Sustainable polymers

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Abstract | Sustainable polymers are materials derived from renewable, recycled and waste carbon resources and their combinations, which at the end of life can be recycled, biodegraded or composted. Sustainable polymers also exhibit reduced environmental impact throughout their life cycle. This Primer presents an overview of the research in and potential of sustainable polymers, with a focus on their life cycle, synthetic routes from renewable carbon feedstocks, production, material characterization, applications, end of life, data reproducibility and limitations faced, and provides a brief outlook. The Primer also briefly covers other carbon feedstocks such as carbon dioxide and wastes, including agricultural and woody residues. Although still in their infancy, new sustainable polymers are already finding applications in packaging, automotive parts and 3D printing. This Primer also discusses the headwinds facing the adoption of sustainable polymers, including complexities of recycling and composting, manufacturing scale-up, data reproducibility, deposition and potential solutions. Development of sustainable polymers will accelerate the age of sustainable polymers and create a truly circular economy for plastics by reducing production and use of virgin plastics from finite resources.

Sustainable
Meeting the needs of the present without compromising the ability of future generations to meet their own needs.

Circular economy
A closed-loop economic system that targets zero waste and pollution throughout material life cycles.

Chemical recycling
Relates to technologies that convert polymeric waste back to monomers, oligomers or other functional chemicals that can be used as raw materials for manufacturing new plastic articles.

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Sustainable polymers are materials derived from renewable, recycled or other lower-carbon feedstocks, which at the end of life (EoL) are managed in environmentally responsible ways through recycling and biodegradation[1–3]. These include natural, modified and synthetic polymers that are sustainable over their life cycle from cradle to cradle in line with a circular economy model[4]. FIGURE 1 presents a schematic of the life cycle of sustainable polymers from feedstock to regeneration. Serious environmental problems caused by the persistence and accumulation of plastic pollution on land and in oceans[5–8] has triggered new research on sustainable polymers and their commercialization, although humankind has a long history of using sustainable polymers (Supplementary Fig. 1).

Major challenges for bringing sustainable polymers to the marketplace include ensuring sustainability and efficiency throughout the entire supply chain; keeping costs and polymer properties competitive and comparable or adding value compared with traditional petroleum-based polymers; and designing materials for environmentally responsible reuse, reprocessing (mechanical recycling)[9], depolymerization (chemical recycling)[10,11] and/or biodegradation[12] at EoL. Research in sustainable polymers should be multidisciplinary, not focusing on one specialized area but being cognizant of the entire life cycle of materials including relevant aspects of polymer science, industrial processing, biology, cost management procurement and environmental science. Nature offers models of sustainable polymer science from which we can learn, using biomimetic principles to maximize the economy and efficiency of chemistry and processes. Using orderly arrangements and combinations of biological (macro) molecules such as amino acids, saccharides, liposomes, ribonucleic acid and others, diverse specialized life functions and life materials can be created. These learnings from nature can inspire and catalyse the development of sustainable polymers[13,14].

Numerous review articles have been published on the development of sustainable polymers and associated challenges[1,2,15–18]. These include investigations of alternative feedstocks such as waste carbon dioxide or various biomass sources for polymer production, potential future applications and the benefits to our society. Exploring new sustainable polymers that can substitute present materials with broader EoL options, enabling easy recycling and/or biodegradation[19,20], is important. This Primer offers a new perspective on sustainable polymers covering the entire life cycle from the broad range of available feedstocks to EoL, and includes experimentation, characterization, applications and sustainability evaluation principles. Our goal is to provide a comprehensive viewpoint for all stakeholders in the field. Monomers and polymers made from natural products and biomass such as lignocelluloses, lipids and

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amino acids via green chemistry are an area of promise and emphasis. Additionally, data deposition and entry requirements for sustainable polymers, which are mostly overlooked, are discussed to guide future rational design of sustainable polymers. This Primer focuses more on sustainable polymers synthesized from (bio)renewable feedstocks, as sustainable bio-based/renewable polymers represent a shift to a bioeconomy[21]. It is an opportunity to use renewable, bio-based carbon, reduce our dependence on fossil fuels and their impact and reduce CO_2 emissions that contribute to climate change[22]. Biomass-based production can have significant impacts on the environment, and so responsible production is key to realizing its true potential.

Experimentation

Development of most sustainable polymers begins with extraction of compounds of interest from biomass feedstocks and ends with processing to products of interest (FIG. 2). Typical steps involve biorefining (producing chemicals from biomass), polymerization, processing (such as post-polymerization modification, injection/blow/compression moulding, 3D printing, self-assembly[23]) and post-processing (reuse, recycling or biodegradation), which can be highly dependent on the specific feedstocks and application. This section discusses the methods used in each step of the development of sustainable polymers.

Drivers for rational design

The drivers for rational design of sustainable polymers come from the accumulated plastic waste crisis and the dependence on non-renewable petroleum resources. Initially, rational design was limited to constructing naturally occurring polymers to improve their performance as nature-inspired alternatives to petroleum-based commodity polymers[24]. However, the preferable rational design of sustainable polymers is built around their synthesis from renewable monomers, and with responsible EoL has been continually (re)oriented for more than two decades. Building sustainable polymers from renewable monomers involves the use of drop-in monomers — which have the same chemical structure as their fossil-based analogues — as well as novel monomers[25,26] for the corresponding production of drop-in and novel bio-based polymers. Examples of these include bio-based polyethylene (bio-PE) and bio-based poly(ethylene terephthalate) (bio-PET) or polylactide (PLA) and poly(ethylene 2,5-furandicarboxylate) (PEF), respectively[27]. Obtained from annually renewable food/agricultural resources with easy extraction, the sourcing of both the drop-in and the novel monomers has rapidly been reoriented towards novel feedstocks such as residues from crop harvesting, algae and used cooking oil. All bio-based polymers cannot be considered as biodegradable[28]. Indeed, whereas some bio-based polymers, such as poly(hydroxyalkanoate)s (PHAs), can degrade in different natural environments upon the action of microbes and/or enzymes[29], others (such as bio-PE) can naturally remain intact[30].

The requirements of the sustainable polymers have also evolved. In the earlier days, they were mainly designed from bio-based materials as commodity plastics with properties comparable with their petroleum-based counterparts. Recent legislations and gathered knowledge have enabled redesign of bio-based plastics to also include EoL scenarios, particularly their recyclability or biodegradability in specific environments with targeted complete bio-assimilation[31]. There are already new concerns at the conception and production stages of sustainable polymers, particularly coming from the land, water and energy consumption during synthesis and extraction of bio-based monomers and bio(-based) polymers[32]. Reducing land resources and occupation seems evident if agricultural co-products are used for extraction of monomers and polymers. Water and energy, however, need a sustainable production approach via good housekeeping practices, process modifications and technology changes. Renewable energy use as well as efficient wastewater/energy recovery all help limit undesired consumption and improve sustainability of the bio-based market. Some of them have already been included in life cycle assessment (LCA) studies during the last decades[33,34].

Sustainable polymers can be viewed within a cycle: plant biomass converts environmental carbon into CO_2 by photosynthesis using water and sunlight; plant biomass is harvested and processed to make monomers or polymers via extraction, fermentation and chemical and microbial conversion; the monomers/polymers are converted to plastic products; and the products are reused or recycled to prolong the product lifetime and to reduce fresh material consumption, or biodegraded to recover and recycle CO_2 through photosynthetic regeneration back to plant-biomass material. LCA is an important tool to identify the potential environmental impacts through the entire life cycle of sustainable polymers. It would be valuable and important to practice life cycle thinking in the design and engineering of new sustainable polymers, especially when the product begins its journey to commercialization, ensuring circularity within adapted economic systems.

Chemistry of monomers and polymers

Natural biopolymers. Renewable feedstocks such as plants and seafood waste are normally composed of cellulose/hemicellulose, starch, lignin, protein, chitin and lipids. These natural biopolymers have a wide variety of structures, properties and functions in nature. When designing for sustainable chemicals or products, cellulose/hemicellulose, starch and chitin — the three

Bioeconomy
An economy where goods are made from responsibly produced biomass.

Life cycle assessment
(LCA). A methodology to assess the environmental impacts associated with each stage of a product's life.

Thermoset
A specific class of polymers that form well-defined, irreversible, chemical crosslinked 3D networks.

Glass transition temperature
(T_g). The temperature at or above which the molecular segments start to move.

major substances obtained — must be first viewed as relevant biopolymers of importance. This follows their extraction, purification and transformation to desired chemicals or products. Cellulose ($(C_6H_{10}O_5)_n$), as the most abundant polysaccharide on Earth, has long been chemically modified into products[35]. Native starches consisting primarily of amylose (20–30%) and amylopectin (70–80%) have been processed to form thermoset resins or thermoplastic starches[36]. Chitin is the second most abundant natural polysaccharide; it is a linear polysaccharide containing nitrogen[37] which has been used directly, or as deacetylated chitosan among many applications[38–40]. These nitrogen-containing compounds have great potential in synthesizing sustainable polymers with advantaged performance, which has not been fully explored. Polypeptides (proteins) composed of amino acid monomers can also be used as sustainable polymers[23,41] via nanoscale/microscale architecture tailoring. Extracted from annually renewable natural resources, natural biopolymers suffer from lack of reproducibility or inherent variability and have so far not been able to perform as conventional plastics without suitable modifications, which has limited their use. The inability of biopolymer-based green plastics to compete with properties of commodity plastics has necessitated a trade-off with biodegradability: modification, blending and additives improve thermo-mechanical properties, but may reduce the biodegradability and compostability[42]. Recent examples of biopolymer-based green plastics using bacterial cellulose[43] and self-assembly of plant protein[23] have shown promise to enhance the properties without reducing the biodegradability.

Monomer extraction. Synthesizing sustainable polymers directly from bio-based monomers is, in many cases, more appealing than natural biopolymers. Bio-based monomers must first be extracted from biomass with transformation and purification for the synthesis of sustainable polymers with properties competitive with established polymers, or those with properties not currently desired in the mainstream. Many bio-based platforms have been successfully developed for extraction of sugar[44,45], lignin[46,47], lipid[48] and protein[49], along with the CO_2 platforms[50] (TABLE 1). The platform for extracting sugar is the oldest, due to the abundance of sugar-containing plants. Historically based on food resources (such as sugar cane and corn), the sugar extraction platform has since been reoriented to waste-based resources (such as wood and grass). Nine sugars are commonly produced and have been transformed to a library of functional monomers consisting of more than 300 monomers, blocks or chemicals[44,51]; 12 of the monomers from this library have been identified as the most promising blocks for synthesizing chemicals and polymers, based on technological and economic feasibility.

Lignin is a heterogeneous polyphenol predominantly containing syringyl, guaiacyl and *p*-hydroxyphenyl units; it is the second most abundant plant-based biopolymer with an approximate annual photosynthetic production of 200×10^8 tons[52,53]. Being one of the few large scale-produced natural aromatic compounds, lignin monomers are suitable precursors to prepare renewable aromatic polymers with useful properties such as high glass transition temperature (T_g)[54–56]. A lack of large-scale implementation of lignin depolymerization strategies to

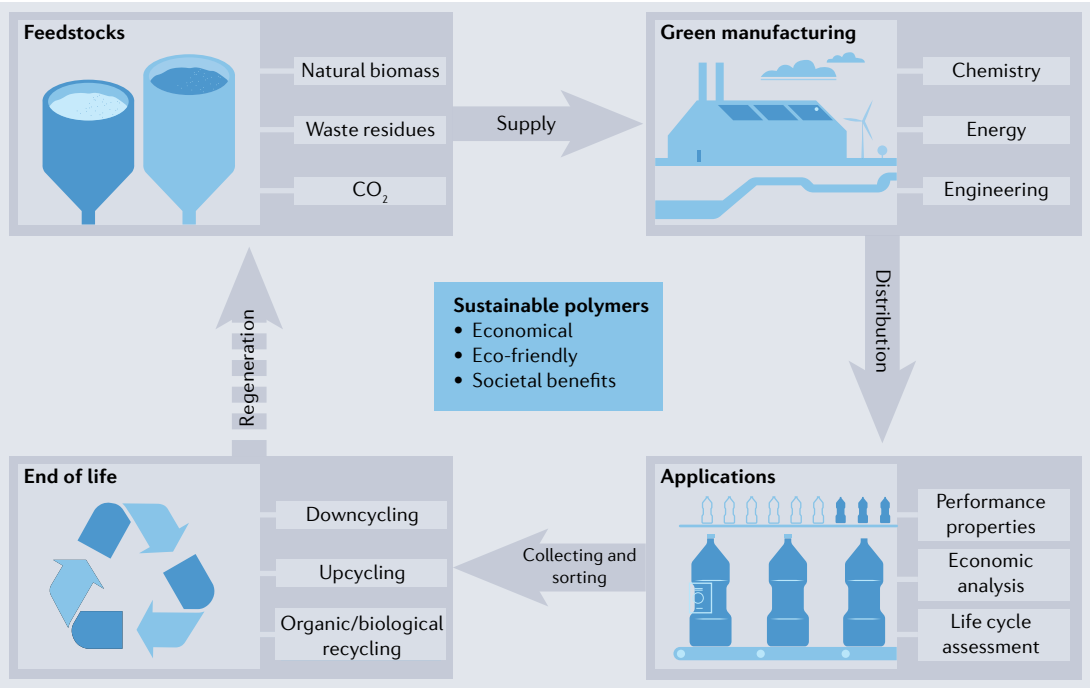


Fig. 1 | **Life cycle of sustainable polymers.** Sustainable polymers begin with renewable, recycled and waste feedstocks, and are processed using green manufacturing (low toxicity, energy/water-efficient manufacturing and less transportation), used in different applications, recovered with responsible end-of-life options and regenerated. This cycle follows the 3R principle — reduce, reuse and recycle — through mechanical, chemical or biological/organic recycling. Dashed arrow indicates that not all end-of-life options are able to go back to renewable resources, such as in mechanical recycling.

