Sustainability of Synthetic Plastics: Considerations in Materials Life-Cycle Management

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ABSTRACT: The sustainability of current and future plastic materials is a major focus of basic research, industry, government, and society at large. There is a general recognition of the positive impacts of plastics, especially packaging; however, the negative consequences around end-of-life outcomes and overall materials circularity are issues that must be addressed. In this perspective, we highlight some of the challenges associated with the many uses of plastic components and the diversity of materials needed to satisfy consumer demand, with several examples focused on plastics packaging. We also discuss the opportunities provided by conventional and advanced recycling/upgrading routes to petrochemical and bio-based materials and feedstocks, along with overviews of chemistry-related (experimental, computational, data science, and materials traceability) approaches to the valorization of polymers toward a closed-loop environment.

KEYWORDS: Plastics, Sustainability, Circularity, Life-cycle management, End of life, Recycling, Plastics waste, Polymers

INTRODUCTION

Plastics play an indispensable role in every aspect of modern life from materials for aircraft, cars, and buildings to clothing and shoes, food and beverage packaging, and biomedical devices. Globally, ∼390 million metric tons of plastic products, including resins, were produced in 2016, and that amount is expected to double in approximately 20 years under various production scenarios.1,2 This increasing demand is a direct result of the burgeoning need for high strength to weight ratio, multifunctional materials.3 Unfortunately, less than 10% of plastics waste is collected worldwide for recycling,4 and even less is actually recycled, exacerbating the environmental impacts associated with landfill usage, degradation byproducts, and aquatic pollution, among other examples, with the weighting of the different impacts varying by region.5,6 Considered a critical tool in the repurposing pathway, mechanical recycling has several challenges with respect to circularity and practical end-of-life outcomes. Similar challenges exist as research efforts expand to incorporate (bio)degradation as a sustainability plastics pathway.7–13 While plastics are often viewed as monolithic, the drive toward durable, lightweight, and functional materials has led to the production of multicomponent systems of increasing complexity and diversity. This trend is most obvious in plastics packaging,14 which demands features such as oxygen barrier properties, mechanical protection, and sensing, and requires several polymeric components, often in a multilayer configuration—along with additives (organic and inorganic fillers) to seamlessly incorporate the requisite features.11,15 These multicomponent plastics necessitate an understanding of how the combination of macromolecules and small molecules contribute to plastics waste and affect downstream sorting and life-cycle approaches.

SETTING THE STAGE—FRAMING SUSTAINABILITY

The messaging around materials sustainability has evolved as more aspects of the plastics lifecycle have been considered. For instance, valorization can refer to increasing the value of a material; however, there is substantial nuance in the terminology. While many in the science and engineering realm consider valorization to describe the application of technology and innovation to increase the value of an output, a significant fraction of the economics (business and policy) community may consider valorization to be the application of government policies, such as regulation and taxation, to influence costs. Overall, the concepts are not mutually exclusive; however, they are different means to potentially achieve the same end. As another example, recycling is a well-recognized term for returning materials to the use stream.
term is sometimes misconstrued to imply solely traditional mechanical reprocessing that can be associated with poor sortation and with outputs that are only useable in lower-grade plastics. However, mechanical reprocessing can have comparatively low climate impacts, and there is substantial demand for mechanically recycled materials. As will be discussed below with a specific emphasis on packaging, mechanical reprocessing on its own cannot handle the volumes of plastics waste or the multicomponent nature of the waste, due to the diversity of formats (e.g., rigid vs flexible), properties of materials outputs, and market size relative to single-use packaging. Hence, additional routes are necessary to improve sustainability through life-cycle management strategies that mesh circularity and end-of-life design. In the following sections, we use the concepts of recycling and degradation (depolymerization or deconstruction) to provide context around circularity vs end of life, current approaches to sustainable plastics, headwinds or challenges, and future opportunities.

