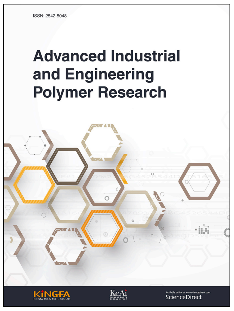

Re-evaluating Bioplastic Blend Wastes through Mechanical Recycling and Chemical Modification

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Re-evaluating Bioplastic Blend Wastes through Mechanical Recycling and Chemical Modification

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Abstract

The escalating environmental challenges posed by conventional plastics have amplified the importance of biodegradable polymers as sustainable alternatives. However, addressing their recyclability and reprocessing is critical to enhancing their environmental and economic viability. This review delves into the multiple reprocessing of biodegradable polymer blends, focusing on mechanical recycling's effects on their structure, properties, and performance. Unlike single polymers, blends offer tailored properties by combining the strengths of individual components, making them more suitable for diverse applications. However, their complex morphologies and phase interactions demand unique strategies for effective recycling.

Key findings highlight that polymer blends, such as PLA/PHB and PLA/PBAT, exhibit greater resilience to repeated processing compared to their pure counterparts, owing to enhanced intermolecular interactions and progressive crystallinity. Compatibilizers, including chain extenders like Joncryl®, play a pivotal role in mitigating degradation by improving phase adhesion and maintaining mechanical and thermal properties. Rheological analyses reveal the critical interplay between phase morphology and processing conditions, emphasizing the importance of tailoring blend compositions and additives for optimal recyclability.

This review sets itself apart by providing the first comprehensive examination of the effects of multiple mechanical reprocessing cycles specifically on biodegradable polymer blends, filling a significant gap in the literature. By addressing current challenges, it offers a roadmap for advancing biodegradable materials toward a circular economy.

Keywords:

Biodegradable polymer blends, Compatibilization, Mechanical Recycling, Multiple re-processing

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Contents

1. Introduction

Plastics are cost-effective, lightweight, and long-lasting substances that can easily be shaped into various items suitable for numerous purposes across diverse fields. As a result, there has been a significant rise in plastic production over the past six decades. Yet, the present rates of their utilization and disposal contribute to various environmental challenges. Approximately 4% of the global oil and gas output, which are finite resources, are utilized as raw materials for plastics production, with an additional 3-4% being consumed for energy during the manufacturing process. A significant amount of plastic manufactured annually is utilized for creating disposable packaging or other temporary goods that are thrown away within a year of production. These facts alone suggest that our current plastic usage is not environmentally viable. Furthermore, due to the long-lasting nature of the polymers used, considerable amounts of discarded plastics are piling up as waste in landfills and natural environments globally [1]. Commercial plastics often consist of a base polymer combined with various additives, plasticizers, pigments, or other polymers. The proportion of polymer in a product can range from nearly 100% to less than 20%, depending on its intended use [2-4]. That is, plastics are integrated into nearly every facet of daily life. However, a few sectors account for the majority of plastic consumption. In the EU member states, packaging represents the largest use of plastics (39%), followed by building and construction (23%), the automotive industry (8%), electrical and electronic equipment (6%), houseware, leisure and sports (4%), and various other applications (20%) (Fig. 1).

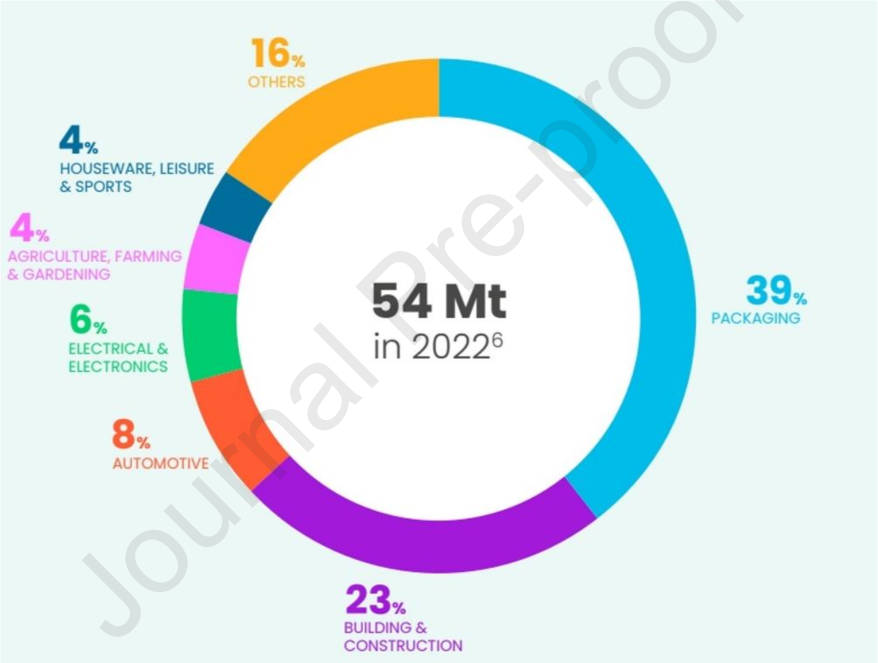

Fig 1. Plastic demand by application in Europe (Plastics – the fast Facts 2023)[5].

Plastics offer several advantages over materials like metal, glass, and paper in various applications. Nevertheless, the disposability of many plastic-based products, particularly those used for disposable packaging, has resulted in a significant impact on the environment. Due to their resistance to microbial degradation, landfills are overflowing with plastic waste jeopardising the environment at a high rate [6-8]. To understand why plastics remain so persistent in the environment, it is crucial to examine their chemical composition and the properties that confer such durability. These factors not only determine how plastics behave during their functional life but also influence their end-of-life fate, including biodegradability and recyclability. Consequently, understanding the specific makeup of plastics is central to developing environmentally responsible alternatives and addressing the current waste crisis. The scientific community has been sounding alarms regarding the risks associated with the worldwide accumulation of plastic waste for over a decade [9, 10]. Despite these warnings, plastic pollution remains a pressing concern. Without a shift in the current global production and disposal system, the projections for 2040 paint a grim picture [11, 12].

Most materials used for temporary and disposable packaging are non-biodegradable, failing to meet the requirements for environmental safety and sustainability. As a result, these materials continue to pose a risk to both human health and the environment [13]. While there are limited studies on detecting microplastics contamination in everyday food, some research has shown the presence of microplastics in common food products, including beverages, plant-based items, marine products, as well as regular additives like salt and sugar, which are summarized in Fig.2 [14].

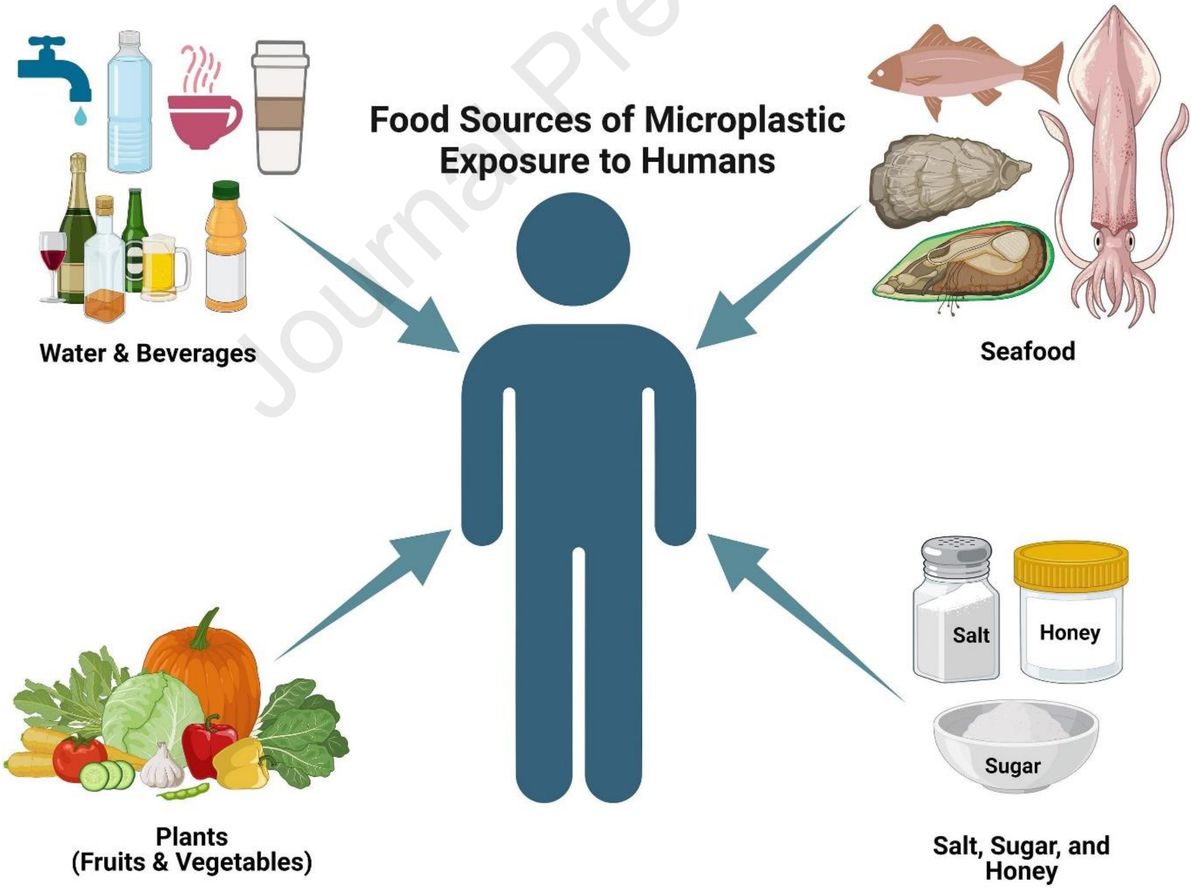

Fig 2. Microplastics in human food chains (reproduced picture) [14].

The environmental footprint of traditional plastics, including their resistance to microbial degradation and their accumulation in natural environments, highlights the urgent need for sustainable alternatives. In response, biodegradable plastics have gained attention as a potential solution to mitigate these challenges. These materials, derived from renewable or fossil resources, offer advantages such as biocompatibility, biodegradability, and non-toxicity. However, their adoption is not without challenges, including high manufacturing costs and limited mechanical performance. To fully understand their potential, it is essential to examine their classifications, properties, and limitations [15-17]. Recently, biodegradable polymers have gained popularity due to their numerous uses in packaging, agriculture, and biomedical devices which is illustrated in Fig.3.

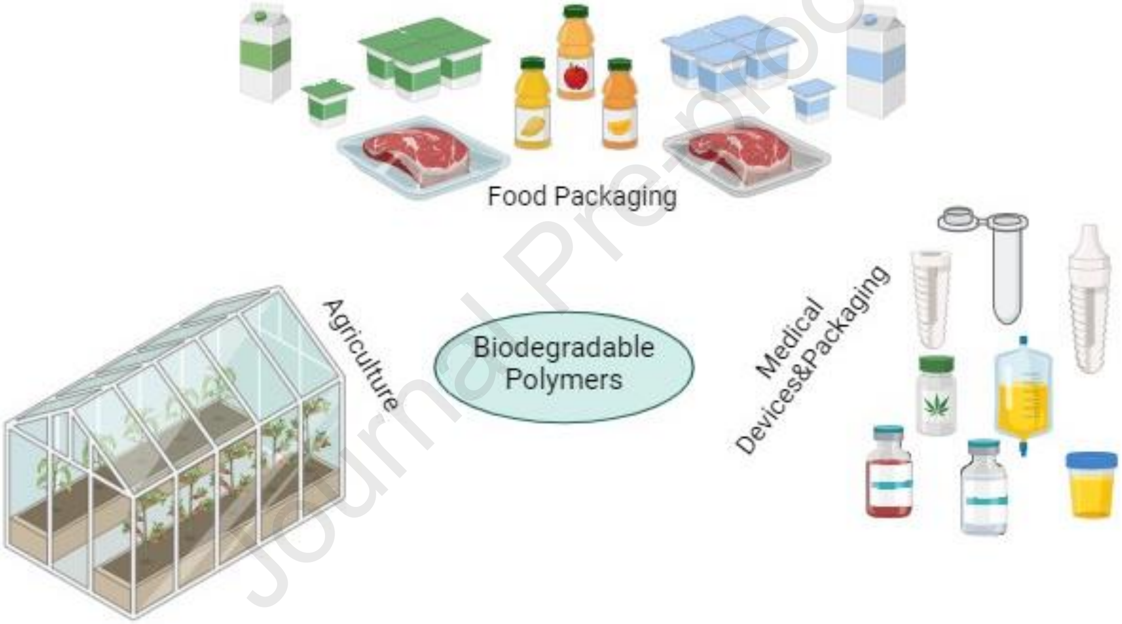

Fig 3. Applications of biodegradable polymers in food packaging, agriculture, and biomedical devices

According to the sources and procedures used in their production, biodegradable polymers can be divided into many categories. In terms of the environment, raw material is classified based on its source: renewables (i.e., biological), and non-renewables/fossil resources (i.e., oil, natural gas, and coal)[18]. Fig. 4 indicates biodegradable polymers based on two resources including renewable and petrochemical resources[19]. Among the common biodegradable polymers, linear aliphatic polyesters such as polylactic acid (PLA), polycaprolactone (PCL), poly (butylene adipate terephthalate) PBAT, and polyhydroxybutyrate (PHB) are the most popular [20-22]. These plastics, whether sourced from biomass or fossil fuels, can serve as a substrate for microorganisms [23].

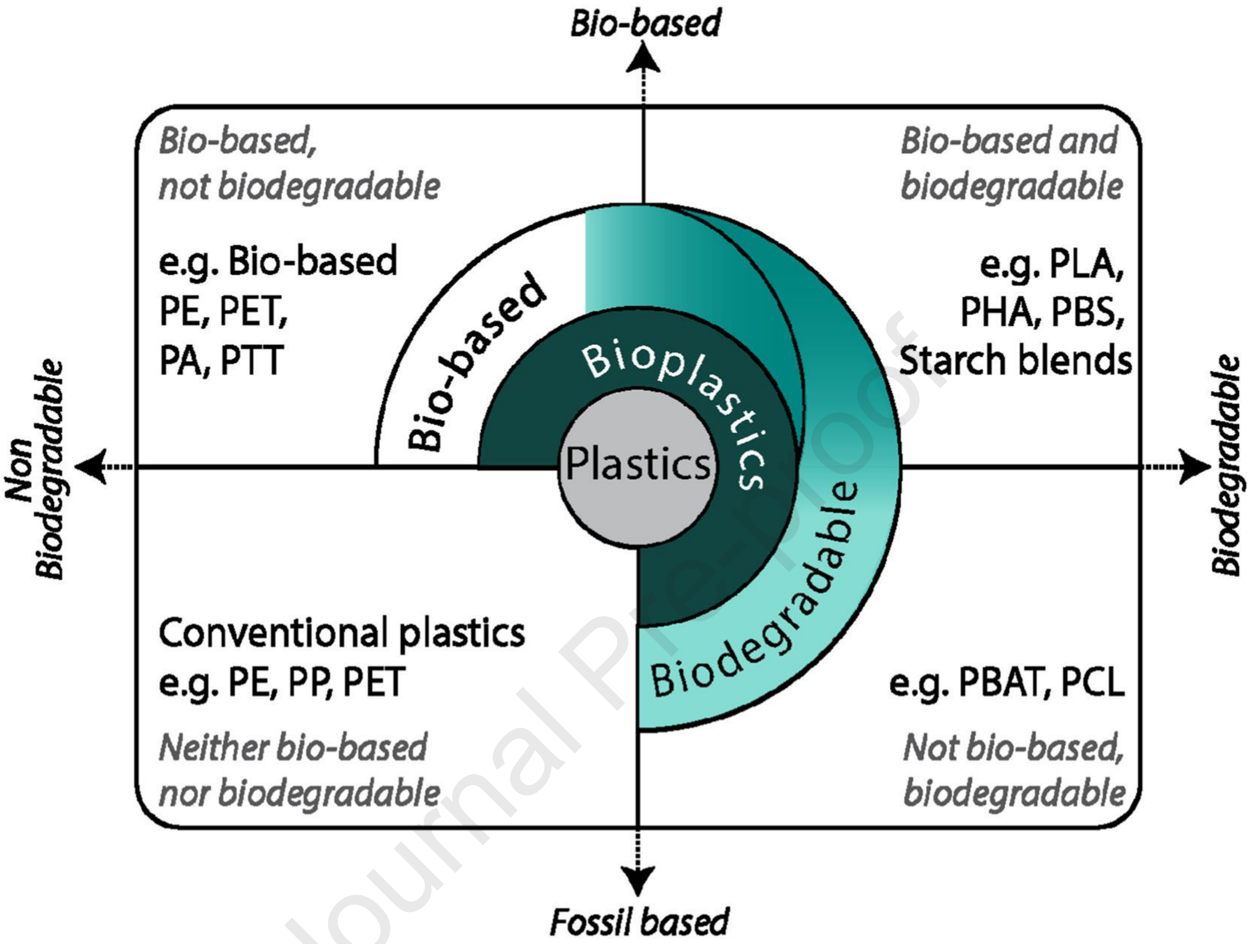

Fig 4. Classification of biopolymers. Retrieved from [19].

However, despite their attractive properties, these biodegradable polymers have limitations such as their high manufacturing costs, insufficient mechanical performance, and inferior thermal stability, preventing them from becoming widely used as disposable packaging in the packaging industry [24-26]. Most biodegradable plastics are made to biodegrade under certain circumstances, most frequently in a commercial composting facility, where temperatures regularly reach 50°C for weeks or months at a time. This, however, contributes to additional costs associated with energy consumption. In essence, these biodegradable polymers are more suited for composting than traditional biodegradation[27]. A study by the packaging platform Sourceful revealed that just 3% of compostable packaging in the U.K. reaches appropriate composting facilities. The report highlighted that due to insufficient composting infrastructure, 54% of this packaging is sent to landfill, while 43% is incinerated [28]. Another study by Purkiss *et al.* [29] indicated that 60% of home-compostable plastics fail to completely decompose in household compost bins, ultimately accumulating in the soil. Therefore, recycling could be considered a better solution for waste management of biodegradable plastics. Biodegradable plastics can be recycled to increase their sustainability and lower their carbon impact before their end-of-life (EOL) degradation. Recycling of all wasted or rejected durable plastics is highly desired and biodegradable plastics are no exception. Sustainable use of biodegradable plastics also includes the condition for resource efficiency, implying that materials should also be recyclable as post-consumer waste[30]. It has been reported that the mechanical recycling and chemical recycling are the most widely practiced methods, which have been focused by most of the studies.1 However, from industrial point of view, the mechanical recycling is the most suitable [31-33]. While considering the benefits of mechanical recycling, it is essential to acknowledge that this strategy is not without its challenges. The most significant issue is that recyclers often lack interest in recycling biodegradable materials, even though these materials are theoretically recyclable. This is primarily because they are seen as contaminants in conventional recycling streams, such as polyolefin or polyethylene terephthalate (PET) recycling[34]. If this issue were to be resolved, and specific recycling facilities were established for biodegradable plastics, like those for conventional materials, another challenge would remain. A significant portion of biodegradable prototypes and commercially available products are blends, as single polymers may exhibit various shortcomings, including brittleness, low thermal resistance, and limited processability [35-38]. Table 1shows several commercial applications for the bio-based blends.

Table 1. Commercial bio-based polymer blends

Brand name	Blend type	Forms & Applications	References
Bio-Fed,(a AKRO-PLASTIC GmbH, Germany)	PBAT/PLA	Films for food packaging	[35]
Terraeck ® BD (Green Dot Bioplastics, USA)	Blends made from synthetic and natural biodegradable polyesters	Film blowing and film casting applications	[39]
Mater-Bi® NF 866	Various grades of thermoplastic starches	Films for food packaging or agricultural mulch films	[40]
PLANTIC ™	Corn harvesting & Starch with PE and PET	Flexible film (PLANTIC™ EF, PLANTIC™ TF, PLANTIC™ PF) Skin (PLANTIC™ RV, PLANTIC™ VL, PLANTIC™ ES) Paper (PLANTIC™ PF, PLANTIC™ PV) Barrier Sealants (PLANTIC™ CF, PLANTIC™ BF) Mono Layer (PLANTIC™ HP) Rigids (PLANTIC™ R, PLANTIC™ E, PLANTIC™ RP, PLANTIC™ RE, PLANTIC™ RM, PLANTIC™ RB)	[41]
BIOTEC	Potato starch and other biologically sourced polymers	Film	[42]
Forzeas ™	Bio PBS ™	Film&Sheet, Polymer&Resin	[43]
TIPA ® Compostable Packaging	Blend of compostable polymers	Compostable films& laminates	[44]
Terrafilum	PLA/ABS&PLA-PP	Filament	[45]
Nonoilen Fillamentum / addi(c)tive polymers	PLA, biopolymers fiber from real wood, polyethylene terephthalate glycol (PETG), polyether-block-amide (PEBA), polyvinylidene fluoride (PVDF) , Olefinic block copolymer (OBC), PC/ABS,	Filament	[46]
Ataraxia Art Flexible	FPLA (Flexible PLA) PLA/TPU	Filament	[47]
Total Energies Corbion	Luminyl® Polylactic acid (PLA) bioplastics	Fibers	[48]
ecovio ® (BASF, Germany)	PLA/PBAT	Resin, Plastic packaging products	[36]
Bio-Flex ® (FKuR, Germany)	PLA/PBAT	Resin	[37]

Agrobiobase	Nature plast PHI001(PHA (corn 55%)	Resin	[49, 50]
Danimer scientific company (Nodex®)	Polyhydroxyalkanoates (PHAs) with other types of biopolymers, like PLA	Plastic resins	[51]
Green Max, Aqua Max Bio	Compostable irrigation system made from corn starch and bioplastic granulate Cradonyl®	Tubes	[52]
ColorFabb	PLA/PHA	Biological scaffolds for tissue engineering applications	[53]
Greener Walker	Bio corn starch blend Eco-friendly	100% Leak Proof Biodegradable Dog Poo Bags	[54]
Polyester Estabio (PL 0640 T05)	Blend of biodegradable polyesters, partially from renewable resources (PLA/PBS)	Cutlery and coffee capsules	[55]

It is interesting to note that, the extent of their recyclability has not yet been thoroughly examined. Thus, it is crucial to systematically investigate the recyclability of these products and explore strategies to enhance it, thereby supporting their overall sustainability.

The blend of two polymers offers a possible solution to overcome these restrictions. Bioplastic-based polymer blends combine bioplastics with other polymers to create materials with desirable properties such as improved strength, durability, flexibility, and heat resistance [16]. Polymer blends are composed of two or more dissimilar polymers that are mixed physically with or without chemical interaction. By using simple physical processes rather than chemical ones, polymer blends can provide desired properties at a lower cost than copolymerization [24]. In addition, polymer blending is an inexpensive and straightforward method for improving the properties of existing materials [56]. It should be also noted that processing conditions as well as post-processing conditions could alter the morphologies of the blends, hence changing the final properties. This implies that recycling of such systems requires more care with respect to retaining their structures and applications. It is due to these challenges that recycling of blend materials is much more difficult than pure materials. In this regard, there have been several review articles addressing the recycling of virgin biodegradable polymers; however, to this date, no comprehensive review on the mechanical recycling of biodegradable blends has been reported. Hence, this review focuses on the effect of multiple processing and recycling on the structure and properties of biodegradable-based polymer blends. We will discuss the mechanical, thermal, rheological, and biodegradation performance of the blends with respect to the number of cycles the blends go through during the mechanical processes, simulating the recycling conditions.

2. Biodegradable blend

There is an increasing demand for polymeric materials with improved properties to fulfill the needs of different applications. Typically, an individual polymer does not have all the necessary characteristics. Creating and producing an entirely new polymer can be both time-consuming and expensive [57]. The idea behind creating a new mixture of two or more polymers is not to significantly change the properties of the individual components but to make the most of the blends optimal performance[58]. A polymer blend is a combination of two or more polymers mixed to form a new material with distinct physical characteristics. Polymer blending has garnered significant interest due to its simplicity and cost-effectiveness in creating versatile polymeric materials suitable for various commercial applications. By carefully choosing the component polymers, the properties of the blends can be tailored to meet specific end-use requirements[59]. There are two primary categories of blends: miscible (homogeneous) blends and immiscible (phase separated structures) blends that can be seen in Fig 5.

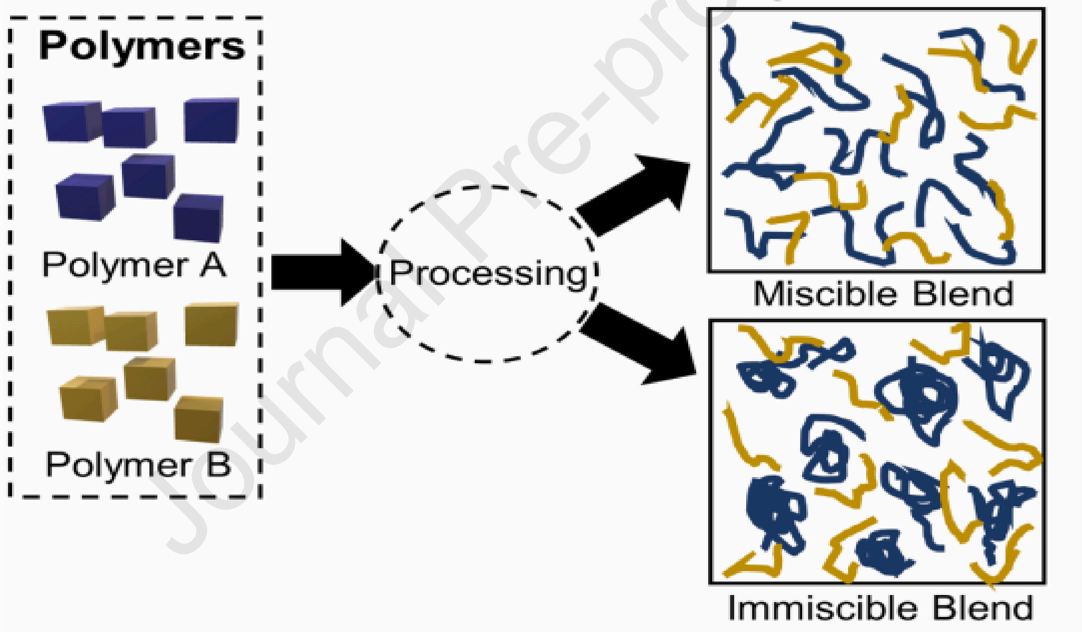

Fig 5. Illustration of miscible and immiscible polymer blends [57].

