REVIEW

Toward polymer upcycling—adding value and tackling circularity

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Plastics have revolutionized modern life, but have created a global waste crisis driven by our reliance and demand for low-cost, disposable materials. New approaches are vital to address challenges related to plastics waste heterogeneity, along with the property reductions induced by mechanical recycling. Chemical recycling and upcycling of polymers may enable circularity through separation strategies, chemistries that promote closed-loop recycling inherent to macromolecular design, and transformative processes that shift the life-cycle landscape. Polymer upcycling schemes may enable lower-energy pathways and minimal environmental impacts compared with traditional mechanical and chemical recycling. The emergence of industrial adoption of recycling and upcycling approaches is encouraging, solidifying the critical role for these strategies in addressing the fate of plastics and driving advances in next-generation materials design.

The rapid rise in global plastics production is largely driven by increasing consumer demands for resilient and lightweight materials, such as in single-use plastics (SUPs), that offer convenience and enhanced functionality. These desires have driven a migration toward multicomponent plastics, particularly in the packaging industry, along with the production of intricate plastic parts facilitated by innovations in manufacturing technology. Complicating this snapshot is the recent surge in SUP usage, including medical packaging, masks, gloves, containers, and utensils, during the ongoing COVID-19 pandemic (*1*, *2*). These pandemic effects may not be long-lasting, but they will certainly influence the plastics waste problem in the immediate future. Unfortunately, because of the increasing demands and complexity associated with plastics production, less than 10% of plastics are recycled world-wide, with less than 1% of plastic being recycled more than once, and only ~12% of plastics are incinerated (*3*). This lack of circularity highlights a serious threat to the environment owing to the escalating prevalence of plastics waste in landfills and aquatic environments. For example, there are over 7.4 billion metric tons of plastic in the Earth system; this number is expected to rise to 40 billion metric tons by 2050, with ~10 million metric tons of that plastic reaching the oceans every year (*4*). The amount flowing into the oceans is dwarfed by the 2.5 billion metric tons of plastics currently in use (*3*); thus, with each passing year, the flux of plastics to surface waters, coastlines, and oceans is amplified—as are the environmental, social, and economic consequences.

Plastics are difficult to recycle from postconsumer waste because of the need for separation prior to reprocessing, and from a raw cost perspective, it is typically cheaper to manufacture new, disposable plastic packaging from virgin feedstocks than to sort and reuse reprocessed material (*5*). This cost differential is exacerbated by the fact that waste disposal infrastructure varies by location, and plastic ends up in the environment through leakages from waste collection, recycling, and disposal systems, or the absence of those systems in general (*3*, *6*).

One major culprit of environmental pollution, SUPs, presents a critical conundrum given the copious volume of plastics waste generated by their use. SUP use has enabled advances in medical technologies and increases in food shelf lives and led to associated reductions in transportation costs and the concomitant lowering in harmful emissions (*7*). Hence, if these materials are banned, there may be substantial and unintended consequences that can have health and socioeconomic impacts across the globe (*4*). In short, one needs to develop and implement policies that minimize SUPs when feasible, while also advancing sustainable reuse and refill, recycling, upcycling, and degradation and composting approaches for cases in which SUPs and other plastics have important economic and performance advantages.

In developing a framework around sustainability, a key challenge is terminology. Recycling typically denotes the reuse of a previously processed or waste material; upgrading refers to the addition of value to an existing chemical or material; upcycling suggests the upgrading and reuse of an existing chemical or material; and circularity implies the act of keeping chemicals and materials in the value chain. However, there is nuance in each of these terms. For instance, the word "recycling" can encompass vastly different processes and outputs depending on the context (*8*–*11*). If one views the recycling processes as a black box, two distinct cases can be considered. First, recycling can describe the separation and deconstruction of a polymer form factor (e.g., bottle, bag, part) through a mechanical approach that usually involves considerable bond breaking of higher-molecular-weight polymer chains. The output is a polymer that normally has reduced toughness, modulus, and other structural characteristics relative to the parent material. Thus, a sizable fraction of virgin (fresh) polymer must typically be added, if the recycled plastic is intended for reuse in its original application (*12*). There are chemical recycling-based dissolution approaches that have potential to address this property degradation issue, but solvent and energy constraints currently limit scalability. Second, recycling can refer to the chemical (pyrolytic or catalytic) or biological (enzymatic) breakdown of plastics into primarily small molecules (*13*–*15*). The depolymerization outputs are commonly used as chemical feedstocks, fuels, and lubricants, but they are rarely reused as precursors to new polymers (*10*), especially in the processing of polyolefin-rich sources (*13*). Although both instances above involve the potential reuse of polymer-derived carbon atoms, a case can be made that neither route is true recycling, in which a plastic material is fully used again for the same application. By contrast, there is a commercially viable route to Nylon-6 made from regenerated caprolactam monomer, exemplified by the Econyl process (*16*), that adds value to chemically recycled (upcycled) carpets and fishing nets through the combination of a "green" marketing opportunity and resilience in materials properties.

