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Recent advances in biodegradable polymer blends and their biocomposites: a comprehensive review

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The growing environmental concerns over plastic pollution and sustainability have led to increased interest in biodegradable polymers as alternatives to conventional plastics. This concern has led to the United Nations resolution of March 2022 calling for urgent action to eradicate plastic pollution globally by 2040 as more than 90% of the global plastic production from 2018 to 2022 was fossil-based, significantly contributing to plastic pollution. In response, there has been a growing shift towards sustainable materials, with biodegradable polymers emerging as a critical solution to mitigate the environmental impacts. However, the properties of biodegradable polymers are at variance with conventional fossil-based plastics in many applications. One way to solve this problem is to re-engineer their properties through polymer blending, a strategy that combines the properties of two or more polymers, aided by compatibilization to improve polymer miscibility and properties. While numerous reviews have focused on biodegradable polymer blends, this article offers a unique contribution by comprehensively examining both biodegradable polymer blends and their reinforced biocomposites within a single review, an area that has seen limited coverage in recent years. This review discusses recent advancements in biodegradable polymer blends and reinforced biocomposites, focusing on material properties, compatibilization techniques, and environmental impact. Key biodegradable polymer blends and reinforced biocomposites based on polylactic acid (PLA), polyhydroxyalkanoates (PHAs), polybutylene succinate (PBS), Polybutylene adipate terephthalate (PBAT), and thermoplastic starch (TPS) are discussed, with a focus on miscibility, compatibilization and the effects on properties. It was found that compatibilizers such as maleic anhydride, dicumyl peroxide, and Joncryl play significant roles in polymer blend miscibility kinetics and compatibility while fillers such as turmeric, cinnamon, coffee ground powder, and rice straw have contributed to improving the mechanical properties and biodegradability of composites. This combined approach of blending and filler reinforcement represents a critical innovation for producing high-performance biodegradable materials. The review examines applications in packaging, agriculture, and biomedical fields, along with the environmental impacts of these materials, such as their biodegradation pathways and ecotoxicity. Lastly, the review discusses future outlooks, including potential breakthroughs and integrating biodegradable polymers into the circular economy.

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1. The manuscript discusses recent developments in biodegradable polymer blends and their biocomposites, focusing on improving mechanical, thermal, and biodegradation properties. Innovations such as nanocomposite reinforcement, compatibilization techniques, and blending biodegradable polymers with bio-based fillers enhance material performance, enhancing environmental sustainability.
2. With increasing global plastic waste concerns, biodegradable polymer blends offer sustainable alternatives to fossil-based plastics. This review is significant for industries like packaging, agriculture, and biomedicine, addressing limitations of biodegradable polymers while enhancing their processability and eco-friendliness. It aligns with circular economy principles and global policies on pollution reduction.
3. Future research will explore advanced compatibilizers, reinforcements, and process optimization for biodegradable blends. This study provides critical insights into designing sustainable materials, helping shape green chemistry by fostering innovation in eco-friendly polymer development.

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1. Introduction

The increasing urgency to address global plastic pollution and environmental sustainability has brought biodegradable polymers to the forefront of materials research and innovation. These polymers, derived from renewable sources, offer the potential to reduce dependence on fossil-based plastics and their associated ecological impact. However, their performance limitations in terms of mechanical strength, thermal stability, and processability restrict their broader application. Addressing these limitations through polymer blending and reinforcement with biocompatible fillers has opened new avenues in biodegradable material development.

The aim of this review article is to discuss the recent advancements in biodegradable polymer blends and reinforced biocomposites, focusing on miscibility, compatibilization, and their effects on properties. The review also examines biodegradation and ecotoxicity of blends and their biocomposites, along with their applications in food packaging, agriculture, and biomedical fields. Lastly, the review discusses the environmental impact assessment and integration of biodegradable polymers into the circular economy. While numerous reviews have focused on biodegradable polymer blends, this article offers a unique contribution by comprehensively examining both biodegradable polymer blends and their reinforced composites within a single review, an area that has seen limited coverage in the last five years (2019–2024).

The review article is structured as follows:

Section 2 gives the general background, section 3 introduces the fundamentals of biodegradable polymer blending, including key design considerations, section 4 discusses recent developments in blends based on PLA, PBAT, PHAs, TPS, PBS, and related systems, emphasizing miscibility and compatibility. This section also discusses the role played by natural and inorganic fillers in enhancing blend performance for the above blend systems. Section 5 examines the biodegradation behaviour of blends and composites, and section 6 covers application areas such as biomedical, packaging, and agriculture. The environmental impact assessment is discussed in section 7, section 8 presents the summary of key findings, while the last section presents the outlined future directions, including integration into circular economy frameworks.

2. General background

Biodegradable polymers are continuously in the research spotlight owing to their perceived benefits compared with non-biodegradable polymers. Growing environmental awareness has driven a shift towards developing biodegradable materials from renewable sources to replace conventional, non-biodegradable plastics, especially in packaging.[1–4] The production of bio-degradable polymers is still low compared with fossil-based polymers; however, increasing awareness is driving global production, with significant increase in the last 5 years (Fig. 1). According to the 2023 Plastics Europe report,[5] global plastic production grew from about 370 million tons (Mt) in 2018 to 400 Mt in 2022. Of these, fossil-based plastic grew from 339.4 Mt to 382, accounting for more than 90% of the global production and bio-based plastics grew from 1.1 Mt to 2.3 Mt, accounting for about 0.5% by 2022[6] (Fig. 1a and b). The plastics industry is increasingly interested in biopolymers, which can reduce carbon emissions by substituting fossil-based polymers.[7,8] Hence there is a need to increase the production of bio-based polymers to replace fossil-based plastic in different applications. While bio-based polymers are environmentally friendly, their higher cost and performance limitations, such as brittleness and low mechanical performance, have hindered their widespread adoption.[9] Consequently, the scientific community is focusing on tailoring their properties to performance and market demands.[10,11] One key approach is to re-engineer their properties through a blend of biodegradable polymers and sometimes other materials to enhance their properties which have been demonstrated in many polymer blends.[12–14] Blends of biodegradable polymers are gradually gaining attention as a critical approach for addressing the drawbacks of single biodegradable polymers, especially in areas such as mechanical properties, processability, and degradation rates. For instance, the blend of a slow-degrading polymer with a fast-degrading one can led to a material that degrades at a rate that benefits the target application.[15,16] Blending offers a cost-effective and efficient way to tailor characteristics while adhering to low carbon footprint regulations.[17,18] By combining two or more biodegradable polymers, scientists can create materials that possess a balance of desired characteristics that single polymers may lack.[19] For example, polylactic acid (PLA), which is known for its mechanical strength and transparency, is often brittle and lacks flexibility. The incorporation of flexible polymers like polyhydroxyalkanoates (PHAs) and polybutylene adipate terephthalate (PBAT) can enhance the flexibility of the material without compromising its strength, making it suitable for a wider range of applications. Blending biodegradable polymers with non-degradable polymers enhances mechanical and functional properties while introducing partial biodegradability, reducing environmental impact compared with entirely non-degradable materials. Additionally, in biodegradable polymer blends, the degradation rates can be controlled especially in applications such as medical implants or agricultural films where the material's lifetime is a factor to consider. This ability to control properties through blending makes biodegradable polymer blends a critical component in the development of sustainable materials.

Also, the need to blend biodegradable polymers arises from the necessity to enhance processability and cost reduction. However, certain biodegradable polymers, although they have good properties, have some drawbacks in terms of processing. For example, they may need low temperatures, have short processing windows, or are difficult to form. When combined with polymers that have improved processing properties, the overall processing of these materials can be made easier and

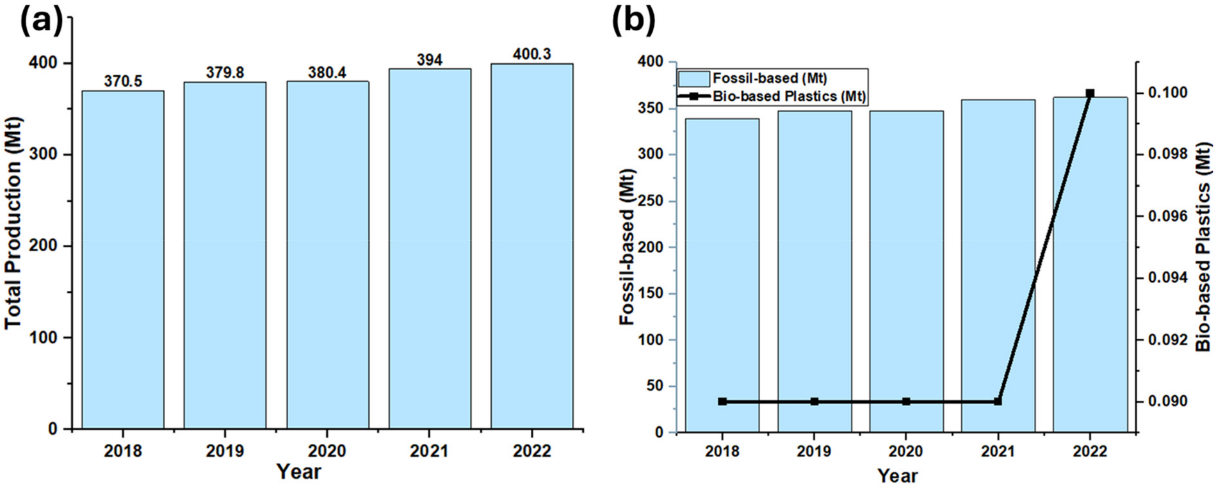


Fig. 1 (a) Global plastic production from 2018 to 2022, (b) global plastic production by polymer type from 2018 to 2022. Data adapted from Plastics Europe 2023 report https://plasticseurope.org/knowledge-hub/plastics-the-fast-facts-2023/ accessed January 4, 2025.

cheaper. Blending biodegradable polymers to make binary, ternary, quaternary blends can balance cost, processability, and performance resulting from the complementary properties of each polymer.[20] These blends can achieve specific mechanical and functional properties such as rigidity and modulus from PHAs, PHBV, and PLA, strength from PLA and PBS, impact strength from PBAT and PCL, and elongation at break from PBS, PBAT, and PCL.[21] Furthermore, these polymers also offer unique advantages, such as being renewable resource-based (PHAs, PLA, PBS), having high heat-deflection temperatures (PHAs, PBS, PHB, PHBV), good flowability (PHB, PHBV), and low cost (PLA, PBAT).[21]

Blending different polymers does have its own drawbacks, including polymer miscibility issues and the need to match the compatibility of polymers to prevent phase separation, which in turn leads to poor performance of the material.[22] Despite these challenges, the advantages of biodegradable polymer blends make them an important development in the pursuit of reducing environmental impact while maintaining material performance. The primary objective is to develop biodegradable materials that can effectively replace traditional plastics in a wide range of applications, such as packaging and medical devices, while also maintaining environmental sustainability.[23]

To address these challenges, recent advancements in biodegradable polymer blends have introduced innovative strategies that enhance material performance and sustainability. A key development is the incorporation of nanocomposites and additives like nanocellulose and nanoclays, which improve mechanical strength, barrier properties, and thermal stability, making these blends more suitable for applications like food packaging and medical devices.[24–27] Compatibilizers have also been introduced to enhance the miscibility of different polymers, addressing the issue of phase separation and resulting in materials with better mechanical properties and consistent biodegradation.[28,29] Table 1 show a summary of different compatibilizers that are commonly used in polymer blends. Additionally, there is a growing focus on fully bio-based polymer blends, combining renewable resources like PLA and polyhydroxyalkanoates (PHA) with natural materials such as starch, cellulose, and natural fibers. These blends not only lower the carbon footprint but also offer enhanced biodegradability, making them ideal for packaging and agricultural uses. In the biomedical field, blends like PLA and polycaprolactone (PCL) are being tailored for specific therapeutic uses in scaffolds, drug delivery systems and tissue engineering where controlled degradation is crucial.[30–32] Moreover, the packaging industry has seen the development of biodegradable blends with improved barrier properties and the ability to interact with packaged products to extend shelf life. These innovations reflect a broader commitment to sustainability in materials science, with ongoing research likely to yield even more sophisticated blends that balance functionality with environmental friendliness.

2.1 Biodegradable polymer blends

Generally, polymers are classified as biobased and fossil or petroleum based. Biodegradable polymers can be biobased, and fossil based. Clearly, not all biobased polymers are biodegradable, and not all biodegradable polymers are biobased. The classification of polymers based on origin and biodegradability is presented in Fig. 2, and Table 2 gives the chemical structures and composition of common biodegradable polymers. Biodegradable polymer blends are a promising class of materials with various applications, including in the biomedical and packaging industries. Biodegradable polymer blends are two or more biodegradable polymers mixed to yield a material with tailored properties such as high strength, improved toughness and ductility, controlled degradation rate, and thermal stability without sacrificing their biodegradability. The process of polymer blending can be done with or without a chemical interaction between the individual polymers.[33] Biodegradable blends offer a likely new frontier in the search for sustainable materials. They offer special properties, including high perform-

Table 1 Various compatibilizers, their pros, cons and recommended condition for use

Compatibilizer	Pros	Cons	Recommended use conditions
Maleic anhydride (MA)	Improves interfacial adhesion and compatibility[46,47]	Requires careful dosing[47]	PE, PP, and biocomposite; blends with polar fillers[46]
Dicumyl Peroxide (DCP)	Initiates grafting, enhances crosslinking[48,49]	Over-crosslinking risk; odor and safety issues[50]	Peroxide-initiated grafting[49]
Joncryl® Series	Chain extension and compatibilization in polyesters[51,52]		PLA/PBAT or PET blends; chain-extending, toughening[51,52]
Epoxidized Soybean Oil (ESO)	Bio-based, improves flexibility, epoxide-reactive[55–57]	Over-plasticization/oil blooming,[53] limited thermal stability at high temperature (above 240 °C)[54]	PLA, PBAT, PBS, starch-based flexible blends[55,56]
Triallyltrimesate (TAM)	Improves crosslinking and stability[52,59]	Can cause loss of Newtonian plateau[58]	Thermoset systems or crosslinked thermoplastics[59]
Tributyrin (TB)	Plasticizer, improves flexibility[60,61]	Less effective in mechanical performance, reduced mechanical strength and risk of migration[60,62]	PLA blends/composite for packaging or biomedical uses[61]
Polyethylene glycol (PEG)	Improves hydrophilicity, compatibility in polar systems[63–65]	Migration risk,[66] may reduce crystallinity[65]	PEG/PLA, PEG/PCL, PBAT, hydrogels[63,64]
MDI (Diphenylmethane Diisocyanate)	Reactive; strong coupling/crosslinker for urethanes/polyesters[67]	Toxicity, moisture sensitive[68]	Polyurethane, polyester blends needing strong covalent bonding[68]
TAIC (Triallyl Isocyanurate)	Crosslinking, thermal stability[70,71]	Can cause brittleness, costly[69]	High-temp or radiation-crosslinked systems[70]

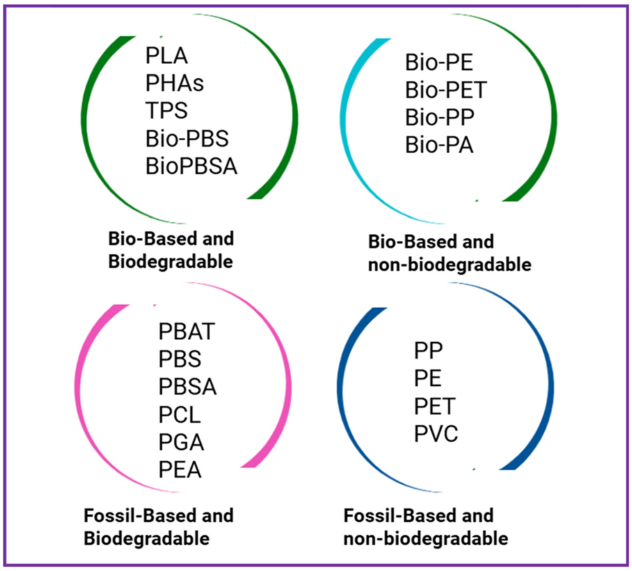


Fig. 2 Classification of polymers based on origin and biodegradability.

ance ability, low costs and easy processability,[34] which in turn would broaden their industrial application prospects.[35]

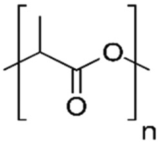
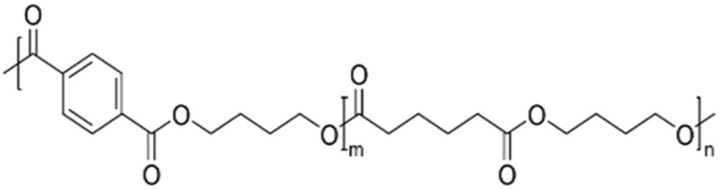
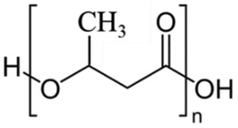
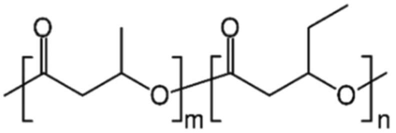
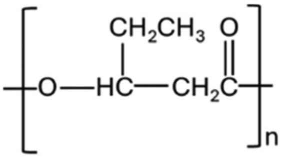
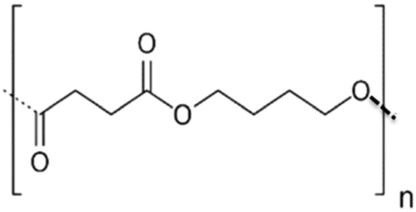
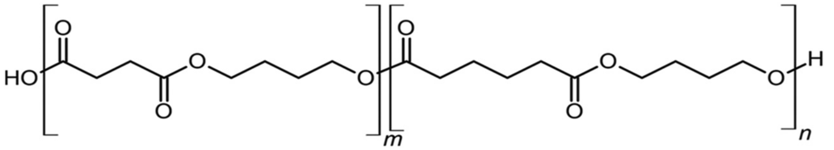
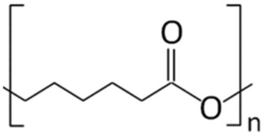
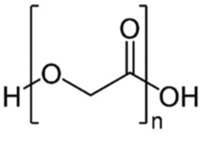
Despite the attractive potential of biodegradable polymers, their widespread use is held back by such considerations as costs, mechanical properties, and poor performance.[35–37] Developing biodegradable blends with acceptable properties can overcome these problems. However, the choice of individual polymers is dictated by their properties (see Table 3). As an example, a more brittle PLA can be blended with tough and flexible PBAT, to increase elongation at break and flexibility. The main focus of polymer blending is to improve the adhesion between the blends, reduce the interfacial tension, and generate limited inclusion phase size and controlled morphology.[38–40] Research has focused on these in recent years which can be effectively addressed by either reactive compatibilization, non-reactive compatibilization and plasticization strategies that can also increase polymer miscibility.[41–45]

2.2 Key considerations in polymer blend design

One key consideration in designing polymer blends is their miscibility. Miscibility and compatibility in polymer blends are important because they contribute to morphology, properties, and performance.[93] One or various properties of different polymers may be desirable in a single polymer blend, or there could be a need to alter the properties of another polymer; here, blending may become a choice. Polymer blends can have homogeneous or heterogeneous properties depending on the individual components in the blend.[94,95]

A homogeneous polymer blend exhibits uniform properties and composition throughout the material.[96] The polymers are miscible at the molecular level, forming a single-phase system with consistent mechanical, thermal, and morphological properties.[97] Heterogeneous polymer blends have distinct phases due to the immiscibility of the polymers.[98,99] The material's properties can vary depending on the distribution and interaction of these phases, often leading to phase-separated microstructures. Since the main challenge in polymer blends is miscibility, many researchers have reported blends to be partially miscible where the polymers mix to some extent, leading to limited interaction between the components, immiscible where the polymers do not mix, forming a multi-phase system with distinct phase boundaries, or miscible where the polymers mix at the molecular level to form a single-phase system (see Fig. 3). The overall properties of any blend depend on blend composition, the properties of individual components, and the structure and interactions of

Table 2 Common biodegradable polymers, sources, structures, and molecular formula (structures are drawn in ChemDraw)

Polymer	Composition	Structure	Molecular formula
PLA	Lactic acid		$(C_3H_4O_2)_n$
PBAT	Adipic acid, 1,4-butanediol, and terephthalic acid		$(–O–(CH_2)_4–OCO–(CH_2)_4–CO–)_X[–O–(CH_2)_4–OCO–C_6H_4–CO–]_y$
PHB	Hydroxylbutyrate		$(C_4H_6O_2)_n$
PHBV	3-Hydroxybutyrate (3HB) and 3-hydroxyvalerate (3HV)		$(C_4H_6O_2)_m$ $(C_5H_8O_2)_n$
PHV	3-Hydroxyvalerate		$C_5H_{10}O_3$
PBS	Butylene succinate		$(C_8H_{12}O_4)_n$
PBSA	1,4-Butane diol, succinic acid, and adipic acid		$H[O(CH_2)4OOC(CH_2)2CO]_m[O(CH_2)4OOC(CH_2)4CO]$
PCL	ε-Caprolactone		$(C_6H_{10}O_2)_n$
PGA	Glycolic acid		$(C_2H_4O_3)$

these components.[100] Imre and Pukánszky[37] reported that interactions are more important in blends because they determine the mutual solubility of the phases, the thickness, and properties of the interphase formed during blending, as well as the structure of the blend. The glass transition temperature (T_g), the morphologies and solubility parameters such as the Hansen and Hildebrand solubility parameters (see eqn (1) and (2)) of the blends are some of the techniques used to study their miscibility.

$$R_a^2 = 4(\delta_{D_1} - \delta_{D_2})^2 + 4(\delta_{P_1} - \delta_{P_2})^2 + 4(\delta_{H_1} - \delta_{H_2})^2 \quad (1)$$

R_a represents the Hansen solubility parameter, δ_D, δ_P, and δ_H are dispersion component (non-polar van der Waals inter-

Table 3 Mechanical properties of different biodegradable polymers

Polymer	Elongation at break (%)	TS (MPa)	TM (GPa)	FS (MPa)	FM (GPa)	Notched IS ($J\ m^{-1}$)	Notched IS ($kJ\ m^{-2}$)	Ref.
PLA	7.85–10.6	40.16–66.02	1.85	89.2	3.8	4.04	3.19	55 and 72
PBAT	385–670	21–25.1	0.034–0.067	4.0–7.5	0.09–1.26	Non-break	—	73–76
PBS	235.7–323	26.6–49.5	0.29–0.93	—	—	6.7–22.5	6.7	77–81
PHB	5–10	20–40	1.0–3.5	29	1.0	28	3 ($kJ\ mm^{-2}$)	82 and 83
PBSA	432.7	18.3	0.138–0.18	19.2	0.439	~500	32	84 and 85
PHBV	2.5–20	25–36.6	1.2–3.28	64.1	2.87	20.4	6 ($kJ\ mm^{-2}$)	74 and 86
Bio-PBSA	313–460.37	17.71–23.72	0.168–0.281	—	—	—	30.7	87–89
TPS	60–76.1	0.55–5.4	0.001–0.077	—	—	—	—	90–92

TS refers to tensile strength, TM to tensile modulus, FS to flexural strength, FM to flexural modulus, and IS to impact strength.

