corrosion and materials degradation

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Review

State-of-the-Art Review of Aliphatic Polyesters and Polyolefins Biodeterioration by Microorganisms: From Mechanism to Characterization

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Abstract: As a result of the exponential growth in the production of plastics and their extended degradation period, strong environmental concerns in association with the disposal of plastic waste have emerged. Pursuing sustainable solutions for managing plastic waste has led to significant interest in plastic biodegradation research, with a specific focus on biodeterioration facilitated by microorganisms. The biodeterioration of plastic by microorganisms is a complex phenomenon that can be influenced by a variety of environmental factors such as humidity, temperature, and pH, as well as polymer properties such as molecular structure, molecular weight, and crystallinity. Toward a better understanding of this phenomenon for resolving the issue of plastic waste, this review article focuses on the biodeterioration of synthetic polymers, in particular aliphatic polyesters and polyolefins, through the enzymatic activities of microorganisms. First, the mechanism of polymer biodegradation via enzymatic activity is discussed, followed by the physical properties of polymers and environmental conditions that influence their biodegradability rates. Then, an overview of experimental approaches and standardized protocols used to assess the biodegradability of polymers by these degrading agents is provided. Finally, current developments in employing biodeterioration for the degradation of aliphatic polyesters and polyolefins are reviewed. The review concludes with a discussion on the complexity of biodegradation by microorganisms, the necessity of proper engineering of polymer properties during production to enhance their biodegradability, and the need for further research to discover sustainable and environmentally acceptable alternatives.

Keywords: polymers; biodeterioration; microorganisms; aliphatic polyesters; polyolefins

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1. Introduction

Although the first synthetic plastic was created in 1907, marking the start of the global plastics industry, it wasn't until the 1950s that the manufacturing of plastics saw a significant increase. Annual plastic production has exponentially grown, skyrocketing during the past seven decades [1]. Before 1980, plastic recycling and incineration were practically non-existent, meaning all of it ended up in landfills. From 1980 to 1990 the incineration and recycling rate increased to 0.7% per year [2]. Due to the high processing costs, the majority of plastic waste, including waste from the textile sector, films, plastics, and nonwovens, is not collected and recycled. Plastics have been a source of concern due to their inability to biodegrade, the closure of landfills due to environmental concerns, and the growing number of environmental issues related to water and land contamination. Over the past two decades, the need for biodegradable polymers and the biodegradation of plastic waste, due to the widespread adoption of plastics in life and the associated constraint on waste disposal facility capacity, has drawn significant attention from researchers [3–16].

At the moment, two strategies are being investigated as potential ways to reduce the damage that the use of polymers causes to the surrounding ecosystem [13]. One strategy is to concentrate on the development of polymeric materials with a long lifespan, which must have better durability and preferably be derived from renewable sources [17–19]. Another

strategy focuses on technological advancements made to produce short-life polymers with the purpose of quick biodegradability [20]. This review specifically concentrates on the second strategy, looking at polymer biodegradability through the lens of organisms' enzymatic activities.

Changes in a material's mechanical, optical, or electrical properties, as well as crazing, cracking, erosion, discoloration, phase separation, or delamination, have all been associated with degradation. Some polymers have the potential to serve as growth substrates for heterotrophic microorganisms, such as bacteria and fungi [21,22]. The primary mechanism of polymer degradation involves the scission of macromolecules' side chains or main chains. In a broad sense, the variables that influence the biodegradability of a polymer can be divided into two categories: the polymer's physical properties and the environmental conditions. In this context, three categories of biodegradable polymers can be distinguished: natural biopolymers such as cellulose, starch, chitin, and polysaccharides, which are highly susceptible to biodegradation by microorganisms; synthetic polymers made by polymerization, such as poly(-caprolactone) (PCL) and polylactic acid (PLA), which are the main focus of this review; and, finally, blend polymers, which are mixtures of the two mentioned categories (e.g., PCL/starch).

Within the scope of this review, the theoretical foundations of the biodegradation mechanism will first be described. Following that, parameters influencing the biodeterioration of synthesized polymers, such as environmental conditions and polymer characteristics, will be highlighted. Then, experimental methodologies for characterizing the biodeterioration of polymers by the assessment of polymer property variation and microorganism evolution footprints, along with available standardized protocols, will be addressed. Lastly, current developments in the biodeterioration of synthetic polymers, highlighting the literature that provided findings through parallel comparative approaches as well as systematic inspection and assessment, will be reviewed.

The procedure for selecting the cited literature in the present review was carried out in accordance with the subsequent steps:

1. The following five key phrases were searched on Google Scholar and Scopus: "polymer biodegradation", "polymer biodeterioration", "polymeric blend biodegradation", "degradation of polyolefin by microorganisms", and "degradation of aliphatic polyesters by microorganisms".
2. The pre-screening process was carried out in order to ascertain the pertinence of the search results. A comprehensive analysis of the articles published after the year 1990 resulted in a total of 229 articles that were deemed principally relevant within the scope of this review article.
3. The selected publications were categorized into five distinct groups: "review articles and book chapters", "original research", "polyolefins", "aliphatic polyesters", and "polymeric blends and composites".
4. The original research publications pertaining to each distinct polymeric family were categorized based on the individual polymer within each family (e.g., polycaprolactone (PCL), polylactic acid (PLA), etc., for polyesters) and the degrading agent involved (e.g., bacteria, fungi).
5. The selected original articles underwent a secondary screening process to determine the primary experimental methodology employed for characterization and the key outcomes. This process resulted in the selection of 187 publications for inclusion in this manuscript.
6. The review articles, book chapters, and original research articles that were not specifically related to the biodeterioration of selected polymers for Section 6 but that presented results that highlighted specific outcomes for the biodeterioration of polymers via microorganisms, such as the effect of influencing parameters or experimental methods used in a creative or critical manner, were used in Sections 1–4 to provide the reader with a clear background.