Miscibility is a thermodynamic concept that indicates the number of phases in polymer blends [24, 60]. Miscible blends form a single phase and exhibit a single glass transition temperature, which is influenced by the individual components [61]. In contrast, thermodynamically immiscible blends have a phase separated structures with large inclusion phases, minimal interaction or adhesion at the interface between components, and two distinct glass transition temperatures, regardless of the specific components in the blend [24]. Thanks to the various morphologies from immiscible blends, properties can be tuned if compositions, interfacial tensions, viscosity ratios and processing conditions are manipulated. Processing conditions such as annealing, screw speed, and other extrusion parameters significantly influence the morphological and mechanical properties of polymer blends. For instance, annealing can induce crystallization, altering the phase distribution and improving properties like stiffness or thermal resistance, while variations in screw speed impact the dispersion of components, affecting the blend's toughness and elasticity. These processing parameters are critical, as they determine the degree of phase separation or compatibility between immiscible polymers. Due to the immiscible nature of most polymer pairs, the blends exhibit phase-separated (segregated) morphologies and they possess lower thermomechanical property as they have poor interfacial adhesion[62]. During each recycling process, the blends undergo consecutive stretching and folding, which alters the original morphology and affects the final properties. This process may use a compatibilizer, depending on the composition and viscoelastic properties of the individual components to minimize these morphological changes during mechanical recycling, a chain extender (as a compatibilizer) will be incorporated into the blend.

3. Compatibilization

The inherent immiscibility of biodegradable polymers, due to unfavorable mixing entropy, leads to phase separation, coarse morphologies, and poor mechanical properties, limiting their applications. This immiscibility causes weak interfacial adhesion, reducing tensile strength and impact resistance, and complicating manufacturing processes. Compatibilization strategies aim to improve interfacial interactions, reduce interfacial tension, stabilize morphology, and enhance mechanical performance[63, 64]. Without compatibilizers, blend properties are significantly lower than those of the original polymers. Effective compatibilization strategies include (i) non-reactive methods using co-solvents or copolymers, (ii) reactive methods generating copolymers in situ, and (iii) nanofiller-induced methods with small amounts of nanofillers. The choice depends on cost, performance, recyclability, and biodegradability[62, 65-67].

Additives known as chain extenders have been designed to enhance melt strength, thermal stability, and serve as compatibilizers in polymer blend. The most widely used of these chain extenders is a multifunctional styrene–acrylic oligomeric chain extender marketed as Joncryl® ADR. Interestingly, Joncryl® is frequently referred to as a synonym for chain extenders; however, the Joncryl® portfolio from BASF SE includes more than just some styrene-acrylic-based oligomers. It is divided into various classes, each serving different functions. These classes include flow modifiers (ADF), plasticizers (ADP), dispersants (ADD), and the widely recognized reactive chain extenders (ADR). According to the supplier, the primary goal of these additives is to enhance the processability and properties of recycled polymers. In fact, the application of Joncryl® in polymer processing is well-established and recognized in both academic and industrial settings. Typical applications of Joncryl® include its basic function as a stabilizing agent during processing, as well as more sophisticated uses, such[68] as in recycling and foaming, where enhanced rheological and melt properties are crucial. Joncryl® ADR initially patented by Johnson Polymer LLC[69], this company was later acquired by BASF SE. Additionally, masterbatches containing this chain extender, which are approved by the Food and Drug Administration (FDA) for food packaging, are available from Clariant AG under the trade name "CESA®-extend." The chain extender Joncryl® ADR was initially intended for use in injection molding and extrusion processes with virgin, post-industrial recycled, and/or post-consumer recycled polyesters such as PET and PBT, as well as polyamides (PA), bisphenol A-based PC, thermoplastic polyurethanes (TPU), polyoxymethylene (POM), and blends thereof to mitigate degradation effects. Additionally, it is frequently used with other polyester polymers like PLA, which is not extensively developed as a common commercial polymeric product[70-72]. Chain extenders allow the reconnection of polymer chains that have been broken by degradation reactions, resulting in an increased Mw of the polymer (Fig 5) [73-75]. Multiple studies have demonstrated that incorporating Joncryl® ADR 4368 significantly enhances the elongation viscosity of PLA, exhibiting notable strain hardening behavior[76, 77]. Villalobos *et al*. [71]demonstrated that even low concentrations (<1.5 wt.%) of Joncryl® ADR 4368 can increase molecular weight to levels comparable to unprocessed PET, while providing a gel-free processing window ideal for stable recycling. This study prompted further research on Joncryl® ADR's impact on recycling behavior. In addition, Salehiyan *et al*.[78] observed Joncryl ® ADR 4468 at 0.5 wt.% resists degradation, showing stable modulus after re-processing with higher viscosity and modulus after re-processing cycles compared to unmodifed PBAT. Therefore, it can be suggested that composites with such outstanding properties are suitable for blending with other biodegradable polymerss. Tavares *et al*. [79] investigated the use of Joncryl® ADR 4368 and Joncryl® 4370 (Polyad PR 002) with neat PET (nPET) and recycled PET(r-PET) during extrusion. Their findings revealed that Joncryl® was more effective in r-PET than in virgin PET due to the higher reactivity of shorter chains in r-PET. Additionally, adding less than 1 wt.% of Joncryl® was sufficient to counteract PET degradation. Cailloux *et al.* [80, 81]have also demonstrated that the use of Joncryl® ADR 4300 can improve the melt rheological properties of PLA. In another study, Makkam *et al*. [82] examined the effect of Joncryl® ADR 4380 on the melt behavior of recycled PET. Although the melt strength of recycled PET is initially too low to measure, it can be enhanced by adding 0.3 wt.% chain extender. At chain extender concentrations of 0.6 and 0.9 wt.%, the melt strength of recycled PET surpasses that of virgin PET. Additionally, its potential as a compatibilizer in the compounding of immiscible polymers will be explored in detail in the following section. Since Joncryl is commonly used in recycling processes to mitigate thermal degradation, slow the molecular weight (MW) loss rate, and act as a compatibilizer in polymer blends, it is likely to be employed in the recycling of such blends, either as a compatibilizer or to compensate for degradation[71, 83-85]. For instance, in a study by Wang *et al*. [86], high-strength and high-toughness PLA/PBAT blends were created using a melting-reactive blending method, incorporating Joncryl ADR-4370S (ADR) as a compatibilizer through an in situ compatibilization reaction. With the addition of ADR, the mechanical properties of all blends were improved. According to the toughening mechanism of PLA/PBAT/ADR, when ADR was not included, the PBAT segments developed droplet-like particles and aggregates within the PLA segments because of the poor compatibility between PLA and PBAT. Under high-impact loads, the substantial aggregation of PBAT resulted in an ineffective toughening of PLA. This was mainly evident as interfacial debonding of the dispersed phase droplets, which was due to weak interactions at the interfaces between the two phases and inadequate energy absorption. In contrast, with the introduction of ADR, the epoxy group of ADR interacted with the terminal carboxyl and hydroxyl groups of PLA and PBAT, creating a PLA-g-PBAT copolymer between the PLA and PBAT segments. This process significantly decreased the surface tension between the two phases. Fig (6) shows toughening mechanism of PLA/PBAT blends[86].

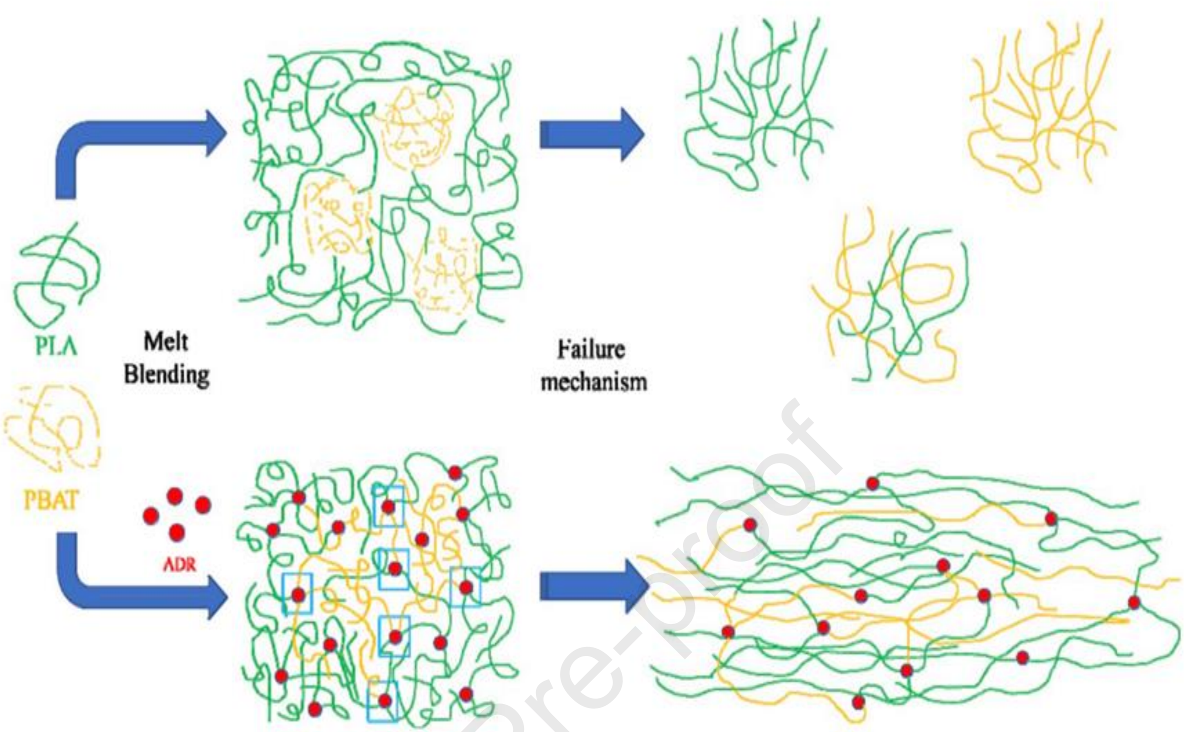

Fig 6. Toughening mechanism of PLA/PBAT blends[86].

3.1. Principle and mechanism of in situ compatibilization of polymer blends with functionalized polymer

Polymers with reactive functional groups (e.g., anhydride, epoxy, isocyanates) can serve as effective compatibilizers in immiscible biodegradable polymer blends due to the presence of reactive functional groups in many biodegradable polymers (Table 2) [24]. During melt blending, these compatibilizers react with functional groups such as hydroxyl or carboxyl groups in the blended components, facilitating the in-situ formation of graft and/or block copolymers.

Table 2. *In Situ* Compatibilization of Biodegradable Polymer Blends with Different Compatibilizer[24].

Blends	**Preparation method**	**Compatibilizer**	**References**
		Compatibilizer with epoxy functionality	
PLA/Starch	Compression molding	PLA grafted GMA	[87]
PLA/PBAT	Blown film	Multifunctional epoxy functionalized PLA	[88]
PLA/PBAT	Injection molding	Random terpolymer (abbreviated T-GMA)	[89]

PLA/Thermoplastic starch	Compression molding	Glycidyl methacrylate grafted poly(ethylene octane)	[90]
PBAT/starch	Blown film	Styrene-maleic anhydride-glycidyl methacrylate terpolymer	[91]
		Compatibilizer with maleic anhydride functionality	
PLA/PCL	Injection molding	MA grafted PLA	[92]
PLA/PBAT	Blown film	MA grafted PLA	[93]
PBAT/Soy protein concentrate	Injection molding	MA grafted PBAT	[94]
PLA/Soy protein concentrate	Injection molding	MA grafted PLA	[95]
PBAT/Starch	Extrusion foam	MA grafted PBAT	[96]
PLA/Starch	Compression molding	MA grafted PLA	[97]
PCL/Starch	Injection molding	MA grafted PCL	[98]
PBS/Starch	Injection molding	MA grafted PCL	[99]
PLA/Starch	Injection molding	MA grafted TOA	[100]
		Polymeric reactive compatibilizer	
PLA/Starch	Injection molding	HDI grafted starch	[101]
PBS/Novatein thermoplastic protein	Injection molding	PEOX and pMDI	[102]
PLA/Soy protein concentrate	Injection molding	PEOX and pMDI	[103]
PLA/Starch	Injection molding	PEG	[104]
PLA/Starch	Blown film	CP-grafted starch	[105]
PLA/Starch	Extruded film	Starch grafted ROM	[106]

PLA/Starch	Injection molding	MLO	[107]
PLA/Starch	Injection molding	ESO	[108]
PET/ EBA-GMA [109]	**Injection molding**	**EBA-GMA**	
Low molecular weight components as compatibilizer			
PLA/PBS	Injection molding	LTI	[110]
PLA/PBS	Compression molding	LTI	[111, 112]
PLA/PBS	Compression molding	MDI	[113]`
PLA/PCL	Injection molding	PDI, LDI, lysine trisocyanate, MDI, 1,3,5-tris(6-isocyanatohexyl)biuret, and 1,3,5-tris(6-isocyanatohexyl)-1,3,5-triazinane-2,4,6-trione.	[114]
PLLA/PCL	Compression molding	MDI	[115]
PCL/Soy protein	Compression molding	MDI	[116]
PLA/Soy protein	Compression molding	MDI	[117]
PLA/PBSA	Injection molding	TPP	[118]
PLA/PBSA	Compression molding	Joncry chain extender	[85]
PLA/PBAT	Blown film	Joncry chain extender	[73]
PLA/PPC	Casting	Joncry chain extender	[119]
PHBV/PBS and PHB/PBS	Compression molding	DCP	[120]
PLLA/PBS	Compression molding	DCP	[121]

PLLA/PBAT	Compression molding	DCP	[122]
PLLA/PBAT	Compression molding	2,5-Dimethyl-2,5-di(*tert*-butylperoxy)hexane	[123]
PLA/PHB	Compression molding	DCP	[124]
Combination of Peroxide with other functional chemicals			
PLA/PHO	Injection molding	DCP and triallyl trimesate	[125]
PLA/PHB	Injection molding	Benzoyl peroxide and MA	[126]
PBAT/Soy meal	Casting	DCP and MA	[127]
PLA/PBAT	Compression molding	Tetrabutyl titanate	[128]
PLA/PBAT	Injection molding	Tetrabutyl titanate	[129]
PLA/PPC-MAH	Compression molding	Tetrabutyl titanate	[130]
PLA/Poly(ω-hydroxytetradecanoic acid)	Injection molding	Tetrabutyl titanate	[131]
PLA/PPC	Compression molding	MA	[132]
PLA/PBAT	Compression molding	PA and BOZ	[133]
PLA/PCL	Compression molding	Glycidyl methacrylate	[134]
PLA/PBAT	Compression molding	Glycidyl methacrylate	[135]
PLA/Starch	Compression molding	DOM	[136]

3.2. Effects of nanoparticles on the morphology of immiscible polymer blends

Using nanoparticles as compatiblizers in polymer blends is an advanced strategy to enhance the compatibility and performance of immiscible or partially miscible polymer systems. Morphology refinement is probably the most renowned consequence of adding nanoparticles to immiscible polymer blends [64]. In blend nanocomposites, the final morphologies are largely influenced by the ultimate distribution of nanoparticles within the blend [64, 137]. The effects of compatibilization can be understood through two primary mechanisms: (i) inhibiting droplet coalescence and (ii) reducing interfacial tension, both of which contribute to smaller droplet sizes. These mechanisms are most effective when the compatibilizer is positioned at the interface. However, in polymer blends with incorporated particles, the mechanisms may differ from traditional compatibilization, as particle localization varies based on the surface energy relationships between the particles and the polymer pairs, which significantly influence their final placement. Additionally, the role of particles as compatibilizers remains unclear, as their ability to reduce interfacial tension is still debated [138-140]. Despite this, droplet size reduction is consistently observed in all particle-compatibilized blends. The mechanisms of droplet size reduction in polymer blends with the addition of particles can be summarized as follows:

i. Particles can disperse within the polymer matrix, increasing its viscosity and thereby lowering the viscosity ratio, which promotes droplet breakup [141, 142].
ii. A more effective approach occurs when particles are positioned at the interface, where they form a physical barrier around the droplets, preventing coalescence[142-145].

Studies have shown that nanoparticles can be initially mixed with the thermodynamically incompatible phase, prompting their migration to the more thermodynamically compatible phase during melt processing[146-148]. This process can result in the interfacial entrapment of nanoparticles, leading to optimized properties[149]. Regarding migration, it is important to note that spherical particles exhibit greater stability at the interface compared to particles with higher aspect ratios[146]. Table 3 shows different types of nanoparticles that were added to the polymer blends as compatibilizers.

Table 3. Morphology stabilization compatibilizers

Sample	Nanoparticles	results	Ref
PLA/PBAT	Cellulose nanocrystals (CNCs)	Co-continuous morphology, and improvement in mechanical properties	[150]
PLA/PBAT	nanoclay	Suppress coalescence and thus stabilize the morphology	[151]
PLA/PBAT	CNTs	Improvement in the interfacial adhesion, and tensile toughness of PLA and PBAT	[152]
PLA/PBAT	CNTs	Excellent mechanical and dielectric properties.	[153]
PLA/PBAT	CNTs	Prevent coalescence	[151]

PLA/PBAT	MWCNTs	Improvement in mechanical performance and increase in electrical resistance suggested their possibility of piezoresistive properties	[154]
PLA/PBAT	MWCNTs	Excellent electrical conductivity	[155]
PLA/PBAT	Al_2O_3	Improvement in impact strength and interfacial interaction	[156]
PLA/PBAT	Silica	Higher a thermal stability,	[157]
PLA/PBAT	Silica	Stabilizing the co-continuous morphology without any diminishing of the mechanical properties	[158]
PLA/PBSA	Organoclay	The numbers of dispersed phases and interfaces increased	[159]
PLA/PBSA	Clay	Stronger interfacial interaction, transfer of applied load from the main matrix to the dispersed phase	[160]
PLA/PBSA	Nanoclay	Good mechanical properties with minimal loss of thermal stability	[161]
PLA/PBS	SiC	Improvement in Young's modulus as well as impact strength	[162]
PLA/PBS	Clay	Increase in mechanical properties for nanocomposites of up to 3 wt.% of clay and improvement in Barrier properties by approximately 26% with the increase of clay content	[163]

4. Recycling

Despite biodegradability of biodegradable polymer blends, the recycling of them has become essential due to mass production of this material where the existing composting facilities may not meet the required market demand. Therefore, biodegradable plastics can be recycled to increase their sustainability and lower their carbon impact before their EOL degradation. It may seem strange to recycle biodegradable polymers, which are currently employed in many applications and are thought to be a sustainable alternative to oil-derived polymers because they are made from renewable resources and are biodegradable or compostable; however, there are several reasons to suggest that recycling of biodegradable polymers is a sensible strategy [164-166]. These are mostly related to the expanding industrial demand and the fact that recycling is essential for reducing the consumption of non-renewable resources (including the energy demand associated with their manufacture). Furthermore, some commercial biodegradable polymers do not undergo severe degradation under normal conditions, and the disposal of bioplastic-made items leads to the disposal of valuable raw secondary materials [33, 167]. Therefore, it is very interesting to investigate pre- and post-consumer recycling of goods manufactured of biodegradable plastics [1]. Recycling biodegradable plastics as opposed to virgin polymers results in greater primary energy demand savings and lessens the impact of global warming [15]. It is crucial to note that, in comparison to single-use items, the use of biodegradable polymers in long-term applications necessitates durability under more harsher circumstances and longer periods. Furthermore, not enough space and facilities are available to meet all the compositing/biodegradation needs. Moreover, mechanical recycling of biodegradable is crucial to ensure their sustainability in the long term. Recycling biodegradable plastics as opposed to virgin polymers results in greater primary energy demand savings and lessens the impact of global warming [168]. On this basis, the waste hierarchy applied to the specific case of biodegradable packaging materials can be considered according to the prioritization model presented in Fig.7 [169].

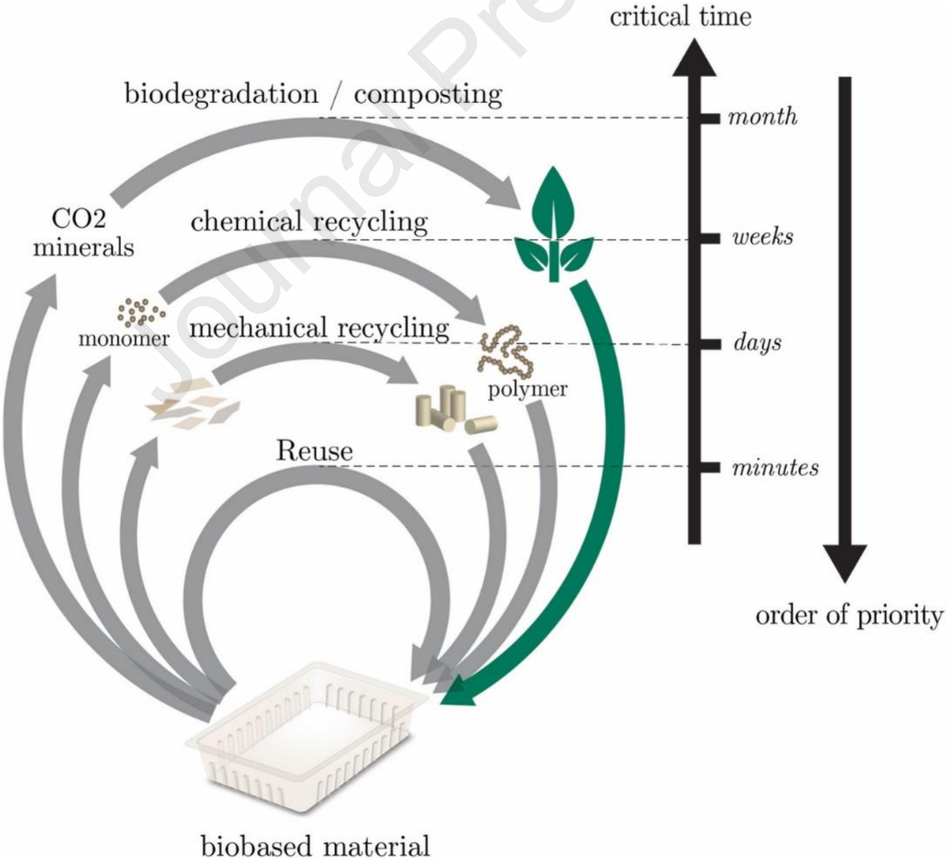

Fig 7. Waste hierarchy approach applied to biodegradable packaging materials [169].

In the previous section the role of compatibilizer in polymer blends were discussed, therefore, as far as compatibilized blends are concerned, their recyclability should be of an interest. For instance In a study by Srimalanon *et al.* [170], interestingly, the number of processing cycles had a notable impact on the elongation at break and tensile strength. The PBS/PLA blends achieved their highest elongation at break and tensile strength during the third processing cycle, which can be attributed to the formation of additional branching and/or cross-linked networks generated by the residual radicals of dicumyl peroxide (DCP) cross linker. The use of Joncryl® as a cross-linker in PLA/PBAT and PLA/PHB blends could significantly enhance phase adhesion, potentially mitigating the reduction in mechanical properties during multiple recycling cycles. However, further investigation is required to validate its efficiency under such conditions.

4.1. Types of recycling and applications of recycled plastics

After plastic waste has been gathered, sorted, and cleaned, there are four potential recycling methods.

Primary mechanical recycling: Primary recycling is a closed-loop recycling technique that is feasible only with high-quality plastic waste of well-documented origin [171, 172]. Primary mechanical recycling involves directly reusing uncontaminated polymer waste to produce new products without any degradation in properties. This process is typically carried out by manufacturers using post-industrial waste, leading to its common designation as closed-loop recycling. While post-consumer waste can also undergo primary recycling, it often presents additional challenges, such as the need for selective collection and manual sorting (Fig 8). These factors can substantially increase the cost of recycled materials, making this approach less popular among recyclers[173-175].

Secondary mechanical recycling: The exact composition and purity level of EOL (end-of-life) and PC (post-consumer) streams are often unknown. As a result, they are processed using secondary mechanical recycling, which includes separation and purification steps, unlike primary recycling. Similar to primary recycling, this process is typically limited to thermoplastic polymers[175]. In secondary recycling, the polymer itself remains unchanged; however, its molecular weight decreases due to chain scissions caused by the presence of water and trace amounts of acids. This reduction can lead to a decline in mechanical properties. To mitigate this effect, measures such as thorough drying, vacuum degassing, and the incorporation of stabilizing additives can be employed[176].

Tertiary or feedstock recycling: Tertiary recycling, also known as chemical recycling, involves chemical processes that break down plastic waste into monomers or other valuable materials. Chemical recycling methods vary widely, with each technique offering specific benefits and drawbacks depending on the type of polymer being processed. Examples of these processes include hydrolysis, pyrolysis, hydrocracking, and gasification and the typical conversion products are liquids and gases, which can serve as feedstock for producing fuels, new polymers, and various other chemicals [173, 175, 177].

Incineration or quaternary recycling: Incineration, used as a method for energy recovery, can also be considered a form of recycling[176]. This type of recycling refers to the energy recovery via incineration of low-grade plastic waste. Plastics are high-yielding energy sources, when burned the resulting heat energy is used to generate steam and electricity. Furthermore, quaternary recycling reduces waste volume to approximately 1% of its original size and effectively breaks down toxic and infectious waste. This makes it particularly suitable for recycling medical applications and hazardous goods packaging. Inorganic components are transformed into inert slag through incineration, which can subsequently be utilized in road construction [175, 177].

Mechanical and chemical recycling are reported to be the most commonly utilized methods, with the majority of the studies reviewed concentrating on these two approaches. However, mechanical recycling offers several advantages over chemical recycling, including lower processing costs, reduced global warming potential, decreased use of non-renewable energy, and lower levels of acidification and eutrophication. According to ISO 15270-2008 [178], mechanical recycling involves converting plastic waste into products or secondary raw materials without significantly altering the chemical structure of the material. Before the recycling process, certain steps are required, such as decontaminating the waste polymer (pre-treatment), identifying the plastic to enable separation, and reducing its size through grinding or shredding [178]. Mechanical recycling involves well-known technological techniques like extrusion and injection molding and has very straightforward requirements as shown in Fig.8 [168, 179]. Both these processes involve the following sequence of steps: (a) heating and melting the polymer, (b) pumping the polymer to the shaping unit, (c) forming the melt into the required shape and dimensions and (d) cooling and solidification [180].