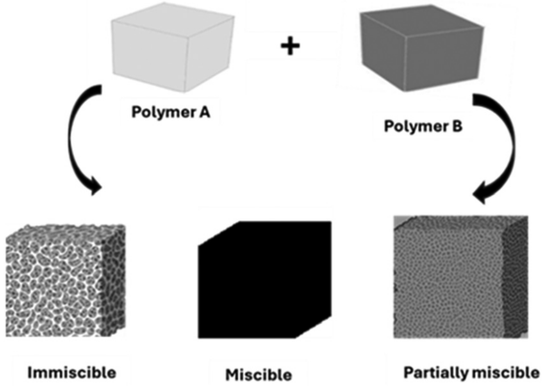


Fig. 3 Types of polymer blend: miscible, partially miscible, and immiscible blends.

actions), polar component (dipole–dipole interactions), and hydrogen bonding components (hydrogen bonding forces).

$$\delta = \sqrt{\frac{\Delta H_\mathrm{v} - RT}{V_\mathrm{m}}} \tag{2}$$

δ is the Hildebrand solubility parameter, ΔH_v, R, and T are heat of vaporisation, gas constant, temperature, and V_m is molar volume.

Other techniques are presented in Table 4. The phase transition and the interfacial tension in the blended polymers can be enhanced through polymer compatibility to promote the dispersion of one phase into another.[103]

2.3 Blending synthesis methods

Biodegradable polymer blending can be achieved using different methods of synthesis (Table 5). The method chosen will be based on the type of polymer and the applications involved. Melt blending is commonly used where polymers are mixed at high temperatures using twin-screw extruders, single screw extruders and similar equipment.[114] The design and architecture of extruders (co-rotating, counter-rotating, screw diameter and length *etc.*) greatly affect the blend morphology and outcome.[115] Process variables such as residence time, temperature, and pressure are also important to achieve desirable properties of the blend; this approach is popular in laboratories and industry because it does not require solvents, can be scaled up and matches extrusion and injection molding processes.[116] In some cases, compatibilizers are added to improve polymer compatibility and prevent phase separation.

Another method of polymer blend synthesis is solution blending. In this method, the polymers are carefully dissolved in common solvent before being cast into films.[117] This method is ideal for laboratory-scale applications where research-

Table 4 Methods for determining miscibility of polymer blends

Method	Description	Miscibility indicators	Ref.
Scanning electron microscopy (SEM)	Visualizes the surface morphology of polymer blends	Absence of phase separation = miscible; distinct domains = immiscible	55 and 104
Transmission electron microscopy (TEM)	Provides high-resolution images of polymer morphology	Similar to SEM: uniform morphology = miscible	74 and 104
Hansen solubility parameters (HSP)	Divides solubility into δ_D (dispersion), δ_P (polar), δ_H (hydrogen bonding)	Small R_a (Hansen distance) indicates good miscibility (see eqn (1))	102 and 105
Hildebrand solubility parameter (δ)	Measures cohesive energy density of polymers	δ values within 1–2 $MPa^{1/2}$ suggest miscibility; large differences = immiscibility (see eqn (2))	101
Differential scanning calorimetry (DSC)	Measures heat flow during transitions (T_g, T_m, T_c)	Single T_g = miscible; dual T_g = phase-separated/ immiscible	55, 56, 106–108
Atomic force microscopy (AFM)	Maps topography and phase at nanoscale	Homogeneous surface = miscible; differences = immiscible	109–111
Optical microscopy	Light-based visualization of morphology	Phase boundaries = immiscible; uniform texture = miscible	112 and 113
Theoretical/computational methods	Uses Flory–Huggins theory to predict blend miscibility *via* the χ parameter	Negative or low χ = miscible; high positive χ = immiscible	105

Table 5 Common methods for synthesizing biodegradable polymer blends

Method	Mechanism	Typical polymers	Pros	Cons	Ref.
Melt blending	Mixing polymers in molten state	PLA, PBAT, PHBV, PBS	Scalable, solvent-free, compatible with extrusion/injection molding	Needs compatibilizers for immiscible blends	114–116
Solution blending	Dissolution in common solvent and casting	PLA, PHBV, PCL	Good miscibility control, useful for lab studies	Solvent use, hard to scale up	117
Reactive extrusion (REX)	Chemical reactions during melt blending using compatibilizers or initiators	PLA/PBAT, PLA/PHBV	Enhances miscibility, improves interfacial bonding	Needs careful control of reaction conditions	118–120
Electrospinning	Electric field-driven fiber formation from solution or melt	PLA, PCL, PHAs	Creates porous nanostructures, suitable for biomedical	Low throughput, setup complexity	121 and 122
Interpenetrating polymer network (IPN)	Two interlaced polymer networks, one formed in presence of another	PLA/PCL, PHAs	Improved toughness, thermal stability	Complex formulation, mostly lab-scale	117
Latex blending	Aqueous dispersion of polymer latex particles then coalesced	Starch-based blends, PLA emulsions	Good particle-level mixing, eco-friendly	Requires drying/coalescence step, limited polymer types	117

ers can have control over blend uniformity. Solution blending is perceived to be less environmentally friendly because of the solvents used and may be difficult for industrial upscale.

For improving compatibility during blending, reactive extrusion is a valuable method. This process involves *in situ* chemical reaction while the materials are being processed.[118] Materials like maleic anhydride (MA) and Joncryl® chain extender are usually added to improve the compatibility and miscibility of the blend.[119,120] Electrospinning is yet another way that polymer blends can be made.[121] The process is mainly applied for blends in biomedical applications.[122] The polymer solution is spun into thin fibers using an electric field. As a result, very porous structures are obtained, but the method is more complex and limited in throughput.

In addition, interpenetrating polymer networks (IPNs) are a unique approach where two or more polymers are combined in a crosslinked state, with at least one network developing during the crosslinking of the other.[117] Because of the interlocked network, IPNs result in mechanically stronger, more thermally stable blends with shape retention which makes them suitable for biomedical and packaging applications. Latex blending is another method of polymer blending where polymers are dispersed as fine particles in an aqueous medium and then coalesced to film.[117] Many coatings, adhesives and biodegradable films use latex blending because it allows for better mixing of particles, less solvent use and more flexibility when using fillers or additives.

3. Recent developments in biodegradable polymer blends; mechanical properties, miscibility and compatibility

Many biodegradable polymer blends have been made to widen the scope of application by enhancing their mechanical, thermal, and barrier properties, improving their processability, and tailoring their degradation rates to meet specific requirements. Some biodegradable polymer blends and their properties are presented in Table 6. Biodegradable polymers demonstrate variations in mechanical properties as shown in Table 3, which significantly influence their use in various industries. Polymers such as polylactic acid (PLA) are known for their high tensile strength and stiffness, but brittleness often limits their use in applications requiring flexibility or impact resistance.[123] On the other hand, materials like polybutylene adipate terephthalate (PBAT) and polycaprolactone (PCL) exhibit excellent ductility and elongation at break, making them well-suited for flexible films and soft packaging.[16] Polybutylene succinate (PBS) offers a more balanced mechanical profile, with moderate strength and flexibility, making it versatile for both rigid and semi-flexible applications.[124] Thermoplastic starch (TPS), while attractive due to its low cost and biodegradability, has low mechanical performance and high moisture sensitivity, which often necessitates blending with other polymers to enhance its usability.[125] The best performance can be obtained by blending strategies to tailor them for specific end-use applications.

Polymer polarity significantly influences the miscibility and functional performance of biodegradable blends.[37,126] To better support this discussion, Table 7 presents the solubility parameters of common biodegradable polymers used in these systems. These parameters help predict blend miscibility by quantifying the interaction potential between different polymer phases. The solubility parameter reflects the cohesive energy density of the polymer, and the closer these parameters are between two polymers, the more likely they are to mix well. Polar–polar blends (*e.g.*, PLA with starch or chitosan) typically show good compatibility due to hydrogen bonding and dipole interactions,[37] leading to enhanced biodegradability and mechanical properties in applications such as packaging and biomedical devices.

Table 6 Mechanical properties of biodegradable polymers blends

Polymer/bled	Composition	Compatibilizer/ plasticizer/additive	Processing method	Elongation at break (%)	TS (MPa)	TM (GPa)	FS (MPa)	FM (GPa)	Notched IS (J m^{-1})	Ref.
PLA/PBS	80/20	—	Melt blending/ compression molding	14.1	60.2	1.79	—	—	8.10	77
	70/30	—		13.2	57.5	1.55			6.30	
PLA/PBSA	90/10	—	Extrusion/injection molding	12.2	52.6	1.012	—	—	—	84
		Joncryl® ADR-4300					—	—	—	
	80/20			121.2	41.2	0.754	—	—	—	
PLA/bioPBS	80/20	—	Fused deposition modeling	—	64.9	3.0	—	—	—	134
	60/40				57.1	2.4				
PLA/PBAT/ESO	40/60/5	Epoxidized soyabean oil	Injection molding	84.6	30.47	—	64.8	3.3	24.01	55
	60/40/5		Injection molding	75.96	19.56	—	39.5	2.1	13.32	
PLA/PHB	85/15	—	Extrusion	140	31	1.2	—	—	—	135
PLA/PHB/OLA	85/15/15	Lactic acid oligomer (OLA)	Extrusion	35	23	1.12	—	—	—	135
	85/15/20			220	18	0.95	—	—	—	
	85/15/30			270	19	0.59	—	—	—	
PHBV/PBAT	70/30	—	Injection molding	3.8	27.9	2.3	51.0	2.79	28.0	74
	50/50			138.6	27.5	1.6	30.6	1.38	293.9	
	30/70			345.3	26.5	6.09	15.8	5.93	no break	
PLA/PBAT/Joncryl ADR-4370S	60/40/0.75	Joncryl ADR-4370S	Reactive compatibilization/ compression molding	579.91	40.88	—	—	—	29.62	136
PLA/PBAT/GR (gum rosin)	80/20/5 parts per hundred (phr.)	Gum rosin	Extrusion/Injection molding	7.3	47.3	1.44	67.2	2.5	8.3	137
PLA/TPS/St-PLA (starch grafted poly (lactic acid))	30/70/5	Starch-*graft*-poly(lactic acid)	Compression molding	4.25	10.2	0.78	—	—	—	138
PHBV/PBAT	70/30/1	2,4′-Diphenylmethan-diisocyanate (MDI)	Twin-screw extrusion/ injection molding	11	26	1.9		4.5	—	139

TS refers to tensile strength, TM to tensile modulus, FS to flexural strength, FM to flexural modulus, and IS to impact strength.

Table 7 Solubility parameters of common biodegradable polymers

Polymer	Hansen (MPa$^{1/2}$)	Hoy (MPa$^{1/2}$)	Van Krevelen (MPa$^{1/2}$)	Hildebrand (MPa$^{1/2}$)	Small's method (MPa$^{1/2}$)	Turbidimetric titration (MPa$^{1/2}$)
PLA	20–21.9[140]	21.31[141]	20.66[141]	20.7[142] 19.9[143]	—	—
PBAT	22.1[140]	21.73[141]	21.22[141]	21.9[142]	—	—
PHB	20.7[140]	—	—	—	—	—
PHBV	20.6[144]	21.6[145]	19.9[145]	—	—	21.9[144]
PBS	22.29[147]	—	—	—	—	22.5[146]
PBSA	—	—	—	—	—	22.3[146]
Bio-PBS	20.7[140]	—	—	—	—	—
Bio-PBSA	23.3[140]	—	—	—	—	—
PGA	20.55[105]	—	—	—	—	—
PCL	21[140]	—	—	—	—	—
TPS	—	—	—	—	8.4[148]	—

Polar–nonpolar blends such as[127,128] thermoplastic starch with HDPE or LDPE offer a tunable balance between biodegradability and durability. However, PHB and PHBV have been greatly studied as the most promising biodegradable poly (hydroxy alkanoates). While it is argued that PHB is extremely brittle, PHBV a co-polymer of PHB is highly flexible and can serve to enhance the toughness of PLA.[129]

3.1 PLA/PHAs-based blends

PLA and PHAs are some of the most studied biodegradable polymers.[2,130,131] Their blends have offered significant solutions in many applications. The main aim for blending PLA with PHAs is to reduce its brittleness and enhance toughness by blending flexible and ductile polymers from renewable sources.[132] Poly(hydroxyalkanoates) (PHAs) such as PHBV (poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate)), PHB (poly (hydroxybutyrate), PHO (poly(hydroxy octanoate)), and PHV (poly(hydroxy velerate)) are biodegradable polymers that have received great attention due to their flexibility and toughness.

3.1.1 PLA/PHB. Studies have shown that incorporation of PHB into PLA can significantly increase the blend's thermal stability and enhance properties such as the crystallization rate.[132,133] Recently, Gao *et al.*[132] analyzed the performance of PLA/PHB binary blend at 10, 15, 20, 30, and 40 wt% of PHB. They noticed phase separation in the blend that possibly resulted in decreased tensile strength and a linear increase in impact toughness. It was shown that PHB wt% above 20 induced a higher crystallization of the blend, thereby lowering the crystallization temperature. The appearance of two melting peaks confirmed immiscibility of the two polymers.

Olejnik *et al.*[149] examined the optimal PLA/PHB ratio for materials with suitable mechanical, processing, and application properties. The PLA/PHB blends with mass ratios of 100/0, 50/10, 50/20, 40/30, 50/50, 30/40, 20/50, 10/50, and 0/100 were prepared using the extrusion process. The result of selected PLA/PHB blends showed two visible melting peaks, which probably signified an immiscible blend or crystalline–amorphous phase. They concluded that the PLA/PHB (50/10) ratio showed optimum performance regarding mechanical and processing properties.

In the processing of PLA/PHB blend, compatibility and miscibility are important factors as confirmed by previous studies. It was shown that the molecular weight plays a significant role in the blending process.[150,151] Low molecular weight PLA is miscible with low molecular weight PHB.[151] Very importantly, PHB/PLA shows extreme rigidity and brittleness that limit processing and use in flexible applications, thereby requiring the use of plasticizers. In some cases the plasticizers may act to improve the compatibility of the individual polymers.[152] Additionally, compatibilizers have been used to impart compatibility and polymer miscibility where needed.

Armentano *et al.*[135] prepared blends of PLA/PHB, using lactic acid oligomer (OLA) as plasticizer. The plasticizer addition was at 15, 20 and 30 wt% by weight, and the blends were extruded for better processability and mechanical properties in flexible film manufacturing. They showed that the lactic acid oligomer plasticizer decreased the PLA/PHB glass transition temperatures, with improved ductile and barrier properties. This is because plasticized blends have higher crystallinity. Field emission scanning electron microscopy (FESEM) images of blend films showed a dispersed PHB phase with a small average diameter in the PLA polymer matrix.

A binary blend of PLA/PHB (60/40) was prepared in the presence of 15 wt% tributyrin (TB) plasticizer.[153] The blend without TB was characterized by a rough fracture surface and phase separation, with two microstructure phases which were accompanied by voids. However, the compatibilized blend caused elastic deformation with fuzzy interfaces and no phase separation, suggesting better adhesion in the blend. The addition of TB negatively affected the gas and vapour barrier which increased with plasticization. The mechanical properties showed reduction in the tensile modulus and strength without an improvement in the impact strength. The PLA/PHB blend displayed a double glass transition, with distinct T_g values for PLA and PHB, indicating their immiscibility. Plasticization caused a reduction in the T_g of the blend, indicating enhanced polymer chain mobility. When propylene oxide block copolymer/ethylene oxide (Synperonic (Syn)) and a mix of adjustable lipophilic–hydrophilic balance liquid surfactants (HLB12) were used as compatibilizers in PLA/PHA (70/30) by Anna *et al.*,[154] it was noticed that adding 0.1 wt% Syn reduced PHB particle size and pull-out in the blend, and HLB12 led to a homogeneous morphology with elongated PHB domains. The cold crystallization of PLA was favoured while progressively increasing the blend crystallinity with a noticeable increase in the elastic modulus of the blend.

3.1.2 PLA/PHBV.

The properties of PLA/PHBV blends are significantly affected by the conditions of processing and their blending proportion. Typically, the addition of PHBV to PLA improves the toughness and flexibility of the blend while reducing PLA's brittleness. The thermal stability of the blend is also enhanced, making it more suitable for applications requiring higher processing or service temperatures.

Typically, the major drawback is the immiscibility of PLA and PHBV, which determines the morphology and properties of the blend. PLA and PHBV are generally considered immiscible due to their different molecular structures and polarities. Polarity is the distribution of electrical charges in polymer molecules. This determines how the molecule interacts with others; this influences their intermolecular forces, such as hydrogen bonding or van der Waals interactions,[155] and plays a significant role in polymer miscibility. This immiscibility often leads to phase separation, which can result in a heterogeneous blend with distinct domains of PLA and PHBV. Polymer blends that are immiscible or partially miscible can exhibit phase separation, leading to distinct regions in the blends, and this can influence the crystallization behaviour of the blend.[156]

It is well known that PLA exhibits a slow rate of crystallization, and this has been addressed by adding nucleating agents.[157,158] The crystallization behaviour of PLA can be influenced by the presence of PHBV as it promotes nucleation, leading to an increase in the crystallinity of PLA, which can improve the mechanical properties of the blend. For instance, the mechanical performance limitations of PLA were addressed by blending it with PHBV and subsequent addition of filler to enhance the properties of the resulting melt-blown nonwovens.[159] PLA and PHBV were blended in various ratios (10, 20, 30, 40 wt%) and processed with a melt-blown technique. When PHBV was added, the cold crystallization peak of PLA significantly decreased, as PHBV acts as a nucleating agent, promoting crystallization during cooling. However, at PHBV contents higher than 10%, cold crystallization of PLA was almost absent. The blend also experienced a mutual crystallization effect where the crystallization of PLA/PHBV phases was mutually hindered and the enthalpy of crystallization for both phases was lower compared with their pure forms, which was attributed to the immiscibility and phase separation in the blends. Novak *et al.*[160] indicated that the addition of PHBV suppressed the cold crystallization of PLA, which negatively influenced the dimensional stability of injection-molded parts.

In the study by Lo *et al.*,[161] electrospun PLA/PHBV membrane (1 : 1, 3/1) showed partial miscibility with a single wide melting peak, and a shift in the glass transition temperature to lower values was observed in the initial heating cycle. The blends had similar tensile strength and percentage of elongation while the 1 : 1 blend exhibited higher Young's modulus. Also, they showed that the mechanical performance of the PLA/PHBV was controlled by the crystallinity of the blend, as the percentage crystallinity was higher (35.2%) in the 1 : 1 blend compared with the 3 : 1 (13.8%).

The contribution of divergent compatibility in a blend has been shown to affect the scope of application of such blend. Compatibilization is essential for biopolymer blends to become viable alternatives to petroleum-based polymers. Compatibilization improves mixing, stabilizes microstructures, and enhances the synergy between biopolymers by reducing interfacial tension and strengthening adhesion, particularly in the solid state, to enable efficient stress transfer.[22] A previous study examined the *in situ* grafting of 2,5-dimethyl-2,5-di(*tert*-butylperoxy)hexane, DBPH, a peroxide initiator, *via* one-step reactive extrusion onto PHBV where the thermal decomposition of PHBV produced unsaturated bonds that graft onto the PLA backbone.[161] The optimal 80/20 PLA/PHBV blend with 0.3 wt% DBPH showed significant improvement in mechanical properties by increasing elongation at break to about 15% from about 10% for blend without the initiator while maintaining tensile strength near that of pure PLA. The thermal stability of the blend was improved, which lowered the glass transition temperature, increased crystallization temperatures, and transformed the blend morphology from a sea-island observed without grafting to more layered structure, demonstrating effective toughening and enhanced performance. Similarly, Pouriman *et al.*[162] grafted maleic anhydride (MA) onto PLA and PHBV to enhance their compatibility in blends by using dicumyl peroxide (DCP) as an initiator during melt compounding. MA and DCP improved the interfacial adhesion in the blend and a possibility of chain-shortening reaction was associated with excessive DCP. Morphologically, MA and DCP resulted in finer dispersion of PHBV within the PLA matrix, suggesting improved phase compatibility and reduced domain size. Table 8 shows blends of PLA with polyhydroxyalkanoates with different additives and the findings from the blending process.

