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Review

Sustainable bioplastics derived from renewable natural resources for food packaging

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SUMMARY

Food packaging is one of the leading sectors for the end use of plastics. Bioplastic is produced from natural renewable resources such as crops, wood pulp, and herbaceous fibers. This article summarizes the rational design of bioplastics from natural resources for food packaging. The bioplastic properties considered include thermal properties, mechanical performance, oxygen/moisture resistance, and biodegradability. Bioplastic degradability and technologies for handling bioplastic waste are discussed. Various aspects of the sustainability of bioplastics (e.g., environmental profile, techno-economic analysis, and societal impact) are investigated. The main challenges of bioplastic application, such as low fracture strain and inferior barrier properties, are discussed. Mitigation approaches to overcome the mechanical properties of bioplastics, such as adding plasticizers, are also discussed. Bioplastic can have properties comparable with fossil-based plastics. Bioplastic can be an alternative to conventional plastic in most applications of food packaging, which can reduce the carbon footprint and environmental impact because of its biodegradability.

INTRODUCTION

Petrochemical plastics are durable and inexpensive materials.[1] They are widely used in construction, packaging, aviation, and electronics.[1] They can be applied in pharmaceutical instruments, 3D modeling, automobiles, and household appliances.[2] Among plastics, polyethylene is widely used as a food packaging material because of its water and gas barrier, thermal stability, low density, and ease of production.[3] However, most plastics (e.g., polystyrene, polyethylene, and polyvinyl chloride [PVC]) need hundreds of years to degrade because of their stable backbone polymer chains.[1] More than 16 million tons of plastic packaging waste were generated in 2016 in the European Union.[4] A third of plastics will end up as marine or terrestrial pollution.[5] Plastic pollution has a highly negative influence on the marine environment and the food chain, including on human beings.[5]

The demand for bioplastics to replace petrochemical plastics is emerging, especially in the food packaging industry.[6] This review defines "bioplastic" as plastic made from natural resources. However, in some literature, bioplastic also includes petrochemical-derived, biodegradable plastic. "Biodegradable" is defined as the capability to be degraded well in a natural environment. Bioplastics take less time to degrade, reduce fossil fuel consumption, create new streams for plastic waste recycling, reduce the space required to manage waste, and reduce the amount of

PROGRESS AND POTENTIAL

Petrochemical plastics bring tremendous convenience to daily life and are widely used in packaging, containers, furniture, electronics, etc. However, most plastics need hundreds of years to degrade because of their stable polymer chains. A third of plastics ends up as marine or terrestrial pollution. Plastic pollution has a highly negative influence on the marine environment and the food chain. Therefore, a demand for bioplastics to replace petrochemical plastics is emerging, especially in the one-time-use food packaging industry. Bioplastics take less time to degrade, save energy during manufacturing, and reduce waste. The main challenges of bioplastics encountered by manufacturers and consumers are adaptability to the existing manufacturing line, performance, and cost.

In this review, a rational design of bioplastics from natural resources for food packaging is evaluated. Management of bioplastic waste regarding degradation and recycling is discussed. Adding plasticizers is an effective approach to improving the mechanical performance of bioplastics. Bioplastic can be an alternative to conventional plastic in most applications of food packaging, with less of a carbon

greenhouse gas (GHG) emissions.[2,7] There are many materials and methods for fabricating bioplastics. Bioplastics comprise a suite of bio-based plastics prepared from a variety of renewable sources such as proteins, wood, potatoes, corn, vegetable oils, and food waste. Figure 1A shows the concept of the use of renewable and biodegradable plastics derived from biomass to replace traditional plastics for food packaging. Petroleum-based and nonrenewable plastics include polycaprolactone (PCL), polycarbosilane (PCS), and poly(butylene adipate-co-terephthalate) (PBAT). Typically, polysaccharides, lipids, proteins, wood, potatoes, corn, vegetable oils, cereal crops, and food waste can be used to prepare bioplastics. Figure 1B shows a visual representation of biodegradable polymers and the four main production pathways. The figure shows the products in each category, what they are made from, and the resulting plastics.

A suitable food packaging plastic is expected to keep its stored product with an acceptable moisture content that prevents microbial growth and spoilage.[3] The main plastics used in food packaging are polyolefins (PE and polypropylene [PP]), polyester, polystyrene (PS), and nylon polycarbonate.[6] These plastics have high strength and excellent mechanical reliability, meeting the basic requirements for food packaging applications.[6] Some researchers have developed strategies to improve bioplastic application in food packaging. For example, Behera et al.[6] found that a higher bentonite concentration (1.5 wt %) can effectively improve yam starch-based bioplastic properties, making it a potential replacement for petrochemical plastic in food packaging applications. Current research on bioplastic for food applications is often focused on mechanical properties. However, to promote its application, other tests are also needed; for example, biocompatibility, food safety, water vapor permeability (to prevent moisture transmission from the environment to the food), and oxygen barrier properties.

Food waste has significant social, economic, and environmental impacts.[9] Approximately one-third of the food in the world goes to waste or is lost before or after it reaches consumers. A major pathway to prevent food waste is developing packaging that protects food from external damage (e.g., oxidation) and extends its shelf life. Innovation in sustainable packaging material to better preserve the freshness and quality of food during its distribution and storage is urgent. Bioplastics have the potential to provide such properties, and they are biodegradable. The demand for sustainable packaging innovation has increased because of the environmental issues related to packaging.[9] For example, hydro-stable and strong lignocellulosic bioplastics have high biodegradability in the natural environment.[1] These bioplastics also have good recyclability. After their end of life, they can be broken into a cellulose-lignin slurry through mechanical recycling.[1]

This review evaluates rational design (i.e., a design that involves a process of reasoning and calculation) of bioplastics from natural resources for food packaging. When traditional petroleum-based plastics will be replaced by bioplastics for food packaging, the main problems faced by manufacturers and consumers include poor performance (e.g., high brittleness), high cost, poor public understanding, and new environmental impact (e.g., unfavorable land use change).[10,11] Current advances and future directions of bioplastic development are discussed. The major properties (e.g., mechanical performance, oxygen/moisture resistance, and biocompatibility) of bioplastics as plastic alternatives for food packaging are discussed. The material processing for bioplastics is investigated, including pulp molding, top-down processing, and bottom-up processing. Figure 2 shows three pathways for rational design of bioplastics from natural resources for food packaging.

footprint and environmental impact. Recycling should be promoted because it can significantly improve the sustainability of bioplastics. The capability of bioplastics to be recycled varies, and appropriate technologies should be selected based on the nature of biopolymers.

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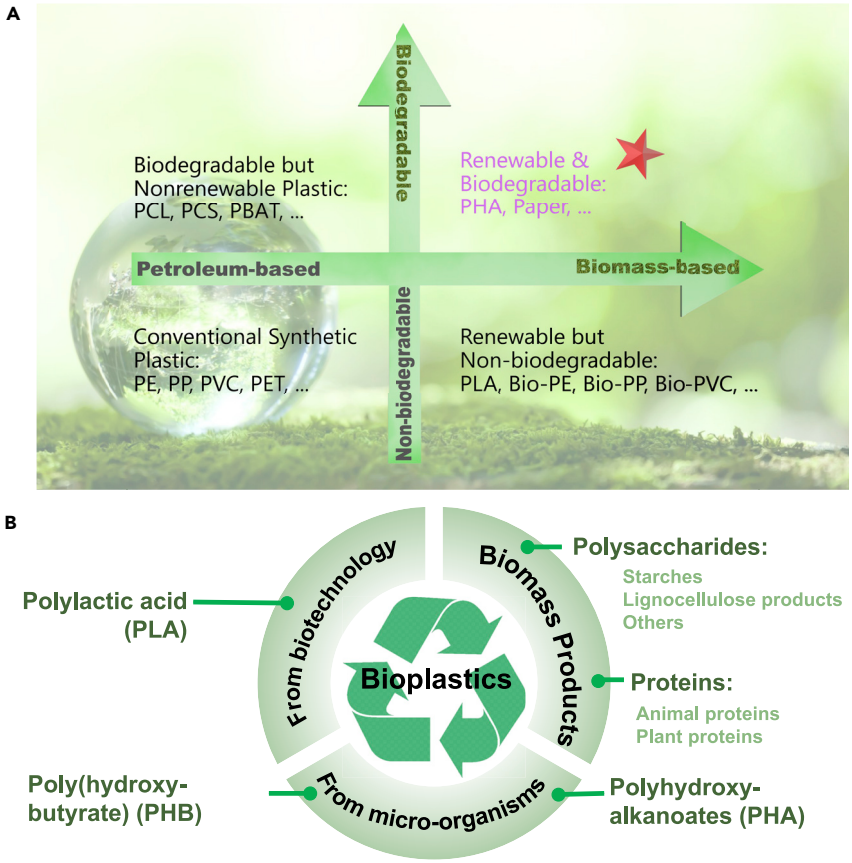

Figure 1. Outline of the review

(A) Conceptual display of use of renewable and biodegradable plastics derived from biomass to replace nonbiodegradable or nonrenewable plastics for food packaging.

(B) Classification of the main bioplastics.[8]

The main challenges in these processing approaches are discussed. Bioplastic management includes evaluation of bioplastic biodegradability and technologies for handling bioplastic waste (e.g., recycling, industrial composting, incineration, and biodegradation). The sustainability of bioplastics, including their environmental profile and societal impact, is discussed. Finally, a perspective for bioplastic development for food packaging applications is given.

BIOPLASTICS FROM NATURAL RESOURCES FOR FOOD PACKAGING

Food packaging is one of the leading sectors for end use of plastics, and plastics are mainly employed as primary packaging in addition to secondary and tertiary packaging (which assists with and facilitates transportation of goods from manufacturers in their distribution chain).[10] In primary packaging, the packaging materials are in direct contact with food products, providing a barrier between products and environmental hazards, which is the main line of protection from bacteria, contamination, and degradation. To maintain substance hygiene and integrity during transportation and storage and to extend food shelf life, it is crucial to consider a few key factors when selecting packaging materials, including chemical-physical, thermal, mechanical, and barrier properties and optical characteristics.

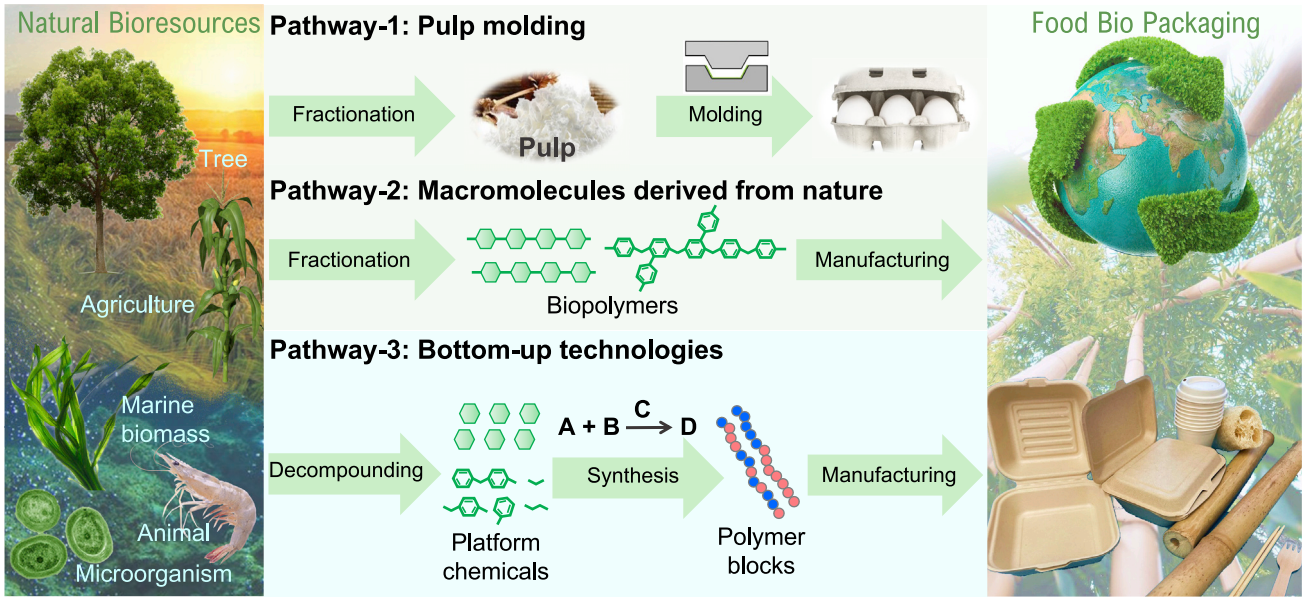

Figure 2. Schematic of three pathways from natural resources to rational design of food bio packaging

Shown are the pulp molding method, macromolecules derived from nature, and bottom-up technologies of synthesis polymers from monomers derived from biomass.