7. Original articles pertaining to the targeted polymers' biodeterioration were utilized in Section 6.
8. A parallel study was carried out using "ASTM International" and the "International Organization for Standardization" to address the standardized procedures for the characterization of the biodegradation of polymers. Due to the shared content of these standards, which is explicitly acknowledged within each standard, only nine of the ASTM International publications were included in this work (Section 5).

2. Biodeterioration Mechanism

There are multiple processes involved in the biodegradation of polymeric materials, which can operate concurrently or independently. Microorganisms can exert their influence by either mechanical, chemical, or enzymatic means. At least two types of enzymes, extracellular and intracellular, are actively involved in the biological degradation of polymers [21,22]. During the degradation process of a polymer, extracellular enzymes (exoenzymes) from microorganisms break down complex polymers, yielding short chains or smaller molecules, such as oligomers, dimers, and monomers. These molecules, which are small enough to pass through the semi-permeable outer membrane of microorganisms, are used by intracellular enzymes (endoenzymes) as carbon and energy sources [10,13,21,22]. The degradability of a material by these agents is greatly dependent on the material's nature and its chemical structure, and it is not guaranteed that this phenomenon will result in material cleavage [23].

While the "biodegradation" of polymers refers to the process by which a polymer undergoes any unfavorable alteration in its material properties brought on by environmental factors such as sunlight and harmful existing compounds (i.e., acids in soils), when this phenomenon is caused by biological agents such as bacteria, fungi, termites, beetle borers, and marine borers, the process is referred to as "biodeterioration" [24]. The definition of this phenomenon emphasizes living organisms as a common causative agent [23,24]. However, abiotic factors such as humidity, temperature, pH, and the presence of pollutants (as nutrients) may play significant roles as initiating parameters for microorganisms to degrade the polymer [13,21,22,25]. This phenomenon is a relatively superficial degradation where the microorganisms are in contact with the polymer, resulting in changes to the mechanical, physical, and chemical properties of the material at the surface [24]. Figure 1 depicts a schematic representation of the different steps of polymer biodeterioration by microorganisms, as well as the product of each stage.

Biofragmentation, also known as depolymerization, is a process in which polymers are lytically, i.e., micro-structurally, destroyed by microorganisms by breaking bonds between components of their atoms, resulting in the formation of oligomers and monomers. In this phase, microorganisms release exoenzymes into the environment to break down polymers with high molecular weights, which cannot penetrate the cell wall and/or plasma membrane, into shorter chain molecules such as monomers, dimmers, and oligomers that are small enough to pass through the cell membrane [10,13,21,22]. The molecular conformation of the polymers is crucial to this process. For instance, crystalline areas, hydrophobic zones, and steric hindrances make it difficult for some enzymes to carry out scission reactions. In this instance, oxidoreductases enzymes participate in the enzymatic oxidation process to alter some molecular structures and enhance the likelihood that the polymer may be fragmented by other enzymes [13].

Assimilation, on the other hand, is the process by which living microorganisms integrate the polymer fragments produced during the biofragmentation phase that passed through the cell membrane and use them as nutrients for energy production, cell structure formation, growth, and reproduction. There are three crucial catabolic routes that create the energy needed to maintain cellular activity, structure, and reproduction: aerobic respiration, anaerobic respiration, and fermentation [13]. These pathways depend on organisms' capacities to develop in aerobic or anaerobic environments. This stage may result in biomass increases, methane production, and the formation of inorganic molecules such as H_2O and

CO_2. Mineralization is the term used for degradation when the final product is CH_4 or inorganic species, such as CO_2 and H_2O [10,21,22].

