Review

# Why is Recycling of Postconsumer Plastics so Challenging?

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**ABSTRACT:** The ubiquitous use of plastics has been driven by their combination of low cost and properties, but these attributes directly challenge waste management schemes for plastic recycling. Some postconsumer recycling programs are now nearly 50 years old, but a significant fraction of plastics still finds landfills or other dumping strategies at their end of life. With the growing concern regarding plastic waste, especially ocean plastics, there is a need for innovation and alternative strategies for the economic translation of plastic waste to valued product(s) that will promote their efficient circular utilization. This review first describes the technical and economic hurdles associated with the recycling of postconsumer plastics, but then it focuses on providing an overview of emergent strategies to recover plastic waste through new polymer design, new recycling processes, and chemical transformations to value-added products. Specific challenges discussed include plastic waste sorting and separations, product variability including additives, and the high efficiency/low cost in which the existing petrochemical industry can produce virgin polymers, in particular polyolefins. Although a wide variety of technical strategies have been demonstrated for recycling of plastics through both mechanical and chemical means, the commercial success of these different strategies is generally limited by either performance, including large variance in key metrics, or economics where the products can match the performance of virgin materials but the recycling process is expensive. Successful capture of postconsumer plastic waste through recycling likely will depend on economic incentives and government regulations.

**KEYWORDS:** sustainability, circular polymers, plastic waste, recycling, reprocessing
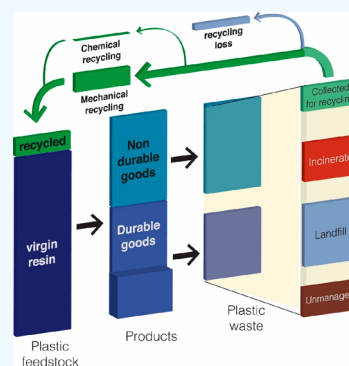
## 1. INTRODUCTION

The question of recycling and reuse of refuse is an old one with evidence of materials recycling back to the Paleolithic era.[1] In more modern times, the rates for recycling wax and wane with the political and economic climate.[2] In the first half of the 20th century, recycling rates correlated well with shortages and the associated push of wartime efficacies from 1914 to 1945, but without these factors, recycling rates fell even in the face of economic challenges.[2] The continued growth of plastic production[3,4] presents a significant challenge in waste (resource) management in the 21st century. The historical data for the recycling of plastics is bleak when considering all plastics produced from 1950 to 2015.[4] Of the 5.8 billion metric tons of plastic produced that is no longer in primary use, 5.7 billion metric tons of plastic have been discarded or incinerated. From more contemporary EPA data on municipal solid waste (MSW) generation in the US from 2018, 292.4 million tons of MSW was generated, of which plastics comprised 12.2%, but they only comprised 4.4% of the 69.0 million tons of MSW that was recycled.[5] This recycled plastic is predominately from two types of polymers: natural high density polyethylene (HDPE, white translucent) and poly-ethylene terephthalate (PET) with recycling rates of 29.3% and 26.8%, respectively. Packaging is the dominant industrial sector for annual plastic production (146 million tons in 2015) and waste generation (141 million tons in 2015) and has a short

product lifetime (mean = 0.5 yr).[4] Only a small fraction of this large potential material stream from packaging materials is recycled, despite governmental efforts to promote recycling.[6] Alternative routes to address the growing plastic packaging waste have included mandates banning some single use plastics[7−9] and larger multinational directives such as the "European Strategy for Plastics in a Circular Economy" through the European Commission.[10] However, the efficacy of limited bans on impacting environmental concerns about plastic waste has been questioned.[7,11,12] Improvements across the waste management chain from disposal practices, recycling technology, government regulations, and public incentives are likely necessary to address global challenges associated with plastic wastes.

The issues associated with plastic waste and the inefficiencies of its recycling have been recognized for some time. For example in 1990, three large German chemical companies (BASF, Bayer, and Hoechst) formed a joint venture to generate ideas on how to best address plastic waste
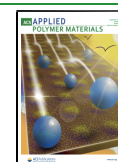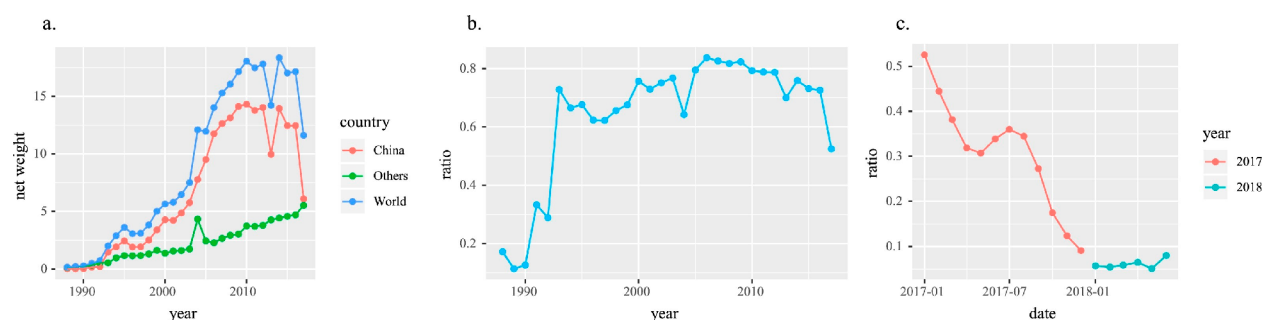
**Figure 1.** (a) Comparison among the world trade volume, the trade volume of China, and the trade volume of the rest of the world for plastic waste. (b) Ratio of China's trade volumes to the total world trade volume. (c) Ratio of China's trade volumes for the year 2017 and the period 2018/01−2018/06. Reproduced with permission from ref 14. Copyright 2020, Elsevier, B.V.

accumulation.[13] However, despite these and many other efforts, the total annual generation of plastic waste continues to grow,[4] and globally, the fraction of plastics that are recycled is not increasing. With much of plastic marked for recycling historically exported to Asia,[14] the import ban implemented by China in 2017 caused a decrease in the waste plastic import to Asia by 52 kilotons in 2018.[15] Figure 1 illustrates the global impact of this change on the plastic waste import ban. This ban has indirectly decreased the value of plastic waste due to lack of alternative waste treatment plants for the recycling of plastics[16] and led to changes in the plastics acceptable by some curbside recycling programs in the U.S. Following the lead of China, other major importers of plastic waste, primarily in southeast Asia, have limited plastic waste imports[15] to further strain the existing recycling infrastructure. With these added pressures on the recycling infrastructure and growing environmental concerns about plastic waste,[17] there have been significant efforts to change the paradigm from the typical downcycling of plastic waste via mechanical means to circular recycling where the material can be reused in the same application and upcycling to provide added functionality or performance during the recycling process.[18] Combined with growing concerns about plastic waste, in particular from single use plastics, these factors have provided an environment to increase industrial efforts in enhancing recycling as part of a cradle-to-cradle approach in materials management as a circular economy.[19] However, there has been some skepticism associated with these approaches that has been labeled by some as greenwashing as the same petrochemical based plastics are continued to be used and produced.[19] With pressure from environmental concerns, government regulations and limited infrastructure to handle the current waste stream, innovations are likely needed to efficiently and economically address the challenges of plastic waste.

In this review, we describe some of the recent innovations to address challenges associated with the recycling of plastics. The technical challenges, in general, have been previously identified in a review more than a decade old that focused on the challenges and opportunities of plastics recycling,[12] but changes in political, environmental, and economic factors have accelerated the need for effective management of plastic waste.[17,20−22] The focus of this review is on postconsumer recycling, which represents a much more challenging problem than reclamation of scrap from the manufacturing process due to the complexity of the mixed plastic waste stream. First, we describe plastic waste with a brief discussion of plastic waste streams in terms of its origins and composition by polymer type. The postconsumer waste stream has several layers of complexities associated with the formulation of commercial plastics that include a variety of additives and the increased engineering of plastic products to enhance properties through multilayer and composite materials. We provide a cursory description on how these attributes contribute to recycling challenges for plastic waste. This is manifested in the current state for the management of plastic waste at the end of product life. To understand why the recovery is limited, the primary method for recycling, mechanical recycling, is then described in detail with discussion of recent advances and the technoeconomic challenges of mechanical recycling. To address these challenges, chemical recycling approaches have been proposed to recycle the polymers as monomers to build virgin plastic resins from recycling, which in theory can be recycled infinitely, but there are significant technical and economic challenges to the realization of a fully circular materials economy for commodity plastics, in particular polyolefins. This review briefly touches on some concepts to redesign polymers from the monomer level to ease the recovery to monomer or fragments that can be repolymerized indefinitely for a truly circular materials economy. By relaxing the circular demands to alternative chemical products, the chemical transformation of waste plastics to value added chemicals is an attractive alternative to a fully circular economy for plastic waste. Finally, this review finishes with a summary of some of the key challenges and opportunities for advancement of the recycling of postconsumer plastics.

## 2. PLASTIC WASTE STREAMS

Plastics are ubiquitous in modern society, from durable goods to packaging to components in personal care products. The disposal of these plastics has been of growing concern based on environmental impact. Improper dumping has led to the proliferation of plastic garbage patches in the oceans,[23] while the breakdown of plastic (waste) as well as microbeads from personal care products present another hazard (microplastics),[24] which is going to necessitate additional considerations due to their presence even in remote locations (far from human activities).[25] Despite these environmental concerns, the plastic generated annually continues to grow as does the plastic waste.[26] This review focuses on the recycling of solid, postconsumer plastic waste and will not consider the additional challenges posed by microplastics[27] or energy recovery through incineration to recover value from plastics,[28] or efforts into plastics from renewable, biobased feedstocks.[29] Understanding the nature of the generated plastic waste stream is important to provide context to the challenges facing nascent technologies and strategies to enhance the recycling of plastics.

**2.1. Composition and Disposition of Postconsumer Plastic Wastes.** At the end of service life for plastic goods, their disposition as waste follows a variety of paths. In general, at this time, plastics tend to be separated into three streams: solid waste for landfilling/disposal, recycling, and incineration.[4] The quantity and characteristics of plastic waste are highly variable based on location and the socioeconomic status of the community in which it is generated,[30] which provides a challenge for its efficient reuse and recycling. In the U.S., municipal solid waste contains approximately 30 million tons of plastic, of which approximately 9% is recycled.[31] In the EU, approximately 26 million tons of plastic waste is generated with nearly 30% of the plastic recycled and 40% incinerated.[31,32] These differences between the U.S. and EU can be partially associated with higher landfill tipping fees leading to increased recycling rates as an economic driver. For example, the introduction of the volume-based fee system for waste in South Korea in 1995 led to significant cost increases for waste disposal and increased the recycling rate from 23.7% in 1995 to 59.1% in 2012.[33] In the U.S., local implantation of unit-pricing for curbside garbage collection increased the mass of recyclable materials by 16%.[34] However, in the U.S., tipping fees for landfilling waste have only increased from $53.04 to $55.11 per ton from 1994 to 2018.[35,36] Economics clearly influences the propensity of the local community to recycle plastics, but even when plastic waste is collected for recycling, there are additional technological challenges that provide limitations to the fraction of plastics that can be recycled.