3.1.3 PLA/PHA blend composite.

PLA/PHA reinforced composites are innovative biopolymer materials that combine the biodegradability of polylactic acid (PLA) and polyhydroxyalkanoates (PHA) with enhanced mechanical and thermal properties, making them ideal for sustainable applications in various industries. Natural fibers are a good filler material in biodegradable composites. They can add to biodegradability and improve properties such as the mechanical, thermal, and barrier properties of composites. A lot of studies have shown the potential of natural fibers as reinforcement in polymer blends despite their limitations.

Boey *et al.*[167] investigated the effect of incorporating spent coffee grounds (SCG) into a poly(hydroxyalkanoate) (PHA) and poly(lactic acid) (PLA) matrix. Various SCG loadings (10–40 wt%) and PHA/PLA ratios were tested for their influence on mechanical properties. A higher SCG content reduced the mechanical performance of the composite in comparison with the blend. The flexural strength dropped from 19 MPa to about 7 MPa. However, adding fungal-treated SCG improved interfacial adhesion, causing the flexural strength to increase by 26%–52% and flexural modulus by 72%–113%. The impact strength was also increased by 56% as a result of better filler–matrix interaction that enhanced stress distribution in the biocomposite.

In PLA/PHBV blends, spent coffee grounds (SCG) were found to influence crystallization behaviour and mechanical properties.[160] SCG increased PLA crystallinity from 8% to 17% by acting as a nucleating agent, accelerating crystallization, but reduced crystallization ability in PHBV-rich blends by 10%–12%. This dual effect impacted the mechanical performance, lowering tensile strength by 15%–20% and flexural strength by 1%–10%, depending on the blend composition. Similarly, Marta *et al.*[168] investigated biochar-reinforced composites of PLA/P(3HB-*co*-4HB) using acetyltributyl citrate (ATBC) plasticizer for sustainable applications. Biochar, used at 10%, 15%, 20%, and 30%, enhanced electrostatic and degradation properties. The surface resistivity decreased significantly from $3.80 \times 10^{12}\ \Omega$ (no biochar) to $1.32 \times 10^{12}\ \Omega$ at 30% biochar, improving electrostatic performance. The degradation study revealed that composites with lower biochar content exhibited accelerated degradation, especially in hydrolytic conditions (70 °C), with accelerated PLA loss at biochar content of 20 wt%. However, while SCG accelerated PLA crystallization, biochar incorporation PLA/P(3HB-*co*-4HB) enhanced stiffness and electrostatic properties but likely impacted flexibility and overall crystallization dynamics, reflecting the complex interplay between fillers, matrix composition, and mechanical performance.

Building on this, Jurczyk *et al.*[169] further investigated PLA/P(3HB-*co*-4HB)/biochar composites at 0, 10, 15, 20 and 30 wt% using ATBC. Their results showed that the composite became stiffer but weaker and more brittle with increasing amount of biochar. Tensile strength dropped to 27.1 MPa from 35.6 MPa for the pure blend at 30 wt% biochar, a 24% decrease. The tensile modulus doubled from 1230 MPa to 2756 MPa as biochar content increased, while the elongation at break sharply reduced from 12% to 4.2% at 30 wt% biochar, a 65% reduction. The impact resistance decreased by 64% (4.47 kJ m^{-2} for the neat blend to 1.61 kJ m^{-2} at 30 wt%). This reduction in strength and impact resistance was attributed to poor bonding between the biochar and polymer matrix, with biochar's porous structure causing cracks to form easily.

3.2 PLA/PBAT-based blends

Blends of PLA and PBAT have been widely researched, with a focus on mechanical properties, rheological behaviour, and biodegradability.[106,107,170–173] Blending PLA/PBAT is a good step forward in improving not only the biodegradability of the blend but also its applications. PBAT can significantly increase the flexibility and toughness of the blend but can also lead to reduced tensile strength and modulus. With these in mind, research has been centered around blend compatibility,

Table 8 The processing methods, additive/compatibilizers used, and properties of PHA-based blends

Blend composition	Aim	Processing method	Compatibilizers/ plasticizer	Result summary	Ref.
PLA/PHBV (10–40% PHBV)	Investigates the blending of PLA with PHBV to enhance PLA's foaming behaviour	Extrusion		• Immiscible blend observed, PHBV exists as dispersed droplets in PLA matrix • The addition of PHBV influenced the crystallization behaviour of PLA, PHBV played nucleating agent, leading to a higher cell nucleation density	159
PLA/PHBV 25/75, 50/50, and 75/25 wt%	Blend property optimization	Solvent casting method		• PLA/PHBV polymer solutions were immiscible with two noticeable T_g • Mechanical properties decreased with the increasing amount of PHBV. Weak polymer interface of PLA and PHBV	163
PLA/PHBV (60 : 40; 50 : 50; and 40 : 60)	Investigate 3D printability of PHBV/PLA	Fused filament fabrication (FFF)	Joncryl grade ADR-4368C	• Printing parameters depend on viscosity and thermal stability • High temperature of 220 °C and speed of 45 mm s^{-1} was recommended for decreased viscosity (8–80 Pa s) • Joncryl compatibilizer improved interface bonding	164
PLA/PHBV (75/25)	Study the antibacterial effectiveness of PLA/PHBV blend film against *L. innocua* by adding ferulic, *p*-coumaric, and protocatechuic antibacterial agents	Melt-blending and compression molding	Polyethylene glycol (PEG 1000)	• 2% of ferulic increases T_g of PLA • Protocatechuic acid caused PHBV supercooling • Thermal stability of blend films was observed with ferulic, *p*-coumaric and protocatechuic acids • All three phenolic acids showed antibacterial activity against *Listeria innocua and Escherichia coli* bacteria	165
PLA/PHB	Investigate better processability and mechanical properties	Electrospinning	Lactic acid oligomer (OLA) (10, 15 and 20 wt%)	• PLA crystallization, decreased viscosity of the blend • 20 wt% OLA produced bead defects and consequent poor mechanical performance • The ternary system PLA-PHB with 15 wt% OLA maintained the mechanical properties	166
PLA/PHO	Study the non-isothermal cold crystallization kinetics of PLA/PHO/talc	Film casting		• The blends displayed two T_g resulting from viscosity mismatch • The blends are not thermodynamically miscible • The crystallization of PLA was increased with a higher amount of PHO and talc	51

increasing the miscibility of the blend for better performance. The compatibilized blends of PLA/PBAT can be achieved through physical and reactive compatibilizers. Block or graft copolymers compatible with PLA/PBAT are commonly utilized as physical compatibilizers. Reactive compatibilizers, on the other hand, can chemically link the PLA and PBAT phases, resulting in high interfacial interaction and superior compatibility.[174] Wu *et al.*[106] studied the impact of ADR4370S, a polyfunctional epoxy-based chain extender/compatibilizer (0 wt%, 0.3 wt%, 0.6 wt%, 0.9 wt%, and 1.2 wt%) on a blend of PBAT/PLA (95/5) and subsequent supercritical CO_2 bead foams. PBAT/PLA compatibility was adjusted by varying the ADR wt%, thus allowing precise foam cell morphology control. The introduction of ADR4370S enhanced the compatibility between PBAT and PLA by facilitating grafting and chain extension reactions, leading to improved interfacial adhesion and a more uniform molecular structure. This resulted in increased viscosity and modulus, contributing to better mechanical strength and elasticity in the blend, which was essential for maintaining foam structure. Differential scanning calorimetry (DSC) analysis showed that the glass transition temperature (T_g) of PBAT increased with higher ADR content as a result of improved compatibility, while crystallinity decreased in the blend because of restricted polymer molecular chain movement. The addition of ADR produced a more homogeneous pore structure, particularly at an optimal ADR content of 0.6 wt%, which achieved the smallest average pore size and the highest foaming ratio.

Epoxidized canola oil (ECO) (5 phr.)-compatibilized PLA/PBAT (80/20, 70/30, and 60/40) blend was prepared by Wahbi *et al.*[107] for application in 3D printing. Their idea was to achieve high impact toughness. In the presence of ECO there were significant improvements in PLA/PBAT compatibility that led to homogeneous PBAT domains within the PLA matrix. The blend containing 30 wt% PBAT showed optimum impact performance of 148 J m^{-1}, an increase of 106% compared with non-compatibilized blends. Analysis of the 3D printed samples showed that there was poor adhesion in the PLA/PBAT strands and larger voids were observed, resulting in brittle failure. The ECO compatibilization induced adhesion of neighboring strands with fewer air voids between deposited strands, resulting in enhanced interlayer adhesion and ductility.

Han *et al.*[108] studied epoxidized soybean oil (ESO)-compatibilized PLA/PBAT blends through reactive compatibilization with varying ESO content at PLA/PBAT ratio of 70/30. The uncompatibilized blend was grossly immiscible with phase separation. The DSC result showed two glass transition temperatures to prove the immiscibility of the blend. They further noted that compatibilization resulted in a single glass transition temperature.

The tensile strength of 70PLA/30PBAT/5ESO increased by 25%, the elongation at break increased more than 6 times, and notched impact strength increased by 400% relative to PLA/PBAT blend.

A biodegradable blend of PLA/PBAT (70/30) was prepared using interfacial stereocomplex crystallites (poly(styrene)-*co*-glycidylmethacrylate) (i-SCs) having 45% epoxy groups per 100 g by reactive blending.[104] The interface compatibilization resulted in enhanced comprehensive performance, which the authors attributed to the "rigid" i-SC, co-continuous morphology. The neat blend showed "sea-island" features having large spherical domain sized-PBAT in the PLA matrix (Fig. 4). This "sea-island" feature had a poor interfacial region, suggesting both polymers were thermodynamically immiscible. Nonetheless, the domain size in the compatibilized blend was greatly reduced due to better interface adhesion (Fig. 5).

Atom transfer radical polymerization *via* surface-initiation is a grafted-from strategy that can mobilize polymers onto the surface of fillers such as cellulose nanocrystal (CNCs). This method can increase the dispersion of the fillers in matrices

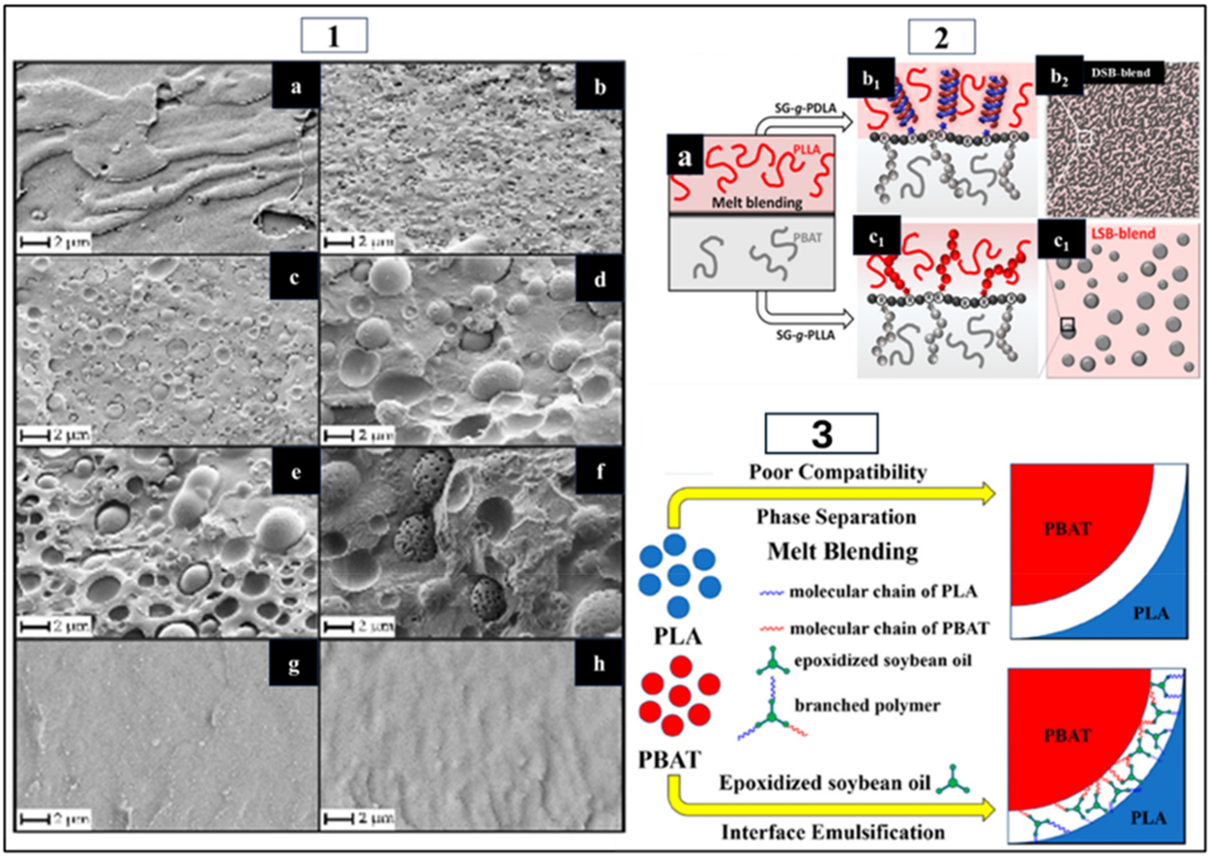


Fig. 4 The morphology of polymer blends using different types of compatibilizers and the effect on polymer phase and miscibility. (1) Field emission scanning electron microscopy (FESEM) images of (a) PLA (b) PLA/PBAT (c) PLA/PBAT5GR, (d) PLA/PBAT10GR, (e) PLA/PBAT15GR, (f) PLA/PBAT20GR, (g) PBAT and (h) PBAT10GR. (2) (a) PLA/PBAT with different reactive compatibilizers; (b) SG-*g*-PDLA reactive compatibilizer to yield "stable co-continuous PLA/PBAT (DSB) blend and (c) SG-*g*-PLLA reactive compatibilizer to yield "Sea-island" LSB Blend. (3) Compatibilization effect of ESO on PLA/PBAT.[104,108,137] (1) was reproduced from ref. 137 under Creative Commons CC license, copyright 2025. (2) was reproduced from ref. 104 with permission from American Chemical Society, copyright 2025 and (3) was reproduced from ref. 108 with permission from American Chemical Society, copyright 2025.

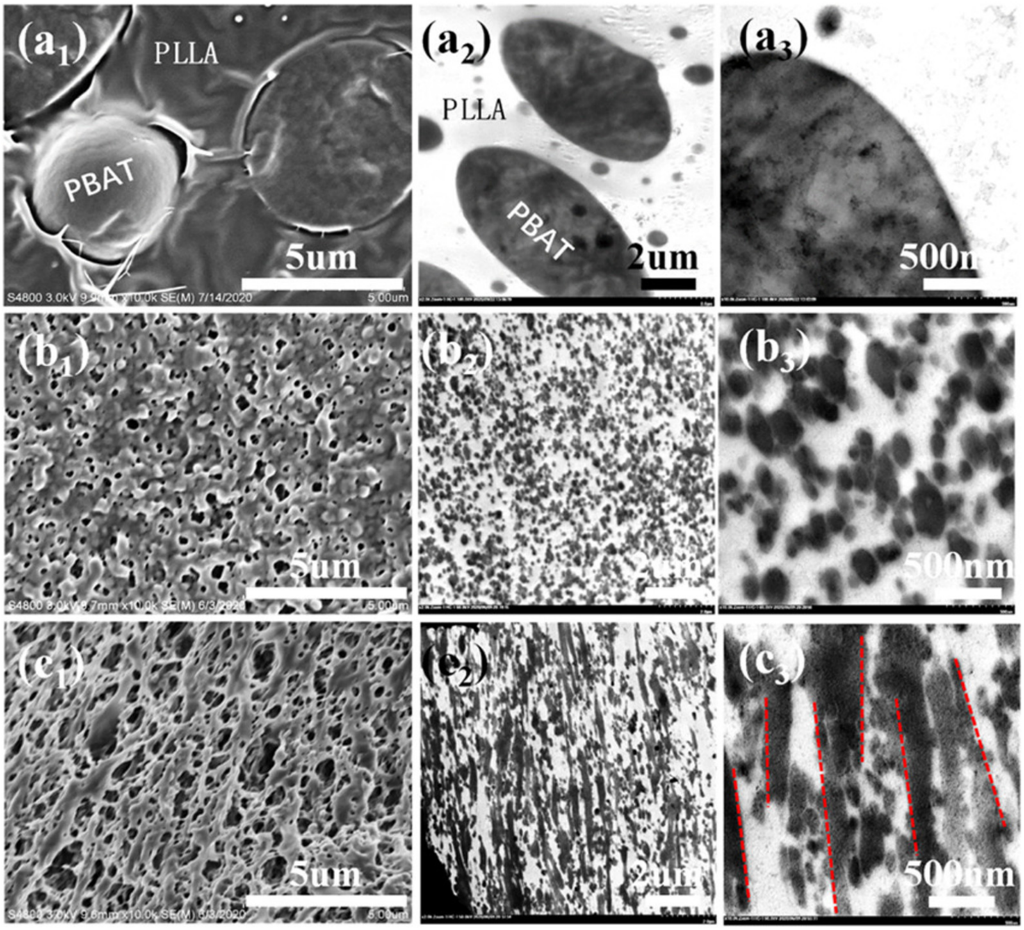


Fig. 5 The influence of compatibilizer on the morphology of PLA/PBAT blend. SEM and TEM images of 70PLLA/30PBAT blends with and without compatibilizers: ($1a_1$–a_3) neat blend, ($1b_1$–b_3) (poly(styrene-*co*-glycidyl methacrylate)-*graft*-(l-lactide)) (LSB) compatibilized PLA/PBAT blend, and ($1c_1$–c_3) (poly(styrene-*co*-glycidyl methacrylate)-*graft*-(D-lactide)) compatibilized PLA/PBAT (DSB) blend.[104] Figure was reproduced from ref. 104 with permission from American Chemical Society, copyright 2025.

and effectively initiate interface compatibility between polymer blends. A dual-purpose incorporation of functionalized cellulose nanocrystal was reported by Sun *et al.*[175] The authors grafted poly(glycidyl methacrylate) (PGMA) (10, 20, and 50%) on the surface of CNC by atom transfer radical polymerization to enhance strength, toughness and compatibility of PLA/PBAT (70/30 wt%) blend. As expected, their result showed that after adding CNC-PGMA, the PLA/PBAT blend (70/30 wt%) reduced in phase size, which they ascribed to improved interfacial compatibility between PLA and PBAT. The PLA/PBAT/CNC1.0-PGMA30 composite exhibited a tensile strength of 49.6 MPa and an elongation at break of 268.5%, which significantly surpassed the mechanical performance of the pure PLA/PBAT and indicated a considerable enhancement in both strength and toughness.

Junwen *et al.*[176] demonstrated that processing parameters and method can significantly impact the properties of a polymer blend. In their work, the blend of PLA/PBAT (80/20) was prepared by combining high mold temperature and strong shear fields during processing. Using the conventional injection molding (CIM) and multi-flow vibration injection molding (MFVIM) techniques led to the formation of highly oriented microstructures, including shish-kebab structures, which increased the blend crystallinity to 48%. The synergistic effect of a high mold temperature and strong shear fields in the MFVIM resulted in tensile strength of 89.07 MPa, tensile modulus of 1664.15 MPa, and impact strength of 10.36 kJ m^{-2}, compared with 53.04 MPa, 1106.47 MPa and 4.61 kJ m^{-2} for the CIM. Additionally, the Vicat softening temperature (VST) of the blends increased to 149.2 °C, indicating a marked improvement in heat resistance. This synergistic method provides a promising approach for developing high-performance PLA materials, expanding their potential applications as sustainable alternatives to conventional plastics. Table 9 shows blends of PLA with PBAT with different additives and the findings from the blending process.

While compatibilization has improved the mechanical performance, interface adhesion, and blend morphology of PLA/PBAT blends, it also presents opportunities to fine-tune their degradation properties by optimizing the blend composition

Table 9 The processing methods, additive/compatibilizers used, and properties of PBAT-based blends

Blend composition	Aim	Processing method	Compatibilizers/plasticizer	Result summary	Ref.
PLA/PBAT (14/86)	Study antimicrobial properties of polylimonene (PLM) and limonene (LIM) both at 5 wt% and 10 wt% in PLA/PBAT blend for sustainable active food packaging	Thermo-compression		PLM enhanced antimicrobial and antioxidant activity; 5% PLM preserved visual quality of cherry tomatoes; 10% LIM showed no antifungal activity and promoted fungal growth	183
PLA/PBAT (0, 10, 20, 30, 40 PBAT)	Study effects of PBAT addition on properties of 3D-printed materials	Twin-screw extrusion, fused deposition modeling (FDM)		PBAT improved crystallinity and ductility but reduced strength; 30% PBAT had highest elongation (10.15%)	184
PLA/PBAT (70 : 30)		Reactive compatibilization/ injection molded	ESO (epoxidized soybean oil) (0, 0.5, 1, 3, 5, 7, and 9 phr.)	ESO improved miscibility (single T_g); 5 phr. ESO increased strength by 25%, elongation by 4 folds, impact strength 400%	108
PLA/PBAT (60 : 40, 40 : 60)		Melt blending/ injection molding	ESO (1.5%)	ESO improved elongation and impact strength; tensile strength decreased; partial miscibility	53
PLA/PBAT (70/30)		Reactive blending	Interfacial stereocomplex crystallites (poly(styrene)-*co*-glycidylmethacrylate) (i-SCs) having 45% epoxy groups per 100 g.	i-SC compatibilization enhanced mechanical properties *via* co-continuous morphology; uncompatibilized blend was immiscible	104
PLA/PBAT			Joncryl ADR-4370S	ADR improved PBAT dispersion; reduced domain size; unmodified blend showed poor adhesion	136
PLA/PBAT (80/20)	Study the plasticizing effects of Gum rosin (GR)	Twin-screw extrusion/ by injection molding, film casting	Gum rosin (GR) (5, 10, 15, and 20 phr)	GR improved miscibility (Fig. 5), processability and reduced PLA T_m and T_g and increased PBAT domain size	137

and compatibilizer selection. The biodegradability of PLA/PBAT blends has been extensively studied due to their potential as eco-friendly alternatives to traditional plastics, with their formulations designed to balance toughness and degradability for better environmental performance. Research demonstrates that biodegradability depends significantly on environmental conditions and the blend composition.[2,177] For instance, under composting conditions, a co-culture of thermophilic bacteria (*Pseudomonas* G1 and *Kocuria* G2) degraded PLA/PBAT blends at a rate of 72% within 15 days, primarily through enzymatic activity that targeted ester bonds.[178] In soil, higher temperatures (58 °C) enhanced the degradation of the PLA/PBAT blend (8.3 wt% of each), achieving 9.2% and 6.1% degradation for PBAT and PLA.[179] However, at a lower temperature of 25 °C, 2.3% of PBAT and 1.7% of PLA degraded respectively, over 33 weeks, while also affecting microbial diversity and soil properties. Similarly, anaerobic digestion (AD) systems showed limited effectiveness for PLA/PBAT blends, with bioplastic bags exhibiting minimal breakdown compared with PLA/PBAT-coated papers, which contributed to biogas production under thermophilic AD conditions.[180] Fungal biodegradation of PLA/PBAT blend was also studied using *Papiliotrema laurentii* S2P4P.[181] The fungus simultaneously degraded PLA/PBAT film, decreasing the half-life of PLA/PBAT blends to about 138 days from 3 years, and produced intermediates like adipic acid and lactic acid. Enzymatic degradation of PLA/PBAT blend (20, 40, 60, and 80 wt% PBAT) using cutinase enzyme from *Humicola insolens* (HiC) demonstrated up to 40% weight loss in PBAT-rich blends within seven days at 70 °C.[182] However, the PLA-rich blends show less degradation. Despite these advances, challenges remain, such as incomplete degradation, persistent residues, and environmental variations affecting performance.

