

Design and control of compostability in synthetic biopolyesters

Paresh Kumar Samantaray ^a, Alastair Little ^a, Alan M. Wemyss ^a, Eleni Iacovidou ^b,
Chaoying Wan ^{a*}

^aInternational Institute for Nanocomposites Manufacturing (IINM), WMG, University of Warwick, Coventry, CV4 7AL, United Kingdom

^bDivision of Environmental Sciences, College of Health, Medicine and Life Sciences, Brunel University London, Uxbridge, UB8 3PH, United Kingdom

*Email: Chaoying.Wan@warwick.ac.uk

ABSTRACT

The aerobic composting and anaerobic digestion of plastics is a promising route to recovering the multi-dimensional value from biodegradable single-use plastics. At present, the collection, separation, and management of biodegradable plastic waste are extremely challenging, and the majority of these plastics still end up in landfills or incineration facilities. This is because not all biodegradable plastics can be treated using organic waste management options (composting). In addition, end-users at a domestic and industrial level are often unaware of the compostability potential of biodegradable plastics, which results in the mismanagement of these types of plastic. A greater understanding of the compostability of biodegradable plastics will generate the required knowledge base for interventions that support their market penetration, use, and proper management. In this review, we clarify the concepts of biodegradability and compostability in bioplastics, in particular commercial synthetic biopolyesters, which have increasing technical and economic importance, and discuss how macromolecular design, blending and additives can be used to modify their compostability. Future trends on the uptake of compostable and biodegradable bioplastics are also commented on.

Introduction

The accumulation of plastic waste in the natural environment and the associated production of microplastics (< 5 mm in size) that contaminate air, water, and soil, have incentivized global action to tackle plastic pollution. This has stimulated the rapid development of bioplastics worldwide.¹ However, the improper disposal of bioplastic waste also has the potential to harm ecosystems, especially when intentionally littered to the natural environment due to misperceptions with regards to their biodegradability. These misperceptions may also result in bioplastics being improperly disposed of with recyclable waste, where they contaminate recyclable petrochemical-based plastics and limit their recycling potential.² Nonetheless, biodegradable bioplastics can have an important role to play in promoting sustainability, especially in applications such as single-use packaging, personal protection equipment (PPE), and agriculture applications. Recycling (as in mechanical reprocessing) alone is not an effective strategy to achieving a circular plastics economy, especially for low-value plastic

components, such as flexible films or bags, heavily contaminated plastics, or multilayered and multicomponent materials. In this regard, the substitution of problematic plastic components (e.g. flexible packaging, or packaging contaminated with food residues), with compostable bioplastics may offer the possibility of end-of-life management options that abide with circularity principles, provided that they are compatible with the existing infrastructure for the management of organic waste, i.e., they can be treated at the same conditions as organic waste streams.

Bioplastics refer to plastic of both bio-based origin and biodegradable character.³⁻⁴ Biodegradable plastics are those that are susceptible to degradation by biological activity (e.g. broken down by microorganisms, such as bacteria or fungi) accompanied by decomposition into environmentally acceptable substances with desirable properties (e.g. water, carbon dioxide (CO₂), methane (CH₄) and biomass).³ Biodegradable polyesters can be produced by microbial biosynthesis (e.g. polyhydroxyalkanoate (PHA)); produced directly from biomass (starch, lignin, cellulose, and chitosan); chemically synthesized from bio-based chemicals (PLA); or synthesized from petrochemicals (PεCL) and PVOH).³ Bio-based plastics are those synthesized using biomass-derived resources, and are not necessarily biodegradable, such as bio-polyethylene (bio-PE), bio-polyethylene terephthalate (bio-PET), and Zytel (bio-nylon). The classification of bio-based plastics and biodegradable plastics is shown in Figure 1.⁴

Many review papers have commented on the management of biodegradable plastics from a life cycle analysis perspective.⁵⁻¹¹ For example, Karamanlioglu *et al.* (2017) overviewed the synthesis, production, degradation (microbial, enzymatic, and environmental), and life cycle assessment of PLA, and summarized the PLA's degradation in composting conditions and soil.⁶ Ahmed *et al.* (2018) focused on biodegradable plastics classification and discussed the effect of exposure conditions such as pH, temperature, and moisture, on their biodegradability potential.⁸ Iwata (2015) in his review highlighted that the key challenge in biodegradable plastics is the control of the biodegradation rate.⁷ Furthermore, Albertsson *et al.* (2017) suggested that weight loss cannot be used as the only indication of degradation, and the use of correct terminology for describing the type of degradation as well as the associated testing conditions are important to avoid misunderstandings and incorrect claims that impact on the sustainable management of polymers.¹² In a recent perspective, Sander (2019) pointed out that the fate of polymers in the soil must be well understood at a mechanistic level to achieve biodegradation of mulch films.¹³

With bioplastics, production is predicted to grow by approximately 39% by 2030,¹⁴ we need to better understand the biodegradability of these materials, under aerobic and anaerobic conditions, from a molecular design and microphase morphology perspective to external environmental effects, in order to move towards a low carbon circular economy requires. In this Perspective, we unravel the key concepts and testing protocols for the biodegradation of polymers, with a focus on the intrinsic compostability of biopolyesters, including their chemical structure, composition, tacticity, crystallinity, phase morphology, and hydrophobicity; as well as extrinsic approaches, such as blending, compositing, and surface treatment. We critically discuss design strategies and their relationships with the

compostability of biopolyesters, to identify and recommend sustainable management solutions for bio-based, biodegradable polyesters. The role of nutrients, microbial consortia, and temperature are also underlined. Finally, we propose applications and future directions of compostable biopolymer packaging.

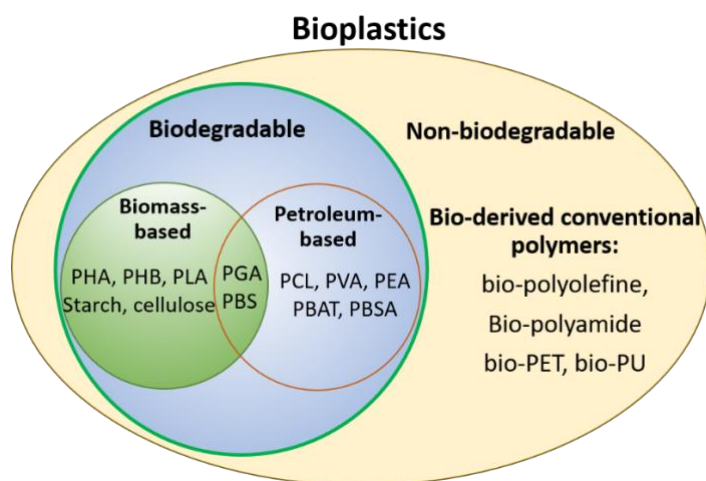


Figure 1. Classification of Bioplastics adapted with permission from Ref. ⁴ Copyright, Royal Society of Chemistry 2020.

