

Recyclability Perspectives of the Most Diffused Biobased and Biodegradable Plastic Materials

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Abstract: The present chapter focuses on the recyclability of both renewable and biodegradable plastics, considering the recovery of matter (mechanical or chemical recycling) from the polymeric materials currently most diffused on the market. Biobased and compostable plastics are carbon neutral; thus, they do not contribute significantly to greenhouse gas (GHG) emissions. Nevertheless, recycling can be beneficial because it allows a prolongation of the material life cycle so that carbon is stored for a longer time up to the final composting. The chemical or mechanical recycling option is linked both to the possibility of reprocessing bioplastics without detrimental loss of properties as well as to the capability of selecting homogenous fractions of bioplastics after waste collection. Moreover, the different structural features of biodegradable bioplastics have resulted in different chemical recycling opportunities and also in different behaviors during the reprocessing operations necessary for recycling. All these aspects are discussed systematically in this review, considering biodegradable bioplastics, their blends and composites with natural fibers.

Keywords: bioplastics; mechanical recycling; chemical recycling; biobased; renewable; biodegradable; compostable; degradation; biobased polymer blends; composites



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1. Introduction

Different classifications related to plastic recycling technologies have been used over time in various fields, for instance, in international congresses and the scientific literature [1–34]. The most complete classification is based on processes and raw materials, and it defines primary, secondary, tertiary and quaternary recycling (Figure 1).

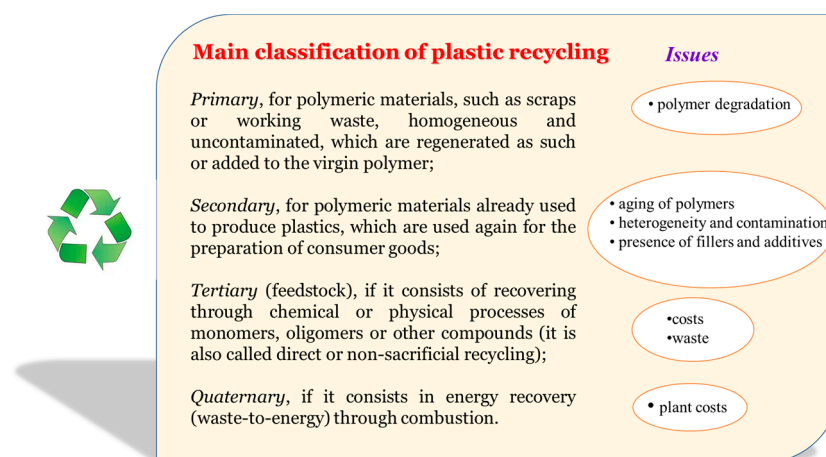


Figure 1. Classification of recycling methodologies based on raw materials and processes.

By primary recycling, also defined as closed loop, we mean the recycling of post-industrial materials, such as scraps (non-contaminated and homogeneous) which are regenerated as such or added in the same process to the virgin polymer.

Secondary recycling means recycling that takes place from a selected material that is reused to produce consumer goods in a plant different from that of primary production. Secondary recycling can belong to the following two classes: post-industrial and post-consumer recycling. Moreover, it can be closed loop, if products to be recycled are collected separately from extensive collection to minimize inhomogeneity, or open loop, if the products are sorted considering their polymer composition from the collected waste.

Tertiary recycling is the recovery through chemical or physical processes of monomers, oligomers or other compounds. Therefore, tertiary recycling involves the use of chemical or physical agents to obtain derivatives from the decomposition or dissolution of polymeric materials. These intermediates can be reintroduced in the same production cycle or they can be used in different ones. Following this approach, the polymeric materials can be considered a reserve of chemicals.

Quaternary recycling is essentially energy recovery through combustion of polymeric materials.

A simpler classification is based on the type of process, considering if it occurs through plastic processing in the molten state (mechanical recycling) using conventional techniques or if it is based on specific and extensive chemical reactions (chemical recycling). Following this classification, it can be noticed that the mechanical recycling is the most widespread since it involves the simple reprocessing of post-consumer or post-industrial materials. When possible, mechanical recycling (both primary and secondary) is generally the preferred method, as it allows for recovering the maximum value contained in the waste [5].

The third classification of recycling technologies considers the final value of recycled products with respect to the original ones. Therefore, open loop recycling [6] refers to a recycling process that preserves the value of the material (for instance, the bottle-to-bottle approach interesting the beverages bottles is a relevant example), whereas downcycling or cascade recycling indicates that the final applications are lower in value than the initial ones. An example is provided by mixed plastics from post-consumer films that are used to produce large-sized objects, such as, for example, profiles for urban furniture. This application is very widespread in the sector of plastics due to the numerous problems of incompatibility between different polymers or materials that make it difficult to produce items with a reduced thickness.

Reasonably, chemical recycling, due to its ability to give intermediate or useful reagents [7], is the one that can more easily lead to up-cycling; however, on the other hand, there are the disadvantages related to plant engineering issues and, therefore, to the overall cost of the recycling operations.

In regard to the development of materials aimed at primary or secondary recycling, it is often necessary to know how to modify the properties of a material with the use of particular additives. For instance, additives such as anti-oxidants or anti-UV agents allow the material to be more stable, while the use of dyes can allow imparting desired aesthetic characteristics.

In the last ten years, interesting bioplastic formulations already available on the market have been developed, mainly addressed at food and packaging applications. These materials from renewable sources are biodegradable and also compostable in many conditions [8]. Their renewability makes them carbon neutral, so, considering the climate change issue, they constitute a better alternative with respect to their fossil counterparts. Materials of this type, which currently have a price comparable with plastics of petrochemical origin, are those based on poly(lactic acid) (PLA) or starch.

In general, the mechanical recycling makes sense if the durability of a material, considering its interactions with microorganisms in the environment, is at least higher than two life cycles; otherwise, only a downcycling can be obtained. From this consideration, two consequences follow:

(1) That it is better if the recycling is conducted by considering long life final applications so that the environmental impact of processing to new products is minimized and a final composting option is considered for the end-of-life management of these products;

(2) That it is more convenient to recycle the bioplastics that show a higher durability.

The most recyclable biobased polymer on the point of view of mechanical recycling is certainly biobased PE, which is not compostable at all, and it is very stable. In any case, as its structure and properties are identical to the petrochemical version, its recyclability is well known [9–11]. The present review considers the biobased and biodegradable plastics because they have the perspective of being recycled one or more times and, when the recycling will be no more feasible, composted (organic recycling), with a better end of life also granting a positive impact on soil fertility [12,13]. These aspects need to be better communicated to stakeholders and consumers, who often are disoriented due to the several options of products' materials present on the market. Moreover, these aspects represent the main contribution of this review to the recycling, environmental and social sectors, since it has the scope of gathering scientific knowledge for the eventual development of new industrial biocircular chains.