3.2.1 PLA/PBAT blend composite.

Incorporating turmeric and cinnamon powder as natural fillers in biodegradable PBAT/PLA blend films for use in active packaging showed the potential of cinnamon as an active filler.[185] Particularly, cinnamon was effective at reducing UV light transmittance and increasing the surface hydrophobicity of the composite film. Cinnamon contains UV-absorbing compounds like cinnamaldehyde and polyphenols that reduce UV transmittance, while its hydrophobic nature increases surface hydrophobicity. The mechanical properties of the blend improved after adding 5 wt% cinnamon, with elongation increasing by 43%. It was observed that a higher elongation at break could be achieved after 2 reprocessing cycles, which improved filler dispersion in the matrix.

Natural cotton stalk (CS) was used as a bio-based filler in PLA/PBAT blends, and the effect on mechanical and barrier properties and compatibility with the blend were studied.[186]

Adding 2 wt% CS increased tensile strength by 21.5% and elongation by 41.6% (Fig. 8). However, at a higher CS content, filler agglomeration and reduced performance were observed. The CS in PLA/PBAT blend improved the water vapor and oxygen barrier properties of the composite, making it more suitable for packaging applications. Jute natural fiber (JF) in PLA/PBAT blend was reported by Sudha *et al.*[187] to improve mechanical and thermal properties. The Young's modulus of the composite was approximately 1832 MPa with 15%JF, while thermal stability rose from 270 °C in pure PLA to 346.37 °C in the composite (Fig. 8). Impact strength reached 49 J m^{-1}, comparable to ABS-based engineering plastics and outperforming recycled ABS/jute composites. There was improved fiber–matrix bonding, enhanced toughness and crack resistance. Hence, PLA/PBAT/JF could be a potential sustainable alternative to conventional plastics for engineering applications.

The role played by compatibilizers and chain extenders in developing natural fiber-filled biodegradable blend composites is critical in enhancing their mechanical, thermal and barrier properties. Compatibilizers can improve interface adhesion, and chain extenders can increase molecular weight to improve toughness of the composite.

The effect of steam and myristic acid treatment on bamboo fiber (BF) in epoxidized soybean oil (ESO)-compatibilized PLA/PBAT blends was studied by Olonisakin *et al.*[55] Treated BF significantly increased the elongation at break (by up to 644%) and impact toughness (by 370%) of the composites (Fig. 6). There was enhanced fiber dispersion and interfacial bonding with PLA/PBAT and improved thermal stability.

Polyethylene grafted maleic anhydride (PE-*g*-MA) served as coupling agent for better interaction of a PLA/PBAT nanocomposite blend with cellulose nanocrystal.[188] A weak interfacial adhesion between the components was observed, due to poor compatibility. The PE-*g*-MA provided an interface bridge where the MA of PE-*g*-MA could bond with the –OH of CNC through hydrogen bonding. The CNC and PE-*g*-MA functioned as nucleating agents, thereby increasing PLA crystallinity and thermal stability.

The addition of rice straw micro-particles to PLA/PBAT blends in the presence of Joncryl® chain extender and maleic anhydride (MA) followed by 500 h accelerated weathering was studied by Mekonnen *et al.*[189] The Joncryl® chain extender was more effective compared with MA in creating a cross-linking network. The addition of rice straw to the blend increased the tensile modulus from 2.3 to 3.4 GPa, a 43% improvement, while the tensile strength decreased (Fig. 6). Rice straw particles served as nucleating agents and lowered the glass transition temperature of the PLA phase from 70 °C to 65 °C. Joncryl®-treated composites retained better mechanical properties after weathering, with a 10–19% reduction in tensile strength compared with 27–30% for MA-treated samples. Similarly, when wood flour and wollastonite was added to PLA/PBAT blends followed by a 1000-hour accelerated weathering (UV exposure), both the blend and the composite exhibited a significant colour change due to the degradation of lignin. The tensile strength and elongation at break declined, with the composite showing a 15% reduction in tensile strength.[190]

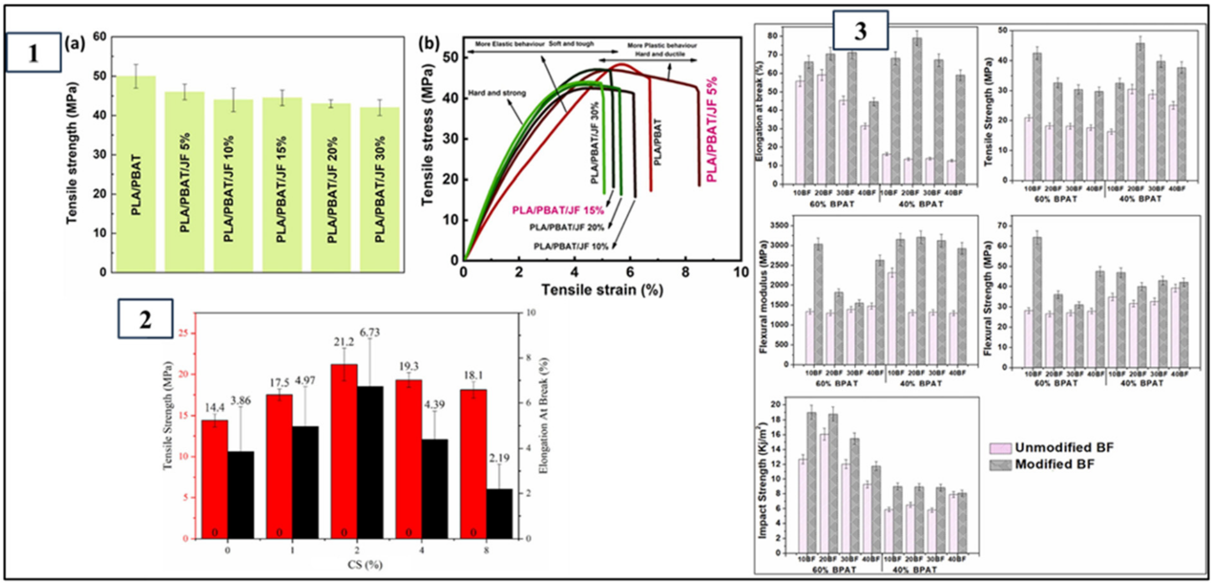


Fig. 6 Effects of different natural fiber on the mechanical properties of biodegradable blend composite. (1) (a) tensile strength of PLA/PBAT with different jute fiber amounts, (b) tensile stress of PLA/PBAT with different jute fiber amounts. (2) tensile strength and elongation at break of cotton stalk-reinforced PLA/PBAT composite, (3) mechanical properties of bamboo fiber-reinforced PLA/PBAT (elongation at break, tensile strength, flexural strength, flexural modulus and notched Izod impact strength). (1) was reproduced from ref. 187 with permission from Elsevier; copyright 2025, (2) was reproduced from ref. 186, with permission from Willey, copyright 2025, (3) was reproduced from ref. 55, with permission from Elsevier, copyright 2025.

3.3 PBAT/PHA-based blends

Polyhydroxy alkanoates (PHAs) and PBAT are two major biodegradable polymers that have received research attention in the search for eco-friendly materials. PHAs are synthesized by many microorganisms, with biodegradability properties and flexibility, while PBAT is a synthetic aliphatic–aromatic copolyester that exhibits great toughness, flexibility, and processability. Combining these two polymers into biodegradable blends has emerged as a viable approach for improving their overall performance and usability as bioplastics in many applications, without compromising their inherent properties.[191]

3.3.1 PBAT/PHBV. Biodegradable PHBV/PBAT blends are important in sustainable packaging applications, so the knowledge of their structure–property relationship is important for tailored applications. A study by Resch *et al.*[139] on the blend of PHBV and PBAT utilized biodegradable citrate ester plasticizer (Citrofol BII), and 2,4′-diphenylmethan-diisocyanate (MDI) (1 wt%), as compatibilizer to improve the processing and mechanical properties of the blend. The PHBV/PBAT blend was prepared in ratios of 90/10, 80/20, and 70/30, and processed by injection molding at 180 °C. The results showed that MDI and the citrate ester increased the elongation and toughness of the blend, but with reduced stiffness and strength. Additionally, PBAT increased the viscosity and melt strength of the blend, making it suitable for blown film extrusion. The MDI chemically reacts with the hydroxyl -OH acid –COOH groups of PHBV and PBAT to form urethane and carbamate linkages, thereby improving compatibility and mechanical properties of the blend.

A recent study[191] explored the blending of biodegradable polyesters, PBAT/PHBV (40/60) using various processing additives like peroxides 2,5-bis(*tert*-butyl-peroxy)-2,5-dimethylhexane (Luperox 101), 2,5-dimethy-2,5-di(*tert*-butylperoxy) hexane (Trigonox101) and Joncryl® ADR 4368, an epoxy-based chain extender. The primary aim was to improve the mechanical, thermal, and morphological properties of the blend, addressing issues like immiscibility and poor mechanical performance. The result was a 35% improvement in tensile strength and a 64% improvement in Young's modulus when 0.02 phr. peroxide and 0.3 phr. chain extender were used. These additives significantly increased chain length and formed complex crosslinked network structures that were indicative of non-Newtonian properties in the blend. However, the effectiveness of the chain extender in combination with the peroxides facilitated crosslinking/branching of the polymer chains, resulting in smaller and finely dispersed PBAT droplets.

Jahangiri *et al.*[192] proposed a novel method to enhance the barrier and mechanical performance of PHBV/PBAT/Joncryl blend by coating (dip coating and bar coating) with PHBV. The addition of 0.3 phr. (per hundred resin) Joncryl to the PHBV/PBAT blend improved the blend's miscibility through chemical reactions between the epoxy groups of Joncryl and the hydroxyl/carboxyl end groups of the polymers. This led to the formation of a more homogeneous interfacial layer, leading to an increase in elongation at break by 102%. In terms of barrier properties, PHBV/PBAT/Joncryl bar-coated with PHBV (10–15% coating weight) showed a 48% improvement in water vapor barrier and a 53% improvement in oxygen barrier compared with the uncoated sheet, whereas the dip-coated PHBV/PBAT had little effect on these properties. The elongation at break reduced significantly with the PHBV coating, particularly with the thicker bar-coating that resulted in a more brittle material.

The morphology and performance relationship of PHBV/PBAT biodegradable blends were studied by Zytner *et al.*[74] through injection molding. The PHBV/PBAT blend was varied at 70/30, 50/50, and 30/30 without any compatibilizer. The result showed that 50 wt% of PBAT could improve the brittleness of PHBV with co-continuous configurations in the blend. The SEM results and glass-transition temperature study indicated that the PHBV/PBAT blend had distinct phase separation due to immiscibility (Fig. 7). PHBV crystallinity decreased with the addition of PBAT according to the DSC analysis. This showed that PBAT can hinder the formation of ordered crystals by PHBV. The studied co-continuous morphology could improve mechanical properties by enabling efficient stress distribution, strain transfer, and a balanced contribution of stiffness from PHBV and flexibility from PBAT. These co-continuous configurations could also improve processability by enhancing melt strength and viscosity uniformity, enabling better extrusion and molding performance.[193]

Pal *et al.*[194] studied the REX of modified nanoclay-PHBV/PBAT nanocomposite films for use in packaging. Compression molded films were compared with cast film extrusion. First, a 20% nanoclay and PBAT masterbatch was made by melt extrusion. Next, a PHBV/PBAT blend was made using the masterbatch of 0, 0.6, 1.2 and 1.8 wt%, with 40 wt%/60 wt PHBV/PBAT. The PBAT served as a toughener to improve toughness and strain at break of the brittle PHBV. Simultaneously, the nanoclay masterbatch aimed to reinforce the blend and improve PHBV/PBAT interfacial adhesion. According to their findings, the PHBV phase was encircled by the continuous phase PBAT in the PHBV/PBAT due to phase separation. However, the nanoclay improved the dispersion of PHBV in PBAT due to improved interface adhesion. The DSC curves exhibited a distinct crystallization peak for the samples prepared by compression molding and cast film extrusion, showing the formation of uniform crystals. However, the nanoclay induced a subtle hump preceding the crystallization peak, suggesting that the nanoclay promoted imperfect crystals or lamellae in contrast to the PHBV/PBAT blend with uniform crystals. The observed improvement in the crystallization temperature with the addition of nanoclay indicated an impact on the blend's solid-state morphology and crystallization kinetics. This effect suggested enhanced compatibility and interaction between the PHBV and PBAT components, signifying a promising development in the overall thermal behaviour of the nanocomposite material.

3.3.2 PBAT/PHB. Interest in biodegradable blends of PLA/PHAs continues to grow, and the scope of applications is increasing with continuous improvement in research and

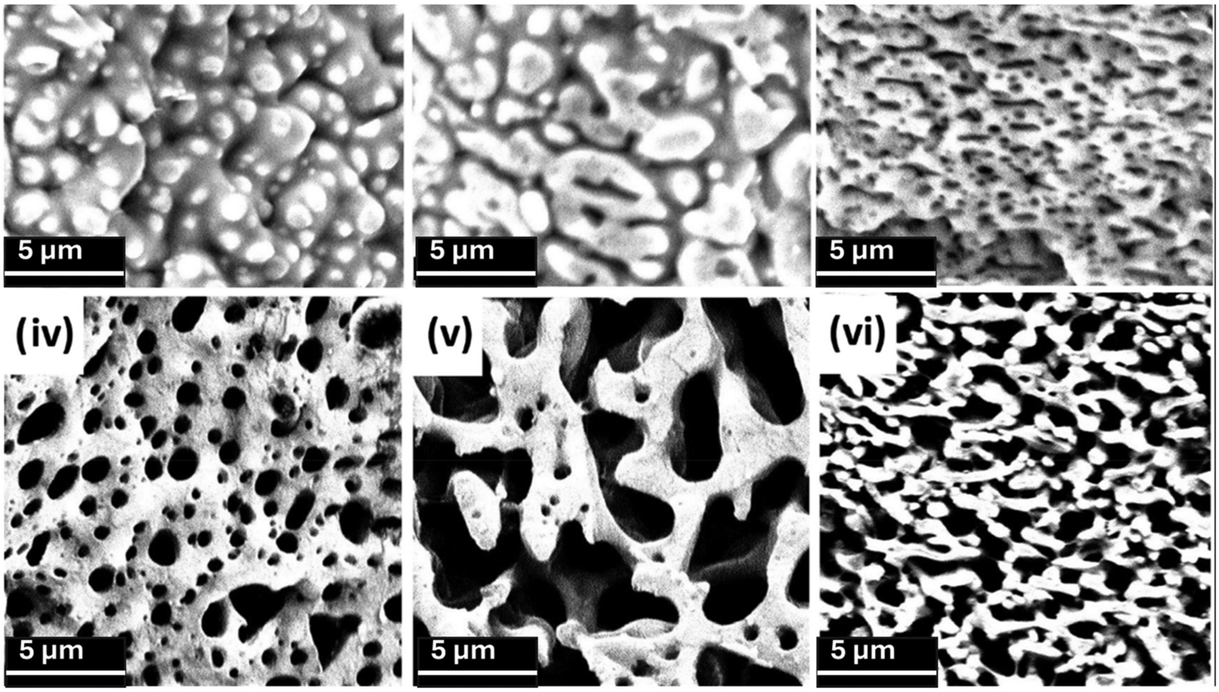


Fig. 7 SEM of impact fractured surfaces of PHBV/PBAT with different amounts of each polymer without any compatibilizer. A distinct phase-separated blend can be observed. (i) PHBV/PBAT (70/30), (ii) PHBV/PBAT (50/50), (iii) PHBV/PBAT (30/70), (iv) solvent-etched 70/30PHBV/PBAT, (v) solvent-etched 50/50PHBV/PBAT, (vi) PHBV/PBAT (30/70), after solvent etching.[74] Figure was reproduced from ref. 74 under Creative Commons CC license, copyright 2025.

development. The performance of the blend depends on factors like composition, processing methods, and the presence of additives. Studies have shown that by adjusting the ratio of PBAT to PHB, the properties of the material can be tailored for specific uses. For instance, a higher PBAT content generally enhances flexibility, while a higher PHB content increases stiffness and thermal resistance.

In addition to mechanical properties, the biodegradability of PBAT/PHB blends is a key focus. These blends degrade more effectively in natural environments compared with conventional plastics, thanks to the microbial breakdown of PHB and the partial biodegradability of PBAT. Researchers are exploring ways to optimize this biodegradation by studying the role played by soil microbiomes and isolating specific microorganisms that can accelerate the process.

Fernandes *et al.*[195] investigated the biodegradation of PHB/PBAT (45/55) polymer blends in soil and the isolation of novel microorganisms capable of degrading PBAT. The biodegradation was achieved at 27 °C in soil reactors that were inoculated with the soil. At the end of the biodegradation, the microorganisms that degrade PHB/PBAT were isolated. The result revealed that over a six-month incubation period, the bilayer PHB/PBAT film exhibited an average mineralization rate of 47 ± 1%, while the reference material (cellulose) reached 75 ± 1%.

The PHB/PBAT film demonstrated minimal biodegradation within the first 11 days, with less than 2% degradation observed. However, the degradation rate increased until day 52, after which it gradually declined. Additionally, CO_2 evolution data indicated higher organic carbon transformation into CO_2. Microorganisms such as *Streptomyces coelicoflavus*, *Clonostachys rosea* and *Aspergillus flavus* were found to degrade PHB, while *Purpureocillium lilacinum* and *Aspergillus pseudodeflectus* degrade PBAT, respectively, at mesophilic conditions.

Although PBAT and PHB are both biodegradable polymers, their compatibility and miscibility are limited. Generally, PBAT is more flexible and ductile, while PHB is more rigid and brittle. Their miscibility is limited due to differences in their chemical structures and intermolecular interactions.

Costa *et al.*[196] confirmed that PBAT and PHB were immiscible. In their research, PHB, PBAT, and a 1 : 1 blend was prepared, and the non-isothermal melt crystallization kinetics of the samples was determined using DSC and the Avrami, Ozawa, and Mo microkinetic models. They stated that the three models were unable to satisfactorily predict the experimental data for the PBAT/PHB blend. The DSC revealed an immiscible incompatible mix between PHB and PBAT due to their differing degrees of crystallinity, melting temperatures, and peak structures.

Additionally, PBAT/PHB blends with 25 wt%, 50 wt%, and 75 wt% of each component were prepared by Beber *et al.*[197] It was shown that blending PBAT with PHB inhibited the crystallization behaviour of both polymers. A similar phenomenon was also reported by Costa *et al.*[196] DSC results revealed the displacement of the PBAT melting peak by a higher temperature in the PBAT/PHB blends than the neat PBAT.

3.4 PBAT/PHA blend composite

PBAT is a biodegradable aliphatic aromatic polyester having good thermal and mechanical properties, especially impact and elongation at break, and it is a suitable toughening agent in brittle aliphatic polyesters.[198] The blends of biodegradable polyhydroxy alkanoantes (PHAs) and PBAT have been researched with immense use in different applications including packaging. To improve the final properties of this blend, researchers in the past decade have incorporated different natural fillers such as lignin,[199] babasu fiber,[200] hemp,[201] starch,[202] cellulose nanocrystal,[203] *etc*. Utilizing these materials allowed them to produce materials suitable for a variety of uses while also enabling the industrial and agricultural wastes to have value.[199,204]

Recently, some research has been documented on natural filler-reinforced PBAT/PHAs. Among this, Yolacan *et al.*[205] did a comprehensive study on PHBV/PBAT, PHBV/PBAT/PHA blends using polyethylene glycol (PEG 400) as plasticizer and adding different amounts of hydroxypropyl methylcellulose (HPMC) (H). Composite films were made through solution casting.

