lower amount of degradation reagent than is typically required for solvolysis reactions. The purified, recovered polyols are close equivalents to the corresponding virgin polyols in terms of their structural and molar mass characteristics. Furthermore, the PUFs made from the recovered polyols have comparable mechanical properties to those made from virgin polyols.

## 4. Conclusion & Perspectives

This review has covered an up-to-date overview of chemical recycling strategies for addressing plastic wastes related concerns related to sustainability and practical feasibility. These include *i*) conventional hydrolytic degradation, *ii*) electrochemical depolymerization, *iii*) catalytic depolymerization of commodity plastics to produce valuable small organics, *iv*) thermodynamics modulation with stable heterocyclic compounds for molecular circularity, and *v*) oligomerization and re-polymerization via addition of junctional compounds. These methods not only enhance depolymerization efficiency but also simplify initial sorting procedures, generate more valuable products, and diminish our dependence on finite fossil resources. As we have reviewed, a key aspect found in some of the most important works in the field is the collaborative effort of a diverse group of scientists on the fundamental questions. While this review deals primarily with depolymerization technologies of polymeric materials, an essential overarching perspective is the effect of macromolecular science on plastic related environmental challenges.

As we have reviewed, there have been several critical challenges, opportunities, and emerging efforts are on the horizon. It still remains difficult to create perfect plastic recycling system totally addressing plastic wastes challenges within a practical manner. A primary example of this difficulty is the preparation of a readily monomer-recoverable polymer while concurrently exhibiting good polymerizability to achieve high molecular weight and mechanical robustness. Prioritizing factors or parameters is also challenging due to the complexity of kinetics, thermodynamics, economics, and materials properties. We believe that expanding polymer design principles into a more innovative methodology, exploring uncharted directions in the context of chemical recycling, will yield significant advancement in resolving the plastic wastes issues. The challenges are substantial, but with concerted efforts, innovative solutions, and a shared commitment to a more sustainable future, we can realize the vision of a world where plastics are not a burden but a valuable resource in continuous circulation.

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