Biodegradability and compostability: key definitions and testing protocols

Biodegradation refers to the breakdown of organic materials by microorganisms, such as bacteria and fungi.¹⁵ The process follows abiotic- and biotic-deterioration, biofragmentation, microbial assimilation, and mineralization, which eventually converts the organic carbon to CO₂ under aerobic conditions, and CH₄ under anaerobic environments (ASTM D6813). The physical breakdown of material into small fragments is classified as **disintegration** (ISO 17088). The **biodegradability** of a polymer is determined by its intrinsic molecular structure and can vary depending on environmental conditions. For example, PLLA takes decades to degrade in seawater, soil, landfills, and at home composting conditions (<37 °C),¹⁶ while it decomposes within 180 days in industrial composting facilities (>60 °C).¹⁷ P₃HB, PHBV, and P ϵ CL rapidly biodegrade in seawater, while PBS and PBAT degrade take significantly longer.¹⁸

As shown in Figure 2, the most common biodegradation process for the management of biodegradable polymers under anaerobic conditions is anaerobic digestion.¹⁹ This biodegradation process generates digestate and biogas (i.e., a mixture of CH₄ (50-65%) and CO₂ (35-50%), with small amounts of H₂S and other gases) in an oxygen-free environment. The digestate is further converted into compost via a post-treatment process, whereas the biogas is used in combined heat and power engines to produce heat and electricity, which is either used in the facility or fed into the grid.

Composting is the most common process used for the management of biodegradable waste in an aerobic environment. In comparison to anaerobic processes, aerobic composting occurs much more quickly and requires less artificial heating, since it releases much more heat. According to ASTM D6813, composting is the controlled aerobic biological decomposition of organic matter into a humus-like product called compost, CO₂, water, and heat.²⁰⁻²¹ In general, the composting process is a human-driven process where biodegradation occurs under controlled conditions involving customized mixtures of microorganisms and temperatures. It is also considered as a consecutive four-phase process that is characterized by a changing temperature pattern as follows: i) initial (mesophilic) phase (25-40 °C), ii) thermophilic phase (35-65 °C), iii) cooling (second-mesophilic) phase and iv) maturation phase.¹⁵ Temperature is a key parameter in regulating microbial activity. The optimum temperature range for open windrow (piles) composting is 40-65 °C,²² where industrial in-vessel composting operates at a temperature >60 °C, and home composting takes place in a domestic setting (ISO 5810-10, EN 13432:2000, and ASTM 4736).²³⁻²⁴

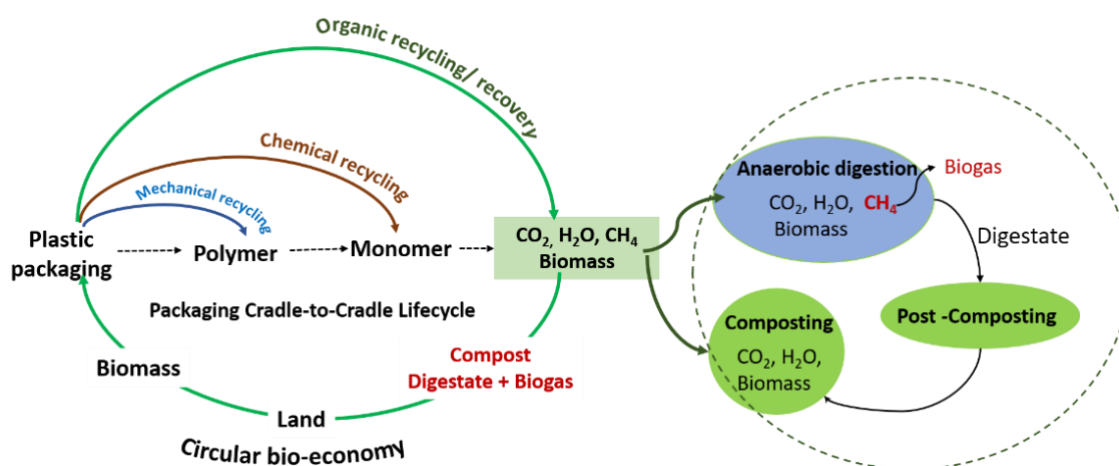


Figure 2. Organic recycling closes the plastic recycling loop

Compostable polymers according to ASTM D6400, ISO 17088, and EN 13432 need to meet three essential criteria (ASTM D6400): they must be intrinsically biodegradable (ASTM D5338); disintegrate in a composting environment without creating the risk for ecotoxicity, or any adverse effects on the final compost; as well as fully degrade to CO₂, water, and biomass without leaving traces of visually distinguishable residues.²⁵ It also must be noted that a biodegradable polymer is not regarded compostable if it leaves behind toxic residues. Moreover, both the EN13432 and ASTM D6400 require a biodegradation rate >90% of the feedstock when treated in an industrial composting facility within a period of 180 days. It is important to note that the active composting phase (thermophilic phase) is short, typically lasting from 3-6 weeks.

For assessing the biodegradation of bioplastics in anaerobic digestion, ASTM D5526 and ISO 14853 can be followed. The digestion may be carried out in either a single-phase reactor system or a

two-phase system. In the two-phase reactor, acid generation (acidogenesis) and hydrolysis occur. This acid is utilized in the second phase for methane generation, which is termed methanogenesis.²⁶ Almost 95% of the anaerobic digestion plants across Europe use single-phase reactors for the digestion of organic waste.²⁷

Understanding the differences between the biodegradability and compostability of biodegradable plastics is critical to supporting their sustainable end-of-life management. An important distinction is that biodegradable plastics can biodegrade under specific conditions and in varying periods of time – meaning that they may not always be designed to biodegrade in the natural environment, can degrade slowly, or break into microplastics. Compostable plastics are designed to biodegrade under composting conditions, which may vary from industrial to home composting, meaning that what is compostable under industrial conditions (incl. anaerobic digestion) may not biodegrade properly at home-composting conditions. However, what is home compostable can usually biodegrade under other composting conditions. It is important to emphasize that the terms ‘biodegradable’ and ‘compostable’ are often used interchangeably,¹² causing confusion and resulting in unintended negative externalities in the entire plastics value chain. Whilst, biodegradable plastics are increasingly promoted as a solution to improving plastics recycling rates via their management with organic waste, and hence promoting circularity in the sector, the biodegradable plastic alternatives that may end up in the organic waste stream, such as plastic bags, packaging, and single-use cups can be labelled as ‘compostable’, ‘biodegradable’ or ‘bio-based’. Therefore, it is very likely that a fraction of ‘biodegradable’ plastics are mixed with ‘compostable’ alternatives, and this can present challenges to composting and anaerobic digestion facilities managers as they need to ensure the quality of their compost / digestate and keep it clean from micro-plastics. As a result, all biodegradable plastics are often seen as contaminants.

To diffuse confusion with regards to ‘biodegradable’ and ‘compostable’ plastic alternatives there is now a plethora of standards developed to assess the compostability of plastics in industrial composting plants, and plastic materials, components, and products fulfilling these standards are now certified and labelled accordingly. Figure 3(a) shows the relevant standards for analyzing the biodegradability of plastics under different environmental conditions, while Figure 3(b) illustrates the key certification labels associated with such plastics, certified by different agencies for communicating biodegradation potential in different environments.