**MANAGING CIRCULARITY VS END OF LIFE**

In the context of sustainability, one can distinguish between circularity and end of life as shown in Figure 1. Circularity involves keeping atoms and materials inside the economic design engine and out of unwanted paths, such as landfill, terrestrial, and aquatic environments. Though there will always be losses and energy costs associated with circularity, the overall concept compares favorably to a linear approach that depends on greater extraction of natural resources and robust end-of-life management. Yet, it also is beneficial to develop end-of-life strategies such that plastics quickly and fully degrade under ambient environmental conditions or in industrial composting facilities. In the near term, however, these same degradable materials have the potential to devalue the dominant plastics waste streams through increased chemical complexity that stresses the collection and sortation infrastructure. One further, currently practiced, end-of-life option worth mentioning is incineration. While burning can lead to significant energy recovery, greenhouse gas and other emission concerns have led to incineration bans in several locations.

**CURRENT AND BURGEONING APPROACHES TO SUSTAINABLE PLASTICS**

Recycling strategies, including mechanical recycling, solvent separations, deconstruction or depolymerization (advanced or chemical recycling), and composting or biodegradation are several common options to improve the sustainability of plastics. The above routes can be potentially applied to both petroleum- and bio-based materials, but there is not likely a one-size-fits-all strategy across all systems. Mechanical recycling is a common approach for numerous plastics in the value chain; however, this route typically does not lead to a closed-loop framework, as polymer chains are usually cleaved during the process, resulting in a degradation of physical properties. Thus, the continued addition of virgin materials is necessary to reproduce consumer products with comparable performance. Furthermore, the additives in plastics affect “sortability” and result in increased levels of contamination within a recycle stream, which also increases separation costs and reduces the utility of recycled materials in higher-value applications. Chemical recycling can occur through pyrolytic, catalytic, enzymatic, or solvent-based routes. The general case of conversion of polymeric materials to small molecules is considered deconstruction, while the more specific case of conversion to monomers is termed depolymerization. Each of the chemical recycling approaches has advantages and disadvantages in terms of substrates, tolerance to impurities/additives and functional groups, yields/product distributions, energy usage, scalability, and environmental impacts. For example, catalytic deconstruction is an attractive approach for handling polyolefin waste. However, the complexity of real plastics waste brings the challenge of catalyst tolerance to impurities. Antioxidant and surfactant additives are known to have a negative effect on many catalytic deconstruction processes, as halogens can poison the most-used heterogeneous catalysts, while functional groups such as nitriles, acetates, and hydroxyls also are suspected to deactivate catalyst sites. Furthermore, it is worth noting that a typical aim for chemical deconstruction is that the outputs are reconstituted as fuels, chemicals, lubricants, and surfactants as opposed to reintroduced into existing materials streams to directly increase plastics circularity.

In addition to the degradation strategies mentioned above, one also can improve the resiliency and extend the service lives of traditional commodity polymers. Such life-extension approaches have significant value in the near term because they can be inserted into the existing supply chains and reduce the current rate of natural resource extraction, though they will
likely never be able to completely replace the need for virgin feedstock. As one example, compatibilizers have been developed to extend the useful life of certain polymers, such as PE and PP, by making the polyolefin constructs more tolerant to polymeric impurities found in plastics waste—facilitating property retention in recycled resins by preventing phase separation and brittle fracture.40–42 As another approach, researchers have focused on self-healing materials that can be repaired operando to extend a material’s overall lifetime; however, many dynamic polymer systems have challenges with creep resistance and fracture toughness at lower temperatures as a tradeoff to adequate reprocessability at higher temperatures.43–45 The drawbacks have motivated researchers to search for routes to mitigate the above problems in various polymer systems.

There also has been a significant shift toward plastics derived from renewable resources to decrease reliance on fossil fuels and reduce the associated plastics waste problems. Generally, two classes of bio-based materials can be established from a recycling/upcycling standpoint.46 The first is bio-based polymers derived from monomers that are chemically indistinguishable from their petroleum-based counterparts; examples include (bio)polysoprene47 and (bio)-polyethylene.48 A positive aspect of these materials is their renewable origin; however, the steps necessary to obtain these compounds from their biosources, relative to their petrochemical analogues, may have substantial environmental impacts that are much greater than the effects of sourcing virgin petroleum feedstocks.49 The second is bio-based polymers with monomeric and macromolecular structures that are chemically distinct from their petroleum-derived alternatives.50 Here, we focus on the latter class of materials, as the former can be considered identical to the petrochemical versions from a recycling, upgrading, or upcycling point of view.