Material properties

Figure 3A summarizes typical thermal (glass transition temperature [T_g] and melting temperature [T_m]), mechanical, and barrier properties of commonly used fossil-based plastics and bioplastics in the commercial market.[10,12–16] Conventional fossil-based plastics mainly include PET, high-density PE (HDPE), PVC, low-density PE (LDPE), PP, and PS. Bioplastics mainly include PLA, thermoplastic starch (TPS), polyhydroxyalkanoates (PHAs), and poly(butylene succinate) (PBS). Generally, bioplastics have properties (e.g., tensile strength) comparable with those of the listed fossil-based plastics. Bioplastics can be adopted as alternatives to conventional plastic materials in some applications of food packaging, leading to a reduced carbon footprint in plastic production and lower environmental impact because of their biodegradability.[17] Figure 3B shows the resin identification codes for plastics.

Major considerations of bioplastics as plastic alternatives for food packaging

T_g characterizes the thermal transition between a soft and rubbery state and a hard and relatively brittle glass state along with decreasing the temperature. Plastics generally undergo drastic changes in physical and mechanical properties over a small range of temperatures around T_g. For packaging, T_g should not fall into the temperature range where the plastic material could be deformed during usage.[19] Likewise, the maximum temperature for end use should be well below T_m, above which the plastic turns into a viscous liquid.

Plastics with a low T_g are desired for food packaging for storage under freezing conditions, but their applications in hot food packaging are limited. Bioplastics and conventional plastics have a wide span of T_g, which can satisfy the typical requirement of temperature range for food storage after packaging, from −18°C to 4°C.[20] Commercially available bioplastics with relatively low T_g (e.g., PHAs) often have poor thermal stability when exposed to high temperatures.[10] Improving the heat resistance of bioplastics certainly expands their usage and facilitates replacement of conventional plastics. Common strategies, including additives or fillers, blending with other

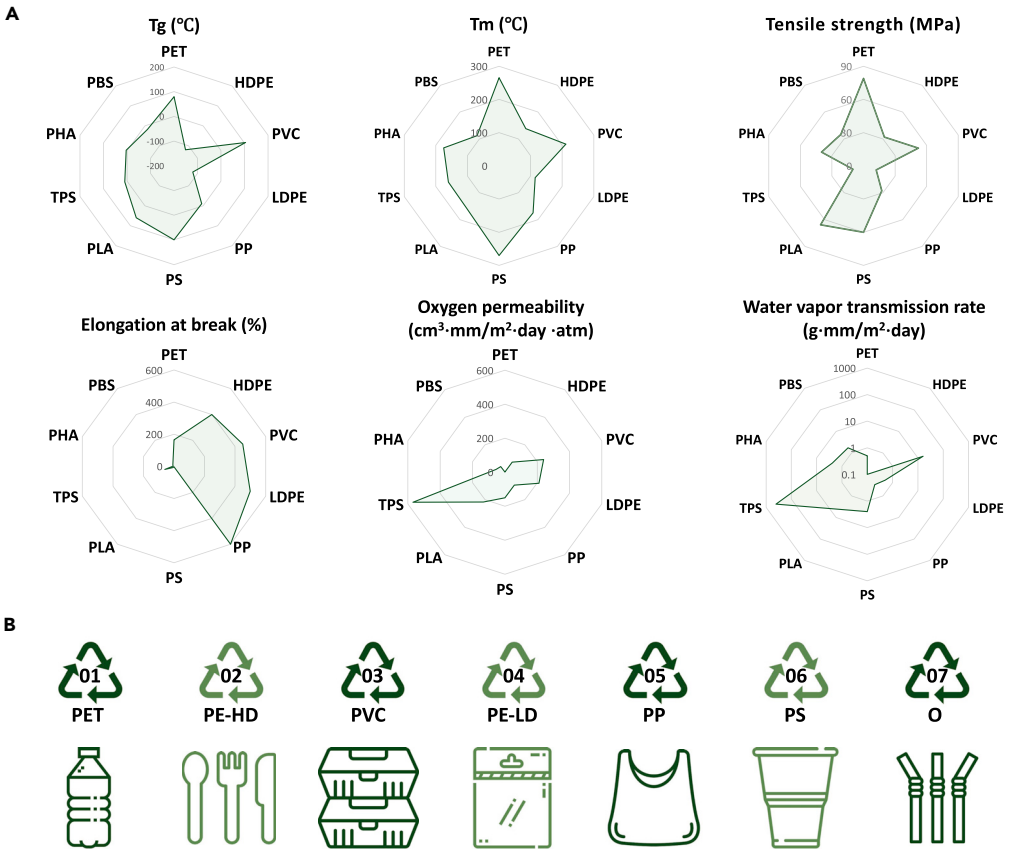

Figure 3. Plastic properties and identification codes

(A) Typical properties of common conventional fossil-based plastics (PET unoriented, HDPE, LDPE, PVC, PP, and PS) and bioplastics (PLA, TPS, PHA, and PBS).[10,12–16]

(B) Resin identification codes (symbols appearing on plastic products that identify the resin of which the product is made) for plastic recycling. The recycling process benefits from separation by resin type, where "O" is other resins (identified by the American Plastics Council).[18]

polymers and compounds, and modifying processing techniques,[20] are applied to improve bioplastic thermal stability. For instance, melt-stretching of amorphous PLA, reducing large spherulitic crystal formation, and inducing nanocrystal formation significantly improve the thermal and mechanical properties of PLA.[21]

The tensile properties of plastics dictate their performance under stress and are important for design of plastic parts and protection of food packaging. The tensile strength depends on the plastic type, chemical modification, additives, and processing conditions.[19] Bioplastics can have a similar tensile strength as conventional plastics, offering comparable mechanical performance that is highly suitable for rigid food packaging needs.[19] Although elongation at break (i.e., fracture strain) typically has a small value (e.g., <5%) for rigid plastics, the value can go up to several hundred percent for packaging polyolefins.[22] Materials with high elongation at break and high tensile strength have high toughness. The larger the elongation at break, the higher the ductility. For food packaging applications, good ductility is often desired. Adding plasticizers is a viable approach for increasing elongation at break and tensile strength.[23]

The barrier properties of plastics are crucial for food packaging applications to retain food quality and shelf life. In general, plastics are relatively permeable to small

Table 1. Characteristics of three groups of plastics

	PLA	Bio-based PET	PCL
Source	first-generation feedstock (e.g., corn, potato)[19,26]	first generation feedstock (e.g., sugarcane)[26]	synthetic feedstock[27]
Characteristics	compostable, renewable, and biocompatible but brittle[26]	one of the most important bioplastics economically[28]	can degrade within 2–4 years;[29] mechanical properties similar to nondegradable synthetic polymers[27]
Manufacturing process	monomer (lactic acid) derived from natural resources by bacterial fermentation[26]	fermentation and distillation from sugar and then *trans*-esterification with terephthalic acid (TPA)[30]	a semicrystalline polyester synthesized by ring-opening polymerization of caprolactone[26]
Main use	packaging, medical devices, food service ware, and coatings[26]	packaging, engineering components, bottles, film equipment[31]	medical implants[29]
Environment impact	CO_2 generation during the biodegradation process is countered by CO_2 consumed from feedstock[19,32]	combined with petroleum-based materials during fabrication process	CO_2 generation during production

molecules such as oxygen, carbon dioxide, water vapor, organic vapors, and liquids, presenting a range of mass transfer characteristics.[24] Among different types of permeants, oxygen and water vapor are two important ones to consider for food packaging because their transfer between the internal and external environment through the plastic packaging barrier affects food spoilage. Specifically, high oxygen content aids oxidation and maintains high respiration rates of fresh produce, leading to a shortened shelf-life; excessive moisture pickup can raise food susceptibility to microorganisms, which rot and damage the food. Generally, bioplastics have inferior barrier properties to oxygen and moisture compared with conventional plastics. The performance can further decrease for hydrophilic bioplastics under humid conditions. These disadvantages are major challenges for application of bioplastics to food packaging.[16,25] Consequently, increasing the barrier properties of bioplastics has drawn significant attention, which promotes sustainable packaging.

Current commodity and niche bioplastics for food packaging and their properties

Table 1 lists the main characteristics of polymers, including the advantages and disadvantages of their manufacturing processes and their environmental impacts. For PLA, the generation of CO_2 during the biodegradation process is indirectly countered by the CO_2 consumed by the first-generation feedstock (the first crops and plants [e.g., sugarcane, corn, and wheat] used to produce bioplastics). PET and PCL use petroleum-based materials during their production, contributing to climate change.

Many factors affect the production and characteristics of plastic. First are the source materials of the three plastic polymers. PLA is produced from renewable first-generation feedstock, such as corn and potatoes. Renewable first-generation feedstock is the best type of bio-based polymer because of its greatly reduced dependence on fossil fuels. Bio-based PET and PCL use a nonrenewable resource in their production process. PET uses some crops, such as sugarcane, and some petroleum-based materials. As PET, PCL uses completely synthetic feedstock. PCL is not biobased but biodegradable, PET is partially biobased but not biodegradable, and PLA is bio-based and compostable.