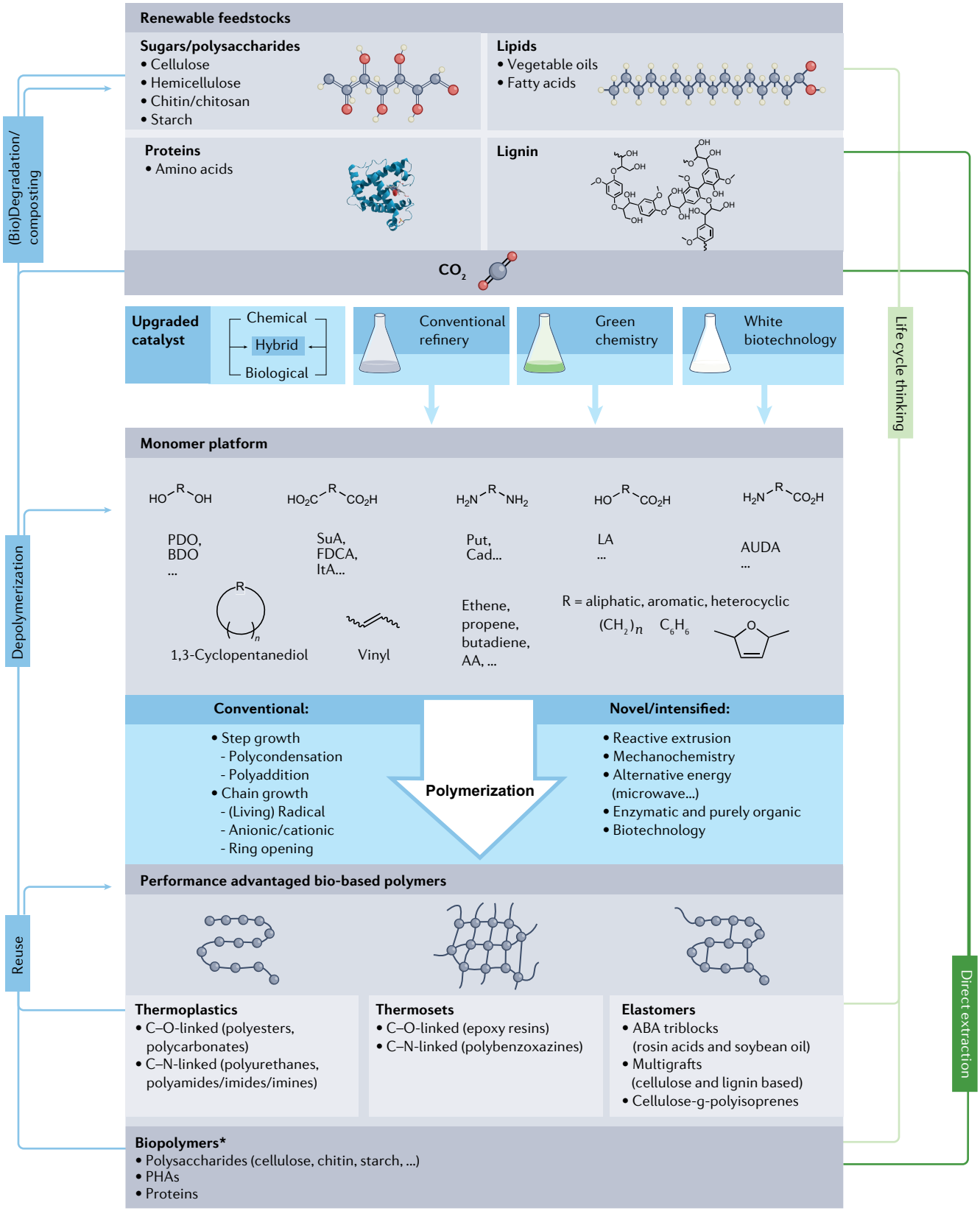


Fig. 2 | **Synthesis of sustainable polymers from renewable feedstocks to bio-based polymers.** Overview of sustainable polymer synthesis, from biomass to monomer, to sustainable polymers including thermoplastics, thermosets and elastomers manufactured. Environmentally responsible end of life (EoL) options include reuse, depolymerization back to monomer and biodegradation back to carbon dioxide, H_2O and cellular biomass. Life cycle thinking is introduced to guide the whole process. Biopolymers (indicated by an asterisk) are the macromolecules (including proteins, nucleic acids and polysaccharides) formed by living organisms. FDCA, furan dicarboxylic acid; PHA, poly(hydroxyalkanoate).

obtain high yield of monomers has limited the commercial valorization of lignin, due to the irreversible lignin degradation and condensation resulting from formation of carbon–carbon bonds[57] during its extraction, and catalyst recovery during hydrogenolysis[58,59]. Novel technologies to block the reactive benzylic positions with a protective agent have been developed to depolymerize lignin, to achieve high-yield monomers for further upgrading[57,60].

Over the past decade, CO_2 has become an attractive C1 resource for monomers and polymers[48]. CO_2 can either be incorporated into backbone chains directly via ring opening, forming carbonyl structures, or reacted to derive monomers such as CO_2-polyol[61] and CO_2-dicarbamates[62]. Use of CO_2 as a feedstock for monomer and polymer synthesis is well summarized elsewhere[48]. In transforming CO_2 into monomers and polymers, the source of energy used to drive the reaction is important not only to accomplish transformation and purification but also for further industrial application of the material. Three approaches have been developed for transforming CO_2 into monomers and polymers: double nitrogen/oxygen electrophile addition via step-growth polymerization; addition of nitrogen/oxygen nucleophile, trapping with carbon electrophile; and use of carbon nucleophile and electrophile. Other interesting but not well-developed platforms rely on the conversion of lipids (vegetable oils, fatty acids and fatty esters)[2], amino acids[50] and pinenes[63].

Prior to developing sustainable polymers, two aspects must be clarified. First, the decision as to which monomers can be extracted from a particular biomass feedstock, and second, the most sustainable methods to extract the monomers from biomass must be identified. Because of the complex nature of monomer extraction, several technical and economic challenges — such as the number of hydrogenation and/or dehydration steps required to reach a desired product, conversion specificity and product throughput — need to be addressed, and new knowledge and better technologies need to be developed.

Sustainable polymer synthesis. Properties for useful life and EoL destiny can be tailored to improve the sustainability of polymers. Fundamentally, the chemistry of polymers must be redesigned to include the use of renewable raw materials beyond fossil fuels in order to conserve energy and valuable resources in chain structures while disassembling the molecular structures after use and maintaining properties after multiple recycling loops[64]. A plastic has been reported with properties comparable with high-density polyethylene (HDPE), with the potential to meet all these criteria. Derived from renewable plant-derived or microalgae-derived monomers, the polymer can be chemically recycled back into its monomers[10]. The large platform of bio-based monomers requires broad-based technologies — such as selective reactions, reversible processes, specific catalysts and so on — for biomass transformation to be developed. This, however, makes product and process sustainability analyses complicated, making the price of the final product a decisive factor[45]. To avoid the discrepancy and better align sustainability to market expectations, pre-identifying useful polymers and, therefrom, specific monomers helps define opportunities for limited research funds, reduce risks and adapt products to existing infrastructure and equipment. The viability of such an approach depends on the success of research performed to transform bio-based monomers into bio-based polymers and their last applicability to technology needs.

The transformation of bio-based monomers to sustainable polymers is performed via chemical or microbial polymerizations. Step-growth and chain-growth processes are the main polymerization classes involved. Among the two, step growth is the more established approach but requires perfect control over reactant stoichiometry, harsh conditions and use of metal-based catalysts, and is both time and energy consuming[65]. The process is largely applicable to bio-based monomers and the resulting polymers can be tuned for (home) composting and/or recycling. Hydroxy acids (such as lactic acid), diols and diacids, and amino and carboxyl functionalities provide the building blocks for some of the most industrially relevant sustainable polymers: polyesters, polyamides and polyurethanes. Chain-growth polymerization more reliably affords high molecular weight polymers, but only applies to cyclic or vinyl monomers. It requires the presence of an initiator that can activate a monomer

Table 1 | **Bio-based platforms for sustainable polymers**

Bio-based platform	Monomer type	Polymer type	Polymerization type	Catalyst	Refs
Sugar	Hydroxy acid, diacids, diols, diamines, cyclics, vinyl	(Co-)Polyesters, polyester polyols, (co-)polyamides, polyurethanes, polyolefins, polyacids	Biological transformation, step growth and chain growth, reactive extrusion	Metal based, organic, enzymes, microorganisms	264–275
Lignin	Acids, alcohols	Polyesters, polybenzoxazines	Step growth	Metal based	276,277
CO_2	Cyclic carbonates	Polycarbonates, non-isocyanate polyurethanes	Step growth and chain growth	Metal based, organic	278–280
Vegetable oils	Triglycerides, fatty acids	Polyesters, polyurethanes, thermosets	Step growth and chain growth	Metal based, organic	281–284
Proteins	Amino acids, (macro)cyclics	Poly(amino acid)s, poly(ester urea)s, polydepsipeptides, poly(ester amide ester)s, peptoids, cationic polymers	Step growth and chain growth	Metal based, organic	68,285–294

Additional information on specific monomers, synthetic pathways and catalysts can be found in Supplementary Table 1.

to run the polymerization (free radical, anionic and cationic)[65]. A special case of chain-growth polymerization is the controlled polymerization enabling polymers of well-defined molar mass, dispersity, functionalities and structures. Lactides with their transformation to PLA via ring-opening polymerization are perfect examples of monomers being industrially polymerized as bio-based polymers using this approach. Indeed, in contrast to step-growth polymerizations, ring-opening polymerization results in polymers having molar masses up to 100 kg mol^{-1} under considerably milder conditions. This synthetic pathway has already been proven, being industrially viable, and is performed in bulk, in solution or in emulsion. A third alternative is microbial polymerization, where metabolically engineered microorganisms convert renewable substrates such as fatty acids into valuable polymers, particularly PHAs[66]. These microbial polyesters show comparable performance with many commodity plastics, while being home compostable. Other aliphatic and aromatic monomers for the synthesis of bio-based polyesters and polyamides can also be produced macrobiotically[67].

The above-mentioned strategies are also applicable in the special case of protein synthesis from amino acids. Performed in solid phase or liquid phase, they have resulted in poly(α-peptoid)s, polyamides or polypeptides, polyesters, polydepsipeptides, poly(ester amide)s, polyurethanes and poly(disulfide amide)s[68–70]. The Ugi reaction — a four-component synthesis involving stoichiometric amounts of acid, amine, isocyanide and aldehyde — has been applied to produce numerous polyamides and polypetoids[50]. Some representative syntheses of sustainable polymers from biomass-based monomers are given in FIG. 3.

Unlike the synthesis of traditional polymers from fossil sources, the synthesis of sustainable polymers has encountered many challenges of monomer diversity and low conversion rates (related to the high complexity of monomer synthesis, extraction and/or purification, often decreasing the purity of the reactants and compromising conversion in polymers), and in some cases, low molecular weight makes these polymers unusable. Monomer diversity and the selectiveness of the blocks transformed from monomers are still major concerns. Another challenge is the mass-based metric, the oldest metric in green chemistry used to quantify sustainability[71,72]. Ideally, any sustainable process must have 100% atom efficiency, meaning that no waste is produced — an E-factor tending to 0. Unfortunately, in the case of polymerization, the E-factor is almost always positive. Chemoselective polymerization[73] and a customized synthesis platform[74] have shown some advantages and efficiencies in the synthesis of sustainable polymers from mixed monomer feedstocks. Catalyst design occupies a leading position for material development[75–77], as all polymerization strategies apply catalysts to decrease the energetic barrier, increase the conversion rate and guide it to high molar mass polymers, including well-defined macromolecular architectures. Conventional catalysts are all metal based, which may prompt significant concerns in terms of biological toxicity, compromising the sustainability of polymers[78]. Many researchers have investigated replacing the metal-based catalysts with purely organic compounds or enzymes, more efficient, less toxic or heterogeneous equivalents[79–83]. However, their use and application in sustainable polymer synthesis remains a question of priorities and compromise, and requires additional steps for polymer purification, often using organic solvents, thus adding cost, and introducing new problems of solvent residues and recovery. With sustainability, scientists have increasingly developed new understanding of the factors required for catalysts in polymer synthesis[84]. New pathways such as the dynamic stoichiometric strategy have been developed and applied to permit significant breakthroughs in sustainable polymer production on an industrial scale[84]. Biocatalysts such as tailored enzymes offer a new opportunity in sustainable polymer synthesis and recycling regarding green chemistries, although they are still rarely developed because of the cost and accessibility[85,86].

Much progress has been made to date (FIG. 3), and different polymer precursors and polymers have been synthesized. But the polymerization methods have experienced problems such as time and energy consumption, conversion issues and product diversity, and use of non-selective/toxic (co-)catalysts, which results in non-effective, polluting and quality-influencing outcomes. These problems are yet to be overcome to satisfy all principles of sustainability[87,88]. This is where innovative and rational design and adaptation of polymerization methods can play a role. Accordingly, a crossover between bio-based monomers and bio-based polymers appears where technology needs meet opportunities. This crossover identifies structures that are most easily obtained from existing conversion processes and finding expanded applications despite their high-risk nature and long-term programmatic commitment.

Processing synthesis. Although concerns associated with the sustainability and environmental impacts of petrochemicals have spurred the development of polymers sourced from renewable resources, it is additionally important to focus on generating property improvements that can expand applications of polymers. For instance, synthesis of sustainable polymers without solvent, such as reactive extrusion, would be accepted by the industries for its environmental friendliness and for cost-efficiency. Processing synthesis and modifications to synthesis that further improve the properties of sustainable polymers for real applications are desired.

There are already many significant industrially relevant methods dedicated to synthesis of sustainable polymers: microwave, gamma or electron beam irradiation, (reactive) extrusion and solid-state modification[89]. Amongst these, reactive extrusion has proven to be sustainable in terms of flexibility, efficiency, time and energy consumption to design new polymers/materials with improved performances[90]. For basic purposes, the process is used to blend bio-based polymers and (nano-) charges[91]. More recently, special designs have been engineered to allow continuous polymerizations and even to combine multiple reactions to achieve bio-based polymer production with tuned properties[92,93]. The versatility of reactive extrusion also allows combining it

Ugi reaction
A multicomponent reaction in organic chemistry involving a ketone or aldehyde, an amine, an isocyanide and a carboxylic acid to form a bis-amide (named after Ivar Karl Ugi).

Mass-based metric
A measurement of the atom efficiency of chemical reactions comparing the mass of desired product with the mass of waste.

E-factor
The ratio of the mass of waste per mass of product in a chemical reaction.

Reactive extrusion
A manufacturing method carried out in an extruder, which combines chemical polymerization and extrusion processing into a single step.