Fig 8. Schematic illustration of mechanical recycling [168].

5. Mechanical and thermal Properties of recyclable polymer blends

In the following, the effects of multiple processing on mechanical and thermal properties of biodegradable-based polymer blends are discussed. Table 4 lists the details of the processing techniques and number of recycling (NOR) of fully biodegradable-based polymer blends.

Table 4. The information about processing techniques and NOR of fully biodegradable-based polymer blends

Blend	Processing Technique	Cycles	Ref
PBAT/PLA	SSE	5	[31]
PBAT/PBS	TSE	7	[181]
PBAT/TPS	TSE	7	[181]
PHBV/PLA	SSE	6	[182]
PLA/PHB	SSE	11	[183]
PLA/PHB	First process= TSE+ (re) SSE Second process= TSE + (re) SSE+ (re) 3D printing	First=9 Second=9	[184]
PLA/PHB	TSE+ Injection	5	[185]
PHBV/PBS PHBV/PBS/sepiolite PHBV/PBS/sepiolite/ PHBV-g-MA	SSE+ Injection	6	[186]
PHBV/PBAT	TSE+ Injection	7	[187]

PLA/PBAT	SSE	2	[188]
PLLA/PBS PLLA/PBS/Flax	TSE+Injection (re-Injection process)	7	[189]
PLA/PBS	TSE+Injection molding	10	[55]
PLA/PBS	TSE	5	[170]
PLA/TPS	TSE	4	[190]
PLA/PCL/SF/NP	TSE	3	[191]

5.1. Fully biodegradable-based polymer blends

5.1.1. PLA/PHB blends

In a study by Zembouai *et al*.[182], the goal was to describe the effects of recycling on the characteristics of (50/50) (PHBV/PLA) melt-molded blends that have undergone up to six additional processing cycles. In this study, blending was achieved with a single screw extruder (SSE). For the (PHBV/PLA) blend, the tensile modulus was slightly higher than that of PLA after the first cycle, but exhibited a reduction of about 9.6 %, decreasing from 4,007MPa to 3,621MPa after six processing cycle .Given the minimal difference observed between these values, it can be concluded that the tensile modulus of the (PHBV/PLA) blend remains relatively constant after six reprocessing cycles. Moreover, adding PLA to PHBV enhances the blend's strain at break. As a consequence, PLA and (PHBV/PLA) blends show less degradation in their mechanical properties after six cycles as opposed to neat PHBV which exhibits a degradation in its properties[182]. Moreover, the differential scanning calorimetry (DSC) analysis was done to evaluate the thermal properties of different samples including PLA, PHBV, and (PHBV/PLA) blend. The results showed that PLA significantly reduces PHBV thermomechanical degradation. Multiple extrusions of a specified composition of the (45/55) (PLA/PHB) blend were also employed by Plavec *et al.* [183], to evaluate the recyclability of the biodegradable blend. SSE was also used in this study for blending (11 extrusion cycles). The tested blend demonstrated relatively constant values of tensile strength of break in the range of 23 to 35 MPa during multiple extrusion processes. Degradation of materials and reduction in polymer chains length can also cause a decrease in relative elongation at break values. Similar to the study by Zembouai *et al*,[182] it can be concluded that the (PLA/PHB) blend is more suitable for multiple processing compared to the individual PLA or PHB polymers. However, unlike the previous study[182] where a reduction in strength was observed after 6 cycles, in this case, no significant decrease in strength was noted even after the 11th processing cycle [183]. Based on the results presented in this work, it can be concluded that the (PLA/PHB) blend shows stronger resistance against degradation at multiple thermomechanical stresses than PLA and PHB polymers alone, which benefits the processing process and mechanical properties of the tested material. In addition, thermal properties of (PLA/PHB) blend were measured by DSC. Results indicated that extrusion time had little impact on the thermal properties of materials. According to DSC data, there was not a major change in the material's thermal characteristics or crystallinity despite some degradation of the substance during multiple processes[183]. In another study, Plavec *et al.* [184] focused on how multiple processing through SSE and twin screw extrusion (TSE) affected the final mechanical characteristics of a (PLA/PHB) blend intended for three-dimensional (3D) printing(SSE+TSE=9 cycles, SSE+TSE+3D=9 cycles). The results showed that partial degradation during processing, which occurred when the molar mass of the polymer blend decreased, is what caused the flexural strength of tested samples values to fall. However, even after nine processing cycles, the maximum flexural strength of polymer blend drop is is just 7% when compared with the original, non-recycled sample. . Additionally, only minimal changes were made throughout multiple processing in the evaluation of Young's modulus and in the case of deformation at the flexural strength [184]. It can be said, the strength characteristics of repeatedly processed samples are not adversely affected. Furthermore, the findings of the thermal properties showed that successive processing during recycling does not significantly alter the ratio of crystalline and amorphous phases in either laboratory or industrial environments[184]. These mechanical and thermal findings align with those reported in [182, 183], in which the material recycling of (PLA/PHB) suitable blends for injection molding was evaluated. Similarly, blends of (70/30)(PLA/ PHB) were prepared by Farias *et al*. [185], and the impact of multiple mechanical recycling (TSE and injection) up to five times on mechanical properties were examined. The results showed that multiple recycling reduced tensile strength, however, this drop was not significant. To explain it more clearly, after five recycling cycles, the tensile strength decreased by about11 %, dropping from approximately 48 MPa in the control sample to around 43 MPa in the sample that had been recycled five times. Notably, multiple recycling did not substantially influence the tensile modulus and elongations at break of samples, and they remained almost stable throughout the processing cycle like the mentioned studies [182-185]. Fig. 9. shows the effect of multiple recycling on tensile properties of (PLA/PHB) samples.

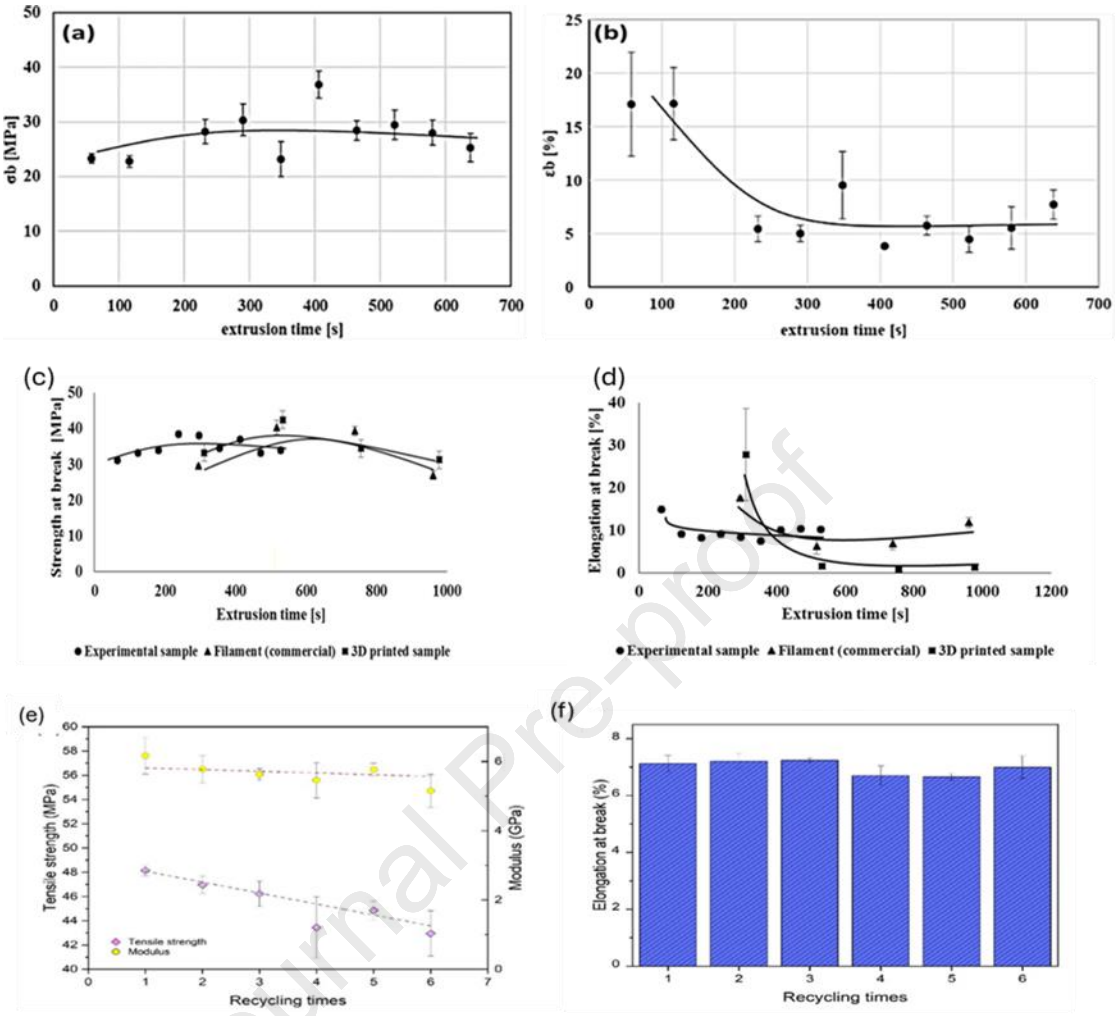

Fig 9. Shows (a,c)the dependency of strength at break on extrusion time of the tested (PLA/PHB) blend[183, 184], (b,d) dependency of elongation at break on extrusion time of the tested (PLA/PHB) blend[183, 184], and (e,f) the effect of multiple recycling on tensile properties of (70/30) (PLA/ PHB) samples[185].

The PLA that was injected multiple times had a similar behavior in all mentioned studies. This trend might be ascribed to the rise of crystallinity throughout recycling cycles. The multiple recycling procedure has the ability to reduce the Mw and degrade the polymer chains, which could deteriorate the tensile characteristics. Nevertheless, the increased crystallinity of the samples across the processing cycles offsets the negative effects of random chain scission in the current blend by enhancing intermolecular bonds within it. Fig.10 presents scanning electron microscopy (SEM) images of the fracture surface of the (70/30) (PLA/ PHB) blend. The micrographs indicated that the dispersion state and droplet morphology altered following the second recycling cycle. Figure 12A and 12B illustrate the presence of dispersed PHB particles with clear boundaries within the continuous PLA matrix. However, after more than two recycling cycles, the droplet size and distribution undergo significant changes, and the sharp boundaries become much less distinct. The size of PHB domains was reduced and more homogeneous discrete phase distributions were produced by SEM data (Fig.10).

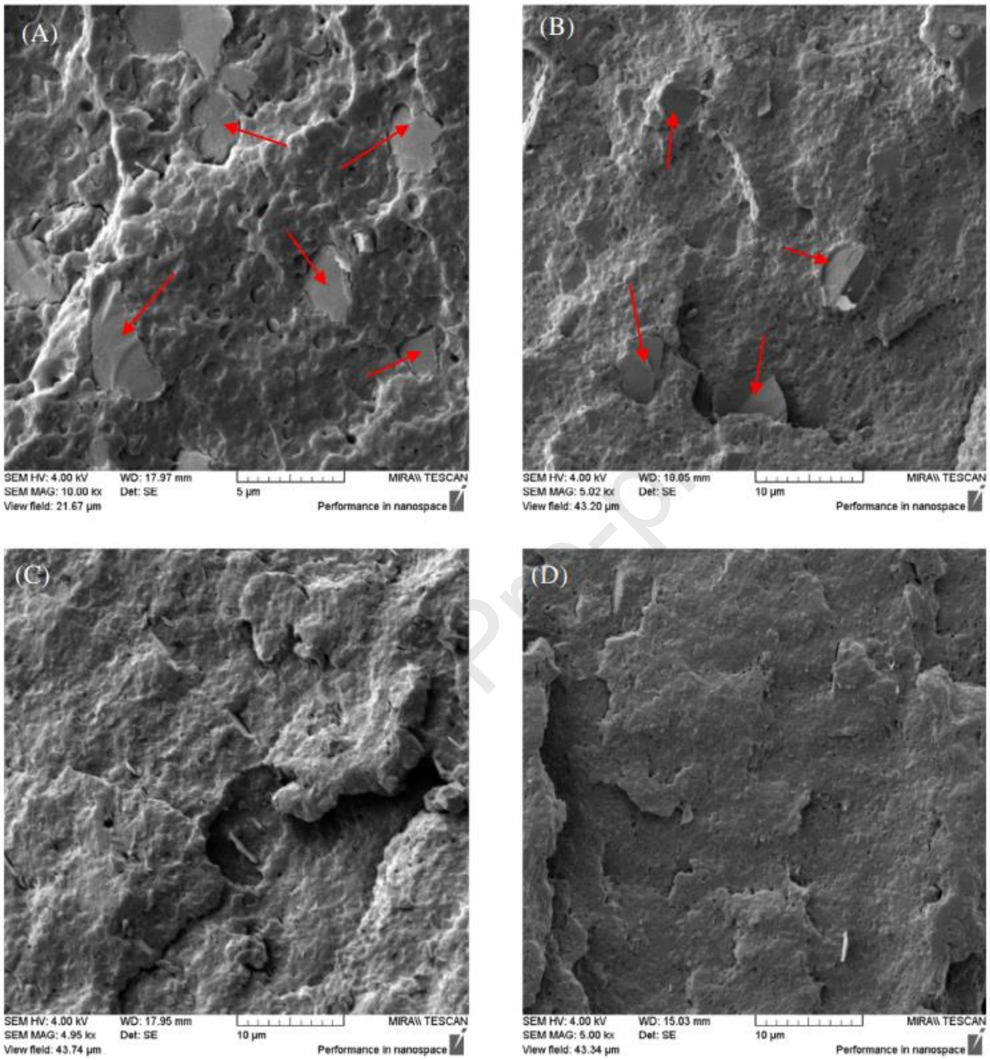

Fig 10. SEM micrographs of (A) control, (B) recycled-1, (C) recycled-3, and (D) recycled-5 (red arrows show some of the PHB droplets in the PLA matrix[185].

As a result, finer PHB particles in recycled sample may act as nucleating agents and help improve the crystallinity of the blended material.

5.1.2. Other PHB-based blends

In a study by Chikh *et al.*[186], by melt mixing, blends of (50/50) PHBV and Poly(butylene succinate)(PBS) were prepared, and PHBV was grafted onto maleic anhydride to obtain a 5 wt.% compatibilizing agent to improve miscibility. In this study, the materials were assessed by examining the impact of repeated extrusion cycles (SSE and injection moulding up to 6) in the presence of sepiolite and compatibilizer. Tensile and Charpy impact tests were used to examine how reprocessing cycles affected the mechanical properties of PHBV, PBS, and the (50/50) (PHBV/PBS) blend. All blend and nanocomposite samples showed a slight decrease in tensile modulus during reprocessing cycles, indicating that recycling has little to no impact on the stiffness of materials (Fig. 11). It can be said that PHBV exhibits the highest tensile modulus after the first reprocessing cycle, with about a 6% decline from 3960 MPa to 3722 MPa after six cycles. On the other hand, all of the tensile strength of samples dropped after six reprocessing cycles, with the exception of PBS, where they surprisingly rose from 29 to 37.4 MPa. The slight improvement in crystallinity of the injected specimen can be used to explain this outcome. Additionally, the effects of reprocessing cycles on the variations in impact strength of PHBV, PBS, (PHBV/PBS) blends, and their nanocomposites were assessed. The results showed that, as compared to neat PBS, all the samples had a lower impact strength. Changes in the molecular chain weight relate to a reduction in impact resistance over reprocessing cycles. Poor chain entanglements caused by shorter molecular chains and a wider chain length distribution reduced the toughness of the multiple processed samples [186]. The thermal stability of the samples was investigated by TGA. Regarding thermal properties, there is only one degradation stage visible in the neat polymers. In contrast, the TGA thermograms for (PBS/PHBV) blends reveal two different deterioration stages. The thermal stability of PHBV is just slightly enhanced after blending with PBS. This is in line with evidence from the literature [192] showing that blending PBS and PHBV improves the PHBV phase's thermal stability. This can be explained by interactions between the molecular chains of PBS and PHBV, which are in charge of delaying PHBV breakdown. In addition, TGA and DSC results showed that the increase in chain mobility with respect to reprocessing cycles can be connected with the reduction in Mw [186]. Mechanical recyclability of (PHBV/PBAT) blends was investigated by Resch-Fauster *et al.*[187] by analyzing the effects of repeated polymer processing (extrusion without additional compounding with virgin material or additives-extrusion for up to seven times) on the mechanical parameters. It was determined that repeated processing of the tested (PHBV/PBAT) blend grade resulted in hydrolytic molecular degradation of PBAT. The mechanical characteristics and, in the advanced stage, also the processability (phase separation of blend components in the molten state) were negatively impacted by the related changes to the molecular structure.

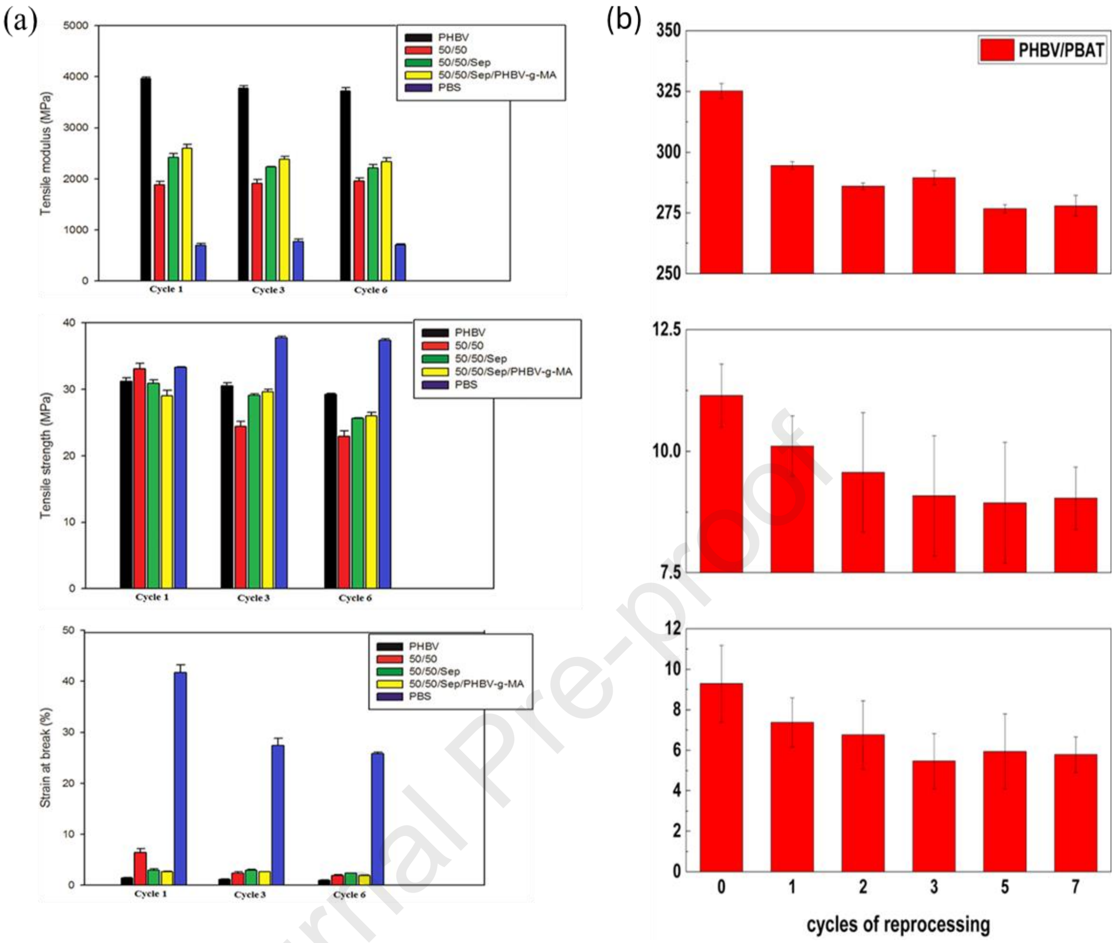

Fig 11. Young's modulus, tensile strength, and strain-at-break of (a)PBS, PHBV/PBS and (b)PHBV/PBAT blend after different reprocessing cycles[186, 187].

However, since (PHBV/PBAT) blend is polyester (formed through a polycondensation reaction), adding chain extenders to the blend can enhance the Mw [167, 187, 193]. In this study, DSC was used to calculate the thermophysical properties. The reprocessing raised the total melting range's temperature (by up to 6 K), regardless of the cycle number. This suggests that repeated processing has increased the size of the crystal lamellae[187].

5.1.3. PLA/PBAT blends

In a study by La Mantia *et al*. [31], the mechanical properties of (PBAT/PLA) blend were tested after being reprocessed for five successive extrusion cycles on samples reprocessed both dry and wet conditions through SSE. The results indicated that there was less material degradation when the sample was processed after drying as compared to when it was processed under wet conditions. Fig. 12 illustrates the two possible degradation mechanisms of PBAT. The carbon atom in the methyl group adjacent to the C=O group is susceptible to free radical attacks, resulting in a radical that can cause both branching and chain scission.

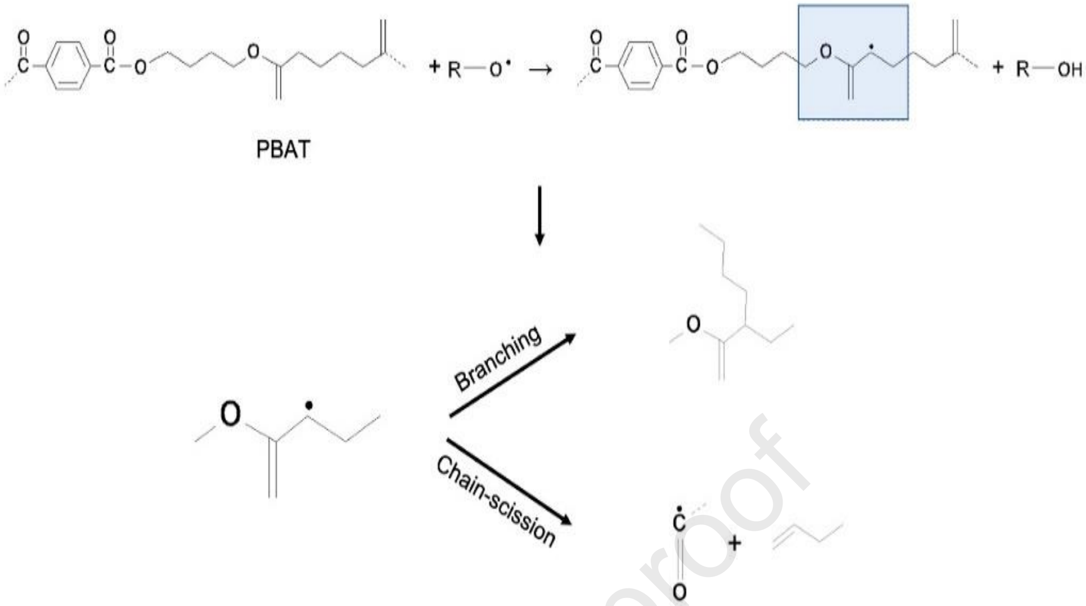

Fig 12. Proposed reaction mechanics[31].

Certainly, both chain scission and branching took place during the reprocessing cycle. However, in the blend reprocessed under wet conditions, chain scission was predominant. Conversely, for the dried sample, branching appeared to be more prevalent. Overall, it can be claimed that in both cases, five extrusion processes did not significantly reduce the mechanical properties of blend, proving that this biodegradable blend can withstand multiple reprocessing cycles. The experiments indicate that reprocessing of this blend would be beneficial, based on the evidence presented in all the results [31]. Thermogram measurements of the virgin sample and the five reprocessed blends under dry and wet circumstances verified the rise in crystallinity, which was responsible for the increase in elastic modulus. In another study Titone *et al.*[188] investigated whether the concentration of recycled biopolymer can significantly affect the mechanical characteristics of biodegradable blends. The Bio-Flex® F2110 (BF), a commercially available combination of PLA and PBAT, was reprocessed utilizing an SSE up to two times in this investigation. The results they acquired show that the elongation at break is notably affected by the number of extrusion cycles, while the impact on the elastic modulus and tensile strength is less significant. Specifically, the elongation at break for sample BF was 121 ± 21%, while BF_{RE1} and BF_{RE2} exhibited values of 93.9 ± 24% and 72.8 ± 18%, respectively. The tensile strength across all samples remained nearly unchanged. However, the elastic modulus of the virgin BF sample, initially 123 ± 10 MPa, increased to 140 ± 12 MPa for BF_{RE1} and 144 ± 10 MPa for BF_{RE2}. It is evident that the number of extrusion cycles significantly affects the elongation at break, while having a lesser impact on the elastic modulus and tensile strength. This outcome is anticipated, as the flow curve (Fig.13) data suggests that a reduction in Mw results in higher crystallinity, which in turn leads to an increase in the elastic modulus[194].

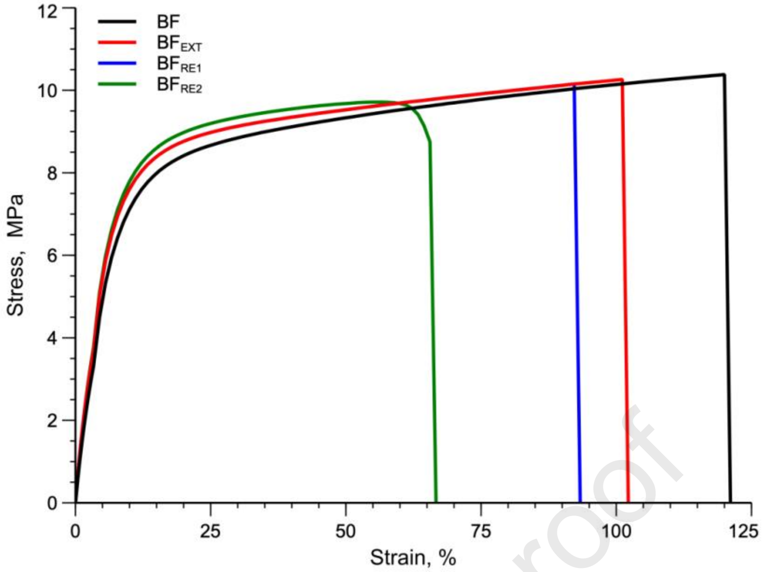

Fig 13. Comparison of stress–strain curves for virgin and extruded BF sample and samples reprocessed 1 and 2 times[188].