Figure 2 illustrates the composition of plastic produced from 2002 to 2014.[4] The dominant plastics are polyolefins (HDPE,
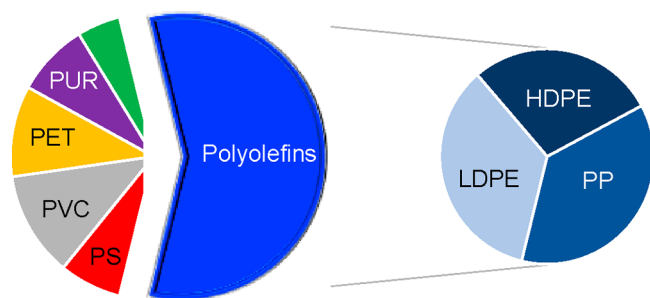


**Figure 2.** Composition of total polymer resin production from data for Europe, U.S., China, and India from 2002 to 2014 as tabulated in ref 4 (Table S2). More than 57% of the mass of plastic produced in this time period was polyolefins with the breakdown between LDPE (and LLDPE), HDPE and PP shown in the breakout. The green slice is all other polymers.

low density polyethylene [LDPE], and polypropylene [PP]), which account for 57.3% of the plastics produced. This production mirrors plastic waste streams, but depending on location, there have been reports of polyolefins comprising more than 80% of municipal plastic waste.[37] The combination of low cost and performance makes polyethylene and polypropylene ideal polymers for a wide variety of applications. In particular, the packaging industry utilizes polyolefins as primary polymers along with PET.[38] When considering plastic waste and recycling, packaging is a critical industry due to its ubiquity and low average lifetime for packaging (0.5 yr).[4] Thus, the development of more effective routes for the efficient recycling of polyolefins and PET will provide the greatest benefit for handling plastic waste, and these plastics should be

the focus of most research efforts associated with plastic recycling.

Figure 3A illustrates the outlier nature of the packaging industry in terms of its proportion of the plastic waste produced annually in comparison to other industrial sectors utilizing plastics. The significance of the packaging industry on plastic waste stream can be clearly inferred from Figure 3A with essentially all of the packaging materials produced transforming to waste when considering annual plastic production and waste generation. When considering plastic waste generation in 2015, nearly 47% of all of the plastic waste was a result of the packaging industry. For improving plastics sustainability, the packaging industry has been targeted in regulations and some corporate policies.[10] The packaging industry has responded to EU regulations with development of alliances and improving integration of the plastic value chain toward the goal of a circular economy.[10] The efforts for plastic recycling tend to be strongly coupled with regulations that influence the economics of recycling. This changing landscape can alter the viability of developed technologies for recycling.

When considering the global plastic economy, the annual input-output provides insight into the flow of plastic goods and their lifecycle. Figure 3B illustrates the worldwide plastic production and waste generation from 2016. Approximately 10% of the plastic feedstock was composed of recycled plastic (either mechanical or chemical). The mass of plastic recycled was comparable to that which was improperly disposed that is critical to the management of ocean plastics.[39] About twice as much plastic was incinerated than was actually recycled. Essentially all of the nondurable plastic produced ended up as plastic waste, while the waste stream of durable goods was significantly smaller than the mass of durable goods produced due to their extended lifetimes and growing plastic production. The limited fraction of the plastic waste stream that is recaptured offers significant opportunities for advancements in sustainable utilization of plastics. Focus on nondurable goods would have the most significant impact in the short term due to its short lifetime and its significant fraction of the total plastic.

**2.2. Complexity of Commercial Plastics.** Although a majority of the plastic waste is based on polyolefins, simply breaking down the waste stream on the basis of the polymer does not provide a full picture of the complexity of this situation. Almost all plastic utilized in commercial products contains a mixture of additives,[40] fillers,[41] pigments, and modifiers to provide processability for producing the product and the final properties critical to the consumer. The additives present depend on the application and the processing used in production,[42] but their details impact the ability to effectively recycle the plastic waste.[43] Hahladakis et al. have reviewed the release of additives from plastics through their lifecycle, including the recycling process.[40] The fate of these additives is another concern when recycling plastic waste,[44] beyond how the additives influence the processes.

Plastic packaging, which comprises a large share of plastic waste produced, has become highly engineered to most economically meet the properties demanded by the customer. One approach to enhance the properties is through the use of multilayer films, which also can challenge recycling.[45] For improvements in barrier properties, metal or inorganic deposited coatings on the plastics have also become common-place.[46] With improvements in deposition techniques, well-adhered nanocoatings can provide added functionality.[47] The
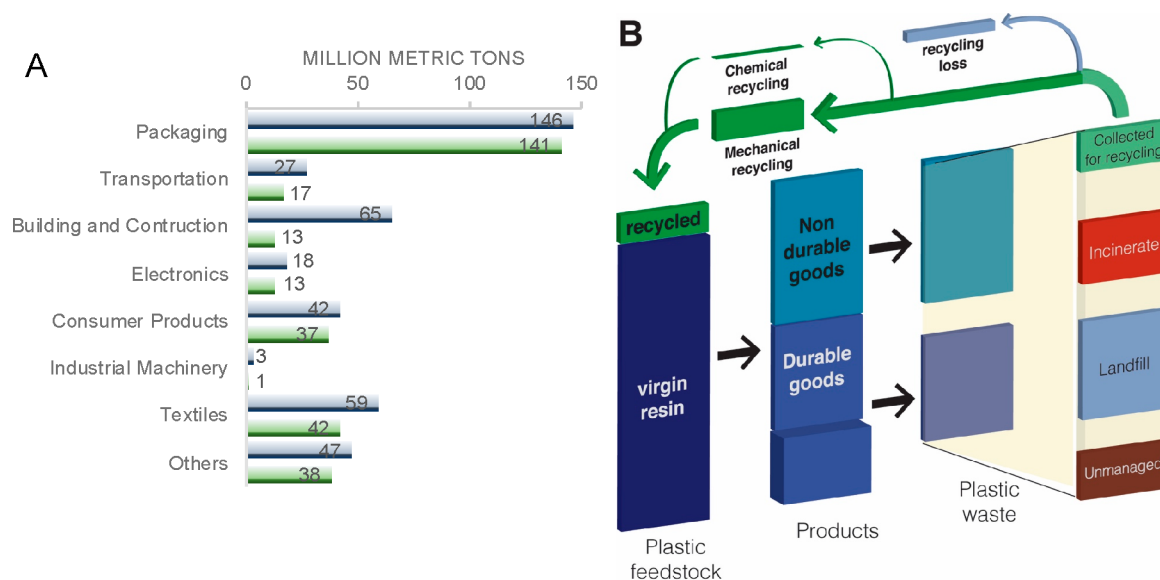
**Figure 3.** (A) Plastic production (blue) and waste generation (green) as a function of industrial sector in 2015. Data from ref 4, Table S1. (B) Visual breakdown of the plastic flow from production to waste worldwide. The size of the blocks is directly proportional to the mass of plastic in the category. 300 million metric tons of virgin plastic were produced, while only 30 million metric tons of plastic was recycled back into plastic feedstock. Data were obtained from ref 247.

drive to transparent high barrier plastic adds an additional challenge to rapid identification of composite plastics in packaging.[48] The highly engineered nature of emergent packaging materials provides additional complexity to the plastic waste stream beyond simple mixtures of different polymers and additives. The impact of these characteristics on recycling will be described in the next sections.

## 3. RECYCLING BY REPROCESSING

**3.1. Definitions and Challenges.** A key issue which complicates recycling protocols, especially those that rely on the deconstruction of chains, is that the free energy of polymerization, $\Delta G_{poly} = \Delta H_{poly} (1 - T/T_{ceiling})$, can be strongly favorable at room temperature ($-\Delta H_{poly} \sim 30-100$ kJ/mol[49]). This leads to many polymers having high thermodynamic stability relative to being depolymerized to a monomer, as reflected in their $T_{ceiling}$ (Table 1; more details are

**Table 1. Reported $T_{ceiling}$ for Some Polymers**

| polymer | $T_{ceiling}$ (°C) |
| --- | --- |
| poly($\alpha$-methylstyrene) | 66 |
| polyisobutylene | 175 |
| poly(methyl methacrylate) | 198 |
| poly(ethylene terephthalate) | 300 |
| polystyrene | 235,[a] 395[b] |
| polypropylene (PP) | 466[a] |
| polyethylene (PE) | 407,[a] 610[b] |
| polytetrafluoroethylene | 1100 |

[a]Data from ref 59. [b]Data from ref 60.

included in the section on Chemical Upcycling). While less stable polymers with lower $T_{ceiling}$ (e.g., PET and polystyrene, PS)[50] are amenable to many "breakdown" pathways,[51] high $T_{ceiling}$ polymers (e.g., polyethylene, PE, and polypropylene, PP, that together constitute >60% of all polymers used[52]) have a much smaller selection. Coates, for example, argues that thermo-catalytically fragmenting polyolefins to monomers is

infeasible because the product free energies are much too high relative to the reactants.[53,54] Some reactions, e.g., polyolefin cross-metathesis, yield smaller, repolymerizable olefins,[55−58] but energy-efficient routes for polyolefin upcycling remain a grand challenge.

While quiescent depolymerization is thus difficult, adding mechanical forces can change the underlying thermodynamic balance and make chain fragmentation a favorable proposition. Frequently, however, this is at the expense of uncontrolled chain end chemistry, where the resulting materials are not repolymerizable. Since Staudinger,[61] it is well-known that mechanical stresses can shift the thermodynamics to favor chain cleavage.[62−68] Historically,[69,70] these homolytic bond scission reactions[71−90] and flow-induced mechanochemistry[78−80,87−102] have been used to reduce polymer molecular weight (including in polyolefins). However, even when the overall force-coupled thermodynamics of scission becomes favorable, significant kinetic barriers can remain. Since force generation is molecular weight dependent and because high forces (>3 nN) are required to overcome kinetic barriers to homolytic bond scission, in practice, mechanical degradation is limited to very high molecular weights.[103]

In this review, we refer to common processing protocols, such as extrusion, as "mechanical" recycling. As noted above, the chain fragmentation that sometimes occurs under the action of these external forces typically does not lead to repolymerizable fragments. In contrast, deliberate strategies to fragment polymers into repolymerizable units is termed as "chemical" recycling.

**3.2. Challenge of Mixed Plastic Waste Streams.** The historically low recycling rates are not by accident. Rather, they are a consequence of a variety of economic and technical challenges that disincentivize recycling. Due to the general poor miscibility of polymer blends, effective sorting of the waste is critical to the quality of products obtained from mechanical recycling, which is the predominant recycling strategy as shown in Figure 3.[104] Even blends of amorphous polyolefins analogous to ethylene/but-1-ene copolymers are

found to be immiscible at room temperature.[105] The general immiscibility of polymers with other polymers leads to strict requirements on the efficacy of the sorting process to minimize contamination by other polymers present in mixed, postconsumer plastic waste. Optical sorting is generally accurate, but the diversity and quantity of plastic associated with a typical recycling operation leads undoubtably to some unwanted plastic in the final recycled product. Separation by flotation can improve the quality of the recycled product,[106] but such processes are never perfect. The phase separation associated with polymer blends in the final recycled feedstock results in additional degradation in mechanical properties, particularly those associated with failure, beyond the shear-induced reduction in properties from decreased molecular weight associated with reprocessing. Thus, routes to increase the tolerance of other polymer impurities in the recycled stream are highly desired to reduce the sorting requirements.