PLA is compostable in an industrial environment [14], whereas poly(butylene succinate) (PBS) [15], poly(butylene adipate-co-terephthalate) (PBAT) [16] and poly(hydroxyalcanoate)s (PHAs) [17] are generally compostable in milder conditions. In fact, in the case of PBAT and PBS, the lower glass transitions can make the material easily hydrolysable at lower temperatures than PLA in different environmental matrices [18]. In good agreement, copolymers of PBS, like poly(butylene succinate-co-adipate) (PBSA), were found to be home and soil compostable [15]. PHA and starch-based materials were found compostable in a very wide range of conditions [15], even if, in the case of PHAs, more research work should be conducted to correlate the structure of the different copolymers belonging to this wide group with their final composting behavior. As a consequence, the mechanical recycling of PLA is certainly more significant, because, for PBS, PBAT and PHA, the faster organic recycling option can be preferred.

An important mention should be given to poly(caprolactone) (PCL), which has provided highly degradable results in several environmental conditions [18], and it was proposed in formulations for highly degradable bioplastic materials [18,19]. Although it will be certainly produced by renewable sources in the future [20,21], currently the commercial product is still fully obtained by fossil sources; therefore, it will be not considered in the present review.

Based on these preliminary considerations, this review is aimed at making a survey about the scientific literature regarding the recycling of biobased and biodegradable polymers, as well as their blends and composites, with natural fillers. The recycling of PLA is mainly considered in the following literature survey, because PLA combines a significantly high durability [22–24] with compostability, showing a high versatility. As a consequence, the mechanical recycling and the chemical recycling of PLA will be discussed. Moreover, insights on the recycling of starch, PBS, PBAT and PHA will be also reported to have a full view regarding these biopolymers. Recycling of blends and recycling of composites with natural fillers will be also surveyed and discussed to have a full understanding about the possibility to extend the life cycle of these carbon neutral materials.

2. Recycling of PLA

PLA is produced by industrial synthesis starting from lactic acid, which in turn is produced by starch fermentation, obtained from vegetables such as corn or sugar cane [25,26]. In industrial polymerization, it is possible to control the lactic acid L or D content of the polymer. In particular, the monomer L is always the predominant one, but the introduction of monomer D in a controlled manner has allowed for the modulation of the properties of the polymer. In particular, the extension of crystallization, which strongly influences the optical and mechanical properties, is strictly related to the content of the D monomer.

2.1. Mechanical Recycling

The case of PLA production, properties and recycling is considered in two extensive reviews focused on PLA published by Farah et al. [27] and Castro-Aguirre et al. [28].

Farah et al. [27] suggests that, in recycling PLA, the most significant issue consists of the decreasing in its molecular weight, which has the consequence of worsening its mechanical properties after repeated injections or molding processing. In recycling, the main problems are both the hydrolytic and thermal resistance of PLA [29]. Hydrolysis occurring during processing is described by several authors [30,31] as the main issue also when the recovery and recycling of post-industrial scraps produced as by-products during the use of PLA in packaging molding was considered. A more specific but very comprehensive review about PLA recycling was published by Badia et al. [32]. It can be noticed that, in all these surveys and critical reviews, the degradation of PLA, in particular hydrolysis due to water and thermal degradation, is extensively discussed since it affects recycling operations, especially when recovered bioplastics are reprocessed by extrusion.

The possible hydrolysis occurring during PLA storage must be considered in view of its mechanical recycling. In fact, it is common that the scraps or items that were recovered (for instance from selective collection) may need to be accumulated or stored for a certain time, for reaching specific quantities or matching the industry activities scheduling, before the successive processing. The durability of PLA has been mainly explored for applications in biomedical products, such as carriers of drug for controlled release, resorbable scaffolds and implants. In this field, understanding the interactions between the polymer and the biological fluids is pivotal. For this reason, most of the available literature is focused on the hydrolysis of different types of PLA (e.g., amorphous, semi-crystalline, copolymerized) having different shapes (e.g., microspheres, plates, cylinders) in aqueous or liquid buffer solutions, which simulate biological fluids. Regarding water in the vapor phase, like the one that can affect the polymer features during storage, only a few studies have been focused on the PLA hydrolysis, and they generally do not consider PLA produced on a large scale. Interestingly, Rocca-Smith et al. [33] stored PLA films at 50 °C in different conditions of relative humidity and showed that they underwent hydrolysis in different ways. In fact, they demonstrated that, when the relative humidity reached a value of 50%, the molecular weight of PLA did not change as a function of time, but when the relative humidity reached the value of 100%, a decrease in molecular weight from 70 K to 10 K was observed in 70 days. It can be noticed that, in this case, the result is the same obtained by maintaining the film of PLA in liquid water. This paper effectively evidenced that the conditions of pellets storage have a strong impact also on PLA recyclability.

Moreover, reprocessing of PLA induces an increase in crystallization during cooling by increasing the number of injection molding cycles. This can be explained considering the higher chain mobility due to chain scission occurring during injection. Several solutions have been studied to avoid this problem. The main scope was to minimize the PLA thermal degradation by the following two main routes: reinforcement by obtaining biocomposites and use of a free radicals' stabilizer. Badia et al. [34] studied the effect of five successive injection molding cycles on the properties and structure of amorphous PLA. Despite there being no evidence of significant changes in the structural properties of the functional groups, a remarkable reduction in PLA molar mass was observed. The presence of a cold-crystallization transition during DSC and DMTA measurements, showing an enthalpy that increased by increasing the number of reprocessing steps, suggested the occurrence of PLA chain scission due to thermo-mechanical degradation.

Bruster et al. [35] investigated the mechanical recycling by reprocessing of PLA and identified acrylated poly(ethylene glycol) (acryl-PEG) as a reactive plasticizer of PLA by a multiscale approach in the case of a lab-developed grade of plasticized PLA. The authors observed that in up to five successive processing cycles, including extrusion and compression-molding, the tensile and impact properties drastically decreased. Chemical and rheological analyses revealed that repeated processing caused PLA chain scission, its crystallization, damaging of the inclusions, a decrease in the size of poly(acryl-PEG) phases

inside the inclusions, and PLA cracking. Regarding this worsening of properties, inclusion damaging and matrix cracking are believed to be mainly responsible for the embrittlement of plasticized PLA after multiple reprocessing, which generally makes it not suitable for being reused with a closed or open loop approach and, thus, for its starting application.