The PHBV/PBAT blend (10/90) exhibited significant mechanical property improvements with a tensile strength of 14 MPa, % elongation of 333%, and elastic modulus of 211 N mm^{-2}, but barrier properties were poor, with an OTR of 297 cc m^{-2} day^{-1} and WVTR of 17 g m^{-2} day^{-1}. The addition of 1% PEG 400, (polyethylene glycol) (P) (PHBV/PBAT/P) improved the tensile strength to 17 MPa, and % elongation increased to 374%, with a slight reduction in elastic modulus (201 N mm^{-2}). The barrier properties (OTR of 233 cc m^{-2} day^{-1} and WVTR of 13 g m^{-2} day^{-1}) were further improved. However, when HPMC at 3% (PHBV/PBAT/P/H3) was added, the maximum tensile strength of 22 MPa, elongation at break of 419%, and elastic modulus of 182 N mm^{-2} were observed without any significant changes in the barrier properties (OTR of 233 cc m^{-2} day^{-1}, WVTR of 13 g m^{-2} day^{-1}).[205]

The ternary PHBV/PBAT/PHA/P blend demonstrated enhanced mechanical properties, with tensile strength reaching 20 MPa, % elongation of 562%, and elastic modulus of 145 N mm^{-2}, but moderate barrier properties with an OTR of 197 cc m^{-2} day^{-1} and WVTR of 9 g m^{-2} day^{-1}. Increasing HPMC in this blend to 3% (PHBV/PBAT/PHA/P/H3) showed a balance, with tensile strength of 19 MPa, elongation at break at 558%, and an elastic modulus of 187 N mm^{-2}, while barrier properties were excellent (OTR of 197 cc m^{-2} day^{-1}, WVTR of 9 g m^{-2} day^{-1}). However, at 5% HPMC (PHBV/PBAT/PHA/P/H5), mechanical performance declined slightly to 17 MPa and 530% elongation at break, though barrier properties remained strong.[205]

Melendez-Rodriguez *et al.*[206] developed a compostable multilayer film for food packaging, combining polybutylene adipate terephthalate (PBAT) and polyhydroxyalkanoate (PHA) with a barrier layer consisting of CNC (cellulose nanocrystal) and an electrospun hot-tack adhesive made from PHBV. The film exhibited excellent oxygen barrier properties, reducing oxygen permeance by over 90% (from 9.3×10^{-15} to 0.5×10^{-15} $m^3\ m^{-2}\ s^{-1}\ Pa^{-1}$) due to the CNC layer. Water vapor permeance (WVP) was also reduced, with values between 2.0 and 3.6×10^{-12} kg m^{-2} s^{-1} Pa^{-1}. While the tensile strength remained stable (around 20 MPa), the CNC significantly reduced the film's flexibility, with elongation at break decreasing by over 90% from 330 and 243% in machine and transverse directions to 27 and 7.6%, respectively. Adhesion between layers was firm, though not very strong, with a peel strength of 0.006 N mm^{-1}. Migration tests showed compliance with food contact standards, and the film fully disintegrated in industrial composting within 60 days, making it a promising sustainable option for high-barrier food packaging.

3.5 Thermoplastic starch-based blends

Extensive research efforts have been directed toward the production of biodegradable starch-based materials for applications ranging from food packaging to potential use in biomedical field. However, the inherent thermodynamic immiscibility resulting from the hydrophobicity of PLA and the hydrophilicity of thermoplastic starch poses challenges, manifesting in poor interfacial adhesion and mechanical properties.[207] Research into biodegradable blends of PLA/TPS is continuously evolving, and new applications and processing methods that aid their compatibility are being reported.

The design of biodegradable polymers and TPS blends using molecular dynamics (MD) simulations to understand interfacial properties has revealed the dynamics of blending TPS with PLA, PBS, and PHB.[208] Four biodegradable polymers PLA, PBS, PHB, and PBAT were blended with unmodified (nTPS) and citrate-modified (cTPS) TPS. Results showed that PBS, PHB, and PBAT diffused effectively into TPS, forming strong interfaces, while PLA exhibited poor compatibility. PBS and PBAT, particularly when combined with cTPS, demonstrated the highest interfacial fracture energy, indicating robust adhesion due to deep molecular interdiffusion, suppression of voids through electrostatic interactions, and energy absorption through molecular chain conformations. PLA and PHB, with weaker electrostatic interactions, were more prone to interfacial fractures. Diffusion analysis revealed that PBS and PBAT penetrated TPS more effectively, especially with cTPS, whereas PLA showed minimal diffusion.

A novel plant-based compatibilizer—pyrogallic acid (PGA)—was used in PLA/TPS blends with glycerol (4/3/1 wt/wt) as plasticizer *via* a one-step twin-screw extrusion. The results showed that PGA significantly improved the blend's mechanical properties, including tensile strength, elongation at break, and thermal stability. Specifically, incorporating 1.5 phr. of PGA yielded the highest tensile strength (23.38 MPa) and elongation at break (16.96%), with 24.7% and 233.2% improvements over neat PLA/TPS blends. Additionally, PGA enhanced the blend's crystallinity, water resistance, and interfacial bonding between PLA and TPS due to stable hydrogen bonding and dehydration-induced crosslinking during extrusion.[209]

The performance of TPS and PBAT blends was enhanced through controlled grafting of a reactive compatibilizer *via* a two-step blending process, adjusting the grafting density of PBAT onto poly(ethylene glycol methyl ether acrylate-*co*-glycidyl methacrylate) (EG) compatibilizer (25 : 25 : 1). By varying the pre-grafting time (0, 5, 10, 20, and 40 minutes), the compatibilizer's location within the blend could be controlled. It was shown that the blend with PBAT grafted for 20 minutes demonstrated improved interfacial compatibility, with the smallest TPS domain size and the highest tensile strength (12.2 MPa), storage modulus, and complex viscosity that suggested effective adhesion. Additionally, B20 exhibited superior water barrier properties and the best balance between strength and flexibility.

Similarly, through dynamic vulcanization, the properties of PBAT/TPS biodegradable composite blow molded films were enhanced. Corn starch was plasticized and crosslinked using 1,3,5-tri-2-propenyl-1,3,5-triazine-2,4,6 (1*H*,3*H*,5*H*)-trione (TAIC). The crosslinked TPS was blended with PBAT, improving their compatibility. At optimum content of 2 wt% TAIC and 30 wt% TPS addition, there was significant improvement in tensile strength (25.43 MPa) and elongation at break (580.83%) due to co-crosslinking of TAIC with TPS and TPS/PBAT. Additionally, the blend demonstrated better thermal stability and biodegradability, retaining good processing properties while showing enhanced performance in soil degradation tests.[72]

The potential application of thermally activated PLA/TPS with enhanced shape memory in biomedicine has been studied by Sessini *et al.*[210] Maleic acid-functionalized oligomeric lactic acid (mOLA) and neat oligomeric lactic acid (OLA) were incorporated as plasticizers (20%). The functionalization of OLA resulted in phase incompatibility and failed to impart the shape memory property to the blend, resulting from the loss of fixed and interchanging phase for shape memory response. However, PLA/TPS with OLA exhibited excellent shape memory properties with a temperature of 45 °C and deformation of 50%. Balancing the plasticizer and compatibilizer is critical for maintaining distinct polymer phases necessary for shape memory function.

Songtipya *et al.*[211] reported on improving the thermomechanical properties of thermoplastic TPS and natural rubber (NR) blends by incorporating polyethylene glycol (PEG) and modified natural rubber such as poly(butyl methacrylate)-grafted natural rubber, poly(methyl methacrylate)-grafted natural rubber and epoxidized natural rubber (NR-*g*-PBMA, NR-*g*-PMMA, and ENR50N). The impact of different TPS/NR blend ratios, PEG content, and types of modified NR on the blends' mechanical, thermal, and biodegradation properties were investigated. Results showed that adding modified NR significantly improved the flexibility and toughness of TPS, with the highest toughness observed in the TPS/ENR50 blend (1628 MJ m^{-3}) containing 1.0 wt% PEG. However, excess PEG reduced mechanical properties due to phase separation. Biodegradation tests revealed that the TPS/NR blends biodegraded over 95% within 120 days.

Ávila-Orta *et al.*[212] utilized reactive extrusion (REX) to thermoplastify native cassava starch. They performed *in situ* chemical modifications by oxidizing the starch with sodium hypochlorite to produce oxidized starch (OS), which was then processed *via* REX to obtain oxidized thermoplastic starch (O-TS). Additionally, malleated thermoplastic starch (MA-TS) was produced by REX processing of native cassava starch with maleic anhydride (MA) and Luperox 101. A dual-modified thermoplastic starch (O-MA-TS) was created by REX of OS with 2% MA and 0.1% Luperox 101. This process resulted in four types of thermoplastic starch: unmodified thermoplastic starch (TS), oxidized thermoplastic starch (O-TS), malleated thermoplastic starch (MA-TS), and dual-modified thermoplastic starch (O-MA-TS). The obtained TS were blended with PLA at 85% and 65% (w/w) *via* melt extrusion and subsequently used to produce non-woven fabrics through melt-blowing. According to the SEM findings, the blend with the unmodified TS was immiscible with the PLA, causing large, extremely coarse TS droplets due to phase separation and weak interface bonding. Many other researchers have developed TPS-based blends using different additives that have resulted in interesting findings, as can be seen in Fig. 8.

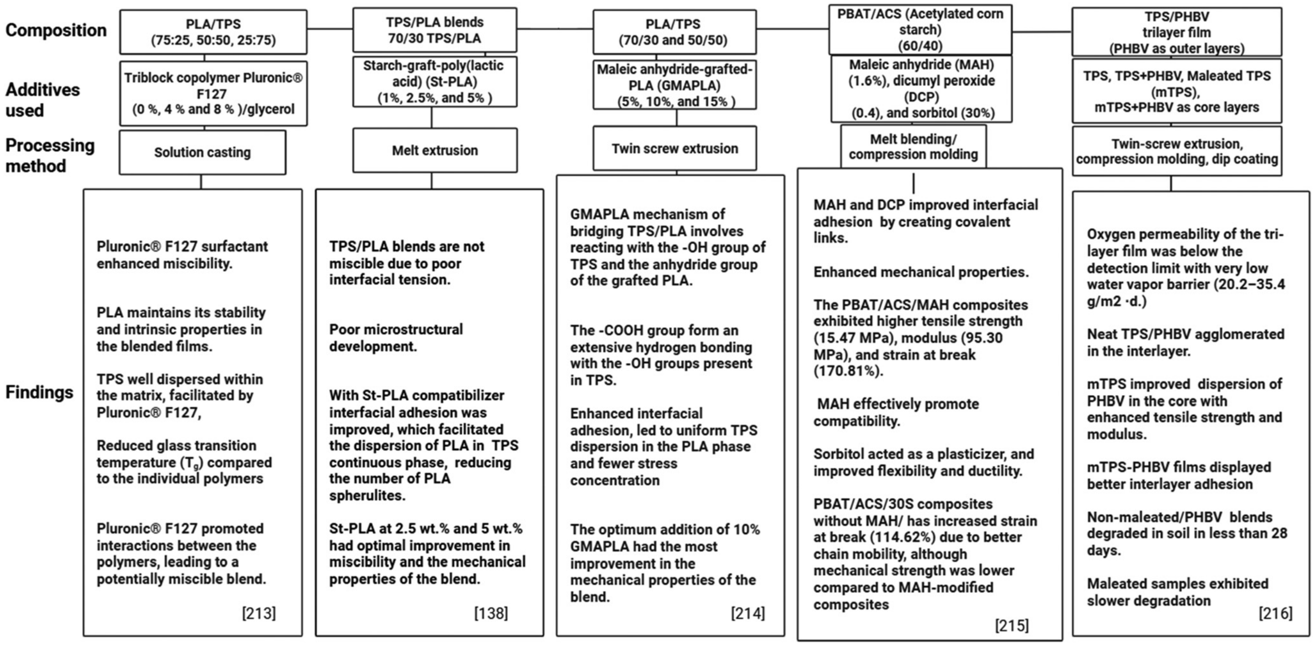
On the other hand, the dual-modified 65PLA/TS blend and the modified 85PLA/TS blend showed better compatibility, which led to a reduced TS phase with a homogeneous shape. The DTG result suggested improved compatibility and interaction between TS and PLA, especially the dual-modified blends exhibiting a shaper peak in the DTG curve.

3.5.1 TPS-based blend composites. Across different studies, fiber reinforcement was shown to improve mechanical properties, thermal stability, and biodegradability. However, high fiber content sometimes reduced tensile strength, making compatibilizers like maleic anhydride necessary to enhance fiber–matrix interaction.

In research on PLA reinforced with rice straw (RS) at 10%, 20%, and 30% weight fractions, the tensile strength decreased as RS content increased, but the addition of maleic anhydride (MA) improved fiber–matrix bonding, resulting in tensile strength increasing by 20% and flexural strength by 14% at 30% RS content. RS fibers increased hydrophilicity, but MA reduced water absorption, improving the material's performance in humid environments.[217]

For PLA–starch laminates reinforced with RS fibers and active extracts, the incorporation of RS significantly improved tensile and flexural properties.[218] These bilayer laminates demonstrated enhanced water vapor and oxygen barrier and fully degraded within 90 days in composting conditions, showing excellent biodegradability, further enhanced by the RS fibers.[218] Similarly, for PBAT-based biodegradable films with thermoplastic starch (TPS) and tea polyphenols (TP), films with 30% TPS degraded fully after 180 days, with increased hydrophilicity and water vapor permeability.[219] Higher TPS content led to enhanced TP release, improving the films' antioxidant and antimicrobial properties, though tensile strength decreased with more TPS.

Aouay *et al.*[220] added lignin-containing cellulose nanofibrils (LCNFs) to a blend of film PBAT and TPS (PBAT/TPS) to reduce the water sensitivity of the blend. By adding 10 wt% LCNFs the tensile strength and modulus were improved due to enhanced



Composition	PLA/TPS (75:25, 50:50, 25:75)	TPS/PLA blends 70/30 TPS/PLA	PLA/TPS (70/30 and 50/50)	PBAT/ACS (Acetylated corn starch) (60/40)	TPS/PHBV trilayer film (PHBV as outer layers)
Additives used	Triblock copolymer Pluronic® F127 (0 %, 4 % and 8 %)/glycerol	Starch-graft-poly(lactic acid) (St-PLA) (1%, 2.5%, and 5%)	Maleic anhydride-grafted-PLA (GMAPLA) (5%, 10%, and 15%)	Maleic anhydride (MAH) (1.6%), dicumyl peroxide (DCP) (0.4), and sorbitol (30%)	TPS, TPS+PHBV, Maleated TPS (mTPS), mTPS+PHBV as core layers
Processing method	Solution casting	Melt extrusion	Twin screw extrusion	Melt blending/ compression molding	Twin-screw extrusion, compression molding, dip coating
Findings	Pluronic® F127 surfactant enhanced miscibility. PLA maintains its stability and intrinsic properties in the blended films. TPS well dispersed within the matrix, facilitated by Pluronic® F127, Reduced glass transition temperature (T_g) compared to the individual polymers Pluronic® F127 promoted interactions between the polymers, leading to a potentially miscible blend. [213]	TPS/PLA blends are not miscible due to poor interfacial tension. Poor microstructural development. With St-PLA compatibilizer interfacial adhesion was improved, which facilitated the dispersion of PLA in TPS continuous phase, reducing the number of PLA spherulites. St-PLA at 2.5 wt.% and 5 wt.% had optimal improvement in miscibility and the mechanical properties of the blend. [138]	GMAPLA mechanism of bridging TPS/PLA involves reacting with the -OH group of TPS and the anhydride group of the grafted PLA. The -COOH group form an extensive hydrogen bonding with the -OH groups present in TPS. Enhanced interfacial adhesion, led to uniform TPS dispersion in the PLA phase and fewer stress concentration The optimum addition of 10% GMAPLA had the most improvement in the mechanical properties of the blend. [214]	MAH and DCP improved interfacial adhesion by creating covalent links. Enhanced mechanical properties. The PBAT/ACS/MAH composites exhibited higher tensile strength (15.47 MPa), modulus (95.30 MPa), and strain at break (170.81%). MAH effectively promote compatibility. Sorbitol acted as a plasticizer, and improved flexibility and ductility. PBAT/ACS/30S composites without MAH/ has increased strain at break (114.62%) due to better chain mobility, although mechanical strength was lower compared to MAH-modified composites [215]	Oxygen permeability of the tri-layer film was below the detection limit with very low water vapor barrier (20.2–35.4 g/m2 ·d.) Neat TPS/PHBV agglomerated in the interlayer. mTPS improved dispersion of PHBV in the core with enhanced tensile strength and modulus. mTPS-PHBV films displayed better interlayer adhesion Non-maleated/PHBV blends degraded in soil in less than 28 days. Maleated samples exhibited slower degradation [216]

Fig. 8 Thermoplastic starch-based blends incorporating different additive and compatibilizers.[138,213–216]

interaction between the TPS and PBAT phases. There was a reduction in moisture sorption, with up to a 50% decrease in moisture uptake, to achieve an effective reduction in water sensitivity. At a 70/30 PBAT/TPS ratio, adding LCNFs altered the film's morphology. Without LCNFs, TPS dispersed as small, uniform 4–5 μm nodules. As LCNF content increased, TPS particle size grew irregularly, reaching 7, 14, and 20 μm for LCNF contents of 6, 8, and 10 wt%, respectively (Fig. 8).

The addition of 5% and 10% cotton fibers to PLA/TPS composites increased the mechanical properties, with 85% PLA, 10% TPS, and 5% cotton fiber showing the highest water absorption due to increased starch content. Cotton fibers slightly improved the hardness and strength of the composites while maintaining biodegradability.[221]

The study on mechanical and thermal properties of coir fiber (CF)-reinforced thermoplastic starch (TPS) and poly(butylene adipate-*co*-terephthalate) (PBAT) composites showed significant improvements in properties with increasing CF content (5%, 10%, 15%, and 20%).[222]

At 20 wt% CF with alkali treatment, tensile strength improved by 393%, and flexural strength increased by 536% compared with the base blend (Fig. 10). Fiber surface treatment resulted in less pullout and increased fiber–matrix adhesion (Fig. 9). The addition of coir fibers also enhanced thermal stability, as the composites exhibited higher glass transition temperatures and storage modulus, indicating improved thermal performance.[222]

3.6 PBS, bio-PBS, bio-PBSA based blends

Polybutylene succinate (PBS) is a biodegradable aliphatic polyester with properties that are comparable to polypropylene. It is produced from succinic acid and 1,4 butanediol.[224] PBS can be synthesized through bio-based and petroleum-based pathways. Its biodegradability, biocompatibility, and promising mechanical properties make it suitable for a variety of applications, especially in environmentally friendly packaging and agricultural films. PBS offers good thermal stability, tensile strength, and flexibility compared with other bioplastics. Its biodegradability can be further enhanced when blended with other biopolymers like PLA or TPS.[225,226] While PBS is primarily petroleum based, research efforts are increasingly focused on producing bio-based PBS (bio-PBS) from renewable sources such as bio-succinic acid, to enhance its sustainability appeal.

Many studies that incorporate PBS, bio-PBS, and bio-PBSA were driven by the need to develop innovative materials that enhance mechanical performance, improve degradation properties, reduce environmental impact, and create cost-effective solutions in packaging, textiles, and biomedical applications.

One study investigated the effects of blending PBS with PLA and adding an epoxy-functionalized compatibilizer to improve mechanical and barrier properties.[227] The addition of organo-montmorillonite Delite®43B (D43B) and random ethylene-methyl acrylate-glycidyl methacrylate terpolymer (ax89) reactive compatibilizer was crucial to improving the oxygen barrier properties of nanocomposite blown films of PBS/PLA. At 3 wt% addition of D43B, the oxygen permeability was reduced by more than 50% and 5 wt% compatibilizer further enhanced oxygen barrier performance and improved elongation at break, in the transverse direction. The blending sequence played a crucial role in determining the compatibility of PBS, PLA, and D43B to influence the mechanical and barrier performance.