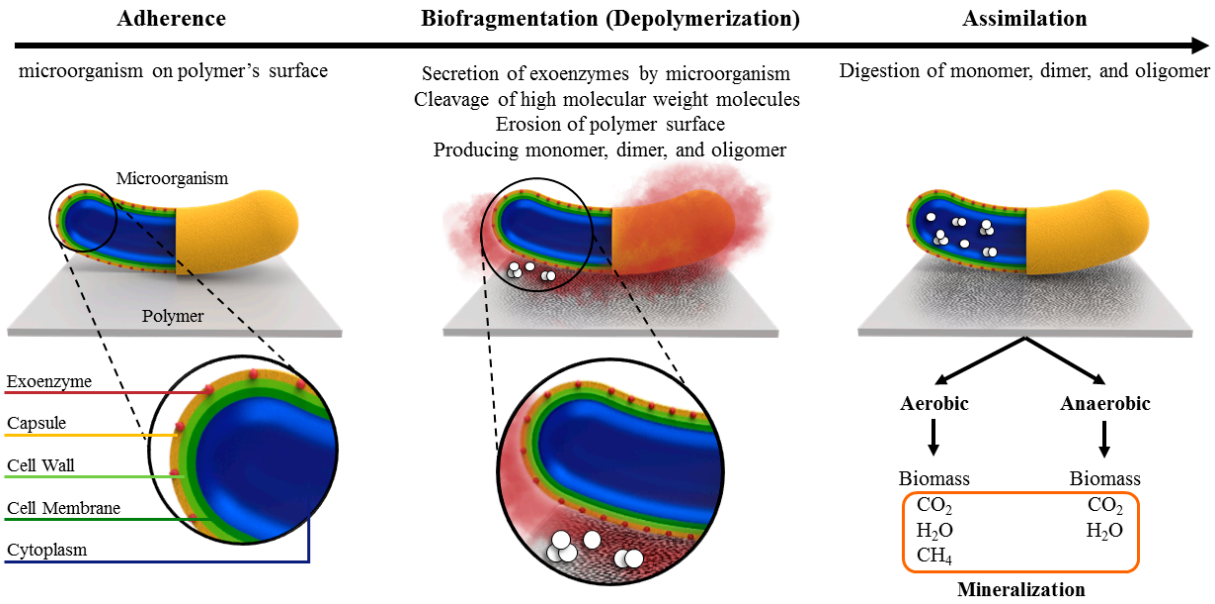


Figure 1. General mechanism of polymer biodeterioration by microorganisms (The white circles depicted in the diagram symbolize monomers, dimers, and oligomers).

3. Factors Affecting Biodegradation

As mentioned before, the variables that influence the biodegradability of a polymer are the polymer's physical properties and environmental conditions. The environmental conditions influencing polymer degradation can be divided into two categories: biotic (organisms and the enzymes they release) and abiotic variables. The polymeric materials may already be degrading due to abiotic factors such as light, temperature (T), humidity, and minerals when they are exposed to the environment [13]. Although UV light photolysis and γ-ray irradiation of polymers frequently lead to cleavage and degradation [20], this section focuses on the factors that influence polymer biodegradation in the context of living organisms (biodeterioration). Figure 2 summarizes the parameters that influence the biodegradation of a synthetic polymer.

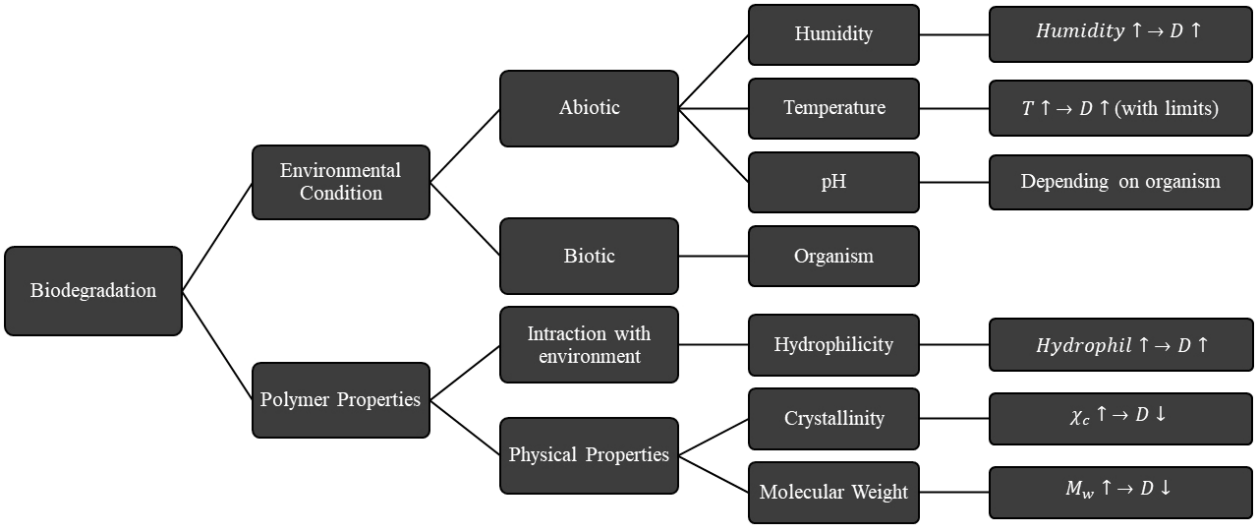


Figure 2. Factors affecting biodegradation of polymers, D: degradation, χ_c: crystallinity, M_w: molecular weight.

3.1. Environmental Conditions

3.1.1. Humidity

Moisture can influence the biodegradation of polymers in two main ways. First, water is a crucial component for the development and reproduction of organisms. Second, environments with a high level of humidity promote the hydrolysis process by increasing the number of chain scission processes. For instance, comprehensive research on the effects of relative humidity on the biodegradation of high-molecular-weight polylactic acid (PLA) films revealed that an increase of 1% relative humidity (RH) in the environment can increase the degradation rate of the studied PLA by 939–2012 M_w/week [26].

3.1.2. Temperature

Generally, temperature has a major effect on the kinetics of enzymatic reactions (similar to any other chemical reactions), especially when it reaches the optimal temperature for enzyme activity [26,27]. A modest adjustment of 1 or 2 °C in the reaction temperature might cause changes of 10% to 20% in the outcomes. Most enzymes' activity increases by 50 to 100% when the temperature rises by 10 °C. However, this rise only lasts while the enzymes' structures are not compromised by the increased temperature. The enzymes cannot be repaired once they have been denatured. Each enzyme has a particular structure as well as unique linkages between amino acids and peptides; hence, each enzyme has unique optimal and denaturing temperatures. Figure 3 shows a schematic representation of how temperature influences enzymatic reaction rates.