The growth market for bioplastics is currently limited by challenges in the synthetic and manufacturing routes required to achieve properties comparable to those of conventional plastics, while retaining high degrees of degradability. There is also much debate about the classification of so-called sustainable plastics as inherently biodegradable, defined as deconstruction or depolymerization within weeks to months under certain conditions.46,47 Furthermore, the costs associated with comprehensive biomanufacturing processes typically lead to materials that are not cost-competitive with petroleum-based alternatives.45,48 Expanding our understanding of the connection between feedstock (renewable vs nonrenewable) selection, performance characteristics, component toxicity, and environmental impact is critical to defining the role of sustainable plastics in life-cycle management.59

As one salient example, poly(lactic acid) (PLA) is a recognizable polyester bioplastic on the market and is primarily employed in single-use applications, such as packaging and containers.50,51 However, PLA suffers from low toughness, high gas permeability, and limited heat resistance, restricting its application space and processability.52 Furthermore, PLA disposal options highlight some challenges in end-of-life management. First, the testing protocols and standards to designate “compostability” are not uniform across industry and academia.16,24,53 leading to disparate predictions of degradability and a need for standardization.53 Second, many compostable or degradable materials, PLA included, decompose best in industrial facilities with tunable thermal conditions that are not readily accessible across most geographic regions.59 Several strategies have been employed to address the mechanical and degradation hurdles associated with polyesters, including toughening via blending and reactive compatibilization, additive incorporation, and copolymerization,54,55 but these approaches have yet to optimize performance and degradation. As one example of a route to balance property improvement with degradation potential, polyester-18,18 is a recently developed bioderivable polymer that mimics many of the thermomechanical attributes of polyethylene due to its longer aliphatic regions in its backbone, yet it degrades through a mechanism similar to that of conventional polyesters.56,57 While this approach is promising, the ultimate product form factor also affects the time scales of degradation and adds to the challenge of formulating strategies for economically advantaged life-cycle management.

One expansion area in the bio-based realm is the utilization of non-food-source biomass, such as lignocellulosic waste.50 Variations in the lignin extraction process from biomass yield heterogeneous fractions that may complicate materials design and require specific fractionation or separation techniques.50,51 However, advances in the catalytic deconstruction of lignin have led to aromatic building blocks that can be cost- and performance-competitive replacements for bisphenols used in everyday thermoplastics, such as polycarbonates, with similar or superior attributes and the potential for reduced toxicity relative to incumbent macromolecules, and as rigid monomers for utilization in thermoset design.55,57 The inherent hydroxy functionality on these lignin-derivable precursors also provides additional pathways for materials construction and manufacturing. However, a key challenge that must be addressed is the lack of inherent biodegradability or recyclability in many lignin-derived constructs—yet, it is worth noting that many bio-based plastics sequester carbon dioxide in the best cases (and carbon in cases for which hydrogenation is a prominent step in bio-based polymer creation), so any potential release of greenhouse gases at end of life must be considered.58,59

### HEADWINDS TO SUSTAINABILITY EFFORTS

When roadmapping the sustainability of the plastics industry, there are several key factors to consider, including scale, complexity of package formats, and consumer perception of plastics. An estimated 9 billion tons of plastic has been produced since 1950; roughly half of this volume is utilized in the packaging industry, in which materials have lifetimes of <1 year.58 The sheer scale of the packaging industry makes it challenging to replace fully fossil based solutions in the near term with new material systems, and without economic penalty, while also maintaining the diversity of functionality achieved with the numerous polymer systems that exist today.