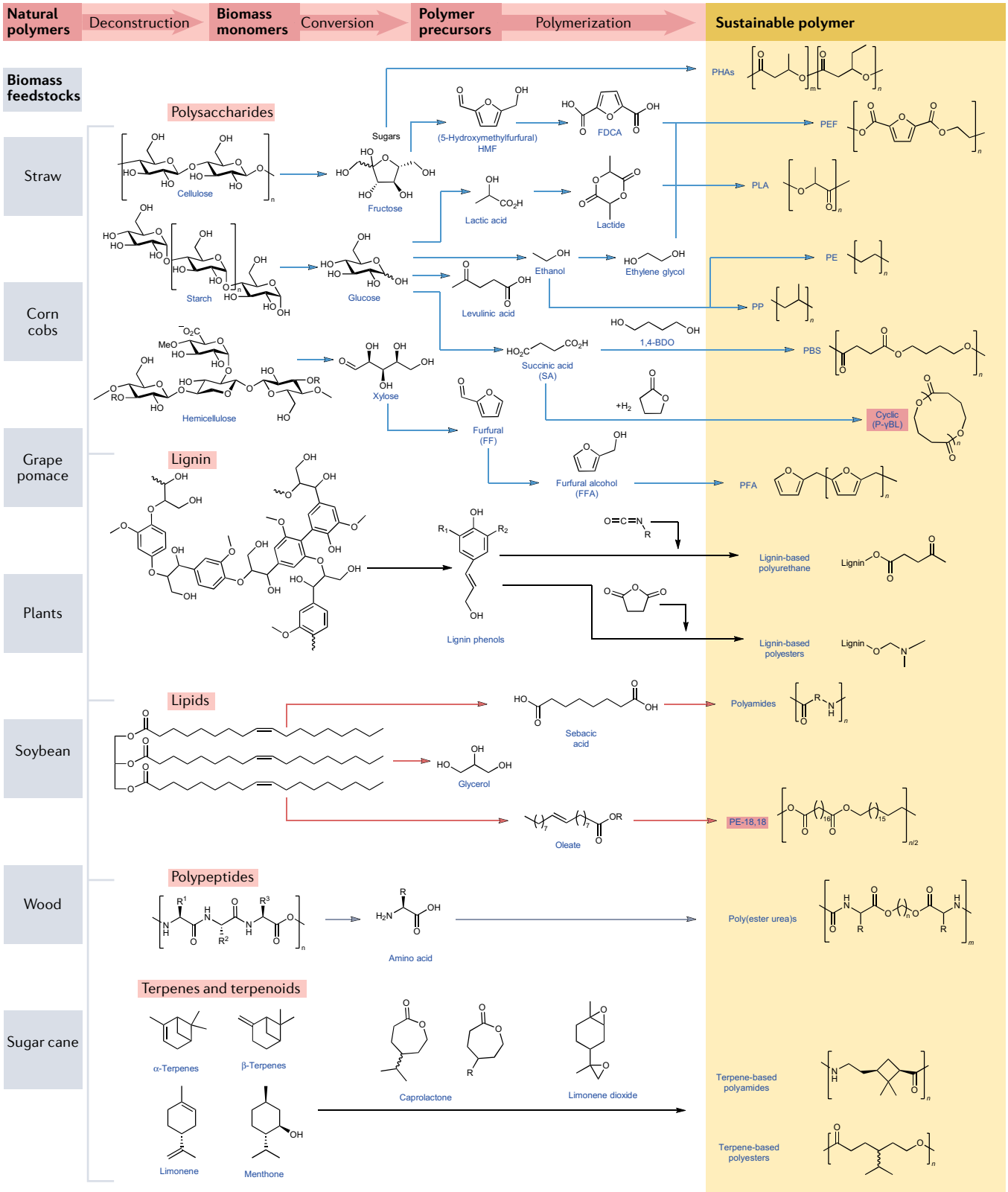
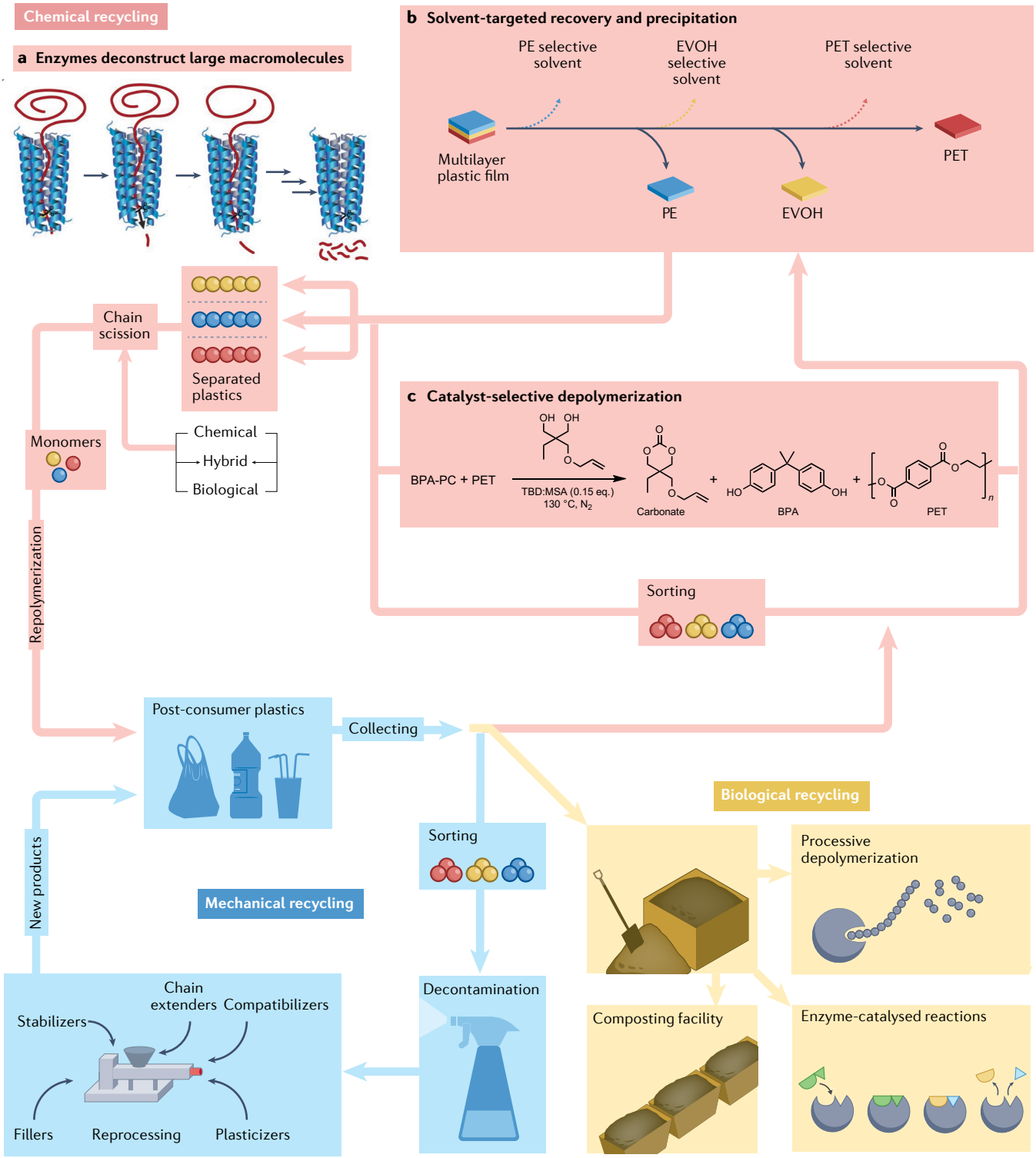


Fig. 3 | **Routes to some representative sustainable polymers synthesized from biomass feedstocks.** Natural biopolymers including polysaccharides, lignin, lipids, polypeptides and terpenes can be extracted from renewable biomass. Through deconstruction and conversion or fermentation, natural biopolymers can be turned into polymer precursors for sustainable polymer polymerization. Listed sustainable polymers, poly(hydroxyalkanoate)s (PHAs), polylactide (PLA) and polybutylene succinate (PBS) are biodegradable/compostable; cyclic (P-γBL)[295] and PE-18,18 (REF.[10]) are chemically recyclable to monomers; and all sustainable polymers are bio-based. FDCA, furan dicarboxylic acid; PE, polyethylene; PEF, poly(ethylene 2,5-furandicarboxylate); PFA, poly(furfural alcohol); PP, polypropylene.



with direct material production. Using as-synthesized polymer directly from the extruder as filament material connected to a 3D printer is an innovative way to produce complex shapes of highly reflective bio-based blends[94,95], bio-scaffolds[96] and reactive mixtures[97]. At large scale, high-rate printing technology would involve two-component thermosetting material mixing at the deposition point and undergoing fast crosslinking with tailored reaction kinetics and rheology. Another approach gaining attention is solid-state polymerization, bringing novel functionalities and EoL scenarios to bio-based polymers. At temperatures between the T_g and the melting temperature (T_m) and in the presence of an appropriate catalyst, solid-state polymerization could impart novel functionalities — such as improved flame retardancy, bioactivity and biodegradability — into the bio-based polymer without compromising the primary thermo-mechanical properties. Mainly developed for poly(butylene terephthalate) (PBT)[98], solid-state polymerization is compatible with reactive extrusion,

Fig. 4 | **End of life of sustainable polymers.** Mechanical recycling, chemical recycling and biological recycling are the basic end-of-life options. Chemical recycling involves depolymerization of macromolecules to monomers followed by repolymerization into original polymers or production of other functional chemicals. To improve efficiency of recycling, various strategies have been developed. **a** | Enzymes deconstructing large macromolecules, in which high-density polyethylene (HDPE) could be depolymerized into diesel and alkanes[110]. The idea promotes development of biological catalyst for polymer recycling. **b** | Recycling multicomponent plastic materials such as multilayer packaging, a method called solvent-targeted recovery and precipitation[296], has been developed where multilayer packaging was separated for subsequent recycling. **c** | Catalyst selective depolymerization[297] has also been used in recycling mixed plastic materials into monomers via selective depolymerization of one component, thus improving recycling efficiency. Mechanical recycling involves reprocessing of mono or mixed plastic materials, normally accompanied by decline in performance. Biological recycling involves biodegradation of macromolecules into carbon dioxide, H_2O and biomass in composting facilities (home or industrial). Different biotechnologies have been developed to promote plastic degradation, such as processive depolymerization and other enzyme-catalysed reactions. Processive depolymerization means that the degradation is programmable and complete via chain end-mediated depolymerization. Enzyme-catalysed reactions involve engineering of microbes with plastic-biodegradation modulus and ability, which can be embedded in the matrix to tailor the biological recycling. EVOH, ethylene vinyl alcohol; PE, polyethylene; PET, poly(ethylene terephthalate). Part **a** reprinted from REF.[110], Springer Nature Limited. Part **c** adapted with permission from REF.[297], Wiley.

enabling users to switch from batch to continuous processing. Microwave-assisted synthesis (combined with reactive extrusion or not) is another important method currently under industrialization[89]. Microwave synthesis has proven to increase the rate and effectiveness of PLA[99] and polybutylene succinate (PBS)[100] synthesis. Control of molecular weight and distribution, the chain structure of the reaction product and reaction mechanism identification are the main issues to be addressed regarding the processing synthesis.

Green composites made of sustainable polymers reinforced with natural biomass such as natural fibres, agro-residues and waste biomass date back to the 1900s when cellulose fibres were used to reinforce phenolic resins[101]. Addition of natural fibres to polymers increases stiffness, strength, toughness and heat resistance, while generally reducing cost. Advanced green composites with high strength and stiffness have been successfully prepared using high-strength and oriented liquid-crystalline cellulose fibres, and bacterial cellulose nanofibres as reinforcement, opening the door for advanced green composites to be used for structural applications in automobile, aerospace and civil structures, replacing glass/Kevlar fibre with registered trademark/epoxy composites[102]. In addition to growing oriented bacterial cellulose nanofibres in narrow channels of polydimethylsiloxane (PDMS) templates or small-diameter PDMS tubes[103], for use in advanced green composites, the oriented cellulose nanofibres may also be used in ropes, parachute/sail fabrics and, possibly, bulletproof vests[102,103]. Blending nano-celluloses such as bacterial cellulose and nano-fibrillated cellulose in transparent polymers would result in strong/tough transparent green composites[104,105], potentially replacing glass in numerous everyday applications (such as automobiles, airplanes, house windows and electronics), making them lighter and/or more fuel efficient. Biomass compositions, forms, size, shape, dispersion in the matrix and biomass/matrix interfacial actions are all known to influence the final performance of the green composites. Using compatibilizers (in situ or ex situ) and surface treatment of the fibres can improve their dispersion, as well as increase fibre/resin interfacial bonding, resulting in better mechanical properties of green composites. Green composites with bio-based but non-biodegradable resins pose the challenge of separating fillers from matrixes (resins) for recycling which are time consuming and energy intensive. However, biodegradable green composites should find applications in many fields due to their ability to be composted at their EoL, once cost and properties are balanced.

End of life

Material selection and application design must be coupled with a suitable EoL management option to facilitate the paradigm shift to fully circular materials (FIG. 4). Mechanical, chemical and biological recycling have become part of global movement to reduce and/or eliminate plastic waste in the environment, and to reuse plastics at the end of their useful life as a valuable resource. The inherent properties of plastics come from the polymer design itself, predetermining the different EoL options: disposal, recycling or composting. As an important class of bio-renewable monomers consisting of ester linkages, lactones can be synthesized into polyesters that are biodegradable[106]. The acetal linkage has also shown its hydrolytic and biodegradable potential. By incorporating acetal functional groups, a PLA showing molecular weight loss in water was reported[15,107]. However, for bio-based polymers formed by addition across a C–C π-bond — such as bio-based polyolefins (bio-PE and bio-based polypropylene (bio-PP)) — biodegradation or chemical recycling to monomer (CRM) remains quite challenging[20], requires harsh conditions and results in mixtures of low-value products. Progress on mechanical recycling with the use of a high-efficiency compatibilizer[9] or upcycling of these polymers resulting in high-value end products such as long-chain alkylbenzenes[11], graphitic carbon[108] or well-defined dicarboxylic acids[109] through catalyst development[11,110], as well as in-chain group incorporation[111], have also been developed. Upcycling is a stratagem adding value to the existing waste materials. Distinguished from chemical recycling that describes technologies that convert polymeric waste to raw materials for manufacturing new plastic articles, upcycling indicates transforming plastics into chemical feedstocks such as waxes, or value-added materials such as carbon-based materials for more advanced applications. The terminology difference between chemical recycling and upcycling[112], and the challenge of using upcycling to repurpose the plastic waste, can be found elsewhere[113]. While realizing sustainability, researchers should maximize resource efficiency and eliminate waste, hazards and pollution when bio-based polymers are used, and guide product designers in designing for circularity with consideration to geographic differences[31]. Moreover, diversity of polymers, additives and pollutants invariably complicates the recycling reality, making post-consumer plastic recycling an insurmountable challenge[114].

Upcycling
A recycling system in which the recycled material is of higher quality and functionality than the original material.

Downcycling
A recycling system in which the recycled material is of lower quality and functionality than the original material.

Ergoneutral
A reaction in which Gibbs free energy is zero under a defined set of reactive conditions.