5.1.4. Other blends

In a study by Bourmaud *et al.*[189] the recyclability of a fully biodegradable L-Poly-(lactide)(PLLA)-PBS-flax biocomposite was investigated. Injection molding was followed by TSE to manufacture the biocomposites (Seven injection cycles). In this study, the results showed that the stiffness and strength at break after compounding fall and a degree of stability in the mechanical characteristics is evident, particularly in stiffness (Fig. 14). To explain it more clearly, regarding the PLLA-PBS matrix, the stiffness and strength at break after compounding are within the range of values previously reported [195]for the pure PLLA and PBS polymers, which are 2070 ± 83 MPa and 39.1 ± 1.8 MPa, respectively. Additionally, the mechanical properties demonstrate a quasi-stability, particularly in stiffness, with values ranging from 1989 ± 40 MPa to 2109 ± 39 MPa. This hypothesis is supported by earlier research[196], which demonstrated a significant reduction in the strength at break of PLLA after multiple processing cycles. With the inclusion of flax fibers, the results differ. The stiffness of the composite shows a consistent decline starting from cycle 5, with a reduction of 27.3% observed between cycles 4 and 7. This notable reduction can primarily be attributed to a slight decrease in the fiber's nanoindentation stiffness, as well as to the degradation of the matrix. Regarding the maximum strength, its value decreases starting from the first processing cycle, with a total reduction of 63.7% after seven injection cycles. A more noticeable decline occurs after the third cycle, further highlighting the significant degradation of PLLA at this stage, which is exacerbated by the presence of flax fibers. Between cycles 1 and 3, the decrease in composite strength and modulus is attributed to both matrix degradation (chain scission) and fiber deterioration (intrinsic modulus reduction and fiber breakage). However, from cycles 4 to 7, the composite's degradation is primarily driven by matrix chain scission, as the flax fibers exhibit minimal degradation during these later stages.

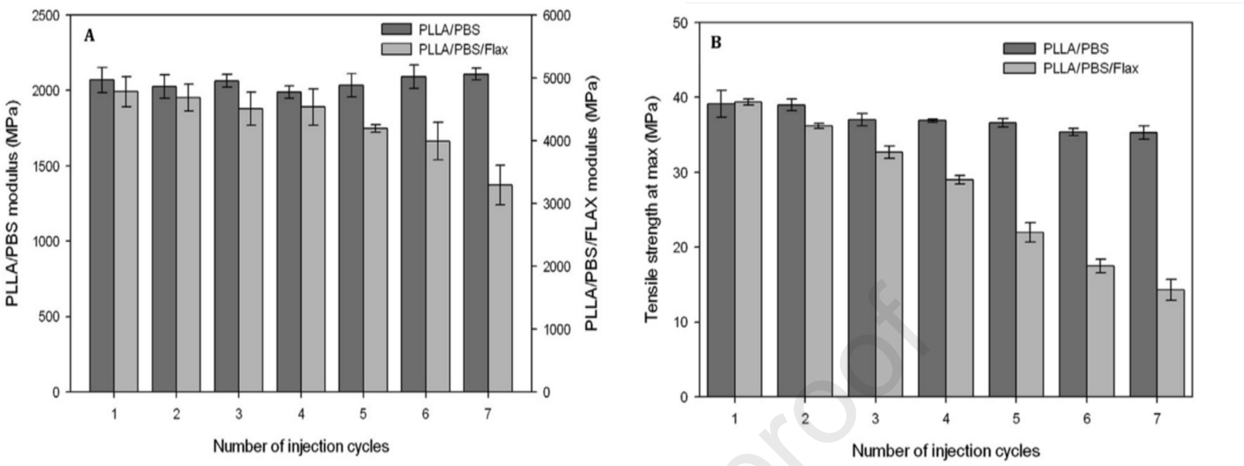

Fig14. Evaluation of the Young’s modulus (A) and strength at max(B) of PLLA-PBS matrix and PLLA-PBS-Flax composites[189].

Moreover, a comprehensive thermal analysis showcased the progression of the matrix, revealing significant PLLA degradation, particularly when flax reinforcements were included. In a study, Bavasso *et al*.[55] aimed to evaluate the potential for reusing waste material generated during the processing of a commercial biodegradable polymer blend (Estabio), widely used in manufacturing disposable products like cutlery and coffee capsules. The polymer underwent multiple extrusion cycles, repeated up to ten times. The results showed that the PLA/PBS blend’s mechanical properties remained mostly consistent throughout the extrusion cycles, although ductility decreased by 53%. Srimalanon *et al.* [170] evaluated the effect of reprocessing cycles on the mechanical properties of hygienic in-situ compatibilized (20/80 and 40/60) PBS/PLA blends doped with 2-hydroxypropyl-3-piperazinyl-quinoline carboxylic acid methacrylate (HPQM). The results revealed that the rupture performance of PBS/PLA blends was influenced by the number of processing cycles. Additionally, the toughness of the blends increased with a higher number of cycles. The third processing cycle yielded the highest elongation at break and tensile toughness, while the tensile modulus and strength remained largely unaffected by the number of cycles. Sirin *et al*.[190], examined how the number of thermal processing cycles for various blend compositions affected the mechanical characteristics of PLA/thermoplastic starch (TPS) blends. Using a TSE in the laboratory, PLA and TPS were compounded up to four times. Two parameters—(i) the number NOR and (ii) the amount of TPS in the blend were used to address changes in the PLA/TPS system’s characteristics. The results demonstrated that when the starch ratio in the blend system increased, the flexural modulus decreased. It is known that modulus of the blends generally obeys the rule of mixtures[197]. Due to TPS’ intrinsically lower modulus than PLA, the addition of TPS decreases the blend’s modulus regardless of NOR. Additionally, the flexural strength of the neat PLA decreased slightly when compared with its NOR counterpart; however, the NOR affected

PLA/TPS blends more strongly after the first recycle. In addition, when the NOR increased, impact strength tended to decline as a result of NOR [190]. Dadras Chomachayi *et al.* [191] investigated how reprocessing affected the properties of PLA/PCL blends reinforced with silk nanoparticles (NPs) and silk microfibers (SF). Polymer blends were prepared using a TSE. Due to the thermo-mechanical degradation of biodegradable, the impact strength of the PLA/PCL blend decreased significantly, by about 65%, from approximately 9 KJ/m^2 to 3 KJ/m^2 after three processing cycles. Additionally, it was reported that increasing the number of reprocessing cycles reduced the impact strength values of the recycled PLA [198]. For PLA/PCL/SF composites, adding 1 wt% of SFs did not result in any noticeable improvement. However, when 5 wt% of SFs was added, the impact strength of the composites slightly increased from about 3 KJ/m^2 to around 5.5 KJ/m^2 compared to the pure PLA/PCL blend, particularly after three reprocessing cycles. This activity could be brought on by the development of a network-like structure of silk fibers in the polymer matrix, which could help recyclable materials fracture more effectively by distributing energy [199, 200]. Compared to SF, the effect of silk NP is stronger. The obtained results demonstrated that the addition of NPs greatly boosts the impact strength value of the neat PLA/PCL blend. Due to a higher contact between the polymer and the NPs than the SF, the results can be attributed to improved transmission of applied stress from the matrix to NPs[201]. It is therefore possible to draw the conclusion that adding small amounts of silk fibroin NP can improve the stress transformation at the interface, thereby enhancing the impact strength of recycled materials[191]. Regarding thermal properties, obtained results indicated that when the number of reprocessing cycles rises, only modest variations in the PLA/PCL/NP nanocomposite's thermograms can be seen, indicating the stabilizing effects of NPs on the thermal properties of recycled materials[33, 202]. In another study by Nomadolo *et al.*[181] the effect of mechanical recycling on mechanical properties of (50/50)PBAT/PBS and (70/30) PBAT/TPS blends was evaluated. The multiple reprocessing cycles had a significant impact on both the tensile strength and strain-at-break, primarily due to the presence of PBS in the polymer blend. The strain-at-break decreased notably from 1013% in the first cycle to 63.9% in the seventh cycle. This decline can be attributed to the continuous reprocessing, which likely induced thermodynamic separation of the blend components. This separation led to the formation of voids and "weak spots" within the polymer matrix, ultimately causing a substantial reduction in the overall strain of the blend. For the PBAT-TPS blend, no significant changes in mechanical properties were observed across seven reprocessing cycles. Notably, the tensile strength of PBAT-TPS was similar to that of neat PBAT, while the strain-at-break showed a significant improvement compared to neat PBAT throughout the reprocessing cycles. These findings suggest that the melt blending process enhanced the compatibility between TPS and PBAT, positively influencing the mechanical properties of the blend. Regarding the impact test, the results demonstrated that the PBAT-TPS specimens exhibited no breakage throughout all reprocessing cycles. In contrast, the impact energy of the PLA, PBS, and PBAT-PBS specimens decreased as the number of reprocessing cycles increased

Table 5 displays the results from the mechanical properties of fully biodegradable-based polymer blends.

Table 5 Mechanical properties of fully biodegradable-based polymer blends[a]

Sample	Tensile Strength (MPa)	Tensile Modulus (MPa)	Elongation at break (%)	Cycles	Loading speed (mm/min)	Ref
PLA	-	-10.7%	-25%	6	1	[182]
PHBV		-11%	-30%			
PHBV/PLA (50/50)		-9.6%	-7.78%			
PLA/PHB	+2%	-	-95%	11	1	[183]
PLA/PHB	+6.45%	-	-32.33%	First=9 Second=9	50	[184]
PLA/PHB	-11.22%	-16.78%	-2%	5	2	[185]
PHBV/PBS	-33.33%	+3.4%	-80%	6	1	[186]
PHBV/PBS/sepiolite	-19.35%	-9.17%	-22.71%			
PHBV/PBS/sepiolite/ PHBV-g-MA	-11.11%	-10.74%	-47%			
PHBV/PBAT	-11%	-5.5%	-20%	7	1 (E) & 50 (TS and ε)	[187]
PLA/PBAT	-2 %	+2.85%	-22.5 %	2	1 (up to 3 min) & 100 (until failure)	[188]
PLLA/PBS	-10%	+2.6%	-82%		1	[189]
PLLA/PBS/Flax	-63.7%	-31.82%	-83%	7		
PLA/PBS	-3.9%	+8%	-	10	10	[55]
PBAT/PBS	-33.33%	-	-93%	7	N/A	[181]
PBAT/TPS	+4%	-	-	7	N/A	[181]

[a] Negative and positive signs indicate a decrease and an increase in the properties after the nth reprocessing cycle, respectively.

Summary

Recycling polymer blends is generally more challenging than recycling single polymers, and with each reprocessing cycle, a reduction in impact strength is often observed due to changes in the molecular chain weights. Material degradation and shortening of polymer chains can also lead to a decrease in relative elongation at break values. However, the PLA/PHB blend exhibits better resistance to degradation under multiple thermomechanical stresses compared to PLA alone. Partial degradation, reflected in the decreased molar mass of the polymer blend during processing, contributed to a reduction in flexural strength. This was counterbalanced by a progressive rise in crystallinity across processing cycles, which strengthened the intermolecular bonds. In notched samples that were recycled multiple times, the finer PHB domains helped mitigate brittleness caused by increased crystallinity and enhanced crack propagation resistance. Stronger intermolecular bonds in the blend may have compensated for the negative effects of recycling on strength by improving crystallinity. Nonetheless, the toughness of multiply processed samples was reduced due to poor chain entanglements arising from shorter molecular chains and a broader chain length distribution. The development of a network-like structure of silk fibers within the polymer matrix could aid fracture resistance by distributing energy more effectively. Moreover, adding small amounts of silk fibroin NPs can enhance stress transfer at the interface, thereby improving the impact strength of recycled materials. In the following, the mechanical properties of partially biodegradable-based polymer blends have been discussed. Table 6 lists the details of the processing techniques and NOR of partially biodegradable-based polymer blends.

Table 6. The information about processing techniques and NOR of partially biodegradable-based polymer blends

Blend	Processing Technique	Cycles	Ref
PLA/HDPE PLA/PC	TSE	6	[30]
PLA/PS	SSE+Injection	4	[56]
PP/PHB/Nanoclay/PP-g-MA/Erucamide	SSE	7	[203]
PLA/LCP	TSE/Injection	7	[204]
LDPE/TPS	SSE/Injection	5,10	[205]
PP/PBAT-TPS	SSE	7	[206]
TPS/EVA /PE-g-MA	TSE	3	[207]
BioPP/TPS+ PE-g-MA	TSE	5	[208]
Corn Starch-Curauá Fiber	Injection molding	10	[209]
PBS/PET	TSE+Injection	N/A	[210]

5.2. Partially biodegradable-based polymer blends

5.2.1. PLA-based blends

The durability of PLA and commercially available blends of PLA with polycarbonate(PC) and high-density polyethylene (HDPE) was assessed by Yarahmadi *et al.*[30] . The materials were extruded using a modular TSE and granulated afterwards (6 cycles). Multiple processing of the blends had no significant effect on the elastic modulus of the materials, although it did impact the elongation at break. However, while elongation at break rose from about 8.5 to 15 % with the number of extrusions for the PLA/HDPE blend, it decreased for the PLA/PC blend (from approximately 10 to 5%). DSC showed that PLA by itself does not crystallize after cooling, especially in procedures with limited orientation and rapid cooling rates, although it can be both thermally and stress-crystallized[211]. Blend components were not miscible, as evidenced by the DSC thermograms, which also demonstrated that PLA's Tg remained unchanged. According to the findings, PC and HDPE both functioned as nucleating agents, lowering the surface free energy barrier in the direction of nucleation and so starting the crystallization process. The only noticeable result of several extrusions is a drop in T_{cc}, which was less noticeable in the PLA/HDPE blend but more noticeable in the PLA/PC blend(approximately from 131 to 108 ̊C)[30]. In a study by Hamad *et al*. [56] the effect of multiple extrusion and injection(4 cycles) of PLA/polystyrene (PS) polymer blend on its mechanical properties was evaluated. The results showed that, after processing cycles, the stress and strain at break of PLA50 decreased. The stress at break of PLA50 dropped significantly after two processing cycles, reducing by 68%. After four processing cycles, the stress at break decreased by 79%. This can be explained by a decrease in Mw following the processing cycles, which results in less cohesiveness in the blend. Moving onto strain at break, the strain at break of the blend reduced by 61% after two processing cycles and by 73% after four processing cycles. These phenomena could be caused by both a drop in chain length and an increase in crystallinity, which both encourage crack propagation above the elastic domain. The decrease in stress and strain at break of PLA50 with processing number was more pronounced than the fall in Young's modulus; the similar phenomenon was observed in pure PLA recycling [196] where it was discovered that the processing number had no effect on tensile modulus despite lowering Mw, and this behavior was linked to an increase in crystallinity in PLA after processing cycles[56]. Viscosity curves showed that pure PLA's zero viscosity dropped, it is possible to deduce that PLA50 has better thermal stability than pure PLA. This difference may be related to PS's strong thermal stability [56, 212]. Another study found a similar impact associated to starch content [213]. de Kort *et al*. [204] investigated recyclability of blends based on PLA and thermotropic liquid crystalline polymers (LCP) . In this study, twin-screw injection molded was used (7 cycles). They found that the PLA was successfully reinforced by the LCP phase because the LCP content quickly increased the modulus and stress at break. However, as the Mw of the PLA reduced and the shape of the LCP changed, a noticeable loss in mechanical performance was noticed. The drop in the Young's modulus of both the PLA and PLA/LCP blends over the course of seven reprocessing steps clearly demonstrates this. The modulus of pure PLA steadily decreased with repeated reprocessing, albeit the overall change is minor (<10%). In contrast, the Young's modulus of the 30 wt. % LCP composites declined by 25% after seven reprocessing steps. A comparable pattern was noted regarding the maximum stress. When it comes to pure PLA samples, changes in Mw don't seem to impact modulus, suggesting that the decrease in mechanical properties post-reprocessing likely arises from impurities building up or additives degrading. The PLA/LCP blends exhibited a notable pattern: the Young's modulus remained steady in blends where the PLA matrix had a Mw of 170 kg mol^{-1} or above, while those with an Mw below 170 kg mol^{-1} displayed a decrease in modulus. Whether the composites were reprocessed or not, the relationship between the modulus and the Mw of the matrix stayed constant. Tensile stress measurements indicated the same tendency, with a constant value when the PLA matrix has a Mw of 170 kg mol^{-1} or above, and decreasing when the Mw of PLA is less than this value. This demonstrates that the mechanical performance of PLA/LCP composites, regardless of thermal history, is primarily determined by the Mw of the PLA phase under the specified processing conditions. However, mechanical performance deteriorates after numerous mechanical reprocessing processes, as the Mw of the PLA matrix gradually decreases [204].

5.2.2. Starch-based blends

In a study by Peres *et al.*[205] the effect of reprocessing on the structure and properties of (50/50) (LDPE/TPS) blends was evaluated. The mechanical properties of LDPE and LDPE after 5 cycles (5ext) and 10cycles (10ext) extrusion cycles were investigated (Fig.15).

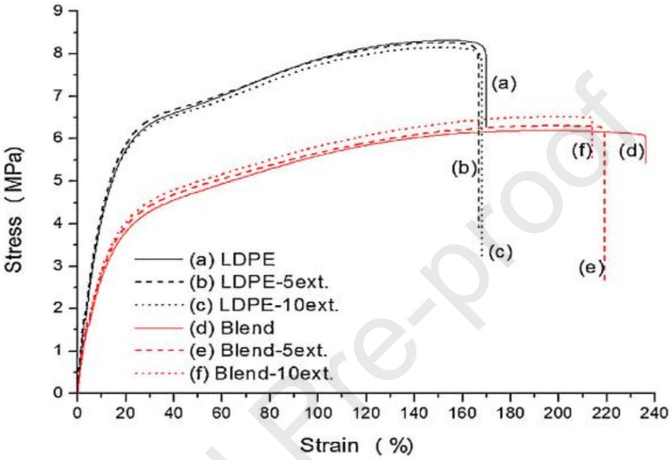

Fig 15. Stress versus strain curves for LDPE and LDPE/TPS blends after 5 (5ext) and 10 (10ext) extrusion cycles[205].

The elastic modulus, tensile strength, and elongation at break of LDPE did not change much as the number of extrusion cycles increased. As a result, the mechanical properties assessed for LDPE were not significantly impacted by at least 10 extrusion cycles, indicating that LDPE has a high level of reprocessability (i.e., little degradation in its mechanical properties as a function of the number of reprocessing cycles). The elastic modulus and mechanical strength of the (LDPE/TPS) blend increased somewhat after 10 extrusion cycles. These minor variations in mechanical properties caused by the increased number of extrusion cycles may be attributed to thermo-oxidative degradation, which can occur, particularly in TPS-rich phases. However, it should be noted that the refinement of the blends' microstructure (reduction in the average size of the TPS-rich phase) during the sequence of several extrusions may have contributed to the improvement of the properties and minimized any potential deterioration in the qualities. Additionally, the oxidation of LDPE chains during reprocessing may have enhanced compatibility between starch and oxidized LDPE chains, thereby contributing to the improved mechanical properties of the reprocessed LDPE/TPS blends. Thermal properties showed that the number of extrusion cycles and the addition of TPS to PE did not significantly alter the crystallization temperatures (T_c) or melting temperatures (T_m) for any of the samples. In another study, Oliveira *et al.* [206] evaluated effect of reprocessing cycles on the degradation of polypropylene (PP)/PBAT-TPS blends . The material underwent SSE processing a total of seven times. Mechanical parameters revealed that reprocessing enhanced the elasticity modulus and yield stress, resulting in a decrease in impact strength. Nevertheless, when the pure PP is reprocessed seven times in comparison to the first cycle, the elasticity modulus decreases by about 10%. The elasticity modulus behavior indicates that after 7 recycling cycles, the thermal degradation of the PP resulted in molecules that were too short to ensure strong linkages between the crystalline and amorphous phases[214]. Furthermore, the blend's elastic modulus decreases by around 33% compared to the pure matrix after the first processing cycle. This phenomenon is most likely due to the low rigidity of the PBAT-TPS agglomerates [215] and the weak interactions between the components of the blend [216]. The observations also reveal a 33.5% increase in the elasticity modulus of (PP/PBAT-TPS) blend by the seventh reprocessing cycle compared to the first. This outcome is most likely due to a decrease in (PBAT-TPS) domains and improved compatibility of the blend's components. According to Saw *et al*. [217], the existence of clusters with weak interfacial interaction hinders the alignment of molecular chains, which decreases the yield strength. In the case of the examined system, the (PBAT-TPS) fraction works as a non-strengthening agent, and the lack of component compatibility prevents applied stress from being passed across the interfaces. Moving on to yield stress, the yield stress of the (PP/PBAT-TPS) blend stayed almost unchanged during reprocessing, showing a slight increase only in the seventh recycling cycle due to better compatibilization of the (PBAT-TPS) domains with the PP matrix. Thermal properties indicated that there was a slight increase in T_m due to an increase in reprocessing cycles. TPS and ethylene vinyl acetate (EVA) blends, compatibilized using polyethylene-graft-maleic anhydride (PE-g-MA), were formulated from different native starches (potato, maize, and waxy maize) and processed through several extrusion cycles by Hejna *et al*.[207]. The origin of the starch has a considerable impact on its composition, and thus the qualities of the produced TPS and their blends with EVA. In this study, three forms of starch were used: potato starch (SZ), maize starch (SK), and waxy maize starch (SKW). The results demonstrated that repeated processing cycles improved the stiffness and tensile strength of all investigated composites. This impact could be attributed to better material mixing. The reaction between maleic anhydride grafted to PE compatibilizer and hydroxyl groups present in TPS structure may also influence compatibility of the material, as well as its mechanical performance [218]. In accordance with other authors' results, tensile strength increased with increasing amylopectin content [219]. As a result of a high amount of amylose in potato starch, the highest values of elongation at break were observed. As a result, it has fewer branched structures, which limit the mobility of polymer chains. Materials with SZ have the most ductile behavior, whereas materials with SKW exhibit brittle break regions. Moreover, DSC results indicated that after composite preparation, EVA crystallinity increased slightly, which was evidenced by the increase in ΔH_m and shifting the peak location of T_m towards higher temperatures[220]. In contrast, when PE-g-MA was mixed with another compound, the crystallinity was disrupted. This may be connected to the interactions that take place between grafted maleic anhydride and the hydroxyl groups found in the structure of TPS [221]. Multiple processing of the PE phase in composites resulted in a modest decrease in its total degree of crystallinity, which is consistent with findings reported in previous research studies [222]. Tavanaie *et al*. [208] studied the melt recycling of bio-based (PP/TPS) compound. In this study, a commercially available granule compound consisting of 50% polypropylene (PP) and 50% thermoplastic starch (TPS) was extruded as many as five times. Furthermore, the once-extruded (SE) PP/TPS granule was blended with virgin (PP/TPS) material in various ratios. The findings indicated that the strength, elongation-at-break, and modulus of the samples remained almost unchanged and comparable to one another, even after being recycled up to four times. However, a significant fall in strength and modulus is found in the fifth stage, indicating the effect of extrusion operation on tensile parameters after the fifth step of extrusion. This implies that the virgin granule can undergo extrusion up to four times without a reduction in tensile strength. Previous studies proved the recyclability of the (PP/TPS compound, and it was expected that no detrimental changes would be noticed in the tensile properties results. The modulus, strength, and strain of the blend samples show very little variation, and the results are extremely similar [208]. Obtained results related to heat treatment indicated that there is no significant difference in the degradation rate of TPS after being heated up to five times. In another study by Lenz *et al*. [209] the influence of reprocessing cycles on the hardness, impact, and tensile properties of corn starch-based biocomposites reinforced with curauá fiber was investigated. In this research, biocomposites were produced using injection molding with a biodegradable polymer matrix made from corn starch and vegetable curauá fiber. These composites were reprocessed up to ten times, both with and without the addition of maleic anhydride grafted polypropylene (MAPP) coupling agents. The addition of the MAPP coupling agent enhanced all properties, especially tensile strength, which remained relatively stable despite repeated reprocessing cycles. Further, results showed an increase in tensile modulus with an increase in reprocessing cycle with the addition of curauá fiber and MAPP coupling agent. Both matrices, with and without MAPP, experienced an increase in tensile modulus after curauá fiber addition. Curauá fiber addition, which enhances stiffness in the biocomposite, lowered elongation at break as expected, with essentially no change detected up to the 10th processing cycle [209]. The findings from TGA analysis revealed that the peak observed around 350°C in the thermograms of the biocomposites is attributed to the cellulose breakdown of the curauá fiber. Additionally, the second peak at approximately 470°C is indicative of the degradation of the matrix material. Up until the tenth processing cycle, no discernible change was seen with the increase in processing cycles[209].

5.2.3. PHB-based blends

Oliveira *et al*.[203] evaluated the effect of reprocessing cycles on mechanical properties of a PP/PHB blend and its nanocomposite. The systems investigated were a) neat PP, b) a PP/PHB blend and c) PP/PHB/organoclay nanocomposite compatibilized with polypropylene-g-maleic anhydride (PP-g-MA) and erucamide. These substances underwent a maximum of seven extrusion cycles at a speed of 60 rpm (SSE). The organoclay acts as the strengthening element in this study, and the interaction between the filler and matrix in the nanocomposite significantly impacts the characteristics of their interface [223]. Overall, the tensile properties of all systems diminished with reprocessing, with the most significant reductions seen in the PP/PHB blend after seven cycles, showing decreases of 50% in stress at break and 37% in elastic modulus. The impact strength of both the PP matrix and the PP/PHB blend generally declined with reprocessing. However, the nanocomposite slightly improved in impact strength, particularly after the seventh cycle, exhibiting an 18% increase.

5.2.4. PBS-based blends

The effect of low PBS content on recycled PET was investigated by Pavon *et al.*[210]. Fig. 16 illustrates the mechanical characterization results of the (PET/PBS) formulations.