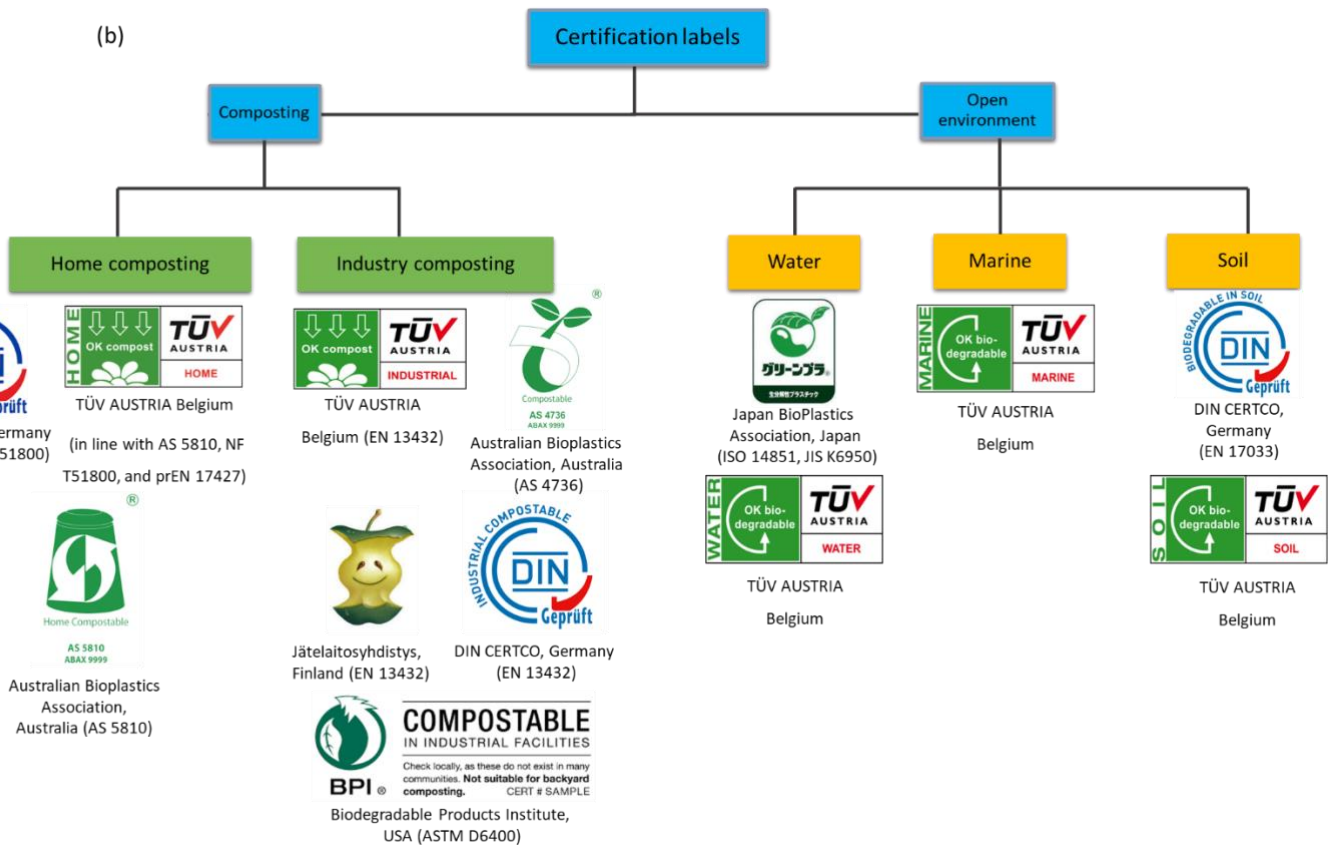
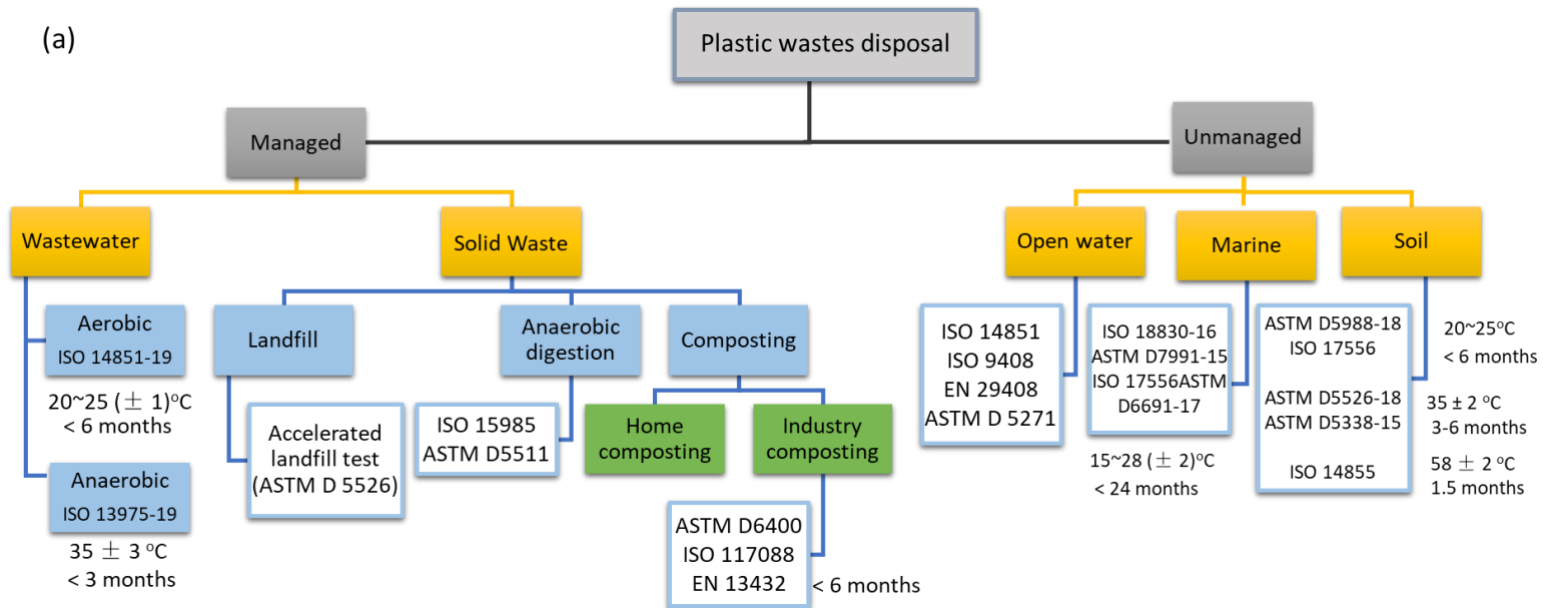


Figure 3. (a) Testing standards for biodegradability analysis of plastics under different environments relevant to plastics waste disposal. (b) Certification labels for biodegradable polymers under different environments.

Certification of plastic materials, components, and products can aid their proper management, given that the set of conditions defined by the certifier are met. In a home composting environment, however, the conditions can vary widely depending on weather, organic materials being mixed, etc., which makes industrially compostable plastic products not suitable for home composting. This is because industrially compostable plastics are designed to biodegrade under specific, controlled conditions in industrial composting plants set by the certifier. According to EN 13432 plastic packaging labelled as 'compostable' may not biodegrade in semi-controlled environments (e.g. home composters), and thus is considered unsuitable for home composting (Figure 3).²³ This points to an urgent need to address ambiguities in the labeling system used in biodegradable plastics materials, components, and products to help producers certify their products properly and to help local authorities and other stakeholders involved in the plastic system to design and/or improve upon their awareness raising campaigns that inform consumers on how to properly dispose of their packaging – be that biodegradable, compostable, home compostable or recyclable. Most importantly, it would help organic waste management facilities managers build trust in the plastic packaging waste input they receive with the organic waste stream and allow its proper management, instead of seeing this as contamination that needs to be removed.

