

Roadmap to Biodegradable Plastics—Current State and Research Needs

Koushik Ghosh* and Brad H. Jones

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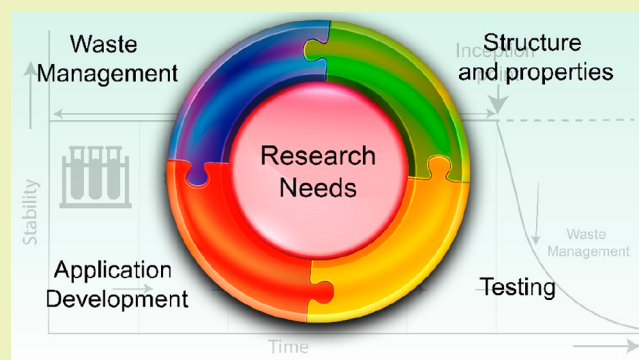
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ABSTRACT: Plastics, with their ubiquitous presence in our daily lives and environment, pose an uncomfortable conundrum. Producers and consumers are aware of the value of these organic ingredients in material flow, yet their persistence and disruption to the ecological milieu desperately stipulate a shift in the status quo. Biodegradable plastics—as the name suggests—has its appeal in ensuring the safe return of carbon to ecosystems by complete assimilation of the degraded product as a food source for soil or aquatic microorganisms. However, despite more than a decade of commercial presence, these plastics are still far from replacing the demand for fossil-fuel-based commodity plastics. We discuss this apparent disconnect herein through a material value chain perspective. We review the current state of commercial biodegradable plastics and contrast it against the desired state of the zero-waste-focused circular economy. To close the gap, we suggest critical research needs concerning the structure and properties of biodegradable plastics, testing standards, application development, and waste management. The ultimate success in displacing conventional plastics with biodegradable alternatives will be predicated on collaboration between all stakeholders across the product value chain.

KEYWORDS: Biodegradable, Persistence, Plastics, Certification, Degradation, Polymers, Microplastics, Waste management, Composting, Ecology, Plastic leakage, Environmental fate



1. INTRODUCTION

Plastics are the definitive symbol of consumer culture with their convenient, cheap, and versatile value proposition. However, plastics are also inexorably linked with persistency, toxicity, fossil fuels, and climate change.^{1–4} While plastics are not the sole culprit in the modern solid waste crisis, they are perhaps the most visible component. Single-use plastics^{5–7} exacerbate the problem because of their short life with the consumer. Although the useful lives of durable plastics—such as Lego blocks and reusable water bottles—may be as long as several years, the useful lives of others—such as candy wraps, sachets, or food wrappers—can be as brief as a few minutes. Fortunately, because of the sheer volume of plastics in solid waste streams, even relatively slight improvements can make a significant difference in the extent of the solid waste crisis.

There is no question that the accumulation of visible plastics—mountains of plastic articles in oceans, waterways, and fields—has captured the attention of scientists, media, and the general public for some time now. More recently, the accumulation of nonvisible plastics—so-called microplastics and nanoplastics—is increasingly understood to have perhaps even more dire consequences^{8,9} in the ecosystems. Conventional plastics do not easily degrade, in fact, they were never formulated

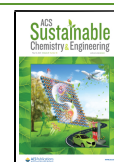
to degrade. In the marine or terrestrial setting, mechanical forces of waves, UV light, or the abrasive force of sediment grains may disintegrate the plastics article, reducing their physical dimension by physical or physio-chemical mechanisms without addressing the concerns of persistence. In contrast, biodegradation invokes an ideal vision of matter, lapsing back into nature without leaving a visible residue. Biodegradability draws attention to how things become “nonthings”.¹⁰ It is no wonder biodegradable plastics offer such promise.

As public awareness around environmental issues such as marine plastic pollution and global warming continues to grow,^{9,11–13} biodegradable plastics are rapidly advancing in consumer markets. Still, unanswered critical issues have hampered the progress of biodegradable plastics. They have been commercially available for several decades; however, this niche market is challenged with a variety of headwinds such as

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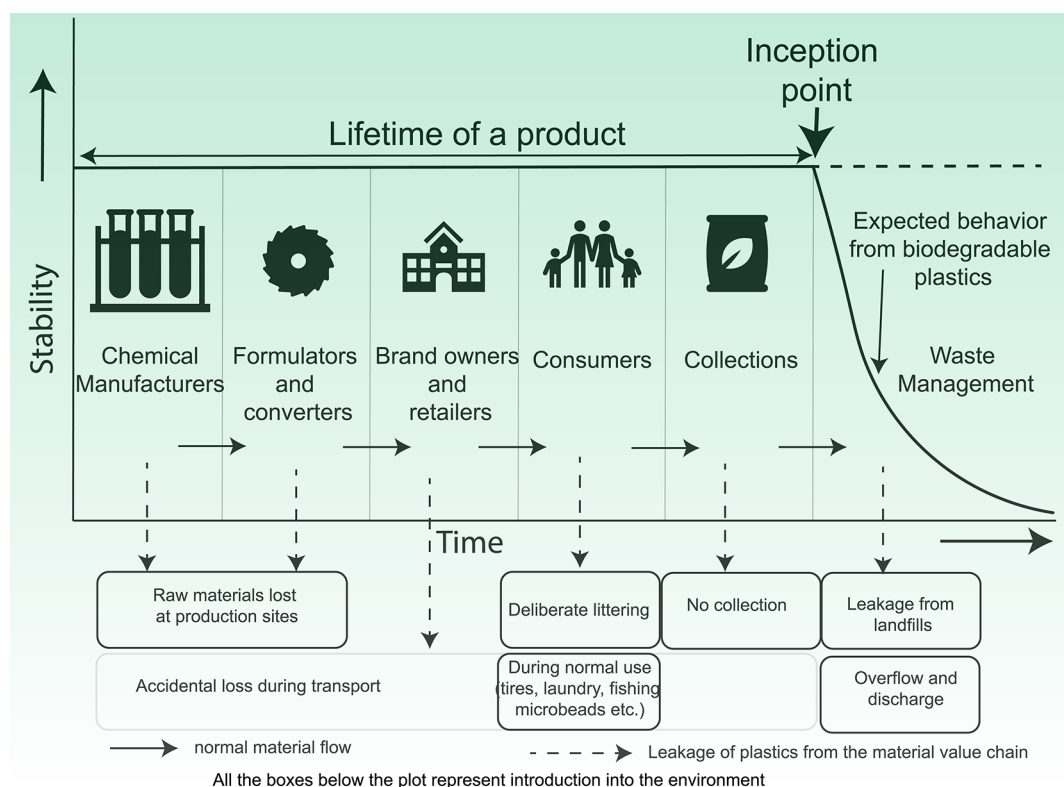


Figure 1. Conceptual model of biodegradable plastics from a material value chain perspective. Biodegradable products, surviving the lifetime of a product, are expected to be completely mineralized in a contained waste management system with triggers induced in those media. However, plastics, irrespective of their nature, inadvertently leak into the environment.

high prices, poor performance, lack of industrial infrastructure, and inconsistent quality standards. New biodegradable plastics are being publicized, accompanied by the consistent withdrawal of other biodegradable products. Also, key companies are entering and exiting this market, which is often representative of new products/markets. New entrants, tightening environmental restraints, and confusing terminologies, among other factors, shape the dynamics of the market. This combination of issues prompts an up-to-date review of the current state-of-the-art technology with an emphasis on knowledge gaps and research needs.

Without a doubt, it is a challenging task that biodegradable plastics set to achieve. Products must survive the entire value chain with a negligible ecological impact, but after disposal in a contained environment, they must completely assimilate as a food source for soil microorganisms, thereby ensuring the safe return of carbon to the ecosystem (Figure 1). Decades of product development efforts were diverted to optimize shelf life stability without considering end-of-life fate. In this modus operandi, sustainable end-of-life criteria pose a seemingly paradoxical challenge. Even if we ascertain this monumental structure–performance–price trade-off, in this context, a key question lingers: will the current biodegradable product meet its goal if it leaks into the environment or if it is disposed of in a nonideal manner, such as in a landfill where greenhouse gas emissions will not be controlled? These queries led us to cast a critical eye across the academic, trade, and policy literature and embrace a holistic view rather than a fragmented interpretation. This perspective—balancing crisis and opportunity—surveys the field, providing succinct summary accounts of current practices, along with a wealth of new research needs for these

products to shape, unsettle, and exceed the negative perception of plastics. The productive implementation and consumer acceptance of biodegradable plastics demands a multidisciplinary collaboration involving all parts of the value chain.

2. SCOPE AND METHODOLOGY

In this perspective, we describe the current commercial state of biodegradable plastics with an analysis of the public databases of certified products. We focus specifically on enzyme-mediated biodegradation of plastics. To clarify the confusion between degradation, biodegradation, and compost, readers are referred to Figure 2. The broad definition of degradation encompasses an irreversible change of materials' chemical and physical properties influenced by multiple environmental factors.¹⁴ Biodegradation is a subset of degradation involving mineralization by microorganisms primarily to CO_2 , H_2O , and CH_4 , which are the final products of aerobic or anaerobic degradation. Compost is

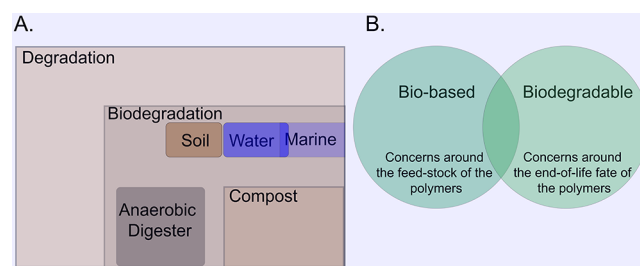


Figure 2. (A) Distinction between degradation, biodegradation, and compost. (B) Difference between the biobased and biodegradable polymers.

one possible biodegradation medium along with water, soil, and marine (Figure 2A). Abiotic processes that occur in the natural environment, such as hydrolysis or oxidation, can accelerate degradation, and biodegradable polymers can be designed to have cradle-to-cradle life cycles.¹⁵ Biodegradable plastics come with discrete labeling relating to the preferred end-of-life treatments (industrial compost, home compost, soil, water, or marine) and are distinct from biobased products. The biobased designation concerns the source of raw material, and it has very little to say on the fate of the product in different environmental conditions. Biobased products may offer different end-of-life possibilities such as biodegradability or compostability, which is often considered as an important environmental advantage and cardinal product functionality. However, not all biobased products are biodegradable or compostable (Figure 2B). We have excluded biobased products that are not biodegradable from the focus of this perspective.

To give the readers a sense of major issues in the field of biodegradable plastics, we have been selective rather than exhaustive. While we discuss most of the major contributions in the field, we focus primarily on depicting a holistic system-level view with multiple stakeholder involvement. This comprehensive view differentiates the scope and breadth of this perspective from the already published review articles in this field.^{16–22} In addition, while the reader should certainly draw inspiration from the existing literature regarding biodegradable polymers for medical or *in vivo* applications^{23–25}—indeed, the development of biodegradable polymers has been driven substantially by such applications—we focus specifically on biodegradable polymers applied as alternatives to environmentally persistent, non-degradable commodity plastics.

We do not discuss additive-mediated degradation of conventional “oxo-degradable” plastics (e.g., polyolefins with transition metal catalyst) as such pro-oxidant additives that are not proven to be effective for complete mineralization,²⁶ and their mechanism is distinct from the enzyme-mediated biodegradation process.

3. PROMISE OF BIODEGRADABLE PLASTICS

Society responded to the challenge of accumulation of plastic wastes in the environment by producing biodegradable plastics. Unfortunately, the promise of biodegradable plastics has been obfuscated by greenwashing.^{27,28} Here, we propose four distinct scenarios. With each scenario, we reason a case for biodegradable plastics.

Scenario 1: Some Plastics Are Difficult to Recycle. The need to divert plastics from landfills is aggravated by difficult-to-recycle materials such as multilayer packaging, food waste, partially degraded or contaminated agricultural products, or other organically soiled packaging. For example, at festivals and sporting events, traditional plastics (e.g., nonbiodegradable foodware) along with food residues and leftovers form assorted, nonseparable, and nonrecyclable waste in a collection stream, where plastic and food residues coexist and contaminate each other.²⁹ Under these situations, the only fate of these contaminated plastics is either landfills or incinerators. However, if foodware (or other contaminated plastics) is compostable then the heterogeneous waste becomes homogeneous from a solid waste management perspective and can be handled through composting. Hence, in this and many other examples, biodegradable plastics promise to increase the available end-of-life options by diverting landfill accumulation or incineration to compost. Compost serves the dual purpose of being a long-term

biofertilizer and soil conditioner. The high temperature resulting from industrial composting guarantees the elimination of pathogens and thus removes the environmental risk of applying compost in the soil.^{30–32} This zero-waste model³³ has seen tremendous success especially at community, sporting, and other large-scale events where reusable foodware and the separation of food waste are not economical or logical. Extrapolating from this case, the advantage of using compostable plastics is evident when there is a risk of cross-contamination between plastics and organic waste. Also, conventional non-degradable, noncertified commodity polymers represent potential contamination of the organic waste stream with a serious source of microplastic in the soil.^{34,35}

Scenario 2: A Landfill Is an Unsustainable Way of Waste Management. End-of-life plastics are amassing in landfills resulting in both management issues and environmental harm.^{36,37} Plastics occupy 5–25 wt % of the total waste in landfills.³⁸ Accumulation, uncontrolled methane release, and leakage of plastics from the landfills exceed the estimated 8 billion USD economic loss^{39,40} in the USA. European legislation⁴¹ (2008/98/EC Waste Framework Directive) and the U.S. Environmental Protection Agency (EPA) placed landfills as the least preferred waste management option. Without a doubt, the system of collecting, landfilling, and incinerating waste is a costly one that contributes to global warming and creates toxic air and water pollution. Put simply, composting of biodegradable plastics provides alternative end-of-life waste management options by diverting the amount of trash sent to landfills and incinerators.⁴²

Scenario 3: Every Material Has a Finite Mechanical Recycling Capability. Mechanical recycling, despite its apparent simplicity in implementation, has several drawbacks. Consumer-discarded plastics are heterogeneous. Either they are contaminated with multiple constituents⁴³ or the additives present are difficult to recover.⁴⁴ Further, the recycled plastics suffer from a loss of quality with each processing cycle as mechanical recycling imposes deteriorated performance in plastic articles.^{45,46} Aiming toward environmental nonpersistence through biodegradable polymers is unarguably a superior alternative if we fail to close the loop with mechanical recycling efficiently.