Bio-derived polymers are created in part to counter the negative effect of regular nonbiodegradable petroleum-based plastics on the environment. PLA is one of the leading candidates for petroleum-based plastic replacement because it is a thermoplastic, high-strength, high-modulus polymer that can be made from annually

renewable resources.[33–36] Unlike other biopolymers, such as PCL or PET, PLA is completely environmentally friendly; however, PLA and PCL are similar in that they can be used for medical applications because they do not degrade to form toxic materials in the human body. PLA, bio-based PET, and PCL have great processibility, allowing them to become film extruding and film forming.[33] PLA has a tensile strength and elastic modulus that is comparable with PET and a slow degradation rate similar to PET.[33] However, PLA has a low toughness, being a very brittle material with less than 10% elongation at break.[33,37] All three of these polymers are used as packaging materials; however, PLA has been most used for commodity applications because it requires 25%–55% less energy to produce relative to petroleum-based polymers.[33]

PLA degrades slowly and produces CO_2 when it degrades. Some of this offset is countered by the fact that the renewable resources used during production lead to some CO_2 being absorbed and utilized. Of course, production of PLA still contributes to global warming with its use of nonrenewable energy. The majority of this comes from resin production and the following transportation of materials.[38] Because of its large availability, biocompatibility, degradability, excellent processability, and mechanical properties close to conventional fossil-based polymers, PLA has the largest share in the global production capacity of all bioplastics.[17,39,40] PLA (or modified PLA) can be applied as a potential alternative to PET, LDPE, HDPE, and PS, and PLA is mostly used in the packaging industry. In particular, its optical transparency gives it a unique advantage for food packaging. PLA is already used for vegetable and fruit packaging as well as food and beverage containers. With further improvement of thermal stability and barrier properties, PLA is expected to expand its packaging applications to more food products.[17,39,40]

Starch-based polymers comprise another major portion of bioplastics produced annually.[17] Starch is a biodegradable, cheap, and widely available natural resource. Its brittle and hydrophilic nature makes native starch unsuitable for packaging applications. However, starch can be converted into thermoplastic by mixing with a plasticizer and/or water under application of thermal and mechanical energy. The obtained TPS is a cost-effective biodegradable material that can potentially be substituted for synthetic polymers (e.g., PS) in packaging.[41] TPS is used as food wraps and layers between food components, although its application in other food packaging is still restrained by the relatively poor water vapor resistance and low mechanical properties. To overcome these limitations, composite formulations of TPS, such as blending with other biopolymers (e.g., cellulose and PLA), are being sought to improve the corresponding properties.[41,42]

Material processing for bioplastics

Pulp molding for plastic alternatives

Pulp molding is a key component in developing alternatives to synthetic plastics for food packaging. However, pulp molding has a significant effect on many of the qualities of the produced materials. To fully optimize creation of molded pulp, more studies need to be done to determine a way to overcome the drawbacks. The secondary fibers, acquired from newspapers and used books, often used in current molded pulp contain leftover residual ink and other chemicals, which is a concern in food packaging. However, switching from recycled wood fibers to virgin wood fibers would not only be counterproductive in terms of helping the environment but would also require more energy because the energy required to produce 1 ton of recycled fiber pulp is 27% lower than that for virgin fiber pulp.[43] In addition to

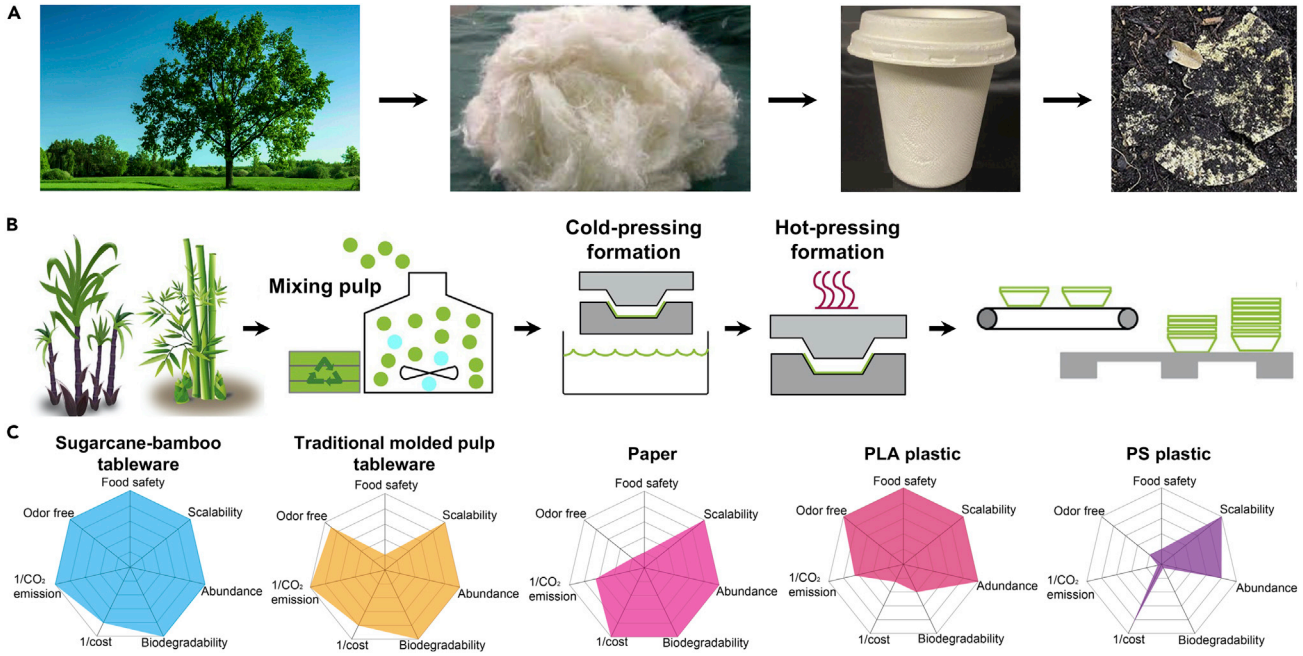

Figure 4. Conceptual design of molding for bioplastics

(A) Biodegradable cup molded from biomass.[47]

(B) Schematic of molded pulp using sugarcane bagasse and bamboo to produce biodegradable tableware as an alternative to plastics used in the food industry.[47]

(C) Components and advantages of sugarcane-bamboo tableware compared with other commercial products.[47]

containing negative chemicals, using recycled materials also has negative effects on the overall mechanical strength of the materials.[44]

Molded pulp is a recyclable, compostable, and environmentally friendly material that has gained increasing interest for food packaging with the developing demand for green and sustainable products.[45–47] Figures 4A and 4B show schematic designs of pulp molding for bioplastics.[47] The major component of molded pulp is cellulose fibers, a type of natural biopolymer in plants. The cellulose fibers interact with each other through hydrogen bonding in the pulp, generating the strength of the final product. Given good tensile and compressive strength, the molded pulp is applied as a plastic alternative (e.g., for PS) and finds application in egg cartons, food trays, and containers.

As shown in Figure 4B, sugarcane bagasse left from the sugar industry as a feedstock was used to prepare fully biodegradable tableware by using a scalable molding method. Long bamboo fibers were mixed with the sugarcane bagasse fibers to enhance the mechanical strength of the resultant plastic alternatives. Compared with traditional molded pulp tableware and commercial products, such as paper, PLA, and PS, the fabricated sugarcane-bamboo tableware has performance as required for food packaging. As shown in Figure 4C, the sugarcane-bamboo tableware has a low heavy metal content (Pb, 0.3633 mg/kg), oil stability (level 6, Technical Association of the Pulp and Paper Industry [TAPPI] standard), excellent hydrophobicity (contact angle, 127°), and high mechanical strength (tensile strength, 35 MPa). The tableware can be mostly biodegraded under natural conditions within 60 days, which is much shorter that degradation of synthetic plastics. In comparison with production of PS (7.4 kg/kg) and traditional paper manufacturing (0.5 kg/kg),

the manufacturing process of this tableware had much less CO_2 emission (0.2 kg/kg). Tableware made from biomass feedstock represents an environmentally friendly and biodegradable alternative to synthetic plastics for food packaging.

Cellulose can be derived from various natural renewable resources, such as agricultural crops and associated waste materials and can be recycled from kraft, paper, and molded pulp products.[48,49] At the industrial scale, cellulose fibers are extracted by mechanical pulping and chemical pulping. The former process leads to a high yield of fibers with low strength because of the high lignin content and hornification. In contrast, chemical pulping, removing lignin, results in high-strength fibers. Because of the hydrophilic nature of cellulose fibers,[50] molded pulp is quite responsive to moisture and loses a lot of strength at high humidity. For food packaging applications, addition of a barrier is required to enhance resistance to water and gases. A simple solution is to laminate the molded pulp with a synthetic polymer film (e.g., PE). However, the synthetic polymer film is difficult to separate after use and hard to degrade in the environment, making the whole product life cycle not fully sustainable. Hence, better coating techniques and additive formulations are needed for application of molded pulp in food packaging. Along with improving the production techniques, pulp materials other than wood fibers, such as fruit pomace and bamboo,[47,51] can be exploited to produce low-cost, environmentally friendly, and biodegradable plastic alternatives.

Addition of nanofillers has been shown to simultaneously increase the barrier properties and mechanical reinforcement, making the derived bionanocomposites a promising material for food packaging applications.[52,53] Nanoclays[52,53] and cellulose nanocrystals[54] are common nanofillers used in industrial applications that have attracted considerable interest for food packaging because of safety, efficacy, and cost considerations. The large variety of these nanofillers allows fine-tuning of properties and, hence, optimization of bio-based food packaging materials.

For composite production, thermomechanical processes with high shearing force, high temperature, and sufficient time are required to ensure filler dispersion in the polymer matrix. Common processing methods include melt compounding, extrusion, compression molding, and injection molding. To achieve satisfactory dispersion of nanofillers, processing techniques such as *in situ* polymerization, solution intercalation, and melt intercalation are available.[55] From an industrial perspective, melt intercalation processing, being more flexible and economical, is preferred and requires a strong filler-matrix affinity that often calls for surface modifications (e.g., cationic exchange and silane/polymer grafting)[52] of nanofillers.

Macromolecules derived from terrestrial plants, ocean biomass, and animals as building blocks for bioplastic

Polyurethane (PU) materials are used in our daily lives for various applications such as foam, coatings, adhesives, sealants, and elastomers. Figure 5 shows bioplastic production from renewable resources. In 2016, PU ranked sixth among all polymers based on annual global production. PU is traditionally produced by a reaction between polyols and isocyanates. Besides petroleum-based polyol, bio-based polyols can be produced from many renewable resources, such as vegetable oil, cashew nut shell liquid, cellulose, lignin, and protein.[56] Unsaturated fatty acids can be converted into polyols via many pathways, such as epoxidation and ozonolysis, and castor oil can be used directly as a polyol because of the free hydroxyl group on ricinoleic acid.

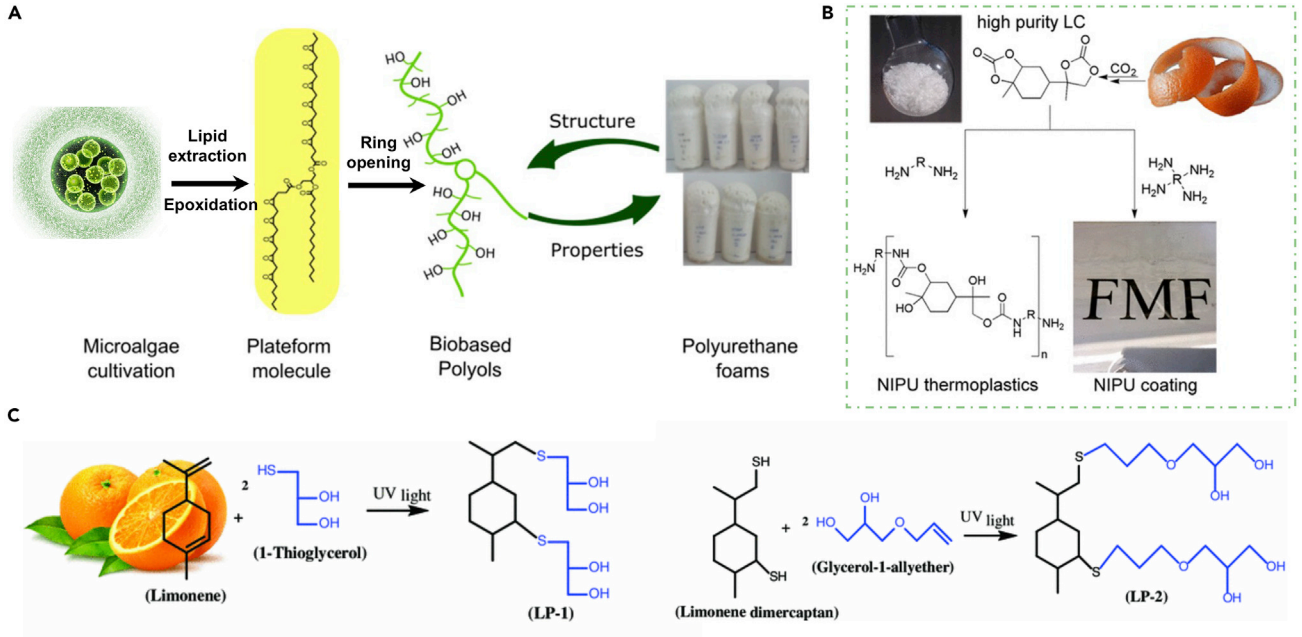

Figure 5. Bioplastics produced from renewable resources

(A) Microalgae oil epoxidized and ring opened to be introduced into rigid PU foams.[57]