Successively, Beltran et al. [36] studied the mechanical recycling of commercial grade PLA that was melt compounded and compression molded. They adopted two different recycling processes. The first one consisted of two different steps in accelerated ageing and melt processing step; the second recycling process included the three different steps of accelerated ageing, a washing process and a second melt processing step. Molecular weight changes were investigated by intrinsic viscosity measurements. The obtained results indicated that both recycling processes induced degradation in PLA. This degradation was more extended in the sample undergoing the washing process. The optical, mechanical and gas barrier properties of PLA were not significantly affected by the degradation undergone during the different recycling processes. Thus, these findings suggested that, albeit with PLA degradation, the impact of the different simulated mechanical recycling options on the final properties was limited and, by properly setting the recycling procedure, the chain scission of the PLA could be controlled. As a consequence, the possible use of recycled PLA in applications related to packaging is not jeopardized.

In the review published by Castro Aguirre et al. [28], the end-of-life scenario for this polymer was considered across the board, considering also the polymer footprint and Life Cycle Assessment approach. Moreover, Piemonte [37], thanks to the Life Cycle Assessment approach, showed for both starch and PLA -based biobased materials and by using different recycling methods (for instance, composting, incineration and anaerobic digestion) that, compared with the mechanical recycling process, the other methods, e.g., chemical recycling, are clearly underperforming from an environmental point of view.

The analysis of the scientific literature has certainly suggested that several methodologies can be adopted to recycle PLA to obtain products for specific industrial sectors and applications. The recycling by melt processing of plastic waste consisting of PLA to produce biodegradable fibers by melt spinning was studied by taking into account several parameters like melt spinning quality, mechanical, structural, thermal and morphological features as well as the dyeing properties of the obtained fibers [38]. Thanks to this systematic analysis, it is possible to state that there is the possibility of carrying out the melt spinning of PLA fibers from recycled waste to obtain yarns with acceptable properties. Looking at the investigated parameters, it was observed that the drying pretreatment undergone by the recovered PLA flakes, the melt extrusion temperature and the drawing operation were fundamental for tailoring both structural and mechanical properties of the recycled fibers made of PLA.

It was suggested that, thanks to the drying, the PLA hydrolysis can be limited or fully overcome. Additionally, chain extenders are reported to allow the melt viscosity of PLA and its blends to be efficiently modulated, as shown by Najfi et al. [39]. These co-authors investigated three specific chain extenders (polycarbodiimide (PCDI), tris (nonyl phenyl) phosphite (TNPP) and Joncryl ADR 4368) in two matrices (PLA and PLA-based nanocomposites containing 2 wt% clay). The authors found that the mechanism of stabilization was a reasonable chain extension that resulted in the formation of longer linear chains nanocomposites and a long-chain branched (LCB) structure in Joncryl-based nanocomposites. The authors found that Joncryl was the most efficient chain extender among those used in this study. Despite this work evidencing that reactive blending and extrusion can be very effective tools in the hand of a recycler, in considering mechanical recycling, it is important to notice that it is necessary to have an uncontaminated starting material. In fact, the contamination can be due to two different reasons: (1) additives yet blended in the PLA matrix or (2) additives present as separated layers. In the former case, generally the compatibility between the PLA matrix and the additive is good. Nevertheless, in the latter case, the successive processing must consider and implement a compatibilization strategy. In a similar way, cellulose esters were used as compatibilizer in wood/PLA composites [40]

or PLA/poly(propylene) (PP) blends [41]. The addition of biobased additives alternative to Joncryl was attempted by Coltelli et al. [42], which used a mixture of epoxidized soy bean oil and succinic acid to induce an increase in melt viscosity in PLA/poly(butylene succinate-co-adipate) (PBSA) blends and in composites of the same blend containing wheat bran. The effect as a function of the additives concentration showed some limits, e.g., if the stoichiometric ratio between -COOH and epoxide groups was higher than one, the chain scission predominated, probably because of the autocatalytic nature of polyester hydrolysis. This investigation showed that bran is a natural filler that induces chain scission in PLA, probably because of its consistent starch fraction. In fact, starch was reported to induce extensive chain scission in PLA [31,43–45]. Other natural fillers, like hazelnut shell powder, that substantially do not contain starch, showed only a limited impact in regard to PLA chain scission [46,47].

The potentialities of biopolymers as additives to modulate PLA melt viscosity was investigated by Beltran et al. [48,49]. In fact, they added chitosan or silk fibroin to PLA, and they observed an increase in the intrinsic viscosity of this polymer. Thus, the authors suggested that the addition of these biopolymers can be effective for modulating the melt viscosity of PLA-based materials during their reprocessing. The authors explained their results considering that the presence of -NH- amidic or amine groups can block the reactivity of -COOH groups thanks to an acid base reaction, thus limiting the occurrence of chain scission. As stated by the authors, previously they have added in PLA halloysite modified with an aminosilane [50], and they have similarly observed an increase in viscosity reasonably due to a similar mechanism. As a consequence, an alternative route for both reinforcing and increasing the melt viscosity of PLA was suggested to be the addition of a nano-inorganic filler, as Scaffaro et al. [51] have added nanostructured hydrotalcite to PLA. However, they have found that its reinforcing effect due to the filler was counterbalanced by the chain scission that the filler induced on the polymer. In particular, the addition of clay [52] was reported to be suitable for improving the PLA mechanical and barrier properties. On the other hand, reprocessing the nanocomposite certainly resulted in some chain scission. An increase in transparency was also observed, and it was correlated to the better dispersion of clay achieved thanks to the further processing of the PLA nanocomposites. Figure 2 summarizes the described strategies adopted to counterbalance PLA chain scission.

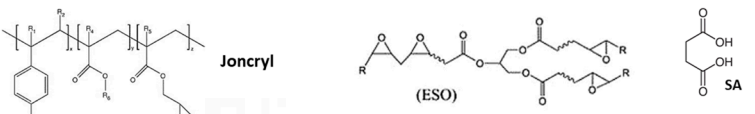
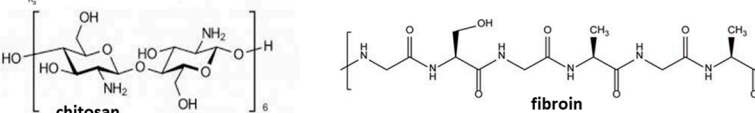
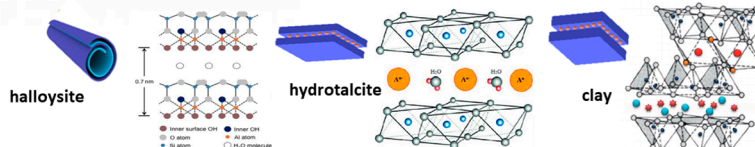
Strategies against PLA chain scission	Tested additives	Reference
Addition of not renewable or renewable chain extenders		Najafi et al. 2012 Coltelli et al. 2021
Addition of proton acceptor biopolymers		Beltran et al. 2021 and 2019
Addition of nano-inorganics to obtain melt reinforced nanocomposites		Beltran et al. 2018 Scaffaro et al. 2017

Figure 2. Strategies adopted to counterbalance PLA chain scission effects in the literature with corresponding references [39,42,48–52].