There has been substantial focus on the upcycling of polymer waste to increase functionality or add performance (*17*). Although upcycling is a valuable strategy in the polymer sustainability portfolio, a few obstacles should be considered: (i) In many cases, upcycling can be more accurately described as upgrading because the resulting products still need to enter the recycling infrastructure; (ii) the volume of a particular plastics waste stream is typically an order of magnitude or more greater than the market size for the corresponding upgraded materials, hence, a single upgrading approach is likely to oversaturate a valuable market; and (iii) the net costs, energy inputs, and environmental impacts associated with upgrading may be greater than those associated with creating the same product from virgin material (*5*, *18*).

In short, one needs to consider a suite of recycling, upcycling, and complementary routes that minimize energy inputs, cost, and environmental impacts to create a valuable output. Furthermore, many of the current approaches

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do not necessarily result in closed-loop processes (e.g., processes in which polymers or polymer by-products are not released into the environment) and instead, "kick the can" several years down the road. Even for those that are approaching closed loop, eliminating losses remains a challenge, particularly for polyolefins, and interactions among polymers, catalysts, and additives in both homogeneous and mixed-waste chemical recycling are major contributors to process inefficiencies.

Highlighting the complexity of plastics waste

The low rate of recycling is exacerbated by increasingly mixed-waste streams that can include polymers of heterogeneous compositions sourced from various petroleum- and bio-based feedstocks (*9*, *19*, *20*). Although the complexity can stem from multilayered and additive manufactured constructs and mixed-waste streams, the compositional heterogeneity of a single-polymer waste stream should not be overlooked. For example, such a stream may contain functional additives (e.g., foaming agents, stabilizers, flame retardants, antistatic agents, plasticizers), colorants (e.g., pigments, dyes), and fillers (e.g., glass or carbon fibers, silica, calcium carbonate) (*21*). Thus, even a relatively simple, "single-component" waste product may harbor agents that kill enzymes and microbes, poison catalysts, migrate into water and soil upon mechanical processing, release harmful by-products at high temperatures, and hamper demixing efforts (*19*, *20*). Hence, it is prudent to consider not only the type(s) of polymer, i.e., high-density polyethylene (HDPE) and low-density PE (LDPE), but also the other components that comprise the material formulation. The continued development of plastics derived either fully or partially from bio-based sources adds further complexity to the recycling or upcycling landscape (*22*, *23*). It is worth noting that two classes of bio-based polymers can be featured—one, in which a bio-based feedstock is used to generate a monomer, and hence a polymer, that is chemically identical to the petroleum-derived analog, and another, in which a bio-based feedstock is employed to produce a monomer, and hence a polymer, with similar functional performance but with a chemically distinct structure [e.g., usually increased heteroatom (oxygen) content]. The first case has no mechanistic impact on the recycling or upcycling process, whereas the second case can introduce considerable materials heterogeneity to the waste stream. Thus, even though the ultimate potential for biosourcing or biodegradation can be advantageous (e.g., increased opportunities for composting or anaerobic digestion, reduced necessity to clean food residue from waste plastics), the gradual inclusion of bioplastics in mixed-waste streams can lead to a constant compositional evolution that complicates established waste treatment approaches (*11*).

The above scientific topics also intersect with policy considerations, such as differences in recycling mandates by region. Although most waste volume comes from plastics packaging in developed countries (*6*), the recycling policies (e.g., multistream versus single-stream or commingled waste collection) may vary widely across national, state, and municipal levels. Single-stream collection has led to increased consumer adoption of recycling initiatives and higher recycling rates (*24*), but this recycling policy has a direct impact on the material and energy inputs (e.g., separation requirements, multicomponent streams, impurities) that affect recyling and upcycling strategies. Thus, the interplay of consumer habits and waste management policies highlights the global effort required to tackle plastics waste pollution. Powered by technoeconomic and life-cycle analyses, valuable opportunities exist to coordinate regulatory frameworks, industrial incentives (*11*), and plastics waste infrastructure to maximize efforts toward the valorization of plastics waste.