From a packaging standpoint, the plastics industry can be further segmented into either rigid or flexible packaging modalities. Rigid structures (e.g., water bottles, shampoo bottles, milk jugs) are traditionally a more favorable format for mechanical recycling infrastructures due to the ease of conveying them in facilities, along with the weight of material that is recovered for recycling.58 These material formats are predominantly monolithic in design with a small percentage of functional polymers, such as poly(ethylene-co-vinyl alcohol), for barrier needs. The above factors are illustrated in the relatively high U.S. recycling rates of poly(ethylene terephthalate) (PET) and high-density polyethylene (HDPE) bottles at ∼28% and ∼33%, respectively, in 2016.59
Flexible packaging formats are more challenging due to the lightweight aspect of the material, form factor in recycling centers, and the ratio of recoverable plastic to contaminants. The majority of the flexibles packaging industry is composed of polyolefin materials, functional additives and other components, such as polyamide, PET, chlorinated polymers and poly(ethylene-co-vinyl alcohol) (in form factors such as multilayers, blends, etc.), and even thin metal layers, which act as contaminants in recycling processes and impede inclusive attempts at circularity.\textsuperscript{62,63} The different components in the multilayer composites can interfere with the catalytic deconstruction as discussed above. The nature of composites also complicates sorting and mechanical recycling in comparison to rigid packaging; therefore, those plastics are mostly incinerated or end up in landfill.\textsuperscript{61} Novel recycling methodologies, such as selective dissolution\textsuperscript{62,63} and melt-blending with compatibilization,\textsuperscript{64} are recently reported in this area. In the general alternatives to address this challenge.

Yet still, the current status of flexible packaging recycling suggests huge ecological and economical potential for advances in this area. In the general flexible packaging case above, the estimated U.S. recycling rates were \textasciitilde 21\% for commercial flexible films and \textasciitilde 4\% for residential films in 2017; the reduced rates, especially for residential flexible waste, are largely a result of a less robust collection infrastructure, along with increased contamination and color (additive) content.\textsuperscript{85} We point out that these numbers can vary by location, as the flexible PE-based film recycling rate in the European Union was estimated to be \textasciitilde 23\% on the basis of 2020 information.\textsuperscript{66}

With respect to consumer sentiment, the mismanagement of plastics waste over several decades has fueled a rise in negative perceptions of plastics.\textsuperscript{6} The economic and environmental benefits, such as total carbon footprint versus alternative mediums, have been usurped by the visual impact of plastic leakage into the environment.\textsuperscript{67} A Harris poll conducted by Sealed Air in 2014 on consumer food waste indicated that the majority of consumers believe that the packaging material is more harmful to the environment than discarded food waste.\textsuperscript{58,69} Yet, life-cycle analysis data from the poultry industry, as an example, indicates that the packaging accounts for roughly 4\% of the total greenhouse gas emissions for the supply chain.\textsuperscript{70,72}

Another significant challenge to a robust circular economy is the economics of closing the loop. Manufacturers of virgin polymers have had decades to optimize production and position plastics as an economical alternative to paper, metals, and glass. The plastics recycling industry has not had the same opportunity to advance purification processes and scale to compete with virgin materials in terms of cost and performance.\textsuperscript{73,74} The added costs of collection, transportation, and sorting of the waste streams place further financial strain prior to arrival at recycling facilities.\textsuperscript{75,76} Depending on the methodology utilized to recycle the materials, circular polymer pricing can demand a significant premium versus its virgin counterpart.

Adding to the aforementioned hurdles, there is no unified global leadership to codify and coordinate plastics waste efforts.\textsuperscript{77,78} This leadership challenge is exacerbated by the accelerating rate of plastic usage, particularly in the midst of the COVID-19 pandemic, as well as the geographical disparities in resource allocation.\textsuperscript{79,80} Sustainability efforts have primarily focused on highlighting the problem of plastics pollution, but a cohesive approach to solution development has just started to gain traction. From a governance perspective, combating the regulatory hurdles and decentralized strategies, even across a single country, requires (1) communication of scientific information to inform decision making, (2) recognition of the current destabilizing economics and politics, (3) science-driven policy development, (4) appreciation of fluctuations and trends in consumer habits, and (5) investments in infrastructure and incentives to promote sustainability.\textsuperscript{78} The complexity of this problem presents a rich opportunity to integrate a systems-based approach, uniting disparate fields and diverse perspectives, to devise solutions.