Block copolymers that are appropriately balanced can effectively stabilize the interface between the polymer phases as demonstrated by Balsara and co-workers,[108] similar to surfactants stabilizing an emulsion of oil−water. Alternatively, Macosko and co-workers illustrated that the kinetic trapping of the dispersed phase in polymer blends can be further controlled with the addition of diblock copolymers using high shear processing, typical of industrial manufacture, and quenching sufficiently fast to minimize significant ripening of the dispersed phase.[109] These approaches can potentially limit the size of the defects associated with the undesired polymer from the perspective of plastic recycling, but the use of multiblock copolymers better addresses challenges associated with mechanical properties.[107,110] As shown in Figure 4, the addition of 5 wt % of a tetrablock copolymer dramatically decreases the size of the dispersed PP phase in a HDPE matrix. This change in morphology converts the HDPE/PP blend from brittle to tough and ductile.[107] It has been suggested that these block copolymers could improve the recycling of HDPE and PP by decreasing sorting requirements, but such improvements in mechanical performance have not yet been demonstrated with a more complex feedstock containing a mixture of different HDPE and PP grades, additives, and fillers that would be present in a postconsumer waste stream. The properties obtained from compatibilization will depend on the exact composition of the plastic waste stream, which will be variable for postconsumer waste. The acceptability of the compatibilized materials will depend on the variance and the acceptable performance for a given application. Additionally, one common challenge with the inclusion of block copolymer additives is their cost, which tends to be prohibitive even at their low loadings for efficacy.

The ubiquitous use of polyolefins is driven by their low cost and design levers that can be utilized to achieve desired performance. For example, packaging commonly requires low oxygen and moisture permeation, which is challenging to achieve without some engineering.[47,111] Thin inorganic or metal coatings can dramatically improve the permeation performance,[46] but are typically difficult to remove and thus are incorporated into the recycled plastic. For example, the properties obtained from an aluminum coated LDPE after mechanical recycling is strongly dependent on the Al dispersion in the LDPE after processing.[112] The growing use of composites will only act to increase the complexity of the waste streams and the challenges with maintaining dispersion to prevent large aggregates that can act as stress concen-
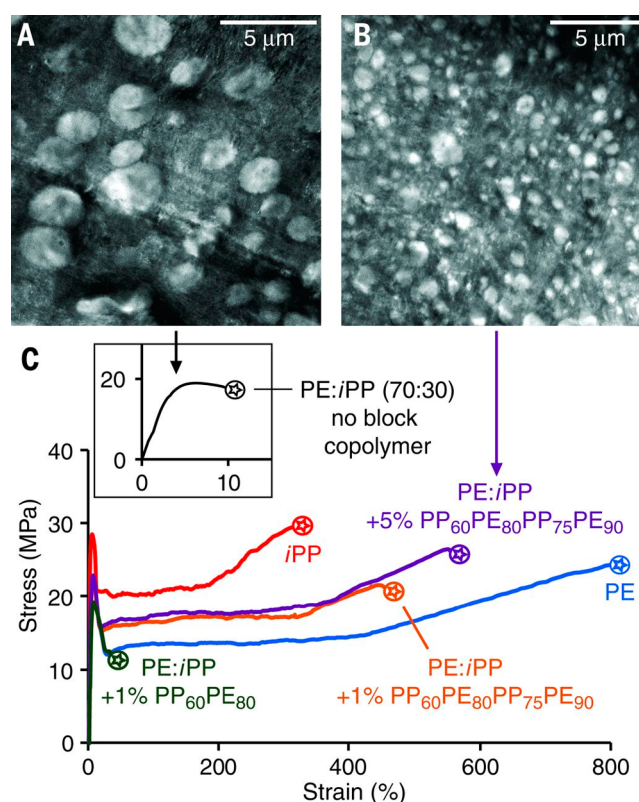


**Figure 4.** Electron microscopy images of a 70/30 blend of HDPE and PP (A) without any block copolymer and (B) with 5 wt % of tetrablock ($PP_{60}PE_{80}PP_{75}PE_{90}$) copolymer. (C) The blend without any block copolymer is brittle and there is only modest improvement with the inclusion of 1 wt % of a diblock ($PP_{60}PE_{80}$) copolymer. The use of the tetrablock copolymer results in a blend with mechanical properties intermediate to the pure (HDPE and PP) components. Reproduced with permission from ref 107. Copyright 2017, American Association for the Advancement of Science.

trators[113] will be an additional challenge to mechanical recycling.[114] Beyond composites, the plastics can be engineered with a variety of additives and multilayer (plastic) materials as a means of providing desired functionality, but these increase recycling difficulty.[115] In particular, developments in delamination and compatibilization of multilayer materials are necessary to enable their effective recycling.[45] Although a reversible cross-linked adhesive between layers has been proposed for the recovery of individual components,[116] the costs of this additional component will challenge adoption in the absence of regulations. Alternatively, the separation of the multilayer materials can be accomplished through appropriate solvent-based processes that selectively dissolve and reprecipitate components from clean multilayer material in high purity.[117] Figure 5 illustrates one recently demonstrated process termed solvent-targeted recovery and precipitation (STRAP) where these selective solvents can enable the recovery of individual components.

Technoeconomic analysis has demonstrated the potential to produce PET from STRAP at a cost point competitive with virgin PET.[117] Every (re)processing step leads to finite damage, which will degrade the properties of the recovered polymer and limit the number of times that a plastic resin can be recycled.[118] With the high purity and control over the additives present, the degradation is expected to be less than with traditional mechanical recycling to increase the number of
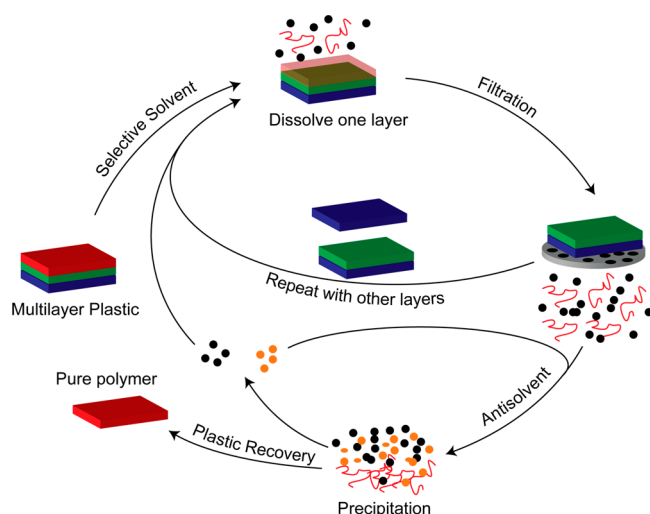
**Figure 5.** Schematic illustrating one of the dissolution−precipitation methods that allow for the recovery of the individual plastics in multilayer plastic films. Scheme adapted from the STRAP method reported in ref 117.



**Figure 6.** Composition of volatile components in postconsumer (PC) recycled polyethylenes determined from gas chromatography and mass spectrometry. A vast majority of the identified compounds resulted from contaminants acquired through the service life and recycling process. Reproduced with permission from ref 125. Copyright 2020, Elsevier B.V.

potential times that a resin can be recycled. Similar separations of multilayer packaging that contain metal components have been demonstrated using switchable solvents[119] and selective dissolution−precipitation processes.[120] The advantage of these solvent separations is that relatively pure polymer can be recovered without loss in properties, but the cost of the solvent and its recycling can be significant when considering polyolefins. Sherwood has recently reviewed a variety of solvent-based methods for recycling of plastics that includes PET, PLA, PVC, and polyolefins and has noted the economic challenges with these approaches, but these solvent-based methods provide an excellent opportunity to maximize reuse of postconsumer plastics as they can produce value-added plastics from difficult to recycle waste streams.[121]

Alternatively, compatibilization of the mechanically recycled multilayer plastic material can be applied, similar to that proposed for tolerance of polymer impurities during sorting of plastic waste. For example, blending recycled multilayer plastic films from food packaging with virgin resin and a maleic anhydride containing compatibilizer provides reasonable processability and mechanical properties.[122] However, these properties are inferior to the virgin resin, so the relative savings with the recycled plastic must be large for this to be economically feasible. Direct mechanical recycling without the need to add virgin resin can be accomplished using block polymers to provide reasonable mechanical properties of PET and HDPE (or LDPE) based blends.[110] However, the efficacy of the compatibilization likely depends on the polymer components present in the multilayer materials and the availability of appropriate compatibilizers for the mixtures.

Additionally, the variety of additives present in plastic waste[40] may differ from the initial formulation due to migration from[123] or to[45] the plastic during their lifetime.[124] Examination of postconsumer recycled LDPE and HDPE identified over 1000 chemicals from chromatography with a majority of these associated with the identified compounds being predominately from contaminants.[125] Figure 6 illustrates the breakdown of the volatile components in terms of origins for the HDPE and LDPE. A minority of these compounds were originally present in the virgin resins and these results
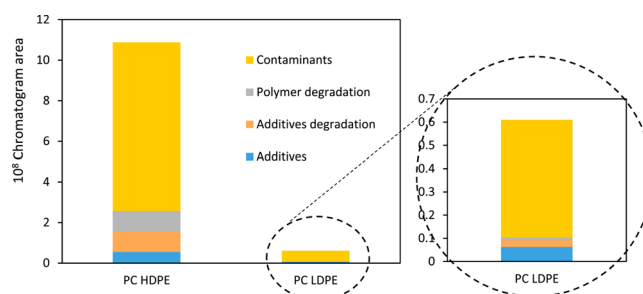
illustrate the need to understand degradation products and potential adsorption from the environment[125] on the properties of recycled plastics. The complexity of the chemical mixtures even in sorted plastic waste due to the formulations and contamination from sorption of chemicals during use and collection will impact the properties of the recycled plastic. These recycled plastics can include banned chemicals as a result of their accumulation through reuse.[126] To comply with governmental regulations, monitoring of plastic waste streams directed to recycling may be necessary and further increase costs associated with recycling. To address unintended inclusion of additives, selective dissolution/precipitation has been proposed for the recovery of clean plastic waste[127] by removing the additives due to their differential solubility with respect to the polymer.[128] This method, however, generates significant quantities of solvent waste and the separation of the solvents from the additives and plasticizers that remain in solution will incur additional costs for the process. De Meester and co-workers have recently reviewed solvent separation methods from the perspective of additive removal to enable recycling of plastic waste with both economic and environmental assessment, which illustrated the potential of these processes to improve the quality of the recycled product.[128] Despite the added costs from the solvent, the high purity polymer product from dissolution−precipitation processes should reduce the variability in the properties of a recycled product and has led to a number of commercial efforts. Purecycle Technologies has been working to commercialize a process to recover high purity PP developed by Proctor & Gamble[129] that exploits a lower critical solution temperature (LCST) to precipitate PP in an economical manner. While they have invested $250 million into their first plant due to be completed in 2022, as of March 2021, PureCycle has not generated any operating revenue and does not expect to until the end of 2022.[130]

Similarly, CreoSolv, developed by the Fraunhofer Institute for Process Engineering and Packaging IVV, has been promoted for closed-loop recycling of plastic to minimize solvent requirements and has been commercialized by PolyStyreneLoop with the initial plant opening in June 2021 for the recycling of polystyrene foam.[131] The coming years will be insightful to determine if these dissolution−precipitation processes are economically competitive, but these offer the potential for improved purity and consistency of the recycled product.