2.2. Chemical Recycling

Interestingly, it was demonstrated [53] through an LCA approach that the production of lactic acid by the depolymerization of PLA should be preferred, as it is convenient from an energy point of view, with respect to the production of lactic acid based on the

fermentation of glucose. As reviewed by McKeon [54], the chemical recycling of PLA can be obtained by three main methods. These three methods are hydrolysis, alcoholysis, and pyrolysis. The hydrolysis of PLA produces lactic acid, and this specific process could potentially be more cost effective than the sugar fermentation. In hydrolysis, PLA is treated with water at a high temperature to yield lactic acid, which can be readily polymerized to high molecular weight PLA. Gironi et al. [55] investigated the PLA solubilization behavior while considering two different organic solvents (acetone and ethyl lactate). They considered different water concentrations with the aim of optimizing the PLA chemical depolymerization process. The obtained results suggested that acetone-based solvents (for instance acetone/water mixtures at different concentrations) can more effectively solubilize PLA rather than the ethyl-lactate. Nevertheless, an increase in water concentration in the solvent phase determined both a reduction in the solvent power and, for the two solvents tested, a reduction in the mass transport coefficient. Iñiguez-Franco et al. [56] investigated the PLA hydrolysis in water–ethanol solutions, performing experiments in 50% ethanol at 40, 60, 70 and 80 °C. Thanks to this study, the kinetic parameters of the hydrolysis, rate constant and activation energy were assessed. For 50% ethanol, no dependence of the hydrolysis rate on pH was noticed. To better understand the effect of temperature on the PLA hydrolytic degradation in water, hydrolysis experiments were also carried out at higher temperatures (up to 90 °C). The final activation energy values for the PLA hydrolytic degradation in 50% ethanol and in water were 9.627×10^4 and 10.474×10^4 J/mol, respectively. These values are in the same range of those that can be found in the literature. Interestingly, the activation energy was 9% higher when PLA was hydrolyzed in water than in 50% ethanol. Thus, this study suggested that using 50% ethanol can be advantageous.

The alcoholysis of PLA was also investigated. Payne et al. [57] developed Zn(II) complexes to promote the degradation of PLA aimed at producing methyl lactate (MeLA) thanks to the use, under mild conditions, of methanol. Majgaonkar et al. [58] have studied the ethanolysis of PLA in acetone at 50 °C and 1 bar as pressure using 1,5,7-Triazabicyclo[4.4.0]dec-5-ene (TBD) as a catalyst. The optimal molar ratio of alcohol per mole of PLA repeating units was found to be 3, and it was understood that the yield of ethyl lactate did not depend on the polymer molecular weight. In addition, the process resulted to depolymerize various post-consumer PLA substrates without showing a significant effect on the ethyl lactate yield. Therefore, the process is both robust and flexible and can provide as product a valuable and green solvent.

Plichta et al. [59] investigated the controlled chain scission of commercial poly(lactic acid) by using the following bi or polyfunctional small molecules: dipentaerythritol, diols, diamines and oligo(ethylene glycol), adipic acid or polyester diols. They evidenced that this method is robust and allows PLA chemical recycling by a controlled chain scission catalyzed with protic compounds. These processes proved to be efficient. Moreover, in the case that a catalytical amount of tin(II) octanoate was used, products like homo- or block-diol-type copolymers or lactic acid oligomers were observed. Diamines proved to be the most efficient, particularly when a catalyst was not used and at quite low temperatures. In contrast, adipic acid at 200 °C reacted only partially. Nevertheless, the molecular weight of the products was much lower than that of PLA. An important issue proved to be persistency of degradation agents in the products' structures. Products consisting of ABA-type triblock copolymers can be obtained by the same chain scission method utilizing macrodiols. PLA degradation with macrodiols can be performed at temperatures of 120 °C in solvent (for instance xylene) or in bulk, as well as in an extruder. Aliphatic copolymers were monophasic, whereas those containing aliphatic and aromatic macrodiols were biphasic. The latter ones showed different morphologies in dependence on composition. In this process, it was observed that the reaction duration strongly depended on temperature, and, generally, the time of reaction was in the range from 30 min to 4 h. Nim et al. [60] investigated the use of microwave-assisted alcoholysis of PLA using ethylene glycol (EG), propane-1,3-diol (PDO), and butane-1,4-diol (BDO). They used tetrabutyl ortotitanate (TBT) as a catalyst and adopted PLA/diol ratios from 1:1 to 4:1 wt/wt. The yields in

higher molecular weight products increased with an increase in the ratios. In contrast, yield decreased with an increase in the ratios from 1:1 to 4:1 wt/wt. The products of the process were successively used polyols in the production of lactide-based polyurethanes thanks to the reaction with 1,6-diisocyanatohexane (HDI) and by using 1,4-butanediol as a chain extender.

Thus, processes leading to intermediate molecules, such as oligomers, were proposed. For instance, the recycling of PLA at high temperatures in the presence of polyols was proposed [44]. These oligomers produced chain scission by reacting with PLA macromolecules. As a result, low molecular weight oligomers with hydroxyl end-groups were formed. The occurrence of the reaction between PLA and multifunctional alcohols, for instance glycerol, glucose, sorbitol and starch, was evidenced by spectroscopic and chromatographic techniques. The resulting amphiphilic products were used as possible compatibilizers for PLA/starch blends. Results coming from several different techniques evidenced that the obtained oligomers improved the interfacial adhesion and enhanced the compatibility and flexibility of both melt-blended and extruded composites consisting of PLA and starch. It was, thus, demonstrated that PLA can be recycled, transforming it into low-molecular-weight compatibilizers. Moreover, it should be noted that this one-pot method required no solvent or catalyst. These features may predict good possibilities for an easy up-scaling.

Pyrolysis was based on heating a material at its thermal degradation temperature under inert atmosphere.

Undri et al. [61] performed the pyrolysis assisted by microwave (MAP) of PLA by using different microwave absorbers (like tire powder, carbon or Fe), different apparatus set-up, and considering the effect of microwave power. They obtained liquid rich in 3,6-dimethyl-1,4-dioxan-2,5-dione (lactide) and organic compounds containing oxygen. From the collected liquids, the pure form of L-lactide was separated. The meso-form of lactide was also present in the liquid and identified through GC-MS analysis. In this process, up to 27.7% of lactide was formed. Furthermore, in recovered liquids, simple acids such as acetic acid and propionic acid (up to 17.1%), carbonyl compounds, and fragments of PLA backbone were also present. The researchers evidenced that the pyrolysis of PLA in the presence of other polymers able to give aroma biocompounds by pyrolysis, such as polymers present in tires, gave cross reactions in the course of the pyrolysis because they can affect both yield and characteristics of the obtained liquid. Interestingly, it was found that aromatic compounds formed from tire pyrolysis prevented the lactide precipitation. More recently, Saeang et al. [62] have found that, during pyrolysis of PLA, the MgO catalyst led to high coke formation. Lactide, lactic acid, and propanoic acid constituted examples of valuable chemicals recovered from the pyrolysis of PLA. The main product of catalytic pyrolysis with zeolite used as catalyst at 400 °C was lactide (up to 79%). The different discussed methodologies of chemical recycling are summarized in Figure 3.

