



# Thermoplastic Starch (TPS)/Polylactic Acid (PLA) Blending Methodologies: A Review

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## Abstract

Poly(lactic acid) (PLA) and thermoplastic starch (TPS) are biodegradable polymers of biological origin, and the mixture of these polymers has been studied due to the desirable mechanical properties of PLA and the low processing cost of TPS. However, the TPS/PLA combination is thermodynamically immiscible due to the poor interfacial interaction between the hydrophilic starch granules and the hydrophobic PLA. To overcome these limitations, researchers studied the modification, processing, and properties of the mixtures as a strategy to increase the compatibility between phases. This review highlights recent developments, current results, and trends in the field of TPS/PLA-based compounds during the last two decades, with the main focus of improving the adhesion between the two components. The TPS/PLA blends were classified as plasticized, compatible, reinforced and with nanocomposites. This article presents, based on published research, TPS/PLA combinations, considering different methods with significant improvements in mechanical properties, with promising developments for applications in food packaging and biomedicine.

**Keywords** Thermoplastic starch · Polylactic acid · Biodegradable polymers · *Polymer blends*

## Introduction

The increase in solid waste from nonbiodegradable polymers and the depletion of petroleum resources have led to environmental awareness and the interest of plastic industries, universities and researchers in the search for raw materials from alternative sources, which has driven the study and

development of biodegradable plastics through the use of renewable resources [1] that have properties comparable to those of current polymeric materials at an equivalent cost [2]. However, biodegradable polymers have some drawbacks, such as low degradation rate, relatively high cost, and high brittleness compared to conventional plastics [3]. In recent years, research has established the potential use of agricultural products for nonfood applications, including polysaccharides, such as cellulose, glycogen, chitin and starch [4]. Among these materials, starch is considered one of the most promising renewable resources, as it is an alternative for synthetic materials due to its availability, easy production, environmental friendliness, profitability and ease of handling [5, 6]. In this context, starches can be obtained from different cereals and tubers, such as cassava [7], potato [8], corn [9], green peas [10], oatmeal [11], rice [12], and barley [13]. In more recent studies, starch can be obtained from species belonging to aquatic ecosystems such as mossy ferns, protozoa, algae and bacteria [14].

Starch is composed of two different types of amylose and amylopectin structures. Amylose is a linear polymer of glucose units with  $\alpha$ - (1  $\rightarrow$  4) and amylopectin bonds, which is highly branched with  $\alpha$ - (1  $\rightarrow$  4) glycosidic bonds in the main chain and  $\alpha$ - (1  $\rightarrow$  6) in branching points [15, 16].

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Amylose and amylopectin constitute 98–99% of the weight of starch [17], and their structure makes these two components differ significantly from each other in both physical and chemical properties; that is, depending on the botanical origin of the native starches, different morphologies, particle sizes and crystallinity structures A, B and C can be distinguished [18, 19]. Despite its wide range of potential, starch is not commonly used for industrial applications due to its hydrophilicity, present because of hydroxyl groups that form intermolecular and intramolecular hydrogen bonds [20], causing starch to erode in the presence of moisture and to have poor mechanical properties. Therefore, it cannot be used directly as a plastic material. To overcome these drawbacks, native starch can be converted into a thermoplastic material through the incorporation of a plasticizing agent such as glycerol, sorbitol and xylitol [21]. The addition of plasticizers reduces intermolecular forces and increases the mobility of polymer chains, decreases the glass transition temperature and increases permeability [22]. The effect of the plasticizer depends on several factors; the most important are the structure of the starch, influenced by the origin of the native starch and processing conditions, including time, temperature, mechanical shear, type and amount of plasticizer [23].

Products manufactured with TPS exhibit good oxygen barrier properties, biodegradability and compostability. However, TPS is sensitive to water, which causes the properties to change according to the relative humidity of the environment and has low mechanical strength compared to commercial plastics [24]. It has been discovered that polymer blends are an effective and simple way to obtain materials with improved properties. Among the strategies studied is the blending of TPS with biodegradable polymers, such as poly(lactic acid) [25].

Poly(lactic acid) (PLA) is a thermoplastic, compostable, biocompatible and nontoxic polymer derived from renewable resources such as corn, sugar beet and potato starch [26]. It offers desirable mechanical and processing properties. PLA is a linear thermoplastic polyester with stiffness, tensile strength and gas permeability comparable to synthetic polymers [27], is considered one of the most promising materials to replace petroleum-based polymers in the packaging industry; however, its production is expensive compared to conventional plastics. Recently, the interactions of components, structure and properties of blends prepared from poly(lactic acid) and thermoplastic starch have been studied, and this combination allows the physical, mechanical and biodegradability properties of the manufactured products to be adapted. This is done to reduce costs and obtain a good-performance material.

This work reviews the state of the art of current studies for the development of biodegradable polymers and examines the properties of thermoplastic starch (TPS)/poly(lactic

acid (PLA) blends. Specifically, a review of the processing, compatibility and reinforcement methods and the influence on the morphology and the mechanical, thermal and biodegradability properties of the compounds is proposed, which were studied and discussed in detail.

## Biologically Based Polymers

### Thermoplastic Starch (TPS)

Thermoplastic starch (TPS) is a homogeneous substance prepared with native starch, water and/or other plasticizers, such as glycerol, sorbitol, and glucose [28]. Other compounds, such as those containing nitrogen (urea, ammonium derivatives, amines), can also be used [29]. The TPS conversion process can be performed using the same processing technologies as conventional plastics, such as compression molding [30, 31], extrusion [32], or injection molding [23, 33–35]. The plasticizer penetrates the starch granules and interrupts the initial crystallographic structure due to temperature and shear forces. The material undergoes a thermomechanical transformation of the semicrystalline starch granules, which are destructured into an amorphous polymeric material that does not exhibit diffraction by rupture of intra- and intermolecular hydrogen bonds (gelatinization) and subsequent rearrangement of the molecular structure (retrogradation or recrystallization). Once the starch is gelatinized and adequately plasticized, the TPS can flow like any synthetic polymer; therefore, TPS has lower degrees of crystallinity than natural starch [36].