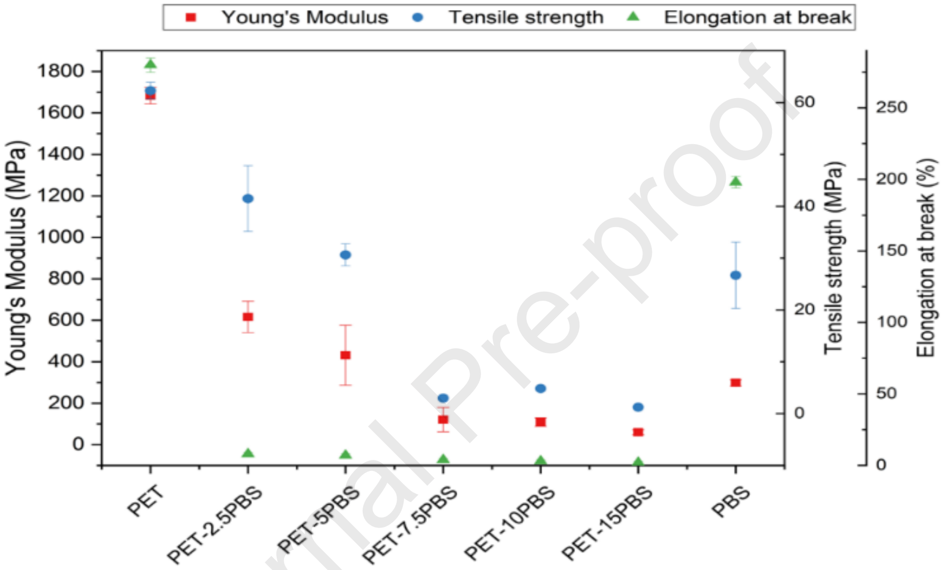

Fig 16. Scatter graphic Young's modulus, tensile strength, and shore D hardness of the PET-PBS formulations[210].

Neat PBS has a tensile strength of 27 MPa, Young's modulus of 299 MPa, and an elongation break of 198%, whereas recycled PET has tensile strength a 62 MPa and 1683 MPa of Young's modulus and 280 % of elongation break [210]. Regardless of the content, properties of PET are generally decreased when PBS is added. The PET tensile strength is reduced by 33 and 51%, respectively, and PET Youngs modulus by 63 and 74%, when PBS is present in the PET content of 2.5 and 5 wt.%. Tensile strength and Young's modulus of the formulations declined by 90% in relation to the value of recycled PET when the PBS level reaches 7.5 wt. % and higher. Additionally, with any PBS content, the PET elongation break decreased significantly. Even at the lowest investigated PBS level (2.5 wt.%), a noticeable decline in the tensile characteristics of recycled PET was found, indicating a lack of miscibility in the system. The findings demonstrate the incompatibility of two polymeric matrices and suggest that PBS contamination during PET recycling could have catastrophic impacts on the mechanical characteristics of the regenerated material. In addition, the PET impact strength is decreased by 37% and 47%, respectively, by adding 2.5 and 5 weight percent of PBS. The impact strength is reduced by 90% at a PBS concentration of 7.5 wt. % and higher. As a result of PBS contamination throughout all contents, the impact strength data demonstrate a reduction in the cohesiveness of the PET structure [210]. Moreover, TGA indicated no significant alteration both before and after the samples were exposed to the composting process, during the thermal deterioration circumstances. According to the degradation and TGA studies, PBS impurities have no effect on PET's heat stability or degradation when it is composted. The information about processing techniques and NOR of partially biodegradable-based polymer blends are indicated in Table 6. Moreover, the obtained results from mechanical properties of partially biodegradable -based polymer blends are shown in Table 7.

Table 7. Mechanical properties of partially biodegradable-based polymer blends [a]

Sample	**Tensile Strength (MPa)**	**Tensile Modulus (MPa)**	**Elongation at break (%)**	**Cycles**	**Loading speed (mm/min)**	**Ref**
PLA/PC	-	+0.87%	-50%	6	0.5 [b]	[30]
PLA/HDPE		-1.43%	+76.47%			
Postconsumer Recycling of PLA/PC		-0.44%	-16.44%	2		
PLA/PS	-78%	-0.27%	-70%	4	50.0	[56]
PP	-5.5%	-4.25%	-	7	50.0	[203]
PP/PHB	-50%	-37%				
PP/PHB/Nanoclay/PP-gMA/Erucamide	+4%	-6.25%				
LDPE	-3.57%	+5%	-1.18%	10	50.0	[205]
LDPE/TPS	+4.84%	+10.42%	-9.32%			
Pure PP	-	-10%	-	7	50.0	[206]
PP/PBAT-TPS		+33.5%				
TPS/EVA (SZ)	+7.41%	+55.74%	-10%	3	1(E)& 50 (TS and ε)	[207]
TPS/EVA (SK)	+51.1%	+169.27%	-44.17%			
TPS/EVA (SKW1)	+46.43%	+54.27%	-62.12%			
PP/TPS	-34%	-28.1%	-20%	5	50	[208]
Matrix	+4.4%	-44%	+8.9%	10	1-500	[209]
Matrix/MAPP	+5.2%	-19.04%	+3%			
Biocomposites	-0.01%	-2.41%	-21%			
Biocomposites/MAPP	+13.89%	-22.51%	+82%			

[a] Negative and positive signs indicate a decrease and an increase in the properties after the nth reprocessing cycle, respectively.

[b] The cross-head speed was initially set at 0.5 mm/ min (for E-modulus evaluation) up to an elongation of 0.5%, and then 50 mm/min until the specimen broke.

Summary:

The elongation at break of polymer blends is influenced by two main factors: polymer degradation, leading to chain scission and reduced elongation, and the dispersion of the phases, which can enhance elongation by reducing domain sizes and interfacial tension. Combining PLA with materials like HDPE or PC complicates the reproducibility of desired qualities. The mechanical performance of PLA/LCP composites is primarily determined by the molecular weight of the PLA phase, regardless of thermal history. Variations in mechanical properties after multiple extrusion cycles are often due to thermo-oxidative degradation. A reduction in the size of the TPS-rich phase during extrusion may improve the properties, while LDPE chain oxidation during reprocessing enhances the compatibility between starch and oxidized LDPE, improving mechanical performance in LDPE/TPS blends. Additionally, the rise in elasticity modulus is linked to smaller PBAT-TPS domains and improved component compatibility. However, incompatibility between components can hinder stress transfer across interfaces, and structural differences may also impact material compatibility. Furthermore, recycled PET exhibits a decline in tensile properties, suggesting a lack of miscibility in the system.

5.3. Comparison between fully biodegradable blends with partially biodegradable blends

The mechanical properties of several fully biodegradable polymer blends were evaluated and compared to those of partially biodegradable blends. For instance, in the partially biodegradable PLA/PC blends, after six cycles of testing, a 0.87% increase in tensile modulus and a 50% decrease in elongation at break were observed [30]. In addition, in the partially biodegradable PLA/HDPE blends, a 1.43% decrease in tensile modulus and a 76.47% increase in elongation at break were noted after six cycles [30]. In contrast, for the fully biodegradable PHBV/PLA blends, both tensile modulus and elongation at break decreased after six cycles, by 9.6% and 7.78%**,** respectively[182]. Moreover, in the PLA/PC partially biodegradable blend, after two cycles, both tensile modulus and elongation at break declined by 0.44% and 16.44%, respectively[30]. On the other hand, in the PLA/PBAT fully biodegradable blend, a 2.85% increase in tensile modulus and a 22.5% decrease in elongation at break were observed[188]. Regarding PHB-based blends, in the PP/PHB partially biodegradable blends, after seven cycles, both tensile strength and tensile modulus decreased by 50% and 37%, respectively [203]. Similarly, in the PHBV/PBAT fully biodegradable blends, both tensile strength and tensile modulus decreased by 11% and 5.5%, respectively [187]. However, the decline in the PHBV/PBAT fully biodegradable blends was lower compared to that in the PP/PHB partially biodegradable blends. The comparison between partially biodegradable and fully biodegradable blends should be conducted under fair and consistent conditions. Key factors to consider include the composite ratio, the dominant phase (biodegradable or non-biodegradable), the use of compatibilizers, the number of processing cycles, and the influence of temperature.

6. Rheological properties

The deformation and flow of matter under stress are the focus of the discipline of physics known as rheology. Particularly, rheology focuses on the properties of matter that determine its behavior when subjected to mechanical forces [224]. Rheology has long been a semi-quantitative technique in the study and engineering of polymers. Two factors make the link between a polymer's structure and rheology of practical interest: First, rheological properties are easier to employ than analytical techniques like nuclear magnetic resonance and are very sensitive to specific aspects of structure. Second, when polymers are in the molten state, their rheological properties govern the flow behavior [224, 225]. In immiscible and partly miscible blends, processing, morphology, and property relationships are very important, and a given processing strategy can produce blends with high added value. Rheological techniques are effective instruments for explaining these interactions. The rheological characteristics of the components have a significant impact on the morphology of blends, and knowledge of these characteristics is necessary to comprehend the processing of blends and the changes that take place during processing. Conversely, rheological properties of blends are affected by their morphology [226]. It is generally true that polymer blends have complex rheological behavior. Rheological properties of immiscible blends depend on a variety of factors, including composition, properties, morphology, phase interactions, interfacial tension, and the history of strains during processing [226, 227]. Even though rheology only provides an indirect estimate of deterioration, it is nonetheless a useful tool because of the proportionate link between viscosity and molar mass [182]. In the following sections, the rheological properties of fully and partially biodegradable-based polymer blends will be discussed.

6.1. Fully biodegradable-based polymer blends

In the study by Sirin *et al*.[190], according to a general trend observed in the phase sizes shown on the SEM, the particle size of the TPS phase increased with higher NOR levels, irrespective of the PLA/TPS ratio(the PLA/TPS ratio was varied as 100/0, 80/20, 70/30, and 60/40 by weight). Two causes can be identified for this: (i) the segregation of the TPS phase during repeated processing, and (ii) the change in the system's rheology brought on by the deterioration. In reality, during repeated processing of immiscible polymer blends, segregation will occur unless they are adequately stabilized with the use of a compatibilizer[228]. In this investigation, no compatibilizer was included in the (PLA/TPS) blends, therefore segregation during repeated processing was unavoidable. The potential for differentiation in the rheological behavior of the components following multiple processing stages is another factor. It has already been established that following repeated processing steps, PLA experiences chain scission and Mw loss brought on by thermo-oxidative degradation, which reduces melt viscosity [75, 229]. On the other hand, frequent processing of TPS may cause the plasticizers to evaporate, resulting in the final transformation of the soft TPS particles into stiff ones. Therefore, by assuming that all processing parameters remained constant, the PLA matrix was unable to provide the necessary shear forces to be applied to the rigidly segregated TPS particles needed for particle break-up, as the Mw reduction of PLA has already reduced its viscosity[190]. In another study, Zembouai *et al*. [182], evaluated the effects of recycling PHBV, PLA, and a (50/50)(PHBV/PLA) blend over six repeated processing cycles on rheological properties using dynamic shear measurements in their molten state. Similar to the previous mentioned study[190] , the complex viscosity of PLA samples exhibits a very minor drop with an increase in the number of cycles. The insignificant reduction in Mw caused by the chain scission phenomenon may help to explain this behavior. The effects of repeated processing on the intricate viscosity of PHBV demonstrated a significant decrease in the complicated viscosity of PHBV. It is possible to associate the significant drop in viscosity with more reprocessing cycles with a reduction in the Mw of the polymers [196, 230]. Multiple reprocessing cycles worsen the tendency of PHBV to experience chain scission during the extrusion process. Turning next to the (PHBV/PLA) blend, when comparing the complex viscosity of (PHBV/PLA) blends from the first extrusion to those from the sixth extrusion, the initial blends exhibit higher viscosity because the latter blends have undergone more reprocessing cycles. Regarding the development of the complex viscosity, it is clear to see the stabilizing influence of PLA on the PHBV, with a better stability of the (PHBV/PLA) blend after reprocessing than the PHBV alone. To explain it more clearly, during various reprocessing cycles, the deterioration appears to be more significant for the virgin PHBV than for the (PHBV/ PLA) blend [182]. In the study by Bourmaud *et al*. [189], melt rheological behavior of fully biodegradable (PLLA/PBS/flax) biocomposite was evaluated. Due to the shear rate during processing, fibers shift and rotate with the polymer flow, altering their orientation and potentially impacting the properties of the composite [231]. Additionally, factors such as fiber length, stiffness, strength, volume percentage, and the type of fiber-matrix interactions may affect flow characteristics [232]. In this case, the orientation of the fibers and, primarily, their length in capillary rheology were the key variables that could affect molten composites. The behavior of the matrix could also affect the viscosity of the compound. Composites had a higher viscosity than a PLLA-PBS hybrid matrix, as would be predicted. In practice, when the polymer is in a molten state, the solid fibers remain dispersed within it, obstructing the polymer's flow and raising its overall viscosity. In comparison to a molten virgin matrix, the molten composite's increased viscosity is more noticeable in areas with low shear rates. The polymer chain entangled and the fibers were disoriented at a reduced shear rate, which was insufficient to maintain the system's mobility and resulted in a rise in viscosity [233]. In this study, no matter what kind of matrix or biocomposite was used, they noticed a clear reduction in apparent viscosity as a function of the NOR cycles. During the initial cycle, the composite exhibited a high viscosity, which was attributed to the presence of long fibers. These fibers hinder the mobility of the matrix's polymer chains due to their interactions. The viscosity of PBS-PLLA-flax dramatically reduced during recycling, demonstrating a high rate of breakdown of the PLLA-PBS matrix as well as a shortening of the fiber length (Figure.17b) [189].

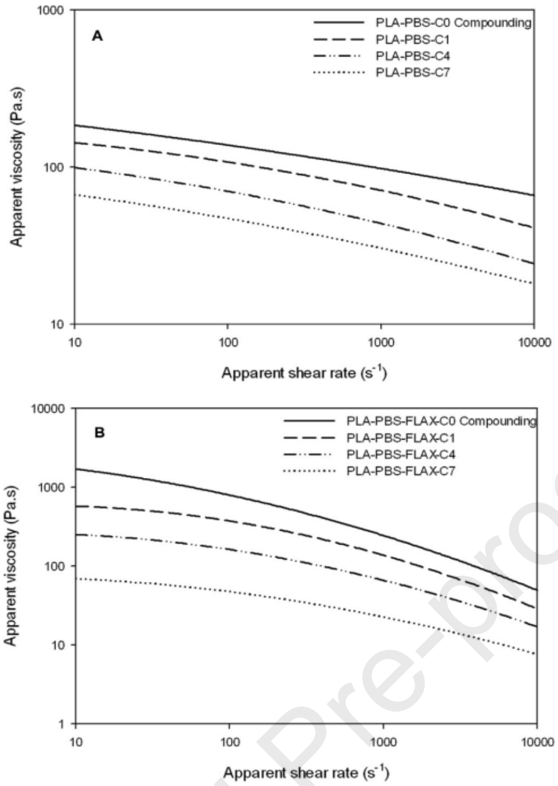

Fig 17. (A)Apparent shear viscosity versus apparent shear rate for the PLLA-PBS matrix (B) PLLA-PBS-flax composites [189].

In contrast, in the study by Bavasso *et al*. [55] a rheological analysis identified evidence of crosslinking, as reflected by an increase in complex viscosity at low frequencies after ten cycles. Similarly, in another study by Srimalanon1*et al.* [170] , it was observed that the storage modulus (G′) of the blend, increased after three processing cycles. This increase was attributed to the formation of more extensive branched or cross-linked networks during reprocessing. However, the storage modulus of the blends declined when the number of processing cycles exceeded three. This reduction was due to chain scission in PLA and PBS, along with the formation of shorter cross-linked structures after more than three cycles. In the study by Chikh *et al*. [186] by performing dynamic shear measurements in the molten state at 175 ̊C, which has been verified to be inside the linear viscoelastic domain, it was possible to examine the rheological behavior of neat PHBV, neat PBS, and their blend after 1, 3, and 6 reprocessing cycles. The complex viscosity consistently decreases as the number of cycles increases, according to the rheological curves of PBS samples (Fig. 18a).

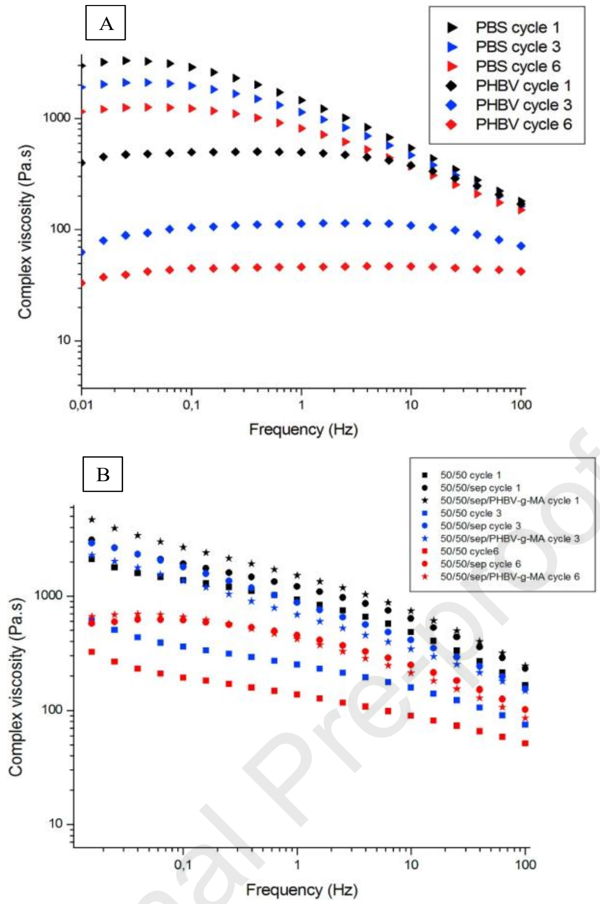

Fig 18. Evolution of the complex viscosity of neat PHBV , neat PBS, and PHBV/PBS blends after 1st, 3rd and 6th reprocessing cycle at 175 ̊C under 0.5% of dynamical shear strain[186].

The complex viscosity values at low frequency, or 0.01 Hz, are 2980, 1910, and 1150 Pa s after 1, 3, and 6 cycles, respectively. An insignificant reduction in Mw caused by the chain scission phenomena may be the cause of this behavior. In particular at low frequencies, PHBV exhibits a more marked reduction in complex viscosity. This reduction suggests that PHBV undergoes chain scission due to thermo-mechanical degradation during the extrusion process. By doing many reprocessing cycles, this effect is improved [182]. Moving onto the (PHBV/PBS) blends (Fig.18b) and their nanocomposites, the stabilizing impact of PBS on the PHBV is highlighted by the complicated viscosity of (PHBV/PBS) blend being higher than that of neat PHBV. When compared to virgin PHBV, the (PHBV/PBS) blend exhibited superior stability after several reprocessing cycles, which is likely a result of the co-continuous morphology's improved material elasticity. Although the recycling also triggered a thermo-mechanical degradation process brought on by a chain scission mechanism, the nanocomposite samples seem to be more stable to recycling than the blends. In fact, after 3 cycles of recording at 0.01 Hz, the complex viscosity of a (50/50) blend is 811 Pa s, while that of a (50/50/sepiolite) blend is 3150 Pa s. When sepiolite is paired with 5 wt. % of PHBV-g-MA compatibilizer, this behavior becomes much more pronounced. The existence of hydrogen bonds between the matrix and the filler may be the cause of the observed improvement in sample stability when both sepiolite and compatibilizer are present. Due to the filler's excellent dispersion state in blends, which is especially favored by the compatibilizer, the viscoelastic properties of the nanocomposite materials significantly improved in the presence of both sepiolite and compatibilizer, suggesting the formation of a 3D structure, i.e. interactions between polymer and polymer as well as polymer and nanofiller[186, 234]. The impact of five consecutive extrusion cycles during reprocessing on the rheological characteristics of the PBAT-based blend on samples treated in both dry and wet circumstances was evaluated by La Mantia *et al*.[31] . Both predrying and postdrying polymers were reprocessed. Because the presence of water also caused the components' hydrolysis in addition to their thermomechanical degradation, the rheological characterization revealed that when the sample was treated after drying, less degradation was seen. To explain it more clearly, as a result of reprocessing their samples after drying, they observed some branching in the PBAT majority component as compared to the chain rupture in the samples processed under wet conditions [31]. After the drying step, processing the sample led to a slight increase in viscosity, especially at lower frequencies, as the number of extrusion cycles rose. In contrast, when the polymer was processed under wet conditions, the sample's viscosity decreased with additional extrusions. Fig. 19 shows the flow curves of the virgin, PLA/PBAT blend, and reprocessed three and five times, are reported for the samples processed in dry and wet conditions.

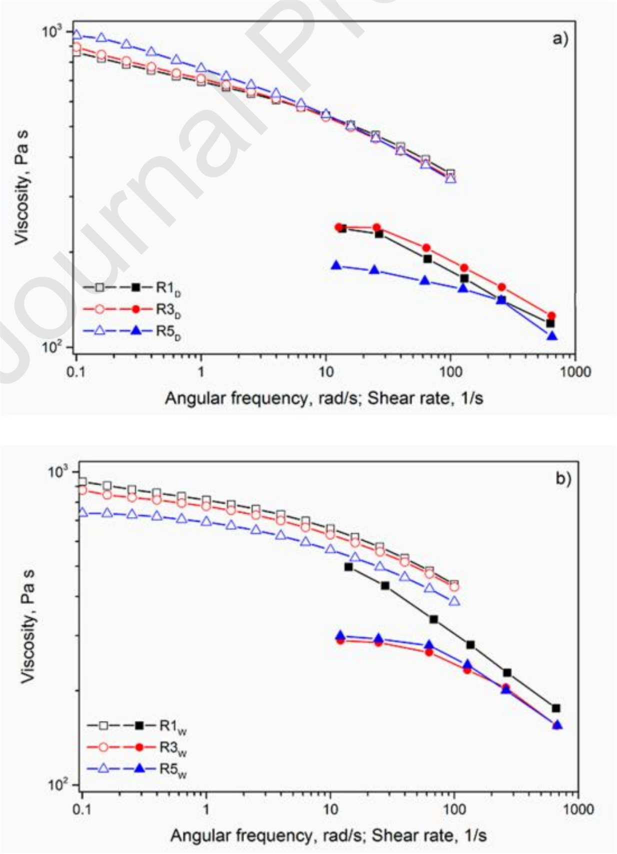

Fig 19. Flow curves of the PLA/PBAT blend after 1, 3, and 5 extrusions steps for (a) dried sample and (b) wet sample. Data taken from rotational rheometer (open symbols) and capillary viscometer (closed symbols)[31].

In another study by Plavec *et al.*[183], the effect of recycling on the viscosity of (45/55)(PLA/PHB) blends was investigated. Results showed that through multiple extrusions, the blended material was reduced in viscosity and partially degraded due to multiple thermomechanical stresses. Measurements of molecular characteristics demonstrated partial degradation. The color space/viscosity was used to determine the degree of degradation of the tested material. It can be concluded that, in contrast to the PLA or PHB polymers individually, the construction of a polymer blend of the PLA/PHB type had a highly favorable effect on the processing stability of biodegradable polymer blends in this situation. In the research [182], which shows how blending PLA and Poly(hydroxyalkanoates)(PHAs) improves processing stability, there was only a slight decrease in viscosity over the course of six processing runs when compared to the non-recycled blend [183]. In another study by Plavec *et al.* [184], the influence of multiple thermomechanical processing of 3D filaments based on (PLA/PHB) blend on their rheological properties was investigated. The viscosity graph illustrated how the complicated viscosity of the material drops as a result of the material's longer retention in the oscillating rheometer, which is a sign that the material is degrading under thermomechanical loading. Multiple processing of the polymeric material also results in a reduction in complex viscosity, and it may be inferred that the examined material deteriorates under thermomechanical stress in the laboratory. Preferably, the process of making filaments causes material degradation. During testing for processing stability, it was found that prolonged extrusion times in multiple processing cycles caused material degradation due to repetitive thermomechanical loading. This was demonstrated by a reduction in the complicated viscosity values of the tested material after several processes. In addition, in this study, experimental and industrial samples were analyzed to determine their complex viscosity. In industrial polymer blending, the material undergoes different levels of thermomechanical loading at various stages compared to the blending process of experimental polymer blends, which experience different shear stresses and retention times. As a result, blending under these conditions causes materials to degrade more rapidly. In the case of the industrial blend, the biggest decrease in viscosity occurred during the blending process. In rheological and morphological studies done by Farias *et al.*[185], (PLA/PHB) biopolymer blends showed some degree of immiscibility. PLA and PHB are particularly susceptible to viscous shear heating during the extrusion process, which can produce viscous shear heating and has detrimental effects on the polymer's viscosity. The evaluation of melt flow index (MFI), which is inversely related to viscosity, is typically used to assess the reduction in viscosity (Fig.20a). Multiple recycling's impact on the MFI of a (70/30) (PLA/PHB) blend measured at 190°C. The MFI grew with each recycling cycle, eventually reaching 33 g/10 min for a sample that had been recycled five times. Although MFI is a commonly used technique in industrial processing, it is not considered an appropriate tool for determining processing windows. This is because it provides only a single point measurement and fails to account for the shear rate and its duration within the viscosity assessment. These problems can be solved by utilizing dynamic rheology. By increasing the number of processing cycles for the various recycled samples, the complex viscosity was consistently reduced (Fig.20b).

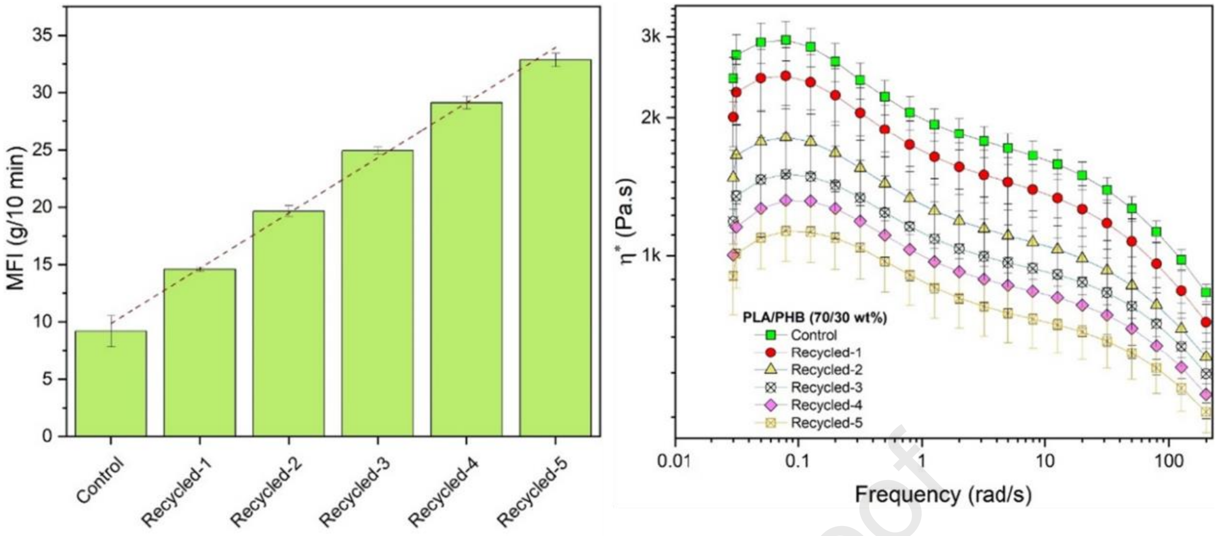

Fig 20. (a)The effect of multiple recycling on the MFI of PLA/ PHB (70/30 wt%) blend, and(b) Complex viscosity of PLA/ PHB blend measured through dynamic viscosity at 190℃ (error bars are standard deviation)[185].