Scenario 4: Plastics Do Leak into the Environment. Biodegradable plastics are designed to mineralize in a controlled waste management environment. However, littering is an uncontrolled event without the space–time constraint, making it pertinent to different environments.²⁹ Larger plastic litter, including everyday items such as drink bottles and other types of plastic packaging, as well as synthetic textiles, migrate to the ocean from land-based sources (Figure 1). Regardless of any improvement in our collection systems, leakage of plastics is unavoidable.^{47,48} Unlike the other three scenarios, this situation straddles a thin boundary between minimizing the impact of accidental leakage and encouraging intentional littering. Biodegradable plastics can alleviate the impact of accidental leakage by lowering the likely permanence time and the risks associated^{29,49} with the product's persistence and accumulation. However, this tacit assumption is not an excuse to litter. We revisit this dilemma in Section 5.4.4.

4. CURRENT STATE

Commercial biodegradable plastics are marketed through a certification (or labeling). Certification of the marketplace products establishes the credibility of sustainability claims. In an

Table 1. Brief Summary of Certified Products from TÜV Austria Database⁵⁶

	No. finished products	No. raw materials	Main constituents	Maximum thickness
Industrial compostable	1463	496	PLA, PHAs, PBS, PBAT, Thermoplastic starch, Regenerated Cellulose, Cellulose Acetate	3.3 mm
Home compostable	739	187	PBAT, Thermoplastic starch, PHAs, Regenerated Cellulose, Cellulose Acetate	1.1 mm
Soil	23	52	PHAs, Thermoplastic starch, Regenerated Cellulose, Cellulose Acetate	Not required ^a
Water	1	20	Regenerated Cellulose, Cellulose Acetate, PHAs	5 mm
Marine	0	20	Thermoplastic starch, PHAs	N/A

^aTÜV Austria does not impose any disintegration criteria for soil biodegradable products.

alternative perspective, the certification translates a set of complex information (lab test protocol, test results) into an easily understandable message across the value chain. Each product (even with the same raw material) needs a separate certification as their composition and end-of-life degradation profile can be different based on the thickness, size, and minor constituents. Generally, the certification process follows three steps. First, the brand owner recognizes the brand to certify and which certification to obtain based on the customer need. Second, a third-party testing laboratory (OWS, AIMPLAS, Innovhub SSI, etc.) conducts the test following the prescribed test protocols by the International Organization for Standardization (ISO), American Society for Testing and Materials (ASTM), European Committee for Standardization (CEN), and others.^{50–54} Third, the certifying body (DIN Certco, Biodegradable Products Institute (BPI), TÜV Austria, and others) reviews the data from the third-party testing lab and makes the decision on certification.

In biodegradation testing, several critical aspects need attention. The first is that biodegradation should principally be determined by measuring carbon to carbon dioxide conversion (and methane in case of anaerobic conditions). For aerobic conditions, oxygen consumption is a good alternative to carbon dioxide titration. Other parameters, such as weight loss, decrease of molecular weight, and deterioration of mechanical characteristics, are only secondary parameters and are not definitive proof of complete degradation. A second important aspect is the necessity to specify the testing environment, as both the level and rate of biodegradation can be different from one environment to another. For example, poly(lactic acid) (PLA), a commonly known industrially compostable plastic, exhibits limited degradation in anaerobic landfill or soil medium.⁵⁵

A summary of certified products is provided in Table 1. Several insights can further be drawn from the detailed analysis of these products:

- Out of all possible biodegradation media (Figure 1A), the industrial compost environment is the most prevalent among brand owners because it can easily be introduced into the current organic waste management systems, and it is the most aggressive medium for plastic degradation. In general, in terms of microbial diversity and the rate of biodegradation: industrial composting > home composting > soil > water > marine. The number of certified products for each media type indirectly confirms this observation.
- The size of an item (i.e., thickness) is a critical factor in determining the certification. Unsurprisingly, the greater the thickness of the article, the longer it takes to disintegrate and biodegrade in the environment.

- PLA is certified only in an industrial composting environment. Poly(hydroxyalkanoates (PHAs), starch, and cellulose or cellulose derivatives are certified for a significantly broader range of environments.
- Garden, agricultural, and horticultural applications dominate the soil biodegradable application list, while food packaging products dominate the compostable products list.
- No acceptable test standards exist for the marine medium, but still, there are significant product development efforts in those areas. Although there are no ASTM standards for home composting, there are French (NF T 51-800) and Australian standards (AS 5810) for home compostability available.

The commercial spectrum of compostable plastics is dominated by aliphatic polyesters and copolyesters (PLA, PHAs), polybutylene succinate adipate (PBSA), polycaprolactone (PCL), polyglycolic acid (PGA), aliphatic aromatic polyesters (polybutylene adipate terephthalate (PBAT)), and carbohydrates (cellulose, cellulose acetate, starch, starch blends). Global consumption of biodegradable polymers was 335,000 t in 2017.⁵⁷ Western Europe remains the largest market for biodegradable polymers (52%), followed by Asia and Oceania (25%) and the U.S. (22%).

Figure 3 outlines the main applications of compostable products based on BPI certification. Foodware, dishes, cutlery, and bag sectors are the major end-use markets, as well as the major contributors for biodegradable polymer consumption. Bans on single-use nondegradable plastic bags remain the most popular policy interventions by governments, and often, biodegradable plastic shopping and produce bags surface as an

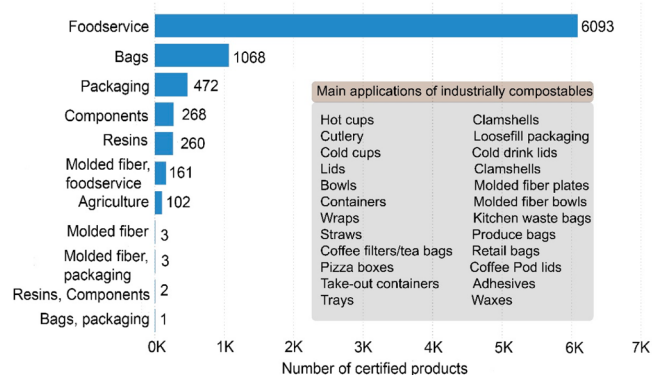


Figure 3. Current landscape of application for industrially compostable products. The figure is generated from the analysis of Biodegradable Plastics Institute (BPI) certified industrial compostable products database.⁵⁸ BPI has over 8000 certified industrial compostable products.

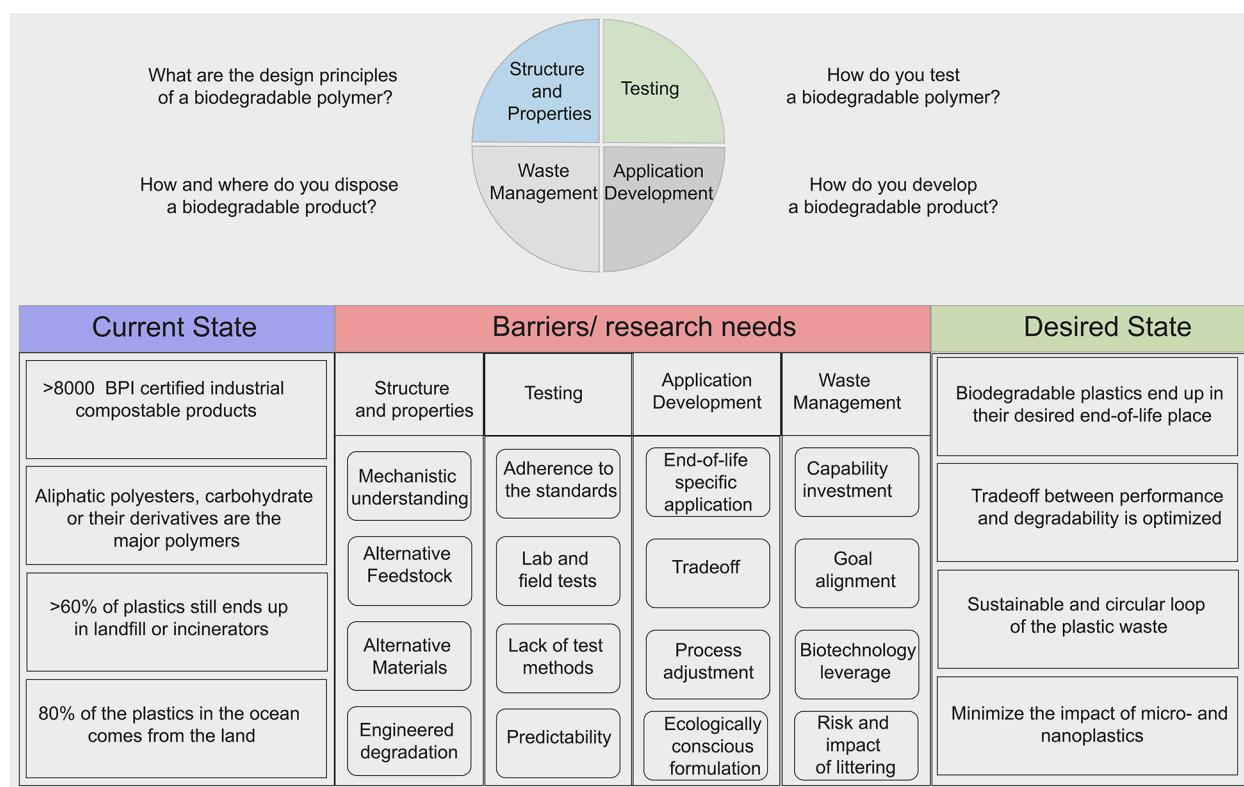


Figure 4. Roadmap to biodegradable plastics.

alternative. Kitchen waste bags are the second-largest end-use segment for biodegradable polymers. The gradual expansion of composting systems and increasing interest in diverting organic waste such as leaves and grass clippings and food scraps from landfills contribute to the growth of this application. The development of starch-based loose-fill packaging is the major reason for the growth in the foam packaging market replacing polystyrene foam materials. Currently smaller but potentially larger volume markets for biodegradable polymers include agriculture and horticulture (mulch films, plant pots, landscaping groundcovers, and more) applications. Paper coatings (extrusion coatings for paper and paperboard for use in cups and cartons), adhesives, colorant, ink for printing, or other additives and minor constituents are growing their share due to the requirements of the certification bodies. Other significant uses include textiles and nonwovens, restorable medical devices (implants, sutures), downhole materials for oil and gas processes, and filaments for additive manufacturing. Legislation is the single most important driver⁵⁹ for biodegradable polymers. For example, bans or taxes on the use of non-biodegradable shopping bags in Italy, France, or coastal cities have led to a substantial surge in the consumption of biodegradable plastics.⁷ In contrast, the growth of biodegradable polymers is slow in places that lack mandates.

5. BARRIERS/RESEARCH NEEDS

Industries have successfully developed and promoted compostable, soil biodegradable, and water biodegradable products (Section 4) in response to the scenarios where such products have the strongest case in the material value chain (Section 3). However, some of the lingering questions remain. Why do the compostable plastics end up in landfills? Are biodegradable plastics promoting an alternative to sound waste management

practices? How are formulators handling these new classes of materials? These questions have, of course, been around for a long time. What is new these days is an influx of ideas, tools, and methodologies outside of traditional chemistry, including microbial ecology, polymer physics, and process science. We acknowledge the current state based on the certified products in the marketplace and speculate the desired scenario based on the goal and aspirations of a circular economy and other sustainable models.^{60–62} To close the gap between the current and desired states, we present the barriers and subsequent research needs in the context of product development, product realization, and waste management efforts. Figure 4 laid out the roadmap by organizing these intertwined research needs. While identifying these 16 research needs, divided over four focus areas, an emphasis has been placed on recognizing significant accomplishments in the relevant directions. To build on those findings, we advocate collaborative research to translate those knowledge, methods, and insights into the other parts of the value chain.

5.1. Focus Area 1: Structure and Properties. Biodegradable polymers inherently carry a greater concentration of heteroatoms than their fossil-fuel counterparts like polyethylene and polypropylene. In addition, biodegradable polymers are often derived from feedstocks considered inferior to their petroleum-based counterparts in several aspects. We identify four areas where these challenges can be addressed.

5.1.1. Mechanistic Understanding. The term biodegradation involves biological activity. If we assume biodegradation is an isolated event, devoid of any environmental variables, three well-defined⁶³ sequential steps—biodeterioration, biofragmentation, and assimilation—can be conceived. However, in the natural environment, biotic and abiotic factors synergistically affect biodegradable polymers in a complex interplay of processes and chemistries (Figure 5).⁶⁴ Conceptually, all these

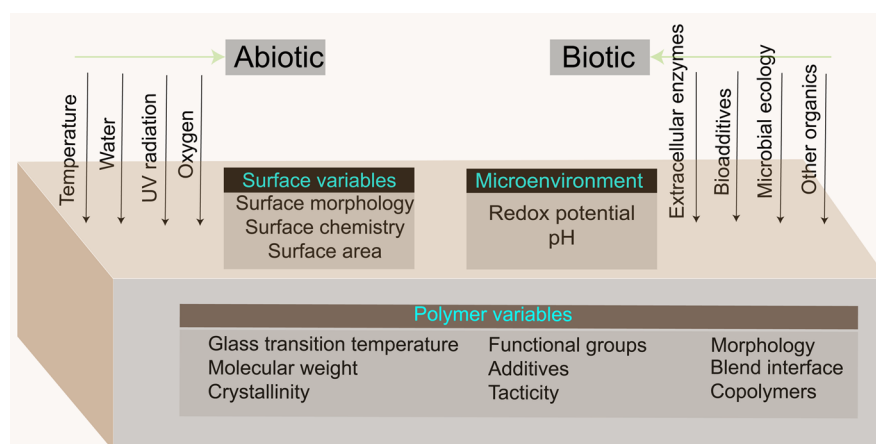


Figure 5. Multivariable space of biodegradation. Microbe–environment–polymer dialogue is dependent on the interplay of abiotic and biotic factors.

aspects can influence the microenvironment (pH, redox potential), interfacial chemistry, and inherent chemistry and physics (chemical composition, crystallinity, polydispersity) of the polymers. Hence, augmented environments in the form of chemical, biological, or mechanical pretreatments (as discussed in Section 5.4.3)—conducive to deterioration and fragmentation—remain attractive methods to achieve the optimal pathways for microbial metabolism.

Microscopic fungi and bacteria or other biological agents (such as earthworms, insects, roots, and rodents) can also fragment the product. To ascertain the microbial assimilation, Zumstein et al.⁶⁵ have reasoned that both CO₂ evolution (in aerobic degradation) and incorporation of polymer-derived carbon into microbial biomass are essential to track carbons. They used ¹³C-labeled PBAT and nanoscale secondary ion mass spectrometry to show microbial assimilation of the labeled carbon from the polymer.