Compostability of Polymers

Natural polymers such as starch, cellulose, and proteins generally degrade via hydrolysis in biological systems. The main chains of natural polymers often contain hydrolyzable groups, such as esters or amides, which can undergo scission in biological conditions. Polyesters containing hydrolytically labile ester linkages are susceptible to abiotic hydrolysis in the presence or absence of enzymes. Their biodegradation process starts from the hydrolysis of the labile ester linkages,²⁸ leading to chemical scission and physical erosion, in tandem to biological actions by enzymes or microorganisms such as bacteria, yeast, and fungi.²⁹

For semi-crystalline polyesters, hydrolytic degradation starts in the amorphous region as it is more prone to the diffusion of water molecules.³⁰ Crystallinity and the chain mobility of the amorphous and crystalline regions are affected by the polymers' glass transition temperature (T_g) and melting temperature (T_m). Therefore crystallinity, T_g and T_m are key factors to consider when designing biodegradable polyesters.

Common biodegradable aliphatic polyesters are shown in Figure 4. Copolymerization and blending of these polymers can lead to materials with tunable compostability. As is exemplified by the range of compostability standards given in Section 2, the degradation rates of these polymers are affected by their environment. However, polymer properties, such as molecular weight, monomeric composition, stereochemistry, and hydrophilicity can be synthetically designed and used to control physical parameters that affect compostability, such as the material's crystallinity, hygroscopicity, or T_m . In this section, we briefly discuss how polymer synthesis can be used to control biodegradability

and highlight some recent advances in synthetic polymer chemistry and materials processing that provide additional control.

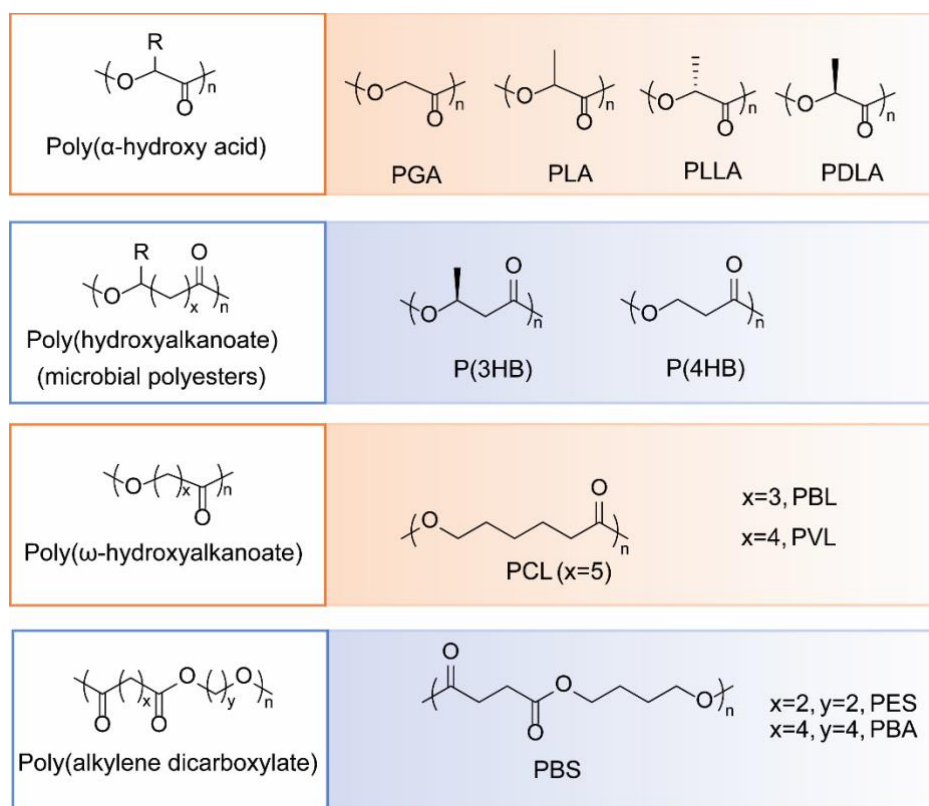


Figure 4. Examples of biodegradable and compostable aliphatic polyesters

Synthetic design of compostable biopolyesters

Compostable polyesters are typically synthesized by either polycondensation or ring-opening polymerization (ROP).³¹ In polycondensation, polyesters are formed through reactions between hydroxyl acids; diols and diacids; or diols and diesters, producing small-molecule byproducts, such as water or methanol. Long reaction times and continuous removal of these byproducts are needed to achieve high molecular weights, which come at a high energy cost due to the elevated temperatures and vacuum conditions required. The main advantage of this method is the large variety of available monomers that can be used, allowing a high degree of control over the structure, backbone, and side-chain functionalities of the products, and therefore their physical properties and composting kinetics.³²⁻³³ The synthesis of compostable materials with higher-order structures is also readily achieved through polycondensation by including multifunctional monomers in the reaction.³⁴ These cross-linked biopolymers are promising candidates as a sustainable alternative to conventional thermosets in several applications.³⁵

Using longer chain aliphatic units can increase the flexibility and lower the T_g of condensation polymers, whilst incorporating rigid units can be used to increase their strength and T_g .³⁶⁻³⁷⁻³⁸

Copolymerization can be used to introduce irregularities in the polymer structure, lowering the crystallinity and increasing the biodegradation rate. For example, the crystallinity, T_g and T_m of PBS can be lowered by copolymerizing with adipic acid to form PBSA.³⁹ As a result, PBSA has a faster biodegradation rate than PBS.⁴⁰ Similarly, incorporating aromatic monomers into a polyester can increase the rigidity and T_g of the polymer, which generally reduces biodegradability, as is observed when incorporating more terephthalic acid or 2,5-furandicarboxylic acid into PBA to form PBAT or PBAF.⁴¹⁻⁴³

Polyesters are also synthesized through the ROP of cyclic esters, such as lactide. This is the preferred method for synthesizing PLA, PGA, and P ϵ CL, as ROP is a chain-growth method that provides a significantly higher degree of control over molecular properties, such as molecular weight and polydispersity, compared with polycondensation. However, the main drawback of this method is the limited choice of monomers available, restricting the ability to tailor the molecular structure.