Mechanical recycling. Mechanical recycling involves processing plastic waste into recycled products without vastly altering the chemical composition of the material, except the possible use of new additives. It is an essential component of the circular economy, which can be identified as primary or secondary recycling[115]. In melt processing, mixed plastic waste mainly undergoes manual and/or automated sorting and reprocessing after cleaning and grinding. Unsorted mixed plastic waste impurities can cause structural weakness in the recycled materials due to their inherent immiscibility. Commodity plastics such as polyethylene (PE), poly(ethylene terephthalate) (PET) and polypropylene (PP) have been approved for recycling programmes and are reprocessed into waste bins, furniture, mobilizers, floors, fibres and bubble wrap using efficient compatibilizers[9]. The value of the recycled products is typically lower so the term downgrading or downcycling is often used. Although all thermoplastics can, in theory, be recycled mechanically with a small to non-existent decline in performance, contamination, lower quality of recycled plastics and high operation costs (caused by storing, sorting and processing) have mainly hampered the capital investment in mechanical recycling[116]. To promote mechanical recycling of plastics, it is critical to design and develop innovative sorting using sensors and artificial intelligence robotics, extrusion technologies and high-efficiency compatibilizers, chain extenders and catalysts, along with fundamental study of degradation mechanism in melt processing[117]. The main challenge is to avoid generating additional, unusable new waste after an item's new useful lifetime, especially when new additives are included. The use of chemical recycling for those materials that can no longer be mechanically recycled would be a great complement.

Chemical recycling. For materials not suitable for mechanical recycling, new processes such as solvolysis, dissolution and/or precipitation or closed-loop CRM need to be developed to convert the waste plastic into original monomers or other value-added chemicals[20,118,119] (BOX 1). These processes hold the possibility to obtain pure monomers by removing additives or colourants and separate mixed plastics for process. They also create an urgent demand for development of green catalysts and their recovery, green solvents and mild yet efficient processes to create more business opportunity[118]. Closed-loop CRM, which theoretically can be repeated indefinitely[3], has recently drawn significant attention. In theory, polymer chains have the characteristic ability to be depolymerized into oligomers or monomers. In practice, however, recovery of monomers is closely related to polymer energetics[20]. Being generally costly and energy intensive, CRM faces the challenges of low monomer selectivity for some plastics[120], low convention rates and possible trade-off between the recyclability and service performance[3]. To lower the energy barrier of bond cleavage in depolymerization, advances in chemical and biological catalysis[85], dynamic polymer networks[121,122] and choosing the right monomer and its polymerization method[123,124] have shown promise. Recyclable polymers derived from γ-butyrolactone[123] and its ring-fused structures (4,5-*trans* six-membered ring-fused γ-butyrolactone)[124] and ring-fused cyclooctenes[125,126] provide examples of polymerization–depolymerization via monomer design, where polymerizations are almost ergoneutral. Successful synthesis of recyclable poly(1,3-dioxolane) (PDXL) exhibiting good thermal–mechanical properties shows the strength of novel catalyst design and polymerization control[127]. Close-loop recycled poly(diketoenamine)s via dynamic polymer network design reveal a novel example of the potential future of chemically recyclable plastics, especially for thermosets[122]. In addition to environmentally benign and economic process (green catalyst, solvent-free polymerization/depolymerization), integrating chemical recyclability with performance-advanced properties in sustainable polymers suggests the future direction of this field. Linear thermoplastics, constituting the largest segment of polymer production and waste generation, is still the biggest challenge for chemical recycling. Recently developed, renewable polycarbonates and polyesters from oleate have exhibited similar thermal–mechanical performances to PE; however, closed-loop recycling into monomers may be a viable strategy for the sustainable utilization of polyolefins[10]. Details of chemical recycling of traditional and novel polymers are well summarized[19,20]. For those plastics that are difficult to recycle chemically or to upcycle — such as crosslinked epoxy[128] and composites containing additives such as flame retardants[129] — laboratory-scale mechanochemistry involving high-energy ball milling has been developed. Scalability operation of mechanochemical recycling is still hampered by energy consumption, time efficiency and proper milling apparatus.

Box 1 | **Chemical recycling terms, categories and new ideas for the circular economy**

Framework

Chemical recycling is distinctly different from physical recycling, mechanical reprocessing and chemical processes that primarily result in fuels or energy recovery. Chemical recycling of polymeric waste can be divided into closed-loop recycling to monomers and other feedstock recycling. Waste polymers can be processed via different technologies including catalytic depolymerization (or chemolysis), solvolysis and pyrolysis.

Technologies

Depolymerization describes technology to break down the backbone bonds of polymers to form monomers. The monomers are identical to virgin monomers used in the preparation of polymers.

Solvolysis involves treating polymeric wastes with solvents and reagents to depolymerize them to low molecular weight chemicals and oligomers[301]. Based on the solvent used, different solvolysis terms are developed. The term hydrolysis is used when the solvent used is water[302], methanolysis when using methanol, ammonolysis when using ammonia and glycolysis when using glycols.

Pyrolysis occurs in the absence of oxygen, using heat to break plastics into a range of basic hydrocarbons. The light oils generated from the pyrolysis process can be cracked to obtain monomers, which is termed feedstock recycling. Heavier products such as wax obtained from pyrolysis are considered as recovery, not chemical recycling. When syngas is generated during very high-temperature thermal cracking, it is termed gasification. The syngas can be transformed into chemicals for plastic production.

The nature-inspired circular economy recycling pearls concept for proteins involves the depolymerization of proteins into amino acids (the monomers) followed by their reassembly into entirely new proteins. This concept involves deriving all plastics from one set-up of building blocks with their particular sequence imparting the properties needed for applications[14].

Biodegradability and compostability. For applications where plastics, especially packaging plastics, are contaminants in the organic waste stream, or where products have high risk of ending up in a natural environment, a certified and verifiable compostable or soil biodegradable material is ideal[130]. As stated previously, conversion of the polymer carbon to CO_2 by microbial metabolism in defined EoL environments must be demonstrated. Biodegradability is uniquely accepted when accompanied by complete microbial bio-assimilation, which is necessary to eliminate harmful impacts associated with microplastics and nanoplastics derived from partial degradation of plastics[131]. Although biodegradation may be able to reduce the persistence and accumulation of microplastics in the environment, the estimated time to complete removal from the natural environment must be calculated using ASTM/International Organization for Standardization (ISO) standards and the intermediate breakdown products need to be carefully evaluated for their environmental impact. In landfill, biodegradable/compostable plastics will show little or no biodegradation and offer no environmental value benefit. Equally important, biodegradation versus recycling (even as an energy source) needs to be delicately balanced and based on application and use.

The challenges of home compostability, biodegradability claims, plastic collection, sorting and recycling are major barriers to successful implementation. Consumers should be made aware of different biodegradation claims, for example, a consumer product based on industrially compostable sustainable polymers may not be completely bio-assimilated under the conditions of home composting processes. A study of biodegradability of mainstream plastics such as PLA, PBS and their blends in different environment has been presented[132]. Biodegradation mechanistic studies such as tracking of carbon evolution during biodegradation[133] and novel technologies such as biocatalysts with embedded enzymes[134] highlight new approaches to biodegradable–compostable polymers. More details on biodegradability and compostability, including their definition, misperceptions (such as bio-based is not equal to biodegradability) and their role in managing plastic waste, can be found in the supporting information (Supplementary Note 1).

There is much work to be done to design an optimal EoL. Currently, much of the efforts are focused on educating consumers and community programmes in collecting, sorting and cleaning, and separation technologies; improving polymer-tracking using covalently bonded identification markers, and recycling methods, including product optimization to reduce the amount of virgin material; and identifying more efficient technologies such as biological factories, and/or selective catalysts and enzymes as has been done for some thermoplastic polyurethanes[135] and PET[136].

Safety considerations. The continuous development of high-quality and high-performance sustainable polymers can bring novel products to market. The focus, progressively, relies on sustainability, from polymer design to the EoL scenario. However, sustainable polymers and materials are relatively new to the consumer market and attention needs to be paid to safety considerations. Based on the principles of green chemistry and engineering[78,87,137], safety considerations encompass feedstock choice, manufacturing and regulations or policymaking.

Green chemistry principles should guide the selection of monomers, catalysts and additives. It is important to identify a product EoL scenario, including availability of waste management infrastructure and overall impact of recycling versus disposal, among other factors. Renewable, waste-derived or recycled feedstock should neither interfere with ecosystem health nor deplete resources. Use of forever and problem chemicals should be excluded in feedstock selection as it interferes with sustainability. Production and manufacturing should be evaluated from unit process mapping (including supply chain, deviations, energy, time, cost) to chemical identification and remain transparent overall. Finally, current loopholes in regulations and policies need to be addressed to prevent greenwashing and misleading consumers. In addition to direct human health effects such as toxicity, carcinogenicity and mutagenicity, consideration is needed for indirect effects, persistence and/or bioaccumulation and physical hazards such as flammability, explosivity and reactivity. Sustainable polymers can only contribute to circularity when correctly used. Further progress in this direction requires public and governmental support and a commitment to research and education to expand the scope of sustainability considerations and address global product and geographic differences.

Results

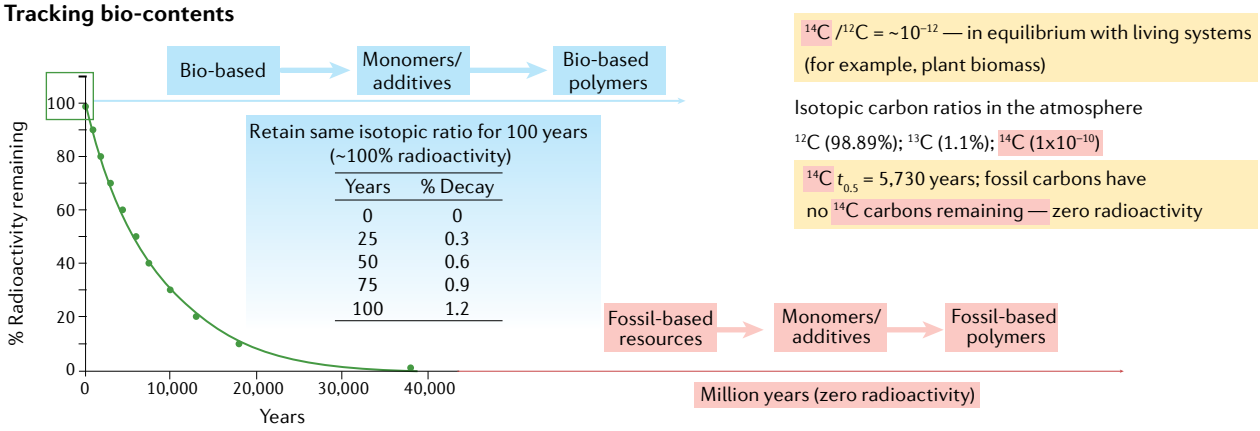
Rational characterization ensuring reproducibility of processing and properties is crucial to bridge the gap between experimental development of sustainable polymers and practical applications. This section describes typical characterization methods and outputs of sustainable polymers from feedstocks to EoL. These techniques inform users about the choices of sustainable polymer design by providing information such as the yield and selectivity of monomers, as well as the evaluation of sustainability by LCA.

Sustainable evaluation

With the migration from a linear economic model to a circular economy, what makes a fully sustainable polymer remains a question to be answered. Fundamentally, renewable feedstocks and environmentally responsible EoL are two basic characteristics that grab our attention. Practically, the carbon footprint through the entire life cycle of the plastic, from feedstocks to products to EoL, should also be tracked to evaluate sustainability. To complete the entire evaluation process, bio-content measurements, tracking the change of carbon into CO_2 during the biodegradation processes and the LCA as shown in FIG. 5 are required. In addition to the sustainability evaluation, other factors including the possible ecotoxicity of the additives (such as the catalyst used), process water and energy and the importance of preventing accumulation of microplastics and nanoplastics in living organisms should also be considered when developing fully sustainable polymers.

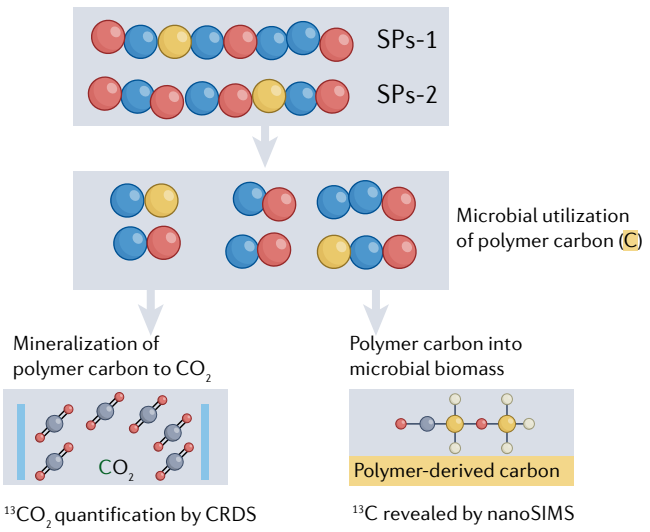
Linear economic model
An economic system consisting of the 'take, make, dispose' model in which most goods end up discarded as waste.