Despite the heterogeneity and diversity of the environmental conditions, several basic mechanistic principles can be extrapolated to all classes of biodegradable polymers:

- For aerobic biodegradation, water is crucial. Hydrolysis of the material proceeds either via a bulk or surface erosion mechanism. In bulk erosion, the degradation process takes place throughout the thickness of the final article, whereas surface erosion proceeds through a decrease in thickness. Different polymers exhibit different dominant mechanisms, based on water diffusivity and reaction rates.⁶⁶
- The higher the surface area is, the greater the rate of microbial degradation is.⁶⁷
- Soil biodegradation follows the Arrhenius equation.⁶⁸
- Microbial degradation rates are enhanced in polymers with lower glass transition temperature,⁶⁹ higher loading of plasticizers,⁷⁰ and lower crystallinity,⁶⁹ each of them contributing to faster rates of diffusion.

Nevertheless, as described in an excellent review by Laycock et al.,⁷¹ our understanding of all interdependent factors controlling degradation in natural environments is not yet sufficiently advanced to permit a lifetime prediction of biodegradable polymers. Such examples of mechanistic understanding in the natural environment applicable to realistic conditions need to be expanded not only to better predict the biodegradation (as discussed in Section 5.2.4) but also to aid in designing a degradable polymer (as discussed in Section 5.1.4.).

5.1.2. Alternative Feedstock. The feedstock of biodegradable polymers—mostly derived from food or another natural source—remains a consistent concern. Agricultural feedstocks raise a serious question around the long-term sustainable prospects.⁷² Alternative solutions can come from effective utilization of renewable feedstock or waste.^{73–77}

The cheap price of the plastics is attributed to the often-overlooked advantage of the platform approach.⁷⁸ The starting material of commodity polymers—ethylene, propylene, butane, benzene, toluene, xylenes, and methanol—are used in multiple applications, and they are sourced from a byproduct of the oil refinery industry. In addition to technological leveraging, these platform chemicals benefit from an economic advantage in which multiple products share investment and infrastructure costs without customized production requirements. To mimic or displace fossil-fuel-based plastics with biobased biodegradable plastics, we need a similar approach buoyed by deep mechanistic understanding. The biorefinery concept analogous to the conventional concept of an oil refinery has potential provided the resulting processes reduce costs, ensue alternative feedstocks, or obtain more valuable organic materials. One of the emerging trends is the combination of two or more waste streams that could avoid the requirement of additional synthetic streams and/or compensate for the nutrients balance in the process.⁷⁹ This includes a pretreatment phase to adjust the waste material to an appropriate feed stream. In the future, the integration of PHA production into processes such as wastewater treatment plants, hydrogen production, or biodiesel factories could enhance its implementation at an industrial scale.⁷⁹

Capture and utilization of carbon dioxide (CCU) technology to drive the sustainable production and consumption of biodegradable plastics⁸⁰ may represent another option in the right direction; it also would avoid the unwanted competition of the plastics industry with the world's food supply.

Tang et al. reported⁸¹ a novel catalytic method to produce copolymers of PHA from the diastereomeric mixtures of monomers. This method avoids the need for a wasteful separation process with an added benefit of enhanced ductility and toughness.

5.1.3. Alternative Materials. To compete with the versatility, performance, and volume of commodity plastics, a continual search for alternative biodegradable plastics should be directed toward a new backbone or side-chain chemistry (or both). The commercial and academic efforts are dominated by aliphatic

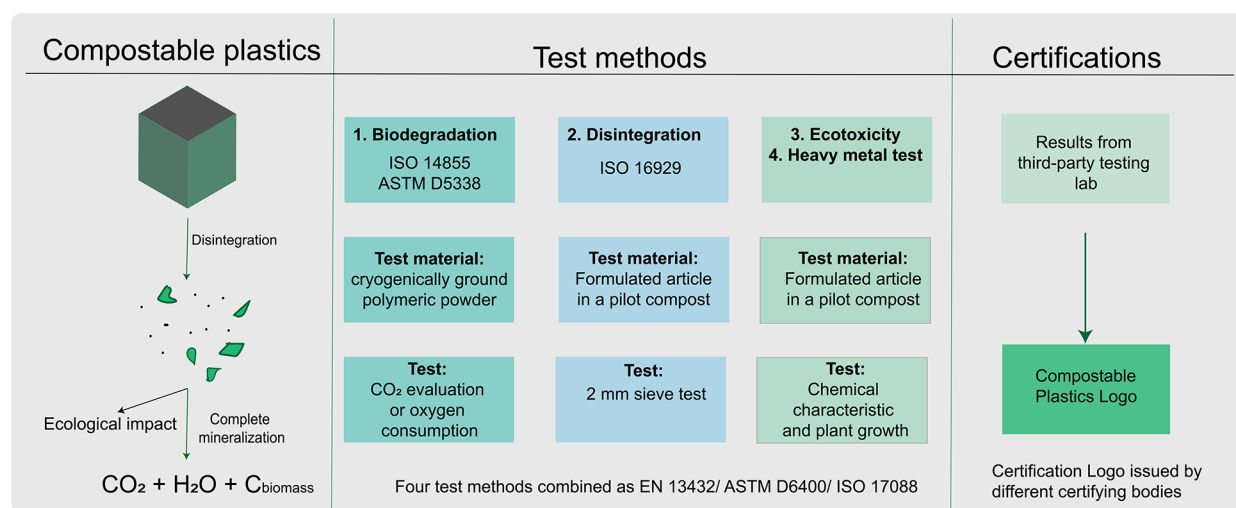


Figure 6. Test methods and certification for industrially compostable plastics. To be certified as an industrially compostable product, each product needs to go through the four test methods.

polyesters based on glycolic acid, lactic acid, and similar α -hydroxy acids or plant-derived polymers.⁸² This lack of diversity is a serious problem because a wide range of degradable materials will be desirable so that the structure–property relationships can be appropriately matched to the specific and unique requirements of each biodegradable application. Some of the recent notable efforts in searching for alternative potential biodegradable materials include the following: (1) utilization of seaweed polysaccharides, such as alginate and carrageenan,⁸³ (2) microalgae-based plastics,^{83,84} (3) 2,5-furan dicarboxylic acid-based polyesters,⁸⁵ and (4) salicylic acid-based aromatic polyesters.⁸⁶

When designing new biodegradable chemistries, special attention needs to be paid to their processing condition (as discussed in Section 5.3.3). For example, proteinaceous material offers a rich combination of functionality and sustainability. However, denaturation, cross-linking, and plasticization are generally unavoidable aspects of protein processing that results in a substantial compromise of the final property.^{87–89}

5.1.4. Engineered Degradation. The design of polymers that are robust and durable in use yet readily degrade if discarded indiscriminately in the environment presents a functional dichotomy (Figure 1). One potential tool to address this dichotomy is to tune the degradation profile of the plastic (while meeting the requirements of a specific application, see Section 5.3), that is, to develop plastics that are carefully engineered to degrade. Consider, for example, work from Hakkarainen et al.,⁹⁰ in which caprolactone/1,5-dioxepan-2-one (CL/DXO) copolymers with various random and block architectures were synthesized. They found that the placement of the more hydrolysis-sensitive DXO units profoundly impacted degradation characteristics of the copolymers in buffered aqueous media. In a similar vein, Terzopoulou et al. reviewed⁹¹ progress in polyesters based on furandicarboxylic acid, noting several pertinent examples in which copolymerization affects degradation in different media. Likewise, Kumar and Maiti have reviewed⁹² the impact of nanoparticles on biodegradation of various biodegradable polymers, identifying scenarios in which degradation rates are both increased and decreased. Very recently, Giundani et al.⁹³ showed that copolymerization of CL with globalide and subsequent functionalization of the globalide

unsaturation by N-acetylcysteine leads to enhanced degradation in enzymatic assays and in activated sludge.

These examples, and many others, indeed illustrate how biodegradable plastics can be engineered to degrade. However, we further stress that there exists a vast, unexplored parameter space regarding application of the same principles to traditionally nondegradable polymers. For example, if a hydrolytically sensitive unit is copolymerized with a polyolefin,^{94,95} can the resulting material be designed such that oligomeric products of hydrolysis become bioavailable in compost? For inspiration, we highlight a recent, excellent piece of work from Haider et al.,⁹⁴ in which orthoester and octadiene sequences were copolymerized and hydrogenated to polyethylene mimics; the unsaturated precursors were shown to exhibit enhanced and tunable hydrolysis rates, although the hydrogenated mimics were not biodegradable under the test conditions employed, leaving a clear opportunity for further development. More generally, can emerging and established principles borrowed from self-healing materials, controlled release, sensory amplification, and transient electronics be translated to engineer degradation?

In advancing such concepts, we must reiterate the importance of participation from stakeholders across the value chain. A cautionary tale involves oxodegradable plastics. These plastics were introduced into the market as biodegradable materials and saw significant commercial success, yet due to a lack of appropriate testing and certification (Section 5.2) coupled with subsequent research showing that such products merely fragment,⁹⁶ withdrawal of such products is anticipated. To avoid this fate, the introduction of new plastics engineered for degradation must be thoroughly supported by fundamental studies, most crucially concerning the corresponding degradation pathways (Section 5.1.1), degradation in realistic conditions (Section 5.2.2), ecological fate of partially degraded products (Section 5.3.4), and potential consequences of littering (Section 5.4.4).

5.2. Focus Area 2: Testing. The lack of complete mechanistic understanding and the heterogeneity of the test medium establishes a need for standard test methods. Despite tremendous progress, the multivariable nature of the test medium and our evolved understanding prompts research needs in four primary directions.

5.2.1. Adherence to Standard Test Protocols. As Haider et al.²² and others^{97,98} have pointed out, in primary literature, the term “biodegradable” is frequently misused, abused, or misinterpreted where the term is in the title or main texts, without any proof or degradation tests being discussed in the actual paper. In some cases, the degradation conditions are often harsh (high temperatures, extreme pH values) or not environmentally realistic (isolated and enriched microorganism/enzymes). To circumvent misunderstanding and incorrect claims, the degradation process needs to strictly adhere to the standard test protocols (Figure 6). Standards are not perfect and deserve scrutiny, but they provide a foundation for a structured database, improve transparency, and increase the reproducibility of the test results across the test laboratories. Standard development is a collaborative, comprehensive, and continual process where the developing test methods are critiqued, approved, and published by recognized standardization bodies. Standardization processes exist at national, continental, and international levels, for example, American Society for Testing and Materials (ASTM), European Committee for Standardization (CEN), and International Organization for Standardization (ISO), respectively. The practice of using correct terminology—to describe and differentiate the multiple pathways of degradation—requires formal education, communication, and training by the publication bodies.

For example, the testing and certification of industrially compostable plastics, as illustrated in Figure 6, provide a framework for designing a formulation appreciating the whole disposal lifecycle of the product. Compostable products, through the disintegration and inherent biodegradation process, should not leave any impact on the ecological system. The only way to ensure that is to follow the testing protocols developed by the standard bodies.

5.2.2. Gap between Lab and Field Tests. The gap between the lab and the actual environmental test is self-evident. The variables in the lab are well defined, controlled, and reproducible. In contrast, polymers are subjected to multiple assaults that are interrelated (discussed in Section 5.1.1) when they are in the environment. This universal paradox is paramount where microbes are an integral and instrumental part of the process. Replication of the microbial ecology—population, diversity, and dynamics—from the field to the lab is a monumental task. An additional issue becomes apparent when recognizing that, in a natural environment, the polymer may not be the preferred substrate in the presence of alternate nutrients.⁹⁹ Lab inoculum may be capable of degrading polymers, but the laboratory tests were limited to these specific microorganisms, where the tested polymer was the sole carbon source for the microorganism. Lab test–field experiment integration initiated by waste management contractors and standardization bodies will close the gap.

Here, we illustrate the crux of the challenge for the example of a centralized industrial compost environment (Figure 7).

Each of the compost medium variables (pH, temperature, moisture content, aeration rate, feedstock composition) does impact the environmental degradation process of the polymer and the diversity of the microbial consortia. For example, higher pH, temperature, and moisture, in the initial stages of composting, accelerate the hydrolysis of the polymer backbone or side chain with alkaline hydrolysis.¹⁰⁰ A typical composting process has three steps:

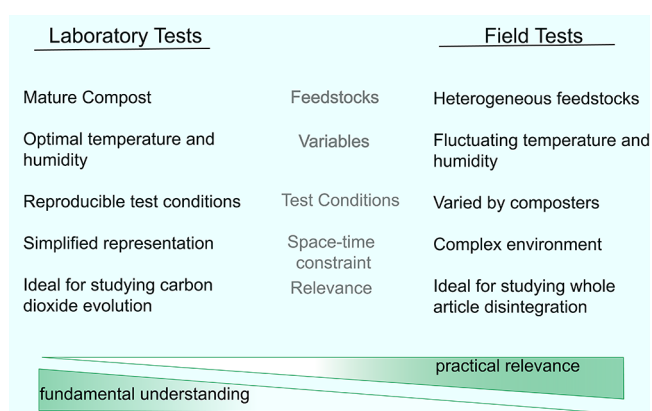


Figure 7. Gap between the laboratory and field tests, relevant to the composting environment.

1. Mesophilic phase (25–50 °C, pH 7–8): lasts for 2–3 days with mesophilic bacteria and fungi as the dominant microbial species.
2. Thermophilic phase (temperature 60–70 °C, pH 8–10): lasts for 10–15 days, where the thermophilic bacteria, actinomycetes, and heat-tolerant fungi survive.
3. Another mesophilic phase (25–50 °C, pH 7–8): lasts for a couple of months, dominated by mesophilic bacteria, actinomycetes, and fungi.

Actinomycetes are responsible for the earthy smells of finished compost and may overwhelm bacterial populations by producing antibiotics. Fungi thrive in acidic, poorly aerated, and cold environments. Anaerobic organisms may survive in the anoxic pockets of the compost. As a general guideline, the variety of organic feedstock determines microbial diversity and population growth.¹⁰¹ A critical balance of feedstock, turning frequency, aeration, and temperature is instrumental for a good quality of compost with optimum C/N ratio, porosity, electrical conductivity, pH, and moisture balance.¹⁰² Importantly, a significant margin^{70,100–102} for optimization exists to facilitate the conducive environment for organic waste mineralization without sacrificing compost quality. The scenario described here presents a strong case to complement lab-scale biodegradation tests with pilot or industrial-scale composting studies.

The development of a reliable test method in soil and marine media present the same issues but in a much larger spatial dimension. Accordingly, a clear distinction between the purpose of laboratory tests (to establish the intrinsic biodegradability under optimal conditions) and the environmental condition (to establish the nonpersistence of plastics) needs to be made. A simulated or mesocosm test media that links the laboratory test and field test is relevant especially where there is a gap between the scale of those two media. Müller⁵¹ addressed this by introducing a three-scale approach, and the Open-Bio Test Development Effort¹⁰³ adopted a laboratory-mesocosm-field-scale test to evaluate marine biodegradation with environmental significance.