Tin octanoate ($\text{Sn}(\text{Oct})_2$) is established as the most commonly used catalyst for the industrial production of polyesters by ROP.⁴⁴ However, whilst having FDA approval for use in food packaging, SnOct_2 is known to be cytotoxic. In an *in vitro* study by Tanzi *et al.*, (1994) the IC50 for $\text{Sn}(\text{Oct})_2$ was found to be 125.9 ppm for human endothelial cells.⁴⁵ Thus, biodegradable polymers made using it may leave toxic residues in compost upon biodegradation. A broad range of less toxic inorganic and organic catalysts have been explored, which often provide greater control over polymerization when compared to $\text{Sn}(\text{Oct})_2$, and have been the subject of several recent reviews.⁴⁶⁻⁴⁹

ROP allows more complex polymer structures to be formed, such as high molecular weight block copolymers. A recent example of this is the synthesis of poly(ϵ -CL-*block*-LLA-*block*- ϵ -CL), which showed a reduced crystallinity compared with the poly(ϵ -CL) homopolymer, and the degradation rate of the block copolymer decreased with increasing L-lactide (LLA) block length.⁵⁰ Similarly, the degradation rate of di- and tri-block copolymers of LLA and 2-(2-hydroxyethoxy)benzoate is slower than that of their respective homopolymers, demonstrating how additional control of the biodegradation rate can be achieved through well-defined block copolymer synthesis.⁵¹

Monomer sequence and distribution also determine the degradation rate of biopolymers, which is highlighted when examining PLGA. The degradation half-life of PLA is reduced from >7 months for the homopolymer to ~1-2 weeks by copolymerization with 50% glycolic acid.⁵² However, the different reactivity rates of lactide and glycolide in ROP also play a role, as their random copolymers possess a block-like structure, with relatively long sequences of either lactide or glycolide units. This leads to three primary sites where hydrolysis occurs at different rates, with the PGA segments degrading fastest, followed by the connections between lactide and glycolide, followed by the PLA segments. Traditionally, the degradation profile of PLGA is an exponential decay. In part, this is due to autocatalysis, where the acids formed from hydrolysis of the esters accelerate the polymers' degradation.⁵³ Li and coworkers (2011) showed that an alternating polymer of poly(lactic-*alt*-glycolic

acid) had a very different degradation profile to the random copolymer, with an approximately linear decrease of molecular weight over an eight-week degradation study.⁵⁴

Comparing the compostability of common biodegradable polymers

PLLA has the slowest enzymatic degradation among all aliphatic polyesters,⁵⁵ and fungal strains can only degrade PLLA oligomers.⁵⁶ A recent study examining the bio-assimilation of its breakdown products showed that microorganisms rapidly utilized lactic acid and lactoyl lactic acid, but they were not effective in assimilating the cyclic lactide. The presence of easily assimilated low molecular weight parts facilitated the initial growth of microorganisms on the film surface.⁵⁷ For biodegradation in compost, various PLA stereo copolymers from mixtures of (D)- and (L)-lactide were exposed to a fungal protease and showed a degradation rate dependent on the (L) repeat unit content.⁵⁸ The crystalline order, the chain stereochemical composition, and the repeating unit sequence distribution affected the interactions between PLA films and *proteinase K*.⁵⁹ When the sample's characteristic crystalline dimension is reduced, the degradation rate is increased and becomes close to the amorphous sample.

PεCL can be readily degraded by bacterial and fungal strains in different environments, such as river water, soil, sewage sludge, and compost. PεCL is completely compostable at 58 °C within 14 days. Its degradation products, such as ε-caprolactone, 6-hydroxyhexanoic acid, cyclic dimers, and trimers, can be easily assimilated by composting microorganisms.⁶⁰ PεCL-starch blends showed 88% biodegradability in 44 days under aerobic conditions (ISO 14851). In comparison, PBS showed only 31% in 80 days under the same conditions.⁶¹ Under controlled composting conditions at 58 °C (ISO 14855-2), PBS degrades slower with a longer incubation time compared to PεCL.⁶² The intermediate products in PBS degradation are 1,4-butanediol and succinic acid, which are readily metabolized by microorganisms through the citric acid cycle.

The biodegradation rate of polyhydroxyalkanoates (PHAs) in the soil is of the order PHB/4HB > PHB3HHx > PHBV > PHB after 35 days at 28 °C.⁶³ PHB films remained nearly unchanged due to their higher crystallinity, while the three copolymers all showed increased crystallinity, demonstrating the fast degradation of the amorphous regions.

Under laboratory composting conditions (58 ± 2 °C, humidity 53.1%), PBSA began to fragment after 4 weeks, and PBS began to fragment after 6 weeks. These times are longer than is typical for plastics in thermophilic anaerobic plants, suggesting that these plastics are unlikely to be directly accepted by these plants at the present time. Neither of these polymers showed fragmentation after 24 weeks in soil or after 1800 h in artificial weathering conditions. The degree of crystallinity of PBS (35%) and PBSA (27%) was increased to 52% after biodegradation in compost, indicating the amorphous parts of the polymers degraded first by hydrolysis and enzymatic degradation, and that PBSA degrades faster than PBS.⁶⁴ Poly(butylene adipate-*co*-terephthalate) (PBAT) films showed the highest biodegradation rate in manure compost (67.3 ± 2.8%), as compared to those in food compost

($44.9 \pm 2.6\%$) and yard compost ($33.9 \pm 1.5\%$), by exhibiting the highest CO₂ emissions and lowest C/N ratio.⁶⁵ The possible presence of extracellular enzymes in manure and food composts may facilitate the hydrolytic reaction, as a highly reduced molecular weight is observed in these composts. Both PHBV and PBSA were rapidly biodegraded within 80 days under lab-scale composting conditions at 58 ± 1 °C.⁶⁶ In both polymers this started with enzymatic erosion at their surface, accompanied by hydrolytic chain scission induced by water diffusion into the bulk polymers. PHBV degraded faster than PBSA despite its higher molecular weight and degree of crystallinity. This was ascribed to the differences in crystal morphology and spatial organization between the polymers. Furthermore, the surface roughness and topography also affect the adhesion of microorganisms on the polymer surface, for example, PBSA has a higher roughness which hinders the colonization of microorganisms on its surface. Also, material stiffness could promote the colonization of the surface by microorganisms.⁶⁷ PBSA (290 MPa) is less stiff than PHBV (4200 MPa), which reduces the propensity of PBSA to be colonized by microorganisms.

Tuning compostability of polymers by blending and composites

The compostability of polymers can also be tuned extrinsically by blending different polymers or introducing additives. Figure 5a compares the thermal transitions in compostable polymers compared to common petrochemical plastics, and 5b shows the biodegradation rates for common compostable bioplastics in different environmental conditions.

Surface roughness, hydrophilicity, and hydrophobicity

To facilitate biodegradation under composting conditions, the microbes present in the test conditions must adhere to the bioplastics' surfaces and initiate the biodeterioration process. This is directly dependent on the hydrophilic-hydrophobic interactions and surface roughness of the polymer substrate. Microbes' primary adhesion on a polymer surface is reversible, and microbial cells can commit to growing on these surfaces by balancing the electrostatic interactions, hydrophobic interactions, and van der Waals forces.⁶⁸⁻⁷¹ In this aspect, rough surfaces favor microbial adhesion and microbial growth.^{68, 72} For the commonly used biodegradable polyesters shown in Figure 3a, the hydrophilicity order $\text{PGA} > \text{PLA} > \text{P}\epsilon\text{CL} > \text{PBS}$, indicating PGA is more susceptible to hydrolysis and degradation than the others. The introduction of hydrophilic polyethers, such as PEO, to PLA by either copolymerization,⁷³ or blending,⁷⁴ can increase its hydrophilicity, and thus increase the biodegradation rate towards that of PGA.