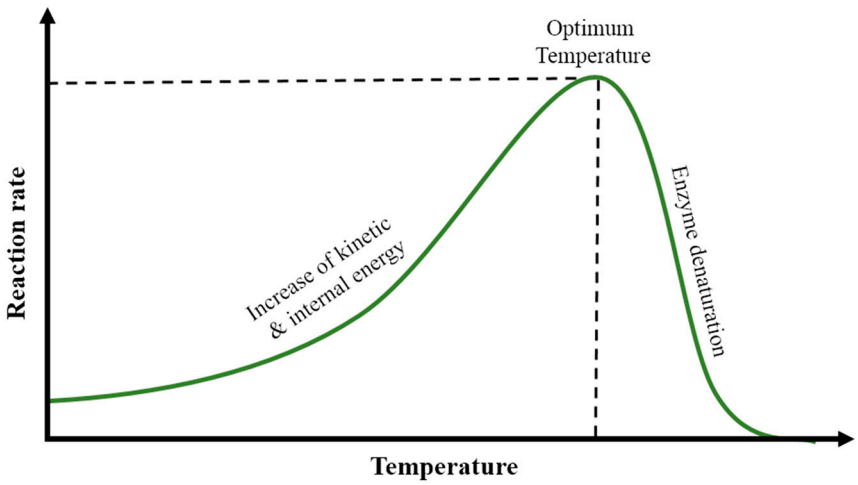


Figure 3. Effect of temperature on reaction rates.

From a kinetic point of view, as the temperature rises, the velocity and kinetic energy of all molecules increase, as do their collisions. The higher the temperature, the more molecules have thermal energies greater than the activation energy required for a reaction. The effect of temperature on a reaction rate usually obeys the Arrhenius equation, which is an inverse exponential of the reciprocal of the absolute temperature. The internal energy of the molecules can include the translational energy, vibrational energy, and rotational energy of the molecules, as well as the energy involved in chemical bonding and nonbonding interactions. This feature might make it easier for enzymes to break down the polymer chain, but if the chemical potential energy rises excessively, some weak bonds that help give active proteins their three-dimensional shapes might be broken, which might cause the protein to become thermally denatured and inactive.

3.1.3. pH

pH is a scale that describes the acidity or alkalinity level of a substance based on the quantity of hydrogen ions or hydroxides present in the substance. Similar to temperature, the optimum pH range for enzymatic activity is unique for each enzyme [28–31]. Changing the pH outside of this range will slow enzyme activity. An extreme pH change will cause the amino acid atoms and molecules to ionize, changing the form and structure of proteins and causing denaturation. In addition to affecting enzymatic activity, pH also has an impact on the substrate's charge and shape, making it difficult for the substrate to bind to active sites or catalyze the formation of a product.

3.2. Polymer Properties

3.2.1. Polymers' Molecular Structures

Although a polymer's physical properties are often referred to as a factor influencing biodegradation, a polymer's chemical structure is the primary component that determines those characteristics and, by extension, its biodegradability. A single polymer molecule can have a linear, branching, or network structure and can consist of anywhere from hundreds to millions of monomers. In biological systems, natural macromolecules such as protein, cellulose, and starch are often broken down via hydrolysis and oxidation. The fact that most synthetic biodegradable polymers have hydrolyzable links along their polymer chains (for instance, amide, enamine, ester, urea, and urethane linkages) makes them amenable to biodegradation by microorganisms and hydrolytic enzymes [20]. Generally, the greater the similarity of a polymeric structure to a natural molecule, the easier it is to break down [21,22]. The impact of a polymer's molecular structure may be viewed from two angles: the impact on the polymer's interactions with the environment and the impact on its physical properties. Most enzyme-catalyzed reactions take place in aqueous environments; therefore, synthetic polymers' hydrophilic-hydrophobic properties have a significant impact on how readily they degrade. For instance, the hydrogen bonds created by the presence of functional groups in a side chain, such as the carbonyl or amide groups of monomers, result in an increase in polymer adhesion and hydrophilicity at the surface, improving the environmental conditions for microorganisms to act [8,20]. Some research indicates that endopeptidases cleave bonds that include amide groups, while lipases and esterases particularly target carboxylic linkages in their hydrolysis-based approach to polymer degradation [32–34]. Regarding the impact of polymers' molecular structures on their physical characteristics, elements such as the degree of crystallinity and molecular weight are important in the hydrolysis and oxidation of polymers by enzymes.