5.2.3. Lack of Test Methods for Marine Biodegradation. Currently, few test methods for the assessment of the biodegradation of materials in the marine environment are available from ISO and ASTM. No European CEN test method has been developed so far. The available test methods concern the biodegradation under aerobic conditions. Two active test methods from ASTM are ASTM D6691-17, which determines aerobic biodegradation of plastics in the marine environment by

a defined microbial consortium or natural seawater column, and ASTM D7473-12, which only addresses disintegration. The other standard, ASTM D7081-05, for nonfloating biodegradable plastics in the marine environment, was withdrawn in 2014, and no replacement has been proposed. In early 2015, the Belgian private nonprofit agency, Vinçotte (acquired by TÜV Austria), introduced a certification scheme for an “OK biodegradation MARINE label” based on the criteria of ASTM D7081.

The challenges of developing a suitable comprehensive marine biodegradation test method^{104,105} are multifaceted. First, 80% of marine litter comes from land. Thus, the quest for a standard on all-encompassing marine biodegradability of plastics requires a consideration of the degradation profile through the lifecycles of plastic litters.²⁷ Second, the marine medium itself is highly heterogeneous. The temperature varies significantly, and microbial profiles¹⁰⁶ fluctuate based on the many marine areas that are low in oxygen (hypoxic) or free from oxygen (anoxic). Also, vast regions are covered with very fine sediment (mud).¹⁰⁷ Third, a major difference between the marine environment and soil is the consequence of biofouling¹⁰⁸ (colonization by micro- and macro-organisms) on the biodegradation process, which has not been investigated in the marine environment, and its effects remain essentially unknown. Fourth, as compared to freshwater, soil, and compost conditions, the marine environment, especially seawater, is considered less aggressive but significant enough to promote the release of dissolved organic carbon¹⁰⁹ from a biodegradation point of view.

Open-Bio is a research project¹⁰³ funded by the European Commission within FP7 (Seventh Framework Program for Research and Technological Development). One part of the project (WPS: in situ biodegradation) concerns the biodegradation behavior in natural environments: soil, freshwater, and marine. Biodegradation in the sandy eulittoral (intertidal beach) zone, in the sublittoral (benthic) zone at the water/seafloor interface, and in the pelagic (free water column) zone are all in consideration to develop a comprehensive marine biodegradation test method.

5.2.4. Lack of Predictability. Biodegradation of plastic is a slow and natural process. The typical timeline for evaluating biodegradation of plastics results from months to years. Industrial compost tests run for three months, and soil biodegradation is for two years, according to the ASTM, ISO, and CEN standards. The process of new formulation development becomes an excruciatingly long process. That sole reason is a sufficient driver for predicting the biodegradation outcome.⁷¹ The goal of lifetime modeling for all classes of polymers is to predict the degradation rate, taking all-controlling variables as input (Figure 5). However, because of the interdependency of all the variables, the aspirational goal of achieving a single-unified predictive model is not a trivial task. Pockets of knowledge from degradation chemistry, drug delivery, and enzyme chemistry should be rendered by using the broader framework of structure–property relations to relate macromolecular and chemical changes to engineering properties.⁷¹

Abi-Akl et al.¹¹⁰ recently described the crux of the challenge inherent to lifetime modeling, in that conventional scaling arguments are insufficient due to the mutually inclusive effects of physical and biochemical processes. They developed a theoretical model that accounts for the biochemo-mechanically coupled kinetics of polymer degradation. They produced biodegraders and examined their capability to both dissociate the material from its external boundaries and to penetrate it to

degrade its internal mechanical properties. Their model quantitatively captures the experimental results and reveals distinct signatures of different bacteria independent of the specific experimental conditions (i.e., particle volume and initial concentrations). In a separate attempt,¹¹¹ with the degradation of the marine oil spill as an example, a quantitative understanding of microbial community structure and function relationship was attempted. Yadav and Hakkarainen¹¹² proposed the use of cellulose acetate (CA) as a model to study the complex interplay between structure, environment, and degradation. CA is produced from a highly biodegradable and chemically robust cellulose backbone through acetylation of some of the hydroxyl groups. The environmental and microbial process introduces a structural change as the lower degree of substitution alters the degradation rates and fate in different environments.

5.3. Focus Area 3: Application Development. Application development of a biodegradable plastic is a challenging task where the performance requirements of the application need to be balanced with the end-of-life degradability requirements (Figure 8). Primary challenges include the expectation of the

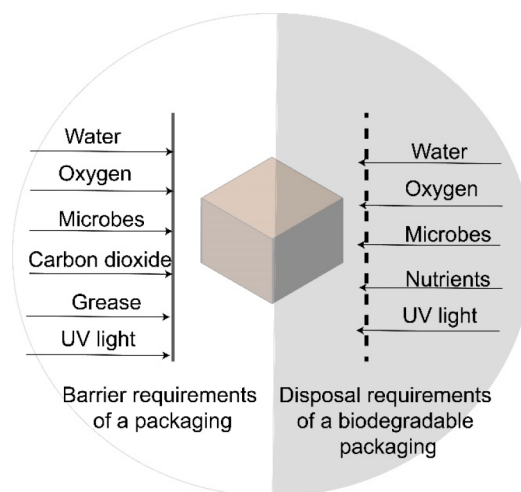


Figure 8. Conflicting performance requirements of a biodegradable product. We have illustrated the paradox with an example of biodegradable packaging where the same variables that are instrumental for biodegradation are detrimental during consumer use and shelf life. Solid lines indicate impermeable criteria of the variables, and the dotted line indicates the facile diffusion criteria during waste management.

formulators to utilize the new materials in their existing equipment and processes and lack of understanding of the certification requirements across the value chain. Considering these circumstances, we have identified four research areas where research and development efforts should be directed.

5.3.1. End-of-Life Specific Application. The penetration of biodegradable products in the marketplace is driven by the regulations around single-use plastics. To advance as a sustainable alternative to the banned counterparts, these products must carry a certification logo so that consumers, product owners, and waste management facilities are aligned with the same objective around the end-of-life fate of these plastics. Additionally, an in-depth knowledge of end-of-life fate helps in the material selection process early in the product development process. With examples of biodegradation of polymeric mulch films in agricultural soils, Sander¹¹³ eloquently assessed the importance of knowing end-of-life knowledge and

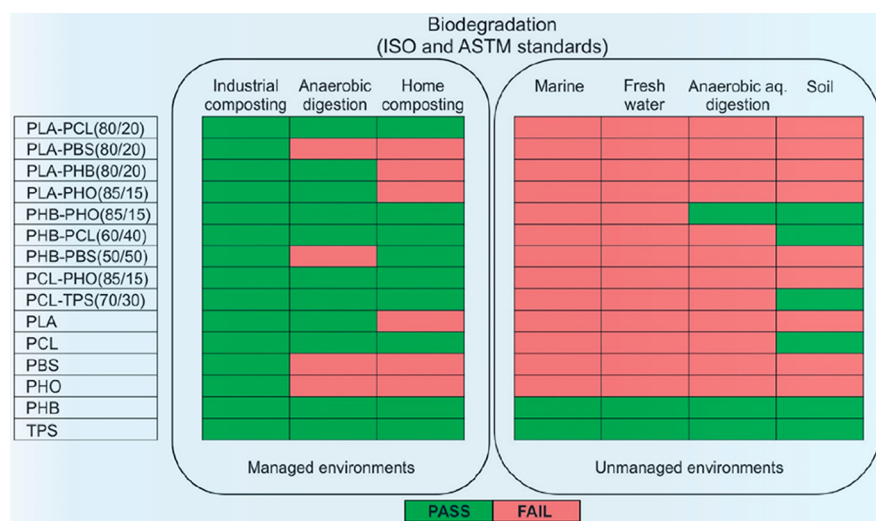


Figure 9. Biodegradation capacity of several bioplastics and their blends in managed and unmanaged environments tested following international biodegradation standards. Adapted from ref 130 with permission.

connecting to material properties. Mulch film addresses the need of crop growth, earliness, and weed control. However, an ideal mulch film, as promised by soil biodegradable compositions, is the one that would degrade at the end of the crop cycle and that would entirely biodegrade in a short time when buried in soil.¹¹⁴ Unfortunately, a significant number of the biodegradable mulch products¹¹⁵ (as registered in GreenPla, Japan Bioplastics Association) are designed from PLA, which does not pass the soil biodegradable test criteria.

European Bioplastics¹¹⁶ recommends that compostable plastics should be promoted if the following criteria apply:

- (1) Plastics are contaminated with food waste.
- (2) Plastics whose expected fate is organic waste collection, and mechanical recycling is not possible.
- (3) Plastics provide a potential to reduce nonbiodegradable plastic contamination of biowaste collection.
- (4) Alternative reusable solutions cannot be redesigned.

Similarly, a series of drivers¹¹⁷ can be identified for promoting a case of soil biodegradable agricultural plastic products:

- (1) There is a short to medium shelf life in the field, on average from one to three seasons (3 years).
- (2) Plastic waste produced at the end of the use is contaminated with soil or other plant residues (e.g., nets for growing crops under greenhouses, geotextile applied in reforestation, or in landscaping).
- (3) Plastic waste is difficult to detach from plant residues or collect (e.g., pheromone dispensers, clips, twines).¹¹⁸ In this case the organic residue is contaminated with plastic.

In the third application, the water biodegradable plastics offer certain benefits where the large quantities of materials migrate into nature as a part of their normal usage. The cosmetic industry, for example, has a regulatory and environmental need to develop lubricants and microbeads that are biodegradable in water.^{119,120}

5.3.2. Trade-Off between Performance and Degradability. While biodegradability is an attractive postconsumer characteristic, these plastics must meet performance objectives in the market before disposal. Nonetheless, many biodegradable plastics often fail to meet material design criteria such as flexibility, strength, and toughness. Unsurprisingly, they do not

match their established nondegradable counterparts for applications such as packaging and thus require modifications.^{121,122} Some of the well-adopted strategies to overcome these constraints include the following:

- (1) Copolymers and homopolymer blends can efficiently disrupt the native crystalline structures of the aliphatic homopolymers to improve ductility and thermal processability.¹²³
- (2) Improvements to processability via viscosity and melt-strength enhancement can be accomplished through chain modifications such as chain extension, branching, and cross-linking.^{124–126}
- (3) Mechanical properties of PHAs and PLA can be tuned by using plasticizers and nucleating agents,^{127,128} which can lead to order-of-magnitude increases in elongation at break, reduced brittleness, and increased crystallinity.

However, each of these modifications comes with the cost of impacting biodegradation potential.¹²⁹ Literature reports often fail to address the trade-off between performance and degradability. The diversity of biodegradable materials and environments makes it difficult to extract simple and generic assessments of their end-of-life fate. One notable effort¹³⁰ investigated the fate of a range of selected biodegradable plastics and their blends, simulating controlled vs uncontrolled environments (Figure 9) to gain a superior understanding of their potential environmental fate and possible future end-of-life management options. They identified a surprising synergy, wherein plastic blends exhibit improved biodegradation in home composting as compared to the constituent individual plastics. Conversely, they also discovered antagonistic situations, primarily in aquatic and soil environments, where plastic blends exhibited poor biodegradation. More studies of this ilk are desperately needed, as the findings expand new end-of-life management considerations but also raise a concern about the potential release of specific material combinations in certain uncontrolled environments.

5.3.3. Process Adjustment. Two of the most commonly studied polymers—PLA and PHAs—are inherently susceptible to thermal and hydrolytic degradation¹³¹ during processing, which can severely limit their applications. For example, poly(3-hydroxybutyrate) (PHB) is often processed slightly below the

melting temperature or at elevated temperatures for only brief periods of time; however, even at submelting temperatures, thermal degradation is still observed. Some of the innovative strategies to overcome processing limitations include the following:

- Heat treatments, such as annealing¹³² and fiber/film-melt drawing,^{133–135} which can impart improvements in crystallinity and morphology that enhance mechanical properties and thermal stability. This approach contrasts the use of copolymers and blends, which reduces crystallinity and makes these materials more ductile and more processable (as discussed in Section 5.3.2). The choice of strategy is dependent on the performance metrics of the application and needs to be balanced with end-of-life specific applications (as discussed in Section 5.3.1)
- Raising the degradation temperature by hindering the degradative mechanisms or lowering the melting temperature by increasing chain mobility.¹³⁶
- Utilizing melt rheological information^{137,138} to optimize screw design for specific bioplastics, composites, or blends of materials, as well as to determine the specific geometry and flight configuration of the feedscrew.

Successful case histories of overcoming processing challenges are often obscured in the trade secret of the company literature. However, the basic knowledge of different shaping technologies and their tolerance to basic polymer chemical and physical parameters (glass transition temperature, melt temperature, degradation point, melt flow index, and more) will save significant time and resources in identifying alternative materials (as discussed in Section 5.1.3) and design principles (as discussed in Section 5.1.4).

5.3.4. Ecologically Conscious formulation. Ecologically conscious formulation demands a thorough end-of-life knowledge for each ingredient and can alter the product design. Plastics are a formulated final product where individual polymers are usually mixed with additives such as strengtheners (e.g., carbon or silica), thermal stabilizers, plasticizers, fire retardants, UV stabilizers, colorants, matting agents, opacifiers, or luster additives. These additives can introduce the possibility of adverse health issues, such as in the case of plasticizers^{139–141} or in the case of black plastics.¹⁴² The compostable standards (EN 13432, ASTM D6400, ISO 17088, AS 4736) specify the limit values for heavy metals and fluorine. It also means that the compost obtained at the end of the composting trial, ultimately containing biomass or undegraded residuals from the final formulated plastics, should not adversely impact the germination and growth of plants (and also earthworms in case of AS 4736). The requirement of ecologically conscious formulation forces us to critically reevaluate the choice of synthetic strategies, selection of catalysis, and option of additives. Zimmermann et al.¹⁴³ studied the toxicity and chemical composition of biobased and biodegradable materials using *in vitro* bioassays and high-resolution nontarget mass spectrometry. Their results show that the majority (67%) of bioplastics and plant-based products carry toxic chemicals. They precisely proposed the selection of materials using green chemistry to “design out” toxicity during the formulation development.

5.4. Focus Area 4: Waste Management. Successful implementation of sustainable waste management^{144,145} of biodegradable plastics requires a thorough understanding of the end-of-life management options (Figure 10) and the

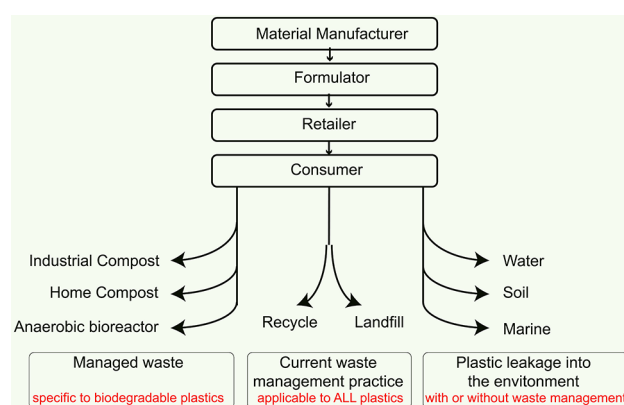


Figure 10. Waste management practice and their importance in handling biodegradable plastics waste.

environmental impact of these polymers, which in turn requires the integration of policies, regulations, and standards. Thus, close collaboration, cooperation, and communication between academia, industry, and government—identified as four research needs—will enable the broader value chain to address these imminent issues.