Polar polymers containing carboxyl, hydroxyl, or amine groups are more hydrophilic than olefin derivatives containing long hydrocarbon chains. Hydrophilic polymers have higher surface energy and hence high wettability to water molecules.⁷⁵ Surface treatment or coatings can change the hydrophobicity of the polymers, thus affecting their biodegradability. In their patent, Kroner *et al.* suggested a coating of polyacetals as a compostable film surface for diaper applications. Polyacetals hydrolyze to form acetaldehyde and diols, which are fully biodegradable under composting

conditions.⁷⁶ In a patent by Suskind, a paperboard was melt-extrusion coated with 90% PεCL and 10% LLPDE. The samples exhibited good biodegradation and a 30% reduction of the coated samples' water vapor transmission rate. This method was suitable for fabricating compostable packaging for the containment of liquids.⁷⁷ Bardi *et al.* studied the biodegradation and ecotoxicity of a coating of ultraviolet/electron beam (UV/EB)-curable printed ink on a blend of PBAT and starch films. While mineralization was reduced up to 35% when samples were cured with printed ink, no eco-toxic effects were observed for *Cucumis sativus* or *Avena sativa* plant models.⁷⁸

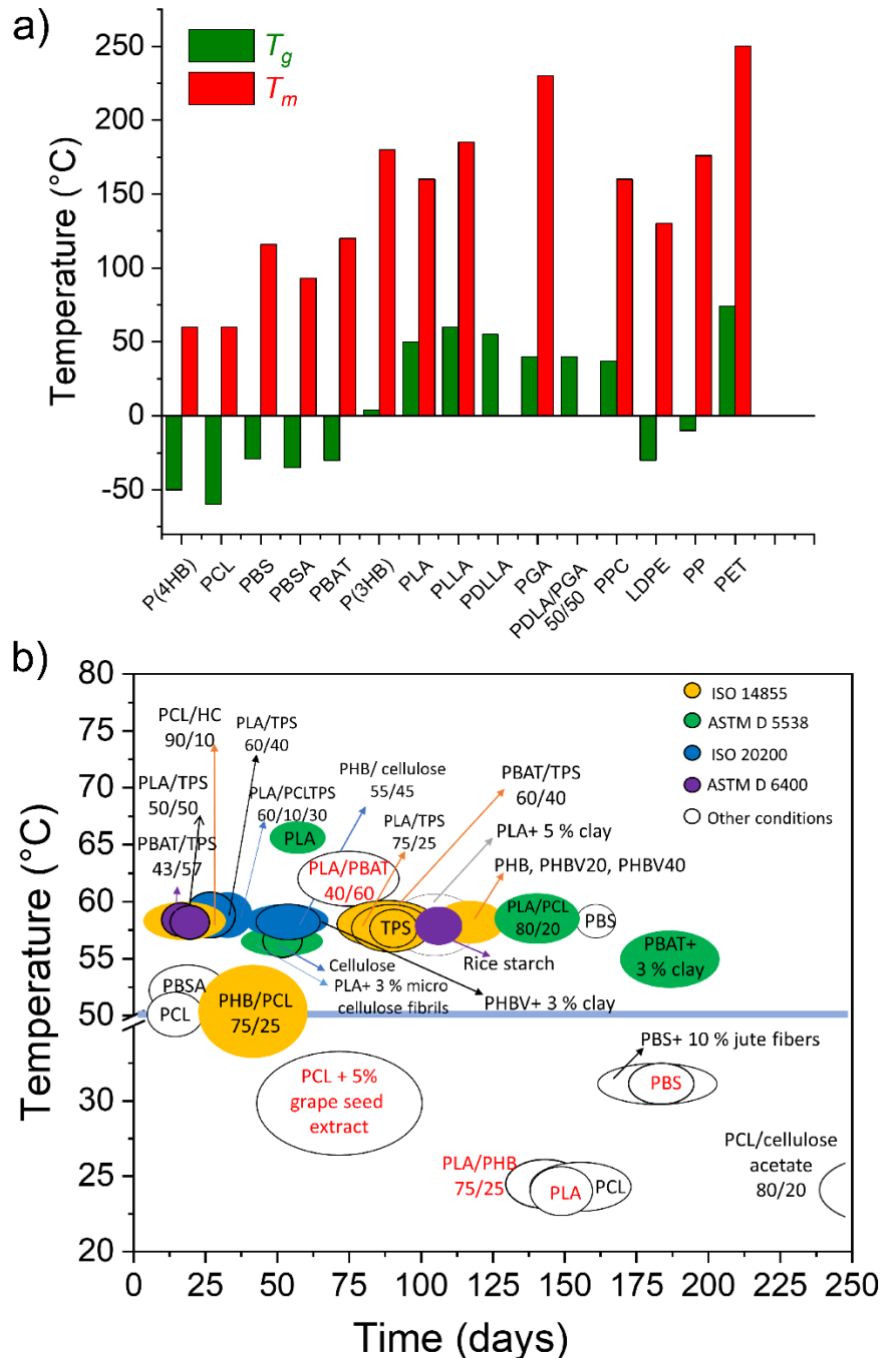


Figure 5. (a) Thermal properties of compostable polymers as compared to common petrochemical plastics; (b) biodegradation of different polymers and their blends as per different test methods. The data was taken from- PLA at 65 °C,⁷⁹ PLA/PεCL 80/20,⁸⁰ PLA/PHB 75/25,⁸¹ PLA + 3% micro cellulose fibrils,⁸² PLA + 5% clay,⁸³ PεCL at 25 °C,⁸⁴ PLA/PεCL/TPS 60/10/30,⁸⁵ PεCL/HC 90/10,⁸⁶ PBAT/PLA 60/40,⁸⁷ PBSA,⁸⁸ PBS,⁸⁹ PLA/TPS 50/50 and PBAT/TPS 43/57,⁹⁰ PBAT + 3% clay,⁹¹ PHB/PεCL 75/25,⁹² PHB, PHBV20, PHBV40,⁹³ PHB/cellulose 55/45,⁹⁴ cellulose,⁸² PHBV + 3% clay,⁹¹ PLA/TPS 60/40,⁸⁵ PBAT/TPS 60/40,⁹⁵ TPS,⁹⁶ rice starch,⁹⁷ PLA, and PLA/PHB 75/25 at 25 °C,⁹⁸ PεCL,⁹⁹ PεCL/cellulose acetate 80/20,⁹⁹ PεCL + 5% grape seed extract,¹⁰⁰ and PBS, PBS + 10% jute fibers.¹⁰¹

Blending antimicrobial agents and nanoparticles

To use biodegradable polymers for food packaging applications, antimicrobial properties are desirable to avoid spoilage of the food products. This can be achieved by coating or blending antimicrobial agents with the polymers to generate antimicrobial films. However, these surfaces or chemical modifications may alter the compostability of the final product. Further, antimicrobial agents should have a low ecotoxicity while rendering good antimicrobial activity.