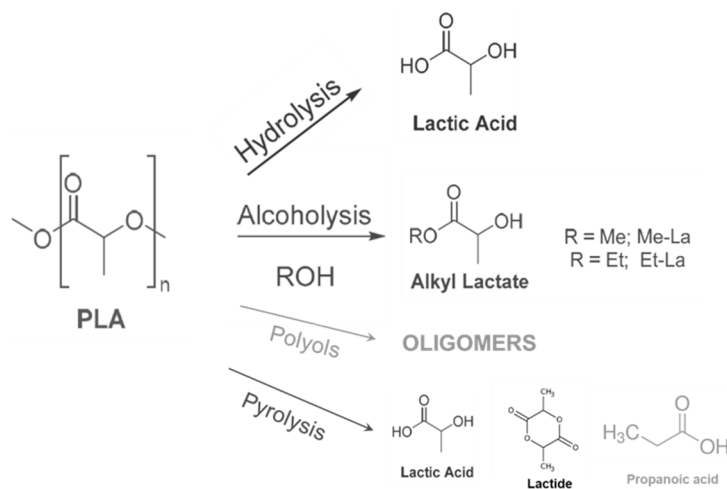


Figure 3. Scheme regarding the chemical recycling of PLA.

3. Recycling of Other Biodegradable Bioplastics

3.1. Poly(Hydroxy Alcanoate)

Poly(hydroxy alcanoate) (PHA) is a class of polymers produced by microorganisms that is easily biodegradable in the environment and in the most hostile conditions. PHB is the most investigated polymer belonging to this class. Vu et al. [63] reviewed the scientific works dedicated to the recycling of PHA. They described the methodologies of mechanical recycling and chemical recycling, and they noticed that research on mechanical and chemical recycling is dominated by two techniques: extrusion and pyrolysis. Despite extended research regarding gasification being almost absent in the literature, it could potentially contribute to the close loop recycling of PHA-based products by the conversion to syngas. In fact, the latter can be used in biological recycling undergoing anaerobic digestion. Volatile fatty acids (VFAs), produced as intermediates, can be used for the production of new polymers. Research gaps exist, especially in regard to the relationships between polymer structure, composition and recycling effectiveness. This aspect is fundamental, as it will influence the need for separation/sorting of the recovered bioplastic items.

Regarding the mechanical recycling, Rivas et al. [64] investigated the trend of mechanical and thermal properties, as well as chemical structural changes, of poly(hydroxybutyrate) (PHB) when undergoing up to three extrusion cycles. An evident decrease in mechanical properties was observed already at the second extrusion cycle. A decrease above 50% in the third cycle was also obtained. Moreover, an increase in the crystallinity index was observed, as crystallization is favored by chain scission. Despite these changes in mechanical properties being observed, detectable changes in the chemical structure or in thermal stability of PHB could not be revealed by using infrared spectroscopy and thermogravimetric analysis, suggesting an important impact of molecular weight on the properties worsening. Bonnenfant et al. [65] obtained similar results, but they used a catechin biobased anti-oxidant that was added to PHBV during extrusion. They obtained a material with a lower temperature of thermal degradation. This is an example of the use of natural anti-oxidants as stabilizers of bioplastics during their reprocessing.

Chemical recycling of PHAs by thermal degradation was investigated by Ariffin et al. [66]. It is well known that PHB produces various kinds of degradation end-products. In particular, monomers such as 3-hydroxybutyric acid from enzymatic hydrolysis and crotonic acid from thermal degradation are produced. The PHA thermal degradation resulted in its conversion into vinyl monomers by using alkali earth compound catalysts. Interestingly, it was found that poly(3-hydroxybutyrate-co-3-hydroxyvalerate)s (PHBVs) were selectively depolymerized into crotonic and 2-pentenoic acids in the presence of CaO and Mg(OH)₂ as catalysts at lower degradation temperatures. Then, the crotonic acid from 3-hydroxybutyrate sequences in PHBV was successively copolymerized with acrylic acid for producing, as new products, water-soluble copolymers, i.e., poly(crotonic acid-co-acrylic acid). This polymer shows a high glass-transition temperature. This copolymerization is an intriguing example of cascade reutilization or chemical recycling of PHAs.

3.2. Poly(Butylene Succinate)

Currently, Poly(butylene succinate) (PBS) is available partially or fully from renewable sources. It has shown a better processability than PHB, as its processing temperature range is less narrow, and it shows a shiny look and flexibility [67]. The potentialities of PBS recyclability were investigated by Georgousopoulou [68] by carrying out consecutive extrusion cycles, setting different temperature profiles so that reprocessing intensive conditions were simulated. The authors have concluded that the reprocessing of PBS at temperatures higher than 190 °C induced reactions consisting of branching/recombination as well as chain scission. These reactions resulted in extrudates of higher viscosity, having a bimodal distribution of the molecular weight. If common stabilizers consisting of hindered phenols were added, the thermo-mechanical degradation of PBS was significantly suppressed. This specific effect clearly suggested the radical character of the degradation reactions. It was observed that a concentration of stabilizers at about 0.1% was enough to efficiently

maintain polymer properties upon reprocessing. However, the higher concentration of 0.5% induced a decrease in the quality of the extrudates. The prepared PBS (degraded and stabilized) showed similar melting and degradation temperatures than virgin material, showing its possibility for mechanical recycling. However, the induced degradation resulted in accelerated melt crystallization and a lower degree of crystallinity. The authors attributed this behavior to the nucleating effect of the branches formed because of slight radical crosslinking. Accordingly, the addition of stabilizers restricted the increase in the melt crystallization rate.

Kanemura et al. [69] investigated the potentialities in terms of processability of PBS as a promising biodegradable and reprocessable material. PBS was immersed in water. The bending strength of PBS was measured, and it was found that it decreased as the immersion time and the immersion temperature increased as a consequence of the hydrolysis responsible for the PBS chain scission; the obtained PBS was thus reprocessed. Unexpectedly, an increase both in the PBS bending strength and molecular weight was observed. The observed increase was explained considering that PBS macromolecules may have an autocatalytic action during the reprocessing by esterification. It was hypothesized that dicarboxylic acids were produced from PBS during the chemical degradation, which ended up in condensation, resulting in PBS with a higher molecular weight. Interestingly, this characteristic of molecular weight increase observed for PBS after reprocessing was not observed for poly(lactic acid) (PLA), one of the most frequently used biodegradable plastics.