The starch cannot be transformed into a thermoplastic material without plasticizers, the application of heat and shear force [37]. Figure 1 shows the plasticization process of the starch. The process parameters, the proportion of plasticizer and the shear forces strongly influence the physical properties of the processed starch [38, 39], such as its tensile strength, fracture strain, elastic modulus, glass transition, and gas barrier [40]. Starch-based thermoplastic materials have potential use for short-life plastic applications, especially for food packaging [38, 41] due to their oxygen barrier properties, biodegradability, compostability and renewability [42]. In addition, compared to synthetic thermoplastics, it is a fairly cheap material. Unfortunately, its applications are limited due to its poor mechanical properties, high sensitivity to moisture and low thermal stability [43] because of its hydrophilicity. To address these deficiencies, several strategies have been used, including choosing starch sources with a higher amylose content, using more efficient plasticizers, chemically modifying the starch, melting TPS with other conventional or biodegradable polymers, reinforcing TPS with fibers and developing TPS/nanofiller compounds

[44], which have become an efficient approach to improve their mechanical performance and preserving the ecological soundness of the final material.

## Poly(lactic Acid) (PLA)

Poly(lactic acid) (PLA) is currently considered the most promising biodegradable polymer because its production is derived from renewable resources, so it has captured the attention of researchers and industry as an alternative to conventional synthetic polymers due to its biodegradability and compostability characteristics. In addition, it has good properties, such as a high melting point, high mechanical strength, high tensile modulus and a high degree of transparency [45], quite comparable with commercially available polymers such as polypropylene (PP), poly(ethylene terephthalate) (PET) and polystyrene (PS). Among its applications are transparent and opaque rigid plastics for packaging, disposable products, plasticulture, textiles, 3D printing, films and fibers [46, 47], making PLA a high-performance polymer attractive from environmental and economic points of view. Other applications of PLA are found in biomedicine due to its biocompatibility and ability to be reabsorbed in contact with living tissues, such as implants, sutures, and for encapsulation of medications [48]. Currently, it is one of the most widely used biopolymers for bone tissue engineering [49, 50] for being recognized as safe (GRAS) and approved by the Food and Drug Administration of the United States (FDA).

Figure 2 illustrates the chemical structure of the poly(lactic acid) polymer, which is a hydrophobic aliphatic thermoplastic polyester that can be semicrystalline or completely amorphous, depending on the stereopurity of the polymer skeleton [51]. PLA is derived from lactic acid produced through the fermentation of dextrose, normally obtained from corn for providing high-purity lactic acid. Another source used as raw material is sugar cane. However, it could be synthesized from other renewable resources, such as starch and sugars. The industrial production of lactic acid is carried out through the fermentation of carbohydrates by bacteria of the genus *Lactobacillus* [26], and there are two optical forms of PLA: L - (+) - lactic acid and D - (-) - lactic acid [52–54], which are generally produced by the polymerization of lactic acid or by ring-opening polymerization (ROP) of lactides [55]. Figure 3 shows the different chemical structures of these two isomers [56].

Unfortunately, the physical properties of PLA have drawbacks, such as brittleness, high permeability rate and water sensitivity; additionally, it has a higher production cost than conventional petroleum-derived plastics, rendering the PLA polymer is unsuitable for a wide range of practical applications [57, 58].

## Blending Processes

In recent years, the interaction of polymer blends has been studied to improve their properties and decrease their production cost. Studies have shown that the mixture of natural polymers and synthetic biodegradable polymers is an alternative for the industrial production of short-life plastics and the reduction of the environmental footprint. Despite its advantages, the combination of these two types of polymers presents some drawbacks in their interaction characteristics, since most natural polymers are hydrophilic materials. On the other hand, most synthetic biodegradable polymers are hydrophobic [59]. As mentioned above, the study of biopolymer blends is enormous; some researchers have studied the combination of TPS thermoplastic starch with biodegradable polymers, such as PLA [60, 61], PCL [2, 62], poly(hydroxybutyrate) PHB [63], poly(butylene adipate-co-terephthalate) PBAT [64] and poly(butylene succinate) PBS [65]. These types of mixtures are an interesting approach to produce low-cost biodegradable materials.

## Plasticized TPS/PLA Blends

The production of biodegradable starch-based materials in combination with biobased and biodegradable biopolymers has been the subject of considerable research effort for food packaging applications and could also be considered for biomedical applications.

Martin and Averous [27] were the first to report on the thermal, morphological and mechanical properties of TPS/PLA blends, observing a dependence of the glass transition temperature ( $T_g$ ) of PLA on the composition of the mixture. The  $T_g$  gradually decreased as the TPS content in the mixture increased, which indicates a reduced compatibility between the components of the mixture. The authors concluded that the lack of interaction between the TPS and the PLA due to phase separation constituted a serious limitation and emphasized the need for some compatibility strategy.

Park et al. [66] investigated the effects of adding glycerol on the characteristics of starch and its mixtures with PLA, observing the appearance of voids formed by the separation of starch particles from the matrix in the PLA and pure starch combination. However, for the PLA/gelatinized starch blends, these voids were not observed, indicating that the gelatinization of the starch improved the interfacial adhesion between the starch and the PLA. The mechanical properties were superior for the PLA/gelatinized blends.

The objective of the research of Ke and Sun [67] was to characterize starch and poly(lactic acid) (PLA) blends

under varying water contents. It was determined that the initial moisture content (MC) of the starch did not have significant effects on its mechanical properties but it did have an effect on the absorption of water which was strongly affected by voids in the mixtures. The thermal and crystallization properties of the PLA in the mixture were not affected by the moisture content, but the crystallinity was affected by the processing conditions.

Ke et al. [68] mixed four starches with different amylose contents and a moisture content of 30 % with PLA at various ratios. The results indicated that the tensile strength and elongation at break of the blends decreased as the starch content increased, and the speed and degree of water absorption of the starch/PLA blends increased with increasing proportions of starch, as it was affected by the amylose content, while the PLA phase became discontinuous as the starch content increased beyond 60 %.