Chitosan is commonly used as an antifungal and antibacterial agent in packaging,¹⁰² and it is biodegradable without any known ecotoxic impact.¹⁰³ Other potential eco-friendly alternatives include essential oil extracts derived from oregano, tea tree, clove, thyme, garlic, or rosemary, which have a broad spectrum of antibacterial and antifungal effects. Apart from the packaging of perishable food items, essential oil based antimicrobial agents are currently used in edible polymer film packaging as well.¹⁰² Antimicrobial enzymes like lysozymes are WHO, and European Union approved food preservatives used in food packaging.¹⁰⁴ These are found in different biological sources like plants, bacteria, fungi, birds, and mammals, and are not persistent, bio-accumulative, or toxic from an ecological perspective. Although lysozyme has a good activity for Gram-positive bacteria,¹⁰⁵ has shown poor antibacterial activity towards Gram-negative bacteria. However, modifications of lysozymes using sustainable approaches can improve its bactericidal efficiency.¹⁰⁴

Apart from the excellent antibacterial properties,¹⁰⁶ natural layered silicates like montmorillonite (MMT) have been widely used for improving the mechanical and barrier properties of plastic packaging. Natural MMT is highly hydrophilic, and so it is often modified with organic surfactants to improve its interfacial interactions and dispersion in polymers. In addition, polymeric compatibilizers are often used to improve the interfacial adhesion between the fillers and polymer matrices. The addition of natural MMT to biopolyesters such as PLA, PBS, and PBAT accelerated biodegradation in different environments. For example, 5 wt.% of MMT in PLA enabled a high degree of mineralization, ranging from 62% to 78% after 90 days of composting, making the PLA/MMT composites compostable.⁸³ The degradation rate of the PBAT/MMT composite was faster than those of neat PBAT and PBAT containing organically modified clay (OMMT) in an aerobic compost medium at 58 °C, as per ASTM D 5338.¹⁰⁷ The improved dispersion of OMMT may hinder the diffusion of enzymes or water,¹⁰⁸ thus slowing down the biodegradation process compared to the poorly dispersed natural clay. Similar results were observed in PBS/MMT composites during soil burial tests in a natural organic humus compost soil, 30 ± 2 °C, and 60~70% relative humidity.¹⁰⁹ After 180 days of soil burial, the total CO₂ evolution is evaluated as PBS (65.2%) > PBS/2%OMMT/PBS-g-MA (62.7%) > PBS/2%OMMT (50.2%). Therefore, the PBS and the compatibilized PBS/OMMT composites are compostable as per ASTM D 6400.

Lignin is a natural biopolymer that has good antimicrobial properties against spoilage microorganisms and can be blended with bioplastics.¹¹⁰⁻¹¹¹ In soil field tests of PHB/lignin blends, the PHB films disintegrated with a 45% weight loss within 12 months. However, the PHB/lignin blends

had only a weight loss of 12% when 10% of lignin was present. The lignin may hamper the colonization by microorganisms, possibly due to its hydrophobicity.¹¹² Other nanoparticles such as silver,¹¹³⁻¹¹⁴ graphene oxide,¹¹⁵⁻¹¹⁶ carbon nanotubes,¹¹⁷ TiO₂,¹¹⁸⁻¹¹⁹ in tandem with eco-friendly antimicrobial agents, can yield compostable polymer nanocomposites with excellent microbial resistance, while having a low ecological impact. The ecotoxicity and compostability a function of filler loading of such combinations are not fully understood in the literature. They can be a booming area of interest for superior nanocomposites in multifunctional compostable packaging applications.

Biodegradable polymers and their blends exhibit unique biodegradability and compostability characteristics under different environmental conditions. In general, if the polymer is biodegradable under a given composting condition, its blend with any other compostable polymer (in minor concentration) will also be compostable under similar conditions. For example, since PLA is suitable for industrial composting, all PLA blends with PLA being the matrix will exhibit >60% biodegradation under industrial composting. Figure 5b consolidates the biodegradation behavior of common biodegradable polymers and their blends under different test conditions from the existing literature. Each color signifies the standard adopted in those papers and the polymers or blends highlighted in red indicate that the biodegradation was less than 60%. (e.g. PLA in room temperature didn't show any biodegradation and blends of PLA/PHB 75/25 showed biodegradation of ~1% in room temperature conditions). From the author's perspective, polymer blends which can be composted under ambient conditions using home composting are ideal for the current packaging requirements. They will serve dual roles; provide the required mechanical and barrier properties, and close the recycling loop via its proper management. Emerging polymers like poly(glycolic acid) (PGA) and its copolymers can biodegrade in ambient conditions⁴ and their blends can have positive impacts in all-compostable packaging applications.

Effect of nutrients, microbial diversity, and type of soil on compostability

The composting of bioplastics is directly influenced by the nutrients present in the soil, consortia of microorganisms, the type of soil, and the temperature parameters used. Ryckeboer *et al.* (2003) defined composting as a self-heating and aerobic biodegradation process in solid-phase under controlled conditions, distinct from natural rotting or putrefaction.¹²⁰ The self-heating results from cellular metabolism in which 1.0 mol of aqueous glucose derivative in aerobic conditions produces 677 kcal/mol of heat.¹²⁰ This evolved heat is also dependent on the type of polymer surface, the soil's relative humidity, and oxygen content.

In the composting process of polymers, organic substrates and compost material provide the carbon compounds with microbial regularity and soil growth. The yield coefficient (amount of carbon incorporated in microbial cells per unit of degraded carbon source) ranges between 10~35% and micronutrients like nitrogen, phosphorous, and potassium play a critical role in microbial growth and hence on the compostability of the polymers.¹²⁰ In limited nitrogen conditions, the composting process

is slowed down, and if an excess is supplied, nitrogen is lost in the form of ammonia gas or via nitrate leaching. The optimum C/N ratio is in the range of 35-45 for composting.¹²⁰ Optimum relative humidity and moisture are also required to achieve compostability. If the moisture is below 30%, the microbial activity decreases, and the organisms enter a dormant phase, while when the moisture content is too high (> 65%), oxygen depletion may occur, and nutrients may leach out from the soil, which is undesirable.¹²⁰

It is also essential to monitor the soil parameters to understand their correlation with composting. ASTM D2974 helps determine moisture, ash, and organic matter content in peats and organic soils. (For mineral soils and rock ASTM D2216 is to be followed.) Different microorganism classes like bacteria, fungi, and actinomycetes have been known to aid in the biodegradation of polymers under composting conditions. The commonly studied fungi for PLA include *Fusarium moniliforme*, *Aspergillus fumigates*, and *Thermomyces lanuginosus*.¹²¹ For PVOH, *Aspergillus*, *Phanerochaete chrysosporium*, *Fusarium*, *Trichosporon laibachii*, *Galactomyces geotrichum*, *Fusarium oxysporum*, and *Fimetariella rabenhorsti* has shown promising results.¹²² Fungal species like *Aspergillus flavus*, *Aspergillus niger*, *Fusarium solani*, *Aspergillus fumigatus*, *Chaetomium globosum*, and *Penicillium funiculosum* have shown promising results in the biodegradation of PεCL.¹²³ Only a few enzymes have been isolated for degrading PLA, such as Pronase, Proteinase K, Bromelain, and some esterase-type enzymes.