(B) Oxidation and subsequent catalytic carbonation of limonene, derived from orange peels, afford high-purity limonene dicarbonate (LC) as a building block for tailoring non-isocyanate PUs (NIPUs) from renewable resources. Incorporation of small amounts of high-purity LC substantially improves NIPU's stiffness, strength, and glass temperature.[58]

(C) Synthesis of polyols based on limonene via a photochemical reaction.[59]

Cashew nut shell liquid is a nonedible chemical (mainly cardanol) extracted from cashew agriculture waste. Cardanol is used to produce polyols for PU applications. The aromatic ring structure in the back bond imparts the PU material with rigidity, and cardanol PU demonstrates good adhesion to metal and improved hydrophobicity.[60,61] Terpene, such as limonene, has been used as a precursor to produce polyol. The ring structure of limonene can provide extra rigidity.[59] Lignocellulosic biomass can be liquefied in polyhydric alcohol (e.g., glycerol and ethylene glycol) to produce precursors for PU production.[62] Liquefaction is usually conducted at 150°C–250°C under ambient pressure using an acid or base as a catalyst. Depending on the biomass and processing parameters, polyols with different properties can be produced. The liquefied product is a mixture of polyol and can be used directly for foams, adhesives, and films.[63]

Protein is also an abundant biomass and has been used for PU production. Kumar et al.[64] modified algal protein into polyol. Protein-based polyols could replace up to 5% of other polyols with comparable performance. Traditional PU production involves isocyanate, which is hazardous and is scrutinized by regulatory agencies across the world. With a growing demand for green chemistry exploiting renewable resources, intensive efforts have been made to synthesize PU via non-isocyanate routes.

One of the most promising pathways to produce non-isocyanate PU (NIPU) is through an aminolysis reaction.[65] A wide range of unsaturated vegetable oil, fish oil, and algal oil has been used to produce NIPU via this approach.[66] Terpene- and cardanol-based chemicals have also been used to produce NIPU.[58] Lignin and cellulose can also be used as precursors to produce NIPU. For example,

Fleischer et al.[67] added cyclic carbonate groups on the surface of cellulose to convert it into a reactive filler for a NIPU composite and demonstrated improved performance. Schmidt et al.[68] synthesized NIPU from a precursor derived from erythritol or butadiene. NIPU has the potential to replace conventional PU as a new-platform polymer. However, there are still several technical hurdles (low reactivity and performance) impeding industrial application of NIPU.[65,66] More research is needed to fully understand the structure-performance relationship to tackle these challenges and advance commercialization of bio-based NIPU.

Bottom-up processing for bioplastics: Microbial synthesis and chemical polymerization

The bottom-up procedures of bioplastics manufacturing include biotechnological conversion of renewable resources and polymerization processes based on monomers. A typical example is industrial production of PLA. Figure 6 shows the lignocellulosic substrates for lactic acid production. The primary feedstock for polymerization, lactic acid, can be prepared from batch fermentation of polysaccharide-based natural products with lactic acid bacteria, which is usually carried out for 3–5 days at 43°C with a pH of 6.0–7.0 for *Lactobacillus bulgaricus*.[69] In contrast to the chemical synthesis approach, bacterial fermentation leads to high production capacity and low manufacturing cost.

Production of PLA can be performed by polymerization through lactide formation or direct condensation polymerization of lactic acid.[71] In the former case, lactide is first prepared from an intramolecular esterification process of lactic acid oligomers. Then the ring-opening polymerization (ROP) of lactide yields PLA with a controlled range of molecular weights (~100,000 g/mol). The properties of PLA depend on its molecular weight and stereochemistry.[72] This procedure has been applied in the industry to produce high-molecular-weight PLA, which is desired for commercial use. However, usage of catalysts for polymerization contaminates the final plastic product, which limits its application in food packaging. Recent developments have focused on selecting nontoxic catalysts[73] and enzyme-mediated processes.[74]

Besides the ROP method, condensation polymerization is another approach adopted for production of PLA, through which lactic acid monomers and/or oligomers react with each other to form longer chains by releasing water molecules. Compared with ROP, direct polycondensation requires fewer manufacturing steps and is believed to be the least expensive. However, because of the difficulty of removing water from the highly viscous reaction mixture, the equilibrium between free acids, water, and polymers in the later stage of polymerization limits formation of longer polymers. Therefore, PLA obtained from direct polycondensation is usually of relatively low molecular weight (<50,000 g/mol). To overcome this drawback, azeotropic condensation polymerization[71,72] and melt-solid polycondensation[75] procedures have been developed to increase the molecular weight of produced PLA (>300,000 g/mol).

In addition to chemical polymerization, production of bioplastics through microbial synthesis is of particular interest in developing the bio-based economy. Such a biosynthetic approach relies on conversion of simple chemicals into complex compounds through living microorganisms. Besides the first-generation bioplastic feedstock, which is carbohydrate-rich crops or plants, the second-generation (e.g., nonfood crops, food loss, waste) and third-generation (e.g., algae, municipal waste) feedstocks can be used as the nutrient source for biosynthesis.[76,77] Biosynthesis usually operates under relatively low energy conditions, providing a highly sustainable production route.[78]

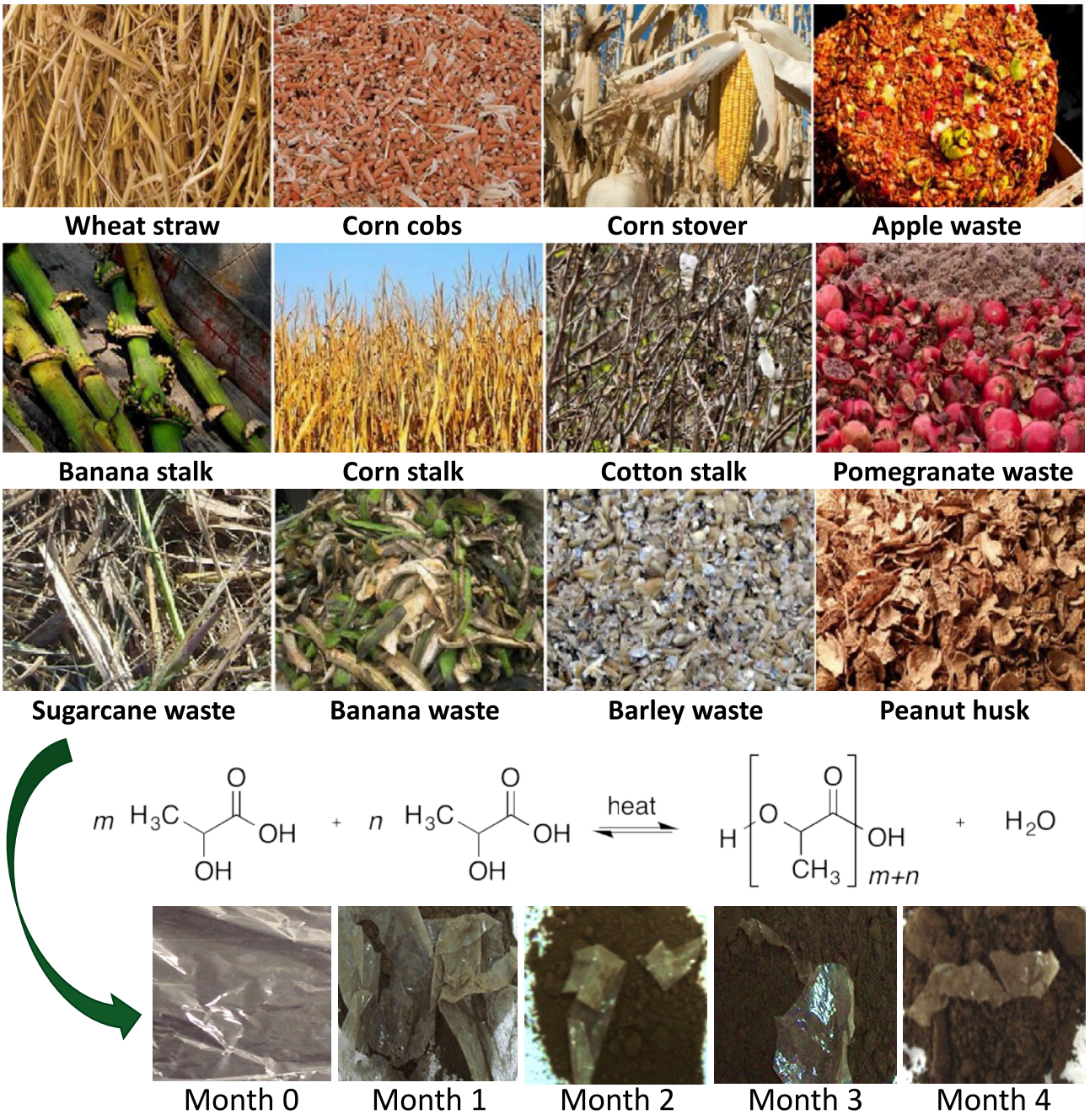

Figure 6. Lignocellulosic substrates for lactic acid production

Commercial production of lactic acid via fermentation depends mainly on the cost of raw materials.[69,70]

Industrial production of PHAs is a representative example of biosynthesis processes.[78,79] PHAs are a family of bacterial polyesters that are nontoxic, biocompatible, and biodegradable. A range of microorganisms can form and accumulate PHAs as storage materials when cultivated under low and unbalanced nutrient conditions. The production procedures of PHAs usually involve microbial fermentation processes and subsequent extraction. The interplay of carbon source, microbes, and pretreatments can lead to a wide variety of PHAs that exhibit diverse physical and mechanical

properties suited for different applications.[79] Currently, PHAs hold only a fairly small market share of bioplastics (~1.7% in 2020)[17] because the high production cost is still a major hurdle for large-scale commercial applications. Along with scaling up the production size of PHAs, improvements through different aspects, such as recombinant microbial strains, mixed cultures, and processes of fermentation, recovery, and purification, help to reduce costs and boost PHA commercialization.[77–79]

BIOPLASTIC MANAGEMENT

Evaluation of plastic biodegradability

Many plastic products are used in a typical human's daily life. As a result, most of the plastic disposed into the environment cause increasing environmental pressure. To address this issue, biodegradation of plastic, which enables assimilation of plastics back into the environment and addresses environmental issues, draws intense research interest and has promising applications.[80] Generally, biodegradation is a process where plastic products are utilized by biological agents.[81] The active ingredients (e.g., gut microbiome) convert (micro)plastic from macromolecules into smaller fragments with several stages. The fragments can be utilized by microorganisms and converted to water, CO_2, or biomass.[80] Figure 7 shows the full progress of a plastic in a biodegradation process. The biodegradability can be evaluated according to the related factors in different routines:

Stage 1: plastic → monomers/oligomers (depolymerization)

Stage 2: monomers/oligomers → biomass (uptake and metabolism)

Stage 3: biomass + O_2 → CO_2 + H_2O (mineralization)

Changes in tested polymers

Plastic samples that have biodegraded have changed, including changes in chemical and physical properties and partial removal of material. Thus, observation of certain sample properties before and after the biodegradation process reveals details of the biodegradation process and is widely used in current studies. For example, weight loss of target samples has been widely used as an indicator of biodegradation because ingestion of plastic by certain microorganisms results in weight loss.[82] The presence of microcolonies of microorganisms and the pits and cracks created during biodegradation, as well as the size change of particle samples, can be observed by microscopy.[83] Accompanied by morphological change, chemical structures might also show some variation during biodegradation. This usually results from biodeterioration as the microorganic grows.[83]