The microstructure and mechanical performance of PLA/PBS melt-blown nonwovens has been enhanced by *in situ* PBS fibrillation technology.[228] First a sea-island morphology in

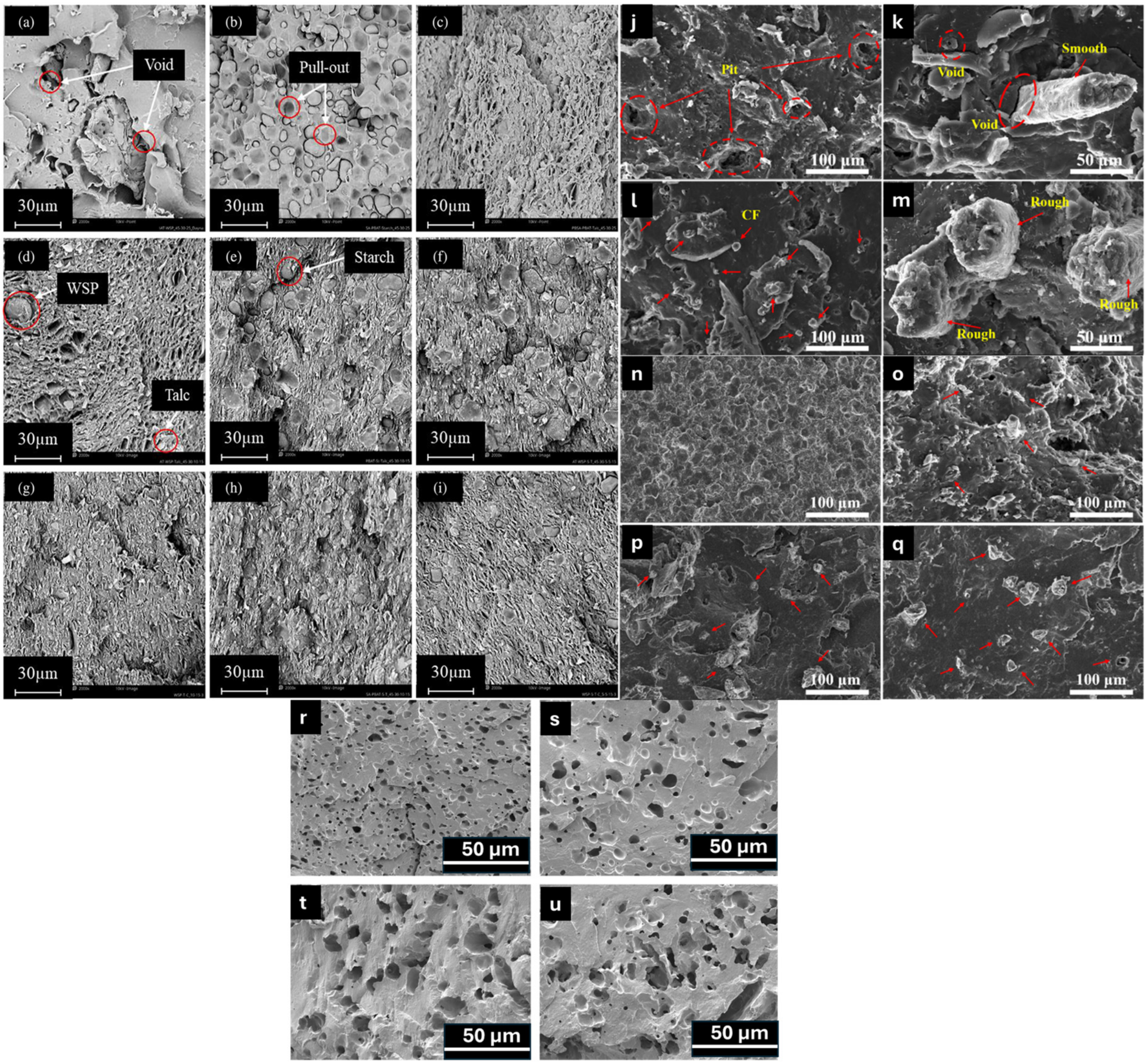


Fig. 9 Scanning electron microscopy image showing the influence of different filler and surface treatment on the morphology of TPS-based and PBSA-based blend composites. (a) PBSA-PBAT-WSP, (b) PBSA-PBAT-St, (c) PBSA-PBAT-T, (d) PBSA-PBAT-WSP-T, (e) PBSA-PBAT-St-T, (f) PBSA-PBAT-WSP-St-T, (g) PBSA-PBAT-WSP-T-Compatibilizer, (h) PBSA-PBAT-St-T-compatibilizer, and (i) PBSA-PBAT-WSP-St-T-compatibilizer, (j and k) tensile fracture surface of 20 wt% non-surface-treated coir-fiber reinforced TPS/PBAT, (l and m) tensile fracture surface of 20 wt% alkaline-treated coir fiber-reinforced TPS/PBAT, (n and o) tensile fracture surface of 5 wt% non-surface-treated coir-fiber reinforced TPS/PBAT, (p and q) tensile fracture surface of 5 wt% alkaline-treated coir fiber-reinforced TPS/PBAT composite, cryogenic fractured PBAT-TPS (70–30) containing different lignin-containing cellulose fiber (r) 0 wt%, (s) 6 wt%, (t) 8 wt%, and (u) 10 wt%. Figures a–i were reproduced from ref. 223 and figures j–q were reproduced from ref. 222 with permission from Wiley, copyright 2025. Figures r and s were reproduced from ref. 220 with permission from American Chemical Society, copyright 2025.

PLA/PBS blends was formed due to their immiscibility, where PBS appeared as droplets in the PLA dispersed phase. The PBS droplets were stretched into *in situ* fibrils under elongational flow during the melt-blown process. The fibrillation process significantly improved the tensile strength and elongation of the blend by 164%, and 672% compared with pure PLA nonwovens. This improvement in mechanical performance, alongside enhanced air and moisture permeability, suggests suitable application of the modified nonwovens in commercial packaging.[228] Table 10 lists some recent studies on PBS-based biodegradable polymer blends additives and key findings.

Blending starch with PBS is a low-cost strategy to stem the high cost of PBS. Rajendran *et al.*[237] argued that PBS is very difficult to market because it is more expensive than fossil-based plastics. Modifying and blending PBS with other polymers could open new applications that reduce unit costs. According to Li *et al.*[238] blending of cassava starch and PBS can be a low-cost PBS approach for developing biocomposites that can replace general-purpose plastics in daily application. For example, a study on modified TPS/PBS/epoxy resin blends suggested that biodegradable modified TPS/PB/epoxy resin blends can be applied in agriculture, packaging and medical

Table 10 PBS-based biodegradable polymer blends

Polymers/additives used	Processing method	Key findings	Ref.
PLA/PBAT/PBS, peroxide, carbodiimide	Reactive extrusion with peroxide and carbodiimide, blown film extrusion	• Reactive extrusion improved rheological properties, for blown film extrusion • Films exhibited tensile strengths up to 57 MPa for tubular films and 53 MPa for champagne films at 30 wt% PBS • Seal strength and heat resistance improved with increasing PBS content, suitable for packaging applications involving warm and hot processed meals	229
PLA/PBS (70/30 blend)	Film extrusion, degradation tested in freshwater and seawater	• PLA degraded most rapidly in warm, light-exposed conditions, generating microplastics • PBS/PLA 70/30 blends showed greater resistance to degradation, without microplastics in cold, dark conditions, indicating slower degradation rates	230
PLA/PBS fibers	Fiber extrusion for textile applications	• Increasing PBS content enhanced fiber elasticity and mechanical properties • A 7% PBS blend showed a 2.8-fold increase in crystallinity compared with pure PLA • Excessive PBS content caused phase separation, reducing mechanical stability	231
PHB/bio-based PBS	Blend preparation for pliable scaffold substrates	• 50 : 50 PHB/PBS blend showed optimal ductility and hydrophobicity, making it suitable for tissue engineering applications • The blend exhibited enhanced degradation stability and reduced swelling, ideal for biomedical bone tissue scaffold applications	232
PBS/PHA	Marine and compost degradation testing	• PBS/PHA blends exhibited faster degradation in compost sediment environments (39.5% in 45 days) compared with pure PBS (31.9%) • In marine environments, PBS/PHA degraded by 25.9% and pure PBS by 20.3% in 180 days, making the blend a promising alternative for reducing plastic pollution	233
PHBV/PBSA/maleic anhydride-grafted PBSA (PBSA-*g*-MAH)	Reactive compatibilization	• Maleic anhydride grafting improved phase interaction, ductility, toughness, and barrier properties, making the blend suitable for commercial green food packaging applications	234
PHBV/PBSA/epoxy-functionalized chain extender JoncrylÂ® ADR-4468	Reactive compatibilization with epoxy-functionalized chain extender (Joncryl)	• The chain extender improved polymer interaction • Reduced dispersed phase domain size, and increased processability • Elongation improved by 45%, transforming the material from brittle to flexible, though impact strength remained unchanged	235
PBS/PGA/ESOn-ECD	Compatibilization using epoxidized soybean oil branched cardanol ether (ESOn-ECD)	• Adding ESOn-ECD improved compatibility between PBS and PGA, reducing the dispersed phase diameter from 2.74×10^{-14} to 1.97×10^{-14} g cm cm^{-2} s^{-1} Pa^{-1} • Tensile strength increased from 15.3 MPa to 19.4 MPa, and elongation increased from 244.5% to 449.0% • Water vapor barrier properties improved significantly	236

fields because of their improved moisture resistance as well as thermal and mechanical properties.[239]

3.6.1 PBS, bio-PBA, bio-PBSA-based composites. Pei *et al.*[240] investigated four types of lignocellulosic biomass fibers—bagasse, bamboo, rice husk, and rice straw—as filler in a PHB/PBS (7 : 3) matrix to develop bio-composites. The fibers were treated with NaOH to enhance fiber–matrix bonding. The mechanical properties were significantly affected by the type of filler. Bamboo fiber addition to PHB/PBS performed best compared with other filler types, with a bending strength of 19.82 MPa, tensile strength of 12.97 MPa, and impact strength of 4.30 kJ m^{-2}. The bamboo composites were thermally stable, with an initial thermal degradation temperature of 248 °C and a residue of 5.81% after thermal degradation. However, rice straw-based composites performed the poorest, with the lowest mechanical and thermal properties.[240]

Micro fibrillated cellulose (MFC) (0.5%, 0.75%, and 1%) was incorporated into a PLA/PBSA blend for packaging applications. It was found that 0.75% MFC provided the best balance of mechanical, thermal, and barrier properties. The elastic modulus increased as MFC content increased, and the elongation at break peaked at 0.75% MFC, achieving 117% elongation. The addition of MFC also reduced oxygen permeability by 28%, improving the film's barrier properties, although water vapor permeability slightly increased due to the hydrophilic nature of MFC. Cardanol improved MFC dispersion, preventing agglomeration and enhancing film performance. The hot tack strength at 0.75% MFC reached 925 g/15 mm, making it the optimal concentration for achieving a balance of flexibility, strength, and barrier properties, which are critical for sustainable packaging applications.[241]

The injection-molded blend of biodegradable PBSA/PBAT (60 : 40) polyester blends with WSP (walnut shell powder), St (corn starch), and talc (T) was studied by McNeill *et al.*[223] Compatibilizer such as maleic anhydride grafted PBSA (MA-*g*-PBSA) was added to improve the compatibility between the different components in the composite.

The addition of 25 wt% talc, WSP, and starch increased the tensile modulus by 234%, 101% and 66% compared with the neat PBSA/PBAT blend (Fig. 10). The flexural modulus also increased by 190%, 97%, and 75%, respectively. These increments were related to the filler type, size, orientation, and shape, with significant contributions to the morphology of the composite. Adding 10 wt% WSP and 15 wt% talc and 5 wt% WSP with 5 wt% St and 15 wt% talc hybrid fillers, the tensile modulus increased by 160% and 162%, while a 147% and 153% increase was observed in the flexural modulus. However, tensile strength decreased. A 25% starch or WSP decreased viscosity and increased water absorption due to the hydrophilic nature of starch.

A dual mechanism involving simultaneous reinforcement with harakeke fiber and compatibilization with dicumyl peroxide (DCP) to improve the mechanical properties of PLA/PBS was proposed by Akindoyo *et al.*[242] The blends without DCP/fiber were observed to have poor miscibility, leading to reduced tensile strength. The addition of harakeke fiber did not significantly improve strength and modulus (Fig. 11). However, the addition of dicumyl peroxide (DCP) as a compatibilizer and harakeke fiber as reinforcement led to a 31% increase in tensile strength and a 148% increase in tensile modulus, with the crystallinity of the blend increasing by 201%. This dual strategy of reinforcement and compatibilization resulted in significantly enhanced mechanical properties compared with unreinforced blends, making it an effective method for improving the performance of biodegradable materials.

Ketata *et al.*[243] hybridized flax fibers (FF) and glass fibers (GF) for reinforcement in a PLA–PBS matrix. The key finding was that hybrid FF/GF resulted in improved mechanical properties compared with using each fiber type alone. The tensile strength of the hybrid composites increased from 42.4 MPa (FF-reinforced composites) to 53 MPa when glass fibers were added. The tensile modulus also improved, increasing from 4.9 GPa for the FF-composite to 5.4 GPa in the hybrid system. However, the addition of glass fibers slightly reduced the elongation at break from 1.7% to 1.5%. Fiber length also played a significant role in reinforcement; while 27% of flax fibers exceeded their critical length in isolation, 34% of fibers surpassed it in the hybrid composite. The glass fibers saw a similar effect, with their percentage increasing from 4% to 19% in the hybrid composite.

In the development of PBS/PLA nanocomposite blown films, Adrar *et al.*[227] added organo-montmorillonite (D43B) and a reactive compatibilizer (ax89), aiming to enhance oxygen barrier properties while maintaining mechanical integrity. Results showed that adding 3 wt% D43B to the PBS/PLA blend reduced oxygen permeability by over 50%, particularly when D43B was localized in the PLA phase. The presence of 5 wt%

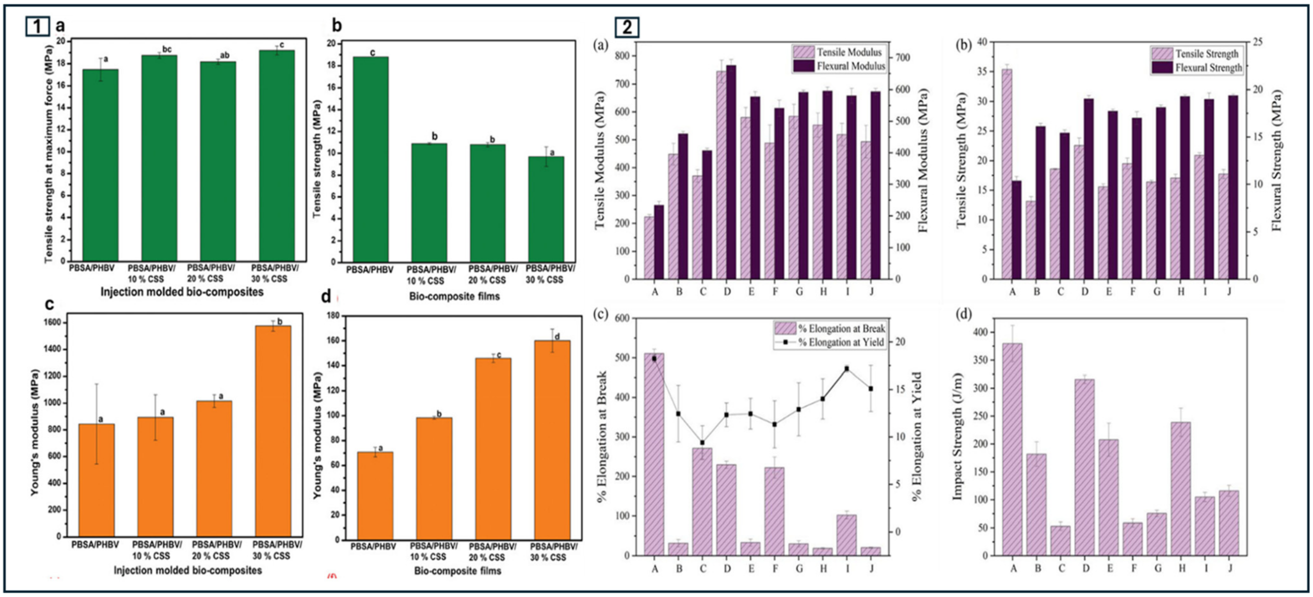


Fig. 10 Mechanical properties of PBS, PBSA and bio-PBSA composites. (1) Composites of PBSA/PHBV reinforced with cowpea lignocellulosic fiber (a) tensile strength of injection molded composite, (b) tensile strength of film (c) Young's modulus of injection molded composite, (d) Young's modulus of film composite. (2) PBSA/PHBV with different fillers (a) tensile/flexural modulus, (b) tensile strength/flexural strength, (c) % elongation at break/yield (d) impact strength. (A) PBAT, (B) PBSA-PBAT-WSP, (C) PBSA-PBAT-St, (D) PBSA-PBAT-T, (E) PBAS-PBAT-WSP-T, (F) PBSA-PBAT-St-T, (G) PBSA-PBAT-WSP-St-T, (H) PBSA-PBAT-WSP/T-compatibilizer, (I) PBSA-PBAT-St-Talc-compatibilizer, and (J) PBSA-PBAT-WSP-St-T-compatibilizer. (1) was reproduced from ref. 244 under Creative Commons CC license, copyright 2025 and (2) was reproduced from ref. 223 with permission from Wiley, copyright 2025.

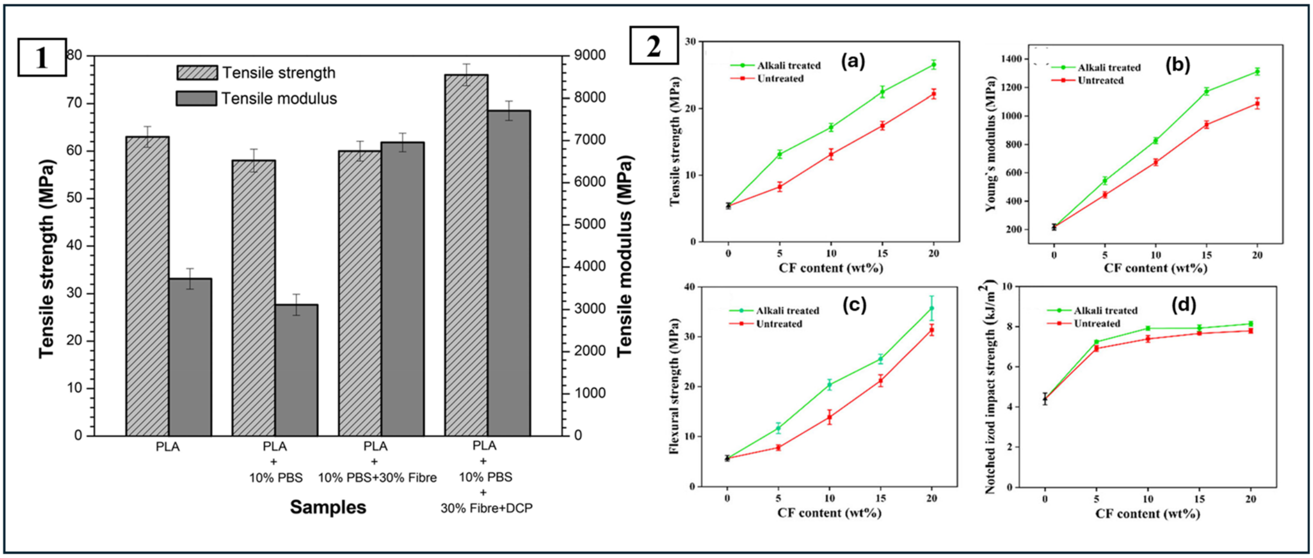


Fig. 11 (1) Tensile strength and tensile modulus of harakeke fiber-reinforced PLA/PBS compatibilized with dicumyl peroxide (DCP), (2) mechanical properties of alkaline-treated coir fiber-reinforced thermoplastic starch/poly(butylene adipate-*co*-terephthalate) composite (a) tensile strength, (b) Young's modulus, (c) flexural strength, (d) notched Izod impact strength. (1)was reproduced from ref. 242 under Creative Commons CC license copyright 2025 and (2) was reproduced from ref. 222 with permission from Wiley, copyright 2025.

compatibilizer further improved the oxygen barrier performance. The addition of the compatibilizer ax89 increased elongation at break in the transverse direction, while D43B reduced elongation in the machine direction. The blending sequence significantly influenced the compatibility between PBS, PLA, and D43B, impacting both barrier and mechanical properties.[227] Masanabo *et al.*[244] investigated the use of cowpea lignocellulosic fibers as a sustainable filler in bio-composites of PBSA/PHBV for rigid and flexible packaging applications. Composites were prepared by injection molding and film casting. The addition of 10, 20, and 30% of the cowpea fibers increased the tensile strength (17.46 MPa) of injection-molded composites at 30% fiber loading (Fig. 10).

However, for film-cast bio-composites, tensile strength decreased from 18.8 MPa to about 10 MPa due to pore formation. Young's modulus increased with fiber content in both injection-molded and film-cast samples, while the tensile strain decreased as fiber addition restricted polymer chain mobility. The thermal analysis showed a reduction in the onset degradation temperature by up to 30 °C for the 30% fiber-loaded composites.[240] Interestingly, water vapor permeability (WVP) and oxygen permeability (OP) were only minimally affected by the fibers, with 20% and 30% fiber bio-composites showing little change compared with fiber-free films.[244]

4. Role played by inorganic, mineral fillers and nanoparticles in biodegradable blends/composites

There are various types of fillers used in biodegradable composites, each with their unique properties, necessitating their choice of use (see Table 11). Inorganic, mineral, and nanoparticle fillers are widely studied for their ability to enhance stiffness, strength, toughness, flame retardancy, and UV resistance of biodegradable composites, making them suitable for more demanding structural and functional applications.

4.1 Inorganic and mineral fillers in biodegradable blends

Significant studies on nanoclay and talc reinforcement in biodegradable composites have been conducted recently aiming to improve their mechanical performance and thermal properties. Nanoclays are efficient for their large surface area and layered structures, providing excellent reinforcement, barrier and mechanical strength to composites. Conversely, talc, a phyllosilicate mineral, is a 2 : 1 tri-octahedral layered silicate complex that consists of an octahedral brucite ($Mg(OH)_2$) layer sandwiched between two tetrahedral silica sheets (Si_2O_5).[245] These minerals have the potential to enhance properties such as barrier and mechanical qualities, making them attractive reinforcing fillers in different industrial applications such as food packaging.

Blend films of biodegradable poly(butylene adipate-*co*-terephthalate) (PBAT), poly(lactic acid) (PLA), and calcium carbonate ($CaCO_3$) were produced in a one-step melt-blending and film-blowing method. The impact of $CaCO_3$ content and particle size on the properties of these films was evaluated.[246] The results showed that increasing the $CaCO_3$ content improved the films' rheology, tensile performance, and tear resistance. The moisture and oxygen barrier values decreased after incorporating $CaCO_3$ particles. The biodegradable PBAT/PLA films with $CaCO_3$ fillers had excellent mechanical, oxygen, and moisture barrier properties, making them a potential option

Table 11 Filler categories used in biodegradable blends/composites[244–250]

Filler category	Examples	Pros	Cons	Recommended conditions for use	Ref.
Natural fillers	Turmeric, cinnamon, coffee grounds	Enhances biodegradability and adds antioxidant properties	May degrade under high heat; colour may affect aesthetics	When biodegradability and bioactivity are priorities (*e.g.*, agriculture films)	160, 167, 185, 248 and 249
	Rice straw, Flax, Kenaf, Jute, Abaca bagasse, Coir Hemp *etc.*	Improves mechanical strength, low cost, biodegradable	Inconsistent quality due to natural variation	In applications needing reinforcement and biodegradability	187, 189, 217, 222, 240, 243 and 255
	Cellulose (MCC, CNCs, CNFs)	Reinforces structure, increases thermal stability	Dispersion issues, may require compatibilizers	Where structural reinforcement and barrier enhancement are needed (*e.g.*, food packaging)	175, 241, 256–258
Inorganic/mineral fillers	Talc, calcium carbonate, hydroxyapatite	Improves stiffness and heat resistance	Can reduce flexibility; may increase weight	When stiffness and thermal resistance are needed (*e.g.*, containers)	250, 251, 252 and 259
	Silica, montmorillonite	Improves barrier properties and nucleation effect	Can lead to agglomeration if not well dispersed	For high-barrier applications (*e.g.*, sealed food packaging)	260 and 261
Nanoparticles	Carbon nanotube, graphene oxide, graphene, carbon dots, metal oxides, carbon black	Enhances mechanical, thermal, and barrier properties at low loadings	Costly, requires complex surface modification	For high-performance applications requiring minimal filler loading	253, 254, 262–264

for food packaging and mulch film in agriculture. Similarly, PLA and inorganic (calcium carbonate (CaCOR$_3$R)) was shown to influence the mechanical, thermal, morphological, and surface properties of PBAT/PLA blend and composites.[247] The incorporation of CaCOR3R increased elongation at break compared with PLA, while the blends exhibited a higher elastic modulus, resulting in greater stiffness than the PBAT matrix.