Nishide *et al.* (1999) investigated the biodegradation behavior of four compostable polymers, PHBV, PεCL, PBS, and PBSA in the soil at different temperatures.⁸⁸ It was observed that PHBV underwent faster degradation at a lower temperature (30 °C) in aerobic conditions while there were no significant differences in the degradation rate of PBSA, PεCL, and PBS under the same conditions. Comparatively, PHBV had the fastest degradation at 30 °C while PBSA had the fastest at 52 °C. Interestingly, all four bioplastics had no degradation in anaerobic conditions after 50 days. Although biodegradation is primarily achieved in polyesters in the presence of bacteria, it was observed that with a fungicide like Daconil (chlorothalonil-2,4,5,6-tetrachloroisophthalonitrile), the biodegradation of all four polymers was suppressed. This suggests that in addition to bacteria, fungi and actinomycetes play a dominant role in biodegradation in soil. The fungal strains which were able to degrade these plastics were also identified. At 30 °C, the fungal strains which were able to degrade the bioplastics were *Mucor sp.* (PHBV), *Aspergillus sp.*, *Cunninghamella sp.* (PBSA), and *Paecilomyces sp.* (PεCL). At 52 °C, the fungal strain isolated from PεCL and PBSA was *Thermomyces sp.*

Outlook and future perspective

Bio-based and biodegradable plastics are terms that are often used interchangeably in the global literature, as well as by the public. This inaccurate use of terminology has created difficulties in the management of bio-based plastics, and particularly biodegradable plastic wastes, which has a negative impact on their sustainability potential. As a result, there is a need to clearly distinguish these terms at

a cross-sectoral level, and moreover, it is important to emphasize that biodegradation may occur in varying degrees and in many ways. Particularly, for biodegradable plastics, their biodegradability depends on their molecular structure and is directly linked to the conditions and standards pertaining to waste management practices. Even though there is a certification system well-aligned with the standards for the management of biodegradable plastic waste, there is still bad practice at the production and consumption stage, which affects the management stage. Whilst ‘biodegradable’ and ‘compostable’ plastics may be technically recyclable, they are currently treated as contamination in the recycling of organic waste streams. Increasing awareness and clear communication in regards to the use of the labels that would enhance its proper disposal could make the recycling of compostable plastics feasible in the future, promoting circularity in the sector. With the bio-waste separate collection being increasingly promoted for moving towards a sustainable bio-waste economy, there is a clear opportunity for integrating the biodegradable plastics into the bio-waste management system; creating synergies between production and waste management sectors. In this review we critically examined the biodegradability and compostability potential of common biodegradable polymers in different environments and discussed the various methods of tuning the biodegradability potential of plastics through chemical synthesis, blending, and addition of additives.

At present packaging applications remain one of the largest market segments of bioplastics, contributing to 53% of the global market production of bioplastics produced in 2019.¹²⁴ Around 62% of the bioplastics produced are biodegradable. Making these biodegradable plastic packages certified compostable plastic packages, especially in the food packaging sector, presents a unique opportunity to deal with the dual problem of plastics contamination with food residues (that often causes their rejection at sorting facilities), and improper food waste management. Replacing conventional plastics that often contain food (e.g. yogurt pots, margarine tubs, salad bags), or are attached to food (e.g. fruit stickers or tea bags), with compostable plastic alternatives can promote their synergistic management of two problematic waste streams, and support the move towards a sustainable plastics and bio-waste management. Besides their use in the packaging sector, bioplastic use in the agriculture and horticulture sectors has expanded tremendously. Mulch films are mainly made of non-biodegradable LLDPE and LDPE, which may disintegrate in the field and contaminate the soil. Currently, the collected waste mulch films are still disposed of in landfills or treated in incineration facilities (with and without energy recovery).¹²⁵ In this regard, compostable polymers with comparable and even better physical properties than LDPE will have the potential to revolutionize plastic use in the agriculture sector. Using non-ecotoxic additives and fillers in biodegradable polymers can yield alternative mulch films that can be composted after use. However, it is critical to note that some natural additives may have eco-toxic impacts.¹²⁶ Hence, monitoring the ecotoxicity potential during and after the biodegradable plastic end-use is a key criterion in the consideration of compostable polymers and their composites in packaging applications. Moreover, compostable polymers and nanocomposites can be modified to deliver essential fertilizer micro-nutrients to the soil during and after its end-use.

Compostable polymers will also bring new functions to other areas. Additive manufacturing has emerged as a new tool to optimize material usage with superior performance. Biodegradable polymers such as PLA, PBAT, PVOH, and PHA have been used as filaments in 3D printing due to their competitive physical properties to conventional petroleum-based polymers.¹²⁷⁻¹³⁰ Compostable PLA/PBAT blends are used for cosmetic media packaging and retained excellent stability during aging in cosmetic media.¹³¹⁻¹³² PLA, PBAT, PBS, and PBSA based compostable covers and cases were designed for smartphones and other electronic gadgets, including laptops.¹³³ This is particularly useful for the fabrication of organic electronic devices that are non-toxic, biocompatible, and biometabolizable.¹³⁴⁻¹³⁵ Biodegradable polymers and their blends can also seek critical applications in sustainable water treatment applications, to replace the non-biodegradable membrane materials.¹³⁶⁻¹³⁸ By tailoring the degradation and water stability, these membranes can help to reduce the carbon footprint of water treatment materials.

Moving forward, the development of new polymer chemistry and macromolecular design approaches, improved recycling infrastructures and consumer awareness, standardized regulations, and policies will help the application and implementation of biodegradable and compostable polymers in a range of areas. These potential applications include, but are not limited to, aerospace industry, automotive, high-temperature packaging, bioelectronic sensors and actuators, additive manufacturing, drug delivery and tissue engineering, clothing, air purification, and water treatment. A clear understanding of the functional design and biodegradation behavior of biodegradable bioplastics will not only revolutionize plastic consumption but will also begin to reverse the impact that humans have had on the ecosystem.

Conflicts of interest

There are no conflicts to declare.

Keywords: bioplastics; biodegradable; compostable; biopolyesters; sustainable

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Biographical

Dr Paresh Kumar Samantaray obtained his B. Tech in Plastics Engineering from CIPET, Bhubaneswar. He pursued his MSc. Eng. and Ph.D. from the Indian Institute of Science under Prof. Giridhar Madras and Dr Suryasarathi Bose. He is a Postdoctoral Fellow working with Dr Chaoying Wan at the International Institute for Nanocomposites Manufacturing (IINM), WMG. His current research interest includes the synthesis and processing of biodegradable polymers and nanocomposites for flexible packaging. He holds adept experience in designing antibacterial polymers and nanomaterials for photocatalysis, desalination, and heavy metal removal.



Alastair Little received an MChem from the University of York in 2019. He is now a PhD student at the University of Warwick supervised by Dr Chaoying Wan and Professor David Haddleton. His research focuses on molecular design, synthesis and processing of biodegradable polymers for packaging applications.



Dr Alan M. Wemyss is a Research Fellow at the International Institute for Nanocomposites Manufacturing (IINM), WMG, University of Warwick. He received his BSc (2009) from the University of Glasgow and PhD (2016) from the University of Warwick, UK. His current research focusses on the design and synthesis of sustainable thermoset materials for a range of applications.



Dr Eleni Iacovidou is a Lecturer in Environmental Management at Brunel University London, leading teaching on environmental governance, management and sustainable development. Her work focuses on performing holistic and integrated sustainability assessments of resources and waste management systems using a system-of-systems approach. She has extensive experience working with both policy and industry



stakeholders on resource recovery projects and initiatives across the UK. She has a background in chemistry, with expertise on environmental engineering and environmental management research following studies at the Department of Chemistry, University of Crete and the Centre for Environmental Policy, Imperial College London.

Dr Chaoying Wan received her PhD degree in Materials Science from Shanghai Jiao Tong University. She was awarded a Marie Curie Individual Fellowship and worked at Trinity College Dublin, Ireland. She joined University of Warwick in 2012, where she is currently a Reader in multifunctional nanocomposites. She specializes in polymer synthesis, characterization and reactive processing of multiphase/multicomponent nanocomposites, smart dielectric elastomers, biodegradable polymers and sustainable rubbers.