Finally, there is a need to manage imposed timelines (real or implied) and expectations that can complement the pathway toward innovation. In essence, one should not expect sustainable materials to immediately meet the cost and performance metrics of incumbent plastics without the benefit of a robust research, development, and regulatory framework.

\section*{OPPORTUNITIES TO FACILITATE IMPROVED LIFE-CYCLE MANAGEMENT}

Several avenues exist for improved approaches to recycling and valorization of plastics waste. One opportunity is to match degradation time scales with performance windows and product shelf lives. As an example of this approach, “unzippable” polymers and polyolefin-like macromolecules with the regular incorporation of sacrificial or degradable bonds are valuable platforms.\textsuperscript{81–84} A major challenge is the need to maintain overall material performance while simultaneously reducing the cost/complexity of the macro-molecular syntheses. Specifically, when cleavable functional groups are added to macromolecules, those moieties often have a significant, or even dominant, effect on the physical and application properties of the resulting materials, unless the monomers are specifically tailored to maintain long enough aliphatic segments, as demonstrated in polyester-18,18.\textsuperscript{55} In recent years, additional strategies, such as the radical copolymerization of ethylene, have provided simpler approaches to access “unzippable” polyolefin-like polymers with appropriate densities of functional groups from an applications perspective.\textsuperscript{85} However, there are still significant opportunities for improvement in achieving higher molecular weights, greater yields, and lower costs in comparison to traditional polyolefins.

Additionally, one can incorporate triggerable chemistries to generate associative (vitrimer) or dissociative networks that theoretically can be re-formed numerous times without a loss in thermomechanical performance.\textsuperscript{86–88} Yet still, the tradeoff between mechanical performance and recyclability is an important conundrum in vitrimer design. Long relaxation times at the recycling temperatures and creep/deformation at the application temperatures also prevent broader utility, although with proper material design the issue can sometimes be mitigated.\textsuperscript{80,89} Furthermore, while the development and maturation of current chemical approaches are necessary, one should further consider targeted and energy-efficient degradation approaches that are not inherently built into the macromolecule, such as catalytic and enzymatic (and microbial) deconstruction routes, to complement the more prevalent pyrolysis technologies.\textsuperscript{84,85,112,13,31,46,70} In all cases, one must account for the relative volumes and demand for upgraded materials versus the supply of plastics waste from which those value-added materials are sourced.

As novel materials are developed to address the leakage of plastic waste into the environment, it is important to consider
the final product format, not just each individual plastic component. Furthermore, as consumer demand for recyclable packaging increases, designing for circularity, without compromising performance, becomes a critical criterion for new product development throughout the value chain. Using an example from the food packaging industry, rigid meat trays with overwrap or lidding films are highly engineered products that ensure food is not wasted throughout the distribution chain, while simultaneously maintaining food safety for consumers. These packaging components are designed for use across a large temperature range from blast-chiller conditions during processing to high-temperature storage scenarios during summer months. It is also important that new product design does not negatively affect operational efficiencies within production plants, including the denesting of trays, sealing times, and integrity during handling, along with stacking efficiencies and pack out for distribution to retail. To maintain package integrity throughout conventional distribution channels, "high-abuse" materials are incorporated to ensure that mechanical failure of the pack does not occur and lead to product spoilage. Because of these and other critical performance attributes, a seemingly simple package for fresh protein can contain numerous polymer families within a coextruded construct.

With so many pathways to consider in handling the return of materials to the supply chain with minimal environmental impacts, and with so many chemistries and processes to consider as part of the solution, the future is both exciting and daunting. Also, given the myriad of potential options and the almost infinite combination of plastics waste form factors, chemistries, additive/filler compositions, and modes of collection, overcoming the challenge of optimizing life-cycle management solely through experimental efforts is an immense and possibly cost-ineffective strategy. Thus, a critical element to success will be building off of the computational tools and machine learning methods that have been developed over the last several decades to accelerate materials discovery and design. Tools under frameworks, such as the Materials Genome Initiative (MGI), will need to be expanded to incorporate the effects of more complex chemistries in bio-based materials and aid selection of the most efficient recovery processes: e.g., different deconstruction or depolymerization chemistries versus solvation-based purification strategies. Though not a primary focus of this perspective, the lack of standardized methods for reporting and reposing data in polymer science is something to consider, as it is a major hurdle in MGI and a significant barrier to concomitant advances in materials and data science.