A Tracking bio-contents

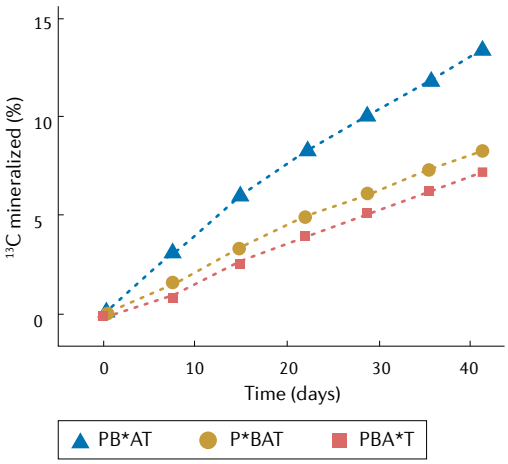


Ba Tracking carbon utilization in biodegradation

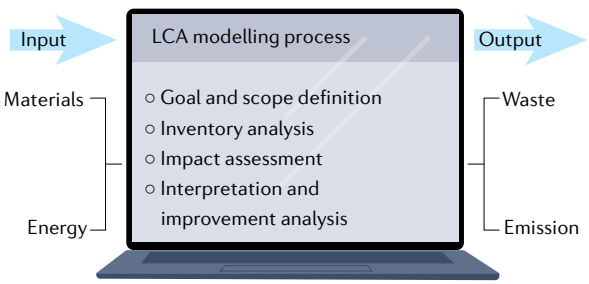
Carbon-labelled biodegradable polymers (e.g. ^{13}C-labelled) in soil incubation



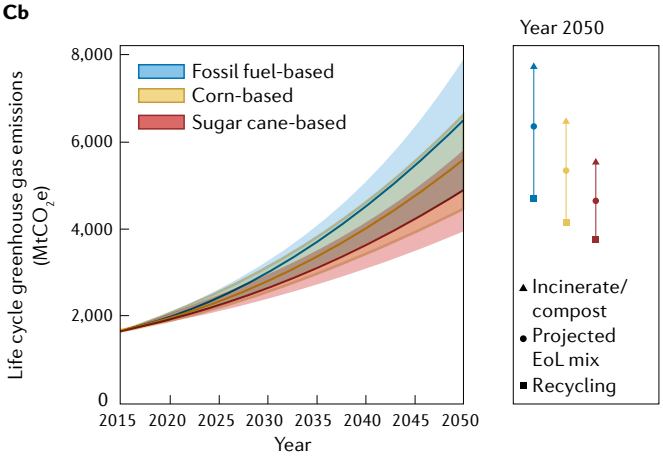
Bb



Ca Life cycle thinking



Cb



The bio-based carbon content of the developed plastics is calculated via radiocarbon analysis based on ^{14}C radioactive decay using accelerator mass spectrometry, benzene analysis and the CO_2 cocktail method[138–140]. Accelerator mass spectrometry is widely accepted for its accuracy in measuring radiocarbon dating[141]. The methodology to measure bio-carbon contents relies on the fact that no ^{14}C isotope is found in petrochemical feedstocks as they are formed over millions of years (FIG. 5A). This approach to measure bio-based carbon

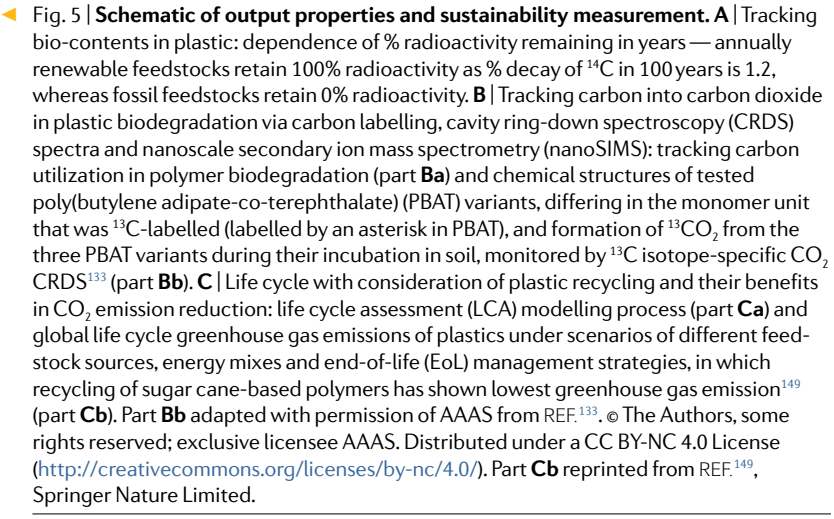


Fig. 5 | **Schematic of output properties and sustainability measurement. A** | Tracking bio-contents in plastic: dependence of % radioactivity remaining in years — annually renewable feedstocks retain 100% radioactivity as % decay of ^{14}C in 100 years is 1.2, whereas fossil feedstocks retain 0% radioactivity. **B** | Tracking carbon into carbon dioxide in plastic biodegradation via carbon labelling, cavity ring-down spectroscopy (CRDS) spectra and nanoscale secondary ion mass spectrometry (nanoSIMS): tracking carbon utilization in polymer biodegradation (part **Ba**) and chemical structures of tested poly(butylene adipate-co-terephthalate) (PBAT) variants, differing in the monomer unit that was ^{13}C-labelled (labelled by an asterisk in PBAT), and formation of $^{13}CO_2$ from the three PBAT variants during their incubation in soil, monitored by ^{13}C isotope-specific CO_2 CRDS[133] (part **Bb**). **C** | Life cycle with consideration of plastic recycling and their benefits in CO_2 emission reduction: life cycle assessment (LCA) modelling process (part **Ca**) and global life cycle greenhouse gas emissions of plastics under scenarios of different feedstock sources, energy mixes and end-of-life (EoL) management strategies, in which recycling of sugar cane-based polymers has shown lowest greenhouse gas emission[149] (part **Cb**). Part **Bb** adapted with permission of AAAS from REF.[133]. Part **Cb** reprinted from REF.[149], Springer Nature Limited.

content is codified into ASTM standard D6866 and ISO 16620 series standards. The ASTM standard is based on organic carbon basis, whereas the ISO standard provides for calculations based on organic as well as biogenic carbon (organic + inorganic carbon). A good example of the calculation of bio-carbon contents in a bio-based ethanol/fossil-based methanol (50/50) mixture has been reported[142]. Corrections for isotopic fractionation and bomb carbon should be completed to obtain accurate values[139]. The bomb carbon corrections result from the open-air nuclear bomb testing that peaked in the 1960s, leading to 7–8% higher ^{14}C activity levels compared with those of the pre-bomb age.

Tracking carbon evolution into CO_2 during biodegradation not only helps us identify the carbon footprint but also facilitates understanding of the biodegradation mechanism, which can promote new biodegradable polymer designs. Tracking is followed by using specifically synthesized carbon-labelled polymers and tracing the labelled carbon, for example, tracking with cavity ring-down spectroscopy (CRDS) using ^{13}C-labelled carbon[133] (FIG. 5Ba). Compostability measurements and specification standards have been established in industrial composting systems (ASTM D6400 and D6868, EN 13432, NF EN 14046 and ISO 17088 and 14855), which include biodegradability, disintegration and ecotoxicity. ASTM/ISO standards have also been developed to test in different natural environments such as soil, ocean and anaerobic digesters[143] as listed in Supplementary Table 2.

Whereas compostability is already measured based on internationally defined standards, chemical recycling processes are still being developed. Therefore, LCA is carried out to evaluate the sustainability of recycling plastics, as well as the reduced greenhouse gas emission. The environmental impact of the selected categories[144] should be quantitatively assessed for sustainable product development, which is included in the principle and framework of LCA, particularly goal and scope definition, inventory analysis, impact assessment and interpretation (FIG. 5Ca). International standards ISO 14040:2006 and ISO 14044:2006 or specific standards for plastic such as ISO 17422:2018 and ASTM F1675-13 have been developed for LCA. Requiring complex calculations, LCA is aided by professional software tools developed by academics and the industry following the above standards, such as SimaPro[144], OpenLCA[145] and GaBi[146]. That using different software tools can generate different results for the same system is well recognized and analysed[146–148], and is attributed to different models and the LCA database adopted into the commercial software tools. Along with the model used, the method in which data are accessed and the selection of system boundaries and carbon boundaries also influence the final LCA output. To have an effective and reliable output, primary data collected directly from operating process plants are preferred over secondary data or models[145]. There is no doubt that renewable feedstocks and an environment-friendly EoL are two essential factors for developing a fully sustainable polymer (FIG. 5Cb); however, LCA analysis and thinking should guide the sustainable polymers' development and application. To conclude, bio-sourcing, biodegradability and/or compostability or a recycling loop without full verification are not sufficient for complete sustainability evaluation. Sustainability covers the entire life cycle of a plastic, including its complete production line (transport issues included), use phase and EoL scenarios (reuse, re/upcycling, composting, biodegradation/bio-assimilation) and ecotoxicity evaluation[149].

Successful sustainable polymers

Sustainable polymers must be accepted by the broad market to be successful. To achieve this target, properties and economic costs of the developed polymers should be comparable with or superior to the conventional products, with the added benefit of sustainability[17]. Unlike petroleum-based plastics whose cost mainly depends on feedstocks, the sustainable polymers manufactured from renewable feedstocks are costly in each step from feedstocks to polymerization. Therefore, appropriate characterization of the following steps is necessary: highly efficient conversion of biomass to monomers and to sustainable polymers with a novel catalyst[150]; precise synthesis of polymers with controlled structures that bring novel properties and selectivity; and the conversion rate of depolymerization. To minimize the energy required to produce sustainable polymers, each step from monomer extraction to polymer processing requires optimization and innovation, including using renewable and minimal energy, using green solvents/catalyst, minimizing the number of transformation steps, increasing the atom economy and developing new and efficient polymerization methods where possible.

Yield and selectivity are major considerations for both extraction and polymerization of renewable monomers. The basic analytical techniques, such as nuclear magnetic resonance spectroscopic analysis, mass spectrometry, microscopy (such as liquid phase electron microscopy)[151] and rheological analysis[152], can be used to identify the chemical structure of monomers or polymers, whereas gas chromatography–mass spectrometry can be used to analyse monomer purity or reaction selectivity. Advanced characterization approaches such as the effective carbon number method have precisely calculated the yield of monomers[60], due to the difficulty

Radiocarbon dating
A method that uses the level of radioactive carbon (^{14}C) to determine the age of carbonaceous materials, as ^{14}C decays over time — older artefacts have less ^{14}C than younger (newer) ones.

Biogenic carbon
The organic and inorganic carbon originating from renewable plant-biomass carbon feedstock.

in obtaining or producing a large quantity of monomers. Once bio-based polymers are produced, the atom efficiency of reactions such as monomer synthesis and extraction, monomer to polymer conversion, absence or presence of by-products and side reactions must be confirmed. Catalysts and any other possible additives should be quantified as precisely as possible based on their sustainability. Depending on the targeted application, thermo-mechanical and/or biological properties are of primary interest, and correlations between structure, processing and properties should also be established. To support and improve sustainability of bio-based polymers for a circular economy, stringent performance criteria for analytical methods should follow fast evolution in characterization methods with consideration of extensivity, solvent use, energy use, mass and time reduction, accuracy and repeatability. Spectrum techniques such as near/mid-infrared Raman spectroscopy and liquid phase electron microscopy attached to chemical reactors (including extruders in reactive extrusion) hold advantages in identifying polymer structure evolution during polymerization, and in turn help control the structures and properties as needed at the upfront. Details of characterization methods and their outputs and actions are presented in Supplementary Table 2.

The next step after validating both processing conditions and structure for sustainable polymers is the measurement of properties for comparison with conventional polymers. At this point, the goal is not to adapt methods or processes to require sustainable polymers to replace conventional plastics using the same norms and needs. An ideal situation would be to reach the required application properties with less complexity. Properties that should be compared with traditional petroleum-based polymers, including processability, crystallization, rheology, mass transportation, mechanical, electrical conductivity and so on, should be evaluated using the corresponding characterization methods.

Although there are numerous approaches to create polymers from renewable feedstocks for successful applications, LCA provides an important tool to measure sustainability and guide future research. However, LCA is not suitable for comparative studies between the sustainability of different systems. Sustainability evaluated by LCA may change with the development of technology and times. Precise characterization of the efficiency, structure and properties of the compound in each step — using correct characterization tools and recognized international norms and claims — is extremely important. Even while international standards and claims are difficult to meet, choosing certified compostable/recyclable products guarantees that material can be swiftly turned into a value-added resource providing benefits to local industry and the circular economy, to avoid landfill, limit pollution and help combat global warming.

Applications

In this section we present several main challenges to promote sustainable applications by discussing sustainable polymers for commodity and advanced fields with promising commercial prospects. Commodity applications, sustainable polymers with high thermal resistance, sustainable polymers for 3D printing and market development of sustainable polymers are reviewed.

Thermal resistance

PLA is a great commercial success story for sustainable polymers, yet high-temperature applications have been greatly limited by its low T_g (REF.[153]). Numerous other sustainable polymers face the same issue of poor heat resistance, which creates a quest to develop bio-based thermoplastics and thermosets with high T_g (REF.[55]). As an example to be used in high-tech applications such as aerospace, the T_g of thermosets should be above 220 °C (REF.[154]). The non-recyclability of thermosets, whether bio-based or petroleum-based, is a hurdle for the development of sustainable products at their EoL. This has created a demand for research on thermosets that either have longer service life, are reusable and/or are recyclable, including malleable, self-healing, biodegradable, chemically recyclable or thermally reprocessable materials[155]. Using covalent adaptable networks with dissociative or associative bonds is a promising approach to achieve thermally reprocessable or chemically recyclable thermosets[156]. Reversible dynamic covalent chemistry can also improve the properties and mechanical recyclability of thermoplastics. As an example, dioxaborolane metathesis was used to turn polyolefins and other carbon–carbon backbone thermoplastics into high-performance vitrimers with improved mechanical recyclability[157]. Bio-based thermosets free of formaldehyde and bisphenol A have also been synthesized[158–164]. To achieve chemically recyclable, degradable or cleavable linkages, disulfide[165–167], boron ester[162], ether[168], imine[169–171] and cyclic acetal[172,173] bonds, among others, have been incorporated into bio-based thermosets[173]. The introduction of reversible cross links, however, can cause a certain degree of trade-off in thermal, mechanical and chemical stability compared with more permanent covalent cross links. Achieving high thermal and mechanical stability, at the same time as the bond-exchange reactions are used at elevated temperatures, to ensure reprocessability and recyclability is a challenge. These materials are still mainly in the research stage.

Although commercial bio-based materials with high T_g are still in infancy, fundamental research on the effect of different structural units on T_g has been performed. Lignin and lignin monomers, benzoxazine, tannins and other bulky compounds such as isohexides, terpenes, terpenoids and rosin acids can induce higher T_g compared with aliphatic and more flexible chains[162,167]. Higher epoxy content[166], higher crosslinking density[174] and group substituents and functionality[175,176] can also increase the T_g. These approaches were also used for design of recyclable thermosets with high T_g, such as digestible vanillin-based thermosets with high crosslink density and strong hydrogen bonding[172,173]. Vanillin-derived spiro diacetal has been used to fabricate a chemically recyclable epoxy thermoset with T_g of 164 °C. This thermoset had a tensile strength and modulus comparable with or better than commercial bisphenol A epoxy resins[177]. Recently, a recyclable super-engineering plastic based on isosorbide was reported[178]. The material was transparent, melt-processable and reprocessable with

Vat polymerization
A photopolymerization method in 3D printing based on light irradiation through a reservoir (vat) filled with photocurable materials.

excellent mechanical properties and an impressive T_g of 212 °C. Furthermore, in vivo and in vitro biocompatibility along with suitability for fabricating transparent flexible electric devices was demonstrated. Vanillin-based thermosets and bismaleimide epoxy resin with ultra-high T_g of 300 °C (REF.[176]) and 380 °C (REF.[179]) have also been reported, but their chemical recyclability was not investigated. Although achieving high T_g or high thermal resistance in degradable or recyclable thermosets is a challenge, introducing bio-based monomers containing rigid groups, such as lignin and terpenes, shows high promise in improving the T_g and enabling development of sustainable polymers with higher thermal resistance for specific applications.