3.2.2. Crystallinity (χ_c)

Crystallinity is a crucial factor affecting biodegradability. A polymer's crystalline section is more resistant than its amorphous portion. The molecules in the amorphous region are not tightly compressed against one another, and this region is more vulnerable to deterioration. Consequently, enzymes primarily target a polymer's amorphous regions. Proteins, for instance, lack regular repeating units along their polypeptide chains, which is one of the key distinctions between them and synthetic polymers. Because of this irregularity, protein chains are less prone to form crystalline structures. It is highly likely that this characteristic helps explain why proteins are readily biodegradable. On the other hand, synthetic polymers typically feature short repeating units, and this regularity promotes crystallization, blocking enzyme access to hydrolyzable groups [20]. Iwata and Doi's [35] investigation of the enzymatic hydrolysis of lamellar single crystals of poly(L-lactic acid) (PLLA) revealed that depolymerization by enzymes occurred mainly against the disordered chain-packing regions of single crystals rather than their chain-folding surfaces. Moreover, Tsuji and Miyauchi [36] discovered that PLLA chains in the confined amorphous regions between crystalline sections are more hydrolysis-resistant than those in free amorphous regions, as in totally amorphous films. Studies have shown that as a semi-crystalline polymer degrades, the amorphous sections of the polymer vanish, causing the

sample's crystallinity to increase quickly initially before levelling off to a much slower pace as the crystallinity approaches 100% [37–39]. Moreover, the degree of crystallinity affects the melting point and glass transition temperature of a polymer. Therefore, the likelihood of biodegradability for the same polymer reduces as T_g and T_m increase [15,40–42].

3.2.3. Molecular Weight (M_w)

As stated in Section 3.2, polymers with molecular structures too wide to pass through an organism's semipermeable cell membrane should be preliminarily fragmented into smaller molecules by exoenzymes. This emphasizes how a polymer's molecular weight affects its capacity to degrade biologically. Plastics, such as PE, PP, and PS, continue to be reasonably resistant to microbial attacks and growth as long as their molecular weight stays high. On the other hand, low-molecular-weight hydrocarbons can be degraded by microbes [20]. The majority of observed discrepancies reported regarding the influence of molecular weight on biodegradation processes for the same polymer may be attributed to the complexity in detecting changes during degradation or, more frequently, the variations in the morphology and hydrophilicity–hydrophobicity of polymer samples of different molecular weights. Studies have shown that alkane-based plastics with molecular weights greater than 400–500 daltons (i.e., more than 30 carbon atoms) must be degraded into smaller molecules before biodegradation through photodegradation, chemical reactions, or other biological processes [20,43]. In fact, a study of PLA of different molecular weights (ranging from 5000 to 25,600) revealed that the biodegradation rate reduced as the molecular weights increased [44]. Research on the degradation of PCL of various molecular weights by *Rhizopus delemar* lipase (endo-cleavage type) found that in PCL with an average molecular weight (M_n) greater than 4000, biodegradation was not affected by M_n, while decreases in M_n below 4000 correlated with increases in the rate of biodeterioration [40].

3.2.4. Physical Form

The physical form of a polymer (e.g., foam, film, powder), which determines its surface-to-volume ratio, can have a significant effect on its susceptibility to biodeterioration by microorganisms. As previously mentioned, biodegradation of polymers by biotic degraders is a superficial phenomenon that requires microorganism adherence to the polymer surface; hence, for the same material, the more surface contact between the microorganism and polymer, the faster the biodegradation. Consequently, polymers in a porous or cellular form, such as foams, are more susceptible to biodeterioration than bulk materials. This is because the open structure of foam materials provides a large surface area that is easily accessible to microorganisms, allowing them to penetrate and colonize the material. Additionally, the pores and cells of foam materials can trap moisture, which provides an ideal environment for the growth and proliferation of microorganisms.

4. Experimental Techniques for Characterization (Analysis of Degradation)

Changes in the physical appearance of polymers, such as surface roughness, the appearance of holes or cracks, defragmentation, color changes, or the development of biofilms on the surface, are one of the first indications of degradation. Biodegradation also has an impact on mechanical properties, including Young's modulus, yielding stress, and elongation at break, which cannot be observed and proved directly. At the molecular level, biodegradation alters the conformation of molecules and functional groups, as well as the degree of crystallinity and certain thermal properties. In this section, we discuss the most commonly used experimental techniques for characterization of the biodeterioration of polymers. It must be noted that the experimental techniques for characterization of biodeterioration are not limited to the categories presented here. Many other methodologies, such as UV–visible spectroscopy [45], UV photo oxidation [46], fluorescence microscopy [47], and high-performance liquid chromatography (HPLC) [48–50], have been employed to assess physical, chemical, and mechanical properties alterations in polymer biodegraded by

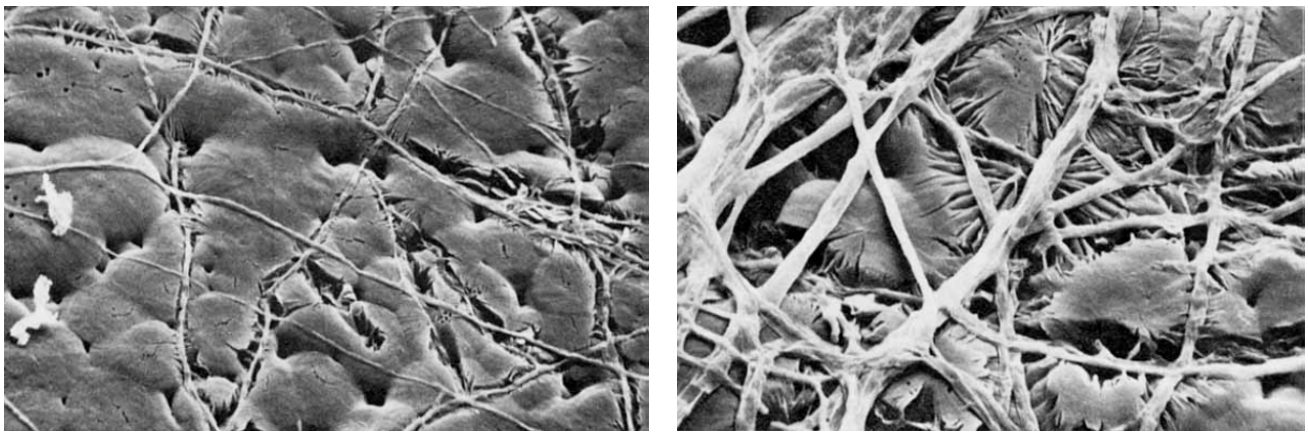
microorganisms. For instance, investigation of a polymer's surface energy in some instances shows that the wettability level of the polymer increases after biodegradation [47,51].