Even only considering a single polymer type within the plastic waste, the molecular weight distribution tends to be engineered to the application to meet the processing and product property demands.[132] Thus, products of nominally the same plastic may have vastly different molecular weight distributions if produced by injection molding in comparison to pultrusion or blow molding. These differences in the initial molecular weight distribution will alter the rheological performance of the recycled resin and its suitability for the variety of melt processing typically associated with the production of plastic products. Recent work has provided theoretical-based insights into the processability of such mixtures of plastics that are encountered in mechanical recycling,[133] which could assist in the processing of recycled plastics. However, variability in the postconsumer waste stream leads to a substantial variance in the molecular weight distribution, additives and fillers present in the recycled product. As these details impact the processability (rheology) and ultimate mechanical properties of the material in the product, key metrics associated with the economics of the product (production throughput and product performance) will in general be poorly controlled in mechanically recycled plastics. Schyns and Shaver have recently reviewed mechanical recycling of packaging plastics that describes material specific considerations for common packaging plastics (PET, LDPE, HDPE, PP, PVC, PS, and their blends).[134]

**3.3. Mechanical Recycling.** The composition of preconsumer plastic waste is well-controlled and this greatly facilitates their recycling. Even for these controlled recycled feedstocks, the maximum recycled content is limited as the mechanical properties of most plastics are degraded by multiple reprocessing steps.[135] For example, the ductility of PET is reduced by 2 orders of magnitude by the third time being mechanically recycled.[118] For postconsumer plastics, mechanical recycling is more challenging as the composition of the waste stream is variable. As shown in Figure 7, the typical mechanical recycling process involves sorting (for postconsumer plastics), grinding, washing, separating, drying, and compounding of the plastic waste to produce thermoplastics ready to be reprocessed into products.

The sorting of the plastics is typically accomplished by optical techniques with separations of plastic waste by polymer type (typically with near IR, NIR) and for the high value plastics (PET and HDPE) by color as well. Certain plastics challenge this NIR sorting, in particular, the black packaging common in food trays as it tends to absorb strongly in the NIR.[136] There have been some developments to modify the black colorant to enable NIR sorting without altering the visible color, such as OnColor IR Sortable Black for Recyclable Packaging,[137] which will increase the fraction of plastics that can be recycled as there is a high PET content for black food packaging. After sorting, the plastics are milled, washed, and separated again by floatation. The clean plastic particles are then reprocessed (sometimes including another grinding step) to produce recycled plastic feedstock to be processed into products. The high shear environment typically encountered during polymer processing (both in recompounding and product fabrication) tends to decrease the average molecular mass of the plastic to reduce its mechanical performance.[138] There have been significant efforts to generate new chemistries and processes[138] to mitigate property degradation and enable increased recycled content in products. For semicrystalline polymers such as PP for injection molding applications, this
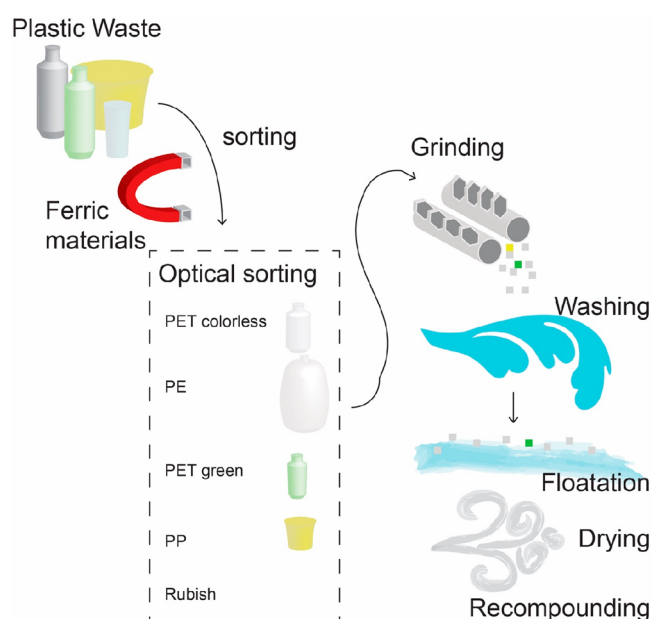


**Figure 7.** Schematic illustrating the general steps for the recovery of plastic waste to plastic feedstock through mechanical recycling. The transportation steps are not included for simplicity. There are a variety of additional processes possible in recompounding to maximize the performance of the recovered plastic.

can be as simple as modifying the molding conditions to generate a more favorable morphology on crystallization.[138] However, this change may adversely impact the cycle time for injection molding to increase the production costs. Alternatively, the decrease in molecular weight can be reversed through chain extension via reactive processing, which is a potentially economical route to mitigate these difficulties.[139,140] These reactive processing routes also offer the potential to enhance the properties depending on the chemistries selected, which may enable upcycling of the waste plastic.[18]

For postconsumer recycling of PET, substantial degradation in performance has been reported due to mechanically facilitated chemical changes in the polymer.[141] Such degradation is a common concern with condensation polymers due to their lower ceiling temperatures and their sensitivity to moisture and alcohols.[142,143] Oxidative changes to PET during mechanical recycling by extrusion tend to include both chain scission to reduce the molecular mass as well as branching induced by chain coupling.[144,145] These tend to decrease the average molecular weight while broadening the polydispersity. Figure 8A illustrates how the molecular mass of PET decreases as the PET is mechanically reprocessed via extrusion. The associated decrease in performance does not hamper its use in some applications, such as textile fibers, that have lower intrinsic viscosity requirements than required by blow or injection molding,[146] but this is essentially downcycling of the PET from high value to low value applications.

Chain extension to reinvigorate degraded feedstocks provides one route to circumvent the breakdown of condensation polymers during mechanical recycling.[139,147] Solid state polymerization to encourage chain extension to counter the loss of molecular mass is a typical mitigating strategy.[148] Selection of an oligomeric chain extender, such as Joncryl-ADR-4368, which is an epoxy-functionalized acrylic,[149] can provide a more effective route to counter decreased
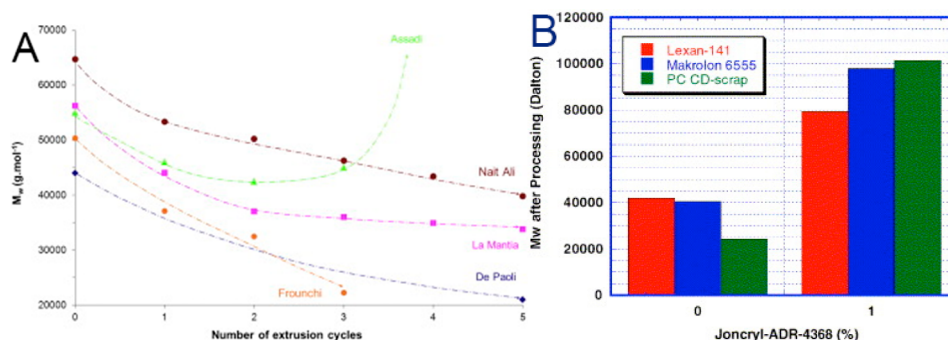
**Figure 8.** (A) Dependence of molecular mass on the number of extrusion cycles to mimic mechanical recycling for several different PET samples reported in the literature from a variety of authors. Reproduced with permission from ref 144. Copyright 2011, Elsevier B.V. (B) Influence of an oligomeric chain extender (Joncryl-ADR-4368) on the postprocessing molecular mass of two commercial PC grades and low value PC scrap from compact disks. Reproduced with permission from ref 139. Copyright 2006, Elsevier B.V.

molecular mass with reactive extrusion.[139] This functionalization enables one chain extender to be applicable to a variety of condensation polymers with effective reversal of molecular mass degradation in postconsumer polyesters, polyamides, polycarbonates, and polyurethanes.[139] Figure 8B illustrates the effect of 1% of chain extender on the molecular mass of several PC samples through a reactive extrusion. The lowest grade PC from compact disk scrap demonstrated the greatest increase, which may be associated with the higher density of reactive chain ends and the lower viscosity initially in the PC to promote the efficacy of the chain extension.[139] The functionality and architecture of the chain extender can dramatically influence the properties of the recycled product.[150] Fillers can also be used to enhance the mechanical properties of mechanically recycled PET to produce properties comparable to virgin PET.[151] However, there are still challenges to "close the loop" to the original application for the materials as regulations associated with food packaging, and the entry of contaminants, constrain the use of mechanically recycled plastics in food contact.[152,153]

Although the chemical stability of polyolefins is greater than condensation polymers, the effect of mechanically induced degradation on the performance of HDPE was simulated by consecutive extrusions. The decreased modulus after 10 reprocessing cycles[154] points to a finite lifetime to the recyclability of polyolefins. The potential for increased crack growth with recycled HDPE due to decreased mechanical performance on reprocessing has limited its use in some long service life applications, such as pipes in construction.[155] Among polyolefins, PP tends to be the most sensitive to mechanical recycling, which results in significant decrease in molecular mass to increase crystallinity and a loss of toughness.[156] When adding thermo-oxidative processes, the decrease in performance of recycled PP is accelerated.[156] The tertiary carbon in the repeat unit of PP is particularly susceptible and thus additional antioxidants tend to be added for mechanical recycling to minimize oxidative chain scission. However, the diversity of additives present in postconsumer PP can significantly alter the properties after recycling.[138]

**3.4. Life Cycle and Technoeconomic Considerations for Improved Viability of Mechanical Recycling.** PET is currently the best example of large volume mechanical recycling. The National Association for PET Container Resources releases an annual report detailing the state of postconsumer recycling of containers.[157] The reported utilization rate of PET in recycling has remained consistently

near 20% since 2005, despite an overall increase in the recycling collection rate for PET through 2017.[157] The vast majority of the virgin PET resin is used in the manufacture of bottles and the plurality (approximately 40−50%) of postconsumer recycled containers are remanufactured into fibers based on data collected since 2001. The prevalence of recycled PET (rPET) in fiber applications is likely derived from two major characteristics.