5.4.1. Capability Investment. Biodegradable plastics can benefit waste management only if efficient collection and sorting systems are implemented (Figure 10).²⁸ Without a dedicated place for containment, biodegradable plastics may adversely impact the current waste management practices.^{18,97} For example, due to their physically indistinguishable nature, biodegradable plastics may end up in the current recycling scheme with the nondegradable counterpart initiating degradation and cross-contamination issues.²⁸

Interestingly, anaerobic digestion and biogasification are controlled waste management options where significant value can be extracted from biodegradable plastics, yet there exists a comparative dearth of facilities. On the basis of the Biocycle Snapshot survey,¹⁴⁶ there are 204 farm-based anaerobic digesters compared to 4713 composting facilities in the U.S. In anaerobic digestion,^{147–149} organic matter is degraded by an anaerobic microbial population producing methane, carbon dioxide, and nutrient-rich residues while producing minimal heat. The biogas can be integrated with a heat and power plant generating electricity and heat or can be upcycled to biomethane. Additionally, anaerobic digestion provides better odor control making them attractive in densely populated areas.¹⁵⁰ In contrast to the polymer degradation behavior in aerobic compost, results in the anaerobic media¹⁵¹ are scarce and suffer from a lack of fundamental understanding and wide-scale replication across the laboratories. Clearly, the discussion and standardization requirements for anaerobic treatments are still in their infancy.

Additionally, the option of innovative pretreatments, leveraged from the mechanistic understanding (Section 5.1.1) and new design principles (Section 5.1.4), should be examined closely to achieve faster cycles (or reduce residence time of plastics) of composting or anaerobic digestion. Some of the pretreatment strategies include the following:

- Mechanical pretreatment,¹⁵² where high energy ball milling can increase the effective surface area producing a faster rate of biodegradation

- (b) Chemical recycling¹⁵³ strategies, where plastic articles can be chemically treated to trigger the production of bioavailable feedstocks
- (c) Biological augmentation strategies,^{154,155} where a conducive environment is created for microbial growth and proliferation

To present an economic case for bioremediation as a plastic-removal mechanism, some of the relevant questions need to be answered: To what extent does the microbial population recirculate the chemical effects of additives (as discussed in Section 5.3.4) into the water and through the food chain? What are the side effects of bioremediation if it is implemented on a large scale? The answers to these questions require an inclusive assessment of microbial communities, time spans, and resources. In other words, if a significant fraction of traditional plastics is ultimately displaced by biodegradable alternatives, then the scale of the bioremediation would be daunting; hence, a sound, reliable techno-economic analysis is required.

5.4.2. Goal Alignment. Most often, composting facilities have unique goals, driven by the fact that the quality of the final compost^{156,157} determines the selling price and consumer favorability. To optimize that goal, composters most often focus on accelerating the compost cycle, which is likely counterproductive to plastics degradation, especially for thicker articles. Additionally, the demand for complete bioremediation of plastics must also be balanced against the economic sustainability of composting methods, which may drive plant operators to prioritize throughput over compost quality.¹⁵⁸ Further, the composting industry itself presents additional challenges¹⁰² that may not align well with plastics remediation goals. It needs to be established whether the plastic has any value in impacting the quality of the compost without impacting the microbial consortia.¹⁵⁹ Biowaste collections lie in the hands of private companies or municipalities, who have no obligation to collect compostable plastics. Out of 4713 composting facilities in the U.S.,¹⁴⁶ only 18% accept food waste and multiple organics. Contamination of compostable plastics with noncompostable wastes or the concern of microplastics or nanoplastics arising from incomplete degradation^{34,35,96} are some of the outstanding concerns contributing to the waste disposal contractors' reluctance to accept any plastics. Another important fact is 90-day disintegration criteria imposed by standard test methods (ASTM D6400 or EN 13432) are not aligned to composting goals. Many industrial composting plants operate at cycles of 8 weeks or even down to 3 weeks to maximize the feedstock and operational cost ratio. This disconnect certainly will not have a simple solution, rather it will require creative research (as discussed in Section 5.1.4 and Section 5.4.3) and perhaps, more crucially, education, discussion, and communication among all stakeholders. To be sure, certification for compostable products and use of the correct logo (Figure 6) is instrumental for ensuring that items have been properly tested, meet international standards, and can be identified as such by composters, municipalities, restaurants, consumers, and others engaged in the diversion of organic waste.

5.4.3. Leveraging Biotechnology. Leveraging biotechnology has an enticing prospect, particularly if coupled to waste management, to address plastics remediation. We do not discuss the what, how, and why of this technology as it has been introduced in multiple exemplary reviews.^{160–162} One of the possible pathways to improve the biodegradation time is to leverage the microbial metabolism—accepted as a safer and

efficient tool for the removal of many organic pollutants.¹⁶⁰ Microbial diversity can be exploited to identify desirable degradative genes and/or properties via the construction of metagenomic libraries.¹⁶³ Subsequently, the genetic information can be transferred to culturable bacteria for enhanced bioremediation.^{164,165} Although the resulting genetically engineered microorganisms (GEMs) are expected to have increased efficiency for bioremediation, they pose a threat to the environment because of the possibility of horizontal gene transfer and uncontrolled proliferation. The construction of containment systems is thus a prerequisite for the release of GEMs in situ to resolve these concerns.^{166,167}

A significant inspiration can be drawn from the reports of mealworms, waxworms, and superworms eating nonhydrolyzable plastics¹⁶⁸ like polyethylene (PE) and polystyrene (PS). However, those results, with some exceptions,¹⁶⁹ will benefit from greater rigor and critique (as discussed in Section 5.2.1). Inderthal et al.¹⁶⁸ have argued the importance of studying the enzymatic degradation of polyethylene, polypropylene, polystyrene, and polyvinyl chloride as these macromolecular structures without any heteroatoms offer significant insight on the autoxidation mechanism which complements the hydrolysis mechanism—the primary pathway through which biodegradable polymers degrade—and the structure-guided knowledge gleaned from these interactions should be directed to enhance enzyme effectiveness and stability.

While developing biocatalysis for plastics degradation, we primarily rely on the one gene-one enzyme-one function concept, originally framed by Beadle and Tatum.¹⁷⁰ Despite their success,^{164,167} the increasing discrepancy between lab tests and field tests (as discussed in Section 5.2.2) points us to the need of deciphering the underlying microbial ecologies and their impact on polymer degradation. Bacteria, fungi, archaea, protists, and a host of viruses and phage represent a diverse microbial population. The mechanism by which they cooperate, compete, and use energy-efficient strategies¹¹¹ to degrade polymeric materials and assimilate in their metabolic pathway remain a daunting challenge.

5.4.4. Risk and Impact of Littered Plastics. Like their nondegradable counterparts, biodegradable plastics may share the same end-of-life fate if they are not a part of controlled waste management systems. Microplastics and nanoplastics, irrespective of their chemical nature, likely present a greater threat to the environment as compared to monolithic plastics. From a biotechnology perspective, microplastics may not be a nutritional carbon source for microorganisms, impeding their assimilation into the environment.¹⁶⁰

Green et al.¹⁷¹ and Green¹⁷² studied the effect of conventional plastics and biodegradable plastics (0.02%, 0.2%, and 2% of wet sediment weight) upon microbial diversity and community growth. The results confirmed that PLA acted as a stressor in sandy sediment, inducing an unfortunate surge in microbe respiration rate. González-Pleiter et al.¹⁷³ explored the toxicity of secondary PHB nanoplastics on three representative aquatic organisms. This study implied that secondary biodegradable nanoplastics, due to partial biodegradation of PHB, induced a significant decline in cell growth in all three aquatic organisms.

Additionally, biodegradable microplastics can act as stronger vectors than conventional microplastics.¹⁷⁴ Zuo et al.¹⁷⁵ disclosed that the adsorption and desorption capacity (with phenanthrene as one of representative organic pollutants) of microplastics from PBAT ($2338 \pm 486 \mu\text{m}$) was greater than that of conventional microplastics (PS, $2628 \pm 623 \mu\text{m}$; PE, 250

μm), suggesting biodegradable microplastics may be a preferred carrier of pollutants and microbial contamination to non-degradable microplastics.

Lack of data prevents us from concluding the threat status of biodegradable microplastics; a true assessment is certainly nontrivial as microplastics enter the food chain through complex, nonlinear, and dynamic interactions with the ecosystems. We reemphasize the need for collection—existing in developed countries, often absent in developing countries—as a primary strategy to prevent leakage. The fate of compostable plastics in open and mixed environments is poorly studied and cannot be predicted. Hence, to stretch the claim of biodegradability as ready for littering may frequently be disingenuous. Degli Innocenti and Breton²⁹ have assessed this critical issue and stressed the importance of a specific methodology for the assessment of the risk and impact of postconsumer littered waste—which is not yet available.

Clearly, the successful prevention of littering requires an enhanced appreciation to the accountability across the supply chain. A proposed integrated block chain¹⁷⁶ that tracks the flow of products, measures, and integrates every player's contribution to waste management in a reliable, transparent, and sustainable way can be a tool to ensure extended producer responsibility (EPR). Several industries, including BASF, already launched a reciChain technology¹⁷⁷ to improve the traceability of their materials and ingredients.

6. CONCLUSIONS

Financial, regulatory, consumer, and technology aspects are cited as the potential driver for biodegradable plastics.^{5,7,42} The overall global production of plastics in 2017 was 335 million tonnes. In comparison, according to an estimate by European Bioplastics⁵⁷ (an association based in Germany representing about 70 members from the entire value chain of bioplastics), global production capacities of bioplastics are predicted to grow from around 2.11 million tonnes in 2018 to approximately 2.62 million tonnes by 2023. This is still insignificant compared to the global plastics market, but with 40% of the global plastics market dominated by packaging,¹⁷⁸ biodegradation polymers have substantial room to grow. However, as we have attempted to convey, broader implementation of biodegradable plastics in the marketplace faces tremendous, intertwined challenges. As we rapidly untangle the complex chemical conversations between the microbes and the abiotic world, we are also extracting valuable information about how to apply this knowledge to improve plastic's nonpersistence. Ultimate success depends on interorganizational and intervalue chain collaboration on the myriad aspects of product realization and waste management. Exploring the synergies between the interested and concerned parties—researchers, government agencies, nongovernment organizations, industry, media, and the general public—enables meaningful progress on material quality, efficiency, and sustainability to be made. By encouraging interdisciplinary knowledge, we align on terms, methods, regulations, and goals, and avoid miscommunication that disenfranchises stakeholders and ultimately delays technological progress.

The environmental and health issues society faces today in the epoch of plastics mostly stem from the fact that the impact of the scales of plastic consumption and disposal was not fully considered until after mass-production reached full scale. A multidimensional value assessment that offers the means of capturing materials and financial flows, interactions, and dynamics between different players is essential for factual

comprehension of the alternative plastics. Lifecycle assessments (LCAs), albeit not without their challenges,^{179,180} will be necessary to ensure that these new choices and alternatives justly reduce the sum of adverse effects, ranging from undesired human exposure in plastics manufacturing and consumer uses to environmental pollution from inappropriate disposal.

While much is still to be learned about the ecological impact of plastics, the emerging science is already having a major influence on adopting the basic philosophy of circularity.^{60,181} As we are at a crossroads of materialization and dematerialization, we propose that rather than continuing to ask, “how should we ban or upcycle plastics?” we might instead ask, “how can we choose the right materials for the right application?” These are not the same thing. We need to take steps to exclude the uses of plastics that have been linked with harmful health effects and choose reusable over single use. The long-term solution may lie in prioritizing which applications are unavoidable and which ones offer short-term convenience only, then developing biodegradable plastics to bridge that gap.

We proposed 16 research needs to integrate efforts over a spectrum of the material value chain. These research needs are interrelated and benefit from the introspection of mechanism, the development of a repertoire of design tools, and appreciation of the multiscale dynamic complex system associated with microbe–polymer–environment crosstalk—all in sync with the needs of different stakeholders. We hope the framework proposed here can serve as a resource—to avoid the disconnect resulting from the rapid progress—to practitioners working on the development of sustainable materials that meet society's demands without compromising the quality of life of current and future generations.

AUTHOR INFORMATION

Corresponding Author

Koushik Ghosh – Sandia National Laboratories, Albuquerque, New Mexico 87123, United States; orcid.org/0000-0002-1188-699X; Email: kghosh@sandia.gov

Author

Brad H. Jones – Sandia National Laboratories, Albuquerque, New Mexico 87123, United States; orcid.org/0000-0003-3674-9075

Complete contact information is available at:
<https://pubs.acs.org/10.1021/acssuschemeng.1c00801>

Author Contributions

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Notes

The authors declare no competing financial interest.

Biographies



Koushik Ghosh is a senior member of the technical staff in the Organic Materials Science Department at Sandia National Laboratories. Before joining Sandia in 2020, Koushik spent six years in the chemical industry. Koushik obtained his Ph.D. degree from the University of Utah under the supervision of Prof. Peter J. Stang in 2010. After his Ph.D., he had two postdoctoral tenures, first from Jeffrey Moore's Laboratory at the University of Illinois, Urbana–Champaign, and the other as a director's postdoc at the Los Alamos National Laboratory. Koushik's research interests include material sustainability and chemistry–biology integration with a system science perspective.



Brad H. Jones is a senior member of the technical staff in the Organic Materials Science Department at Sandia National Laboratories. He obtained his Ph.D. degree in materials science and engineering from the University of Minnesota in 2011 under the supervision of Prof. Timothy Lodge. He joined Sandia as a postdoctoral researcher in 2014, where he studied stimuli-responsive peptide self-assembly before converting to his current position in 2015. His research interests include the chemistry and physics of cross-linked polymers, stimuli-responsive materials, and olefin metathesis.