Beyond the tools necessary to innovate in chemistry and materials science of polymers, all of these pathways are intimately affected by economic considerations associated with manufacturing, markets, trade, etc. To support decision making, comparable tools that account for all these factors and environmental impacts (e.g., greenhouse gas emissions, water usage) must be developed that can move traceable data between organizations and stakeholders. Furthermore, the community should better integrate expertise in life-cycle assessment and techno-economic analysis so that greater trust, transparency, and accountability will support decision-making with respect to investments in infrastructure and policy. One tool of recent note is the Materials Flows through Industry (MFI) platform from the National Renewable Energy Laboratory that can aid in this process.

Tracing recycled plastic through any chemical manufacturing process is complex, especially because the nature of organic molecules makes them inherently difficult to track through operations such as pyrolysis and cracking. Tools that combine an understanding of chemical processes with the practical realities of modern chemical manufacturing, so that credible and traceable claims can be made about the incorporation of recycled materials into new products, are critical for the success of those pathways. Certification programs, such as Mass Balance Accounting (MBA), represent an emerging field for polymer engineering that combines concepts from chemistry and manufacturing with economics and data security. The development of MBA tools, which have been practiced in other industries for many years, is critical to the success of advanced chemical recycling of plastics, and the adoption of such frameworks may determine whether or not chemically recycled materials are valued, viable technologies in the marketplace.

Altogether, progress in the sustainable plastics arena requires a coordinated effort that transcends geographical location, incorporates initiatives that span government, industry, and academic sectors, and involves policies that promote consumer adoption. Governmental agencies have proposed new regulations (e.g., plastic bag bans, export bans), alignments with global commitment strategic plans, and additional measures (e.g., lower tax rates) to tackle plastics waste pollution. These global policy adoptions connect to public–private industry partnerships that utilize these frameworks to promote sustainability efforts, from supply chain management to new product design. From an academic perspective, significant investments from funding agencies have sparked massive research efforts in catalysis, chemistry, materials science, manufacturing, artificial intelligence, and process design toward sustainable materials innovations. If one considers the fate of plastics, it is critical that this research framework be expanded to encompass the spectrum of life-cycle management, including toxicity, policy, behavioral science, (socio)economic, and environmental impact assessments. Initiatives that foster the convergence of these aspects are vital in the race toward sustainable plastics. Multi-disciplinary scientific innovations, policy and regulatory leaps, and academic/industry alliances are only pieces of the sustainability puzzle. Shifting consumer demand for sustainable materials from appreciation to intentional practices and investments is the often-forgotten link.

THE TAKE-HOME MESSAGE

The structural and performance characteristics of polymers and plastics vary for different applications, especially in the packaging space, and each use necessitates unique and tailored macromolecular chemistries and architectures. As such, the route to sustainability is likely to be different for each polymer class, as no single solution exists. In some cases, the continued use of traditional building blocks is appropriate, provided that routes to long-term waste mitigation and life-cycle management are developed. For other cases, the cultivation of new building blocks is an opportunity to leverage bio-based and bio-inspired components, while also taking advantage of selective degradation and reversible chemistries that allow the reimagination of macromolecular constructs. In still other instances, there may be opportunities to use traditional feedstocks for new applications via upgrading approaches. For all scenarios, however, an understanding of...
chemistry and engineering combined with toxicity, environmental science, economic analysis, data science, materials traceability, and life-cycle assessment strategies are important for the design and utilization of materials with reduced and verifiable impacts. In short, persistence on all fronts associated with circularity in plastics is vital to ameliorate the current plastics end-of-life challenges.

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**Notes**

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