Another approach to improving the heat resistance of sustainable polymers is to incorporate rigid segments into renewable polymers. A successful example is the high-T_g PLA prepared via ring opening of a cycloadduct derived from lactide[180,181]. The introduction of cyclopentadiene into lactide gives a bifunctional lactide derivation, resulting in a copolymer with high T_g of ~190 °C, far higher than that of PLA (~60 °C). Furan dicarboxylic acid (FDCA) can also act as a rigid monomer to increase the T_g of its copolymers[182]. It was reported that the poly(*p*-phenylene furanamide) synthesized from 2,5-furan dicarboxylic acid and aromatic diamines showed higher T_g as compared with aliphatic analogues. The T_g for these furan polyamides was as high as 280 °C (REF.[183]).

3D printing of sustainable polymers

3D printing or additive manufacturing is an eco-friendly production technique that can promote sustainability by minimizing production waste or unwanted by-products, and reducing the quantities of materials used[184]. The 3D printing market is currently increasing at a compound annual growth rate of around 20% per year and the potential applications are unlimited, from drones to spare parts for automotive products, washing machines, fashion, sports, personalized toys, education[185] and medical and biomedical products[186]. With its flexibility and diversity, this technique offers a direct pathway to sustainability through innovation and industrial viability as illustrated with examples in FIG. 6.

Biodegradable PLA is one of the most common commercial 3D printing materials due to its suitable rheological properties and slow crystallization ability making it easy to print good-quality prints[187]. Another established commercial bio-based material is polyamide 11 (PA11), which is suitable for high-performance applications. 3D printing is also ideal for recycling of plastics because it can work with small amounts of material and small series production, partially removing the need for large volumes of material with constant quality. Several companies are producing commercial filaments from factory waste streams, post-consumer plastics and even ocean plastics. Versatile and affordable, 3D printing can turn sustainable materials into innovative design or personalized products such as shoes[188], and has been used by many designers and start-ups. The first compostable and recyclable surfboard was 3D-printed with PLA/algae-based filament[189], whereas DUS Architects 3D-printed bio-based urban cabins (a micro retreat space in cities)[190]. In 2018, the Liberation Collection was presented as the first full collection of 3D-printed clothing, bags and jewellery created from biodegradable or recycled polymers[191]. Promising future applications are smart textiles incorporating electronics. For example, 3D-printed electronic textiles composed of carbon nanotubes as a conductive core and silk fibroin as a dielectric sheath could harvest biomechanical energy from human motion or be utilized as a supercapacitor for energy storage[192]. Elastic and ionically conductive 3D-printed carbon nanofibre monoliths could find applications in soft electronics, flexible sensors and thermal isolation[193]. Another innovation is 4D printing: 3D-printed products able to morph into different shapes as a response to environmental stimuli. For example, exciting PLA-based contemporary furniture, aerodynamic helmets or armour have been demonstrated[194]. The vast array of new 3D printing techniques holds endless possibilities in the (bio)medical field including slow-release drug formulations, tissue engineering such as the heart[195] and human ear[196], bioprinting of cells, sensors and many other applications[197,198].

Vat photopolymerization, such as stereolithography and digital light processing, are 3D printing techniques utilizing light-curable resins. Research is currently concentrated on the development of bio-based photocurable resins, but commercial resins are still mainly fossil based. Bio-based photocurable resins formulated by terpenes[199], natural phenols[200], vanilla[201] and various multi-acrylate monomers[202–204] extracted from biomass, waste cooking oils[205] and elastomers from bio-based poly(glycerol sebacate) (PGS)[206] have been 3D-printed via vat photopolymerization. In situ reactions initiated with different chemistries such as thiol–ene click chemistry and photo-curing allow controlling of structures and functions[207]. Adequate curing speed, continuous 3D printing technologies (FIG. 6c), bio-based contents and reuse or recyclability of the printing materials should be noted in the development of bio-renewable monomer-based 3D printing. Reported multi-acrylate monomers are often partially bio-based[202]. The incorporation of dynamic crosslinking and physical crosslinking methods such as hydrogen bonding[208] have been developed to improve the sustainability of the EoL. Dynamic crosslinking can also increase the layer adhesion, such as the thermally reversible Diels–Alder reaction in PLA to obtain tougher PLA objects[209]. In a complementary way, vitrimers and 3D printing are combined to prepare sustainable thermosets for a wide range of applications. The 3D printing of vitrimers is carried out using dynamic crosslinking which can be triggered by external stimulus such as UV, digital light or thermal energy[210]. Because of the dynamic cross links, these sustainable vitrimers exhibit reprocessability or recyclability, and at least four[210] or five[211] recycling cycles have been demonstrated.

Natural biopolymers such as DNA[212], different proteins[41,213–216], polysaccharides[196,217–220] and lignin[221] have been widely researched for bioprinting. Dissolving or melting feedstocks and maintaining control of their viscosity are critical factors for smooth realization of 3D

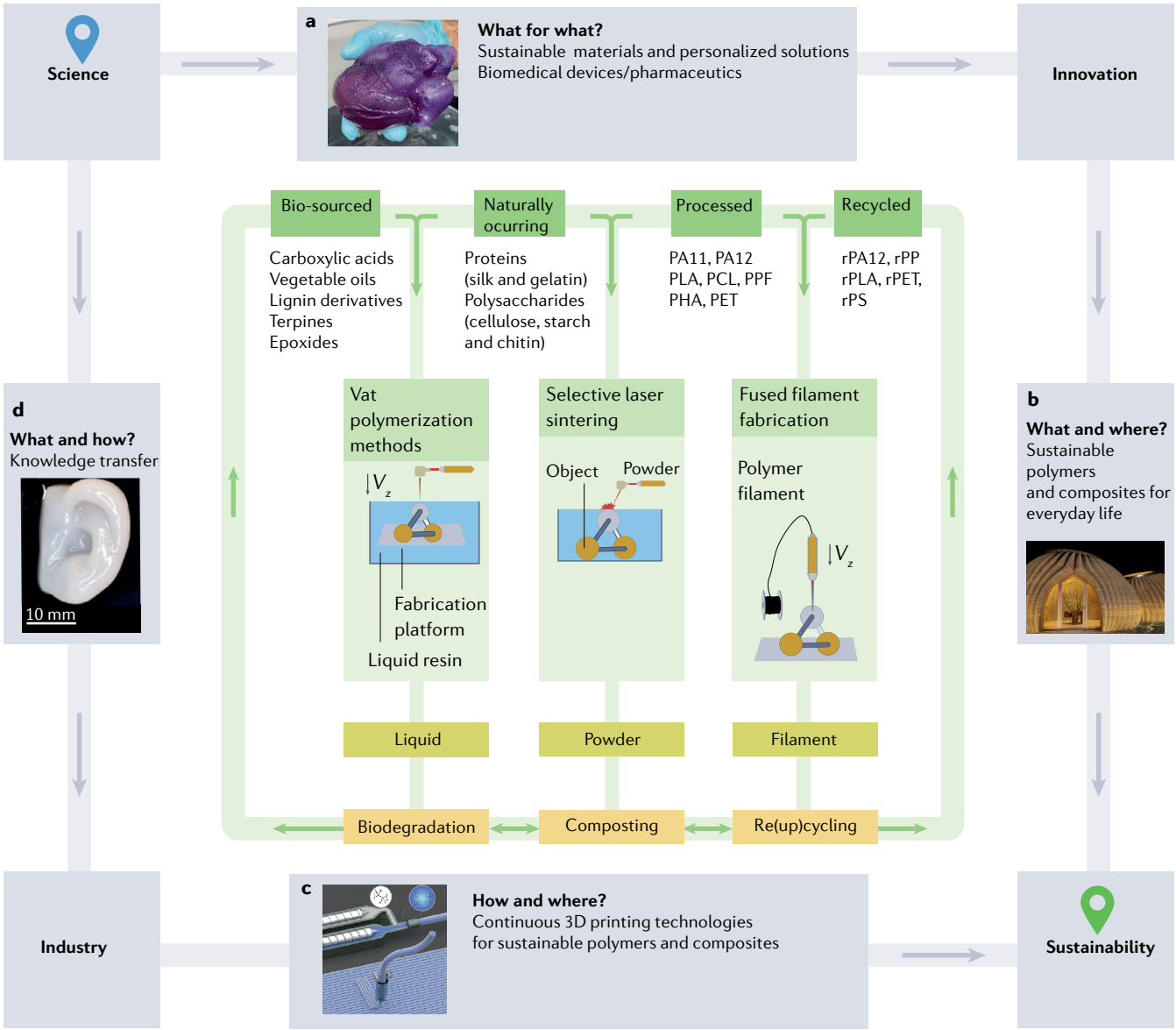


Fig. 6 | **3D printing of sustainable polymers and their applications.** Vat photopolymerization, selective laser sintering and fused filament fabrication are three main methods used to print various sustainable polymers depending on their origin (bio-sourced, natural occurring, processed or recycled), statue (liquid, powder or filament) and end of life (inner cycle). Path from 3D printing science to sustainability is inseparable from two directions. One is transferring knowledge/data to industry who fabricate 3D-printed products with continuous 3D printing technologies; the other is through innovative ideas and designs by using 3D printing technology to realize application of sustainable polymers and composites in our daily life (going from science to sustainability through industry and innovation via decision-making; outer cycle). **a**–**d** | Examples adapted, with permission: science to innovation, a 3D-printed heart (part **a**); innovation to sustainability, 3D-printed house (part **b**); industry to sustainability, continuous 3D printing technologies for sustainable polymers and composites (part **c**); and science to industry, 3D-printed shoes[188] and 3D-bioprinted human ear (part **d**). PA11, polyamide 11; PA12, polyamide 12; PCL, polycaprolactone; PET, poly(ethylene terephthalate); PHA, poly(hydroxyalkanoate); PLA, polylactide; PPF, propylene fumarate; rPA12, recycled PA12; rPET, recycled PET; rPLA, recycled PLA; rPP, recycled PP; rPS, recycled polystyrene; V_z, velocity direction. Part **a** adapted with permission from REF.[195], American Chemical Society. Part **c** adapted with permission from REF.[298], American Chemical Society. Part **d** (ear) reprinted with permission from REF.[196], American Chemical Society.

printing processes of these materials. 3D-printed food products are already in commercial production, and these renewable feedstocks also have high promise for more advanced applications such as tissue regenerative engineering medicine including cartilage[196], bone and wound healing[217]. The biggest advantage of 3D printing is the ability to create variable but precise models and controllable properties from the bottom up. This is unmatched by traditional processing methods.

Market approach

Established and emerging commercial products and applications need to be developed in line with the circular economy. Examples are given in FIG. 7 and Supplementary Table 3. Bio-based, biodegradable and/or recycled materials are already in commercial use in packaging, consumer goods, agriculture, automotive parts and textiles, although volumes are still small compared with traditional petroleum-based virgin materials[222].

Packaging and consumer goods. Flexible and rigid packaging, grocery bags, food wraps and coffee capsules are currently the largest commercial applications of bio-based polymers, including compostable and non-compostable grades (FIG. 7a). The PlantBottle made of bio-PET and launched by Coca-Cola is estimated to have reduced CO_2 emissions since 2009 in quantities equivalent to 1 million vehicles being removed from the streets[223]. Bio-based drop-in materials such as bio-PET and bio-PE preserve and protect their content comparably to their petroleum-based counterparts, whereas the emerging commercial material PEF outperforms current plastic packaging in oxygen, CO_2 and water barrier capabilities[224], but is currently cost-prohibitive for most applications.

Common biodegradable packaging alternatives such as PLA and starch blends are ideal as food packaging; however, their generally low T_g and limited heat resistance make them unsuitable for hot drink or meal applications. Total Corbion recently introduced a PLA grade with heat resistance up to 100 °C that could replace PS, PP and ABS in high heat packaging and engineering applications[225]. Many commercial fibre-based products, for example those from compressed agricultural residuals such as wheat bran and wood or vegetable fibres, have appeared during the last few years as replacements for fossil fuel-based plastic packaging and consumer products. Toys made of sustainable plastics and their packaging are high on the agenda of toy manufacturing companies. LEGO, for example, aims to use only sustainable packaging by 2025 and all LEGO bricks will be produced from either bio-based or recycled plastics by 2030 (FIG. 7d). Since 2018, some LEGO bricks are produced from sugar cane-based PE and bricks from recycled PET bottles are at a prototype stage[226,227].

In terms of EoL, bio-PE, bio-PP, bio-PET and PEF can be mechanically or chemically recycled with their fossil-based counterparts. As a relatively unique phenomenon in mechanical recycling (where polymer mixtures typically lead to deterioration of properties),