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REFERENCES

- (1) Geyer, R.; Jambeck, J. R.; Law, K. L. Production, use, and fate of all plastics ever made. *Sci. Adv.* **2017**, 3 (7), e1700782.
- (2) Lehner, R.; Weder, C.; Petri-Fink, A.; Rothen-Rutishauser, B. Emergence of Nanoplastic in the Environment and Possible Impact on Human Health. *Environ. Sci. Technol.* **2019**, 53 (4), 1748–1765.
- (3) Rochman, C. M.; Browne, M. A.; Halpern, B. S.; Hentschel, B. T.; Hoh, E.; Karapanagioti, H. K.; Rios-Mendoza, L. M.; Takada, H.; Teh, S.; Thompson, R. C. Policy Classify plastic waste as hazardous. *Nature (London, U. K.)* **2013**, 494 (7436), 169–171.
- (4) North, E. J.; Halden, R. U. Plastics and environmental health: the road ahead. *Rev. Environ. Health* **2013**, 28 (1), 1–8.
- (5) Hale, R. C.; Song, B. Single-Use Plastics and COVID-19: Scientific Evidence and Environmental Regulations. *Environ. Sci. Technol.* **2020**, 54 (12), 7034–7036.
- (6) Schnurr, R. E. J.; Alboiu, V.; Chaudhary, M.; Corbett, R. A.; Quanz, M. E.; Sankar, K.; Srain, H. S.; Thavarajah, V.; Xanthos, D.; Walker, T. R. Reducing marine pollution from single-use plastics (SUPs): A review. *Mar. Pollut. Bull.* **2018**, 137, 157–171.
- (7) Xanthos, D.; Walker, T. R. International policies to reduce plastic marine pollution from single-use plastics (plastic bags and microbeads): A review. *Mar. Pollut. Bull.* **2017**, 118 (1), 17–26.
- (8) Rochman, C. M. Microplastics research—from sink to source. *Science* **2018**, 360 (6384), 28–29.
- (9) de Souza Machado, A. A.; Kloas, W.; Zarfl, C.; Hempel, S.; Rillig, M. C. Microplastics as an emerging threat to terrestrial ecosystems. *Glob Chang Biol.* **2018**, 24 (4), 1405–1416.
- (10) *Accumulation: The Material Politics of Plastic*; Gabrys, G. H., Gabrys, G., Michael, M., Eds; Routledge, London, 2013. DOI: 10.4324/9780203070215.
- (11) Rillig, M. C.; Bonkowski, M. Microplastic and soil protists: A call for research. *Environ. Pollut.* **2018**, 241, 1128–1131.
- (12) Lamb, J. B.; Willis, B. L.; Fiorenza, E. A.; Couch, C. S.; Howard, R.; Rader, D. N.; True, J. D.; Kelly, L. A.; Ahmad, A.; Jompa, J.; Harvell, C. D. Plastic waste associated with disease on coral reefs. *Science* **2018**, 359 (6374), 460–462.
- (13) Neu, L.; Bänziger, C.; Proctor, C. R.; Zhang, Y.; Liu, W.-T.; Hammes, F. Ugly ducklings—the dark side of plastic materials in contact with potable water. *npj Biofilms and Microbiomes* **2018**, 4 (1), 7.
- (14) Vert, M.; Doi, Y.; Hellwich, K.-H.; Hess, M.; Hodge, P.; Kubisa, P.; Rinaudo, M.; Schué, F. Terminology for biorelated polymers and applications (IUPAC Recommendations 2012). *Pure Appl. Chem.* **2012**, 84 (2), 377–410.
- (15) Göpferich, A. Mechanisms of polymer degradation and erosion. *Biomaterials* **1996**, 17 (2), 103–114.
- (16) Iwata, T. Biodegradable and bio-based polymers: future prospects of eco-friendly plastics. *Angew. Chem., Int. Ed.* **2015**, 54 (11), 3210–5.
- (17) Kale, S. K.; Deshmukh, A. G.; Dudhare, M. S.; Patil, V. B. Microbial degradation of plastic: a review. *J. Biochem. Technol.* **2015**, 6 (2), 952–961.
- (18) Shen, M.; Song, B.; Zeng, G.; Zhang, Y.; Huang, W.; Wen, X.; Tang, W. Are biodegradable plastics a promising solution to solve the global plastic pollution. *Environ. Pollut. (Oxford, U. K.)* **2020**, 263, 114469.
- (19) Narancic, T.; O'Connor, K. E. Plastic waste as a global challenge: are biodegradable plastics the answer to the plastic waste problem? *Microbiology (London, U. K.)* **2019**, 165 (2), 129–137.
- (20) Shah, A. A.; Hasan, F.; Hameed, A.; Ahmed, S. Biological degradation of plastics: a comprehensive review. *Biotechnol. Adv.* **2008**, 26 (3), 246–65.
- (21) Schneiderman, D. K.; Hillmyer, M. A. 50th Anniversary Perspective: There Is a Great Future in Sustainable Polymers. *Macromolecules (Washington, DC, U. S.)* **2017**, 50 (10), 3733–3749.
- (22) Haider, T. P.; Voelker, C.; Kramm, J.; Landfester, K.; Wurm, F. R. Plastics of the Future? The Impact of Biodegradable Polymers on the Environment and on Society. *Angew. Chem., Int. Ed.* **2019**, 58 (1), 50–62.

- (23) Ikada, Y.; Tsuji, H. Biodegradable polyesters for medical and ecological applications. *Macromol. Rapid Commun.* **2000**, *21* (3), 117–132.
- (24) Tian, H.; Tang, Z.; Zhuang, X.; Chen, X.; Jing, X. Biodegradable synthetic polymers: Preparation, functionalization and biomedical application. *Prog. Polym. Sci.* **2012**, *37* (2), 237–280.
- (25) Manavitehrani, I.; Fathi, A.; Badr, H.; Daly, S.; Negahi Shirazi, A.; Dehghani, F. Biomedical Applications of Biodegradable Polyesters. *Polymers (Basel, Switz.)* **2016**, *8*, 20.
- (26) Vazquez, Y. V.; Ressia, J. A.; Cerrada, M. L.; Barbosa, S. E.; Vallés, E. M. Prodegradant Additives Effect onto Comercial Polyolefins. *J. Polym. Environ.* **2019**, *27* (3), 464–471.
- (27) Nazareth, M.; Marques, M. R. C.; Leite, M. C. A.; Castro, Í.B. Commercial plastics claiming biodegradable status: Is this also accurate for marine environments? *J. Hazard. Mater.* **2019**, *366*, 714–722.
- (28) Zhu, J.; Wang, C. Biodegradable plastics: Green hope or greenwashing? *Mar. Pollut. Bull.* **2020**, *161*, 111774.
- (29) Degli Innocenti, F.; Breton, T. Intrinsic Biodegradability of Plastics and Ecological Risk in the Case of Leakage. *ACS Sustainable Chem. Eng.* **2020**, *8* (25), 9239–9249.
- (30) Onwosi, C. O.; Igbokwe, V. C.; Odimba, J. N.; Eke, I. E.; Nwankwoala, M. O.; Iroh, I. N.; Ezeogu, L. I. Composting technology in waste stabilization: On the methods, challenges and future prospects. *J. Environ. Manage.* **2017**, *190*, 140–157.
- (31) Fernandes, L.; Zhan, W.; Patni, N. K.; Jui, P. Y. Temperature distribution and variation in passively aerated static compost piles. *Bioresour. Technol.* **1994**, *48* (3), 257–263.
- (32) Roland Mote, C.; Griffis, C. L. Heat production by composting organic matter. *Agric. Wastes* **1982**, *4* (1), 65–73.
- (33) Hottle, T. A.; Bilec, M. M.; Brown, N. R.; Landis, A. E. Toward zero waste: Composting and recycling for sustainable venue based events. *Waste Manage.* **2015**, *38*, 86–94.
- (34) Zhang, B.; Yang, X.; Chen, L.; Chao, J.; Teng, J.; Wang, Q. Microplastics in soils: a review of possible sources, analytical methods and ecological impacts. *J. Chem. Technol. Biotechnol.* **2020**, *95* (8), 2052–2068.
- (35) Sintim, H. Y.; Bary, A. I.; Hayes, D. G.; English, M. E.; Schaeffer, S. M.; Miles, C. A.; Zelenyuk, A.; Suski, K.; Flury, M. Release of micro- and nanoparticles from biodegradable plastic during in situ composting. *Sci. Total Environ.* **2019**, *675*, 686–693.
- (36) Barnes, D. K. A.; Galgani, F.; Thompson, R. C.; Barlaz, M. Accumulation and fragmentation of plastic debris in global environments. *Philos. Trans. R. Soc., B* **2009**, *364* (1526), 1985–98.
- (37) Tansel, B. Persistence times of refractory materials in landfills: A review of rate limiting conditions by mass transfer and reaction kinetics. *J. Environ. Manage.* **2019**, *247*, 88–103.
- (38) Canopoli, L.; Fidalgo, B.; Coulon, F.; Wagland, S. T. Physico-chemical properties of excavated plastic from landfill mining and current recycling routes. *Waste Manage.* **2018**, *76*, 55–67.
- (39) Garcia, J. M.; Robertson, M. L. The future of plastics recycling. *Science* **2017**, *358* (6365), 870–872.
- (40) Unfinished Business: The Case for Extended Producer Responsibility for Post-Consumer Packaging, 2012. *As You Sow*. <https://www.asyousow.org/reports/unfinished-business-the-case-for-extended-producer-responsibility-for-post-consumer-packaging> (accessed April 2021).
- (41) Directive 2008/98/EC of the European Parliament and of the Council on Waste and Repealing Certain Directives. <https://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2008:312:0003:0030:en:PDF> (accessed 2021 January 18).
- (42) Dilkes-Hoffman, L. S.; Pratt, S.; Lant, P. A.; Laycock, B. The role of biodegradable plastic in solving plastic solid waste accumulation. *Plastics to Energy: Fuel, Chemicals, and Sustainability Implications* **2019**, 469–505.
- (43) Eriksen, M. K.; Christiansen, J. D.; Dagaard, A. E.; Astrup, T. F. Closing the loop for PET, PE and PP waste from households: Influence of material properties and product design for plastic recycling. *Waste Manage.* **2019**, *96*, 75–85.
- (44) Ugduler, S.; Van Geem, K. M.; Roosen, M.; Delbeke, E. I. P.; De Meester, S. Challenges and opportunities of solvent-based additive extraction methods for plastic recycling. *Waste Manage.* **2020**, *104*, 148–182.
- (45) Schyns, Z. O. G.; Shaver, M. P. Mechanical Recycling of Packaging Plastics: A Review. *Macromol. Rapid Commun.* **2021**, *42*, 2000415.
- (46) Hopewell, J.; Dvorak, R.; Kosior, E. Plastics recycling: challenges and opportunities. *Philos. Trans. R. Soc., B* **2009**, *364* (1526), 2115–26.
- (47) Thompson, R. Environment: A journey on plastic seas. *Nature (London, U. K.)* **2017**, *547* (7663), 278–279.
- (48) Chamas, A.; Moon, H.; Zheng, J.; Qiu, Y.; Tabassum, T.; Jang, J. H.; Abu-Omar, M.; Scott, S. L.; Suh, S. Degradation Rates of Plastics in the Environment. *ACS Sustainable Chem. Eng.* **2020**, *8* (9), 3494–3511.
- (49) Pico, Y.; Barcelo, D. Analysis and Prevention of Microplastics Pollution in Water: Current Perspectives and Future Directions. *ACS Omega* **2019**, *4* (4), 6709–6719.
- (50) De Wilde, B. Biodegradation Testing Protocols. *Degradable Polymers and Materials: Principles and Practice* **2012**, ACS Symp. Series 1114, 33–43.
- (51) Müller, R.-J. Biodegradability of Polymers: Regulations and Methods for Testing. *Biopolymers Online* **2002**, DOI: 10.1002/3527600035.bpola012.
- (52) Tillinger, R.; De Wilde, B.; De Baere, L. Standard test methods for polymer biodegradation in solid-waste treatment systems. *Biodegradable Plastics and Polymers* **1994**, *12*, 323–330.
- (53) De Wilde, B.; Boelens, J. Prerequisites for biodegradable plastic materials for acceptance in real-life composting plants and technical aspects. *Polym. Degrad. Stab.* **1998**, *59* (1–3), 7–12.
- (54) De Wilde, B.; De Baere, L.; Tillinger, R. Test methods for biodegradability and compostability. *Meded. - Fac. Landbouwk. Toegepaste Biol. Wet. (Univ. Gent)* **1993**, *58* (4A), 1621–1628.
- (55) Karamanlioglu, M.; Preziosi, R.; Robson, G. D. Abiotic and biotic environmental degradation of the Bioplastic polymer poly(lactic acid): A review. *Polym. Degrad. Stab.* **2017**, *137*, 122–130.
- (56) Certified Products. TÜV Austria. <https://www.tuv-at.be/green-marks/certified-products/> (accessed 2020 October 23).
- (57) Bioplastics Market Development Update 2019. *European Bioplastics*, 2019. https://www.european-bioplastics.org/wp-content/uploads/2019/11/Report_Bioplastics-Market-Data_2019_short_version.pdf (accessed 20 November 2020).
- (58) Biodegradable Products Institute. <https://www.bpiworld.org/> (accessed 2020 October 25).
- (59) Chen, Y.; Awasthi, A. K.; Wei, F.; Tan, Q.; Li, J. Single-use plastics: Production, usage, disposal, and adverse impacts. *Sci. Total Environ.* **2021**, *752*, 141772.
- (60) Hahladakis, J. N.; Iacovidou, E. Closing the loop on plastic packaging materials: What is quality and how does it affect their circularity? *Sci. Total Environ.* **2018**, *630*, 1394–1400.
- (61) Loorbach, D.; Frantzeskaki, N.; Avelino, F. Sustainability Transitions Research: Transforming Science and Practice for Societal Change. *Annual Review of Environment and Resources* **2017**, *42* (1), 599–626.
- (62) Niero, M.; Hauschild, M. Z. Closing the Loop for Packaging: Finding a Framework to Operationalize Circular Economy Strategies. *Procedia CIRP* **2017**, *61*, 685–690.
- (63) Lucas, N.; Bienaime, C.; Belloy, C.; Queneudec, M.; Silvestre, F.; Nava-Saucedo, J.-E. Polymer biodegradation: Mechanisms and estimation techniques – A review. *Chemosphere* **2008**, *73* (4), 429–442.
- (64) Min, K.; Cuiffi, J. D.; Mathers, R. T. Ranking environmental degradation trends of plastic marine debris based on physical properties and molecular structure. *Nat. Commun.* **2020**, *11* (1), 727.
- (65) Zumstein, M. T.; Schintlmeister, A.; Nelson, T. F.; Baumgartner, R.; Woecken, D.; Wagner, M.; Kohler, H.-P. E.; McNeill, K.; Sander, M. Biodegradation of synthetic polymers in soils: Tracking carbon into CO₂ and microbial biomass. *Science Advances* **2018**, *4* (7), eaas9024.