First, PET mostly exists as either bottle grade or fiber grade,[158] with the viscosity of the fiber grade typically being lower. Thus, the degradation of the viscosity of bottle grade polyesters typically encountered during mechanical recycling can be acceptable for fiber applications. Furthermore, the extension of the solid-state polymerization technique used to produce high viscosity PET to recycling streams can boost the molecular weight to produce a viscosity appropriate for fiber applications. This facile recovery of molecular weight makes PET somewhat unique among high use commodity plastics, and thus, there is more latitude economically to recover PET for mechanical recycling. The lack of a similar, commercial scale processes for other step growth polymers limits their mechanical recycling. For example, polyurethane foams, commonly used in cushions and insulation, are difficult to reprocess for use in a virgin-like state. Typically, these materials are shredded, mixed with a binder and consolidated into lower value carpet underlayment.[159]

Second, the large PET fiber market provides an accessible target industry for the rPET with market pull from consumer demands for environmentally friendly fabrics and materials leading to fibers with increasing content of recycled material in commercial products. These market conditions have enabled commercial success for companies like Unifi with their Repreve material, but virgin resins (>97%) still dominate in the production of textiles.[160]

The economic drivers for mechanical recycling require a convergence of factors, which has primarily limited mechanical recycling from an economic perspective to PET and natural HDPE. There are additional societal questions associated with the impact of mechanical recycling. Significant resources are utilized in mechanical recycling, including the fuel associated with collection and transportation, water use in cleaning, and energy in reprocessing. The optimal approach to mechanical recycling from lifecycle analysis (LCA) may not be generating extremely clean, virgin-like materials[161] as is typically the goal. In particular, the processes used to clean and reprocess recycled materials tend be significantly more resource intensive

than simple mechanical recycling with minimal value addition. Creating clean, efficient processes in combination with advanced compatibilizers can fill gaps in technology for mechanical recycling to improve both the economic and LCA considerations.

## 4. CIRCULAR POLYMER ECONOMY BY DEPOLYMERIZATION

With the challenges associated with mechanical recycling of postconsumer plastic waste in terms of sorting, degradation in molecular weight during reprocessing, and compatibilization of unknown mixtures, there has been growing interest in the recycling of polymers by fully chemical means.[162,163] Figure 9
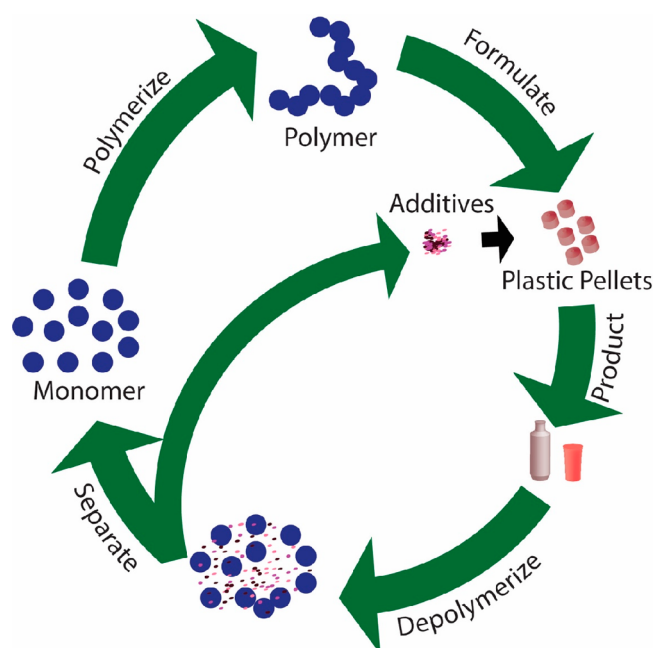


**Figure 9.** Circular economy for plastics through polymerization and depolymerization cycles. The recycled polymer can be essentially identical with the virgin polymer or completely reformulated to meet requirements for another high value application.
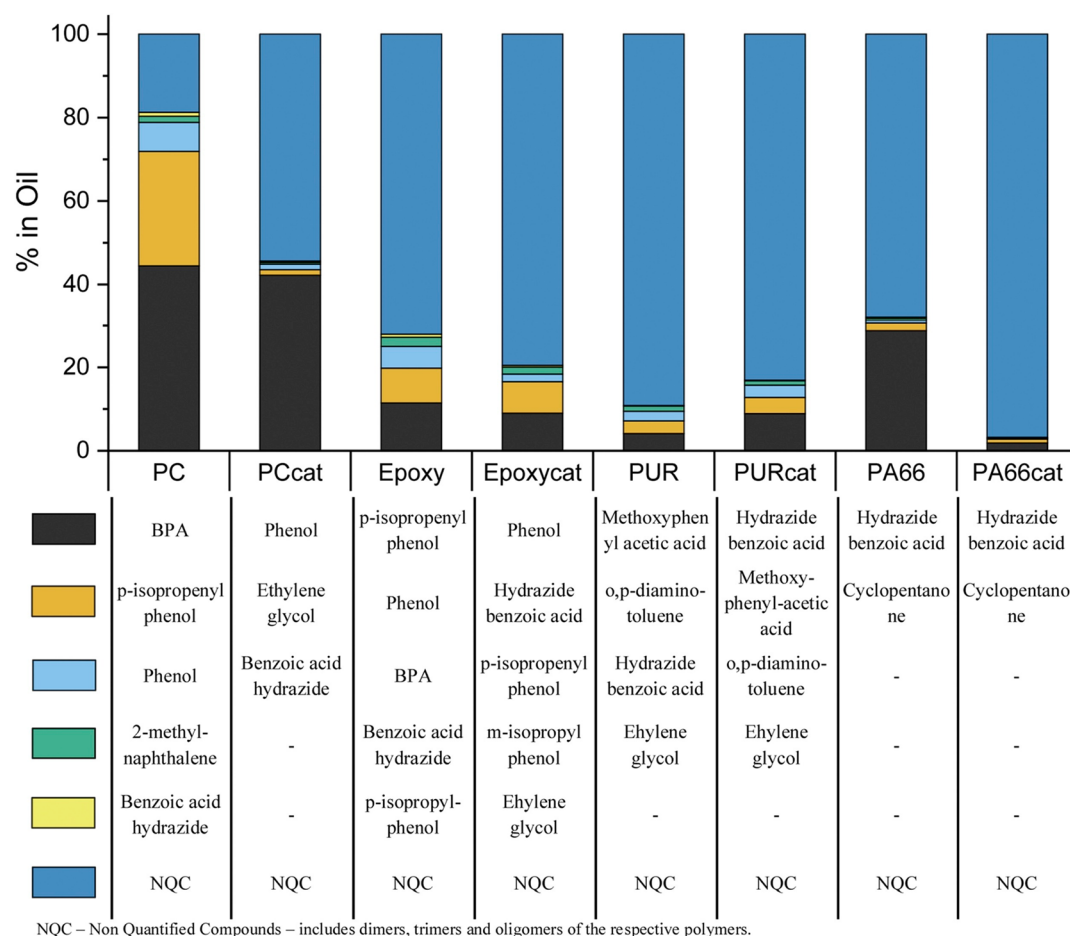
schematically illustrates the circular economy approach where monomers are polymerized to make products. At the end of their service life, the polymer is then depolymerized back to monomer. The advantages of chemical recycling are associated with the depolymerization to monomers, which can be more readily purified, and then repolymerization to make recycled plastics that are indistinguishable from virgin resins. Chemical recycling can potentially address the challenges with multilayer plastic films in packaging to transform the plastics present back to monomers without addressing the solid state separation challenge.[164] The conversion back to the monomer allows for a closed materials loop, where the plastics of tomorrow are made from monomers derived from the plastics of today. This circular economy for plastics[165] could have a significant impact on the overall demands for oil-based or biobased monomers in the future as the plastic in place now theoretically could be chemically recycled perpetually through the depolymerization–repolymerization.[166] However, the additives that provided challenges in the mechanical recycling of plastics can also adversely impact their chemical recycling back to monomer through poisoning of catalysts that enable the depolymeriza-

tion.[167] There are catalysts that are insensitive to pigments and other additives, such as Brookhart's Ir(III) catalyst,[168] which provide insights into catalyst designs that would be tolerant for postconsumer plastic waste. While the interest has been recently rekindled, the concept of chemical recycling is not particularly new with patents in the 1950s describing the depolymerization and chemical recycling of PET.[169] Despite the considerable fraction of PET in the plastic waste stream (Figure 2), the contribution of chemically recycled plastics to the overall recycling stream is vanishingly small (Figure 3). In the following sections, the challenges associated with realization of chemical recycling back to monomer on an industrial scale are described along with some strategies being undertaken to improve the viability of these methods to enable a circular economy for plastic feedstocks.

**4.1. Brief Thermodynamic Considerations for Depolymerization.** As the circular economy through chemical recycling is associated with the depolymerization of the polymer, generally back to its monomer(s), examination of the thermodynamics provides a floor for the energy requirements for chemical recycling (as previously discussed above). The decrease in entropy associated with the conversion of small molecules (monomers) to polymers requires polymerization to be exothermic for a negative Gibbs free energy of reaction ($\Delta G_{poly}$). The enthalpy–entropy compensation point where $\Delta G_{poly} = 0$ is the ceiling temperature for the polymerization, $T_{ceiling}$.[170,171] This temperature provides insight into the ease of depolymerization of different plastics as $\Delta G_{poly}$ is a state function associated with the (de)polymerization and does not consider the kinetic pathways. As shown in Table 1, there is significant variance in the reported values for $T_{ceiling}$, but one general trend is that polyethylene exhibits the highest $T_{ceiling}$ and thus will be challenging to depolymerize back to ethylene.

The limitation associated with the ceiling temperature limits depolymerization to monomer at lower temperatures. Pushing equilibrium toward the monomer is possible with condensation polymers through the use of solvents (water, alcohols, glycols, amines) that can potentially be products of polymerization.[169] However, as these are equilibrium processes, the products tend to be a mixture of monomers and oligomers plus the excess solvent required to shift equilibrium toward monomers.[172] The recovery of monomers to complete the circular economy is thus typically limited by the ability to effectively purify the products from the equilibrium depolymerization.

The role of the addition of byproducts from the polymerization on depolymerization is best illustrated through hydrothermal processes that can convert PET, polycarbonate, and polyamide-6 to monomers as well as a variety of other chemicals, while limited depolymerization of PP or PS occurs under the same conditions.[173] However, even for condensation polymers, the product distribution can be messy. Figure 10 illustrates the product distribution from hydrothermal treatment for a variety of polymers. Of the polymers examined, polycarbonate provided the highest fraction of monomer (BPA), but this was still under 50% of the total mass.[173] To increase the yield of the desired monomer, more extreme conditions are typically required. The depolymerization of PET,[174] polycarbonate,[175] or polyurethane[176] to monomers in high yield is possible without any catalysts in supercritical water or alcohols, but the relatively high temperatures and pressures associated with supercritical fluids tends to adversely

| | PC | PCcat | Epoxy | Epoxycat | PUR | PURcat | PA66 | PA66cat |
|---|---|---|---|---|---|---|---|---|
| ■ (black) | BPA | Phenol | p-isopropenyl phenol | Phenol | Methoxyphenyl acetic acid | Hydrazide benzoic acid | Hydrazide benzoic acid | Hydrazide benzoic acid |
| ■ (orange) | p-isopropenyl phenol | Ethylene glycol | Phenol | Hydrazide benzoic acid | o,p-diamino-toluene | Methoxy-phenyl-acetic acid | Cyclopentanone | Cyclopentanone |
| ■ (light blue) | Phenol | Benzoic acid hydrazide | BPA | p-isopropenyl phenol | Hydrazide benzoic acid | o,p-diamino-toluene | - | - |
| ■ (green) | 2-methyl-naphthalene | - | Benzoic acid hydrazide | m-isopropyl phenol | Ehylene glycol | Ehylene glycol | - | - |
| ■ (yellow) | Benzoic acid hydrazide | - | p-isopropyl-phenol | Ehylene glycol | - | - | - | - |
| ■ (dark blue) | NQC | NQC | NQC | NQC | NQC | NQC | NQC | NQC |

NQC – Non Quantified Compounds – includes dimers, trimers and oligomers of the respective polymers.