Wang *et al.*[259] evaluated the influence of lignin, calcium carbonate ($CaCO_3$) (PPC), wollastonite (PPW), and talc (PPT) on the properties of PLA/PBAT composites. These fillers improved the mechanical, thermal, and microstructural properties of the composites to varying degrees. The study found that PPC ($CaCO_3$) composites exhibited the best mechanical properties, with tensile strength improved by 15%, impact strength by 27.9%, and bending strength by 32.6%. Thermal analysis revealed that both $CaCO_3$ and wollastonite enhanced thermal stability, with PPC and PPW showing higher crystallinity, particularly for PLA (22.2% for PPC and 24.5% for PPW). Talc, however, decreased thermal stability. These findings suggest that using these fillers, especially $CaCO_3$ and wollastonite, enhances both the mechanical and thermal performance of biodegradable PLA/PBAT composites, making them suitable for various applications.

In the presence of Joncryl chain extender, tensile strength and modulus of PBAT/post-industrial wheat starch (PPWS) increased by 5 and 517%. At the same time, talc functioned as a nucleating material to increase thermal stability of the composites.[265]

Nanoclay, as earlier mentioned, has great potential as a filler in biodegradable polymer blends. The influence of montmorillonite (MMT) (2 and 5 phr) on the mechanical and thermal properties PBAT/TPS was studied by Peidayesh *et al.*[260] Results showed that adding MMT nanoparticles slightly decreased tensile strength, with tensile stress dropping from 21.3 MPa for the 10% TPS blend to 18.7 MPa with 2% MMT and 18.8 MPa with 5% MMT. However, MMT increased the Young's modulus from 68.7 MPa (no MMT) to 76.0 MPa with 2% MMT, indicating a stiffer material. Elongation at break decreased with MMT addition, confirming the material's increased brittleness. Dynamic mechanical thermal analysis (DMTA) revealed that MMT slightly restricted the mobility of the glycerol-rich regions in the TPS domains, improving stiffness but reducing flexibility.[260]

4.2 Nanoparticles in biodegradable blends

Nanoparticles are also being applied to reinforce biodegradable polymers, improving their performance for a variety of uses, alongside mineral and inorganic fillers. Nanoparticles are incorporated into polymer blends to improve their properties, especially in the field of food packaging. For example, Khonakdar *et al.*[261] investigated how nanosilica can improve thermal stability and reduce thermo-oxidative degradation of PLA/PBAT blend nanocomposites. The PLA/PBAT blends (90/10 and 75/25 wt/wt) were prepared by adding 1, 3, and 5 phr hydrophilic (HPL) nanosilica and hydrophobic (HPB) nanosilica. The addition of HPB nanosilica improved the thermal stability of nanocomposite. The blend containing 5 phr. HPB nanosilica exhibited the highest degradation activation energy. It was found that nanosilica localized at the PLA/PBAT interface. This enhanced the material's thermal stability. Further observation showed that at higher nanosilica loading, the degradation processes of the composite could be slowed down, making these materials promising for packaging applications.

A multilayer film from PLA/PBAT blends with sodium alginate and coatings of chitosan (CS), zinc oxide nanoparticles (ZnONPs), or silver nanoparticles (AgNPs) for food packaging was assessed for its barrier performance.[262] These coatings improved the water vapor and oxygen barrier properties and increased the mechanical strength of the films, especially the AgNPs. The film coated with AgNPs decreased the water vapor transmission rate (WVTR) by 40%, going from 5.77×10^{-14} g cm^{-2} Pa^{-1} s^{-1} for the uncoated film to 3.41×10^{-14} g cm^{-2} Pa^{-1} s^{-1}.

In addition, the film exhibited a growth in tensile strength of 8%, going from 264.79 to 286.22 MPa, and displayed strong antibacterial effects on *Escherichia coli* and *Staphylococcus aureus*. These films, coated with AgNPs, are ideal for prolonging the shelf life of food products.[262]

Pozza Junior *et al.*[263] conducted research on using PLA/PBAT/graphite as an electrochemical sensor for detecting the toxic pollutant 2,4,6-trichlorophenol (TCP). The sensors were prepared by electrospinning. It was demonstrated that these sensors had low detection limits (7.84×10^{-4} mol L^{-1} or 0.0155 mg L^{-1}) and great sensitivity for TCP. Thermal analysis showed that the addition of graphite decreased the crystallinity of the blend and increased the surface area available for analyte adsorption, improving the electrochemical response of the sensor. The sensor had a linear detection range from 1.00×10^{-7} mol L^{-1} to 2.00×10^{-6} mol L^{-1} with a correlation coefficient of 0.993, which makes it a good candidate for environmental monitoring.

Zhu *et al.*[266] developed a nanofibrous membrane made of electrospun PHB/PLA composite, incorporating organic photochromic dye (OPD) and silver nanowires (AgNWs), for potential applications in smart textiles, medical care, and counterfeit prevention. The inclusion of 0.05 wt% AgNWs significantly enhanced the antibacterial efficacy against *E. coli* and *S. aureus*, exceeding 98%. Breaking stress and breaking strain of the OA-PHB/PLA NFM were 1.61 ± 0.22 MPa and 42.53 ± 5.93%. The membrane had great thermal stability and maintained a filtration efficiency of 99.9% with a minimum pressure drop of 73.4 Pa.

Erick *et al.*[264] sought to enhance the strength, heat resistance, and biodegradability of PLA/PHBV blend by incorporating graphene nanoplatelets (GNP) and multi-wall carbon nanotubes (MWCNT). Adding 2% MWCNT and 5% GNP to the PLA/PHBV blend resulted in a 195% increase in ultimate tensile strength and a 200% increase in strain at break. There was a 30% increase in the elastic modulus of the nanocomposites. PHBV increased thermal degradation temperatures by 10–20 °C. Under composting conditions, the nanocomposites retained good biodegradability, with about 45% degradation in 150 days. Hybrid nanocomposites containing MWCNT and GNP can greatly improve the performance of PLA/PHBV blends while still maintaining biodegradability, making them ideal for sustainable packaging applications.[264]

Gu *et al.*[267] created m-TiO_2 by modifying TiO_2 with 3-glycidoxy-propyltrimethoxy-silane and incorporating it into PLLA/PBS blends through reactive blending. The m-TiO_2 played the role of interfacial compatibilizer using the epoxy group on its surface. This resulted in simultaneous improvement in the tensile yield strength, notched impact strength and elongation at break. The composite's photocatalytic degradation and antibacterial activity performance were enhanced compared with the neat PLLA/PBS films.

A study on Zinc Oxide Nanoparticle (ZnONP) in PBAT/TPS films with varying ZnONP concentrations (0.5%, 1%, 3%, 5%) and their effects on the films' properties revealed that higher ZnONP content (5%) significantly improved UV blocking and antimicrobial activity. However, water solubility was reduced, with a decrease to 8.49%. In acidic food simulants, ZnONP migration increased, causing reduced thermal stability, while in aqueous simulants, migration was lower.[268]

5. Biodegradation of polymer blends and composites

Biodegradable polymer blends and their composites are designed to degrade under natural environmental conditions, and in controlled environments such as industrial and home composting environments, reducing long-term plastic waste accumulation.[269] While some polymers degrade well in aerobic conditions like composting, they may not perform as effectively in anaerobic environments such as landfills or anaerobic digestion systems.[270] The biodegradability of these polymer blends depends on factors such as their composition, the presence of natural fillers, and external environmental conditions, molecular weight, crosslinking, water solubility, degree of substitution, and crystallinity.[271,272] For instance, polylactic acid (PLA) is a widely used biodegradable polymer, but its slow degradation rate in soil and marine environments necessitates blending with other biodegradable materials like thermoplastic starch (TPS) or polybutylene adipate-*co*-terephthalate (PBAT) to enhance its decomposition.[179,273,274] Studies have shown that the addition of natural fibers such as barley straw, rice straw, or nanocellulose further improves biodegradation by increasing water absorption and microbial colonization, which accelerates polymer breakdown.[275]

In composting environments, PLA/TPS and PBAT-based blends demonstrate significantly enhanced degradation, with some studies reporting over 40% weight loss within 90 days (Table 12). The biodegradability of polymer composites is also influenced by the nature of additives and processing conditions. Plasticizers like polyethylene glycol (PEG) and compatibilizers such as maleic anhydride (MA) can modify the polymer matrix, improving flexibility and water uptake, thereby facilitating microbial degradation. Additionally, factors such as temperature, humidity, and microbial diversity play a crucial role in determining the rate of biodegradation. Research findings indicate that biodegradation at higher temperatures leads to faster polymer breakdown compared with soil burial, where degradation is often limited by environmental fluctuations.

Table 12 Biodegradation studies of polymer blends and composites

Blend composition	Filler	Additives	Biodegradation environment	Biodegradation conditions	Standard used	Findings	Ref.
PBAT/PLA (20, 40, 60, 80 wt% PBAT)	—	—	Enzymatic degradation with *Humicola insolens* cutinase	70 °C for 7 days	—	PBAT-rich blends degraded up to 40% weight loss, while PLA-rich blends showed lower degradation	182
PLA/Tapioca (TS) 65.7/27.9 wt%	—	PLA-*g*-MA, epoxidized palm oil (EPO)	Soil burial	23 °C and 30 °C, for 60 days	ASTM D5988	PLA/TS blends exhibited a biodegradation rate of 2.84%. Degradation occurs in the amorphous domains of starch chains	276
PLA (80 wt%)/TPS(Cassava) (20 wt%)	—	Glycidyl methacrylate (GMA 1 wt%)/benzoyl peroxide (BPO 0.1 wt%)	Soil burial	30 ± 2 °C, 90 days	—	PLA/TPS showed degradation rate, up to 40% weight loss in 90 days	277
PLA (95 wt%)/PBSA (5 wt%)		Joncryl (3 wt%)	Soil burial	30 ± 2 °C, 90 days	—	PLA/PBSA film exhibited 8.6% degradation in 90 days of soil burial	277
TPS/natural rubber (90/10 wt%)	—	PEG, modified natural rubber	Soil burial	120 days	—	More than 95% degradation in 120 days; suitable for short-term applications	211
PHB/PBAT (45/55 wt%)	—	—	Soil microbiome	27 °C, 180 days	—	PHB layer degraded faster than PBAT achieving a 47% mineralization in 180 days. PHB increased crystallinity indicated preferential biodegradation of amorphous regions	195
PLA/TPS (56/30 wt%)	—	PLA-*g*-MA (14 wt%)	Composting	58 and 37 °C, 180 days	ASTM D5338-15 (2021).	High mineralization observed at 58 ± 2 °C. At 37 ± 2 °C, improved degradation was observed compared with PLA, with mineralization more than 57%	278
TPS/PLA/PBAT (60/30/10 wt%)	Jute fibers (5, 10, 15 wt%)	—	Composting	37 ± 2 °C and 58 ± 2 °C	ISO14855-1: 2012	Jute fibers acted as a reinforcing agent but slowed down the biodegradation rate Biodegradation rate was faster at 58 ± 2 °C than at 37 ± 2 °C	279
PLA/PHB/cellulose paper (75/25 wt%) (sandwich structure)	Cellulose paper	—	Soil burial	8 months	—	16% weight loss under the action of soil microorganisms, water and heat was observed after 8 months of soil burial.	280
PLA/PHB (75/25 wt%)	—	—	Soil burial	8 months	—	PLA/PHB blends achieved a weight loss of 14% over 8 months	280
PLA/PBAT film	—	—	Fungal degradation (*Papiliotrema laurentii*) in mineral salt medium (MSM)	30 °C, for 30 days	—	Weight loss of 14% observed within 30 days The half-life of film reduced from nearly 3 years to 138 days	181
PBAT/TPS (40 to 60 wt% TPS)	—	Compatibilizer (MA, PBAT-*g*-MA)	Laboratory composting	58 °C at a 10 mL min^{-1} air, moisture content less than 50%	ISO 14855-2:2018	PBAT/TPS (40–60 wt%) without compatibilizer, the biodegradation rate was 82–87% after 90days. PBAT/TPS blends with PBAT-*g*-MA showed reduced biodegradation rates due to the presence of PBAT-*g*-MA with degradation rate decreasing to 72–74% after 90 days	281

Table 12 *(Contd.)*

Blend composition	Filler	Additives	Biodegradation environment	Biodegradation conditions	Standard used	Findings	Ref.
PLA/PHB (85/15 wt%)	Keratin (1–20 wt%)	Acetyl tributyl citrate (ATBC) (15 wt%)	Hydrolytic degradation	0.01 M NaOH solution (pH of 11.8), extra-pure water (MilliQ, pH of 7), 25 °C	—	PLA/PHB/Keratin composites showed improved degradation under hydrolytic conditions achieving 50% degradation in 20 days	282
PLA/PHB (85/15 wt%)	—	Acetyl tributyl citrate (ATBC) (15 wt%)	Hydrolytic degradation	0.01 M NaOH solution (pH of 11.8), extra-pure water (MilliQ, pH of 7), 25 °C		About 50% hydrolysis occurred after 50 days Polymer crystals of PHB were more hydrolytically stable and caused slower degradation	282

6. Applications of biodegradable composites

The application of polymer composites depends largely on matrix–reinforcement relationships, the type of polymer matrix, and the type of reinforcement. Nanofillers, synthetic fillers, natural fiber, proteins, starch granules, and biological macromolecules have opened up different applications for reinforced polymer composites. Most recently, and importantly, biodegradable reinforced polymer composites have been utilized across diverse industries due to their versatility (Fig. 12). These composites are gaining increasing attention due to their potential applications in various industries, resulting from their unique properties such as low weight, high strength, environmental friendliness, and biodegradability.

6.1 Biomedical applications

In medicine, biodegradable reinforced composites are used for implantable devices, such as drug delivery systems and scaffolds for tissue engineering, bones, and dental resin-based composites.[130] These materials are attractive due to their biodegradability, which eliminates the need for a second surgery to remove the implant. Their high strength and low weight make them suitable for orthopedic braces and prosthetics. It has been demonstrated that bone regeneration is feasible with a biodegradable piezoelectric poly(L-lactic acid) nanofiber

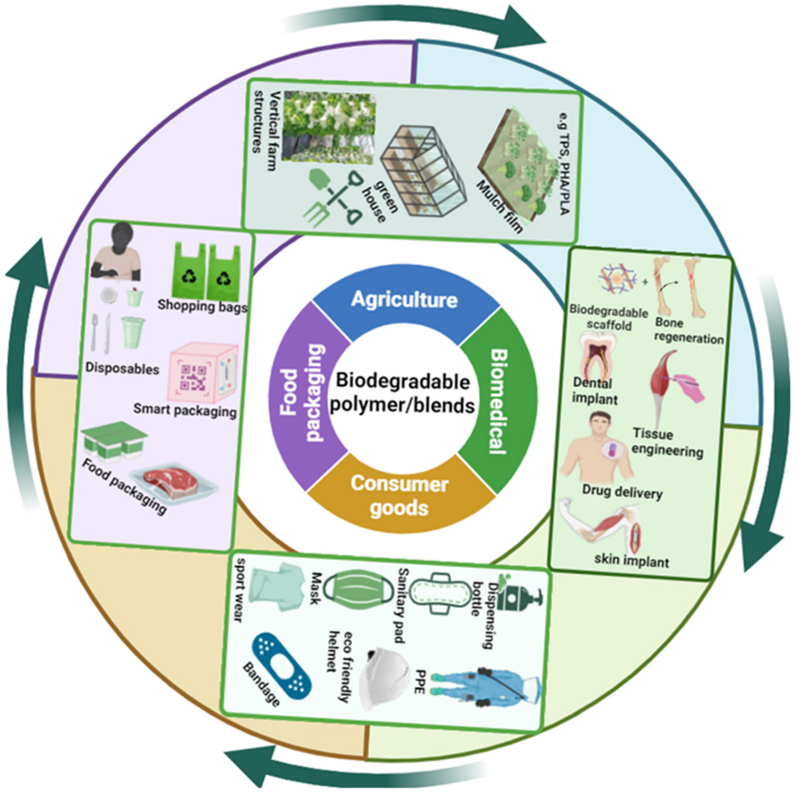


Fig. 12 Applications of biodegradable polymers and their blends.

scaffold with ultrasound that can be externally controlled. This is a hybrid of electrical stimulation (ES) and tissue-engineering approaches (biomaterial scaffolds).[283] The triblock copolymer of poly(L-lactic acid)-*block*-aniline pentamer block-poly(L-lactic acid) (PLA-AP) with poly(lactic-*co*-glycolic acid)/hydroxyapatite (PLGA/HA) scaffold exhibits an improved cell proliferation and better *in vitro* osteogenesis differentiation for effective bone healing in rabbits.[284] In the pursuit of advancing potential biomedical applications, Borah *et al.*[285] developed glycine *N*-hydroxysuccinimide (NHS) ester-modified polyaniline (PANI)/chitosan nanocomposites for tissue engineering. The nanocomposites showed better fibroblast morphology, proliferation, adhesion, and spreading.

Drug delivery is another area of medicine where biodegradable polymers and their composites are relevant owing to their biocompatibility and degradability. PLA's biodegradability and biocompatibility make it an ideal vehicle for parenteral-controlled drug delivery systems because its microparticles can control the rates of drug delivery that could last for a few days to several weeks and up to a year.[286] Several studies have reported the potential efficiency of PLA in active drug delivery, including PEG-PLA loaded with Arenobufagin nanoparticles for enhanced cancer therapy and reduction,[287] and PEG-*b*-PLA micelles and PLGA-*b*-PEG-*b*-PLGA sol-gels for drug delivery.[288] PLA could present a water-tight coating membrane to significantly reduce the hygroscopicity of mildronate by more than two times without negatively impacting the physical state of the drug, representing a milestone in the cardioprotective drug hygroscopicity, thereby preserving its bioavailability.[286,289]

6.2 Food packaging

In the packaging industry, reinforced biodegradable matrix composites have been used because they can provide an eco-friendly and sustainable alternative to traditional food packaging materials. These packaging materials are synthesized into film comprising fillers such as starch, cellulose, lignin, and natural fibers, nanoparticles, which micro-organisms in the environment can break down. The reinforced biodegradable matrices are designed to provide superior strength and barrier properties for food packaging, allowing for extended shelf life and improved product safety. Additionally, these matrices are often designed to be compostable, meaning they can be broken down into natural environmental components. These materials have the potential to reduce the amount of plastic waste generated by conventional packaging materials like polyethylene and help to reduce the impact of food packaging on the environment.

PLA is one of the most researched biodegradable polymers for composites. Composite films incorporated with different active fillers have provided functional properties including barrier and antimicrobial functions, and enhanced degradability. Chen *et al.*[290] reported the oxygen permeability of laminated multilayer CNC/PLA film to be 70 times lower than pure PLA film, and the water vapour permeability decreased 7-fold. The CNC was made into a suspension with 15 wt% polyvinyl alcohol (PVA) and then coated on a PLA substrate, followed by lamination. Their idea was that a multilayer composite film could prove successful in improving the barrier of PLA to moisture and oxygen. Also, incorporating cholecalciferol (Vitamin D3) (CC) into PLA solvent-casted composite film could provide an effective barrier to UV-B light at 320 nm, antioxidant activity, and an improved mechanical and oxygen barrier with antibacterial activity against food-borne bacteria (*S. aureus* and *E. coli*).[291] The functionality of PLA composite film in active packaging has been further improved to include sensing components. Curcumin incorporated into a PLA/poly (propylene carbonate) (PPC) blend has the potential as a smart indicator (sensors that monitor the condition of the food inside the package to provide information to consumers) for food packaging and industrial ammonia (NH_3) gas monitoring applications.[292] Other fillers in biodegradable polymers have been reported as potential barrier materials in food packaging. These include calcium carbonate, lignin/tannic acid, in PBAT, Pickering emulsions (PE) of essential oils[293,294] stabilized by nanocellulose in thermoplastic starch,[295] and talc in polybutylene succinate-*co*-adipate (bio-PBSA).[89]

6.3 Agriculture

In agriculture and fisheries, composite materials are made into greenhouses for vegetables, flowers, granaries, feed stores, septic tanks, drains, spray, flower pots, milk delivery vehicles, and manure transport vehicles.[296] The transition to modern systems of agriculture necessitates the introduction of innovative technologies and environmentally friendly measures to counter the adverse effects of traditional farming. Biodegradable composites have found their use in controlled environment agriculture (CEA). This is the production of plants in an enclosed space like green houses or vertical farms, whereby several environmental variables like temperature, humidity and light among others are carefully controlled.