Circularity and upcycling through synthetic innovations

For plastics and other classes of polymers to positively contribute as renewable resources to the economy, a shift toward circularity is needed. Circularity in polymer life cycles is a challenge of thermodynamics, both in the sense that no process is 100% efficient, and every process is irreversible. Furthermore, circularity in polymer recycling cannot be achieved with mechanical recycling alone owing to the chemical heterogeneity of waste streams and property deterioration; thus, chemical approaches must continue to emerge. Chemical recycling offers the conceptual ability to remove additives efficiently, and recover and refine monomers and polymers for remanufacturing; however, it is somewhat difficult to design complete pathways in a manner that requires a lower energy input than the synthesis of the polymers from virgin, small-molecule sources.

For chemical recycling by depolymerization, innovations in both catalysts and processes often go hand-in-hand. These innovations have enabled circularity opportunities for poly(ethylene terephthalate) (PET) via catalytic solvolysis (*25*) (e.g., methanolysis—Loop Industries, Indorama, and others; glycolysis—IBM and Axens) and enzymatic hydrolysis (Carbios) (*26*); note that PET is one of the easiest polymers to either mechanically or chemically/enzymatically recycle. Similarly, HPDE can be deconstructed catalytically by pyrolysis to naphtha, which reenters the manufacturing cycle for ethylene monomer and PE resin (BASF, Sabic, Plastic Energy, and others). Pyrolysis also is an effective strategy for the depolymerization of polystyrene (PS) to styrene monomer (Ineos Styrolution, Agilyx, and others), and in the special case of Nylon-6, chemical circularity has been achieved using ring-closing depolymerization (BASF, Aquafil, and others) (*27*). These thermally driven, chemical recycling processes remain energy intensive, often are intolerant to common polymer additives or impurities, give broad product distributions, and have relatively low yields of higher-value products, all of which hamper scalability. Additionally, given the intensity of the thermal processes required, current approaches are projected to generate large amounts of greenhouse gas (GHG) emissions.

For chemical recycling by dissolution–precipitation, advances have instead focused on identifying solvents for specific polymers that are nonsolvents for other polymers in mixed-plastic waste streams, as well as finding sorbents that can dissociate polymers from additives. Notably, the dissolved and precipitated polymer must be dried before repelletization for remanufacturing, which can be energy intensive. Currently, dissolution–precipitation has gained industrial traction for the recycling of isotactic polypropylene (iPP) and PS as practiced by PureCycle and Polystyvert, respectively. Selective dissolution–precipitation also has been shown to be effective in the deconstruction of multilayer films, e.g., comprising PE and PET and a soluble tie layer between them (*28*). However, there are open questions as to whether similar processes can be broadly applied to other commodity or specialty polymers. It is also unclear whether dissolution–precipitation processes are lower in life-cycle energy or GHG intensity than competing thermal depolymerization approaches. Specifically, whereas chemical recycling to monomer consumes energy by breaking chemical bonds and through refinement of chemical and monomer feedstocks, dissolution–precipitation methods require energy for evaporation and solvent recovery. A comparative study of the life-cycle impacts for depolymerization versus dissolution–precipitation processes would clarify the benefits and trade-offs of each route in pursuit of meeting global sustainability goals.

There are a growing number of new polymer chemistries (Fig. 1) for which circularity in chemical recycling is a part of the macromolecular design (*9*, *29*, *30*). The incorporation of bonds or implementation of catalysts to tailor the energetics of addition–elimination reactions underlying solvolysis or hydrolysis of condensation polymers, similar to the characteristics of PET, has emerged as a powerful strategy for deconstructing polymer chains to linear monomers. Such processes have facilitated the closed-loop chemical recycling of linear and networked conventional polyesters, polycarbonates, and polyimines, as well as new polymers based on polydiketoenamines (*7*). Notably, polydiketoenamines hydrolyze at ambient temperature in mixed-waste streams, and life-cycle assessments

have shown that the GHG and energy intensities associated with monomer recovery and refinement are lower than those for the primary production of commodity polymer resins (*31*). Similarly, and analogous to caprolactam recovery from Nylon-6, exerting kinetic control over ring-chain equilibria has been leveraged for efficient recycling of emerging classes of polymers, such as polydisulfides and polycycloolefins, from cyclic monomers (*32*). These recent demonstrations highlight how one can depolymerize polymers by inverting the thermodynamic stability of the monomer relative to the polymer as dictated by the temperature, monomer structure, polymer chain tacticity, chemical environment, and, in some cases, the catalyst. However, an added complexity arises when several monomers are combined in a copolymer to tailor properties for a specific end use. Upon depolymerization, a mixture of monomers is obtained, and refining this mixture requires processes that effectively manage the materials entropy along with the chemical entropy associated with making and breaking bonds to achieve a more sustainable life cycle (*33*).