Shin et al. [69] investigated the rheological properties of the TPS/PLA blends and explained their relationship with their morphology. To improve the dispersion and interfacial affinity of the thermoplastic starch (DTPS)/PLA blend, Wang et al. [70] used glycerol, formamide and water alone or in combination as plasticizers for starch. The results showed poor dispersion and compatibility between glycerol plasticized starch and PLA. When formamide and water were added, the plasticization of the starch improved, increasing the dispersion of the mixture and decreasing the interfacial tension; however, the mechanical properties of the TPS/PLA blends did not satisfy the practical application, especially the elongation at break. Wang et al. [71] studied the compatibility of TPS/PLA, plasticizing starch in the presence of glycerol and formamide, excluding water. The DTPS and PLA blends were compatible to some extent by the effect of formamide.

Li and Huneault [72] investigated the properties of TPS/PLA blends, where the TPS phase is plasticized with sorbitol, glycerol and glycerol/sorbitol. It was found that in all the mixtures investigated, the PLA formed the continuous phase and the TPS formed a dispersed phase; however, the TPS phase plasticized with sorbitol can be more finely dispersed and distributed with greater uniformity in the PLA matrix. Even in the absence of any compatibility, the TPS/PLA blends plasticized with sorbitol showed a much higher tensile strength and modulus. It was found that the glycerol/sorbitol ratio has a significant effect on the properties of the mixture.

Shirai et al. [73] evaluated the effect of adipate and citrate esters as plasticizers in PLA/TPS blends. Diethyl adipate was reported to improve elongation and water vapor permeability. The incorporation of adipate or citrate esters improves the mechanical properties and processability of PLA/TPS sheets.

## Compatibilized TPS/PLA Blends

The hydrophobic nature of PLA and the hydrophilicity of thermoplastic starch are thermodynamically immiscible, which leads to low interfacial adhesion and compatibility between the mixture of TPS and PLA [70]; this lack of affinity leads to weak mechanical properties [74], evidenced in the thermal and morphological characteristics reported in the literature [27, 66]; in their research, it was determined that the main drawback is related to the nonmiscibility of polymers due to differences in chemical structures [75].

The term miscibility describes the interactions between the components in a polymer blend and the level at which they are homogeneous. On the other hand, the term compatibility indicates the final properties of the mixture and is related to miscibility; consequently, polymer blends that are classified as miscible and partially miscible are compatible, while immiscible blends are incompatible [55]. A strategic approach implemented to improve the properties of these blends is the addition of a coupling or compatibilizing agent with functional groups capable of interacting with the hydroxyl groups of the starch to improve the interaction between the two phases. Figure 4. represents the expected changes in the TPS/PLA starch blends after the addition of compatibilizer, showing a relatively homogeneous distribution compared to blends without compatibilizer. To achieve blends with improved properties, copolymerization, grafting, transesterification and the use of reactive coupling agents have been applied successfully [76]. According to the literature, compatibilization strategies can be classified as chemical crosslinking, amphiphilic bridging, component modification and interfacial transition [53], Table 1. presents some of the studies on PLA-TPS blends with compatibilizers.

With this objective, various compatibilizers and additives have been investigated to improve the interfacial interactions of these mixtures. Huneault and Li [77] obtained TPS/PLA blends compatible with maleic anhydride grafts in PLA, which showed a much finer dispersed phase size. The mixtures exhibited better ductility and elongation at break compared to the control. In a similar study, to improve the compatibility of the poly (lactic acid)/thermoplastic starch blends, Akrami et al. [78] grafted starch with polyethylene glycol grafted with maleic anhydride (mPEG-g-St). The results indicated that the interfacial adhesion was improved, possibly due to the interaction between the carboxylic acid groups of the free end of the compatibilizer and the active groups of the TPS and PLA phases. Several studies have explored the use of compatibility with grafts such as acrylic acid (PLA-g-AA/starch) [79], amylose grafted with PLA (PLA-g -A) [80], glycidyl methacrylate (GMA) (PLA-g-GMA) [81, 82], polylactide (PLA) grafted on the surface of starch granules

(St) (St-g-PLA)[83, 84], and oligo (lactic acid) (OLA-g-starch)[85], and it was demonstrated that more homogeneous morphologies improved the mechanical properties and thermal stability of the compatibilized TPS/PLA blends [36].

Ferrarezi et al.[74] proposed a new pathway for preparing TPS/PLA blends using poly(ethylene glycol) (PEG). The addition of PEG resulted in increased crystallization of PLA due to its plasticizing effect and the improvement of the interfacial interaction between TPS and the PLA matrix. The results show that PEG addition increased the impact strength of the ternary mixture and that the elastic modulus remained similar to that of the TPS/PLA blend.

Many researchers have explored the effects of isocyanate-derived compatibilizers to cross-link PLA and starch molecules; the isocyanate functional groups can react with the starch hydroxyl group or the terminal carboxyl group of PLA to form urethane bonds and improve interfacial adhesion [53, 86]. Ke and Sun [87] made mixtures of triethyl citrate as a plasticizer and methylenediphenyl diisocyanate (MDI) as a compatibilizer. It was found that triethyl citrate improved the elongation at break and toughness; however, it did not improve the coupling effects of MDI in starch and PLA. Recently, the effect of phenyldiisocyanate (PDI) on the properties of mixtures was studied with the use of starch modified with citric acid (CA), and it was discovered that PDI resulted in a better size distribution of the TPS particles in the PLA matrix and better interfacial interactions between two phases [86]. Another approach used by the researchers to improve the compatibility of the TPS/PLA mixtures are ternary mixtures with the incorporation of another polymer that provides a transition phase that should have better compatibility and interfacial adhesion, these coupling agents result in greater ductility and elongation at break compared to pure PLA and better dispersion of the TPS phase in the PLA matrix, which have been demonstrated in the studies of TPS/PLA and poly (ethylene octane) [82]; PLA/TPS/polycaprolactone (PCL)[88–90]; PLA/TPS/poly (butylene adipate-co-terephthalate) (PBAT)[91] and poly (ethylene-co-vinyl acetate) (EVA) blends [92].