Chemical and physical changes on the microscale lead to changes in the mechanical properties of tested samples.[84] Ingestion of plastic during biodegradation might lead to molecular change of the tested samples.[84] The structure changes of the plastic might be reflected in other properties, such as crystallization, thermal properties, and surface energy. Therefore, the effectiveness of biodegradation in test samples can also be evaluated through X-ray diffraction, differential scanning calorimetry, and thermogravimetric analysis.[85]

All of the aforementioned changes to tested plastic samples indicate variation during degradation. However, characterization of the tested plastic might not reflect the actual biodegradation. Other factors in biodegradation, such as temperature, pH, environmental conditions (water, soil), and components under the preceding circumstances influence the biodegradation process. For example, plastics could be

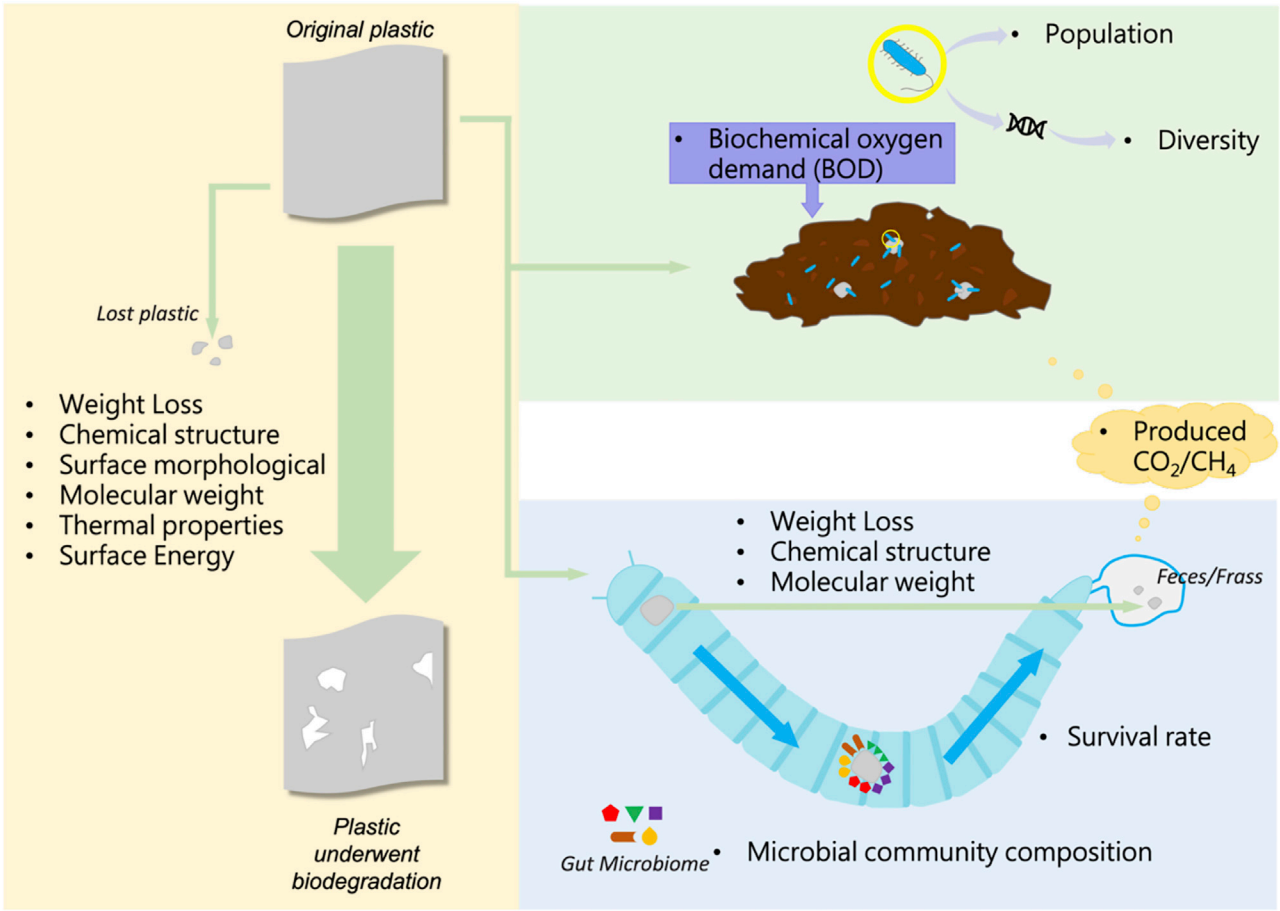

Figure 7. Schematic of the general biodegradation processes of plastics
Yellow area on the left, depolymerization; green area at the top on the right, uptake and metabolism; blue area at the bottom on the right, mineralization.

mechanically and chemically broken down in the near-sea environment.[86] If the weight of the plastic sample was compared in this process, then the weight at time might be underestimated because of the lost microplastic in the collision or aging,[86] indicating an overestimated weight loss and thereby a higher degree of biodegradation. Therefore, it is important to carefully evaluate biodegradation by characterizing the plastic samples solely, and some more direct evidence related to the bioprocess should be involved.[87]

The most direct evidence of biodegradation of plastic is the property changes of plastic that has been digested by and discharged from biological agents, although such plastics are usually hard to harvest in cases of soil, compost, and marine biodegradation. In the case of biodegrading plastic in insects (e.g., mealworm), the residual plastic in the egested frass was recovered and then compared with the original samples in terms of molecular weight and chemical structure. Considering that the difference between plastic consumed and discharged by the insect is the portion that experiences the real biodegradation, the chemical structure change,[88] weight loss,[89] and percentage decrease[88] and molecular weight change[89] of the residual plastic in the feces/frass could be used to estimate the real biodegradation percentage that has been used in some research.

Changes in the metabolic process

The pivotal process of biodegradation is that the biological agents use plastic as a resource in their metabolism. Thus, the activity of the metabolism related to ingestion of plastic provides direct evidence of biodegradation. In the aerobic metabolism of biological agents, the oxygen needed to degrade the plastic (e.g., PHA) is a clue regarding the metabolic activity, which is the indicator for the biodegradation percentage.[90] Correspondingly, CO_2 evolution in the biodegradation process has also been used to evaluate the consumption of carbon content from plastic.[82,91] The same idea could also be applied to anaerobic biodegradation when methane is generated.[92] A biodegradation percentage could also be obtained by dividing the net methane amount by a theoretical methane amount according to the elemental composition.[93]

Microorganisms play an important role in biodegradation. Inoculums, including bacteria and fungi in the soil, compost, or marine environment and the gut microbiome in some insects, are essential participants in biodegradation.[88,89,91,94] The metabolism and reproduction of the microorganism that ingests plastic (e.g., PBS) could reflect the biodegradation process. Therefore, microbial communities with respect to population and family and genus diversity can be used to understand the biodegradation progress in tests. For microbials used in soil, compost, and marine environments, drop plate methods[94] can be used to monitor the relative bacterial change over time. For biodegradation by insects, the community dynamics of the gut microbiome also illustrate the biodegradation process through the survival rate, population, and microbial community composition of the microbials in the insects.[88,89]

Technologies for handling bioplastic waste

Recycling

The majority of plastic waste is burned, buried in landfills, or finds its way into the oceans. Such waste is not recycled because it is usually much cheaper to produce new plastic products than to recycle existing ones. Bioplastics should be recycled at the end of their life to be reused when possible, achieving a circular economy to conserve natural resources. Although bioplastics are plant based, they could be designed to be identical to petroleum-based plastics from a chemical structure and physical property viewpoint, making them difficult to recycle or degrade at end of their life. Despite great efforts in the field of bioplastic recycling, current bioplastic recycling is more of a downward spiral than a closed-loop process, and much work is still needed.

There are generally four ways to recycle bioplastics: (1) closed-loop reuse of the polymer materials (route I, primary), (2) downgrading into less demanding products via mechanical recycling (route II, secondary), (3) chemical recycling into monomeric units (route III, tertiary), and (4) energy recovery via incineration (route IV) (Figure 8A).[44,95,96] Each technology has advantages and disadvantages. For example, primary closed-loop recycling could only be applied to near-pristine waste. Mechanical recycling (e.g., extrusion and injection molding) normally has a relatively low processing cost and is thus the only widely adopted technology for large-scale plastic recycling; however, the quality of the polymer might be downgraded because of the change of polymer chain length, and this method does not address the microplastics waste crisis. These issues make lower downgraded plastic unsuitable for use in areas such as food packaging. Chemical recycling (e.g., hydrolysis, aminolysis, glycolysis, and alcoholysis) offers great opportunities too and recovers initial monomers and/or generates value-added chemical building blocks, but the process is laborious and expensive and thus not suitable for industrial applications. The development of an efficient catalyst that could facilitate selective depolymerization of

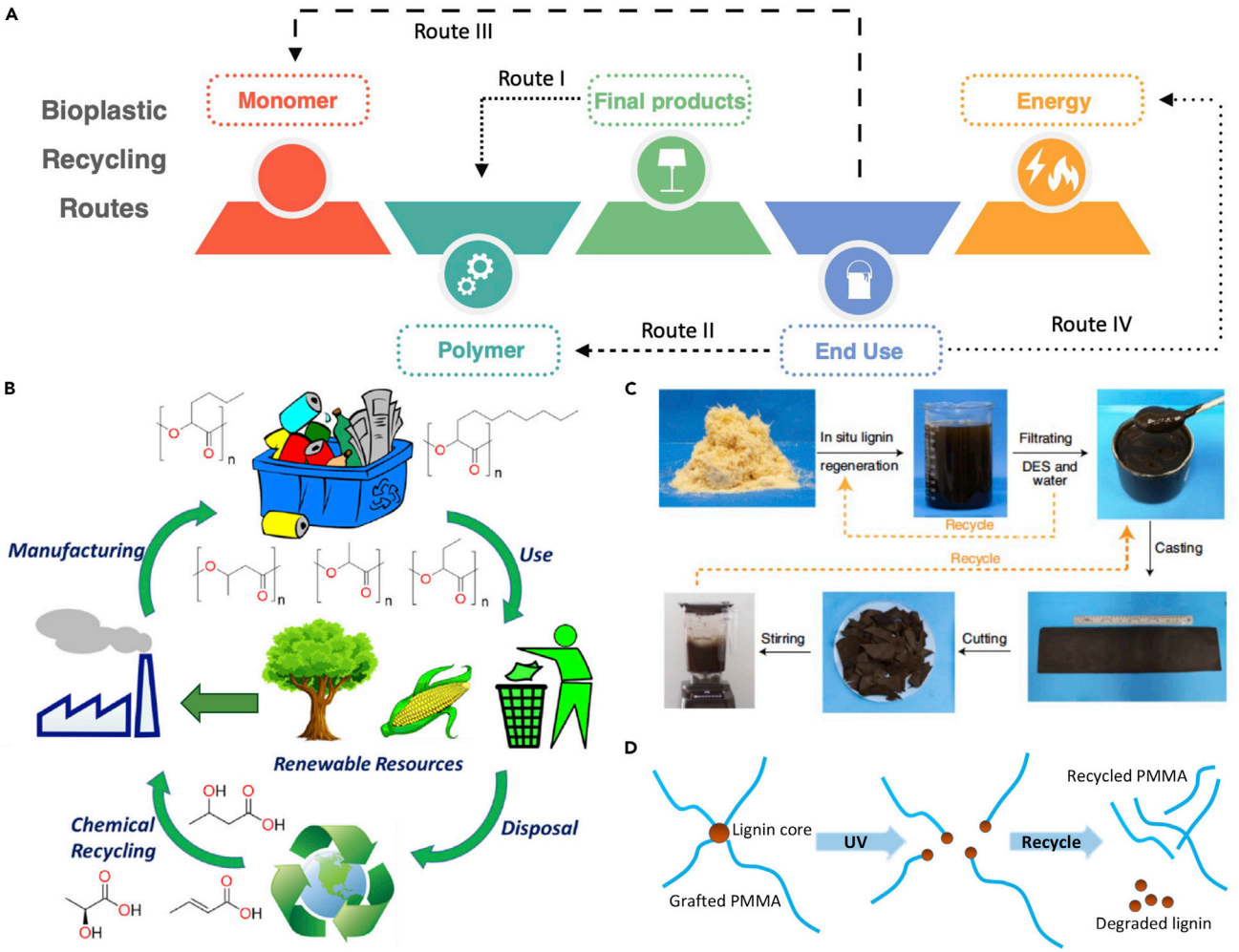

Figure 8. Recycling of bioplastics

(A) Common bioplastic recycling routes.