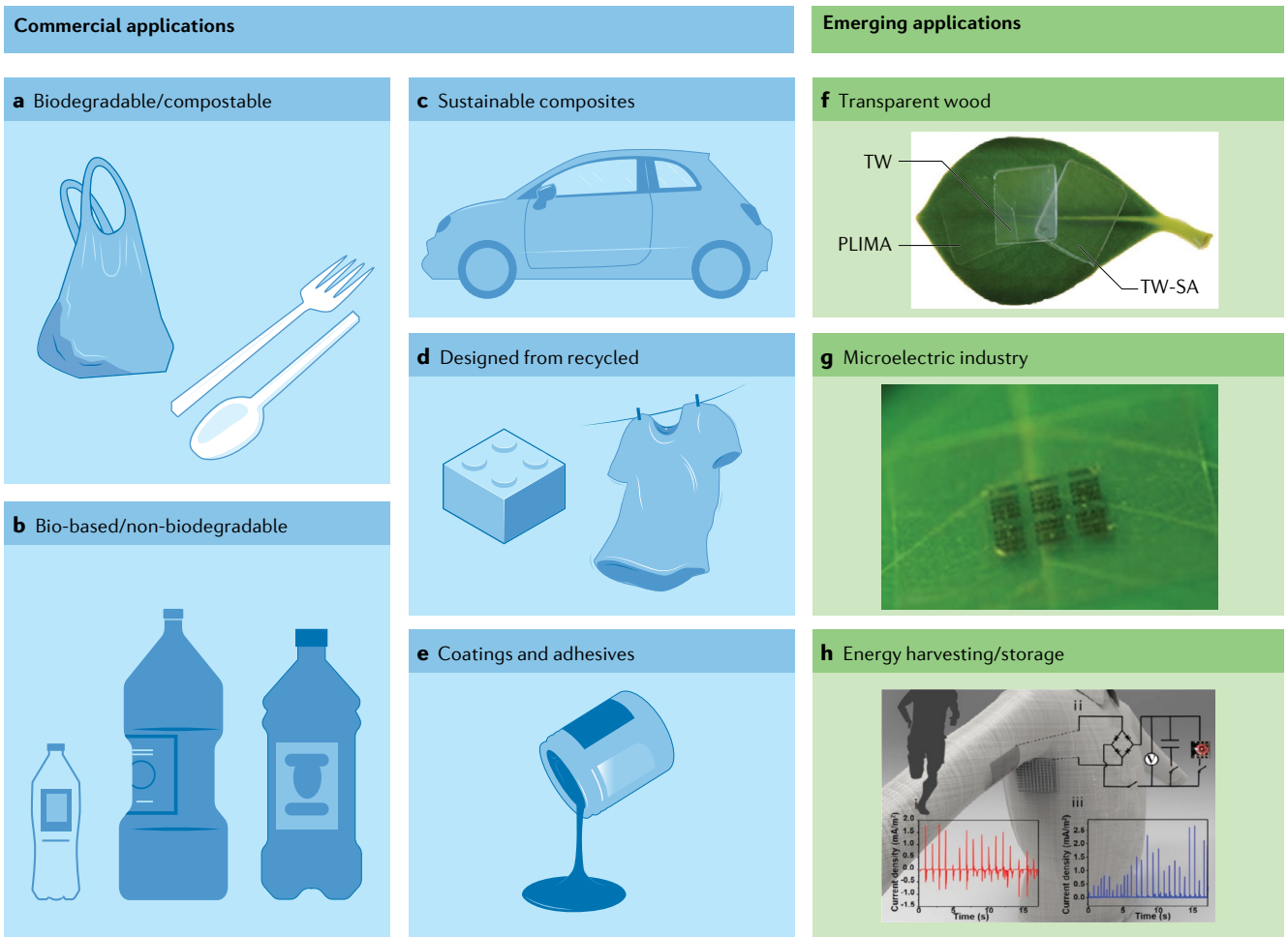


Fig. 7 | **Successful commercial and emerging application examples of sustainable polymers.** Commercial applications include biodegradable/compostable products such as shopping bags and cutlery (part **a**), bio-based but not yet biodegradable products such as bottles (part **b**), sustainable composites used in vehicles (part **c**), products designed from recycled plastics such as LEGO bricks and clothes (part **d**) and bio-based coatings and adhesives (part **e**). Some emerging (not yet common) commercial applications include fully bio-based transparent wood (part **f**), paper-based microelectronics (part **g**) and energy-harvesting/storage applications such as the printable smart pattern for multifunctional energy-management e-textile and paper-based electronic printable energy-harvesting and storage textiles based on silk fibres and carbon nanotubes (part **h**). PLIMA, poly(limonene acrylate); TW, transparent wood; TW-SA, transparent wood with succinylation. Part **f** adapted from REF.[244], CC BY 4.0 (https://creativecommons.org/licenses/by/4.0/). Part **g** reprinted from REF.[299], CC BY 4.0 (https://creativecommons.org/licenses/by/4.0/). Part **h** adapted with permission from REF.[192], Elsevier.

limited amounts of PEF entering a recycling stream of PET can improve the properties of PET[228]. For eating and drinking on the go (FIG. 7b) and other applications where packaging gets contaminated by organic matter, industrially (PLA) or industrially and home compostable (PBAT, PHA, starch, cellulose acetate) materials are ideal choices.

Agriculture and horticulture. Use of plastics in agriculture and horticulture has steadily increased due to many benefits; use of agricultural films was estimated to be 7.4 million tons in 2019 (REF.[229]). These materials are often used for short periods of time and significantly degraded or contaminated, making collection and recycling challenging. Appropriately designed biodegradable plastics are ideal for application such as mulching, greenhouse covers, plant pots, tree shelters, protective films for fruits and other related products. Several biodegradable mulching films are in commercial production. They fully degrade upon soil burial, eliminating collection and soil cleaning, while giving comparable function to linear low-density polyethylene (LLDPE) mulching[230]. Products designed from agri-residues and/or recycled plastics, such as recycled PP, have been commercialized at different levels of production and many more are under development providing more sustainable items for agriculture and horticulture.

Automotive, construction, coatings and adhesives. The automotive industry has been among the forerunners in using biocomposites, bio-based plastics and recycled plastics. The first prototype modern car made completely from bioplastics was demonstrated in 2018 (REF.[231]). Commercially, biocomposites are mainly used in car interiors (such as door and floor panels, dashboards, packaging trays and so on) (FIG. 7c), but structural components are also under development; Porsche, for example, has begun designing car bodies made of hemp composites. Biocomposites reduce weight and lower CO_2 emissions during both production and use[232]. In the European Union alone, the car industry uses 80,000 tons of wood and plant fibres each year[233]. Natural fibres are less expensive than glass fibres and offer several performance benefits. Replacing conventional polymer matrix with plant-derived options and using agriculture, forestry and post-industrial residues can further increase sustainability. Significant research efforts are dedicated to the development of more sustainable bio-based and/or recyclable thermoset composites for lightweight and high-strength construction parts. Bio-based coatings and adhesives[234–236] are under active development. The first commercial bio-based coil coating, GreenCoat, for exterior building applications was developed from rapeseed oil. The production of sustainable GreenCoat, now a large-scale commercial product, was launched in 2012 by SSAB AB (earlier Swedish Steel AB)[237] (FIG. 7e).

Sustainable textiles. The traditional textile industry, including cotton cultivation, causes significant negative environmental impacts. Bio-based and recyclable polymer fibres, such as PET and PTT, can lower these impacts[238]. New bio-based and biodegradable textiles are emerging at prototype or pilot scale. Examples are PHB-based textiles produced from biogas (Mango Materials), microbial cellulose fabrics (Nanollose) or vegan leather produced from mushroom mycelium (Mylo) or cactus leaves (Desserto)[239]. About 85% of textiles get discarded in landfill. Large-scale recycling of textiles needs to be developed particularly if fast fashion is to become more sustained. PET and PTT fibres are recyclable, but mixed fibres are challenging. Re:NewCell is a textile to textile recycling company that has developed a commercial breakthrough technology to produce Circulose, a material chosen for *Time*'s list of the 100 best inventions in 2020 (REF.[240]). The recycled fibres are already being used by major companies including Levi's and H&M to produce sustainable jeans and other items (FIG. 7d). Another commercial success is polyester fabric developed from ocean plastics by Parley Ocean plastic and Adidas. By the end of 2020, 30 million shoes had been made from the polyester fabric corresponding to 330 million PET bottles[241].

Bio-based and/or biodegradable electronics. Electronic devices have penetrated all areas of our lives, from smart packaging to air and water filters, eHealth, data and energy storage, and the internet of things. Over time, there has been a shift from durable solid devices to flexible portable devices often used for short periods of time. This creates challenges in ensuring not only their reliable function but also their biocompatibility and EoL management. Bio-based, biodegradable and/or recyclable electronics could in the future provide sustainable solutions for various disposable devices. Scientific work has demonstrated that bio-based polymers are promising materials for applications from portable sensors to monitor our health, freshness of food or environmental contaminants to portable electrochemical energy storage devices, solar cells, flexible OLEDs and electronic biomimetic skin[242] (FIG. 7g,h). 3D printing techniques are ideal for future production of bio-based energy storage materials, soft electronics and sensors[243]. Other potential commercial applications include transparent wood[244,245] (FIG. 7f), when it goes into large-scale low-cost fabrication.

Many companies have made ambitious public commitments around sustainability goals, and some have strategized to continually increase their use of sustainable bioplastics. Although they may not yet utilize 100% sustainable plastics, because of limited access, mechanical properties, durability, performance or simply because it is too costly, small steps have been taken. For example, soy-based foams where up to 25% of the polyol is derived from hydroxylated soybean oil are used commercially in mattresses, home furnishings and car seats. In 2018, IKEA began introducing recycled plastics, aiming at 100% renewable and/or recycled plastic by 2030. In addition to the bio-PET-based PlantBottle by Coca-Cola, in 2020 Dasani introduced their hybrid bottle which uses equal parts plant-based and recycled PET. In the automotive industry, great strides have been made using natural fibres to replace both mined talc and fibreglass. Ford Motor Company introduced the largest natural fibre-reinforced automotive injection moulded part in the console of the

Lincoln Continental in 2019. Although the console is not much more recyclable at EoL than the glass-filled composite it replaced, it is significantly lighter in weight, improving fuel economy, and represents a step towards more sustainable materials. More commercial examples can be found in Supplementary Table 3. These companies recognize a pull from consumers to reduce the environmental costs of petroleum-based plastics, along with the opportunity to differentiate their brand. Furthermore, these first commercial implementations at high volume will initiate future transformation of the plastics industry from conventional to new sustainable polymer technologies.

Reproducibility and data deposition

Reproducibility

In the development of sustainable polymers, reproducibility starts at the beginning of the material life cycle, with the reproducibility of feedstocks. The composition ratio of feedstocks is influenced by the growing season, collecting methods/places and storage methods during transportation and sorting; each of these affect factors such as the material's sugar content. It is important to consider this reproducibility to ensure the consistency of yield and selectivity of monomers extracted from biomass for the large-scale production of polymers. Other reproducibility issues arise from energy efficiency and recycling ability, mass/heat/quantum transformations during polymerization and EoL treatment conditions.

To ensure the reproducibility of data in sustainable polymer development, either for consistent properties or production, standard processing/characterization methods should be adopted. For example, cellulosic fibres and resins based on proteins and starches are hydrophilic because their structures contain plenty of –OH, –COOH and $–NH_2$ groups. They absorb different amounts of moisture at different relative humidity and temperature conditions, resulting in significantly varying properties with slight environmental changes. As a standard protocol, it is important to characterize mechanical, chemical and thermal properties — as per ASTM or other standards — of several sets of specimens prepared at different times and use the averages as representative values. This makes it necessary to maintain precise humidity and temperature conditions in the laboratory along with conditioning times. Other standards, such as degradation standards in different environments, should be strictly followed with blank and control groups, normally cellulose. Methods for ensuring compostability and chemical recyclability should suit the targeted product or package, and provide reliable scientific evidence that all components will integrate into the chosen process in a safe and timely manner either in an appropriate industrial facility or in a home compost pile or device. Although natural environments are never considered environmentally responsible disposal environments, the rate and extent of biodegradation is useful information for comparative analysis with other sustainable polymers, and to estimate worst-case scenarios. The fate and effect of biodegradable plastics, their incorporated additives, degradation products and fragments (microplastics and nanoplastics) on soil and marine ecosystems should be further investigated[246–249].

Data deposition in repositories

The deposition and sharing of data in the sustainable polymers field is crucial, especially for biodegradation or composting. As a result of uncertain data consistency and non-reported failed trials, numerous research projects have wrongfully concluded that some certified biodegradable plastics are non-biodegradable or non-compostable in their experimental results, or some doubtful plastics are biodegradable from mass loss or virtual observation but without respirometry measurements[250]. Data deposition related to the characterization conditions (such as compost constituent, pH, temperature and humidity) and intuitive data display (such as the dependence of CO_2 release with time) should be provided by researchers, making the data comparable between experiments. Additionally, the establishment of related databases can help to better obtain big data for collective research in sustainable polymers synthesis and processing. Requirements for data deposition take on the following format in line with the Ellen MacArthur Foundation Circularity Model[4] (BOX 2).

The use of plant-biomass feedstocks (such as renewable bio-based carbon) is an important component for the manufacturing of sustainable polymer materials. Researchers need to experimentally measure the bio-based carbon content of their polymer using radiocarbon analysis following the standards, which is more important for industrial partners as bio-contents should be accurately reported to prevent greenwashing and misleading consumers. The percent bio-based carbon content is reported based on total organic carbon or total carbon (organic + inorganic), and the calculation equations are presented in BOX 2.

For sustainable biodegradable polymers, the percent biodegradability in the targeted EoL environment should be experimentally determined. The report on biodegradability requires direct experimental demonstration of the microbial conversion of the test polymer carbon by quantifying the conversion to CO_2 (or $CO_2 + CH_4$ under anoxic conditions) using approved international standards. Such analyses may be complemented by tracing the plastic's carbon into microbial biomass using isotopically labelled polymer[133].

Recyclability for sustainable bio-based, non-biodegradable polymers should be reported. For durable industrial products and polymer composites, biodegradability and compostability are not a suitable EoL option or strategy. Therefore, a strategic road map for recovery from the waste stream and a recycling approach should be developed for these products. Experiments to demonstrate mechanical or chemical recycling should be developed and integrated into the design of the sustainable polymer. More importantly, standards should be established for plastic recycling, including their composition, their source and correct, stamped identification of the recycled products, to help consumers better distinguish between different recycled polymers, and to improve the ease and accuracy of using the correct disposal method. Such standards would speed up the consumer acceptance of recycled plastic products.

Box 2 | **Data deposition of sustainable polymers**

Beginning and end of life (EoL) of sustainable polymers
A data table should be completed.

Data table format

	Beginning of life	EoL: 90% + biodegradability in natural and managed environments		
Polymer	**% Bio-based carbon content**	**Soil (Yes/No)**	**Ocean (Yes/No)**	**Composting industrial home/community (Yes/No)**
x	x	x	x	x

Follow ASTM International/International Organization for Standardization (ISO) test methods and specification standards for biodegradability reporting.

	Beginning of life	EoL: recycling			
Polymer	**% Bio-based carbon content**	**Mechanical**	**Chemical biochemical depolymerization**	**Chemical pyrolysis gasification**	**Physical dissolution**
x	x	x	x	x	x

For recycling, check boxes with yes or no and process details: catalyst, temperature, reaction time report.

Bio-based carbon content measurement
Bio-based carbon products will retain 100% of their radioactivity from ^{14}C, whereas fossil carbons will show 0% radioactivity as they were formed millions of years ago. Comparing radioactivity of the test sample with a 100% bio-based standard product, the bio-based carbon content can be calculated:

$$\%\ \text{Biobased carbon content} = \frac{\text{Mass of biobased organic carbon}}{\text{Mass of total organic carbon}} \times 100\%$$
(Total organic carbon basis)

$$\%\ \text{Biobased carbon content} = \frac{\text{Mass of biobased organic carbon}}{\text{Mass of total (organic + inorganic) carbon}} \times 100\%$$
(Total carbon basis)

$$\%\ \text{Biogenic carbon content} = \frac{\text{Mass of biobased biogenic carbon}}{\text{Mass of total carbon}} \times 100\%$$
(Biogenic carbon = Organic + Inorganic carbon)

Establishment of a database will play a significant role in facilitating the research on sustainable polymers, such as finding suitable enzymes to degrade polymers or catalysts that promote chemical recycling. This requires researchers to share their data and continuously ensure their reliability. Recently, the Plastics Microbial Biodegradation Database (PMBD)[251] has been established, based on the National Center for Biotechnology Information (NCBI) UniProt database as well as collected and confirmed literature results. The PMBD enables the prediction of genes encoding enzymes able to degrade various plastics, although it remains under construction. Databases such as UniProt[252] should be constructed to facilitate the sharing of data.