**Figure 10.** Product distribution in the oil phase from hydrothermal treatment for polycarbonate (PC), epoxy, polyurethane (PUR) and nylon (PA66) with and without KOH as an alkali catalyst. Reproduced with permission from ref 173. Copyright 2020, Elsevier B.V.

impact the economics of such depolymerization. In general, the depolymerization of addition polymers, such as polyolefins, back to monomer tends to be challenging due to thermodynamics.[170]

**4.2. Chemical Recycling of Common Plastics.** The relatively low ceiling temperature of PET along with the ability to shift equilibrium to favor the monomer with appropriate solvents has led significant efforts over the past several decades to develop depolymerization methods,[164,167,169,177−179] but large scale implementation industrially remains rare. For chemical depolymerization of PET, the bench scale yield of monomer can be nearly 90% with 95% purity from virgin PET at temperatures near 200 °C, but this tends to decrease with pigmented or multilayer PET common in plastic recycling streams.[180] With such performance in depolymerization to monomer, there have been efforts associated with commercialization of depolymerization processes to recover monomers, bis(2-hydroxyethyl) terephthalate (BHET), dimethyl terephthalate (DMT), and ethylene glycol. In the 1990s, Eastman Chemical developed a commercial PET product line that used 50% chemically recycled content. The technology was originally developed as Eastman Kodak to chemically recycle the polyester in photographic film. However, this product line was short-lived due to lack of demand. Similarly starting in 1994, DuPont operated a chemical recycling (methanolysis) facility for production of DMT from PET waste instead of *p*-xylene.[181] In 2019, BP announced the plans for a pilot plant to

depolymerize difficult to recycle (nonclear PET) postconsumer PET using the BP Infinia technology.[182] A plan by a joint venture between Loop Industries and Indorama Ventures was announced in 2019 to depolymerize nearly 40 thousand metric tons of PET annually with a low temperature/low pressure process, but the associated production unit has been delayed in completion from 2020 to 2023 and one contract with a major customer has already been canceled due to the delays.[183] More recently, Eastman Chemical announced plans in early 2021 to build the world's largest chemical recycling facility for plastics to convert 100 000 t of PET, which cannot be mechanically recycled, into monomers for "premium specialty plastics made with recycled content".[184] This announcement marks a circular return to chemical recycling after the failure of a prior product line based on chemically recycled PET. It should be noted that the scale of the investment by Eastman for the methanolysis facility to chemically recycle PET is an order of magnitude greater than the investment by DuPont in the 1990s for a similar technology. For more information about chemical recycling of PET, readers are directed to a recent review focused solely on this subject.[185]

The textile industry has been interested in the depolymerization of polyester to improve the sustainability of synthetic materials,[186] but the presence of pigments and dyes can hamper the depolymerization and lead to challenges in obtaining the requisite purity of the obtained monomers for repolymerization. Instead of repolymerization to primary

fibers, the depolymerized product can act as a softener[187] or improved dye for fabrics.[186] There are a multitude of additional issues that also influence the technoeconomic viability of chemical fiber recycling in terms of a circular economy.[188] Chemical recycling to monomers for condensation polymers has advantages in that the conditions for depolymerization tend to be relatively mild, but the purity requirements for monomers for condensation polymerization are relatively strict as small stoichiometric imbalance in end functionalities can significantly decrease the average molecular weight. Additionally, the solvent costs to drive the depolymerization is estimated to represent nearly 40% of the cost to produce BHET by glycolysis due to the large excess of glycol required to produce a high yield of the BHET monomer.[180] This high cost of glycol is likely the rationale for methanolysis to produce less desired products (DMT and EG) for large scale facilities.[184] Alternatively, recent developments with engineered depolymerases have demonstrated the ability to biologically degrade PET back to monomer that can be recovered for repolymerization,[189] but the costs of the engineered depolymerases need to be decreased to be cost competitive.

The recovery of monomers from chain growth polymers tends to be more challenging with increased stability of the polymer and generally limited functionality along the backbone. From the perspective of significantly impacting plastic waste, the ability to recycle HDPE, LDPE, and PP is a critical consideration as these polymers represent a majority of the plastic waste generated. Simple pyrolysis requires temperatures in excess of the ceiling temperature to crack polyolefins to light olefins (monomers) that could be used again in the synthesis of polyolefins, but the residence time must be short to avoid reduction to methane and other secondary reactions.[190] For HDPE, the yield of light olefins (ethene, propene, and butene) was maximized at 700 °C.[191] The pyrolysis of LDPE, HDPE, and PP yields a mixture of hydrocarbon liquids and gases that are primarily aliphatic,[192] which requires additional separations to produce a usable product for repolymerization. As an alternative to the chemical approach, catalytic pyrolysis of HDPE to light olefins can be accomplished in a two-step process; HDPE is first cracked at 500 °C to long alkanes and waxes and then converted catalytically to light olefins over a zeolite catalyst at 550 °C with about 60% of the carbon from the HDPE being captured in ethene, propene, and butene products.[193] The temperature for the conversion of long alkanes into olefins is within the reported range of the ceiling temperature for polyethylene (Table 1). The product distribution was mostly propene for the olefins produced and their yield was reported to decrease significantly at lower temperatures as would be expected.[193]

Figure 11 illustrates the product distribution of light alkanes/alkenes from a two-step catalytic cracking of HDPE using a continuous fixed bed reactor.[193] The product yield is dependent upon the catalyst space time with longer exposure of the HDPE to the catalysts resulting in higher yield of the light olefins. At longer space times, the yield of propene increases, which reaches a maximum of approximately one-third of the product.[193] The catalytic conversion of HDPE to heavier olefins (C3–C6) can be accomplished at lower temperatures (290 °C) as the ceiling temperature limitation of polyethylene depolymerization can be sidestepped with these longer aliphatic chains as products. Higher yields of light olefins (75%) can be obtained from LDPE when operating at
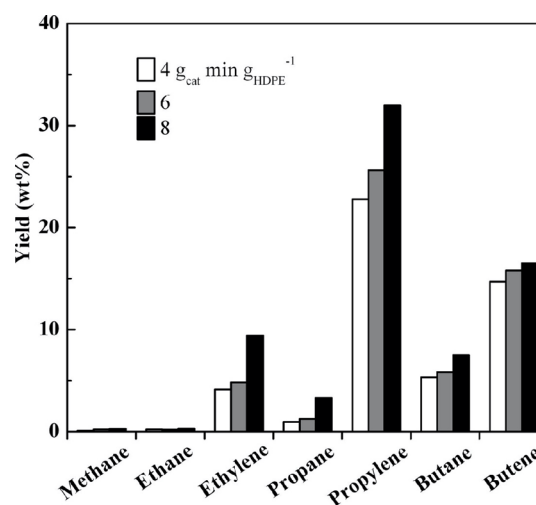


**Figure 11.** Gaseous products obtained from HDPE based on a two step thermal and catalytic process where the HDPE is first pyrolyzed at 500 °C, followed by catalytic cracking over HZSM-5 zeolite at 500 °C. Reproduced with permission from ref 193. Copyright 2012, Elsevier B.V.

800 °C.[194] The challenges with plastic waste has reinvigorated efforts in the development of catalysts for chemical recycling to increase the circularity in the materials economy for plastics,[195] but the commercial realization of chemical recycling has been limited to date (Figure 3) in comparison to mechanical recycling. Several reviews have recently focused on the catalysts used for chemical recycling of plastic waste.[196,197]

The recovery of light olefins (ethene and propene) that would provide chemical feedstocks for a majority of the plastics produced require high temperatures for the depolymerization due to the stability of the C–C σ bonds. This temperature requirement adds significant energy requirements that are compounded by the modest product selectivity, which will necessitate additional separations. Even with chemical recycling by pyrolysis (thermal and catalytic), there are some sorting requirements as PVC needs to be removed to avoid generation of significant HCl.[198] Similarly PET tends to adversely impact the yield of desired products from pyrolysis.[199] The low cost of olefins obtained from petrochemical processing of fossil fuels challenge the economics associated with chemical recycling to monomers due to high energy requirements (as well as the adoption of biobased plastic alternatives).[200] Miao et al. have recently reviewed depolymerization technologies for common plastics, which provides more depth on the depolymerization challenges.[201]

**4.3. New Polymers Designed for Depolymerization.** With the economic challenges associated with obtaining high quality monomers from traditional thermoplastics through chemical recycling, there has been a concerted effort in recent years to re-examine the plastics used to rationally design the polymer from the monomer up to enable recycling and sustainability,[202] while retaining the desired physical properties of the plastic.[195,203,204] A variety of chemistries, such as bridged thioesters,[205] lactones,[206] and dynamic covalent diketoenamine bonds,[207] have been developed that provide intrinsically recyclable plastics through depolymerization/repolymerization. The challenge with these polymers tends to be the costs. In order for these new polymer designs to make a major impact toward chemical recyclability of plastic waste streams, they
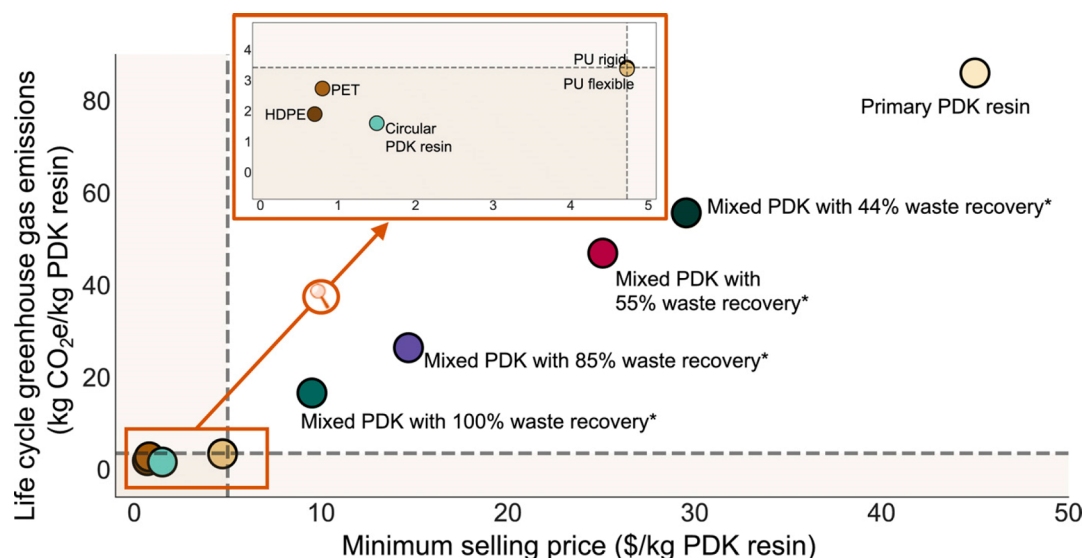
**Figure 12.** Role of recovery and effective chemical recycling of polydiketoenamines on the economics and environmental impact. With a fully circular economy (no new chemicals), the cost can approach polyolefins and PET, but ineffective recovery of PDK dramatically increases the cost. This image is adapted from ref 209 and is licensed under CC BY 4.0.

must be able to effectively replace the primary packaging materials (PET, HDPE, LDPE, and PP, which comprise a significant fraction of the plastic waste), while being cost competitive. A condensation polymer based on oleates from natural sources can produce polyethylene like structures, including crystallinity, but can be nearly fully recovered as building blocks for repolymerization to provide a simple route to circular analogs to polyethylene.[208] Although carbonate linkages between C18 alkane in the backbone enable facile depolymerization, the relative economics of this system compared to polyethylene is not known. Recent analyses of the circular economy for polydiketoenamines (PDK), which are intrinsically recoverable as monomers, has demonstrated that the recycled resin cost is within a factor of 2 of the production cost of HDPE and PET.[209] However, as illustrated in Figure 12, the recovery of the polymer is critical to provide any potential for a cost-effective resin. A circular (fully chemically recycled) PDK has a minimum selling price of $1.5/kg, but the loss due to efficiencies in manufacturing even when 100% of the PDK is recovered raises the minimum sale price by nearly an order of magnitude. Given the current collection rates of plastics for recycling,[5] the cost of PDK is likely to be noncompetitive for commodity plastic replacement without significant government incentives or new regulations that limits the use of polyolefins and PET in some applications.