(B) Chemical recycling of biodegradable plastics can help close the loop toward a lower environmental impact in the LCA of the material.[102]

(C) The recyclability of a high-performance lignocellulosic bioplastic, where "DES" is deep eutectic solvent.[1]

(D) Lignin-enabled phototunable recycling of plastics. A polymeric film with superior mechanical performance is developed, where photo-degradable lignin serves as the core to be cleaved by ZnO nanocatalysts and enable recycling of polymethylmethacrylate (PMMA) after ultraviolet irradiation.[103]

plastic to its monomers while preserving its key functional groups in the generated monomers is essential.[97] For a detailed discussion of the technologies available for chemical recycling and upcycling of commercial plastics, the reader is referred to several recent specialized reviews.[98–100] Last, plastic identification/separation and decontamination technologies for rapid sorting and cleaning to access high-quality plastic materials are also needed for efficient recycling and have been summarized in a recent review.[101] The capability to recycle bio-based plastics varies, and appropriate technologies should be selected based on the nature of biopolymers. The optimal recycling route for a particular bioplastic should start with the possibility of reuse, followed by mechanical recycling until its physical properties deteriorate, and finally by chemical recycling to recover the virgin monomers or chemical upgrading to produce value-added products.

PE furanoate (PEF), a rising star among well-developed bioplastics, has started to replace traditional synthetic resource-based PET in the beverage packaging industry

in recent years. PEF is produced from the polymerization reaction between the renewable carbohydrate-derived 2,5-furandicarboxylic acid (FDCA) and ethylene glycol (EG).[104] Although PEF is not biodegradable or compostable, it could be recycled to CO_2 by incineration, which eventually takes up by lignocellulosics. The opportunity to chemically recycle PEF has not been ignored in the literature, but only a few routes have been put forward. PEF could be enzymatically recycled into its monomer FDCA at a much faster rate than conventional PET.[105] Many plastic bottles and packaging films are difficult to recycle because multiple polymers, such as PU and nylon, are used in conjunction with PET. If a monolayer PEF film could be used alone or along with traditional PET, plastic bottles could be recycled because PEF is chemically similar to PET, providing good compatibility with PET recycling technology.[106]

PHA is another intriguing bioplastic for convenience food packaging.[107] Development of its own recycle stream is still in its infancy and is currently dominated by mechanical recycling. A significant reduction in the tensile strength of regenerated PHA-based materials has been reported.[108] The most studied chemical approach for PHA recycling is thermal degradation via pyrolysis.[109] Hydrolysis and methanolysis of PHB to propylene[102] and methyl 3-hydroxybutyrate have also been reported.[110] Figure 8B shows how chemical recycling of biodegradable plastics can help close the loop toward lower environmental impact. For a more elaborate discussion of the state of the art of chemical recycling strategies for PHA-based bioplastics, including pyrolysis, hydrocracking, hydrolysis, and gasification, the reader is referred to two recent dedicated reviews.[111,109]

Besides developing efficient technologies to recycle bioplastics, strategies for designing bio-based polymers with improved recyclability are also well documented. Chen et al.[112] and Sangroniz et al.[113] synthesized some novel polymers that demonstrated complete and quantitative chemical recyclability by mild-temperature (~120°C) chemolysis or high-temperature (~300°C) thermolysis, representing a closed-loop economy approach to producing plastic packaging bioplastics. Lignocellulosics and lignin-containing recyclable bioplastics have received considerable attention in recent years.[103] Xia et al.[1] manufactured a plastic using a lignin-nanocellulose fibril matrix. The bioplastic film could be easily obtained and demonstrated excellent biodegradability and recyclability by simple mechanical disintegration (Figure 8C). Lignin, the most abundant aromatic on earth, has also been used as a renewable feedstock to produce recyclable bioplastic that leverages its photodegradability, biocompatibility, and biodegradability (Figure 8D).[114,115,116]

Industrial composting

Composting, a subset of biodegradation, is a promising route to produce a humus-like product (e.g., fertilizer, heat, water, and CO_2) from biodegradable plastics in an aerobic environment.[117] A compostable plastic is typically considered biodegradable; however, there is no guarantee that a biodegradable plastic is always compostable. Compostable plastics need to meet specific criteria in terms of biodegradability, ecotoxicity, time, environmental conditions, and quality of the compost being produced. Although nowadays bioplastics are designed to be compostable, in most cases, their compostability requires a strictly controlled conditions (~60°C, 50%–55% solids) that can be offered only by industrial composting facilities.[118]

Natural polymers such as cellulose/hemicellulose and starch can be easily degraded via hydrolysis. Bio-polyesters with hydrolytically labile ester linkages are also susceptible to abiotic hydrolysis; thus, they are largely considered compostable

bioplastics. Some common compostable polyesters include PLA, PHA, PCL, and PBS. Blending or copolymerization of these kinds of polymers could also lead to new materials with reasonable compostability. A polymer's physicochemical and thermal properties, such as molecular weight, surface roughness, hydrophilicity, and crystallinity, are all believed to affect the compostability of the final plastic product.[119] For example, molecular interaction (hydrophilicity, electrostatic, and van der Waals forces) between microbes and the plastic surface could significantly affect the adhesion force and growth of microorganisms; thus, bioplastics with a rough surface and high hydrophilicity favor composting.[120] In the packaging industry, antimicrobial agents (e.g., chitosan and lignin) are frequently added to bioplastic and have been reported to significantly affect compostability.[119] How macromolecular design, blending, and additives can be used to modify the compostability of synthetic biopolyesters as well as the effect of microbial diversity, type of soil, and nutrients on compostability are summarized in a critical review.[119]

The conditions for industrial and home composting are very different. Degradation of amorphous PLA in home composting (approximately 20°C) is estimated to take more than 100 years.[121] However, for industrial composting, the environment typically has a high temperature (approximately 60°C–70°C), high oxygen content (≥6%), high humidity (approximately 60%), and controlled pH (e.g., 8.5). PLA, for example, can be degraded in an industrial composting environment in 45–60 days by microorganisms in the compost.[121] Appropriate selection of the most convenient method for bioplastic waste management depends on the nature and properties of plastic and its applications. Composting offers unique advantages in applications such as packaging polymeric materials for food waste collection, where separation of an individual type of plastic and other organic matter is limited or not possible. On the other hand, plastic recycling or upcycling has the potential to produce valuable monomers and value-added new materials and expand the plastic life cycle.[122]

Incineration and biodegradation

Incineration is one of the traditional methods of plastic waste management. Compared with other approaches, incineration has a significant advantage in reducing plastic waste through a rapid reaction.[123] Heat energy can be recovered for electricity generation.[124] However, a known disadvantage of incineration is release of large amounts of CO_2, which contributes to global warming.[11] Carbon monoxide, particulate matter with an aerodynamic diameter of 2.5 μm or less (PM 2.5), persistent organic pollutants, and other harmful byproducts could be generated during the incineration process.[123,125] However, bioplastics can be degraded by microorganisms. Biodegradation provides more green options for handling bioplastic wastes. This also highlights the importance of developing sustainable bioplastics from renewable resources to replace traditional plastics, helping to create a convenient and sustainable human lifestyle.

Regarding biodegradation, degradation of cellophane-based film is adversely affected by the degree of substitution.[126] Leppänen et al. compared cellophane with 13 other cellulose-based materials, including carboxymethyl cellulose, methyl cellulose, and cellulose acetate, and found that, although cellophane is among the highest-tensile-strength polymers, it is also readily degradable. They also showed that pilot-scale composting of cellophane is surprisingly slow after 2 weeks compared with enzymatic hydrolysis after 2 days. The chain length of the substituent also affects cellulose degradation because it could also hinder binding of cellulase enzymes.[126]

Coatings have been applied to the surface of cellophane to enhance its barrier properties for the food and pharmaceutical industries. The coating layers, however, could also lower its biodegradability. Benyathiar et al.[127] have shown that uncoated cellophane mineralized more (71%) compared with nitrocellulose-coated cellophane (55%) after 141 days in a simulated aerobic composting environment. Irradiation with gamma and electron beams was used for sterilization and could also reduce the biodegradability of cellophane.[127]

A significant advantage of sustainable bioplastics is that they can be biodegraded in an environmentally friendly manner. Various kinds of microorganisms (e.g., *Acidovorax facilis*) have been reported to have the capacity to degrade bioplastics.[128] With a polymer structure, bioplastics are generally unable to permeate the cell membrane to enter the microbial cells. In turn, depolymerases (e.g., cutinase, lipase) are secreted by the microbes to disrupt the interunit linkages (e.g., ester bonds) of bioplastics and release oligomers, dimers, and monomers (Figure 9A). Then these compounds are taken in by the microbial cells and used as carbon sources for microbial metabolism.[10] Figure 9B shows the closed loop demonstrating the biodegradability and circularity of cellulose.

There are various biodegradable plastics in terms of chemical composition and physical properties. Identifying and obtaining efficient bioplastic-digesting microorganisms is essential to achieve decomposition of bioplastics. Because bioplastics are generally not soluble in aqueous media, traditional screening methods are time consuming. Shin et al.[129] developed an efficient microspray-based system for screening bioplastic-degrading microorganisms. By spraying bioplastic microparticles on a plate, a large interface between bioplastic substrates with microorganisms is realized. Employing this facile method, the researchers were able to effectively isolate bioplastic-digesting microorganisms. This simple and straightforward microspray-based screening system is a powerful tool for searching for and selecting suitable microorganisms to efficiently degrade bioplastics. Designing engineered microbial strains by applying systems and modifying key enzymes are emerging approaches to upgrading bioplastic degradation.[130]

Along with biotic factors, the accessibility of bioplastics significantly influences biodegradation efficiency. Only by sufficient contact between the bioplastic substrates and microbial cells and/or enzymes can efficient degradation be realized. Gamerith et al.[131] employed a kind of cutinase to hydrolyze PET. They found that the degradation rate increased by 180% when the particle size of PET powders was decreased from 0.25 to 0.05 mm.[131] Therefore, to improve bioplastic biodegradation efficiency, even a pretreatment operation to reduce the bioplastic's molecular weight is an effective method. With development of advanced techniques such as structural biology, systems and synthetic biology, and molecular dynamics, the biodegradation mechanisms of bioplastics are becoming more and more clear, guiding efforts toward more efficient and sustainable biodegradation of bioplastics.