Yang et al.[93] studied the preparation and characterization of thermoplastic starches and their mixtures with poly(lactic acid) by chemical modification of the starch without plasticizer, and two new types of TPSs were successfully prepared (MA-g-starch and Epicard-g-starch). The results showed a dispersed phase size for Epicard-g starch, improving the interfacial adhesion between PLA. On the other hand, Noivoil and Yoksan [94], improved the compatibility of the TPS/PLA blends by replacing the TPS of native cassava starch (TPSN) with the TPS of acetylated starch in different proportions. Scanning electron microscopy (SEM) confirmed a better dispersion and a smaller phase size for the TPSA/PLA blend films with a low degree of acetylation.

The compatibility strategy that has attracted attention recently is the incorporation of epoxidized vegetable oils in PLA and starch blends through the chemical modification of the components so that it has better affinity. Several studies have reported that the addition of epoxidized vegetable oils can improve the ductility of PLA and reduce the cost of products without compromising biodegradation. Przybytek et al.[95], obtained compositions of PLA/TPS/epoxidized soybean oil (ESO), the results showed that the addition of ESO in the PLA/TPS composition improved water resistance and impact strength. In a similar study, Xiong et al.[96] proposed a mixture of polymers with epoxidized soybean oil (ESO) as a reactive compatibilizer, and the starch granules were grafted with maleic anhydride (MA). The characterization of the mixtures demonstrated the formation of a more compatible compound. The elongation at break and the impact strength of PLA/ESO/starch increased significantly compared to those of the PLA/starch blends.

Recently, blends of starch and poly(lactic acid) were produced, including the epoxidized oil of thistle seeds, and it was evidenced that the plasticizing action of the oil induced improvements in the interfacial adhesion between PLA and TPS but a weak miscibility at the molecular level [97]. Turco et al.[97] prepared biodegradable films based on poly(lactic acid) (PLA) and thermoplastic corn starch (TPS) and added epoxidized thistle oil (ETO) plasticizer. Improvements were observed in the interfacial adhesion between hydrophobic PLA and hydrophilic TPS. However, there were no improvements in the mechanical properties due to phase separation between the PLA and TPS polymers.

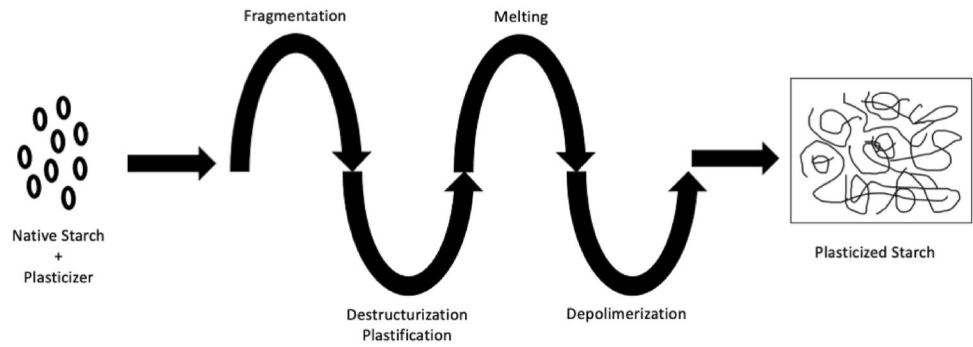
Several studies have reported the plasticizing-compatibilizing effect provided by epoxidized vegetable oils. Ferri et al.[60], obtained TPS/PLA blends with maleinized linseed oil (MLO) used as a compatibilizer. The MLO improves the compatibility in mixtures of immiscible or partially miscible polymers, as well as an increase in the elongation at break.

Other additives were incorporated into the TPS/PLA systems. Citric acid increases dispersion and decreases the interfacial tension between TPS and PLA [21, 98]. Tween 60, linoleic acid and zein were used as additives for the production of thermoplastic starch (TPS), and the amphiphilicity facilitated the processability, fluidity and extensibility of the blends. Linoleic acid was a more effective additive to improve the fluidity of TPS-PLA blends than Tween 60 and zein [99]. Table 2 presents the results obtained in the above-mentioned investigations regarding the formulations that offer the highest degree of compatibility concerning the proportion of TPS/PLA mixtures and compatibilizers used.

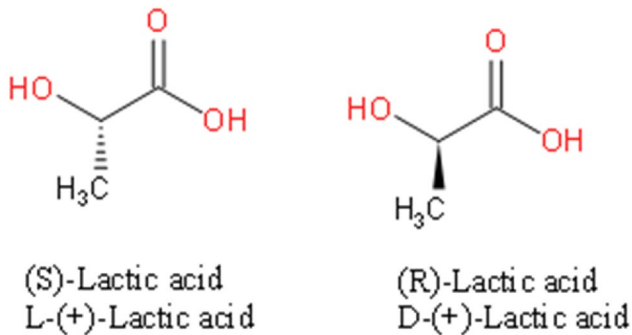
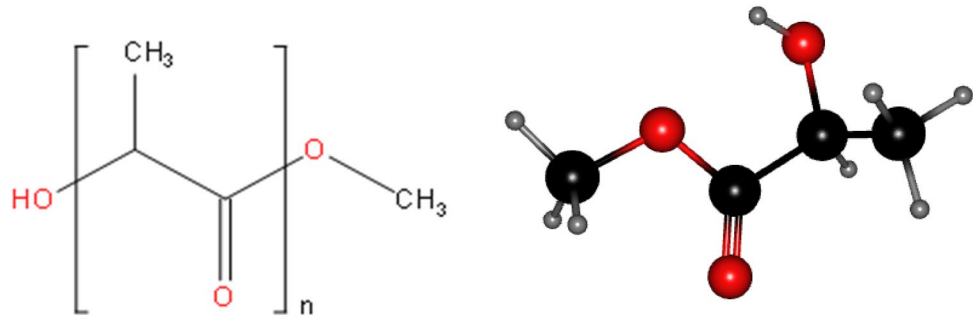
## Reinforced TPS/PLA Blends

The PLA blends with thermoplastic starch lead to changes in their characteristics. Generally, if a polymer is not miscible