3.3. Poly(Butylene Adipate-co-Terephthalate)

Poly(Butylene adipate-co-terephthalate) (PBAT) is a biodegradable polymer that is synthesized by fossil butandiol (BDO), terephthalic acid and adipic acid. Furthermore, the partial conversion regarding the use of fully biobased sources is ongoing. In fact, BDO has been obtained through industrial biological fermentation so that fossil BDO can be replaced. Then, other similar polymers based on sebacic acid, as a substitute of adipic acid, coming from castor oil, have been used as a monomer to prepare poly(butylene sebacate-co-butylene terephthalate) (PBSeT) copolyesters. Moreover, 2, 5-furandicarboxylic acid (FDCA) has been considered as one of the most promising bio-based aromatic monomers. Hence, in a few years it will be possible to have semiaromatic fully biobased polymers [70]. The partial biobased commercial PBAT was considered in several applications, because it is flexible and rubbery-like at room temperature. Thus, it was found suitable for toughening the more brittle PLA [71]. Regarding its recyclability, it was found that the commercial polymer with a molecular weight of about 25,000 undergoes chain scission upon melt processing, but in a less relevant way with respect to PLA [29]. A blend based on PBAT, containing PLA, was reprocessed both after pre-drying and without pre-drying by single screw extrusion (temperature in the range 150–180 °C) and the rheological characterization suggested that, if the sample was processed after drying, a less extensive degradation was observed [72]: in the samples reprocessed after drying, branching in the PBAT component became slightly predominant with respect to the chain scission. On the other hand, the opposite was observed for those samples that were reprocessed without a preliminary drying. Nevertheless, five extrusion steps did not significantly worsened the mechanical properties of the blend in both conditions, thus demonstrating the feasible mechanical recyclability of PBAT-based blends.

In general, Scaffaro et al. [73] observed mechanic—chemical degradation of several bioplastics within film processing due to the high surface to volume ratio of these manufactures. This effect suggested some limitations to their ultimate performance and to the possibility of recycling. Nevertheless, the strategy of anti-oxidant addition showed promising potentialities in limiting this kind of degradation.

4. Recycling of Starch-Based Materials

Materials based on starch are widely employed, especially in pouches and bags. The material consists of starch (consisting in turn of amylose and amylopectin) blended with

biodegradable polymers and plasticizers. The main component of the material is therefore starch, but other biobased polymers or molecules are present in the different formulations available on the market. Accinelli et al. [74] studied the degradation of carrier bags in soil, compost and two aquatic ecosystems (a littoral marsh and seawater). Carrier bags were rapidly deteriorated in soil and compost. In contrast, little deterioration was observed for specimens that were buried in soil under field conditions or exposed to water of a littoral marsh and of the Adriatic Sea. Interestingly, the authors demonstrated that end-of-life carrier bags can be effectively recycled to produce lactic acid by the fungus *R. oryzae*, so by exploiting biotechnology for producing a valuable molecule to be used, for instance, for producing PLA.

Saieh et al. [75] studied the effect of low concentrations of cellulose nanofibers and nanoclay particles on physical, mechanical, and morphological properties of biodegradable composites made of recycled thermoplastic starch (TPS) and mixed industrial sawdust coming from Iranian wood species, including walnut, hornbeam, and rush. The results showed an improved tensile modulus, tensile strength and flexural modulus. The authors demonstrated that water absorption and swelling were reduced by about 20%. When these biocomposites were exposed at high temperatures, nanoparticles conferred a greater stability to the composite structure. Thus, it was demonstrated that cellulose nanofibers and nanoclay particles can be successfully used as additives for improving the physical and mechanical characteristics of the biodegradable composites made of thermoplastic starch and industrial sawdust.

Thermoplastic starches were obtained from different sources, and they were used to prepare TPS/poly(ethylene-co-vinyl acetate) (EVA) blends (having a content of TPS of 65%) compatibilized with polyethylene grafted with maleic anhydride [76]. The impact of multiple processing cycles, mimicking possible reprocessing, on the structure and performance of prepared blends was determined. An increase in the processing time of multiple extrusion induced an increase in the yield of the reaction between maleic anhydride and starch hydroxyl groups. It was observed that this reaction also had a noticeable effect on the mechanical performance of materials. As a consequence of the increasing in compatibility with each processing cycle, both the tensile strength and elastic modulus of TPS/EVA blends also increased. This specific effect was also corroborated by SEM characterization that suggested a more homogenous structure of the analysed blends as a consequence of a longer processing time.

The mechanical reprocessing of PBAT/starch blends was strongly affected by hydrolytic degradation; therefore, a commercial epoxide functionalized chain extender was used to control it [77]. In contrast, blends based on polypropylene/PBAT/starch proved to be suitable for several processing cycles [78], but their end of life proved to be problematic. In fact, they were not, for instance, biodegradable, if tested by exploiting a specific and very effective mushroom-based biotechnology [79]. Degradation of PLA/PBSA blends containing starch as a minor additive was also observed [9,45]. In general, the mechanical recycling of starch-based materials seems highly problematic because of the severe degradation they undergo during use and because of the chain scission they induce in biopolyesters.

5. Recycling of Blends

The polymer that is currently the most widely available on the bioplastic market, and which is biobased and industrially compostable, is PLA. The recycling behavior of PLA blends has been considered interesting by many researchers.

The recyclability of PHA biopolymers like poly(hydroxy butyrate-co-valerate) (PHBV), PLA and PHBV/PLA blend was studied through repeated reprocessing cycles (up to 6) [80]. The morphology and the structure of PHBV, PLA and PHBV/PLA were affected in different ways by repeated reprocessing cycles. Considering their stability as a function of the number of processing steps, PLA proved to be less sensitive to reprocessing cycles with respect to PHBV, which was significantly degraded. Thus, PHBV degradation occurring during reprocessing was revealed by a large decrease in the molecular weight and viscosity

attributable to chain scission. This work highlighted that the thermo-mechanical PHBV chain scission was significantly limited in the presence of PLA. As a consequence, the mechanical properties of PLA and PHBV/PLA blends are less affected after six cycles with respect to neat PHBV. The latter showed a significant decrease in these properties. These results were confirmed by Plavec et al. [81], who studied the multiple reprocessing of a commercial PLA/PHB 45/55 blend. The authors suggested that the reprocessing is possible up to 11 times.