- (66) Burkersroda, F.v.; Schedl, L.; Göpferich, A. Why degradable polymers undergo surface erosion or bulk erosion. *Biomaterials* **2002**, 23 (21), 4221–4231.
- (67) Chinaglia, S.; Tosin, M.; Degli-Innocenti, F. Biodegradation rate of biodegradable plastics at molecular level. *Polym. Degrad. Stab.* **2018**, 147, 237–244.
- (68) Pischedda, A.; Tosin, M.; Degli-Innocenti, F. Biodegradation of plastics in soil: The effect of temperature. *Polym. Degrad. Stab.* **2019**, 170, 109017.
- (69) El-Hadi, A.; Schnabel, R.; Straube, E.; Müller, G.; Henning, S. Correlation between degree of crystallinity, morphology, glass temperature, mechanical properties and biodegradation of poly (3-hydroxyalkanoate) PHAs and their blends. *Polym. Test.* **2002**, 21 (6), 665–674.
- (70) Phuong, V. T.; Verstiche, S.; Cinelli, P.; Anguillesi, I.; Coltelli, M.-B.; Lazzeri, A. Cellulose acetate blends - effect of plasticizers on propertie and biodegradability. *J. Renewable Mater.* **2014**, 2 (1), 35–41.
- (71) Laycock, B.; Nikolić, M.; Colwell, J. M.; Gauthier, E.; Halley, P.; Bottle, S.; George, G. Lifetime prediction of biodegradable polymers. *Prog. Polym. Sci.* **2017**, 71, 144–189.
- (72) Brizga, J.; Hubacek, K.; Feng, K. The Unintended Side Effects of Bioplastics: Carbon, Land, and Water Footprints. *One Earth* **2020**, 3 (1), 45–53.
- (73) Wu, J.; Eduard, P.; Thiagarajan, S.; Jasinska-Walc, L.; Rozanski, A.; Guerra, C. F.; Noordover, B. A. J.; van Haveren, J.; van Es, D. S.; Koning, C. E. Semicrystalline Polyesters Based on a Novel Renewable Building Block. *Macromolecules (Washington, DC, U. S.)* **2012**, 45 (12), 5069–5080.
- (74) Wang, Z.; Ganewatta, M. S.; Tang, C. Sustainable polymers from biomass: Bridging chemistry with materials and processing. *Prog. Polym. Sci.* **2020**, 101, 101197.
- (75) Gandini, A. Polymers from Renewable Resources: A Challenge for the Future of Macromolecular Materials. *Macromolecules (Washington, DC, U. S.)* **2008**, 41 (24), 9491–9504.
- (76) Jang, Y.-S.; Kim, B.; Shin, J. H.; Choi, Y. J.; Choi, S.; Song, C. W.; Lee, J.; Park, H. G.; Lee, S. Y. Bio-based production of C2-C6 platform chemicals. *Biotechnol. Bioeng.* **2012**, 109 (10), 2437–2459.
- (77) Posvyashchennaya, A.; Volgina, T.; Novikov, V.; Zinoviyev, A. Lactide Production from Polymer Waste. *Key Eng. Mater.* **2018**, 769, 17–22.
- (78) Huo, J.; Shanks, B. H. Bioprivileged Molecules: Integrating Biological and Chemical Catalysis for Biomass Conversion. *Annu. Rev. Chem. Biomol. Eng.* **2020**, 11 (1), 63–85.
- (79) Rodriguez-Perez, S.; Serrano, A.; Pantiñón, A. A.; Alonso-Fariñas, B. Challenges of scaling-up PHA production from waste streams. A review. *J. Environ. Manage.* **2018**, 205, 215–230.
- (80) Kumar, M.; Sundaram, S.; Gnansounou, E.; Larroche, C.; Thakur, I. S. Carbon dioxide capture, storage and production of biofuel and biomaterials by bacteria: A review. *Bioresour. Technol.* **2018**, 247, 1059–1068.
- (81) Tang, X.; Westlie, A. H.; Watson, E. M.; Chen, E.Y.-X. Stereosequenced crystalline polyhydroxyalkanoates from diastereomeric monomer mixtures. *Science* **2019**, 366 (6466), 754–758.
- (82) Hillmyer, M. A. The promise of plastics from plants. *Science (Washington, DC, U. S.)* **2017**, 358 (6365), 868–870.
- (83) Zhang, C.; Show, P.-L.; Ho, S.-H. Progress and perspective on algal plastics – A critical review. *Bioresour. Technol.* **2019**, 289, 121700.
- (84) Marrone, B. L.; Lacey, R. E.; Anderson, D. B.; Bonner, J.; Coons, J.; Dale, T.; Downes, C. M.; Fernando, S.; Fuller, C.; Goodall, B.; Holladay, J. E.; Kadam, K.; Kalb, D.; Liu, W.; Mott, J. B.; Nikolov, Z.; Ogden, K. L.; Sayre, R. T.; Trewyn, B. G.; Olivares, J. A. Review of the harvesting and extraction program within the National Alliance for Advanced Biofuels and Bioproducts. *Algal Res.* **2018**, 33, 470–485.
- (85) Papageorgiou, G. Z.; Papageorgiou, D. G.; Terzopoulou, Z.; Bikiaris, D. N. Production of bio-based 2,5-furan dicarboxylate polyesters: Recent progress and critical aspects in their synthesis and thermal properties. *Eur. Polym. J.* **2016**, 83, 202–229.
- (86) Kim, H. J.; Reddi, Y.; Cramer, C. J.; Hillmyer, M. A.; Ellison, C. J. Readily Degradable Aromatic Polyesters from Salicylic Acid. *ACS Macro Lett.* **2020**, 9 (1), 96–102.
- (87) Pomet, M.; Redl, A.; Morel, M.-H.; Domenech, S.; Guilbert, S. Thermoplastic processing of protein-based bioplastics: chemical engineering aspects of mixing, extrusion and hot molding. *Macromol. Symp.* **2003**, 197 (1), 207–218.
- (88) Zhang, Y.; Liu, Q.; Rempel, C. Processing and characteristics of canola protein-based biodegradable packaging: A review. *Crit. Rev. Food Sci. Nutr.* **2018**, 58 (3), 475–485.
- (89) Chmielewska, A.; Kozłowska, M.; Rachwał, D.; Wnukowski, P.; Amarowicz, R.; Nebesny, E.; Rosicka-Kaczmarek, J. Canola/rapeseed protein – nutritional value, functionality and food application: a review. *Crit. Rev. Food Sci. Nutr.* **2020**, 1–21.
- (90) Hakkarainen, M.; Hoeglund, A.; Odelius, K.; Albertsson, A.-C. Tuning the release rate of acidic degradation products through macromolecular design of caprolactone-based copolymers. *J. Am. Chem. Soc.* **2007**, 129 (19), 6308–6312.
- (91) Terzopoulou, Z.; Papadopoulos, L.; Zamboulis, A.; Papageorgiou, D. G.; Papageorgiou, G. Z.; Bikiaris, D. N. Tuning the Properties of Furandicarboxylic Acid-Based Polyesters with Copolymerization: A Review. *Polymers* **2020**, 12, 1209.
- (92) Kumar, S.; Maiti, P. Controlled biodegradation of polymers using nanoparticles and its application. *RSC Adv.* **2016**, 6 (72), 67449–67480.
- (93) Guindani, C.; Candiotto, G.; Araújo, P. H. H.; Ferreira, S. R. S.; de Oliveira, D.; Wurm, F. R.; Landfester, K. Controlling the biodegradation rates of poly(globalide-co-ε-caprolactone) copolymers by post polymerization modification. *Polym. Degrad. Stab.* **2020**, 179, 109287.
- (94) Haider, T.; Shyshov, O.; Suraeva, O.; Lieberwirth, I.; von Delius, M.; Wurm, F. R. Long-Chain Polyorthoesters as Degradable Polyethylene Mimics. *Macromolecules* **2019**, 52 (6), 2411–2420.
- (95) Wilson, R. B.; Jonasdottir, S. Hydrolytically Degradable Olefin Copolymers. U.S. Patent US6534610B1, 2001.
- (96) Markowicz, F.; Szymańska-Pulikowska, A. Analysis of the possibility of environmental pollution by composted biodegradable and oxobiodegradable plastics. *Geosciences (Basel, Switz.)* **2019**, 9 (11), 460.
- (97) Zumstein, M. T.; Narayan, R.; Kohler, H.-P. E.; McNeill, K.; Sander, M. Dos and Do Nots When Assessing the Biodegradation of Plastics. *Environ. Sci. Technol.* **2019**, 53 (17), 9967–9969.
- (98) Albertsson, A.-C.; Hakkarainen, M. Designed to degrade. *Science* **2017**, 358 (6365), 872.
- (99) Ru, J.; Huo, Y.; Yang, Y. Microbial Degradation and Valorization of Plastic Wastes. *Front. Microbiol.* **2020**, 11, 442.
- (100) Sedničková, M.; Pekařová, S.; Kucharczyk, P.; Bočkáj, J.; Janigová, I.; Kleinová, A.; Jochec-Mošková, D.; Omaníková, L.; Perdochová, D.; Koutný, M.; Sedlářík, V.; Alexy, P.; Chodák, I. Changes of physical properties of PLA-based blends during early stage of biodegradation in compost. *Int. J. Biol. Macromol.* **2018**, 113, 434–442.
- (101) Rudnik, E. Compostable polymer materials. *Compostable Polymer Materials* **2019**, 1–410.
- (102) Awasthi, S. K.; Sarsaiya, S.; Awasthi, M. K.; Liu, T.; Zhao, J.; Kumar, S.; Zhang, Z. Changes in global trends in food waste composting: Research challenges and opportunities. *Bioresour. Technol.* **2020**, 299, 122555.
- (103) Open-Bio: Opening Bio-Based Markets via Standards, Labelling and Procurement. *BioBasedEconomy*. <https://www.biobasedeconomy.eu/projects/open-bio/> (accessed 2020 November 20).
- (104) Lott, C.; Eich, A.; Unger, B.; Makarow, D.; Battagliarin, G.; Schlegel, K.; Lasut, M. T.; Weber, M. Field and mesocosm methods to test biodegradable plastic film under marine conditions. *PLoS One* **2020**, 15 (7), e0236579.
- (105) Oberbeckmann, S.; Osborn, A. M.; Duhaime, M. B. Microbes on a Bottle: Substrate, Season and Geography Influence Community Composition of Microbes Colonizing Marine Plastic Debris. *PLoS One* **2016**, 11 (8), e0159289.