SUSTAINABILITY OF BIOPLASTICS

Bioplastics derived from renewable resources are receiving more and more attention from industry because petro-plastics threaten the environment, and the reserves of fossil fuels are limited.[132–134] Among them, PLA, TPS, and PHB are of most interested because they have properties similar to synthetic polymers (PHBs) or can be directly composted or landfilled for disposal (TPS).[133] Sustainability is

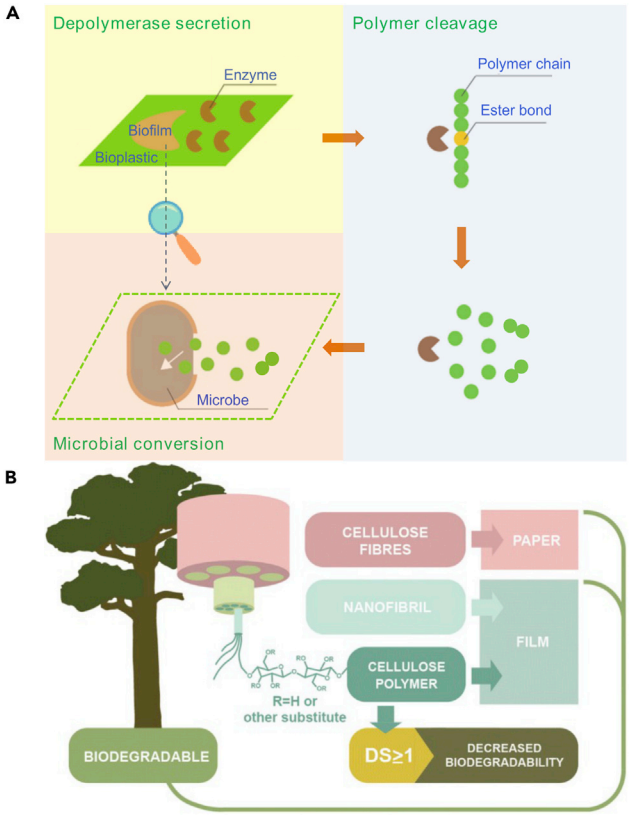

Figure 9. Biodegradation of bioplastics

(A) Mechanisms of the biodegradation process of bioplastics.[10]

(B) Schematic of the hierarchical structure of cellulose from wood fibers and potential applications as paper and films. Chemical grafting of cellulose increases the degree of substitution, leading to decreased biodegradability. The closed loop demonstrates the circularity of cellulose. The "DS" is degree of substitution.[126]

usually regarded as the significant advantage of bioplastics over petro-plastics and can be evaluated in terms of life cycle assessment (LCA), life cycle costing (LCC), and social LCA (S-LCA).[135,136]

The environmental impact of bioplastics over the entire value chain can be quantified by LCA. The most notable indicator of LCA is GHG emissions. Bioplastics do not inherently behave better than petro-plastics regarding GHG emissions, and a detailed LCA with unbiased impact categories, adequate input data, and consequential modeling should be conducted for individual cases.[137–139] Yano et al.[140] quantified the reduction in life cycle GHG emissions when replacing fossil-derived plastics with degradable, biomass-derived plastics (100% PLA) for household use. A reduction in life cycle GHG emissions of 14%–20% was achieved by replacing fossil-derived plastics with bioplastics. The researchers also observed that adapting effective source separation and recycling could promote a reduction in GHG emissions contributed from waste plastics. A reduction of 31.9% in GHG emissions could be achieved when increasing the separate collection rate from 39% to 100% and replacing fossil-derived plastics with biomass-based materials.

Many researchers use a cradle-to-gate method to investigate the GHG emissions of bioplastics. However, the disposal phase should be taken into account. Figure 10A

shows the life cycle of bioplastic produced from maize, where land competition with increased CO_2 emission occurs because of indirect land use changes. Benavides et al.[141] studied the environmental effects of PLA, bio-PE plastics, and fossil-derived plastics such as HDPE and LDPE. Bio-derived plastics exhibited the lowest GHG emissions (−1.0 and 1.7 kg CO_2e/kg for bio-PE and PLA with no biodegradation, respectively, where CO_2e is carbon dioxide equivalent) and fossil energy consumption (29 and 46 MJ/kg of bio-PE and PLA, respectively). However, taking landfill and composting emissions into account could result in an increase in the life cycle emissions of PLA from 16% to 163% depending on the degradation condition. Bohlmann[142] conducted an LCA for PLA derived from corn and PP derived from natural gas. The LCA revealed that PLA was more energy efficient than PP for food packaging, whereas the difference in GHG emissions for the two polymers was almost negligible when assuming that the carbon in PLA was fully sequestered in a landfill condition. Another example shows that the disposal phase can generate GHG, and this phase should be taken into account in an LCA.

Although PLA is the first commercially available bio-based material, its barrier properties are much worse than those of petroleum-derived plastics.[145,144] Thus, improving the tensile strength and barrier properties by introduction of fillers is necessary to broaden application of PLA. Incorporation of functional fillers can not only reduce the cost but also change the properties (e.g., density, thermal expansion) by generating multiphase systems with micro/macrostructures.[143] Guo et al.[146] studied the effect of functional fillers of PLA-based plastics on the environment. The results indicated that incorporating fillers (e.g., calcium carbonate) into PLA bottles could reduce the amount of PLA particles, and a sensitivity analysis showed that using 5% less PLA particles could result in a decrease of 3.4% in the overall environmental impact of PLA/$CaCO_3$ bottles (Figure 10B).

Compared with PLA, bio-PET has better barrier properties, although it is nonbiodegradable. However, the environmental impact of bio-PET is uncertain. Tsiropoulos et al.[147] compared cradle-to-gate environmental profiles of fully bio-based HDPE and partially bio-based PET with the fossil fuel-derived counterparts.[147] Approximately 70% of partially bio-based PET's weight was petrochemical PTA. GHG emissions from bio-based HDPE were 140% lower than those of petrochemical PE, and the reduction in consumption of nonrenewable energy was approximately 65%. For partial bio-PET, GHG emissions were similar to its petrochemical counterparts, but consumption of nonrenewable energy was lower by up to 10%. Chen et al.[148] designed a novel process to convert lignocellulosic biomass from forest residues to bio-PET bottles.[148] They compared the cradle-to-gate impacts of petroleum-based PET bottles, partially bio-based PET bottles, and 100% bio-based PET bottles. Their results showed that PET bottles from woody biomass had 21% less global warming potential and consume 22% less fossil fuel than petroleum-based PET bottles.

Other processes and resources have been reported for producing bioplastics.[149] Kim et al.[150] designed an integrated process with several energy-intensive units to produce FDCA as a plastic monomer from lignocellulosic biomass. The integrated process showed less (53%) fossil depletion but more (29%) climate change than production of petroleum-derived terephthalic acid (TPA). They also identified that most of the climate change and fossil depletion in the integrated process came from generation of electricity. Applying renewable feedstocks (e.g., biogas, woodchips) and renewable technologies (e.g., hydropower, wind power) to electricity generation could make the integrated process more environmentally friendly.

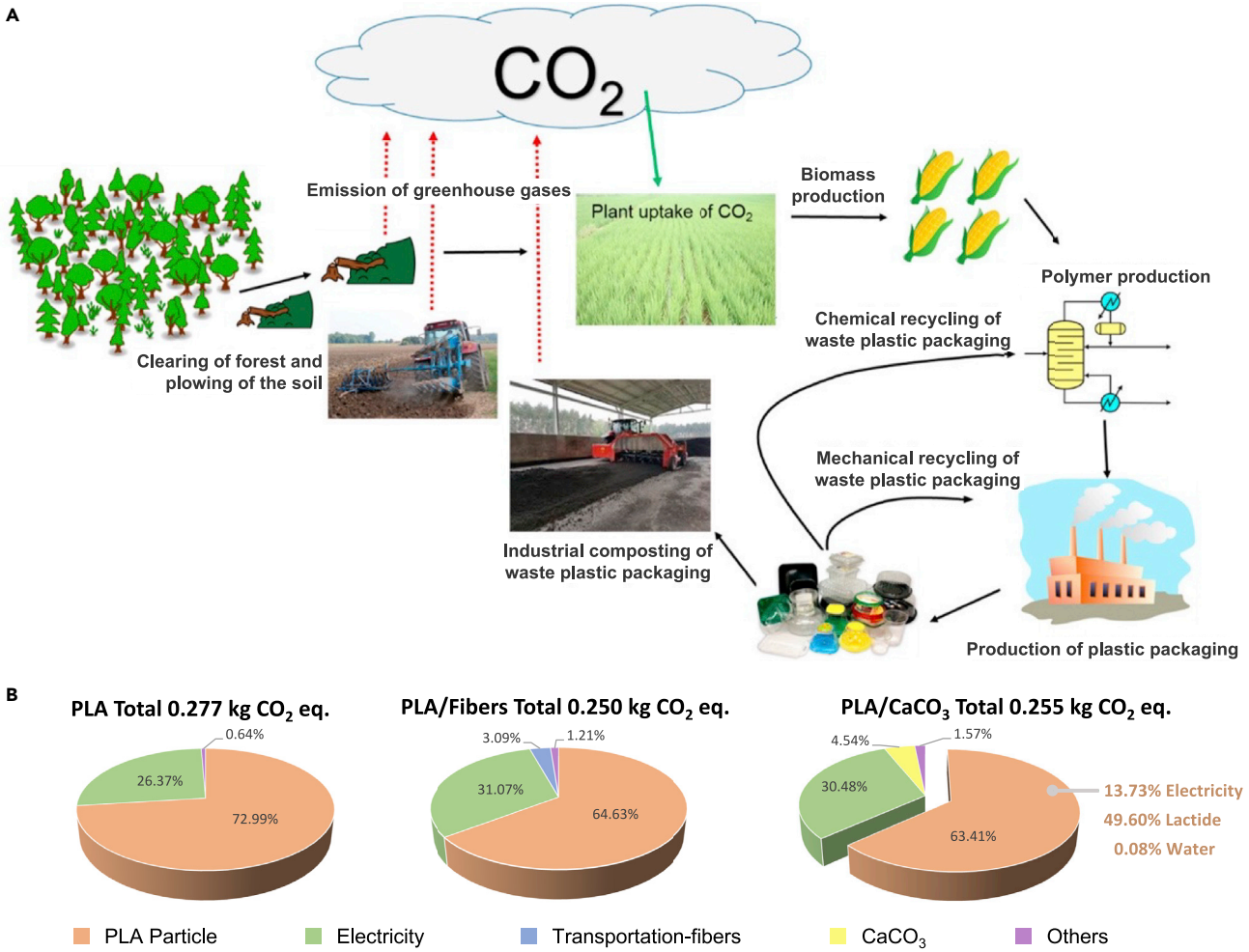

Figure 10. Environmental impacts of bioplastic production

(A) Life cycle of bioplastic produced from maize. When first-generation biomass is used for packaging production, land competition with increased CO_2 emission occurs from indirect land use changes. This induces climate changes and environmental impacts from fertilizers, pesticides, and so on.[143]

(B) Global warming potential of three kinds of PLA-based bottles.[144]

Apart from GHG emissions, other environmental indicators, such as land use, eutrophication, and acidification, should be evaluated in detail for bioplastics.[133,151] Even application of bioplastics can result in a reduction in GHG emissions, and the total environmental impact can be worse than with petro-plastics in some cases when considering these environmental indicators.[133,152] In a study by Chen et al.,[148] woody biomass-based PET bottles were worse regarding environmental indicators such as ecotoxicity and ozone depletion impacts. Tsiropoulos et al.[147] found that the impact of bio-HDPE and bio-PET on human health and ecosystem quality was higher by factors or even orders of magnitude compared with petro-HDPE and petro-PET.

Eerhart et al.[153] studied the energy and GHG balance when producing bio-PEF from corn-derived fructose. The cradle-to-grave analysis showed that a reduction in nonrenewable energy use (40%–50%) and GHG emissions (45%–55%) can be achieved through this PEF process compared with production of petro-PET. The analysis also revealed that the reductions from the PEF process were higher than

the reductions obtained from producing other biobased plastics, such as bio-PLA or bio-PE. Genovesi et al.[154] studied the LCA of disposable and reusable tableware. When considering a landfill scenario as end of life, multiuse bioplastics had a lower impact than fossil-based plastics in the categories of human toxicity water and ecotoxicity water chronic. The impact of the bioplastic reuse system depended on the number of reuses and the washing method. Washing could be improved by adopting certified power. The bioplastic reuse system was suitable in the baseline scenario because the washing method was efficient, and the number of reuses was high. At present, it is still not clear which type of bioplastic outperforms the rest because of the scarcity of comprehensive studies. For future research, the sustainability assessment should be analyzed for the entire life cycle, with unbiased impact categories and reasonable system boundaries. Recycling and reusing should be promoted because they can significantly improve sustainability.[155,151]

Techno-economic analysis of bioplastics

Techno-economic analysis (TEA) has been used to discuss the effects of possible technologies on converting bio-feedstocks to the target plastic products and identify the correlation between various technologies and major economic indicators (e.g., minimal selling price and fixed capital investment).[156,157] A few processes for bioplastic production have been analyzed by TEA and compared with petrochemical plastics, as discussed below.