The degradation of the blend's polymers, specifically PHB, is responsible for the (PLA/PHB) viscosity loss at low frequencies. It is commonly known that PHAs have a small processing window, which has restricted their commercial applicability [235]. The dynamic rheology test was conducted with a frequency sweep from higher to lower frequencies, and this circumstance could cause the blend’s residence time in the rheometer to surpass the thermal stability of the polymer at the lower frequencies, leading to degradation. Titone *et al*. [188] determined the effect of small amounts of reprocessed polymer on the rheological properties (shear and elongation). Results indicated that as the number of reprocessing stages grew, the viscosity of the reprocessed PLA/PBAT blends dropped over the entire examined frequency or shear rate. The findings indicated that the Mw of BF samples declines during reprocessing due to the thermomechanical stress applied to the melt. [188]. Fig. 21 presents the flow curves obtained from rheological tests using a rotational rheometer (showing complex viscosity, η* versus angular frequency) and a capillary viscometer (displaying shear viscosity, η versus shear rate) for both virgin samples and samples reprocessed one and two times.

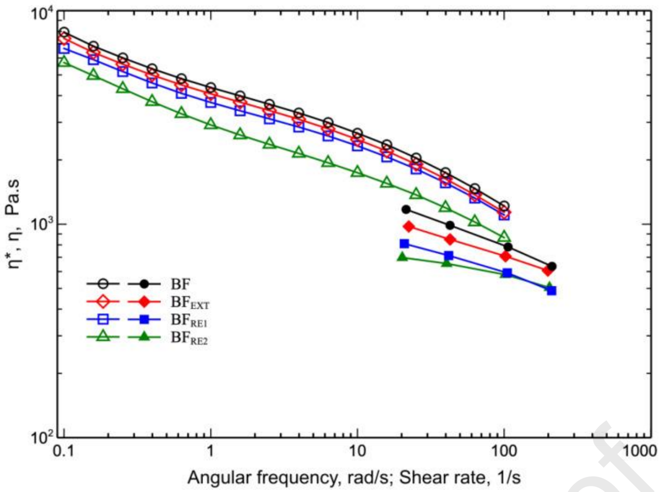

Fig 21. Flow curves of virgin and extruded BF sample, and samples reprocessed 1 and 2 times. Data taken from rotational rheometer (open symbols) and capillary viscometer (closed symbols)[188].

In another study by Dadras Chomachayi *et al*. [191], the repeated extrusion technique was applied to the PLA/PCL-based composites, and their inherent viscosity was studied and compared. The intrinsic viscosity (IV) values of samples in relation to the quantity of reprocessing cycles were supplied. The results show that mechanical recycling of the (PLA/PCL) blend decreased IV values, which is due to the thermo-mechanical breakdown of polymers that takes place during recycling. Following this, the effect of silk fibroin NPs and microfibers on the rheological properties of reprocessed (PLA/PCL) blends was evaluated. In comparison to the neat (PLA/PCL) blend, the remarkable lowering of IV values of recycled (PLA/PCL/NP) nanocomposite was significantly restricted by the inclusion of silk nanoparticles. This behavior supports the stabilizing effect of silk NPs can lessen polymer breakdown during reprocessing, perhaps as a result of blocking the terminal carboxyl groups of polyesters [191, 236]. In the case of (PLA/PCL/SF) composites, there is not a noticeable distinction between the IV values of pure PLA/PCL blends and those of (PLA/PCL/SF) composites in the first and second reprocessing cycles. The SEM and Fourier-transform infrared spectroscopy (FTIR) data revealed that the (PLA/PCL) blend and silk fibers had a relatively weak intermolecular connection, which could be the cause of this behavior. Overall, according to the results from this study it can be said that the thermo-mechanical degradation of biodegradable during mechanical recycling was slowed down by the presence of both NPs and SFs. The outcomes demonstrated that the reprocessed composites had greater IV values than the pure (PLA/PCL) blend. The above-mentioned qualities significantly improved when using NPs as opposed to SFs, which was attributed to NPs' superior intermolecular interaction with biodegradable [191].

6.2. Partially biodegradable-based polymer blends

The impact of numerous(50/50) (PLA/PS) extrusions and injections on its rheological properties was discussed by Hamad *et al*.[56] . Results showed that as the number of processing steps was increased, the apparent viscosity gradually decreased. These findings were explained by the fact that the Mw decreased throughout the processing cycles. Overall, the rheological findings demonstrated that the blend's apparent viscosity reduced with increasing processing number, which was explained by the drop in M_w because of the processing cycles. In addition, it was discovered that after the recycling procedure, the blend's flow behavior becomes more susceptible to temperature and shear rate[56]. In other study by Peres *et al*.[205], LDPE and the(50/50) (LDPE/TPS) blend were evaluated at 150°C for their viscosities depending on the extrusion cycle number(Fig.22).

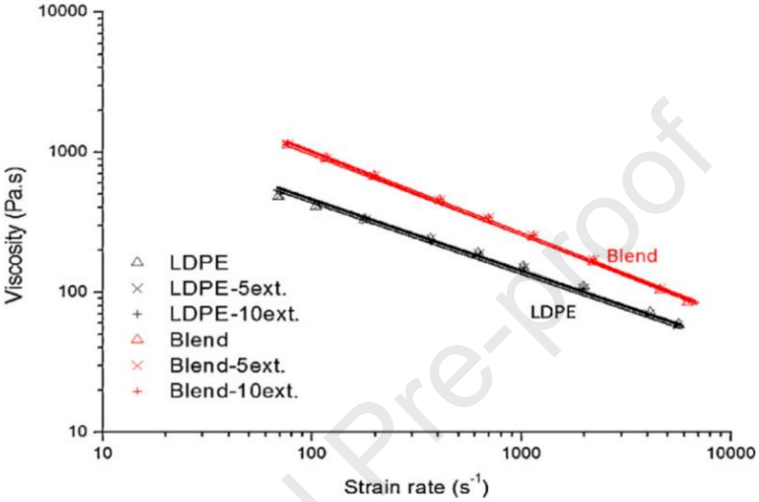

Fig. 22. Viscosity as a function of shear rate for LDPE and the LDPE/TPS blends measured by capillary rheometry at 150 °C [205].

As a function of the number of extrusions, neither the standard LDPE samples nor the reprocessed samples showed a significant change in viscosity. The (LDPE/TPS) blend also showed similar results when subjected to multiple extrusion cycles, i.e., no major changes in viscosity were observed following the reprocessing. Based on these results, any thermo-oxidative reactions that might have taken place during the 5–10 extrusion cycles for LDPE and (LDPE/TPS) did not result in significant chain damage. The addition of TPS to LDPE reduced the flow of the LDPE chains by restricting the flow of the LDPE chains when compared with LDPE alone [205]. In a study by Melcova *et al*. [237], the in-situ processing of two biopolyesters, PHB and PLA, with five reactive agents was investigated while a melt torque was simultaneously recorded. The melt viscosity of both was increased only by Raschig, an additive based on oligomeric carbodiimides. To put it more simply, Raschig was utilized in the subsequent trials to stabilize a (70/30) (PHB/PLA) blend. A maximum increase in melt torque during kneading in comparison to the non-stabilized PHB/PLA blend was reached with the addition of 0.5 wt. % of Raschig. As a result of Raschig addition, the Mw of the mixes was affected in a more complicated manner. Unreacted Raschig residue caused the average Mw to drastically decrease and polydispersity to significantly increase over 0.5 wt. %. Using melt volume-flow measurements, the processing stability of a PHB/PLA blend through six different extrusion cycles was established. When compared to the non-stabilized sample after the first extrusion cycle, the melt volume-flow index (MVI) of Raschig blend was lower after six cycles. This demonstrates that Raschig can effectively stabilize a PHB/PLA blend throughout processing[237]. In the study by Tavanaie *et al*. [208], they tested the effect of shear on the viscosity of (PP/TPS) compounds that were recycled through extrusion up to five times(Fig. 23a). They also examined virgin (PP/starch) samples blended with up to 50% of single-extruded samples by weight. Results for five times recycled compounds containing PP/TPS showed two important results (Fig.23b).

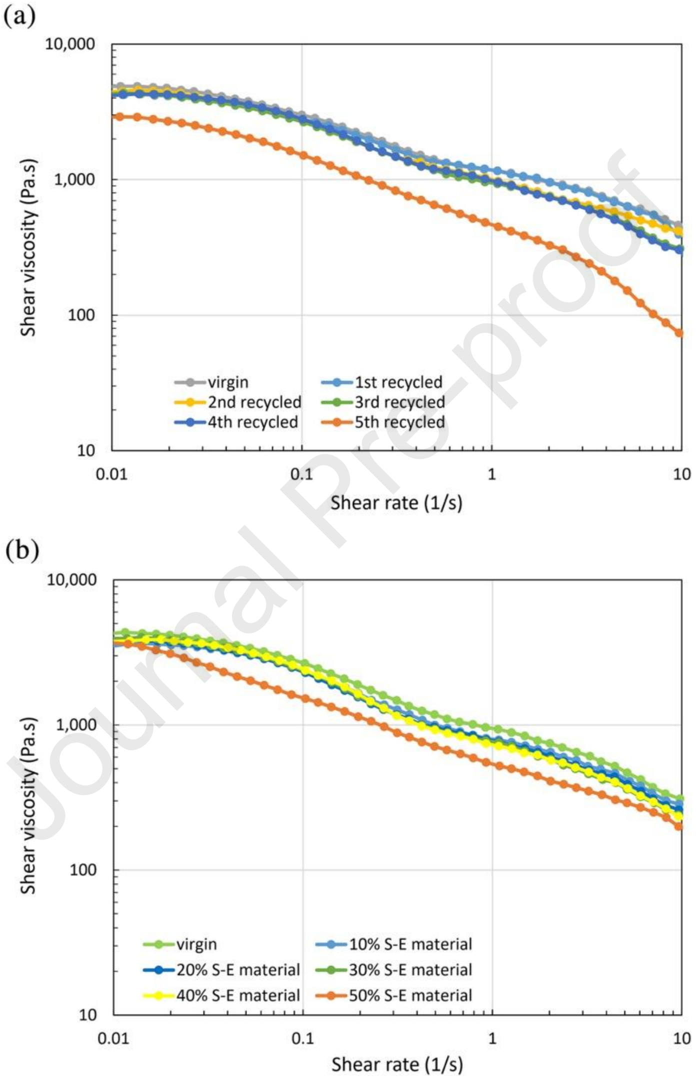

Fig 23. Viscosity changes curves with changing the shear rate for: (a) recycled samples up to five times in the extrusion process; and (b) virgin polypropylene (PP)/thermoplastic starch (TPS) blend mixed with different percentages of single-extruded samples up to 50% by weight[208].

First, as the number of times the compound was recycled increased, neither viscosity nor shear stress increased, and all the curves were almost identical. Second, the trend of changes in viscosity and shear stress after recycling the extruded polymer five times was nearly comparable, showing that the material's rheological properties have a high potential for recycling. However, it was only after the fifth time that the first signs of change become apparent, whether they were quantitative or in the form of a trend (curve shape). The heat hydrolysis of materials, particularly PP and maybe TPS, during the thermal recycling process was the cause of this large difference for the fifth recycled sample, which may result in structural unevenness of this sample. As a result, the outcome of analyzing the rheological properties supported the prior findings and showed no changes up to four times when employing this type of PP compound based on TPS. Moreover, the results related to the mixture of virgin (PP/TPS) blend with single extruded (PP/TPS) blend, indicated two important outcomes. First, when shear rate increased, the trend of increases in viscosity and shear stress remained constant and as the virgin sample and the single-extruded sample of the samples were mixed further, all of the curves were almost identical. Second, viscosity variations and shear stress followed a constant trend for single-extruded PP/TPS compounds up to 50% of the total percentage composition and this demonstrates that the compounds containing single-extruded material up to 50% do not show a significant alteration in their rheological characteristics. Furthermore, the composition of (50/50) virgin and single-extruded material just slightly changed, which was a very tiny variation[208].

Summary:

1. Thermal degradation during multiple processing cycles, leading to a reduction in Mw and subsequently affecting rheological properties.
2. In immiscible blends, morphologies can undergo multiple changes during re-processing, involving numerous droplet coalescence-breakup processes. As a result, there is a possibility of significant morphological changes after each cycle. Since these morphologies directly impact final properties, it becomes essential to stabilize them. That’s where the importance of compatibilizers is highlighted.

Rheological analyses serve a dual purpose. They not only allow us to replicate processing conditions when similar shear rates are observed, aiding in understanding how blends will flow at specific cycles for improved manufacturing processes, but they also enable the analysis of blend structures. Consequently, rheological analysis can help identify degradation and morphology changes more sensitively than other solid-state analytical tools. In a critical review article writing we need to critique the existing literature and suggest ideas. For example, we could say, the studies explored the effect of recycling, solely used rheological tools to analyse the stability of the blends after each cycle. It would be also beneficial to correlate the flow properties resembling the practical processing conditions in favour of best optimised re-processing conditions. For instance, if viscosities drop after a few cycles, maybe one approach to minimize the losses is to continue the actual processing conditions at lower shear rates and temperatures to avoid fast degradation. Alternatively, it is advisable to establish a correlation between rheological properties and morphology changes. This involves assessing the extent of droplet size variation after each cycle and determining whether droplet breakup or coalescence dominates the morphological changes during processing and up to which cycle each phenomenon prevails.

7. The effects of compatibilizers on recycling

According to the results from several studies it can be said materials tends to degrade throughout the various stages of mechanical recycling. This degradation lowers the polymer's average Mw, which negatively impacts the performance of the recycled material and decreases its intrinsic viscosity which is an essential factor in polymer processing [238]. Moreover, repeated recycling significantly reduced the impact strength. The material properties alter due to changes in crystallinity, which resulted from a decrease in Mw with each reprocessing. Consequently, the recycled materials become stiffer, less flexible, and exhibited lower water absorption and impact resistance compared to the original material [239]. Based on the findings from multiple studies, it can be concluded that enhancing the mechanical recycling properties of materials made from plastic waste blends can be achieved by using appropriate compatibilizers. To explain it more clearly, with the addition of the chain extenders as compatibilizers, the polymers Mw increase, leading to a restoration of its properties. Results showed that there is a reduction in MFI and crystallinity, along with enhanced thermal stability and mechanical properties compared to recycled blends that lacked the chain extender[239]. Moreover, compatibilizers improve the interfacial adhesion between the macromolecules of the polymers in the blend[240]. Overall, in polymer blends, different polymers often do not mix well due to differences in their chemical structures, leading to phase separation. Compatibilizers reduce interfacial tension between the phases, improving their adhesion and creating a more homogeneous blend. This enhanced compatibility ensures that recycled materials maintain acceptable mechanical and thermal properties after multiple processing cycles. By improving the dispersion of the phases, compatibilizers prevent defects like cracks or voids from forming at the interface of the polymers. This results in a stronger, more durable blend, which can withstand the stresses of repeated recycling without significant degradation in properties such as toughness or flexibility. Compatibilizers help stabilize the morphology of polymer blends during repeated processing, ensuring that the structure of the recycled material remains consistent with the original. This helps maintain product quality even after several cycles of recycling. [11, 63, 241, 242].

8. Conclusion and Future marks

The increasing demand for sustainable materials has emphasized the need for effective recycling strategies for biodegradable-based polymer blends. This review has highlighted the significant potential of blending biodegradable polymers, such as PLA and PBAT, to enhance the performance characteristics required for various applications. However, the inherent differences in properties and miscibility of these polymers often pose challenges for recyclability and mechanical performance. Using compatibilizers has emerged as a pivotal strategy to address these challenges, improving interfacial adhesion and enhancing the overall recyclability of polymer blends. By optimizing compatibilizer types and blending ratios, researchers have demonstrated the possibility of achieving better mechanical and thermal properties while maintaining biodegradability. Moreover, recycling these improved blends not only contributes to a better circular economy but also reduces plastic waste and environmental impact. Future research should focus on developing more efficient compatibilizers and exploring advanced recycling techniques to further improve the performance and sustainability of biodegradable-based polymer blends. Collaborative efforts between academia, industry, and policymakers will be essential to scale these advancements and ensure practical implementations in the plastics industry. In this way, blending and compatibilization strategies represent a promising path toward achieving a more sustainable and environmentally friendly approach to polymer recycling. In this review, we specifically concentrated on evaluating the effects of reprocessing on the mechanical and rheological properties of biodegradable-based blends, considering it as a key aspect of the recycling process. It is important to note that, mechanical recycling under real-world conditions involves multiple steps, including collection, separation, sorting, cleaning, drying, and grinding of waste materials. Moreover, polymers in real-world cases that are used in recycling process may have gone through an aging process due to exposure to various environmental conditions such as prolonged weathering, contaminated with food wastes, agricultural fertilizers and pharmaceutical products. Such conditions could lead to an extra degradation process prior to actual recycling processing. However, these conditions were not accounted for in the present studies. To better simulate real-world scenarios, future research should incorporate these steps [243].

Furthermore, there are various approaches to enhance the properties of biodegradable-based blends. For instance, using specific blending sequence procedures to evaluate the affinity of each polymer with different compatibilizers like chain extenders [244-247] can optimize the mechanical and rheological characteristics of the blends. Additionally, the incorporation of other bio-based compatibilizers, such as tannic acid, gallic acid, and epoxidized soybean oil into immiscible blends offers a promising strategy to improve compatibility and performance. Moreover, the antioxidant properties of tannic acid and gallic acid can mitigate oxidative degradation, enhancing the durability of the materials. As renewable, naturally derived additives, they contribute to sustainable material development, reducing dependency on petroleum-based solutions and aligning with the goals of green chemistry.

Vitrimers, a distinct class of polymers defined by their dynamic covalent bond networks, can also be harnessed as compatibilizers due to their unique adaptive properties. These bonds do not create permanent crosslinks, enabling recycling and reprocessing. Additionally, vitrimers can contribute to self-healing properties, enhancing the durability and lifespan of the blends [248, 249]. These compounds enhance the compatibility of immiscible polymers through their ability to form covalent and hydrogen bonds, leading to improved interfacial adhesion. Their use also results in better mechanical properties, such as increased tensile strength and toughness, while improving the thermal stability of the blends. These advancements hold significant potential for industrial applications, particularly in film food packaging and film casting, where enhanced mechanical, thermal, and barrier properties are crucial for performance and sustainability.

Furthermore, the fast-growing role of AI and machine learning in all technological aspects is undeniable, and this field is no exception. Machine learning can play a significant role in tailoring and optimizing material properties more efficiently and effectively by identifying patterns, predicting behavior, and guiding the design process, ultimately accelerating development and improving performance[250, 251]. In fact, AI and machine learning can revolutionize recycling polymer blends by improving material sorting and identification processes through advanced image recognition and spectroscopy analysis. They can also predict optimal recycling parameters, such as temperature and processing conditions, to maintain material properties during reprocessing. Furthermore, machine learning models can help design innovative compatibilizers and additives to enhance the recyclability and mechanical performance of blended polymer.

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References

[1] J. Hopewell, R. Dvorak, E. Kosior, Plastics recycling: challenges and opportunities, Philosophical Transactions of the Royal Society B: Biological Sciences 364(1526) (2009) 2115-2126.
[2] R. Hoffmann, D.D.S. Morais, C. Braz, K. Haag, R.M. Wellen, E. Canedo, L. De Carvalho, K. Koschek, Impact of the natural filler babassu on the processing and properties of PBAT/PHB films, Composites Part A: Applied Science and Manufacturing 124 (2019) 105472.
[3] L. Filiciotto, G. Rothenberg, Biodegradable plastics: Standards, policies, and impacts, ChemSusChem 14(1) (2021) 56-72.
[4] M.S. Kim, H. Chang, L. Zheng, Q. Yan, B.F. Pfleger, J. Klier, K. Nelson, E.L.-W. Majumder, G.W. Huber, A Review of Biodegradable Plastics: Chemistry, Applications, Properties, and Future Research Needs, Chemical Reviews (2023).
[5] P. Europe, Plastics – the fast Facts 2023, (https://plasticseurope.org/).
[6] K. Mosnáčková, M. Danko, A. Šišková, L.M. Falco, I. Janigová, Š. Chmela, Z. Vanovčanová, L. Omaníková, I. Chodák, J. Mosnáček, Complex study of the physical properties of a poly (lactic acid)/poly (3-hydroxybutyrate) blend and its carbon black composite during various outdoor and laboratory ageing conditions, RSC advances 7(74) (2017) 47132-47142.
[7] J.R. Jambeck, R. Geyer, C. Wilcox, T.R. Siegler, M. Perryman, A. Andrady, R. Narayan, K.L. Law, Plastic waste inputs from land into the ocean, Science 347(6223) (2015) 768-771.
[8] I. Hussain, A. Aitani, Z. Malaibari, H. Alasiri, M. Naseem Akhtar, O. Fahad Aldosari, S. Ahmed, Chemical Upcycling of Waste Plastics to High Value-Added Products via Pyrolysis: Current Trends, Future Perspectives, and Techno-Feasibility Analysis, The Chemical Record 23(4) (2023) e202200294.
[9] D.K. Barnes, F. Galgani, R.C. Thompson, M. Barlaz, Accumulation and fragmentation of plastic debris in global environments, Philosophical transactions of the royal society B: biological sciences 364(1526) (2009) 1985-1998.
[10] W.W. Lau, Y. Shiran, R.M. Bailey, E. Cook, M.R. Stuchtey, J. Koskella, C.A. Velis, L. Godfrey, J. Boucher, M.B. Murphy, Evaluating scenarios toward zero plastic pollution, Science 369(6510) (2020) 1455-1461.
[11] S.B. Borrelle, J. Ringma, K.L. Law, C.C. Monnahan, L. Lebreton, A. McGivern, E. Murphy, J. Jambeck, G.H. Leonard, M.A. Hilleary, Predicted growth in plastic waste exceeds efforts to mitigate plastic pollution, Science 369(6510) (2020) 1515-1518.
[12] P. Feijoo Domínguez, A. Mohanty, A. Rodriguez-Uribe, J. Gámez Pérez, L. Cabedo, Biodegradable blends from bacterial biopolyester PHBV and bio-based PBSA: Study of the effect of chain extender on the thermal, mechanical and morphological properties, (2022).

[13] P. Europe, Plastics the facts 2014/2015: an analysis of European plastics production, demand and waste data, Plastic Europe, Brussels (2015).
[14] S. Giri, G. Lamichhane, D. Khadka, H.P. Devkota, Microplastics contamination in food products: Occurrence, analytical techniques and potential impacts on human health, Current Research in Biotechnology (2024) 100190.
[15] R. Kumar, K. Sadeghi, J. Jang, J. Seo, Mechanical, chemical, and bio-recycling of biodegradable plastic: A review, Science of The Total Environment (2023) 163446.
[16] R.M. Albuquerque, H.M. Meira, I.D. Silva, C.J.G. Silva, F.C.G. Almeida, J.D. Amorim, G.M. Vinhas, A.F.S. Costa, L.A. Sarubbo, Production of a bacterial cellulose/poly (3-hydroxybutyrate) blend activated with clove essential oil for food packaging, Polymers and Polymer Composites 29(4) (2021) 259-270.
[17] R. Salehiyan, S. Soleymani Eil Bakhtiari, A review on rheological approaches as a perfect tool to monitor thermal degradation of biodegradable polymers, Korea-Australia Rheology Journal (2024) 1-23.
[18] J. Rydz, W. Sikorska, M. Kyulavska, D. Christova, Polyester-based (bio) degradable polymers as environmentally friendly materials for sustainable development, International journal of molecular sciences 16(1) (2014) 564-596.
[19] M.C. Lavagnolo, V. Poli, A.M. Zampini, V. Grossule, Biodegradability of bioplastics in different aquatic environments: A systematic review, Journal of Environmental Sciences 142 (2024) 169-181.
[20] K.M. Nampoothiri, N.R. Nair, R.P. John, An overview of the recent developments in polylactide (PLA) research, Bioresource technology 101(22) (2010) 8493-8501.
[21] R.M. Rasal, A.V. Janorkar, D.E. Hirt, Poly (lactic acid) modifications, Progress in polymer science 35(3) (2010) 338-356.
[22] Y. Zhong, P. Godwin, Y. Jin, H. Xiao, Biodegradable polymers and green-based antimicrobial packaging materials: A mini-review, Advanced Industrial and Engineering Polymer Research 3(1) (2020) 27-35.
[23] S. Pilla, Engineering applications of bioplastics and biocomposites-An overview, Handbook of bioplastics and biocomposites engineering applications (2011) 1-15.
[24] R. Muthuraj, M. Misra, A.K. Mohanty, Biodegradable compatibilized polymer blends for packaging applications: A literature review, Journal of Applied Polymer Science 135(24) (2018) 45726.
[25] J. Rydz, M. Musioł, B. Zawidlak-Węgrzyńska, W. Sikorska, Present and future of biodegradable polymers for food packaging applications, Biopolymers for food design (2018) 431-467.
[26] R. Salehiyan, S. Soleymani Eil Bakhtiari, Bioplastic Based Multifunctional Conductive Blend Nanocomposites, Reference Module in Materials Science and Materials Engineering, Elsevier2024.
[27] M.R. Havstad, Biodegradable plastics, Plastic waste and recycling, Elsevier2020, pp. 97-129.
[28] Are Compostable Bags As Environmentally Friendly As We Think They Are?, https://www.forbes.com/sites/noelfletcher/2025/01/21/the-epa-is-investing-24-million-to-bring-new-technologies-to-market/?
[29] D. Purkiss, A.L. Allison, F. Lorencatto, S. Michie, M. Miodownik, The Big Compost Experiment: Using citizen science to assess the impact and effectiveness of biodegradable and compostable plastics in UK home composting, Frontiers in Sustainability 3 (2022) 942724.