PLA was found effective as a structural component in macroalgal cultivation (settlement substrates) and cricket rearing (housing) for its resistance, rigidity, and direct material–organism interactions.[295]

It tolerated corrosive cultivation conditions and provided a suitable substrate with no adverse effect on the macroalgal physiology or nutritional composition, and served as a recyclable shelter for crickets. PHB-reinforced barley fiber composites showed promising potential as a permeable biodegradable composite in agriculture.[297] The added fiber increased water uptake capacity to allow water permeation and facilitated biodegradation.

According to Maraveas *et al.*,[298] polysaccharide derivatives (DS), PHB, PCL, PHA, and PLA were shown to be highly biodegradable materials for fabricating anti-insect, anti-hail, and windbreak plastic nets in agriculture. The introduction of biodegradable composites into such systems ensures the optimisation of sustainable production of commodities with a reduction in environmental impact. Such substrates degrade into non-harmful by-products, making controlled environment agriculture more friendly for the environment. This bifunctionality supports the overall purpose of developing sustainable, efficient agricultural systems.

Besides structural applications, biodegradable composites can be used in the development of biodegradable mulches, used for soil management in controlled environment agriculture. Soil moisture, temperature, and weed growth can be effectively controlled using mulching. For instance, TPS, PHA/PLA, and bio-based polybutylene succinate (BioPBS) used as mulching and fruit (tomatoes and peaches) protection bags improved soil quality and reduced blossom end rot.[299] The produced peaches had uniform colour (without red blush), a required characteristic for peaches. Their biodegradability in the soil was about 6 months, showing their viability. A problem with traditional plastic mulches is disposal, hence, biodegradable mulches from composite materials provide an effective alternative to traditional non-degradable mulches. During the growing season, these mulches offer the needed agronomic benefits and decompose into residue that enriches the soil humus.

7. Environmental impact assessment

End-of-life considerations and ecotoxicity assessments are crucial aspects in the evaluation of biodegradable polymer blends and composites. Ecotoxicity refers to how chemical, physical, or biological stressors impact ecosystems and organisms including fish, insects, microorganisms, wildlife, and plants.[300] As society moves towards more sustainable materials, study of the lifecycle to understand their impact on the environment is essential. It is well understood that biodegradable polymer blends and composites can significantly reduce the burden of traditional plastics on the environment. A thorough assessment of their end-of-life behaviour and potential ecotoxicity is essential. At the end of their useful life, biodegradable polymer blends and composites are designed to undergo industrial or home degradation processes, breaking down into simpler compounds under environmental conditions, such as sunlight, microbial activity, and moisture. The degradation process, rate, and extent are largely dependent on the presence of additives, polymer structure, chemical, molecular weight, and polymer composition. Hence, for precise prediction of the degradation time in different environments, an understanding of the degradation kinetics is essential.

The big question is whether the biodegradation of biodegradable polymer blends and composites raises concerns regarding the degradation of by-products and their potential environmental impact. While biodegradation offers the advantage of reducing the accumulation of plastic waste in landfills and oceans, concerns have been raised about the potential of releasing harmful substances. Under controlled industrial or laboratory conditions, assessing biodegradation based on standard procedures is simpler than in real-life contexts including soils and oceans, which have varying microbial populations.[301] According to Ali *et al.*,[302] because biodegradation involves a mixture of identified and unidentified substances that may be hazardous to animals and soil health, it can both pose challenges and offer advantages in complex ecosystems. Unknown substances with roughly 32% cytotoxicity, 23% anti-androgenicity, 42% oxidative stress, and 67% baseline toxicity in bioassays have been shown to be present in bioplastics according to some studies. These compounds have the potential to negatively impact soil and animal health.

Ecotoxicity assessments play a crucial role in evaluating the environmental implications of biodegradable polymer blends and composites. These assessments involve evaluating the potential toxicity resulting from the degradation of various organisms in aquatic and terrestrial environments. Such testing includes a focus on endpoints such as reproductive effects, acute and chronic toxicity, and bioaccumulation potential. To be deemed environmentally benign, these polymers, their blends, and composites must pass toxicity testing and break down into fragments within a given amount of time. However, one of the major setbacks in assessing the ecotoxicity of biodegradable polymers is the lack of standardized testing procedures and regulatory frameworks, unlike the biodegradation and compostability of plastics that have standardized and established testing procedures with clearly outlined regulatory requirements.[303] Unlike biodegradation and controlled composting, there seems to be no specific standard for assessing the ecotoxicity of these materials in different environments.

In fact, two ASTM standards, *i.e.*, ASTM D5152-91; practice for water extraction of residual solids from degraded plastics for toxicity testing[304] and ASTM D5951-96(2002); standard practice for preparing residual solids obtained after biodegradability standard methods for plastics in solid waste for toxicity and compost quality testing[305] were withdrawn in 1998 and 2011, respectively, with no replacements. Hence the assessment of the ecotoxicity of biodegradable materials presents unique complexities as a result of their various compositions and degradation pathways. The multifaceted complexities in the assessment of the ecotoxicity of polymer blends and composites require ecotoxicity testing procedures suited for biodegradable polymers for understanding the possible environmental problems that may be associated with them. A study by Palsikowski *et al.*[306] on ecotoxicological assessment of PLA, PBAT, and their compatibilized blend in soil found that the biodegradable materials and their blends did not exhibit phytotoxic (adverse effects on plant growth, physiology, or metabolism), cytotoxic (quality of being toxic to cells), genotoxic (damaging to DNA or genetic material), or mutagenic (causing genetic mutations) effects on the meristematic cells of *Allium cepa*, except for a chromosomal aberration index observed in one experiment with the blend 25/75. Sforzini *et al.*[307] used biotests to evaluate the ecotoxicity impact of Mater-Bi, a biodegradable plastic, on soil organisms. The ecotoxicity tests on Mater-Bi biodegradable plastic showed no harmful effects on soil organisms, including autotrophic organisms, *Daphnia magna*, and *Eisenia andrei*. In their discussion of the environmental fate and ecotoxicity assessment of biodegradable polymers, Farachi *et al.*[308] emphasized the importance of assessing the possible ecological dangers that may be connected to biodegradable materials. Carteny *et al.*[309] hypothesized that in marine environments, biodegradable microplastics may accumulate more contaminants than conventional microplastics, despite emitting fewer additives. Life cycle analyses (LCAs) are the most complete tools to evaluate the entire lifecycle of recyclable polymers, blends and their composites, starting from the raw material stage to the final disposal stage or end of life. Through LCAs, factors such as greenhouse gas emissions, energy consumption, and ecotoxicity potential are evaluated. These analyses help identify challenges and opportunities to enhance the sustainability of biodegradable materials.

8. Conclusion

8.1 Summary of key findings

In response to the growing environmental awareness and shift towards developing biodegradable materials from renewable sources, research has increasingly focused on addressing the limitations of conventional, non-biodegradable plastics. An important strategy in enhancing the properties and performance of biodegradable polymers is the use of polymer blends and reinforcements, which improve compatibility and miscibility through plasticization, as well as reactive and non-reactive compatibilization. These techniques enhance mechanical strength, flexibility, and overall functionality by reducing interfacial tension in polymer blends, making biodegradable polymer blends more viable alternatives to conventional plastics. For example, PLA/PHAs blends, such as PLA/PHB and PLA/PHBV, exhibit enhanced toughness, flexibility, and thermal stability, making them suitable for packaging, biomedical implants, and 3D printing. PLA/PBAT blends, known for their high flexibility and impact strength, are widely used in food packaging and compostable bags. PBAT/PHA blends, such as PBAT/PHBV, provide improved ductility and biodegradability, making them ideal for sustainable packaging and controlled degradation applications. The incorporation of fillers further enhances these blends, with natural fillers such as coffee grounds, rice straw, and lignocellulosic fibers improving biodegradability and mechanical reinforcement for sustainable composites. Inorganic and mineral fillers, such as nanoclays and calcium carbonate, significantly enhance mechanical strength, barrier properties, and thermal stability, making them suitable for high-performance packaging and structural applications, while nanoparticle reinforcements, such as cellulose nanocrystals (CNCs), graphene, and hydroxyapatite nanoparticles, contribute to improved mechanical, thermal, and antimicrobial properties, particularly in biomedical implants, tissue engineering, and antimicrobial packaging.

9. Future directions

9.1 Integration with circular economy concepts

An innovative method for sustainable manufacturing and waste management is offered by integrating biodegradable polymers and composites into the circular economy (Fig. 13). This paradigm shift is essential for addressing the dual issues of resource depletion and environmental degradation, which are caused by the traditional linear economic model of "take-make-dispose" (Fig. 14). Biodegradable polymers and composites are a great ally of the circular economy, which stresses the ideas of reduce, reuse, repurpose, and recycle.[310,311] The circular economy may incorporate biodegradable polymers and composites in many ways, such as waste management, product design and production, and policy formulation. Every stage offers opportunities and challenges to establish a more sustainable cycle of material use and recycling.[8]

The design phase is critical for establishing the lifecycle impact of a product. Designing for biodegradability requires not just choosing appropriate materials, but also considering product usability, durability, and disposal at the end of life.[312] Products can be built to decompose under certain conditions, such as in industrial composting facilities, diverting them away from landfills and into usable compost.[313] Furthermore, innovation in the combination of biodegradable polymers with natural fibers often results in unique composites with superior properties, expanding their application range and replacing more traditional, non-biodegradable materials in some sectors. The circular economy principle is embodied also in the production process. Efficient, waste-minimizing, and energy-saving manufacturing processes are fundamental steps toward achieving circularity.[314,315] Modern manufacturing procedures like additive manufacturing (3D printing) make it possible to design and optimize customized composite systems with no material wastage or energy-demanding machining methods compared with traditional manufacturing. It can reuse recycled materials like plastics, turning waste into new products and supporting a circular economy. This process is energy-efficient, especially for small-batch or complex designs, and reduces the need for global shipping by enabling local, on-demand production. Thus, closed-loop recycling systems including production scrap and end-of-life product recovery, and closed-loop water systems during manufacturing can help to lessen the environmental footprint of these materials.[316–318]

During utilization, biodegradable polymer composites display an advanced functionality that exhibits variability among sectors like packaging, construction, transport system, and consumer goods.

Applying the circular economy principle, the manufacturers make all types of composite products such as formats and transportation systems durable, repairable and refurbishable, which increases their life cycle. Similarly, the pay-per-use policy such as product-as-a-service models or leasing arrangements which incentivize reuse and resource conservation, encourage the growth of the circular economy mindset in consumers and businesses.[8,319] In the circular economy, waste management is essential, and biodegradable polymers, blends, and their composites offer significant advantages.

The two most important methods for handling these materials at the end of their lifespan and turning trash into useful resources like compost and biogas are anaerobic digestion and composting.

The establishment of infrastructure, such as commercial composting facilities, along with customer involvement in

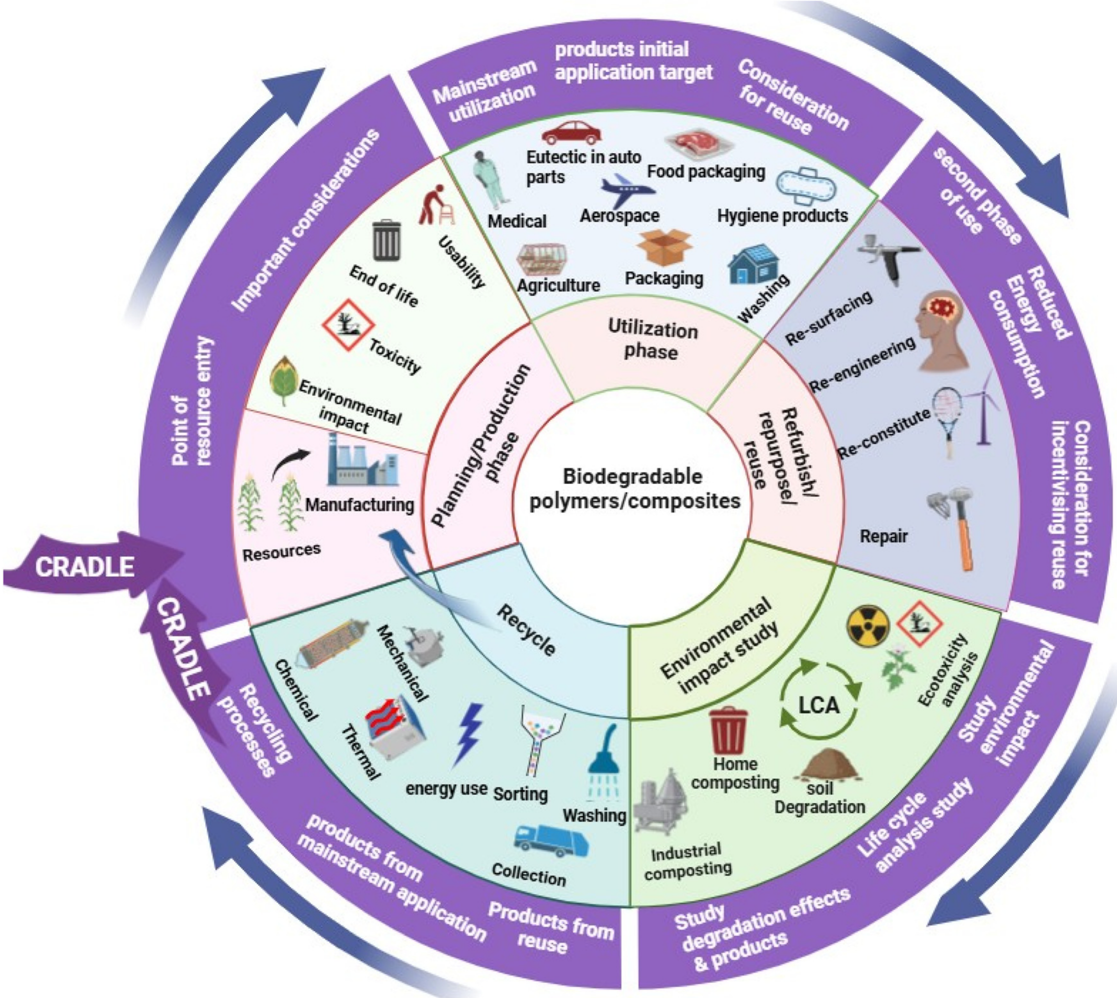


Fig. 13 Circular economy concept: cradle to cradle for biodegradable polymers, blends, and composites. Figure was made with Biorender.

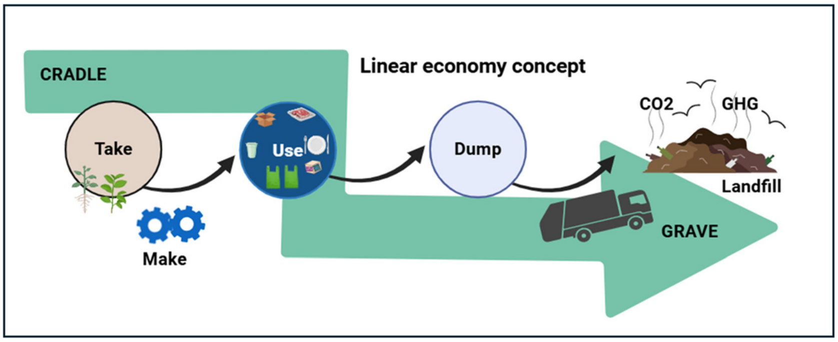


Fig. 14 Linear economy concept: from cradle to grave. Figure was made with Biorender.

appropriate waste sorting, are prerequisites for the success of these initiatives. Moreover, new chemical recycling techniques are being developed that may decompose polymers into their monomers, providing a means of repurposing biodegradable waste to make new materials and thereby closing the loop.

Many studies have focused on the significance of product design, effective utilization, waste management, and reuse. As a step toward a circular economy, sustainable woody-like composites derived from recycled Tetra Pak cellulose and poly (butylene succinate) were fabricated.[320] McKeown *et al.*[321] concentrated on PLA chemical recycling, highlighting how end-of-life disposal strategies of biobased polymers are vital for a successful transition to a sustainable circular economy. In the framework of a circular economy, the utilization of waste coffee trash as a renewable source for creating sustainable PBS biocomposites was highlighted.[322] The creation of sustainable polymer materials in a way that improves sustainability through material design, waste management, renewable energy sources, biodegradability, biotechnological approaches, and enzymatic recycling within the framework of an international circular (bio)economy can foster efficient resource utilization and management.[323]

9.2 Biodegradability and recyclability in the circular economy

While biodegradable polymers offer clear environmental benefits especially in mitigating plastic pollution,[8,324] the shift toward a circular economy requires a broader, system-level perspective. In this framework, recyclability often takes precedence over biodegradability. This is because materials that can be efficiently reused or reprocessed preserve their resource value and reduce waste generation across multiple life cycles. Blending biodegradable polymers is a common strategy to enhance mechanical, thermal, or barrier properties. However, this approach introduces significant challenges for recycling. Most polymer blends consist of immiscible or only partially miscible phases, leading to material heterogeneity that complicates mechanical or chemical recycling.[325] These incompatibilities can result in phase separation, property degradation, and overall reduced recyclability, even when compatibilizers are used. As Ragaert *et al.*[326] and Titone *et al.*[325] clearly emphasized, mechanical recycling remains a key pillar of circular material flow, and mono-material streams are generally more favorable for maintaining quality in closed-loop systems.

Biodegradability, while valuable in open environmental contexts where plastic leakage is inevitable, may not offer the same benefits in structured waste management systems lacking industrial composting infrastructure. In fact, in the hierarchy of circularity, recyclable materials are often prioritized because they better support long-term material retention and reuse.[327,328] To align polymer blend development with circular economy goals, future strategies must go beyond traditional property enhancement. This includes designing blends with recyclability in mind through reversible chemistries, dynamic covalent networks, or advanced compatibilization techniques that allow effective reprocessing without compromising performance. Additionally, the concept of '*design-for-recycling*' should be embedded from the outset, ensuring that material formulations account for their entire lifecycle, not just their immediate utility or degradability.[328]

Ultimately, achieving true circularity in polymer systems will require striking a careful balance between biodegradability, performance, and recyclability. Rather than viewing these qualities as mutually exclusive, the next generation of materials must be engineered to embody all three to support both environmental sustainability and practical circular economy implementation.

9.3 Potential breakthroughs

The horizon of biodegradable polymer blends and filler-reinforced composites is promising, and some breakthroughs are waiting to happen that could change the field of sustainable materials. With the ongoing efforts by industry and researchers to promote green practices and the emergence of alternatives to petroleum-based plastics, the development and incorporation of biodegradable polymers are expected to be key in making the world greener.

One significant area of future breakthroughs in the development of biodegradable polymer blends is the progress in material design and processing techniques. New developments may lead to biodegradable polymer blends with superior qualities, such as barrier, mechanical and thermal properties, through further refinement of the blend composition, structure, and processing conditions. This would make them more appropriate for a wider range of applications than traditional plastics.

Particularly for biomedical applications, such as implantable devices, drug delivery systems, tissue engineering scaffolds, orthopedic braces and prosthetics, biodegradable reinforced composites are most appropriate. Future progress in the field of biomaterials may be marked by the discovery of new materials with improved biodegradability, biocompatibility, and efficacy for regenerative medicine and personalized healthcare.

The future circular economy integration will be driven by the development of closed-loop systems for biodegradable materials, innovative recycling technologies, and eco-friendly product design approaches that emphasize sustainability at every stage of the product lifecycle.

Author contributions

Kehinde Olonisakin: investigation, methodology, data analysis, visualization, writing—original draft preparation. Amar Mohanty; project conceptualization, investigation, validation, funding acquisition and supervision, writing – Review and editing. Mahendra Thimmanagari: investigation, validation, writing – review and editing. Manjusri Misra: project conceptualization, investigation, methodology, validation, administration, resources, funding acquisition and supervision, writing – review and editing. All authors contributed to the discussion, reviews, and approval of the manuscript for publication.

Conflicts of interest

Authors have no competing-conflicting interests to declare.

Abbreviations

ASTM	American Society for Testing and Materials
CNCs	Cellulose nanocrystals
CNPs	Cellulose nanoparticles
DMA	Dynamic mechanical analysis
DSC	Differential scanning calorimetry
ESO	Epoxidized soybean oil
FESEM	Field emission scanning electron microscopy
FTIR	Fourier-transform infrared spectroscopy

GPC	Gel permeation chromatography
GR	Gum rosin
HA	Hydroxyapatite
Hec-g@OA	Hectorite modified with octadecylamine
LCA	Life cycle analysis
MA	Maleic anhydride
MDI	4,4′-Diphenylmethane diisocyanate
NC	Nanoclay
NHS	*N*-Hydroxysuccinimide
NMR	Nuclear magnetic resonance
OLA	Oligomeric lactic acid
O-MA-TS	Dual modified thermoplastic starch
PANI	Polyaniline
PBAT	Polybutylene adipate terephthalate
PBS	Polybutylene succinate
PCL	Polycaprolactone
PE	Polyethylene
PET	Polyethylene terephthalate
PGA	Poly(glycoic acid)
PGMA	Poly(glycidyl methacrylate)
PGV	Poly(vinyl chloride)
PHAs	Polyhydroxyalkanoates
PHB	Poly(hydroxy butyrate)
PHBHHx	Poly(3-hydroxybutyrate-*co*-3-hydroxyhexanoate)
PHBV	Poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate)
PHO	Poly(3-hydroxyoctanoate)
PLA	Polylactic acid
PLA-AP	Poly(L-lactic acid)-*block*-aniline pentamer
PLGA	Poly(lactic-*co*-glycolic acid)
PP	Polypropylene
PPC	Polypropylene carbonate
REX	Reactive extrusion
SEM	Scanning electron microscopy
SI-ATRP	Surface-initiated atom transfer radical polymerization
TAM	Triallyltrimesate
TEM	Transmission electron microscopy
TGA	Thermogravimetric analysis
TPS	Thermoplastic starch
UV-Vis	Ultraviolet–visible spectroscopy
WVTR	Water vapor transmission rate
XRD	X-ray diffraction

Data availability

No primary research results, software or code have been included, and no new data were generated or analysed as part of this review.

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