Athough recycle-by-design concepts are still in their nascency, the market remains uncertain, if not skeptical, regarding the prospects of introducing new polymers to make products more sustainable. This position is warranted; it has taken decades to tailor commodity polymers and additives to yield manufacturable formulations that perform well in their desired end uses. Also uncertain is whether recycle-by-design concepts can advance in stride with a transition to more sustainable, bio-based feedstocks for the chemicals needed for resin production, addressing the growing market pull for bio-based circular plastics. In this regard, pairing polymer design for circularity with performance-advantaged bio-monomers may turn the traditional cost–benefit analysis of biochemicals in resin production on its head, toward disruptive and more sustainable innovations and solutions. It may be possible to use artificial intelligence and machine learning approaches to accelerate advances in polymer research and development, such as the identification of new atom- and energy-efficient strategies that hasten a transition to circular manufacturing.

Adding value beyond chemical recycling strategies

Another approach to upcycling or upgrading polymer waste is focused on innovative transformations that impart precise functionality and tailored material properties. The C–H bond has been heralded as a strategic target for property expansion of commodity polymers, such as PS, PP, polycarbonate, and poly(ethylene glycol), with chemoselectivity that facilitates upgrading of plastics waste, improves compatibilization, enables spatial patterning, and promotes the development of copolymers otherwise inaccessible through traditional synthetic methods (*34*). This suite of material diversification pathways builds upon recent advances in photoredox catalysis, radical-mediated techniques, and C–H activation (*14*, *34*, *35*). A key advantage of this upgrading strategy is that small amounts of functionalization lead to pronounced modification of material properties (e.g., adhesion, surface tension) without polymer chain degradation, generating functional materials without the synthetic complexity (e.g., monomer reactivity considerations) associated with copolymerization (*34*). These approaches also are readily applied to aromatic polymers. In the case of PS, upgrading via C–H activation to generate sulfonated polymers for electronic materials applications (*34*, *36*) may be particularly advantageous given the substantial gulf between PS's widespread usage in the food industry as packaging, containers, and utensils and its low rates of recycling because of sorting constraints and limited reuse (*12*, *37*, *38*). Moving forward in the context of life-cycle management and circularity, critical questions emerge: (i) Are these functionalization methods suitably tolerant to mixed waste or contaminated waste streams? (ii) What opportunities exist to install latent functionality to target both property tuning and closed-loop recycling? (iii) Are these upgraded materials now even more challenging to recycle and/or separate with conventional strategies? (iv) Is the market size for the differentiated upgraded materials commensurate with the supply of recycled precursor? and (v) Will polymers designed for easier recycling and upcycling generate hazardous by-products owing to their breakdown mechanisms, or will they have unintended failure mechanisms that affect their useful lifetime? Implementation of these upgrading and upcycling strategies in the current paradigm of the plastics waste dilemma will require strong academic–industry–municipality partnerships, with a systems-level focus to maximize innovation impacts from application and materials management perspectives.