4.1. Morphological Analysis

4.1.1. Scanning Electron Microscopy (SEM)

At an early stage of degradation, when the amorphous component of a polymer is deteriorating rapidly, the slower-degrading crystalline parts (spherulites) remain and protrude from the surface of the material. At this stage, the physical changes at the polymer's surface are not apparent to the naked eye; hence, scanning electron microscopy is beneficial for monitoring these changes on a much smaller scale. For example, in a study by Li et al. [52] which used scanning electron microscopy to assess the biodegradability of blends of thermoplastic starch (TPS) with poly(lactic acid) (PLA) and low-density polyethylene (LDPE), the influence of a highly biodegradable phase in a mixture with a significantly less biodegradable component on the surface morphology of the biodegraded composite was highlighted. Although this approach is one of the most used techniques for the observation of biodegradation, it is inconclusive and is limited to qualitative evaluations of surface morphology. Nonetheless, this technique has been utilized to show that the geometries of holes on decaying surfaces vary depending on the biodegradation media [53–55].

Figure 4 portrays scanning electron microscopic visualizations of polycaprolactone biodegradation by the filamentous fungus *Penicillium funiculosum*; the amorphous portions deteriorate first, leaving the radial spherulite arms (Figure 4a), followed by the crystalline structure (Figure 4b).



(**a**) (**b**)

Figure 4. SEM image of *Penicillium funiculosum mycelium* degrading polycaprolactones (**a**) biodeterioration of amorphous regions, (**b**) complete degradation of amorphous regions and partial degradation of crystalline phase. Adapted from [54] with permission from John Wiley and Sons.

4.1.2. Atomic Force Microscopy (AFM)

A high-resolution image of the three-dimensional shape (topography) of a sample surface during or after the degradation process can be obtained using atomic force microscopy. In other words, this approach gives a morphological characterization of the nano-scale surface roughness. Since this technique can also measure some mechanical properties such as Young's modulus, the simultaneous acquisition of topographical images and locally measured mechanical properties, displayed as an image with a similarly high resolution, provides much more quantitative characterization of the degradation process. For instance, an AFM examination of polyethylene that was deteriorated by wax moth caterpillars revealed a 140% increase in surface roughness [56]. In another study, AFM techniques were used to evaluate the lamellar thicknesses of single PLLA crystals before and after enzymatic degradation [35]. Atomic force microscopy was paired with scanning electron microscopy to investigate the possibility of polyethylene biodegradation by bacillus strains (YT1 and

YP1) from the guts of plastic-eating waxworms, providing evidence of surface deterioration with cavities of approximately 0.3 and 0.4 µm depths after inoculation [47] (see Figure 5).