Although properties similar to current commercial packaging have been demonstrated for polymers designed to be intrinsically recyclable,[206] the scale of production of polyolefins, which is approaching one-quarter of a billion metric tons annually, may be challenging to replicate with the new chemistries. Additionally, with this scale, small differences in the price compared to polyolefins will be magnified substantially by any major user of the plastics. Considering an increase of price in the production of the recyclable plastic by 0.1 cent per kg in comparison to commodity polyolefin would lead to an overall cost to the producers of the polyolefins of more than $230 million (based on historical data shown in Figures 1 and 2). This sensitivity to the production cost and thin margins in the commodity plastic market have limited the number of new large scale polymers introduced in

the past 50 years. In 2007, Eastman Chemical successfully introduced the Tritan line of copolyesters to replace bisphenol A (BPA)-based polycarbonates (PC) in food packaging. This new plastic was driven by the voluntary removal of PC from food packaging due to concerns of BPA as an estrogen-mimic. Note that PC is a minor overall component in plastic waste, and only about 3% of PC is used in food packaging, while BPA-based PC is still common in nonfood contact products. Poly(hydroxyalkanoates) have provided some hope for viable alternatives to polyolefins, but a previous venture in this class of polymers (Metabolix) failed to reach true market viability.[210] This prior history associated with the introduction of new polymers in commodity plastics illustrates some of the potential pitfalls and need for a clear market pull to successfully introduce alternatives to polyolefins that are intrinsically recyclable.

**4.4. Life Cycle and Technoeconomic Considerations.** Closed Loop Partners[211] has identified 98 different organizations involved from lab scale proof of concept to scaled up commercial processes that process plastic to monomer feedstock. Most of these efforts focus on PET due to its prevalence and potential for ester cleavage chemistry through hydrolysis, methanolysis, or glycolysis.[211] For example, Carbios relies on an engineered enzyme to induce hydrolysis at pilot scale, which has been demonstrated to change the paradigm from bottle-to-fiber, the norm for mechanical recycling, to fiber-to-bottle through enzymatic chemical recycling.[212] While claims of great technology for depolymerizing PET are common, the leap from pilot or small commercial scale to a major source of PET monomer has been daunting, although Eastman has recently announced intentions for a large scale plant for depolymerization of PET.[184] Other step growth polymers have also been examined, but the challenges are generally greater than with PET for commercial viability. Nylon-6 depolymerization to caprolactam as feedstock to spin new yarns is being investigated by Aquafil.[211] The Rampf group has developed and commercialized chemical recycling of waste polyurethanes with this technology planned to be scaled to 2000 t annually for converting polyurethane foams to polyols.[213]
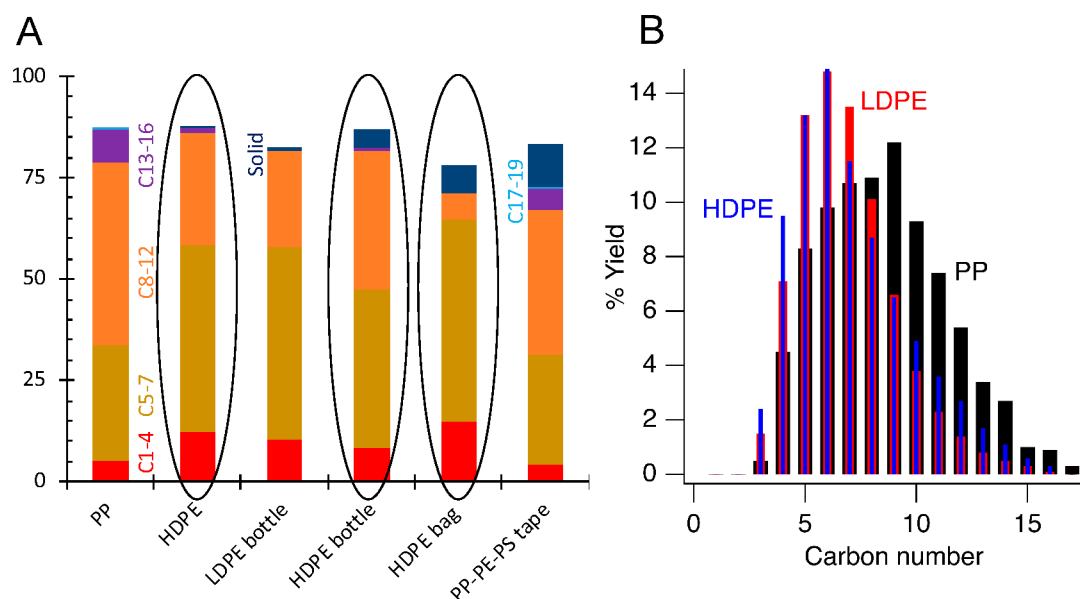
**Figure 13.** (A) Products from a dual catalyst approach for the conversion of pure PP, pure HDPE, LDPE bottle, HDPE bottle, HDPE bag, and composite (45 vol % PP/45 vol % PE/10 vol % PS) tape at 250 °C and 30 bar $H_2$. The three different HDPE materials examined are circled to illustrate the product dependence on the material source. (B) Detailed breakdown of the hydrocarbon products in terms of number of carbons. Data were extracted from ref 220.

Although there appear to be economic obstacles for widespread adoption of chemical recycling, regional production of monomers from waste plastics for circularity may have significant advantages from a LCA perspective with improved economic viability, in particular for PET. In 2017, 52% of the PET production capacity of 30.3 million tons worldwide was in Asia.[214] The shale gas boom in the United States has reduced the availability of terephthalic acid (PTA), which is traditionally obtained from the naptha cut in cracking of heavy hydrocarbons in the manufacture of ethylene. This shift has been blamed for recent supply disruptions in PTA in North American. Domestic supply chain capability for resilience is generally desired, but this is challenged by the lack of infrastructure and feedstock availability in North America and the economic disincentives from an overcapacity of raw materials in Asia. Advances in chemical PET recycling could provide supply chain security as well as energy and pollution savings from minimizing the shipment of PET overseas. However, adoption will likely require the costs for the PET to match the delivered price of PET from petrochemical sources.

## 5. CHEMICAL CONVERSION OF PLASTICS TO VALUE-ADDED CHEMICALS

Since the thermodynamics associated with depolymerization for polyolefins indicates significant energy requirements, the value of the light olefins is low, and the depolymerization tends to produce a distribution of products, there is need for other chemical approaches to address this major component of plastic waste. The conversion of waste plastics to other chemicals offers the potential for the upcycling of the carbon in low value polyolefins to higher value organics. Thus, the energy requirements associated with depolymerization to monomers can be avoided and conditions associated with the cracking of hydrocarbons can be used to produce a variety of chemicals.[162] This assertion is borne out by a recent technoeconomic analysis of waste polypropylene (PP), which has indicated that fast pyrolysis to produce heavy gases and oils is favored unless

there are tremendous breakthroughs in mechanical recycling of PP.[215] Design of catalysts and the details of the plastic feedstock can be used to tune the products being produced,[166] e.g., from fine chemicals to fuels.[192,216] These processes tend to rely on pyrolysis to convert the plastics into liquid organics. An industrial process has been implemented by RES Polyflow LLC to convert postconsumer plastic waste directly to petroleum mimics.[217] The process uses multiple reactor zones to convert the plastics to a variety of chemicals in >90% yield to a liquid that mimics crude oil and can be processed with existing petrochemical infrastructure.[217] Due to the prevalence of polyolefins in the waste streams, pyrolysis tends to produce alkane-rich streams with characteristics best suited for fuels (after additional cracking).[218,219] There have been some examples of partial depolymerization to oligomers that provide raw materials to other applications.[139,177] In general, the economic incentives for chemical recycling of commodity plastics to monomer have been limited by the low cost of the monomers from petroleum products.[139] This has driven most commercial efforts associated with pyrolysis of waste plastics to focus on fuels as the product.[162] Recently, a dual catalyst approach has been demonstrated to convert polyolefins to fuels and light lubricants at modest temperatures.[220] Figure 13 illustrates the product distribution from a number of different polyolefins. The approach generally avoids overcracking of the polyolefins to low value, light alkanes ($C_{1-4}$), but there is an increase in the residual solids for the commercial plastics, potentially due to additives, and in the case when other plastic (PS) is present.

However, this ultimate combustion of the carbons from the plastic is detrimental to the circular materials economy as the molecular structures are destroyed to yield $H_2O$ and $CO_2$.[118] The generation of fuel from waste plastics is an intermediate approach between recycling back to a usable material and the common incineration for energy recovery approach (Figure 3). Davidson et al. have provided a critical review of chemical recycling from the perspective of life cycle assessment.[221]

Beyond fuels, appropriate cracking catalysts can transform polyethylene into liquid lubricants and waxes.[222] Fine chemicals, such as alkylaromatics, can be generated from polyethylene via tandem catalytic conversion.[223] However, the reaction rates for the catalytic conversion of polyolefins in the melt tend to be sluggish due to polymer diffusivity limiting catalyst turnover. A recent review has highlighted pyrolysis technologies for polyolefins and provides additional details on catalysis, reactor design, and material integration in existing refinery processes.[224] For a more detailed description of chemical recycling of common postconsumer plastics into fuels and chemical feedstocks, interested readers are directed to several recent reviews.[162,225,226]

## 6. KEY CHALLENGES AND OPPORTUNITIES

Despite the challenges associated with plastic waste, there are clear societal benefits to the utilization of plastics. Prime examples include the improvement in hygiene in healthcare enabled by sterile plastic packaging as well as the improvements in fuel economy of automobiles through lightweighting with plastics and plastic composites replacing metal parts.[227] Trends to improve the sustainability of plastics have focused on the circular economy with an end goal of a closed loop industry for plastics where the input to new plastic materials is solely from waste plastics. Much research has been devoted to fundamental studies to develop new paths in the lifecycle of polymers that could integrate better into a circular economy.[215,228,229] However, beyond these technical challenges, there remain questions about the economic and environmental realities. A value chain analysis of the plastic recycling market in the Nordics identified several key reasons for low demand of recycled plastics: price, traceability, and design deficiencies in the recyclability of plastics from products.[230] These aspects relate to the economics of the processes involved in recycling and the quality of the recycled product, which in general are intertwined.