**Fig. 1** Process of plasticized starch [29]



**Fig. 2** Chemical structure of poly (lactic acid) (PLA)

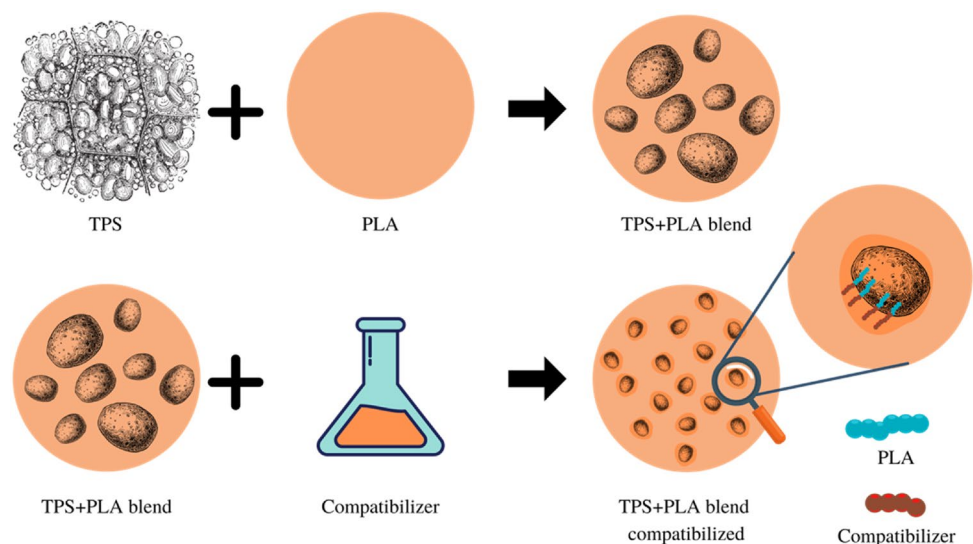


**Fig. 3** Chemical structure of l (+) and d (-) lactic acid

with starch, the mixture can be expected to have lower mechanical properties compared to both individual components. Although the compatibility between the PLA and TPS can be improved with the coupling agents, this may not sufficiently improve the mechanical properties [100]. However, the objective of the TPS/PLA blend is to minimize costs with acceptable properties. In recent years, attention has been given to strengthening mixtures with the incorporation of reinforcements to overcome their mechanical weaknesses.

Different investigations of biocomposites have incorporated natural fibers due to their numerous advantages, such as easy processing, low cost, biodegradability and better mechanical properties [101]. Additionally, the use of natural

**Fig. 4** Compatibilization process of TPS / PLA blends



**Table 1** Compatibilization strategies

Interphase compatibilization	Compatibilizer	Main changes	References
Chemical crosslinking	PLA- <i>g</i> -MA Maleic anhydride (MA)	The compatibilized blends contained 1–3 $\mu\text{m}$ TPS particle sizes, compared to 5 and 30 $\mu\text{m}$ in the blends without compatibilizer, they showed a much finer dispersed phase size The grafted MA acts as a compatibilizer at the domain/matrix interface, which influences the flexibility of the blends	[77] [129]
	OLA- <i>g</i> -starch Oligo (lactic acid)-grafted starch	TPS dispersed phase with smaller size and better distribution was obtained by adding OLA- <i>g</i> -starch	[85]
	Grafting of glycidyl methacrylate (GMA) in the presence of benzoyl peroxide (BPO)	The grafting favored the dispersion and processability of the blown films, improving the interaction between PLA and TPS due to GMA carboxyl or hydroxyl groups. The resistance to elongation at the break of blown films also improved	[130]
Amphiphilic bridging	(A- <i>g</i> -PLA) PLA-grafted amylose  Tween 60 Linoleic acid (LA) Zein  (PLA- <i>g</i> -MTPS) poly(lactic acid) grafted with maleated thermo-plastic starch (St- <i>g</i> -PLA) grafted with poly (lactic acid)	Amylose grafted with PLA increased the tensile strength of PLA/TPS blends containing 60% by weight of TPS by 60% The incorporation of amphiphiles resulted in better plasticization and processability. Likewise, it provided greater extensibility and crystallinity. However, the addition of amphiphiles decreased the stiffness, tensile strength, and stiffness of TPS materials The mechanical properties of the mixture were remarkably improved compared to those of the normal mixture The presence of starch- <i>g</i> -PLA results in changes in the particle surface and an increase in the elongation of the thermoplastic material	[80] [99] [131] [84]

Table 1 (continued)

Interphase compatibilization	Compatibilizer	Main changes	References
Compositional modification Compatibilization of the TPS/PLA mixture given they are thermodynamically incompatible. It is done by modifying the components so that the modified component has a higher affinity [53, 132]	Epoxidized Sesame Oil	The epoxidized oil acted as a coupling agent in the blends, improving their interfacial adhesion by enhancing PLA-TPS molecular interactions Surface heterogeneity in composite films is decreased with the addition of ESO, suggesting that the epoxidized oil acts as a coupling agent increasing the deformation and the loss of stiffness as well as the tensile strength	[133] [134]
Interfacial transitioning The TPS/PLA blend is supplied with a transition phase that allows a better interfacial adhesion in both PLA and TPS phases. Therefore, to have good compatibility the transition component must have affinity in both phases of the blend and the interfacial tension of the transition component must be lower than that of TPS/PLA [53]	Starch acetylation  Maleinized linseed oil (MLO)  PLA/ABS/TCSP Poly(lactic acid (PLA), Acrylonitrile-Butadiene-Styrene (ABS), and Tapioca cassava starch powder (TCSP) Poly ( $\epsilon$ -caprolactone) (PCL) and methylene diphenyl diisocyanate (MDI)  (GPOE) glycidyl methacrylate grafted poly (ethylene octane)	Acetylated starch blends exhibit improved mechanical properties due to the replacement of the hydroxyl groups in the starch molecule by acetyl groups [135]  The addition of MLO leads to elongation at a break of 140–160% and allows it to interact with the poly (lactic acid) chain ends and hydroxyl groups on the thermoplastic starch, leading to compatibilization [60]  Significant improvements are observed in ternary blends compared to pure and binary polymer blends. The blends also show good homogeneity and particle distribution [136]  The addition of methylene diphenyl diisocyanate (MDI) improves the interaction of TPS/PLC/PLA mixtures, probably due to the urethane bonds formed by the hydroxyl groups of TPS and the carboxyl groups present in PLC and PLA [89]  The distribution of particles in ternary mixtures is smaller than in binary mixtures, due to the reaction between TPS and GPOE [82]	