- (106) Amaral-Zettler, L. A.; Zettler, E. R.; Mincer, T. J. Ecology of the plastsphere. *Nat. Rev. Microbiol.* **2020**, *18* (3), 139–151.
- (107) Madsen, T.; Rasmussen, H. B.; Nilsson, L. Anaerobic biodegradation potentials in digested sludge, a freshwater swamp and a marine sediment. *Chemosphere* **1995**, *31* (10), 4243–4258.
- (108) Fitridge, I.; Dempster, T.; Guenther, J.; de Nys, R. The impact and control of biofouling in marine aquaculture: a review. *Biofouling* **2012**, *28* (7), 649–669.
- (109) Romera-Castillo, C.; Pinto, M.; Langer, T. M.; Alvarez-Salgado, X. A.; Herndl, G. J. Dissolved organic carbon leaching from plastics stimulates microbial activity in the ocean. *Nat. Commun.* **2018**, *9* (1), 1430.
- (110) Abi-Akl, R.; Ledieu, E.; Enke, T. N.; Cordero, O. X.; Cohen, T. Physics-based prediction of biopolymer degradation. *Soft Matter* **2019**, *15* (20), 4098–4108.
- (111) Roling, W. F.; van Bodegom, P. M. Toward quantitative understanding on microbial community structure and functioning: a modeling-centered approach using degradation of marine oil spills as example. *Front Microbiol.* **2014**, *5*, 125.
- (112) Yadav, N.; Hakkarainen, M. Degradable or not? Cellulose acetate as a model for complicated interplay between structure, environment and degradation. *Chemosphere* **2021**, *265*, 128731.
- (113) Sander, M. Biodegradation of Polymeric Mulch Films in Agricultural Soils: Concepts, Knowledge Gaps, and Future Research Directions. *Environ. Sci. Technol.* **2019**, *53* (5), 2304–2315.
- (114) Martín-Closas, L.; Costa, J.; Pelacho, A. M. Agronomic Effects of Biodegradable Films on Crop and Field Environment. In *Soil Degradable Bioplastics for a Sustainable Modern Agriculture*; Malinconico, M., Ed.; Springer: Berlin, Heidelberg, 2017; p 67–104. DOI: 10.1007/978-3-662-54130-2_4.
- (115) GreenPla. Japan Bioplastics Association. <http://www.jpaweb.net/english/> (accessed 2020 December 20).
- (116) EUBP Proposes Criteria and Product Examples for Preferable Use of Compostable Plastics. *European Bioplastics*. <https://www.european-bioplastics.org/eubp-proposes-criteria-and-product-examples-for-preferable-use-of-compostable-plastics/> (accessed 2020 November 21).
- (117) Guerrini, S.; Borreani, G.; Voojis, H. Biodegradable Materials in Agriculture: Case Histories and Perspectives. In *Soil Degradable Bioplastics for a Sustainable Modern Agriculture*; Malinconico, M., Ed.; Springer: Berlin, Heidelberg, 2017; pp 35–65. DOI: 10.1007/978-3-662-54130-2_3.
- (118) *Soil Degradable Bioplastics for a Sustainable Modern Agriculture*; Springer-Verlag: Berlin Heidelberg, 2017. DOI: 10.1007/978-3-662-54130-2.
- (119) Nam, H. C.; Park, W. H. Aliphatic Polyester-Based Biodegradable Microbeads for Sustainable Cosmetics. *ACS Biomater. Sci. Eng.* **2020**, *6* (4), 2440–2449.
- (120) Ammala, A. Biodegradable polymers as encapsulation materials for cosmetics and personal care markets. *Int. J. Cosmet. Sci.* **2013**, *35* (2), 113–124.
- (121) Zhao, X.; Cornish, K.; Vodovotz, Y. Narrowing the Gap for Bioplastic Use in Food Packaging: An Update. *Environ. Sci. Technol.* **2020**, *54* (8), 4712–4732.
- (122) Hu, H.; Zhang, R.; Jiang, Y.; Shi, L.; Wang, J.; Ying, W. B.; Zhu, J. Toward Biobased, Biodegradable, and Smart Barrier Packaging Material: Modification of Poly(Neopentyl Glycol 2,5-Furandicarboxylate) with Succinic Acid. *ACS Sustainable Chem. Eng.* **2019**, *7* (4), 4255–4265.
- (123) Su, S.; Kopitzky, R.; Tolga, S.; Kabasci, S. Polylactide (PLA) and its blends with poly(butylene succinate) (PBS): a brief review. *Polymers (Basel, Switz.)* **2019**, *11* (7), 1193.
- (124) Najafi, N.; Heuzey, M. C.; Carreau, P. J.; Wood-Adams, P. M. Control of thermal degradation of polylactide (PLA)-clay nanocomposites using chain extenders. *Polym. Degrad. Stab.* **2012**, *97* (4), 554–565.
- (125) Di, Y.; Iannace, S.; Di Maio, E.; Nicolais, L. Reactively modified poly(lactic acid): Properties and foam processing. *Macromol. Mater. Eng.* **2005**, *290* (11), 1083–1090.
- (126) Rytlewski, P.; Malinowski, R.; Moraczewski, K.; Zenkiewicz, M. Influence of some crosslinking agents on thermal and mechanical properties of electron beam irradiated polylactide. *Radiat. Phys. Chem.* **2010**, *79* (10), 1052–1057.
- (127) Anderson, K. S.; Schreck, K. M.; Hillmyer, M. A. Toughening Polylactide. *Polym. Rev. (Philadelphia, PA, U. S.)* **2008**, *48* (1), 85–108.
- (128) Chou, P. M.; Mariatti, M.; Zulkifli, A.; Todo, M. Changes in the crystallinity and mechanical properties of poly(L-lactic acid)/poly(butylene succinate-co-L-lactate) blend with annealing process. *Polym. Bull. (Heidelberg, Ger.)* **2011**, *67* (5), 815–830.
- (129) Zenkiewicz, M.; Malinowski, R.; Rytlewski, P.; Richert, A.; Sikorska, W.; Krasowska, K. Some composting and biodegradation effects of physically or chemically crosslinked poly(lactic acid). *Polym. Test.* **2012**, *31* (1), 83–92.
- (130) Narancic, T.; Verstichel, S.; Reddy Chaganti, S.; Morales-Gamez, L.; Kenny, S. T.; De Wilde, B.; Babu Padamati, R.; O'Connor, K. E. Biodegradable Plastic Blends Create New Possibilities for End-of-Life Management of Plastics but They Are Not a Panacea for Plastic Pollution. *Environ. Sci. Technol.* **2018**, *52* (18), 10441–10452.
- (131) Signori, F.; Coltelli, M.-B.; Bronco, S. Thermal degradation of poly(lactic acid) (PLA) and poly(butylene adipate-co-terephthalate) (PBAT) and their blends upon melt processing. *Polym. Degrad. Stab.* **2009**, *94* (1), 74–82.
- (132) Wang, Y.; Li, M.; Shen, C. Effect of constrained annealing on the microstructures of extrusion cast polylactic acid films. *Mater. Lett.* **2011**, *65* (23–24), 3525–3528.
- (133) Pilla, S.; Kim, S. G.; Auer, G. K.; Gong, S.; Park, C. B. Microcellular extrusion-foaming of polylactide with chain-extender. *Polym. Eng. Sci.* **2009**, *49* (8), 1653–1660.
- (134) Dorgan, J. R.; Williams, J. S.; Lewis, D. N. Melt rheology of poly(lactic acid): Entanglement and chain architecture effects. *J. Rheol. (Melville, NY, U. S.)* **1999**, *43* (5), 1141–1155.
- (135) Antipov, E. M.; Dubinsky, V. A.; Rebrov, A. V.; Nekrasov, Y. P.; Gordeev, S. A.; Ungar, G. Strain-induced mesophase and hard-elastic behaviour of biodegradable polyhydroxyalkanoates fibers. *Polymer* **2006**, *47* (15), 5678–5690.
- (136) Jin, F.-L.; Hu, R.-R.; Park, S.-J. Improvement of thermal behaviors of biodegradable poly(lactic acid) polymer: A review. *Composites, Part B* **2019**, *164*, 287–296.
- (137) Vlachopoulos, J.; Polychronopoulos, N. *Basic Concepts in Polymer Melt Rheology and Their Importance in Processing*; John Wiley & Sons, Inc.: Berlin, 2012; pp 1–27. DOI: 10.1002/9781118140611.ch1.
- (138) Lim, L. T.; Auras, R.; Rubino, M. Processing technologies for poly(lactic acid). *Prog. Polym. Sci.* **2008**, *33* (8), 820–852.
- (139) Koch, H. M.; Calafat, A. M. Human body burdens of chemicals used in plastic manufacture. *Philos. Trans. R. Soc., B* **2009**, *364* (1526), 2063–78.
- (140) Oehlmann, J.; Schulte-Oehlmann, U.; Kloas, W.; Jagnytsh, O.; Lutz, I.; Kusk, K. O.; Wollenberger, L.; Santos, E. M.; Paull, G. C.; Van Look, K. J. W.; Tyler, C. R. A critical analysis of the biological impacts of plasticizers on wildlife. *Philos. Trans. R. Soc., B* **2009**, *364* (1526), 2047–62.
- (141) Hahladakis, J. N.; Velis, C. A.; Weber, R.; Iacovidou, E.; Purnell, P. An overview of chemical additives present in plastics: Migration, release, fate and environmental impact during their use, disposal and recycling. *J. Hazard. Mater.* **2018**, *344*, 179–199.
- (142) Turner, A. Black plastics: Linear and circular economies, hazardous additives and marine pollution. *Environ. Int.* **2018**, *117*, 308–318.
- (143) Zimmermann, L.; Dombrowski, A.; Völker, C.; Wagner, M. Are bioplastics and plant-based materials safer than conventional plastics? In vitro toxicity and chemical composition. *Environ. Int.* **2020**, *145*, 106066.
- (144) Zhang, F.; Zhao, Y.; Wang, D.; Yan, M.; Zhang, J.; Zhang, P.; Ding, T.; Chen, L.; Chen, C. Current technologies for plastic waste treatment: A review. *J. Cleaner Prod.* **2021**, *282*, 124523.
- (145) Kliopova, I.; Staniškas, J. K.; Stunženas, E.; Jurovickaja, E. Bio-nutrient recycling with a novel integrated biodegradable waste

management system for catering companies. *J. Cleaner Prod.* **2019**, *209*, 116–125.

(146) The State of Organics Recycling in the U.S. *BioCycle*. <https://www.biocycle.net/state-organics-recycling-u-s/> (accessed 10 January 2021).

(147) Mali Sandip, T.; Khare Kanchan, C.; Biradar Ashok, H. Enhancement of methane production and bio-stabilisation of municipal solid waste in anaerobic bioreactor landfill. *Bioresour. Technol.* **2012**, *110*, 10–17.

(148) Wan, S.; Sun, L.; Douieb, Y.; Sun, J.; Luo, W. Anaerobic digestion of municipal solid waste composed of food waste, wastepaper, and plastic in a single-stage system: Performance and microbial community structure characterization. *Bioresour. Technol.* **2013**, *146*, 619–627.

(149) Pelleria, F.-M.; Pasparakis, E.; Gidarakos, E. Consecutive anaerobic-aerobic treatment of the organic fraction of municipal solid waste and lignocellulosic materials in laboratory-scale landfill-bioreactors. *Waste Manage.* **2016**, *56*, 181–189.

(150) Grosso, M.; Nava, C.; Testori, R.; Rigamonti, L.; Viganò, F. The implementation of anaerobic digestion of food waste in a highly populated urban area: an LCA evaluation. *Waste Manage. Res.* **2012**, *30*, 78–87.

(151) Quecholac-Pina, X.; Hernandez-Berriel, M. D. C.; Manon-Salas, M. D. C.; Espinosa-Valdemar, R. M.; Vazquez-Morillas, A. Degradation of Plastics under Anaerobic Conditions: A Short Review. *Polymers (Basel, Switz.)* **2020**, *12* (1), 109.

(152) Jones, D. Grinding waste away. *Nature* **2000**, *403* (6772), 847–847.

(153) Worch, J. C.; Dove, A. P. 100th Anniversary of Macromolecular Science Viewpoint: Toward Catalytic Chemical Recycling of Waste (and Future) Plastics. *ACS Macro Lett.* **2020**, *9* (11), 1494–1506.

(154) Sharma, B.; Jain, P. Deciphering the advances in bioaugmentation of plastic wastes. *J. Cleaner Prod.* **2020**, *275*, 123241.

(155) Nair, N. R.; Sekhar, V. C.; Nampoothiri, K. M. Augmentation of a Microbial Consortium for Enhanced Polylactide (PLA) Degradation. *Indian J. Microbiol.* **2016**, *56* (1), 59–63.

(156) Rodrigues, L. C.; Puig-Ventosa, I.; López, M.; Martínez, F. X.; Ruiz, A. G.; Bertrán, T. G. The impact of improper materials in biowaste on the quality of compost. *J. Cleaner Prod.* **2020**, *251*, 119601.

(157) Echavarri-Bravo, V.; Thygesen, H. H.; Aspray, T. J. Variability in physical contamination assessment of source segregated biodegradable municipal waste derived composts. *Waste Manage.* **2017**, *59*, 30–36.

(158) Pandeyaswargo, A. H.; Premakumara, D. G. J. Financial sustainability of modern composting: the economically optimal scale for municipal waste composting plant in developing Asia. *Int. J. Recycl. Org. Waste Agricult.* **2014**, *3* (4), 4.

(159) Karamanlioglu, M.; Preziosi, R.; Robson, G. D. The Compostable Plastic Poly(lactic) Acid Causes a Temporal Shift in Fungal Communities in Maturing Compost. *Compost Sci. Util.* **2017**, *25* (4), 211–219.

(160) Shen, M.; Zeng, G.; Zhang, Y.; Wen, X.; Song, B.; Tang, W. Can biotechnological strategies effectively manage environmental (micro)-plastics? *Sci. Total Environ.* **2019**, *697*, 134200.

(161) Aguilar, A.; Wohlgemuth, R.; Twardowski, T. Perspectives on bioeconomy. *New Biotechnol.* **2018**, *40*, 181–184.

(162) Wei, R.; Tiso, T.; Bertling, J.; O'Connor, K.; Blank, L. M.; Bornscheuer, U. T. Possibilities and limitations of biotechnological plastic degradation and recycling. *Nature Catalysis* **2020**, *3* (11), 867–871.

(163) Purohit, J.; Chattopadhyay, A.; Teli, B. Metagenomic exploration of plastic degrading microbes for biotechnological application. *Curr. Genomics* **2020**, *21* (4), 253–270.

(164) Hauer, B. Embracing Nature's Catalysts: A Viewpoint on the Future of Biocatalysis. *ACS Catal.* **2020**, *10* (15), 8418–8427.

(165) Zhang, X.; Fevre, M.; Jones, G. O.; Waymouth, R. M. Catalysis as an Enabling Science for Sustainable Polymers. *Chem. Rev. (Washington, DC, U. S.)* **2018**, *118* (2), 839–885.

(166) Bilal, M.; Iqbal, H. M. N.; Guo, S.; Hu, H.; Wang, W.; Zhang, X. State-of-the-art protein engineering approaches using biological

macromolecules: A review from immobilization to implementation view point. *Int. J. Biol. Macromol.* **2018**, *108*, 893–901.

(167) Sheldon, R. A.; Woodley, J. M. Role of Biocatalysis in Sustainable Chemistry. *Chem. Rev.* **2018**, *118* (2), 801–838.

(168) Inderthal, H.; Tai, S. L.; Harrison, S. T. L. Non-Hydrolyzable Plastics – An Interdisciplinary Look at Plastic Bio-Oxidation. *Trends Biotechnol.* **2021**, *39* (1), 12–23.

(169) Yang, Y.; Yang, J.; Wu, W.-M.; Zhao, J.; Song, Y.; Gao, L.; Yang, R.; Jiang, L. Biodegradation and Mineralization of Polystyrene by Plastic-Eating Mealworms: Part 2. Role of Gut Microorganisms. *Environ. Sci. Technol.* **2015**, *49* (20), 12087–12093.

(170) Beadle, G. W.; Tatum, E. L. Genetic Control of Biochemical Reactions in *Neurospora*. *Proc. Natl. Acad. Sci. U. S. A.* **1941**, *27* (11), 499.

(171) Green, D. S.; Boots, B.; Sigwart, J.; Jiang, S.; Rocha, C. Effects of conventional and biodegradable microplastics on a marine ecosystem engineer (*Arenicola marina*) and sediment nutrient cycling. *Environ. Pollut. (Oxford, U. K.)* **2016**, *208*, 426–434.

(172) Green, D. S. Effects of microplastics on European flat oysters, *Ostrea edulis* and their associated benthic communities. *Environ. Pollut. (Oxford, U. K.)* **2016**, *216*, 95–103.

(173) González-Pleiter, M.; Tamayo-Belda, M.; Pulido-Reyes, G.; Amariei, G.; Leganes, F.; Rosal, R.; Fernandez-Pinas, F. Secondary nanoplastics released from a biodegradable microplastic severely impact freshwater environments. *Environ. Sci.: Nano* **2019**, *6* (5), 1382–1392.

(174) Frere, L.; Maignien, L.; Chalopin, M.; Huvet, A.; Rinnert, E.; Morrison, H.; Kerninon, S.; Cassone, A.-L.; Lambert, C.; Reveillaud, J.; Paul-Pont, I. Microplastic bacterial communities in the Bay of Brest: Influence of polymer type and size. *Environ. Pollut. (Oxford, U. K.)* **2018**, *242*, 614–625.

(175) Zuo, L.-Z.; Li, H.-X.; Lin, L.; Sun, Y.-X.; Diao, Z.-H.; Liu, S.; Zhang, Z.-Y.; Xu, X.-R. Sorption and desorption of phenanthrene on biodegradable poly(butylene adipate co-terephthalate) microplastics. *Chemosphere* **2019**, *215*, 25–32.

(176) Sekhri, P. *Harvesting the Plastic We Have Sowed: Costs and Challenges in, and a Novel Application of Blockchain for Implementing Extended Producer Responsibility in Chile*. Thesis: System Design and Management Program, Massachusetts Institute of Technology, 2018. <http://hdl.handle.net/1721.1/118522> (accessed April 2021).

(177) BASF Introduces Innovative Pilot Blockchain Project to Improve Circular Economy and Traceability of Recycled Plastics. BASF. <https://www.basf.com/us/en/media/news-releases/2020/02/basf-introduces-innovative-pilot-blockchain-project-to-improve-c.html> (accessed 2021 January 18).

(178) *Our World in Data*. <https://ourworldindata.org/> (accessed 2021 January 10).

(179) Hellweg, S.; Milà i Canals, L. Emerging approaches, challenges and opportunities in life cycle assessment. *Science* **2014**, *344* (6188), 1109.

(180) Clift, R.; Druckman, A. *Taking Stock of Industrial Ecology*; Springer International Publishing, 2015. DOI: 10.1007/978-3-319-20571-7.

(181) Korhonen, J.; Honkasalo, A.; Seppälä, J. Circular Economy: The Concept and its Limitations. *Ecological Economics* **2018**, *143*, 37–46.