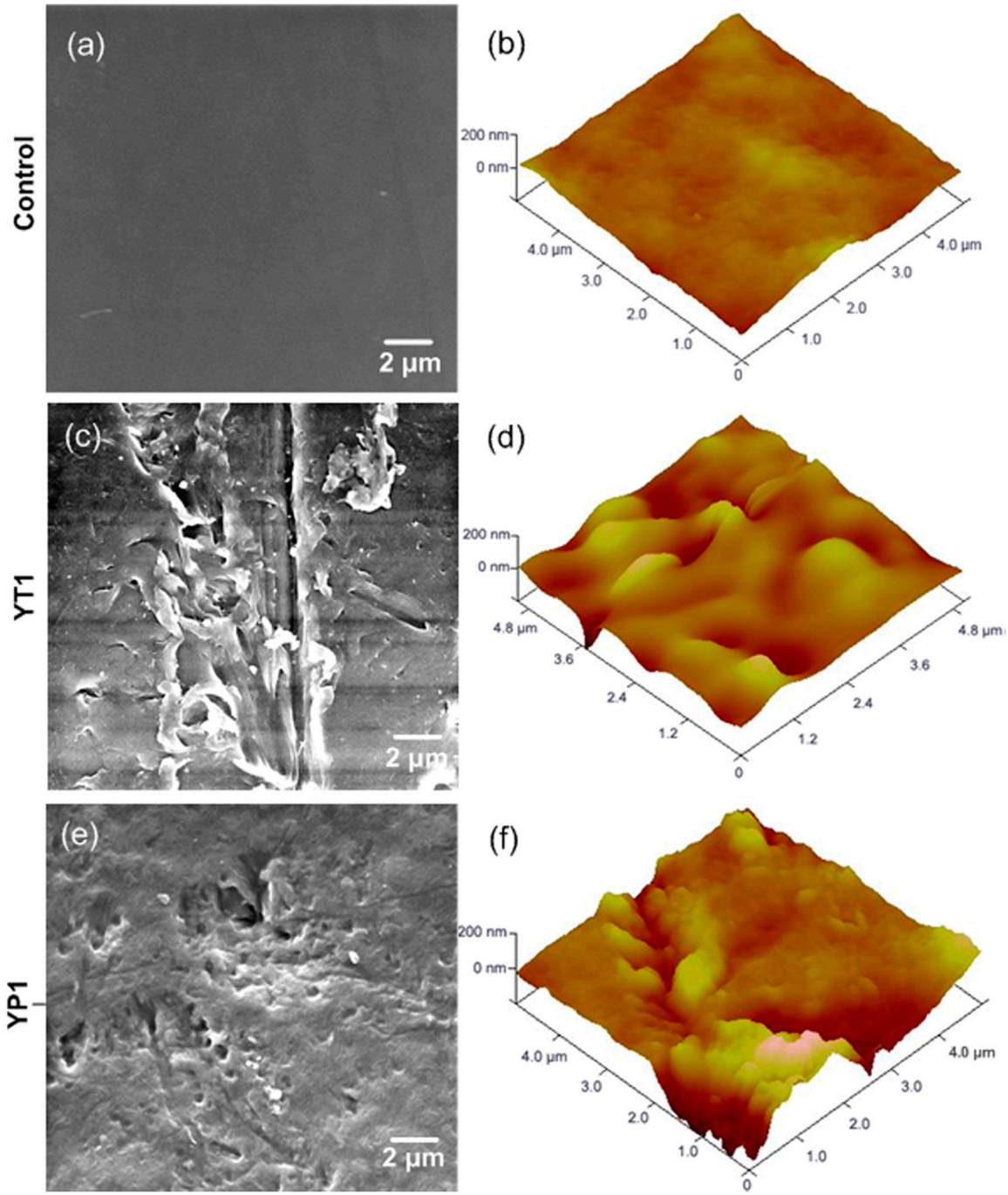


Figure 5. Combined SEM and AFM techniques for characterization of the bioditerioration of polyethylene sheets after 28 days, (**a**,**b**) SEM and AFM images of the sterile control sample, (**c**,**d**) SEM and AFM images of the specimens degraded by Bacillus strain YT1, (**e**,**f**) SEM and AFM images of the specimens degraded by Bacillus strain YP1. Adapted with permission from [47] the American Chemical Society, copyright 2014.

4.2. Gravimetric Measurements

The measurement of the weight loss of test specimens such as films or test bars is a preliminary quantitative step and the most extensively utilized method of assessment. The primary benefits of this approach are its simplicity and adaptability, but a disadvantage is that it requires a large number of samples to obtain results with a high level of accuracy. Since weight loss may involve processes dominated by chemical hydrolysis and the breakdown of polymers during exposure, especially when elevated temperature and humidity are applied, obtained data via gravimetric measurements should be interpreted with caution. Another issue could arise from improper cleaning of the buried specimen or from severe material disintegration. When small fragments of significantly disintegrated specimen cannot be collected from the test environment, the acquired gravimetric measurements will be overstated. In this scenario, as described in DIN V 54900 for the full-scale composting process, the samples can be put into small nets to aid the recovery [10].

4.3. Respirometry Measurement

This method is the backbone characterization technique for all the available standardized protocols for the determination of polymer biodegradation (see Section 5). In an aerobic environment, microorganisms oxidize carbon with oxygen (O_2) to produce carbon dioxide (CO_2) as their main metabolic end product. The respirometry technique measures the oxygen consumption or carbon dioxide evolution (modified Sturm test) in a closed system with controlled air or oxygen circulation as an indicator for polymer degradation. When other conventional methods of application are unsuccessful, this technique can be used to measure the degradation of soluble, powders, and delicate polymeric compounds.

This method is particularly suited for verifying degrees of mineralization and has a high level of accuracy. In the context of mineralization as the objective of a study or experiment, the system containing the candidate polymer must be continuously monitored for CO_2 evolution or O_2 consumption. When working with synthetic mineral media, there are typically only a low number of alternative carbon sources present in addition to the polymer itself; hence, only a relatively low level of background respiration must be determined.

Automated and continuous measurements provide benefits, but they can also have drawbacks. If gradual degradation processes are to be assessed and the CO_2 concentration or decline in O_2 concentration to be detected is very small, the possibility of systematic errors increases. Consequently, the signals of the detectors must be steady for long periods of time.

4.4. Fourier Transform Infrared Spectroscopy (FTIR)

Fourier transform infrared spectroscopy is a quick, cost-effective, simple, and non-destructive technology that allows the identification of functional groups which may form or disappear during biodegradation, including branches, co-monomers, unsaturation, and the presence of additives such as antioxidants. For example, this approach has been utilized to characterize carbonyl group formation during the photodegradation of polyethylene and polypropylene from both a qualitative and quantitative perspectives [57]. It bears mentioning that this technique can be complemented by X-ray photoelectron spectroscopy (XPS) [47] or nuclear magnetic resonance (NMR) spectroscopy to determine a molecule's chemical structure [58].