**Table 2** Summary of formulations that increase compatibility in TPS / PLA blends

Compatibilizer	Strategy	Mix ratio compatibilized			Compatibility		References
		TPS w%	PLA w%	Compatibilizer w%	Control	Response	
Poly (ethylene glycol) (PEG)	Interfacial Transitioning	56	19	25	TPS/PLA blends	↑	[74]
Maleic anhydride (MA) PLA-g-MA	Chemical Crosslinking	27	73	2	TPS/PLA blends	↑	[77]
Acrylic acid (AA) PLA-g-AA	Chemical Crosslinking	40	60	5,96	PLA unmodified, and starch/PLA blends	↑	[79]
PLA-grafted amylose (A) A-g-PLA	Amphiphilic Bridging	60	40	1	TPS/PLA blends	↑	[80]
Glycidyl methacrylate (GMA) PLA-g-GMA	Chemical Crosslinking	50	50	15	TPS/PLA blends	↑	[81]
Glycidyl methacrylate grafted poly (ethylene octane) PLA-g-GPOE	Chemical Crosslinking	20	80	10	TPS/PLA blends	↑	[82]
Grafted Starch (St) PLA St-g-PLA	Amphiphilic Bridging	–	75	25	TPS/PLA blends	↑	[83]
Oligo (lactic acid) grafted starch OLA-g-starch	Chemical Crosslinking	50	50	3–5	TPS/PLA blends	↑	[85]
Phenylene diisocyanate (PDI)	Interfacial Transitioning	40	60	1	TPS/PLA blends with citric acid	↑	[86]
Methylenediphenyl Diisocyanate (MDI)	Interfacial Transitioning	45	55	0,5	TPS/PLA blends with triethyl citrate	↑	[87]
Polycaprolactone (PCL)	Interfacial Transitioning	50	40	10	TPS/PLA blends	↑	[88]
		–	70	30	PCL/PLA, PCL/TPS and PCL/PLA/TPS varied formulations	↑	[90]
Poli (adipate-co-terefalato de butileno) (PBAT)	Interfacial Transitioning	50	20	30	TPS/PLA blends	↑	[91]
Epoxidized cardanol grafted starch (Epicard-g-Starch)	Chemical Crosslinking	–	70	30	Maleic anhydride grafted starch (MA-g-starch)/ PLA	↑	[93]
TPS from acetylated starch (TPSA)	Componential Modification	–	50	50	TPS/PLA blends	↑	[94]
Epoxydized soybean oil (ESO)	Componential Modification	25	75	2	TPS/PLA blends	↑	[95]
Citric acid (CA)	Chemical Crosslinking	50	50	1–4	Poly (lactic acid)/thermoplastic dry starch (PLA/DTPS) blends	↑	[98]

Arrows up in the response column indicate that compatibility increased under the presented formulation compared to the control blend for each case

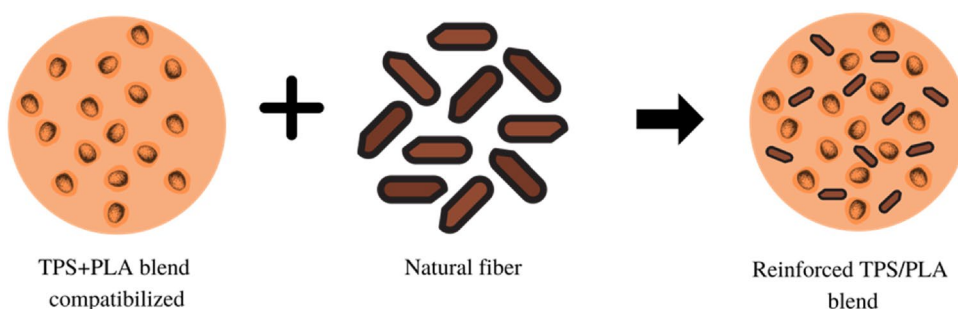
fibers such as kenaf, hemp, flax, jute, and bamboo, among others, are excellent ecological alternatives to synthetic fibers for the sustainable development of the bioindustry [102]. Figure 5 shows the general dispersion of natural fibers used as reinforcements in TPS/PLA starch blends.

Teixeira et al. [100] studied the addition of cassava bagasse, which is a by-product of the extraction of starch, to

produce reinforced thermoplastic starch. The results revealed that the incorporation of bagasse fiber possibly acted as a filler in the PLA matrix since there was no increase in the tensile strength of the TPS/PLA blends.

Bocz et al. [103] developed fully biodegradable compounds consisting of TPS/PLA biopolymer matrices reinforced with chopped flax fibers, resulting in outstanding

**Fig. 5** Reinforcement with natural fibers of TPS / PLA blends



mechanical performance and reduced flammability through a synthesized multifunctional additive system.

Smithipong et al. [104] studied the effects of pineapple leaf fiber (PALF) as a filler reinforcement on the properties of compounds with a TPS and TPS/PLA matrix. This new TPS with PALF/PLA compounds presented better mechanical properties and water resistance than the TPS/PLA blend.