[30] N. Yarahmadi, I. Jakubowicz, J. Enebro, Polylactic acid and its blends with petroleum-based resins: Effects of reprocessing and recycling on properties, Journal of Applied Polymer Science 133(36) (2016).
[31] F.P. La Mantia, L. Botta, M.C. Mistretta, A. Di Fiore, V. Titone, Recycling of a biodegradable polymer blend, Polymers 12(10) (2020) 2297.
[32] K. Hamad, M. Kaseem, F. Deri, Recycling of waste from polymer materials: An overview of the recent works, Polymer degradation and stability 98(12) (2013) 2801-2812.
[33] J. Badia, A. Ribes-Greus, Mechanical recycling of polylactide, upgrading trends and combination of valorization techniques, European Polymer Journal 84 (2016) 22-39.
[34] L. Alaerts, M. Augustinus, K. Van Acker, Impact of Bio-Based Plastics on Current Recycling of Plastics. Sustainability 2018, 10, 1487.
[35] J.V. Azevedo, B. Hausnerova, B. Möginger, T. Sopik, Effect of Chain Extending Cross-Linkers on the Disintegration Behavior of Composted PBAT/PLA Blown Films, International Journal of Molecular Sciences 24(5) (2023) 4525.
[36] Y.Q. Gill, S.M.H. Mehdi, U. Mehmood, Enhancement of polylactide/poly (butylene adipate-co-terephthalate) blend [ECOVIO] properties by carboxylated nitrile butadiene rubber (XNBR) addition for smart packaging applications, Materials Letters 306 (2022) 130881.
[37] R.C. Rebelo, L.P. Gonçalves, A.C. Fonseca, J. Fonseca, M. Rola, J.F. Coelho, F. Rola, A.C. Serra, Increased degradation of PLA/PBAT blends with organic acids and derivatives in outdoor weathering and marine environment, Polymer 256 (2022) 125223.
[38] J.R. Rocca-Smith, O. Whyte, C.-H. Brachais, D. Champion, F. Piasente, E. Marcuzzo, A. Sensidoni, F. Debeaufort, T. Karbowiak, Beyond biodegradability of poly (lactic acid): physical and chemical stability in humid environments, ACS Sustainable Chemistry & Engineering 5(3) (2017) 2751-2762.
[39] Green Dot Bioplastics. Biodegradables. , 2023.
[40] M. Aldas, C. Pavon, J. Ferri, M. Arrieta, J. López-Martínez, Films Based on Mater-Bi® Compatibilized with Pine Resin Derivatives: Optical, Barrier, and Disintegration Properties. Polymers (Basel) 13: 1506, 2021.
[41] Plantic Technology, https://plantic.com.au/.
[42] BIOTEC, https://www.biotec.de/company/.
[43] Mitsubishi Chemical's FORZEAS™ (https://us.mitsubishi-chemical.com/).
[44] TIPA COMPOSTABLE PACKAGING, https://tipa-corp.com/bio_plastic_technology/.
[45] Terrafilum, https://www.terrafilum.com/.
[46] Fillamentum addi(c)tive polymers, https://fillamentum.com/collections/.
[47] Ataraxia Art https://ataraxiaart.com/?srsltid=AfmBOop17fKTWPtuOE4qise-9JuHSSzXmiMUfvnsAqSana9OjLDBQozm.
[48] TotalEnergies Corbion, https://www.totalenergies-corbion.com/.
[49] Agrobiobase, https://www.agrobiobase.com/.
[50] F. Kučera, J. Petruš, J. Jančář, The structure-hydrolysis relationship of poly (3-hydroxybutyrate), Polymer Testing 80 (2019) 106095.
[51] Danimer scientific company (Nodex®), https://danimerscientific.com/.
[52] Greenmax.eu/en/solution, https://greenmax.eu/en/solutions/.
[53] J.G. Ausejo, J. Rydz, M. Musioł, W. Sikorska, M. Sobota, J. Włodarczyk, G. Adamus, H. Janeczek, I. Kwiecień, A. Hercog, A comparative study of three-dimensional printing directions: The degradation and toxicological profile of a PLA/PHA blend, Polymer degradation and stability 152 (2018) 191-207.

[54] GREENER WALKER, https://www.greenerwalker.com/.
[55] I. Bavasso, M.P. Bracciale, G. De Bellis, A. Pantaleoni, J. Tirillo, G. Pastore, S. Gabrielli, F. Sarasini, Recycling of a commercial biodegradable polymer blend: Influence of reprocessing cycles on rheological and thermo-mechanical properties, Polymer Testing 134 (2024) 108418.
[56] K. Hamad, M. Kaseem, F. Deri, Effect of recycling on rheological and mechanical properties of poly (lactic acid)/polystyrene polymer blend, Journal of materials science 46 (2011) 3013-3019.
[57] H.W. Toh, D.W.Y. Toong, J.C.K. Ng, V. Ow, S. Lu, L.P. Tan, P.E.H. Wong, S. Venkatraman, Y. Huang, H.Y. Ang, Polymer blends and polymer composites for cardiovascular implants, European Polymer Journal 146 (2021) 110249.
[58] L. Yu, K. Dean, L. Li, Polymer blends and composites from renewable resources, Progress in polymer science 31(6) (2006) 576-602.
[59] D. Paul, Control of phase structure in polymer blends, Functional polymers, Springer1989, pp. 1-18.
[60] J. Parameswaranpillai, S. Thomas, Y. Grohens, Polymer blends: state of the art, new challenges, and opportunities, Characterization of polymer blends (2014) 1-6.
[61] C.-F. Yang, H.-C. Wang, C.-C. Su, Enhancing the Compatibility of Poly (1, 4-butylene adipate) and Phenoxy Resin in Blends, Materials 10(7) (2017) 692.
[62] S. Aparna, D. Purnima, R.B. Adusumalli, Review on various compatibilizers and its effect on mechanical properties of compatibilized nylon blends, Polymer-Plastics Technology and Engineering 56(6) (2017) 617-634.
[63] L.A. Utracki, Compatibilization of polymer blends, the Canadian journal of chemical Engineering 80(6) (2002) 1008-1016.
[64] M.S. de Luna, G. Filippone, Effects of nanoparticles on the morphology of immiscible polymer blends–challenges and opportunities, European Polymer Journal 79 (2016) 198-218.
[65] G. Fredi, A. Dorigato, Compatibilization of biopolymer blends: A review, Advanced Industrial and Engineering Polymer Research 7(4) (2024) 373-404.
[66] L.A. Utracki, C.A. Wilkie, Polymer blends handbook, Kluwer academic publishers Dordrecht2002.
[67] M.M. Coleman, P.C. Painter, J.F. Graf, Specific interactions and the miscibility of polymer blends, Routledge2017.
[68] A. Vozniak, R. Hosseinnezhad, I. Vozniak, A. Galeski, PET mechanical recycling. A new principle for chain extender introduction, Sustainable Materials and Technologies 40 (2024) e00886.
[69] W.G. Blasius Jr, G.A. Deeter, M.A. Villalobos, Oligomeric chain extenders for processing, post-processing and recycling of condensation polymers, synthesis, compositions and applications, Google Patents, 2006.
[70] T. Standau, M. Nofar, D. Dörr, H. Ruckdäschel, V. Altstädt, A review on multifunctional epoxy-based Joncryl® ADR chain extended thermoplastics, Polymer Reviews 62(2) (2022) 296-350.
[71] M. Villalobos, A. Awojulu, T. Greeley, G. Turco, G. Deeter, Oligomeric chain extenders for economic reprocessing and recycling of condensation plastics, Energy 31(15) (2006) 3227-3234.
[72] S. Shankar, J.-W. Rhim, Effect of types of zinc oxide nanoparticles on structural, mechanical and antibacterial properties of poly (lactide)/poly (butylene adipate-co-terephthalate) composite films, Food Packaging and Shelf Life 21 (2019) 100327.

[73] L.C. Arruda, M. Magaton, R.E.S. Bretas, M.M. Ueki, Influence of chain extender on mechanical, thermal and morphological properties of blown films of PLA/PBAT blends, Polymer Testing 43 (2015) 27-37.
[74] H. Li, M.A. Huneault, Effect of chain extension on the properties of PLA/TPS blends, Journal of Applied Polymer Science 122(1) (2011) 134-141.
[75] R. Al-Itry, K. Lamnawar, A. Maazouz, Improvement of thermal stability, rheological and mechanical properties of PLA, PBAT and their blends by reactive extrusion with functionalized epoxy, Polymer Degradation and Stability 97(10) (2012) 1898-1914.
[76] N. Najafi, M.-C. Heuzey, P.J. Carreau, D. Therriault, C.B. Park, Rheological and foaming behavior of linear and branched polylactides, Rheologica Acta 53 (2014) 779-790.
[77] J. Wang, W. Zhu, H. Zhang, C.B. Park, Continuous processing of low-density, microcellular poly (lactic acid) foams with controlled cell morphology and crystallinity, Chemical Engineering Science 75 (2012) 390-399.
[78] R. Salehiyan, M.C. Kim, T. Xia, S. Jin, M. Nofar, L. Chalmers, K. Hyun, Enhanced re-processability of poly (butylene adipate-co-terephthalate)(PBAT) via chain extension toward a more sustainable end-of-life, Polymer Engineering & Science (2024).
[79] A.A. Tavares, D.F. Silva, P.S. Lima, D.L. Andrade, S.M. Silva, E.L. Canedo, Chain extension of virgin and recycled polyethylene terephthalate, Polymer Testing 50 (2016) 26-32.
[80] J. Cailloux, O.O. Santana Pérez, E.A. Franco Urquiza, J. Bou, F. Carrasco, J. Gamez-Perez, M.L. Maspoch, Sheets of branched poly (lactic acid) obtained by one step reactive extrusion calendering process: Melt rheology analysis, (2013).
[81] J. Cailloux, O.O. Santana, M.L. Maspoch, J.J. Bou, F. Carrasco, Using viscoelastic properties to quantitatively estimate the amount of modified poly (lactic acid) chains through reactive extrusion, Journal of Rheology 59(5) (2015) 1191-1227.
[82] S. Makkam, W. Harnnarongchai, Rheological and mechanical properties of recycled PET modified by reactive extrusion, Energy Procedia 56 (2014) 547-553.
[83] M. Guclu, Y. Alkan Göksu, B. Özdemir, A. Ghanbari, M. Nofar, Thermal stabilization of recycled PET through chain extension and blending with PBT, Journal of Polymers and the Environment (2022) 1-9.
[84] M.-B. Coltelli, V. Gigante, L. Aliotta, A. Lazzeri, Recyclability Perspectives of the Most Diffused Biobased and Biodegradable Plastic Materials, Macromol 4(2) (2024) 401-419.
[85] V. Ojijo, S.S. Ray, Super toughened biodegradable polylactide blends with non-linear copolymer interfacial architecture obtained via facile in-situ reactive compatibilization, Polymer 80 (2015) 1-17.
[86] X. Wang, S. Peng, H. Chen, X. Yu, X. Zhao, Mechanical properties, rheological behaviors, and phase morphologies of high-toughness PLA/PBAT blends by in-situ reactive compatibilization, Composites Part B: Engineering 173 (2019) 107028.
[87] J. Liu, H. Jiang, L. Chen, Grafting of glycidyl methacrylate onto poly (lactide) and properties of PLA/starch blends compatibilized by the grafted copolymer, Journal of Polymers and the Environment 20 (2012) 810-816.
[88] J. Schneider, S. Manjure, R. Narayan, Reactive modification and compatibilization of poly (lactide) and poly (butylene adipate-co-terephthalate) blends with epoxy functionalized-poly (lactide) for blown film applications, Journal of Applied Polymer Science 133(16) (2016).
[89] N. Zhang, Q. Wang, J. Ren, L. Wang, Preparation and properties of biodegradable poly (lactic acid)/poly (butylene adipate-co-terephthalate) blend with glycidyl methacrylate as reactive processing agent, Journal of Materials Science 44 (2009) 250-256.

[90] Q. Shi, C. Chen, L. Gao, L. Jiao, H. Xu, W. Guo, Physical and degradation properties of binary or ternary blends composed of poly (lactic acid), thermoplastic starch and GMA grafted POE, Polymer Degradation and Stability 96(1) (2011) 175-182.
[91] D. Wei, H. Wang, H. Xiao, A. Zheng, Y. Yang, Morphology and mechanical properties of poly (butylene adipate-co-terephthalate)/potato starch blends in the presence of synthesized reactive compatibilizer or modified poly (butylene adipate-co-terephthalate), Carbohydrate polymers 123 (2015) 275-282.
[92] L. Gardella, M. Calabrese, O. Monticelli, PLA maleation: an easy and effective method to modify the properties of PLA/PCL immiscible blends, Colloid and Polymer Science 292 (2014) 2391-2398.
[93] A. Teamsinsungvon, Y. Ruksakulpiwat, K. Jarukumjorn, Preparation and characterization of poly (lactic acid)/poly (butylene adipate-co-terepthalate) blends and their composite, Polymer-Plastics Technology and Engineering 52(13) (2013) 1362-1367.
[94] F. Chen, J. Zhang, In-situ poly (butylene adipate-co-terephthalate)/soy protein concentrate composites: Effects of compatibilization and composition on properties, Polymer 51(8) (2010) 1812-1819.
[95] R. Zhu, H. Liu, J. Zhang, Compatibilizing effects of maleated poly (lactic acid)(PLA) on properties of PLA/soy protein composites, Industrial & Engineering Chemistry Research 51(22) (2012) 7786-7792.
[96] Y. Nabar, J.M. Raquez, P. Dubois, R. Narayan, Production of starch foams by twin-screw extrusion: Effect of maleated poly (butylene adipate-co-terephthalate) as a compatibilizer, Biomacromolecules 6(2) (2005) 807-817.
[97] M.A. Huneault, H. Li, Morphology and properties of compatibilized polylactide/thermoplastic starch blends, Polymer 48(1) (2007) 270-280.
[98] C.-S. Wu, Physical properties and biodegradability of maleated-polycaprolactone/starch composite, Polymer degradation and stability 80(1) (2003) 127-134.
[99] R. Mani, M. Bhattacharya, Properties of injection moulded blends of starch and modified biodegradable polyesters, European Polymer Journal 37(3) (2001) 515-526.
[100] Z. Xiong, C. Li, S. Ma, J. Feng, Y. Yang, R. Zhang, J. Zhu, The properties of poly (lactic acid)/starch blends with a functionalized plant oil: Tung oil anhydride, Carbohydrate Polymers 95(1) (2013) 77-84.
[101] Z. Xiong, L. Zhang, S. Ma, Y. Yang, C. Zhang, Z. Tang, J. Zhu, Effect of castor oil enrichment layer produced by reaction on the properties of PLA/HDI-g-starch blends, Carbohydrate polymers 94(1) (2013) 235-243.
[102] K. Ku-Marsilla, C. Verbeek, Compatibilization of protein thermoplastics and polybutylene succinate blends, Macromolecular Materials and Engineering 300(2) (2015) 161-171.
[103] B. Liu, L. Jiang, H. Liu, J. Zhang, Synergetic effect of dual compatibilizers on in situ formed poly (lactic acid)/soy protein composites, Industrial & engineering chemistry research 49(14) (2010) 6399-6406.
[104] M.M.F. Ferrarezi, M. de Oliveira Taipina, L.C. Escobar da Silva, M.d.C. Gonçalves, Poly (ethylene glycol) as a compatibilizer for poly (lactic acid)/thermoplastic starch blends, Journal of Polymers and the Environment 21 (2013) 151-159.
[105] P. Jariyasakoolroj, S. Chirachanchai, Silane modified starch for compatible reactive blend with poly (lactic acid), Carbohydrate polymers 106 (2014) 255-263.
[106] B. Kosan, F. Meister, T. Liebert, T. Heinze, Hydrophobic modification of starch via grafting with an oxazoline-derivative, Cellulose 13 (2006) 105-113.

[107] J. Ferri, D. Garcia-Garcia, L. Sánchez-Nacher, O. Fenollar, R. Balart, The effect of maleinized linseed oil (MLO) on mechanical performance of poly (lactic acid)-thermoplastic starch (PLA-TPS) blends, Carbohydrate polymers 147 (2016) 60-68.
[108] Z. Xiong, Y. Yang, J. Feng, X. Zhang, C. Zhang, Z. Tang, J. Zhu, Preparation and characterization of poly (lactic acid)/starch composites toughened with epoxidized soybean oil, Carbohydrate polymers 92(1) (2013) 810-816.
[109] K. Bocz, F. Ronkay, K.E. Decsov, B. Molnár, G. Marosi, Application of low-grade recyclate to enhance reactive toughening of poly (ethylene terephthalate), Polymer Degradation and Stability 185 (2021) 109505.
[110] M. Harada, T. Ohya, K. Iida, H. Hayashi, K. Hirano, H. Fukuda, Increased impact strength of biodegradable poly (lactic acid)/poly (butylene succinate) blend composites by using isocyanate as a reactive processing agent, Journal of applied polymer science 106(3) (2007) 1813-1820.
[111] V. Vilay, M. Mustapha, Z. Ahmad, T. Mitsugu, Characterization of the microstructure and mode I fracture property of biodegradable poly (L-lactic acid) and poly (ϵ-caprolactone) polymer blends with the additive lysine triisocyanate, Polymer-Plastics Technology and Engineering 52(8) (2013) 768-773.
[112] T. Takayama, M. Todo, Improvement of impact fracture properties of PLA/PCL polymer blend due to LTI addition, Journal of materials science 41 (2006) 4989-4992.
[113] L. Li, G. Song, G. Tang, Novel biodegradable polylactide/poly (butylene succinate) composites via cross-linking with methylene diphenyl diisocyanate, Polymer-Plastics Technology and Engineering 52(12) (2013) 1183-1187.
[114] M. Harada, K. Iida, K. Okamoto, H. Hayashi, K. Hirano, Reactive compatibilization of biodegradable poly (lactic acid)/poly (ε-caprolactone) blends with reactive processing agents, Polymer Engineering & Science 48(7) (2008) 1359-1368.
[115] M. Ma, H. Zheng, S. Chen, B. Wu, H. He, L. Chen, X. Wang, Super-toughened poly (L-lactic acid) fabricated via reactive blending and interfacial compatibilization, Polymer International 65(10) (2016) 1187-1194.
[116] Z. Zhong, X.S. Sun, Properties of soy protein isolate/polycaprolactone blends compatibilized by methylene diphenyl diisocyanate, Polymer 42(16) (2001) 6961-6969.
[117] K. Fang, B. Wang, K. Sheng, X.S. Sun, Properties and morphology of poly (lactic acid)/soy protein isolate blends, Journal of applied polymer science 114(2) (2009) 754-759.
[118] S. Karagoz, G. Ozkoc, Effects of a diisocyanate compatibilizer on the properties of citric acid modified thermoplastic starch/poly (lactic acid) blends, Polymer Engineering & Science 53(10) (2013) 2183-2193.
[119] Q. Sun, T. Mekonnen, M. Misra, A.K. Mohanty, Novel biodegradable cast film from carbon dioxide based copolymer and poly (lactic acid), Journal of Polymers and the Environment 24 (2016) 23-36.
[120] P. Ma, D.G. Hristova-Bogaerds, P.J. Lemstra, Y. Zhang, S. Wang, Toughening of PHBV/PBS and PHB/PBS blends via in situ compatibilization using dicumyl peroxide as a free-radical grafting initiator, Macromolecular Materials and Engineering 297(5) (2012) 402-410.
[121] R. Wang, S. Wang, Y. Zhang, C. Wan, P. Ma, Toughening modification of PLLA/PBS blends via in situ compatibilization, Polymer Engineering & Science 49(1) (2009) 26-33.
[122] P. Ma, X. Cai, Y. Zhang, S. Wang, W. Dong, M. Chen, P. Lemstra, In-situ compatibilization of poly (lactic acid) and poly (butylene adipate-co-terephthalate) blends by using dicumyl peroxide as a free-radical initiator, Polymer Degradation and Stability 102 (2014) 145-151.

[123] M.-B. Coltelli, S. Bronco, C. Chinea, The effect of free radical reactions on structure and properties of poly (lactic acid)(PLA) based blends, Polymer Degradation and Stability 95(3) (2010) 332-341.
[124] W. Dong, B. Zou, Y. Yan, P. Ma, M. Chen, Effect of Chain-Extenders on the Properties and Hydrolytic Degradation Behavior of the Poly (lactide)/Poly (butylene adipate-co-terephthalate) Blends, International journal of molecular sciences 14(10) (2013) 20189-20203.
[125] M. Nerkar, J.A. Ramsay, B.A. Ramsay, A.A. Vasileiou, M. Kontopoulou, Improvements in the melt and solid-state properties of poly (lactic acid), poly-3-hydroxyoctanoate and their blends through reactive modification, Polymer 64 (2015) 51-61.
[126] P. Jandas, S. Mohanty, S. Nayak, Morphology and thermal properties of renewable resource-based polymer blend nanocomposites influenced by a reactive compatibilizer, ACS Sustainable Chemistry & Engineering 2(3) (2014) 377-386.
[127] T. Mekonnen, M. Misra, A.K. Mohanty, Fermented soymeals and their reactive blends with poly (butylene adipate-co-terephthalate) in engineering biodegradable cast films for sustainable packaging, ACS Sustainable Chemistry & Engineering 4(3) (2016) 782-793.
[128] M.-B. Coltelli, C. Toncelli, F. Ciardelli, S. Bronco, Compatible blends of biorelated polyesters through catalytic transesterification in the melt, Polymer Degradation and Stability 96(5) (2011) 982-990.
[129] S. Lin, W. Guo, C. Chen, J. Ma, B. Wang, Mechanical properties and morphology of biodegradable poly (lactic acid)/poly (butylene adipate-co-terephthalate) blends compatibilized by transesterification, Materials & Design (1980-2015) 36 (2012) 604-608.
[130] L. Zhou, G. Zhao, W. Jiang, Effects of catalytic transesterification and composition on the toughness of poly (lactic acid)/poly (propylene carbonate) blends, Industrial & Engineering Chemistry Research 55(19) (2016) 5565-5573.
[131] S. Spinella, J. Cai, C. Samuel, J. Zhu, S.A. McCallum, Y. Habibi, J.-M. Raquez, P. Dubois, R.A. Gross, Polylactide/poly (ω-hydroxytetradecanoic acid) reactive blending: A green renewable approach to improving polylactide properties, Biomacromolecules 16(6) (2015) 1818-1826.
[132] M. Yao, H. Deng, F. Mai, K. Wang, Q. Zhang, F. Chen, Q. Fu, Modification of poly (lactic acid)/poly (propylene carbonate) blends through melt compounding with maleic anhydride, Express Polym Lett 5(11) (2011) 937-949.
[133] W. Dong, B. Zou, P. Ma, W. Liu, X. Zhou, D. Shi, Z. Ni, M. Chen, Influence of phthalic anhydride and bioxazoline on the mechanical and morphological properties of biodegradable poly (lactic acid)/poly [(butylene adipate)-co-terephthalate] blends, Polymer international 62(12) (2013) 1783-1790.
[134] B.Y. Shin, Compatibilization of immiscible poly (lactic acid)/poly (ε-caprolactone) blend through electron-beam irradiation with the addition of a compatibilizing agent, Radiation physics and chemistry 83 (2013) 98-104.
[135] M. Kumar, S. Mohanty, S. Nayak, M.R. Parvaiz, Effect of glycidyl methacrylate (GMA) on the thermal, mechanical and morphological property of biodegradable PLA/PBAT blend and its nanocomposites, Bioresource technology 101(21) (2010) 8406-8415.
[136] J.F. Zhang, X. Sun, Mechanical and thermal properties of poly (lactic acid)/starch blends with dioctyl maleate, Journal of Applied Polymer Science 94(4) (2004) 1697-1704.
[137] S.S. Ray, R. Salehiyan, Nanostructured immiscible polymer blends: migration and interface, Elsevier2019.

[138] F. Tao, D. Auhl, A.C. Baudouin, F.J. Stadler, C. Bailly, Influence of multiwall carbon nanotubes trapped at the interface of an immiscible polymer blend on interfacial tension, Macromolecular Chemistry and Physics 214(3) (2013) 350-360.

[139] S. Sinha Ray, M. Bousmina, Compatibilization efficiency of organoclay in an immiscible polycarbonate/poly (methyl methacrylate) blend, Macromolecular rapid communications 26(6) (2005) 450-455.

[140] R. Salehiyan, S. Sinha Ray, Processing of polymer blends, emphasizing: melt compounding; influence of nanoparticles on blend morphology and rheology; reactive processing in ternary systems; morphology–property relationships; performance and application challenges; and opportunities and future trends, Processing of Polymer-based Nanocomposites: Processing-structure-property-performance relationships (2018) 167-197.

[141] S.S. Ray, M. Bousmina, A. Maazouz, Morphology and properties of organoclay modified polycarbonate/poly (methyl methacrylate) blend, Polymer Engineering & Science 46(8) (2006) 1121-1129.

[142] R. Salehiyan, H.Y. Song, W.J. Choi, K. Hyun, Characterization of effects of silica nanoparticles on (80/20) PP/PS blends via nonlinear rheological properties from Fourier transform rheology, Macromolecules 48(13) (2015) 4669-4679.

[143] L. Elias, F. Fenouillot, J.-C. Majesté, P. Cassagnau, Morphology and rheology of immiscible polymer blends filled with silica nanoparticles, Polymer 48(20) (2007) 6029-6040.

[144] S.S. Ray, S. Pouliot, M. Bousmina, L.A. Utracki, Role of organically modified layered silicate as an active interfacial modifier in immiscible polystyrene/polypropylene blends, Polymer 45(25) (2004) 8403-8413.

[145] F. Fenouillot, P. Cassagnau, J.-C. Majesté, Uneven distribution of nanoparticles in immiscible fluids: Morphology development in polymer blends, Polymer 50(6) (2009) 1333-1350.

[146] M. Nofar, R. Salehiyan, S.S. Ray, Influence of nanoparticles and their selective localization on the structure and properties of polylactide-based blend nanocomposites, Composites Part B: Engineering 215 (2021) 108845.