Su et al. [82] evidenced that the main destination of PLA/PBS blends should be composting. Additionally, for recovering materials, chemical recycling can be an effective method. In fact, in chemical recycling, polymer chains are broken down into small molecules (for instance monomers), which can then be re-fed to polymerization reactions. Tsuneizumi et al. [83] have investigated and carried out the chemical recycling of PLA/PBS blends using two different routes. The first route consists of the separation of PLA and PBS by exploiting their different solubility in toluene. In the other route, the sequential degradation of PLA/PBS blends using a lipase first to degrade PBS into cyclic oligomer was considered. The obtained oligomers were then repolymerized in order to produce new PBS. Moreover, PLLA was degraded into repolymerizable lactide oligomers. The mechanical recyclability of PLA/PBSA blends was studied by Coltelli et al. [84], who showed that it was possible to control the decrease in melt viscosity by using commercial fossil-based chain extenders with epoxydic groups (Joncryl) as additives or by using alternative renewable chain extenders. These compounds were based on epoxidized soybean oil and malic or succinic acid. Although a reference commercial fossil chain extender proved to be much more effective, this novel bicomponent renewable chain extender seemed quite promising. In fact, it proved to be more effective on composites containing natural fibers than on PLA/PBSA blends. The interest in poly(lactic acid)/poly(butylene succinate-co-adipate) (PLA/PBSA) blends is the consequence of their suitability for film packaging applications. In fact, in some compositional ranges, these blends proved to be flexible, resistant, and compostable. A significant study made regarding industrial PLA/PBSA films was also carried out that suggested good recyclability for these films [84]. These films were reprocessed through extrusion in order to simulate recycling, and they successively characterized in terms of their melt flow rate, which was studied as a function of PBSA content. In this work, films produced by extruding the PLA/PBSA 60/40 blend were reprocessed several times by injection molding. The variations in melt fluidity and thermo-mechanical properties proved to be negligible in up to three injection molding cycles. This result was observed for both the PLA/PBSA blend and pure PLA granules. For the blend, the change in color (evaluated in colorimetric studies as yellowing and darkening) was more evident (Figure 4). Moreover, some slight local compositional changes in injection-molded items could be evidenced. Interestingly, a slight decrease in the crystallinity of PBS as a function of the number of injection molding cycles was also observed. Thus, it was concluded that this PLA/PBSA blend can be effectively recycled by extrusion or injection molding before being composted.

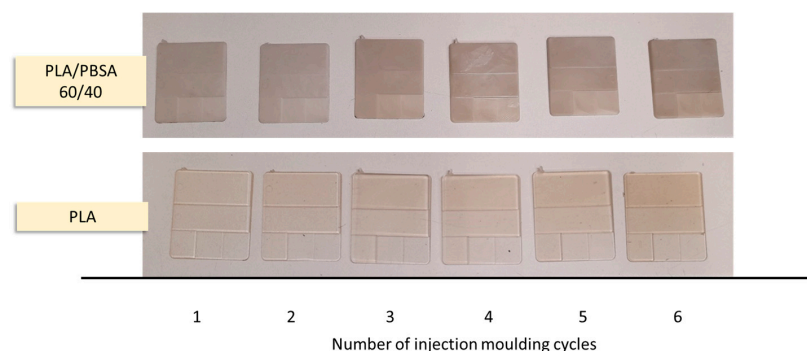


Figure 4. Comparison of color change as a function of number of injection molding cycles for PLA/PBSA blend (**top**) and for pure PLA (**bottom**).

6. Recycling of Natural-Fiber-Reinforced Composites

Natural-fiber-reinforced composites are interesting because they represent the declination of engineering composite materials, characterized by a high modulus and low density, in their biobased version, and also because these materials offer the possibility of valorizing fibers coming from agro-food-forest waste, thus providing a concrete opportunity for the application in industrial cycles of circular economy principles.

Lopez et al. compared the properties of three biodegradable matrices with the ones reinforced with cellulose fibers, in particular softwood chemi-thermo-mechanical pulp (CTMP) [85]. The effect provoked by repeated injection-molding of three biodegradable matrices (PLA, a commercial aliphatic polyester and starch-based Mater-Bi) was studied, and it was found that, by successive reprocessing steps, tensile, flexural and impact strength decreased. The decrease that was observed in mechanical properties has been attributed to the matrices' degradation, as confirmed by the melt flow and molecular weight results. Despite reprocessing not showing a significant effect on glass transition temperature, it diminished the degradation temperature and melting point of polymers. The authors stated that neat PLLA could undergo to up to five reprocessing steps without a drastic loss in mechanical and thermal properties. Moreover, they stated that the aliphatic polyester could be recycled by up to 10 times. On the other hand, for post-consumer starch-based Mater-Bi, the composting was suggested by the authors, as the recyclability is very poor for this material. The reprocessing of composites reinforced with softwood chemi-thermo-mechanical pulp (CTMP) respected the tendencies observed for the neat matrices. Whilst CTMP behaved mainly as a filler in PLA, reinforced thermoplastic starch-based composites showed improved recyclability and mechanical properties.

In general, the addition of natural fibers was found to be detrimental for the biocomposites in the biopolyester matrix, because natural fibers, being rich in hydroxylic groups due to their mainly polysaccharidic composition, can induce a chain scission on polyester. Moreover, they are generally hygroscopic, so they can contain some humidity that can induce hydrolysis in biopolyesters during melt processing. Accordingly, Costa et al. [86] noticed that two different types of filler, obtained from two different layers of the babassu palm fruit (mesocarp and epicarp), greatly promoted incipient degradation of the PHB/PBAT blend when they were processed in an internal laboratory mixer. The most heterogeneous and anisometric filler was the one inducing the highest level of chain scission. Hence both chain scission and shape effect-induced degradation were evidenced. Similar results regarding hydrolysis due to natural filler were noticed in PLA/PBSA blends reinforced with wheat bran [42]. To counterbalance the observed degradation, biobased chain extenders were prepared using epoxidized soybean oil (ESO) and dicarboxylic acids (DCA), in particular malic acid (MA) and succinic acid (SA). It was observed that the mechanical properties were not significantly affected by the different chain extension systems. Moreover, in terms of melt fluidity reduction, the commercial Joncryl proved to be more efficient (Figure 5a), and the investigated biobased chain extenders could be promising for the processing of biobased composites reinforced with natural fibers. Additionally, they could be also considered in the future as effective additives to allow for biocomposite recycling. In fact, it should be noticed that the use of non-biobased chain extenders can be detrimental for the biodegradability of the material. In contrast, thanks to this fully biobased approach, biocomposites could be processed and recycled, and then, in their final stage of life, they could be fully composted, thus overcoming the issue of generating possible residues of microplastics.