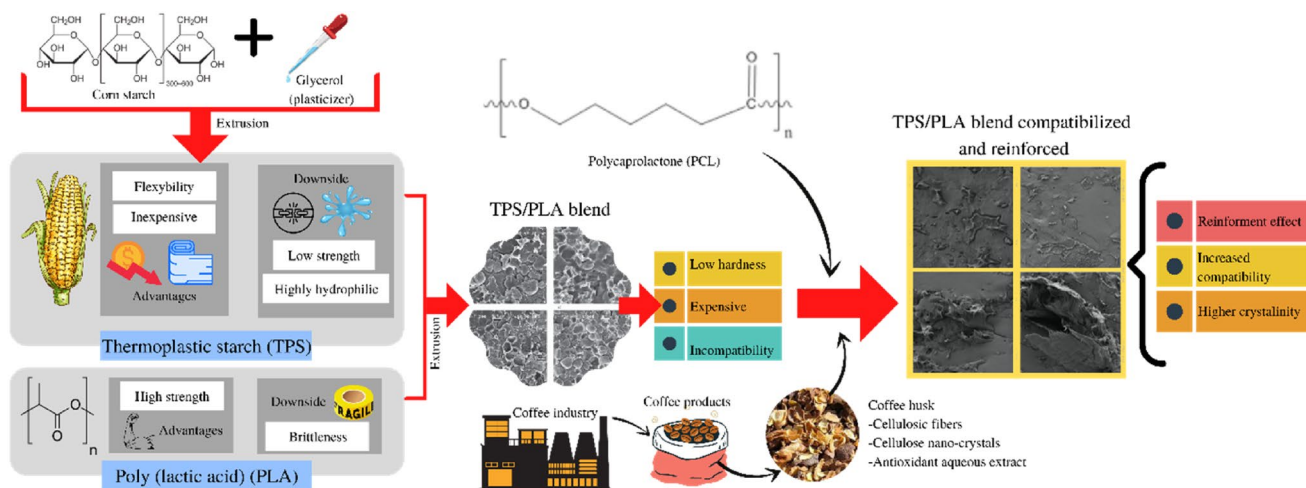
Iovino et al. [105] studied the aerobic biodegradation of compounds formed by poly(lactic acid) (PLA), with and without the addition of maleic anhydride (MA), acting as a coupling agent, thermoplastic starch (TPS) and short natural coconut fiber. In a similar publication, Surin et al. [106] prepared compounds with PLA and TPS modified with maleic anhydride (MA) using coconut fiber as reinforcement. The results suggested that the addition of coconut fibers increased the fragility of the TPS/PLA and MTPS/PLA compounds. However, the fiber content had a great effect on biodegradation only for the MTPS/PLA compound. Recently, the addition of coconut fiber (CF) as a reinforcing agent was studied again, and blends of TPS/PLA/CF biocomposites were obtained. The results suggest that CF improved the dispersion and compatibility of the TPS and PLA phases because they exhibited a greater rigidity and hardness and a

lower extensibility, tensile strength and impact strength than the TPS/PLA blend [3].

Nunes and Dos Santos [107] prepared mixtures reinforced with natural cotton fibers, seeking to increase tensile strength without compromising biodegradation. The reinforcement of the samples with cotton fibers (10 and 20 %) contributed to reduce the cost and limiting the deterioration of the simulated water and the soil.

N.R et al. [108] investigated the effect of wood fiber reinforcement (WF) on the mechanical properties of TPS/PLA mixtures, together with the use of a PLA copolymer (MA-g-PLA) grafted with maleic anhydride as a coupling agent. The study showed improvements in its mechanical properties and revealed a homogeneous mixture of TPS and PLA, as well as a uniform distribution of wood fibers in the matrix. The improvement in the properties was due to the synergy between the reinforcement with wood fiber and the use of a coupling agent.

Collazo-Bigliardi et al. [109] proposed the incorporation of lignocellulosic fractions (cellulosic fibers: CF and cellulose nanocrystals: CNC) of coffee husks to improve the functional properties of the compatibilized PLA and starch film, it was found that the previously incorporated CNC in the starch phase was effective in reinforcing the



**Fig. 6** Coffee husk used as reinforcement in a compatibilized TPS / PLA mixture [109]

tensile properties of the material and in the reduction of water vapor permeability and oxygen permeability of the films. Figure 6 synthesizes the effects of coffee husk lignocellulosic fractions on the compatibilized S-PLA films.

Jullanun and Yoksan [110] evaluated the effects of cassava pulp (CP) on the properties of a thermoplastic mixture of cassava starch (TPS)/poly(lactic acid) (PLA). It was determined that CP could act as a reinforcement depending on the concentration, with the objective of improving the tensile strength and Young's modulus of the compounds and as a nucleating agent for PLA in the biocomposites.

### Nanocomposite TPS/PLA Blends

Interest in nanotechnology has increased in recent years, and scientists, engineers from academia and industry are investigating the use of nanostructures since it was discovered that they could be built from a polymer and layered nanofiller [111] to achieve better mechanical, thermal, rheological and biodegradation properties of polymers compared to conventional microcomposites.

Liao and Wu [112] reported the preparation of biodegradable nanocomposites from poly(lactic acid) (PLA) or poly(lactic acid) grafted with acrylic acid (PLA-g-AA), titanium tetraisopropylate and starch by an in situ sol-gel process. According to the results, the PLA-g-AA/TiO<sub>2</sub>/starch hybrids could significantly improve the thermal and mechanical properties of a PLA-g-AA/starch hybrid, since it provides a smaller starch phase size and a dispersion at the nanometric scale of TiO<sub>2</sub> in the polymer matrix.

Arroyo et al. [113] investigated the properties of nanocomposites combining thermoplastic starch (TPS), polylactic acid (PLA) and natural montmorillonite (MMT). The results suggest that the clay particles showed a higher affinity for the TPS phase or at the mixing interface. They also reported an improvement in modulus and tensile strength but a reduction in fracture strength, demonstrating that TPS/PLA mixtures and TPS/PLA/MMT compounds have stable mechanical properties over a period of 300 days.

Zhang et al. [44] studied the effect of the polymer chain extender on the properties of the TPS/PLA mixtures, and the results suggested that the dispersion and compatibility between TPS and p-PLA improved significantly, increasing the tensile strength, elastic limit and elongation at break.

Ayana et al. [114] prepared biodegradable nanocomposites of thermoplastic starch (TPS)/polylactic acid (PLA)/sodium montmorillonite (NaMMT) blends highly exfoliated to evaluate the effects as a compatibilizer on the properties of the TPS/PLA/nanoclay nanocomposites. The results suggest an increase in the mechanical and thermomechanical properties in the presence of PLA and a greater influence in the presence of clay; in addition, a considerable decrease in the water absorption in the nanocomposites is observed.

Ferreira et al. [115], unlike the works cited above [113, 114], suggested the incorporation of an organically modified clay with increasing contents (1.0–10.0% by mass) into the polymeric components. The results revealed a better adhesion between the phases in comparison with the unmodified binary mixtures; likewise, the shear viscosities indicated a better compatibility as the content of organoclay increased.