Because production of bioplastics involves a series of sequential unit operations (e.g., mixing, separation, drying, and reaction), a prerequisite for their production is quantifying their individual contribution to the overall cost and identifying the significant ones. Kim et al.[150] designed a process for producing FDCA from lignocellulosic biomass to replace petroleum-derived TPA. Using a discounted cash flow methodology, the minimum selling price of FDCA was determined to be $1,024 United States dollar per ton (USD/T) for the proposed process and $1,380 USD/T for the alternative process (production of furfural from C5 sugar). Both costs were lower than the price of petroleum-derived TPA ($1,445 USD/T). The cost contributions for the proposed process revealed that the feedstock cost (26.2% of the total cost), 5-hydroxymethylfurfural production (23.5%), heat and power generation (18.2%), and biomass fractionation (13.3%) were the major cost contributors. Sensitivity analysis indicated that the minimum selling price of FDCA was most sensitive to the total capital investment, discount rate, and feedstock costs.

To deviate from the dependence on food crops for bioplastics production, more and more research focuses on converting waste to bioplastics.[158] However, the properties of different types of waste vary, which might increase the operational cost. Bassi et al.[149] quantified the budget costs, externalities, and final societal LCC of producing PHA from municipal food waste and wastewater sewage sludge in five geographical regions. Budget costs were lower than PU and PHA derived from first-generation biomass, but the costs were less clear compared with LDPE. Source-separated food waste collection in the biorefinery (76% of the budget cost of the biorefinery plant) was identified as the most relevant contributor to budget costs.

Rajendran and Han[159] conducted a TEA of producing PLA and biodiesel from food waste (Figure 11A). Table 2 shows the economical parameters of PLA and biodiesel production from 50 tons of food waste. The process consisted of milling and drying, oil extraction, biodiesel production, lactic acid production, lactide production, and PLA production. Geological and societal characteristics could influence the

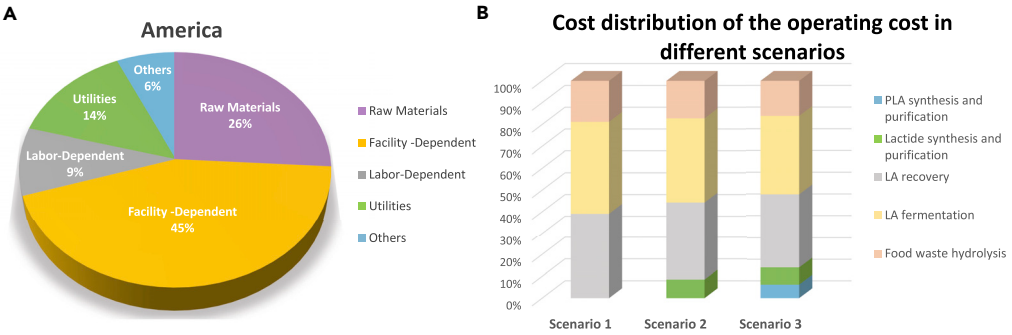

Figure 11. TEA of bioplastic production

(A) Annual operating cost breakdown of PLA and biodiesel production in the United States.[159]

(B) Distribution of operating costs for three different scenarios with different products: (1) lactic acid, (2) lactide, and (3) PLA.[160]

properties of the feedstocks and products. The minimal selling price of PLA from this process was $4.2–$5.3 USD/kg, which was much more expensive than the price of PP ($1.8 USD/kg) and HDPE ($1.5 USD/kg) because of too many unit operations and high consumption of solvents. The byproducts (biodiesel), as value-added products, could help reduce the price of PLA in this process, depending on the selling price of biodiesel.

Kwan et al.[160] designed a plant with a 20-year lifetime for producing lactic acid, lactide, and PLA from food waste powder at a rate of 10 MT/h (Figure 11B). For every 10 MT of food waste powder, 3.1 MT of 80% lactic acid, 1.7 MT of lactide, or 1.3 MT of PLA could be obtained from the plant. The minimum selling prices of lactic acid, lactide, and PLA made this process economically feasible. Of the three processes, production of 80% lactic acid showed the best economic performance in terms of annual net profits ($22,184,169), net present value, internal rate of return (31.1%), and payback period (5.1 years), assuming a discount rate of 5%. Sensitivity analysis revealed that the profitability of the plant was determined by the prices of lactic acid, lactide, PLA and byproducts as animal feed. Relevant TEA studies of bioplastics production from various feedstocks are still scarce. Considering the tremendous consumption of petro-plastics in the world, significant improvements in process efficiency (energy and materials) and technology are necessary to make bioplastics competitive in price. Process design should be considered to take advantage of valorization of value-added byproducts.

Societal impacts of bioplastics

The market share of bioplastics is increasing in the plastics industry. Compared with production of petro-plastics, growth and processing of biomass involve using chemicals such as pesticides, fertilizers, and organic solvents,[161] whose effects are manyfold. First, use of chemicals increases the environmental impact of bioplastics. Tsiropoulos et al.[147] found that the impacts of bio-HDPE and bio-PET from sugarcane ethanol on human health and ecosystem quality were up to 50 times and 2 orders of magnitude higher than for petro-HDPE, respectively. The major causes were pesticide use, land occupation, and preharvest burning practices. Improvements to these causes could significantly reduce the impact of the bio-based polymers. Second, chemical use can result in safety issues in food packaging. For packaging materials, especially those contacting food, the tendency of substances migrating from the packaging into food should be lower than the specific migration limits established by regulators.[162]

Table 2. Economic parameters of PLA and biodiesel production from 50 tons of food waste[159]

Parameters	China	India	Brazil	United States
Total capital investment	41,891,000	45,065,000	48,217,000	47,320,000
Operating cost ($/year)	12,252,000	13,651,000	15,027,000	12,782,000
Main revenue	9,807,000	12,994,000	15,489,000	14,669,000
Unit production cost	6.51	5.48	5.06	4.54
Return on investment	8.35	12.42	15.12	17.11
Payback time (years)	11.98	8.05	6.62	5.84
Internal rate of return (%)	4.14	10.55	15.55	18.36
Net present value	−12,532,000	1,456,000	18,271,000	26,902,000

Because nanoparticles might be added to reinforce the properties of bioplastics, their effect on human health should be studied. Lian et al.[163] studied the impact of nano-TiO_2 on properties of polyvinyl alcohol-chitosan films and the migration behavior of TiO_2 nanoparticles from films to food simulants. Improvements in water vapor, the gas barrier, and mechanical and antimicrobial properties were obtained through incorporation of TiO_2 nanoparticles. Distilled water, acetic acid, ethanol, and olive oil were used as food simulants to determine the migration behavior of TiO_2 particles into food. Only olive oil could induce migration of a trace amount of TiO_2, and migration of TiO_2 nanoparticles could be significantly reduced by high hydrostatic pressure treatment at 200–400 MPa.[163,164] Unintentionally added substances might exist in bioplastics during the manufacturing process, and they could be released during the life cycle of the bioplastic.[165] Therefore, more relevant research is needed.

The common feedstocks used for bioplastics are from agricultural crops such as corn or sugarcane (first-generation feedstock), lignocellulosic biomass and wood (second-generation feedstock, such as straw and corn stover), and engineered crops, algae, and urban residues (third-generation feedstock).[166–168] Only the first-generation feedstock can currently provide materials for production of bioplastics at the industrial level. This could inevitably lead to the dilemma of food versus bioplastics when scaling up bioplastics production.[169] These agricultural crops are known to require large amounts of water. The water crisis might be aggregated by the bioplastics industry, especially in areas with limited water resources. When converting a natural landscape to production of feedstocks for bioplastics, adverse environmental impacts (e.g., soil erosion, eutrophication, biodiversity loss, and carbon emissions) can emerge.[167] In these scenarios, bioplastics from waste are more appealing. Depending on the feedstocks, the popularity of bioplastics can vary among different groups of people. For example, vegan consumers will not accept bioplastics with slaughterhouse waste as feedstock for packaging applications.[169] Thus, certifications from neutral organizations are important for the consumers and suppliers of bioplastics.

Similar to production of petro-plastics, biorefineries and polymerization processes are highly energy consuming because of the existence of many energy-demanding processes (e.g., separation, heating). Although production of bioplastics is considered less hazardous than production of petro-plastics, various hazards still exist in the process, such as biological agents, pesticides, solvents, and volatile organic compounds.[170] These hazards result in increased emission of carbon and a detrimental effect on the safety of workers. A detailed analysis of the effect of bioplastic production on the health and safety of workers can be found in Álvarez-Chávez et al.[161] As a capital- and energy-intensive process, production of bioplastics from biorefineries might contribute to growth in the job market. The structural path analysis performed by Kikuchi et al.[157] clarified that producing biopolymers from

domestic biomass resources might have positive effects on local economies (e.g., increasing added value or employment).

Currently, the manufacturing costs and sale prices of bioplastics are relatively higher than for petro-plastics, and a study revealed that consumers and plastic processors were not willing to pay more for bioplastics.[169] Many consumers are skeptical about the sustainability of bioplastics. It is expected that the price of bioplastics can be driven down by technology innovations and that the acceptability of bioplastics can grow through decreased prices and more well-informed consumers. One focus of technology development can be in the direction of developing durable bioplastics for multiple reuses.

CONCLUSION AND PERSPECTIVES

Bioplastic is a renewable and cost-effective polymer. Bioplastic typically exhibits properties comparable with fossil-based plastic, and it can be an alternative to fossil-based plastic in most applications of food packaging, with a smaller carbon footprint and less environmental impact. This article systematically reviews bioplastic fabrication and management of bioplastic waste regarding degradation and recycling. The main challenges of bioplastic applications include low fracture strain, inferior barrier properties, and water vapor resistance. Adding plasticizers is an effective approach to improving the tensile strength and fracture strain of bioplastics. PLA is one of the most promising bioplastics because of its superior mechanical performance and easy processability.

Despite great efforts having been made in bioplastic recycling, how bioplastic recycling is performed is more of a downward spiral than a closed-loop process. The capability of bioplastics to be recycled varies, and appropriate technologies should be selected based on the nature of biopolymers. An optimal recycling route for a particular bioplastic can start with the possibility of reuse, followed by mechanical recycling until its physical properties decrease, and finally chemically recycling to recover the virgin monomers or chemical upgrading/upcycling to produce value-added products. Degradation by microorganisms also provides a green option for handling bioplastic wastes.

With regard to the sustainability of bioplastics (e.g., environmental profile, TEA, and societal impact), recycling should be promoted because it can significantly improve sustainability. For example, PET bottles derived from woody biomass have 21% less global warming potential and consume 22% less fossil fuel than petroleum-based PET bottles. Future work needs to be performed to investigate bioplastics with higher mechanical and barrier properties. Bioplastic recycling based on a closed-loop process also requires more studies. The mechanism behind biodegradation of bioplastics needs to be thoroughly investigated. Sustainability assessments should be analyzed for the entire life cycle with unbiased impact categories and reasonable system boundaries.

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AUTHOR CONTRIBUTIONS

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DECLARATION OF INTERESTS

The authors declare no competing interests.

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