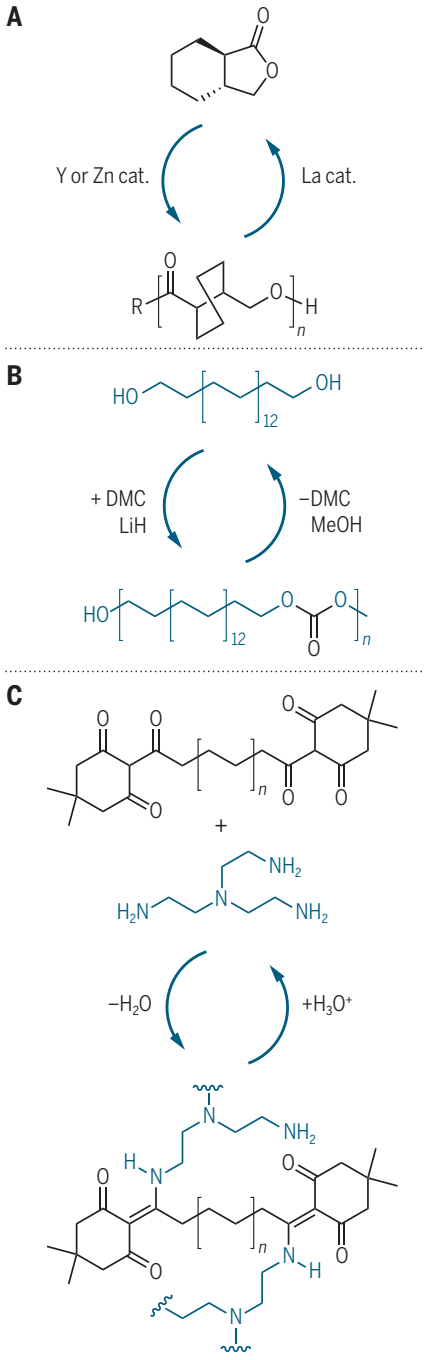

Fig. 1. Circularity in novel polymers. (**A**) Catalytic ring-opening polymerization and catalytic ring-closing depolymerization of a bicyclic lactone monomer (*29*). (**B**) Polymerization and solvolysis of a polyethylene-like polycarbonate using dimethyl carbonate (DMC) (*30*). (**C**) Polymerization and hydrolysis of a polydiketoenamine resin (*7*).

Striving for global equity

China's import ban on plastic waste in 2018 left much of the developed world with few options (*39*). The United States, Germany, the United Kingdom, and Japan initially redirected their waste shipments to Vietnam, Thailand, and Malaysia, where officials responded with their own restrictions to limit imports. Indonesia has emerged as the next epicenter for waste disposal, and the environmental impacts to crops, marine life, and air quality are growing. With global trade restrictions in place, however, more countries, provinces, and municipalities will be left to decide what to do with the growing piles of waste at home. Poor communities appear increasingly at risk to bear the brunt of the burden, and the toxic legacy of plastic waste in these communities likely will affect select regions for many years to come.

Much in the same way that the aluminum and glass industries have worked with communities to contribute to the development of recycling infrastructure for those materials, it may be time to do a similar exercise for

plastics. The European Union has been more forthcoming in this regard (*40*), where SUPs bans and extended producer responsibility regulations are in place, stimulating innovations and upgrades to materials recovery facilities, along with nucleating partnerships for managing logistics and accountability. In the United States, however, various headwinds have so far been able to stymie most legislative efforts at municipal, state, and national levels, despite general commitments to improving the sustainability of materials and products. There is growing sentiment to tax virgin resin or provide other economic incentives to level the playing field and spur increased incorporation of recyclates in resin production (*41*). For example, the United Kingdom has just introduced a new tax (*42*) that applies to plastic packaging produced in, or imported into, the United Kingdom that does not contain at least 30% recycled plastic, which is driving innovation and changes in practice. If such measures succeed in increasing recycling rates and lowering the volume of waste that is landfilled or incinerated in under resourced communities, a more equitable outcome may be within reach. Jobs creation tied to plastics recycling also could provide substantial economic benefits, factoring into the sustainability equation (*43*).

Building a more sustainable future

The plastics waste dilemma is a global challenge that requires urgent intervention (*11*, *44*) and a concerted effort that links partners across industrial, academic, financial, and government sectors buttressed by large investments in sustainability. Solutions must balance an appreciation for how advances in plastics technology have enhanced the human condition with necessary circularity considerations and life-cycle management strategies toward a sustainable ecosystem. Many current approaches tend to focus on singular aspects (e.g., waste management, process design, manufacturing strategies, catalytic innovations, or synthetic pathways) of the recycling and upcycling value proposition. Polymer upcycling will certainly be a critical piece of the solution, but it is not the silver bullet to address the entire plastics dilemma. It needs to be part of a plastics circular economy that embraces the varied forms of a reduce, reuse, and recycle philosophy. To achieve a more sustainable future, integration of not only technological considerations but also equity analysis, consumer behavior, geographical demands, policy reform, life-cycle assessment, infrastructure alignment, and supply chain partnerships is vital (*43*). Only by adopting a systems-based, multidisciplinary strategy can we implement effective and long-lasting solutions to the staggering amount of plastics waste and minimize the impacts of current and next-generation plastics.

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ACKNOWLEDGMENTS

L.T.J.K. and T.H.E. acknowledge the US Department of Energy, Energy Frontier Research Center program (DOE DE-SC0021166; Center for Plastics Innovation), and the National Science Foundation, Growing Convergence Research program (NSF GCR CMMI—1934887), for financial and administrative support during the writing of this manuscript. B.A.H. acknowledges support from the EERE Office of Strategic Programs and Advanced Manufacturing Office under US Department of Energy contract no. DE-AC02-05CH11231. A.J.R. acknowledges support from the Grantham Foundation for the Protection of the Environment and the UKRI Smart Sustainable Plastic Packaging project, Many Happy Returns - Enabling Reusable Packaging Systems (NE/V010638/1).The views expressed in this perspective do not necessarily represent the views of the US DOE or the US government.

10.1126/science.abg4503

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Science **373** (6550), 66-69.
DOI: 10.1126/science.abg4503