Limitations and optimizations

Sustainable polymers are still facing challenges towards large-scale prototypes and broad commercialization. These challenges mainly result from two missing factors — lack of efficiency and unified conversion methods, and the lack of a clear understanding of the target products[45]. Because of these missing components, there is inadequate production of replacement products for commodity petrochemical polymers with required synthetic routes and performance. These two problems have been acknowledged since the start of investigations into alternative sustainable polymers, but have not been effectively addressed, even with a surplus of research investment. The products synthesized from a single technique can be diverse, leading to different monomers and polymer structures, each requiring information on atom economy and conversion prospects[45]. This divergence brings research uncertainty, including issues related to product performance and synthesis pathways. Sustainable polymers encounter two main commercialization drawbacks — the constraint of effective post-processing (recycling, reusing, biodegradation and compostability) at product EoL and the limitation of scaling up the manufacturing process and cost.

Limitation of EoL

For a sustainable polymer, the EoL considerations are critical. Beyond mechanical recycling, a higher focus is required on chemical recycling aimed towards the production of monomers and oligomers. A significant amount of research has examined the chemical recycling of plastics such as PET and PE, but limitations still exist. First, recycling needs to be cost and energy-efficient. Second, the real-life plastic waste stream is more complex and often mixed and contaminated compared with those modelled. There is also a need to develop scaling up and unifying of chemical recycling methods. Regarding biodegradation, the composting process for sustainable polymers often requires specific conditions such as high moisture, pH and temperature as well as longer degradation times (~6 months) which require additional energy. Increasing the rate of composting will reduce time and conserve energy. However, this needs to be balanced against the rate and time scales of natural biological processes. A report on nanoscopic dispersion of 2 wt% enzymes with deep active sites in enzyme-protected polymer complexes suggests that semi-crystalline PLA can be degraded primarily via chain end-mediated depolymerization in 6 days at 50 °C (REF.[134]). These are promising new approaches; however, as stated previously, conversion of the polymer carbon to CO_2 by microbial metabolism in defined EoL environments must be demonstrated. Equally important, biodegradation versus recycling (even as an energy source) needs to be delicately balanced and based on application and use[18].

Scaling up manufacturing limitation

In order to manufacture sustainable polymers on a larger scale than in a laboratory setting, existing downstream processing systems need adjusting and potentially new synthesis platforms need to be developed. Large-scale sustainable polymer manufacturing is limited by factors such as the biomass conversion rate, selectivity, catalyst efficiency, contradictions between polymerization and depolymerization (for chemical recycling) and diversity of polymers from renewable feedstocks. Although

some alternative plastics such as PLA, PHAs, bio-PET, PEF and bio-PE have been developed, production costs are much higher compared with their petroleum-based counterparts. Efficient conversion technology, either using novel catalysts or synthetic methods, deserves greater attention as a potential solution for cost issue. With the use of next-generation industrial biotechnology[253], PHA copolymers containing short-chain and medium/long chain-length monomers (abbreviated as SCL-co-MCL/LCL) were successfully obtained with a customized synthesis platform for objects[74]. An effective synthesis platform can greatly reduce the production cost of new sustainable polymers. This platform has also proved to be capable of efficient synthesis of PHA copolymers containing unsaturated fatty acid side chains, along with structural diversity for further chemical modification of PHA. Catalysts, undoubtedly, play a critical role in sustainable polymer development, both in their synthesis and EoL[75,78]. Since the invention of a Ziegler–Natta catalyst to produce PE/PP, catalysis for producing various petroleum-based polymers has experienced rapid development. The development of catalysts and manufacturing approaches for sustainable polymers will also go through a similar developing process.

To address the main limitation of scaling up sustainable polymer manufacturing with recycling and composting EoL, a multidisciplinary approach is needed, which will enable cooperation among researchers from polymer science, environmental engineering, biotechnology, computational sciences, economics and emerging manufacturing and processing technologies.

Outlook

Significant efforts have been dedicated to sustainable polymer research and development during the last two or three decades, via incorporating renewable monomers into well-defined high-performance polymers and devising innovative solutions for their biodegradation and recycling. The rapid developments are inseparable from the increasingly fundamental understanding and regulation of polymer structure and performance, judicious catalyst selection and preferred EoL options. New science developed in fast-growing versatile polymer synthetic routes, structuring of polymers, triggered depolymerization methods, computer science (high-throughput experimentation and artificial intelligence)[254], biotechnologies[255,256] and LCA have together contributed to the bank of knowledge. The goal of fully replacing conventional, petroleum-based polymers and composites with sustainable ones in all applications is still in the distant future. Some of the future research areas will include developing low-cost and multifunctional polymers that are recyclable or compostable, high-performance polymers, fibres and composites for high-temperature applications, primarily using agricultural and food processing wastes and non-edible raw materials, and facile green, no-waste and low-energy processes, among others. Close collaborations among research scientists from different fields, industrial partners, policymakers and social organizations need to be established, as well as increasing the public's awareness of Sustainable Development Goals.

In addition to the renewable feedstocks from sugars, lipids and proteins (amino acids), sources from lignin-based aromatics and CO_2 as starting materials have drawn our interest as the former can expand the applications of sustainable polymers in high-temperature applications whereas the latter is directly related to carbon footprint reduction. Concerning emerging commercial bio-based sustainable products, a recent European study screened the most important product value chains currently under development. This included large-volume biomass, low-volume high-value biomass and urban biowaste-based products[256]. Using urban waste as feedstocks for monomers or building blocks[257], value-added chemicals[258] and bio-based polymers will be one of the future focuses of sustainable polymer development (FIG. 8). The identified products included biocomposites bio-resin prepregs, carbon fibres and biopolymers from urban waste. However, when evaluating a feedstock, it is the feedstock's impact on the environment and people that matters, not solely on its generation-based classification. Focus should be on the availability of resource-efficient technologies that can convert the feedstock into a bioplastic, chemical or other material with minimal energy, water and other inputs while delivering performance, which is on par with their fossil-based equivalents. These technologies must also work at commercial scale.

Global legislative regulations, such as the circular economy plan proposed by European Union legislations in March 2020 (REF.[259]), requiring carbon neutrality will be a key stimulator to promote research innovations and market acceptance of sustainable polymers by businesses and customers. Public awareness has been awakened by visible and increasingly perceivable threat of environmental changes in our lives[260]. This will undoubtedly promote and accelerate the development of a sustainable economy. The next 5–10 years are believed to be the window for rapid development of sustainable polymers, and polymer scientists are in the best position to lead these efforts. Material circularity, industrial ecology, sustainable materials and cradle to cradle design are no longer simply buzzwords but form the new guiding principles that will lead us to the next generation of sustainable polymers.

Facing the challenges of sustainable polymer development, from the initial biomass screening to performance improvement and EoL options, the next decade of sustainable polymers will maintain focus on molecular design, catalytic technology, reducing waste discharge and promoting recycling of materials including composting, as well as the establishment of databases and information centres for data deposition and sharing. As massive amounts of plastic waste have already been accumulated environmentally and more new plastics continue to be produced, developing recycling technology to upgrade the existing legacy polymer waste into high end-value monomers, chemicals and polymers should be prioritized for the coming decade, by using either mechanical or chemical recycling. Technologically, developing sustainable polymers with performance-advantaged properties in scaling up manufacturing is what we all strive for. To maintain

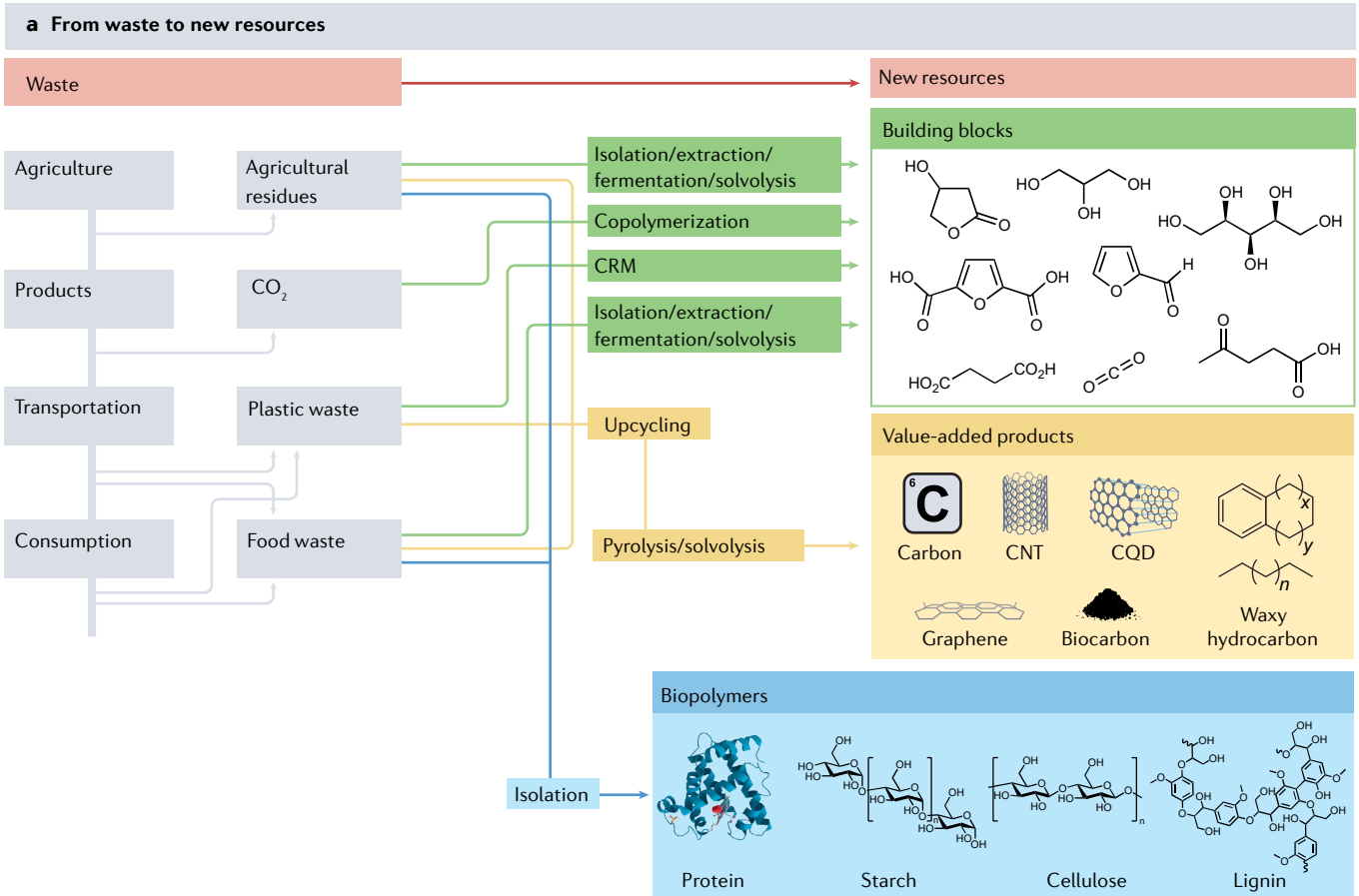


Fig. 8 | **The future of sustainable polymers.** Transforming waste to value-added resources (part **a**) and related examples (parts **b**–**e**). **a** | Feedstock choice is complex, and there is no list of sustainable or non-sustainable feedstocks. Efforts to develop novel feedstocks to become realistic at a larger scale are in progress. Different from traditional agriculture feedstocks, novel feedstocks include agriculture residues, industrial by-products, algae, captured carbon dioxide and downstream waste such as used cooking oil[300]. **b** | Upcycling of waste polyethylene (PE) to waxy hydrocarbon: upcycling conditions and catalyst used, mass percentage of recovered hydrocarbons. **c** | Building blocks — lactic acid produced from food waste via fermentation. **d** | Use of CO_2 to synthesize copolymers. **e** | Upcycling of mixed plastic waste to graphene by flash Joule heating. CQD, carbon quantum dot; CNT, carbon nanotube; CRM, chemical recycling to monomer; HDPE, high-density polyethylene. Part **b** adapted with permission from REF.[11], AAAS. Part **c** adapted with permission from REF.[257], Elsevier. Part **d** adapted with permission from REF.[50], Royal Society of Chemistry. Part **e** adapted with permission from REF.[258], American Chemical Society.

sustainability, the 3R principle — reduce, reuse and recycle — should always be a top priority.

Eco-friendliness, financial viability and social benefit are at the heart of sustainable polymers. More appropriate tools and robust databases are needed to assess the sustainability of plastics and their impact on the planet. Incentives from governments will stimulate the business growth related to bioplastics production and plastic recycling. Concurrently, several recycling and/or upcycling technologies have been set up by industrial enterprises, such as the VolCat developed for recycling PET (by IBM)[261] and IrgaCycle and ChemCycle developed for mechanical and chemical recycling of plastics (by BASF)[262]. Consumer behaviour and awareness that plastics made from waste are not different from the products made from virgin resins[263] can also promote the acceptance of sustainable polymers in our society.

Conclusions

The Age of Plastics in the 1950s, which ironically began in a bid to conserve natural resources, has brought undeniable benefits in terms of safety, energy savings and protections to the world. However, the planet is now facing massive levels of plastic pollution, adding to climate change concerns. We desperately need a second Age of Plastics that leaves a much lower footprint on our planet — rightfully called the Age of Sustainable Polymers. The journey towards research, development, scaling and implementation of more sustainable polymers is still in its infancy. The petrochemical industry is comparatively mature and has found applications for all of its high-volume refinery products, making it extremely difficult for newer, more environmentally responsible materials to compete. There is potential emerging legislation, including bans and taxes on single-use plastics to address the flood of plastic litter, especially in emerging economies such as those of Asian and African countries. Academic and industry partners willing to pay more for sustainability are experimenting with marketplace reactions to plastics with lower environmental impact, and consumer preference will play a role in adoption. Emerging sustainable materials may also have unanticipated advantages to fossil-based plastics, such as improved energy or noise absorption, so researchers must remain vigilant in searching for those enhanced properties, as these will add to their desirability over their fossil analogues. There is room for improvements in every facet of the development of sustainable polymers including the use of waste feedstocks such as lignin, CO_2 and non-edible biomass, using energy/water-efficient manufacturing directed by green chemistry principles, designing materials for environmentally responsible EoL and improving separation, reuse and recycling technologies for conventional plastics.

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