According to Shayan et al., an evaluation was made on the effect of modified thermoplastic starch (MTPS) and montmorillonite nanoclay particles (MMT) on the mechanical properties and biodegradability of chemically cross-linked PLA (XPLA). The results showed that the modification of thermoplastic starch with maleic anhydride (MA) improved the dispersion of starch in the XPLA matrix, consequently improving the tenacity and biodegradability of the final product [116, 117].

Other research conducted for mixtures of thermoplastic starch and polylactic acid is the incorporation of nanoclays, according to Paglicawan et al. [118] and Ebrahimi et al. [119]. They prepared and studied the morphological and mechanical properties of blends of PLA and thermoplastic starch with nanoclays, and the results showed that the addition of nanoclay increased the mechanical properties due to better adhesion between the two phases. Tensile studies revealed that the increase in nanoclay content contributed to an increase in the Young's modulus and a decrease in tensile strength.

Jeziorska et al. [120] investigated the properties of PLA/TPS/silica nanocomposites hardened with polydimethylsiloxanol in the presence of maleated PLA (MPLA), which was used as a compatibilizer. The results indicate that polydimethylsiloxanol improves the ductility and initial thermal stability of the plasticized blend. Likewise, it was found that the addition of silica significantly increased the elongation at break of the material.

Recently, the effect of graphene nanoplates has been studied to improve the properties of thermoplastic starch/poly(lactic acid) polymer blends. It was found that the graphene nanoplatelets were located at the interface of the PLA and TPS phases. In addition, it was found that the percentage of crystallinity for all samples containing graphene is higher than that of the TPS/PLA mixtures [121].

Nazrin et al. [122] prepared bionanocompounds of thermoplastic starch blends reinforced with sugar palm nanocellulose fibers (TPS)/poly(lactic acid) (PLA) and evaluated the effects of different proportions of TPS (20%, 30%, 40%, 60%, and 80%) and constant sugar palm nanocellulose fibers (0.5%). The results indicate that the increase in the TPS fill decreased the general mechanical properties of the bionanocomposites of the mixture due to the fragility of the TPS.

## Future Perspectives

Research on renewable polymers has been carried out for a long time. However, the interest in using material through various types of blends and composites has become increasingly prosperous due to the limitation of crude oil and the increased concern for environmentally friendly material [123], as is the case of PLA/TPS blends that have a great prospect as biocomposites due to both components possessing a biological basis as biodegradable [53].

According to Nofar et al. [36], the development of PLA/starch blends is of great interest for food packaging applications and could also be considered in biomedical applications. While Hamad et al. [55], claim that the high stiffness of compatibilized PLA/TPS blends limits their applicability as food packaging films. Plasticizers are generally needed to improve the flexibility of this system. Considering several points, including the rapid biodegradation and high toughness of the compatible blend, in addition to the high flexibility achieved by using plasticizers, PLA/TPS/compatibilizer/plasticizer is a suitable candidate for food packaging film applications.

Every day the research on the development of PLA/TPS blends seeks to improve their final properties, in order to give different applications to the existing ones in new areas, such is the case of the work developed by Castillo et al. [124], who adds poly (butylene adipate-co-terephthalate) that increased the elasticity of PLA materials, while the presence of thermoplasticized starch promoted a higher rate of biodegradation of the materials investigated. Furthermore, the prepared materials do not show a phytotoxic effect on the plant, which demonstrates their potential application in the forestry industry as a biodegradable carrier for plant multiplication. On the other hand, Rydz et al. [125], discuss recent trends in agricultural application including the use of natural and synthetic (bio)degradable polymers such as agar, starch, pectins, alginates, chitin, lignin, cellulose, and its derivatives, PLA and PCL to replace traditional plastics. Soil retention films, agricultural films, seed or compost tapes, and binding materials, threads, clips, stalks, compost bags, silage envelopes, seed trays, and pots are being replaced by (bio)degradable equivalents and disposable items so that the (bio)degradable used do not have to be removed after use.

The current and future trends in the use of TPS, PLA, and their blends can be an effective solution to different environmental problems, since they combine good processability with biodegradation and the use of renewable raw materials [126], which is why their applications in the areas of food, forestry, and agricultural packaging are growing every day, requiring a great demand for materials for their development. Despite the biodegradability of the constituent materials of the blends, Dorigato [126] assures that the growing presence

of biopolymers in the plastic waste stream makes the recyclability of bioplastic-based blends particularly important for both academia and the plastics industry. Therefore, further research on PLA/TPS blends should be considered to further improve their final properties, using plasticizers, compatibilizers, and other additives, thus achieving application interest in other areas, developing biocomposites that can be integrated within a circular economy (CE) model to ensure a sustainable product that is environmentally-friendly [127].

## Conclusions

The research carried out has the common objective of obtaining a new biodegradable material generated from low-cost renewable sources with properties comparable to petroleum-derived polymers. Many researchers and industries have focused their efforts on the preparation of compounds based on thermoplastic starches from different sources and plasticizing agents that seek to reduce hydrophilicity. On the other hand, polylactic acid (PLA) is considered the most promising biopolymer for the replacement of conventional plastics, but its production cost limits its application. PLA has hydrophobic characteristics, so it is immiscible with most of the available polymers, which makes processing the mixture with thermoplastic starch difficult. Even so, to improve their compatibility, different strategies such as the addition of plasticizers, coupling agents, reinforcements and nanocomposites seek to improve the interfacial adhesion of the two materials and obtain a final product with good mechanical, thermal and homogeneous morphological properties without phase division. In addition, the combination of different strategies that provide a wide range of methodologies in the TPS/PLA blend was found, increasing the properties and efficiency of the combination.

The research carried out over the last few years for the development of blends based on TPS/PLA offers a convenient option for generating novel properties with high performance for applications in different areas of materials science, manufacturing technologies, polymers, agriculture and agri-food. Therefore, future research works still have a lot of space to work on the processing and compatibility of these materials, which will continue to grow in the search for a new low-cost material that is completely biodegradable and easy to process and produce on an industrial scale to compete with petroleum-derived plastics.

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**Authors Contribution** KMV: Research, Formal analysis, Visualization, Writing—Original Draft, MJAT: Conceptualization, Investigation,

Supervision, Writing—Original Draft, JU and DEM: Writing—Reviewing and Editing.

### Declarations

**Conflict of interest** The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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