An appropriate assessment model that accounts for material value, economics and environment is key to provide insights into decisions about what is (are) the most sensible route(s) to address plastic waste. For example, the consideration of the energy and material worth of constructed C−C bonds in plastics in comparison to $CO_2$ from its incineration will influence decision trees.[118] In some cases, the incineration with waste energy recovery can have lower environmental impact than mechanical recycling for addressing plastic waste.[231] A recent study found that the optimum for waste management of plastics is recycling only 25% of plastics while the remainder is better suited for waste-to-energy incineration.[232] With the variety of recycling routes and potential products, considering economic and sustainability issues during process design using an optimization algorithm provides a route to assess the favorability of different route to recycle plastics. Even for the pyrolysis of HDPE, the selection of processes can significantly alter the economic viability and environmental impacts.[233] Figure 14 illustrates the trade-offs between economics and environmental impact for this process with a major improvement in the sustainability possible with only a small impact on the economics. However, this may be challenging to implement for some publicly traded companies as activist hedge funds can influence the corporate boards into avoiding strategies that do not maximize profits as well as the direct tying of executive compensation to short-term profits.[234]
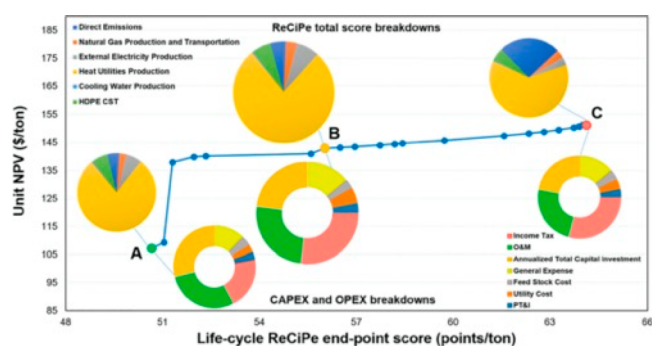


**Figure 14.** Illustration of the trade-offs between economics and sustainability for the pyrolysis of HDPE. The most environmentally friendly design is shown by point A where a lower ReCiPe score (a standardized method for calculation of life cycle impact) is favored. Conversely, the most profitable (highest net present value, NPV) configuration is shown as point C. A small hit to the return on investment can dramatically decrease the environmental impact (point B). Reproduced with permission from ref 233. Copyright 2020, American Institute of Chemical Engineers.

Even if mechanical recycling is the best from a technoeconomic perspective, the question of improving plastic recycling rates is multifaceted and requires examination of challenges from all viewpoints. One of the first challenges is the wide diversity of polymers, additives, fillers, and other components that comprise the plastic waste that is dominated by packaging with short lifetime. This creates a challenge for recycling as the heterogeneities in the waste stream can lead to high variability for mechanical recycling and can alter the product distribution with chemical recycling. The recent California recycling commission report included a recommendation to develop policy proposals that eliminate some packaging that impedes recycling.[235] With the size of the economy of California, implementation of these policies may decrease the overall variety of plastic generated in postconsumer waste to ease some of the challenges in recycling.

For postconsumer waste, there are two problems associated with decisions made by individuals about curbside recycling programs. First, materials that could be recycled can be landfilled to decrease the fraction of materials recycled. Potentially even more problematic from an economic perspective is the inclusion of plastics that cannot be recycled and contaminants in the plastics[38] in the mixed recycling streams,[236] which necessitates significant added costs associated with sorting and disposal of the waste materials. Sorting technology of plastic waste is key to the efficacy of both chemical[50] and mechanical[237] recycling. Low level contaminants can be nearly impossible to remove, and this tends to degrade the quality of mechanically recycled plastics.[38] There have been efforts to separate plastic out of standard municipal waste, but it is unlikely to ever be economically viable.[238] There are potentially some opportunities for informal sorting efforts that could help to build toward improved recycling efficiency in developing countries.[239] Irrespective of all these facts, the efficient sorting of plastic waste is a critical shortcoming in current technologies, despite significant improvements in the past years.[240] Alternative methods associated with selective dissolution−precipitation improves the product quality at additional costs;[127,128] scaled processes associated with selective dissolution are being commercialized by Polystyvert,[241] APK, and Creacycle. It remains to be seen if

this solution-based separation methodology will overcome the shortcomings of mechanical and optical sorting to produce higher quality recycled plastic products at a cost that the market will bear.

The intrinsic variability in the plastics being recycled leads to similar variation in the properties of mechanically recycled plastics. This variation is attributed to differences in molecular mass of the polymers in the recycling stream, additives, and the efficacy of the sorting. Examination of the product portfolio of Avient (formerly PolyOne), which is one of the largest plastic suppliers to end-users, shows that there are more than 4000 grades of resin currently being offered, which does not include the variation in the variety of additives associated with their Masterbatches to supply plastics to a variety of industries, including those with a strong focus on packaging. This variability is predominately in the molecular weight distributions, which are tuned to the application to provide high throughput for production while also producing the required mechanical performance of the plastic part. The engineered nature of plastic resins to match processing and application can lead to some challenges in the performance of mechanically recycled polymers due to the uncontrolled nature of the mixtures in the recycled product, even with perfect sorting. These subtle differences sometimes drive manufacturers with different colors for their product to sometimes use different sourced resins as this can influence the color of the product, even when the resin is technically the same polymer. These factors can lead to variation in performance of recycled plastics that tends to limit applications for the recycled product with downcycling of the plastic to a lower performance application.[177] Removal of additives through dissolution– precipitation based separations appears to improve the performance of mechanically recycled products,[117] but this will need to be further investigated for a broader range of plastics to understand how mixtures of base resins from different suppliers interact to influence the properties of mechanically recycled plastics. An alternative route to remove some additives (in particular, ink and dye) from recycled plastics is being commercialized by Cadel Deinking through surfactant-based extractions.[242] Depolymerization to recycle monomers in a circular materials economy will avoid any challenges with the mixed molecular weight distribution, but the impact of the variety of additives on the (catalytic) depolymerization reactions will remain a challenge to the efficacy of chemical recycling.

Beyond these technical and economic challenges, there are regulatory issues that can influence the favorability of recycling of plastic waste. For example, the EU has implemented a series of policies that promote the development of a circular economy for plastics.[243] The packaging industry has responded with improved integration of the plastic supply chain to enhance the sustainability of the plastics used.[10] Both the EU and California have both been aggressive in their policies and regulations to promote recycling and the vision of a circular materials economy for plastics, which will impact the viability of certain recycling processes as well as the use of some plastics in impacted applications. Regulations can also adversely impact the potential uses of recycled plastics. For example, the contamination and regulatory requirements associated with food contact represent a significant hurdle for the closed loop recycling of food packaging.[237] These regulatory factors present opportunities to dramatically shape how plastics are used and recycled, but there is also a danger of unintended

consequences.[8] The bans of single use plastic bags have been popular with local governments,[9] but these complete bans can actually be unfavorable from a LCA perspective if water use is considered, due to water use for the production and cleaning of multiple use natural textile bags.[11] The use of regulations and mandates[244] is likely to be a critical factor to the success of initiatives to increase the recycling rates for postconsumer plastics, as the economics to promote recycling has been lacking historically.[240]

## 7. CONCLUSIONS

There has been tremendous progress made in developing new materials and processes for the recycling of plastics in the past decade with the mounting pressures associated with the ever-growing plastic waste generation. However, challenges remain to address the limited recycled fraction of postconsumer plastic waste. Many of these are rooted in economics as one of the drivers of the large scale use of certain plastics is the combination of low costs and properties to meet the requirements of the application. This is particularly true for polyolefins, which comprise nearly 60% of the plastics produced annually. Low cost routes to recycle polyolefins mechanically tend to lead to inferior plastics, while chemical recycling offers the potential to produce indistinguishable plastics but the costs may not be competitive with those for virgin resins. This cost competitiveness with polyolefins is particularly challenging in the era of shale gas, which has dramatically decreased the cost of ethylene. From the fourth quarter of 2017 to the fourth quarter of 2019, the price of polyethylene dropped from $1.488/kg to $1.036/kg.[245] This reduction in value of polyethylene places added strain on its effective recycling, but also limits economic pressure to reduce plastic use. Historically, the mechanical recycling of natural HDPE is an exception where the properties obtained tend to be suitable for some applications, which is responsible for the relatively large fraction of HDPE that is recycled mechanically in comparison to other polyolefins. Development of low-cost processes (competitive economically) to produce recycled LDPE and PP with properties comparable to the virgin resins is a key challenge to increase the fraction of plastics that can be readily recycled. The complex nature of the plastic formulations is also a challenge with the variety of additives, fillers, pigments, etc. used in commercial applications. The development of any new technologies for plastic recycling needs to take into account the complexities of the plastic waste as the different additives can adversely impact the quality of the recycled product or the recycling process itself, especially for chemical recycling. Low-cost removal of these compounds or understanding of the influence of their interactions on mechanical properties or catalyst performance for mechanical and chemical recycled plastics, respectively, would greatly benefit the ability to produce high performing recycled plastics. Additionally, the variability in the properties of the recycled plastics significantly limits their applications. This variability arises from the ever changing, diverse, postconsumer plastic waste feedstock for the recycling. Routes to improve the uniformity in properties from a highly variable feedstock (waste plastic) would provide new applications for recycled plastics. The redesign of plastics toward intrinsic chemical recycling would provide a simple solution to enable a circular materials economy, but the cost of these plastics in comparison to polyolefins is likely to limit their adoption in high volume applications, namely packaging which accounts for a majority

of the plastic waste generated annually. The wild card for the adoption of redesigned plastics is government policies and regulations, which could through mandates require full circularity where the finite nature of mechanical recycling would limit commodity polyolefins. The recently implemented EU tax on nonrecycled plastic packaging waste provides some economic advantages to improving recyclability of packaging materials, but this is likely insufficient to cover the added costs over polyolefins at only €0.80/kg of nonrecycled waste.[246] The complexity of postconsumer plastic waste will likely necessitate a multipronged approach with technoeconomic analyses critical to directing the most appropriate recycling processes (mechanical, chemical to monomer, and chemical to high value chemicals).

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## ■ ABBREVIATIONS

BHET, bis(2-hydroxyethyl) terephthalate; BPA, bisphenol A; DMT, dimethyl terephthalate; EU, European Union; HDPE, high density polyethylene; LCA, lifecycle analysis; LCST, lower critical solution temperature; LDPE, low density polyethylene; LLDPE, linear lower density polyethylene; NPV, net present value; PDK, polydiketoenamines; PET, polyethylene terephthalate; PP, polypropylene; PS, polystyrene; PTA, terephthalic acid; PUR, polyurethane; PVC, poly(vinyl chloride); rPET, recycled PET

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