The presence of bran makes the system SA+ ESO more effective, and this enhancement in reactivity was explained by considering the reaction between dicarboxylic acid and bran and the further reaction with epoxide groups of ESO that can result in the production in the melt of extended macromolecular systems (Figure 5b). Interestingly, the results obtained by replacing bran with hazelnut shell powders (HSP) [47] showed that the composite degradation was less affected by the presence of the filler, probably because of the different composition of the HSP, which contained less starch than bran. In a wider concentration

range, the ESO/SA system proved to be more effective than in HSP composites. In the latter biocomposites, malic acid was more effective than succinic acid. Again, the differences can be tentatively attributed to the yet underlined different chemical compositions of these two agro-waste-based fillers. In general, PLA composites containing HSP could be also used in additive manufacturing, resulting in an advantageous decrease in the porosity of prepared specimens [87].

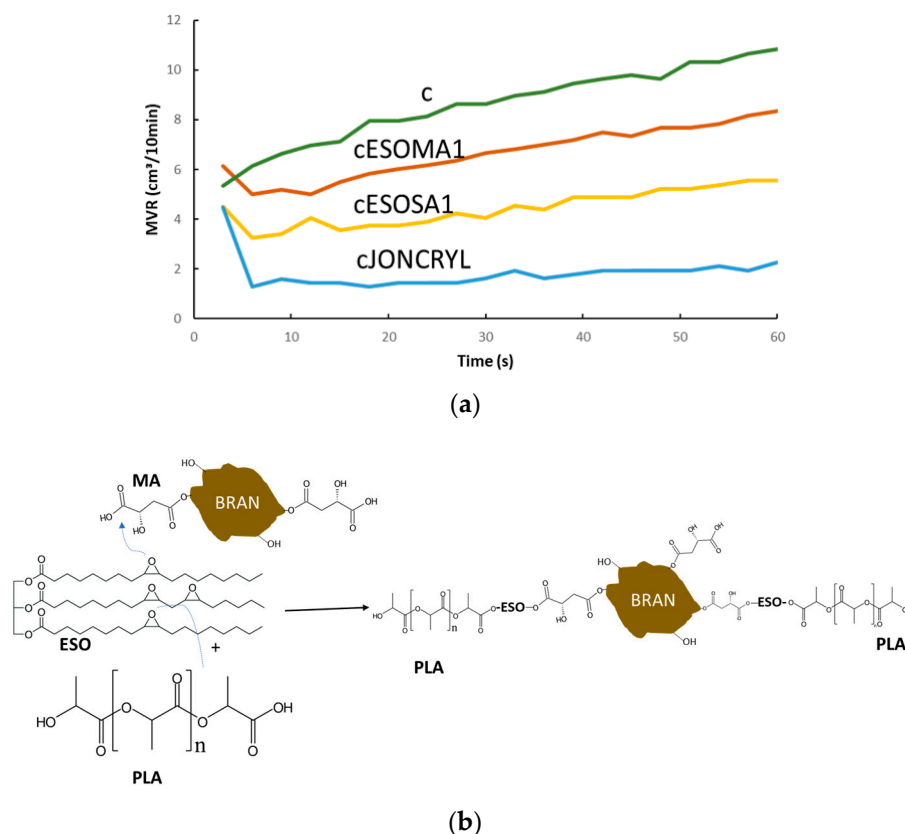


Figure 5. Reproduced from reference [42] by courtesy of MDPI. **(a)** Trend of Melt Volume Rate, depending on melt fluidity, as a function of time for PLA/PBSA composites reinforced with 20% of wheat bran. Legend: c = composite // cESOMA1 = composite containing ESO and MA // cESOSA = composite containing ESO and SA // cJONCRYL = composite containing Joncryl commercial non biobased chain extender; **(b)** scheme explaining the effect of bran in potentiating the reactivity of malic acid/epoxidized soybean oil chain extending system.

A high content of cellulose fibers was found to be detrimental in PLA composites tested in six successive injection molding cycles. In particular, composite material with 20 wt% of recycled content could withstand six cycles relatively well, while the composite with 50% of fiber degraded much more quickly [88].

The mechanical recycling of biocomposites based on PLA and flax fiber was investigated by Duigou et al. [89]. After mechanical recycling, the tensile properties at different fiber contents (20% and 30% by weight) were conserved until the third cycle. The reprocessing by injection molding resulted in lower molecular weight, reduction in fiber length and separation of fibre bundles. However, the biocomposite properties after three cycles indicated a promising acceptable recyclability of this material. Ngaowthong et al. [90] prepared biocomposites, incorporating sisal fibers in PLA, and they observed that the PLA matrix remarkably increased its crystallinity by the addition of fibers and with repeated recycling. They also observed that, by increasing the amount of fibers, water absorption was increased. In contrast, water absorption was reduced by increasing the recycling time.

In general, all these papers showed that the content of cellulosic fibers is an important parameter to be considered in the preparation of biocomposites.

Grozdanov et al. [90] have studied the possible recycling of biocomposites of PLA reinforced with rice husks and kenaf fibers. Milled recycled biocomposites (milled size of 0.050 mm) were used in polymer mortars and concrete structures for the low-cost building industry.

7. Conclusions

The possible separation of bioplastics can allow their recycling through mechanical or chemical recycling. Mechanical recycling can be considered the best option because it does not require specific industrial facilities, but it can be affected by degradation occurring during reprocessing. The effect of degradation is generally different for biopolyesters on the basis of their structure, molecular weight and operative conditions. However, currently, a systematic understanding of all these variables is not fully available.

It is clear that materials containing starch (also as a component of natural fibers) are more degradable than neat biopolyesters. Biocomposites undergo a chain scission with an extent depending on natural fiber content, composition of the filler, shape and humidity content.

In all the cases, mechanical recycling can be facilitated by the use of chain extenders and anti-oxidants during reprocessing, but it will be important to develop new biobased version of these additives, because they often are not yet commercially available.

Chemical recycling also seems possible, and this approach offers the possibility of obtaining several chemicals, ranging from monomers to oligomers.

On the whole, the present chapter suggests that the use of biobased and biodegradable bioplastics allows for a more sustainable end of life with respect to fossil alternatives. In fact, in several cases they can be chemically or mechanically recycled, and, when this recycling approach is no longer possible, they can be finally composted.

Instead, fossil plastic can be recycled, but after several cycles, and when the properties of the material are no longer suitable for any applications, they should be incinerated or landfilled. Thus, especially for non-durable and widely diffused products (packaging, sanitary, personal care, etc. . .), and to replace multi-material fossil products, biodegradable bioplastics and biocomposites could be a valuable option. The future perspectives in research about recycling biobased and compostable polymers will address several topics, such as the possible investigations of renewable stabilizing systems for reprocessing these biopolyesters and biocomposites, more deep studies on the degradation paths of biopolymers during reprocessing, as well as a more systematic knowledge of the correlations between macromolecular design, morphology and recyclability.

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