[147] G. Filippone, D. Acierno, Altering the onset of cocontinuity in nanocomposite immiscible blends by acting on the melt-compounding procedure, Journal of Applied Polymer Science 122(6) (2011) 3711-3718.

[148] L. Elias, F. Fenouillot, J.-C. Majesté, P. Alcouffe, P. Cassagnau, Immiscible polymer blends stabilized with nano-silica particles: Rheology and effective interfacial tension, Polymer 49(20) (2008) 4378-4385.

[149] A.-C. Baudouin, C. Bailly, J. Devaux, Interface localization of carbon nanotubes in blends of two copolymers, Polymer Degradation and Stability 95(3) (2010) 389-398.

[150] M. Mohammadi, M.-C. Heuzey, P.J. Carreau, A. Taguet, Interfacial localization of CNCs in PLA/PBAT blends and its effect on rheological, thermal, and mechanical properties, Polymer 233 (2021) 124229.

[151] R. Salehiyan, M. Nofar, K. Malkappa, S.S. Ray, Effect of nanofillers characteristics and their selective localization on morphology development and rheological properties of melt-processed polylactide/poly (butylene adipate-co-terephthalate) blend composites, Polymer Engineering & Science 60(11) (2020) 2749-2760.

[152] Z. Xiao, G. Li, C. Liu, H. Li, J. Lin, The carbon nanotubes effects on the morphology and properties of poly (lactic) acid/poly (butylene adipate-co-terephthalate) blends, Polymer Composites 43(12) (2022) 8725-8736.

[153] P. Wang, S. Gao, X. Chen, L. Yang, X. Wu, S. Feng, X. Hu, J. Liu, P. Xu, Y. Ding, Effect of hydroxyl and carboxyl-functionalized carbon nanotubes on phase morphology, mechanical and dielectric properties of poly (lactide)/poly (butylene adipate-co-terephthalate) composites, International Journal of Biological Macromolecules 206 (2022) 661-669.

[154] K.N. Dhakal, B. Krause, R. Lach, A. Wutzler, W. Grellmann, H.H. Le, A. Das, S. Wießner, G. Heinrich, R. Adhikari, Electrically conductive nanocomposites based on poly (lactic acid)/flexible copolyester blends with multiwalled carbon nanotubes, Journal of Applied Polymer Science 139(4) (2022) 51554.

[155] Y. Liu, H. He, G. He, J. Zhao, Y. Yang, G. Tian, Segregated polylactide/poly (butylene adipate-co-terephthalate)/MWCNTs nanocomposites with excellent electrical conductivity and electromagnetic interference shielding, Journal of Applied Polymer Science 139(8) (2022) 51668.

[156] J. Chen, R.R. Hu, F.L. Jin, S.J. Park, Synergistic reinforcing of poly (lactic acid) by poly (butylene adipate-co-terephthalate) and alumina nanoparticles, Journal of Applied Polymer Science 138(16) (2021) 50250.

[157] H. Khonakdar, S.S. Khasraghi, A.H. Yazdanbakhsh, S.R. Mousavi, S. Ahmadi, H. Arabi, M.A. Nobre, H.A. Khonakdar, An assessment of the role of nanosilica in thermal/thermo-oxidative degradation mechanism of poly (lactic acid)/polybutylene adipate terephthalate blend nanocomposites, Polymers for Advanced Technologies 35(4) (2024) e6374.

[158] E.J. Dil, N. Virgilio, B.D. Favis, The effect of the interfacial assembly of nano-silica in poly (lactic acid)/poly (butylene adipate-co-terephthalate) blends on morphology, rheology and mechanical properties, European Polymer Journal 85 (2016) 635-646.

[159] T. Malwela, S.S. Ray, Enzymatic degradation behavior of nanoclay reinforced biodegradable PLA/PBSA blend composites, International journal of biological macromolecules 77 (2015) 131-142.

[160] V. Ojijo, H. Cele, S. Sinha Ray, Morphology and Properties of Polymer Composites Based on Biodegradable Polylactide/Poly [(butylene succinate)-co-adipate] Blend and Nanoclay, macromolecular Materials and Engineering 296(9) (2011) 865-877.

[161] V. Ojijo, S. Sinha Ray, R. Sadiku, Effect of nanoclay loading on the thermal and mechanical properties of biodegradable polylactide/poly [(butylene succinate)-co-adipate] blend composites, ACS applied materials & interfaces 4(5) (2012) 2395-2405.

[162] P. Somdee, M. Shettar, N. Prasoetsopha, M.A. Ansari, Reinforcing poly (lactic acid)/poly (butylene succinate) biodegradable blends with silicon carbide (SiC): A silane-coupled approach for enhanced mechanical and thermal performance, Polymer Composites (2024).

[163] A. Bhatia, R. Gupta, S. Bhattacharya, H. Choi, Effect of clay on thermal, mechanical and gas barrier properties of biodegradable poly (lactic acid)/poly (butylene succinate)(PLA/PBS) nanocomposites, International Polymer Processing 25(1) (2010) 5-14.

[164] R. Scaffaro, A. Maio, F. Sutera, E.F. Gulino, M. Morreale, Degradation and recycling of films based on biodegradable polymers: A short review, Polymers 11(4) (2019) 651.

[165] R. Scaffaro, F. Lopresti, L. Botta, A. Maio, Mechanical behavior of polylactic acid/polycaprolactone porous layered functional composites, Composites Part B: Engineering 98 (2016) 70-77.

[166] M. Morreale, A. Liga, M.C. Mistretta, L. Ascione, F.P. La Mantia, Mechanical, thermomechanical and reprocessing behavior of green composites from biodegradable polymer and wood flour, Materials 8(11) (2015) 7536-7548.

[167] M. Niaounakis, Biopolymers: reuse, recycling, and disposal, William Andrew2013.

[168] R. Kumar, K. Sadeghi, J. Jang, J. Seo, Mechanical, chemical, and bio-recycling of biodegradable plastics: A review, Science of the Total Environment 882 (2023) 163446.

[169] I. Dedieu, S. Peyron, N. Gontard, C. Aouf, The thermo-mechanical recyclability potential of biodegradable biopolyesters: Perspectives and limits for food packaging application, Polymer Testing 111 (2022) 107620.

[170] P. Srimalanon, I. Martín-Fabiani, B. Prapagdee, T. Markpin, E. Wimolmala, N. Sombatsompop, Influence of reprocessing cycles on mechanical and antibacterial performances of hygienic in situ compatibilized PBS/PLA blends doped with HPQM, Journal of Material Cycles and Waste Management 23(5) (2021) 1893-1904.

[171] S. Al-Salem, P. Lettieri, J. Baeyens, Recycling and recovery routes of plastic solid waste (PSW): A review, Waste management 29(10) (2009) 2625-2643.

[172] S. Kumar, A.K. Panda, R.K. Singh, A review on tertiary recycling of high-density polyethylene to fuel, Resources, Conservation and Recycling 55(11) (2011) 893-910.

[173] S. Al-Salem, P. Lettieri, J. Baeyens, The valorization of plastic solid waste (PSW) by primary to quaternary routes: From re-use to energy and chemicals, Progress in Energy and Combustion Science 36(1) (2010) 103-129.

[174] C. Baillie, D. Matovic, T. Thamae, S. Vaja, Waste-based composites—Poverty reducing solutions to environmental problems, Resources, Conservation and Recycling 55(11) (2011) 973-978.

[175] I.A. Ignatyev, W. Thielemans, B. Vander Beke, Recycling of polymers: a review, ChemSusChem 7(6) (2014) 1579-1593.

[176] S. Dimitris, L. Achilias, L. Andriotis, I. Koutsidis, D. Louka, N. Nianias, P. Siafaka, I. Tsagkalias, G. Tsintzou, Material Recycling-Trends and Perspectives, Greece: Department of Chemistry, Aristotle University of Thessaloniki (2012).

[177] A. Brems, J. Baeyens, R. Dewil, Recycling and recovery of post-consumer plastic solid waste in a European context. Therm. Sci. 16, 669–685, 2012.

[178] ISO, Plastics–Guidelines for the recovery and recycling of plastics waste, ISO 15270: 2008 (E) (2008).

[179] T. Ramos-Hernández, J.R. Robledo-Ortíz, M.E. González-López, A.S.M. del Campo, R. González-Núñez, D. Rodrigue, A.A. Pérez Fonseca, Mechanical recycling of PLA: Effect of weathering, extrusion cycles, and chain extender, Journal of Applied Polymer Science 140(16) (2023) e53759.

[180] J. Vlachopoulos, D. Strutt, Polymer processing, Materials science and technology 19(9) (2003) 1161-1169.

[181] N. Nomadolo, A. Mtibe, O. Ofosu, C. Mekoa, J. Letwaba, S. Muniyasamy, The Effect of Mechanical Recycling on the Thermal, Mechanical, and Chemical Properties of Poly (Butylene Adipate-Co-Terephthalate)(PBAT), Poly (Butylene Succinate)(PBS), Poly (Lactic Acid)(PLA), PBAT-PBS Blend and PBAT-TPS Biocomposite, Journal of Polymers and the Environment (2024) 1-16.

[182] I. Zembouai, S. Bruzaud, M. Kaci, A. Benhamida, Y.-M. Corre, Y. Grohens, Mechanical recycling of poly (3-hydroxybutyrate-co-3-hydroxyvalerate)/polylactide based blends, Journal of Polymers and the Environment 22 (2014) 449-459.

[183] R. Plavec, S. Hlaváčiková, L. Omaníková, J. Feranc, Z. Vanovčanová, K. Tomanová, J. Bočkaj, J. Kruželák, E. Medlenová, I. Gálisová, Recycling possibilities of bioplastics based on PLA/PHB blends, Polymer Testing 92 (2020) 106880.

[184] R. Plavec, V. Horváth, S. Hlaváčiková, L. Omaníková, M. Repiská, E. Medlenová, J. Feranc, J. Kruželák, R. Přikryl, S. Figalla, Influence of multiple thermomechanical processing of 3D filaments based on polylactic acid and polyhydroxybutyrate on their rheological and utility properties, Polymers 14(10) (2022) 1947.
[185] N.C. Farias, I. Major, D. Devine, M. Brennan Fournet, R. Pezzoli, S. Farshbaf Taghinezhad, M. Hesabi, Multiple recycling of a PLA/PHB biopolymer blend for sustainable packaging applications: Rheology-morphology, thermal, and mechanical performance analysis, Polymer Engineering & Science 62(6) (2022) 1764-1774.
[186] A. Chikh, A. Benhamida, M. Kaci, A. Bourmaud, S. Bruzaud, Recyclability assessment of poly (3-hydroxybutyrate-co-3-hydroxyvalerate)/poly (butylene succinate) blends: combined influence of sepiolite and compatibilizer, Polymer Degradation and Stability 142 (2017) 234-243.
[187] K. Resch-Fauster, A. Klein, E. Blees, M. Feuchter, Mechanical recyclability of technical biopolymers: Potential and limits, Polymer Testing 64 (2017) 287-295.
[188] V. Titone, M.C. Mistretta, L. Botta, F.P. La Mantia, Toward the Decarbonization of Plastic: Monopolymer Blend of Virgin and Recycled Bio-Based, Biodegradable Polymer, Polymers 14(24) (2022) 5362.
[189] A. Bourmaud, D. Åkesson, J. Beaugrand, A. Le Duigou, M. Skrifvars, C. Baley, Recycling of L-Poly-(lactide)-Poly-(butylene-succinate)-flax biocomposite, Polymer degradation and stability 128 (2016) 77-88.
[190] H. Sirin, B. Tuna, G. Ozkoc, The effects of thermomechanical cycles on the properties of PLA/TPS blends, Advances in Polymer Technology 33(S1) (2014).
[191] M. Dadras Chomachayi, A. Jalali-arani, J. Martinez Urreaga, A comparison of the effect of silk fibroin nanoparticles and microfibers on the reprocessing and biodegradability of PLA/PCL blends, Journal of Polymers and the Environment 29 (2021) 2585-2597.
[192] W. Zhu, X. Wang, X. Chen, K. Xu, Miscibility, crystallization, and mechanical properties of poly (3-hydroxybutyrate-co-4-hydroxybutyrate)/poly (butylene succinate) blends, Journal of Applied Polymer Science 114(6) (2009) 3923-3931.
[193] J. Scheirs, Polymer recycling: science, technology and applications, (No Title) (1998).
[194] F.P. La Mantia, M.C. Mistretta, V. Titone, An additive model to predict the rheological and mechanical properties of polypropylene blends made by virgin and reprocessed components, Recycling 6(1) (2021) 2.
[195] A. Bourmaud, Y.-M. Corre, C. Baley, Fully biodegradable composites: Use of poly-(butylene-succinate) as a matrix and to plasticize l-poly-(lactide)-flax blends, Industrial Crops and Products 64 (2015) 251-257.
[196] I. Pillin, N. Montrelay, A. Bourmaud, Y. Grohens, Effect of thermo-mechanical cycles on the physico-chemical properties of poly (lactic acid), Polymer Degradation and Stability 93(2) (2008) 321-328.
[197] C. Simões, J. Viana, A. Cunha, Mechanical properties of poly (ε-caprolactone) and poly (lactic acid) blends, Journal of Applied Polymer Science 112(1) (2009) 345-352.
[198] M. Żenkiewicz, J. Richert, P. Rytlewski, K. Moraczewski, M. Stepczyńska, T. Karasiewicz, Characterisation of multi-extruded poly (lactic acid), Polymer Testing 28(4) (2009) 412-418.
[199] M. Dadras Chomachayi, A. Solouk, S. Akbari, D. Sadeghi, F. Mirahmadi, H. Mirzadeh, Electrospun nanofibers comprising of silk fibroin/gelatin for drug delivery applications: Thyme essential oil and doxycycline monohydrate release study, Journal of biomedical materials research part a 106(4) (2018) 1092-1103.

[200] M. Dadras Chomachayi, A. Solouk, H. Mirzadeh, Improvement of the electrospinnability of silk fibroin solution by atmospheric pressure plasma treatment, Fibers and Polymers 20 (2019) 1594-1600.
[201] M. Dadras Chomachayi, A. Jalali-Arani, F.R. Beltrán, M.U. de la Orden, J. Martínez Urreaga, Biodegradable nanocomposites developed from PLA/PCL blends and silk fibroin nanoparticles: study on the microstructure, thermal behavior, crystallinity and performance, Journal of Polymers and the Environment 28 (2020) 1252-1264.
[202] E.-C. Chen, T.-M. Wu, Isothermal crystallization kinetics and thermal behavior of poly (ε-caprolactone)/multi-walled carbon nanotube composites, Polymer degradation and Stability 92(6) (2007) 1009-1015.
[203] R.R.d. Oliveira, T.A.d. Oliveira, L.R.C.d. Silva, R. Barbosa, T.S. Alves, L.H.d. Carvalho, D.T. Rodrigues, Effect of reprocessing cycles on the morphology and mechanical properties of a poly (propylene)/poly (hydroxybutyrate) blend and its nanocomposite, Materials Research 24 (2021) e20200372.
[204] G.W. de Kort, L.n.H. Bouvrie, S. Rastogi, C.H. Wilsens, Thermoplastic PLA-LCP composites: a route toward sustainable, reprocessable, and recyclable reinforced materials, ACS sustainable chemistry & engineering 8(1) (2019) 624-631.
[205] A.M. Peres, R.R. Pires, R.L. Oréfice, Evaluation of the effect of reprocessing on the structure and properties of low density polyethylene/thermoplastic starch blends, Carbohydrate polymers 136 (2016) 210-215.
[206] T.A. Oliveira, R.R. Oliveira, R. Barbosa, J.B. Azevedo, T.S. Alves, Effect of reprocessing cycles on the degradation of PP/PBAT-thermoplastic starch blends, Carbohydrate Polymers 168 (2017) 52-60.
[207] A. Hejna, J. Lenża, K. Formela, J. Korol, Studies on the combined impact of starch source and multiple processing on selected properties of thermoplastic starch/ethylene-vinyl acetate blends, Journal of Polymers and the Environment 27 (2019) 1112-1126.
[208] M.A. Tavanaie, A.H. Ghahari, A study on melt recycling of bio-based polypropylene/thermoplastic starch compound, Journal of Applied Polymer Science 138(43) (2021) 51282.
[209] D.M. Lenz, D.M. Tedesco, P.H. Camani, D. dos Santos Rosa, Multiple reprocessing cycles of corn starch-based biocomposites reinforced with Curauá fiber, Journal of Polymers and the Environment 26 (2018) 3005-3016.
[210] C. Pavon, M. Aldas, D. Bertomeu, H. de la Rosa-Ramírez, M.D. Samper, J. López-Martínez, Influence of the Presence of Poly (butylene succinate) in the Poly (ethylene terephthalate) Recycling Process, Clean Technologies 5(1) (2023) 190-202.
[211] D. Battegazzore, S. Bocchini, A. Frache, Crystallization kinetics of poly (lactic acid)-talc composites, Express Polym. Lett 5(10) (2011) 849-858.
[212] K. Pielichowski, J. Njuguna, T.M. Majka, Thermal degradation of polymeric materials, Elsevier2022.
[213] J. Lawton, Effect of starch type on the properties of starch containing films, Carbohydrate Polymers 29(3) (1996) 203-208.
[214] K. Wang, N. Bahlouli, F. Addiego, S. Ahzi, Y. Rémond, D. Ruch, R. Muller, Effect of talc content on the degradation of re-extruded polypropylene/talc composites, Polymer degradation and stability 98(7) (2013) 1275-1286.
[215] A.G. Pedroso, D.d.S. Rosa, Mechanical, thermal and morphological characterization of recycled LDPE/corn starch blends, Carbohydrate Polymers 59(1) (2005) 1-9.

[216] E.G. Bajsić, I. Šmit, M. Leskovac, Blends of thermoplastic polyurethane and polypropylene. I. Mechanical and phase behavior, Journal of applied polymer science 104(6) (2007) 3980-3985.
[217] L.T. Saw, N.A.A. Rahim, C.X. Viet, Processing degradation of polypropylene-ethylene copolymer-kaolin composites by a twin-screw extruder, Polymer Degradation and Stability 111 (2015) 32-37.
[218] W. Shujun, Y. Jiugao, Y. Jinglin, Preparation and characterization of compatible thermoplastic starch/polyethylene blends, Polymer Degradation and Stability 87(3) (2005) 395-401.
[219] R. Zullo, S. Iannace, The effects of different starch sources and plasticizers on film blowing of thermoplastic starch: Correlation among process, elongational properties and macromolecular structure, Carbohydrate Polymers 77(2) (2009) 376-383.
[220] M. Sabetzadeh, R. Bagheri, M. Masoomi, Study on ternary low density polyethylene/linear low density polyethylene/thermoplastic starch blend films, Carbohydrate polymers 119 (2015) 126-133.
[221] M.K. Abdul Wahab, H. Ismail, N. Othman, Compatibilization effects of PE-g-MA on mechanical, thermal and swelling properties of high density polyethylene/natural rubber/thermoplastic tapioca starch blends, Polymer-Plastics Technology and Engineering 51(3) (2012) 298-303.
[222] B. Itim, M. Philip, Effect of multiple extrusions and influence of PP contamination on the thermal characteristics of bottle grade recycled PET, Polymer Degradation and Stability 117 (2015) 84-89.
[223] J.F.M. Moreira, T.S. Alves, R. Barbosa, É.M. de Carvalho, L.H. Carvalho, Effect of Cis-13-docosenamide in the Properties of Compatibilized Polypropylene/Clay Nanocomposites, Macromolecular Symposia, Wiley Online Library, 2016, pp. 68-75.
[224] O. Sadiku-Agboola, E.R. Sadiku, A.T. Adegbola, O.F. Biotidara, Rheological properties of polymers: structure and morphology of molten polymer blends, Materials Sciences and Applications 2(01) (2011) 30.
[225] I. Babbar, G. Mathur, Rheological properties of blends of polycarbonate with poly (acrylonitrile-butadiene-styrene), Polymer 35(12) (1994) 2631-2635.
[226] P. Carreau, M. Bousmina, A. Ajji, Rheological properties of blends: facts and challenges, Progress in Pacific Polymer Science 3: Proceedings of the Third Pacific Polymer Conference Gold Coast, Queensland, December 13–17, 1993, Springer, 1994, pp. 25-39.
[227] M. Fahrländer, C. Friedrich, Rheological properties of polymer blends with sphere-in-sphere morphology, Rheologica acta 38(3) (1999) 206-213.
[228] L. Utracki, Reprocessing of Commingled Polymers and Recycling of Polymer Blends, Frontiers in the Science and Technology of Polymer Recycling, Springer1998, pp. 333-354.
[229] K. Nuñez, C. Rosales, R. Perera, N. Villarreal, J. Pastor, Nanocomposites of PLA/PP blends based on sepiolite, Polymer bulletin 67 (2011) 1991-2016.
[230] N. Touati, M. Kaci, S. Bruzaud, Y. Grohens, The effects of reprocessing cycles on the structure and properties of isotactic polypropylene/cloisite 15A nanocomposites, Polymer Degradation and Stability 96(6) (2011) 1064-1073.
[231] R. Guo, J. Azaiez, C. Bellehumeur, Rheology of fiber filled polymer melts: Role of fiber-fiber interactions and polymer-fiber coupling, Polymer Engineering & Science 45(3) (2005) 385-399.

[232] G. Kalaprasad, G. Mathew, C. Pavithran, S. Thomas, Melt rheological behavior of intimately mixed short sisal–glass hybrid fiber-reinforced low-density polyethylene composites. I. Untreated fibers, Journal of applied polymer science 89(2) (2003) 432-442.
[233] G. Gamon, P. Evon, L. Rigal, Twin-screw extrusion impact on natural fibre morphology and material properties in poly (lactic acid) based biocomposites, Industrial Crops and Products 46 (2013) 173-185.
[234] W. Chow, A.A. Bakar, Z.M. Ishak, J. Karger-Kocsis, U. Ishiaku, Effect of maleic anhydride-grafted ethylene–propylene rubber on the mechanical, rheological and morphological properties of organoclay reinforced polyamide 6/polypropylene nanocomposites, European Polymer Journal 41(4) (2005) 687-696.
[235] T. Gerard, T. Budtova, Morphology and molten-state rheology of polylactide and polyhydroxyalkanoate blends, European polymer journal 48(6) (2012) 1110-1117.
[236] R. Patwa, A. Kumar, V. Katiyar, Effect of silk nano-disc dispersion on mechanical, thermal, and barrier properties of poly (lactic acid) based bionanocomposites, Journal of Applied Polymer Science 135(38) (2018) 46671.
[237] V. Melcova, R. Prikryl, P. Mencik, J. Tochacek, Processing stabilization of poly (3-hydroxybutyrate)/poly (lactic acid) blends with oligomeric carbodiimide, Materials Science Forum, Trans Tech Publ, 2019, pp. 7-12.
[238] F. Beltrán, I. Barrio, V. Lorenzo, B. Del Río, J. Martínez Urreaga, M. de La Orden, Valorization of poly (lactic acid) wastes via mechanical recycling: Improvement of the properties of the recycled polymer, Waste Management & Research 37(2) (2019) 135-141.
[239] M.F. Cosate de Andrade, G. Fonseca, A.R. Morales, L.H.I. Mei, Mechanical recycling simulation of polylactide using a chain extender, Advances in Polymer Technology 37(6) (2018) 2053-2060.
[240] M. Żenkiewicz, M. Kurcok, Effects of compatibilizers and electron radiation on thermomechanical properties of composites consisting of five recycled polymers, Polymer Testing 27(4) (2008) 420-427.
[241] E. Dal Lago, C. Boaretti, F. Piovesan, M. Roso, A. Lorenzetti, M. Modesti, The effect of different compatibilizers on the properties of a post-industrial PC/PET blend, Materials 12(1) (2018) 49.
[242] M. Pracella, L. Rolla, D. Chionna, A. Galeski, Compatibilization and properties of poly (ethylene terephthalate)/polyethylene blends based on recycled materials, Macromolecular Chemistry and Physics 203(10-11) (2002) 1473-1485.
[243] M. Gammino, C. Gioia, A. Maio, R. Scaffaro, G. Lo Re, Chemical-free Reactive Melt Processing of Biosourced Poly (butylene-succinate-adipate) for Improved Mechanical Properties and Recyclability, ACS Applied Polymer Materials (2024).
[244] M.-B. Coltelli, A. Bertolini, L. Aliotta, V. Gigante, A. Vannozzi, A. Lazzeri, Chain extension of poly (Lactic acid)(pla)–based blends and composites containing bran with biobased compounds for controlling their processability and recyclability, Polymers 13(18) (2021) 3050.
[245] P. Feijoo, A.K. Mohanty, A. Rodriguez-Uribe, J. Gámez-Pérez, L. Cabedo, M. Misra, Biodegradable blends from bacterial biopolyester PHBV and bio-based PBSA: Study of the effect of chain extender on the thermal, mechanical and morphological properties, International Journal of Biological Macromolecules 225 (2023) 1291-1305.
[246] W. Luo, Y. Yang, Y. Weng, Z. Huang, C. Zhang, Epoxidized cardanol oleate (ECD-OA) as an effective biobased chain extender of polylactic acid (PLA), International Journal of Biological Macromolecules 282 (2024) 136903.

[247] J. Sun, A. Huang, S. Luo, M. Shi, J. Song, H. Luo, Effect of chain extender on morphologies and properties of PBAT/PLA composites, Journal of Thermoplastic Composite Materials 36(3) (2023) 1175-1186.
[248] K. Yokoyama, Z. Guan, A Vitrimer Acts as a Compatibilizer for Polyethylene and Polypropylene Blends, Angewandte Chemie 136(20) (2024) e202317264.
[249] Z. Chen, C. Cui, C. Jin, X. Li, Y. Zhou, Y. Shao, L. Ma, Y. Zhang, T. Wang, Tough and thermostable polybutylene terephthalate (PBT)/vitrimer blend with enhanced interfacial compatibility, Macromolecular Rapid Communications 44(10) (2023) 2200972.
[250] Z. Liang, Z. Li, S. Zhou, Y. Sun, J. Yuan, C. Zhang, Machine-learning exploration of polymer compatibility, Cell Reports Physical Science 3(6) (2022).
[251] S. Takada, T. Suzuki, Y. Takebayashi, T. Ono, S. Yoda, Machine learning assisted optimization of blending process of polyphenylene sulfide with elastomer using high speed twin screw extruder, Scientific Reports 11(1) (2021) 24079.

Conflict of Interest

The authors have declared no conflict of interest

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