4.5. Thermal Analysis

4.5.1. Differential Scanning Calorimetry (DSC)

Differential scanning calorimetry (DSC) measures the amount of heat transferred to or from a sample that is undergoing a physical or chemical change. This method is used to examine a polymer's melting and crystallization characteristics as well as any heat gain or loss that occurs during the corresponding phase transitions. As noted in Section 3.2, during the initial phases of biodeterioration, microorganisms target the amorphous fraction

of polymers first. Once this portion has been broken down, the crystalline part of the polymer begins to slowly deteriorate. As a result, it is reasonable to anticipate that a polymer's degree of crystallinity will increase in its initial stages and then slightly fall thereafter. The use of DSC across the various stages of degradation for the purpose of measuring the crystallinity of the polymer at each stage can provide further insights regarding the progression of the biodeterioration of the polymer. For instance, a comparison between the heat of fusion from the DSC analyses of polyethylene before and after 140 days of degradation by *Aspergillus niger* revealed an increase of 50 kJ/kg [59]. A DSC investigation of biodegraded poly(lactic acid) revealed a reduction in the cold crystallization temperatures (T_c) for all biodegraded materials compared to their initial values, as well as the appearance of a low-temperature endothermal shoulder upon melting [60]. For a more thorough examination, DSC can be combined with X-ray diffraction (XRD) [61,62], a technique for determining the crystallographic structure of a material as well as phase identification. Additionally, XRD provides details on how internal stresses and flaws cause the actual structure of a polymer to diverge from the ideal one [38,52,63].

4.5.2. Thermogravimetric Analysis (TGA)

TGA is an analytical technique used to determine a material's thermal stability and its fraction of volatile components by monitoring its weight variation as a function of a constant heating rate in a controlled atmosphere. Several thermogravimetric studies on the biodegradation of polymers revealed that the decomposition temperatures of polymers shift to lower temperatures with longer temperature spans after degradation in comparison to polymers that have not been degraded [58,64–67]. In some of these studies, TGA was combined with simultaneous differential thermal analysis (DTA) and derivative thermogravimetry (DTG) to gain a better understanding of the thermal properties of the biodegraded studied polymers [66,67].

4.6. Molecular Mass Characterization

4.6.1. Viscosimetry

The average molecular weight of a polymeric material has a direct effect on its intrinsic viscosity; the higher the average molecular weight, the greater the intrinsic viscosity. In other words, the longer the chains, the more difficult it is to get them to flow since they are more entangled. Several experimental techniques, including rehometry [52], the melt flow index (MFI), and capillary viscosimetry [46,68–70], can be used to describe a polymer's molecular mass using its viscosity. In the context of the biodegradation of polymers, capillary viscosimetry is one of the most widely used methods for the determination of the average molecular mass due to its cost efficiency. In a study on the biodegradation of low-density polyethylene (LDPE) by ultraviolet light and a bacteria consortium, the capillary viscosimetry test on specimens after 30, 60, and 90 days of incubation revealed more detailed information concerning the effects of UV light and bacteria on biodegradation and evaluated a final 34% reduction in molecular weight [46]. It must be highlighted that this technique estimates one single value as the average molecular weight and does not provide any insight about the molecular weight distribution.

4.6.2. Chromatography

Size-exclusion chromatography (SEC), gel permeation chromatography (GPC), and gel filtration chromatography (GFC) are names interchangeably used to describe the same liquid column chromatographic technique. In comparison to capillary viscosimetry, SEC is a more sophisticated technique that not only allows measurement of the average molecular mass but also provides information about the molecular mass distribution. This method has been used to characterize the polydispersity index and average molecular weight variation in biodegraded nylon 66 [71] and nylon 12 [69]. Since during degradation the amorphous fraction of a polymer goes through chain scission via the enzymatic attacks of microorganisms, the molecular weight distribution of the polymer will change significantly, which

cannot be detailed by average molecular mass characterization. For instance, size-exclusion chromatography studies before and after microbial contamination of PLA demonstrated a significant shift in the molecular weight distribution curves toward lower molecular weights and longer retention times [60].

4.7. Surface Hydrolysis and pH Level Characterization

Another novel technique entails the monitoring of the pH variations in a degradation system. A rise in acidity is a reliable sign that polymers have been exposed to enzyme-induced surface hydrolysis. This technique has the benefit of requiring a minimal amount of material for experimentation but suffers from the fact that it can only be used with polymers containing ester bonds, as those can be easily cleaved through hydrolysis reaction under natural conditions, meaning its potential scope is somewhat restricted. Consequently, this technique has been used to characterize the biodegradation of aliphatic–aromatic copolyesters [72], poly(3-hydroxybutyrate) [50], and poly(hydroxybutyrate-co-hydroxyvalerate) (PHBV) [34]. It is evident that this kind of system may not be adequately applicable for simulated environmental circumstances containing microorganisms or when biochemical activities carried out by microorganisms are required. Nonetheless, it has been used to characterize the biodegradation of low-density polyethylene by *Bacillus amyloliquefaciens* [73] and *P. chrysosporium* [68], as well as the biodegradation of PA66 [74].

4.8. Mechanical Characterization

In general, a polymer's mechanical properties, particularly Young's modulus, yield stress, strain at yield stress, and stress and strain at break, alter dramatically due to deterioration. A polymer's mechanical characteristics following degradation in various incubation environments can be used to determine the optimal environmental conditions (temperature, humidity, and microorganism type) for biodegradation. According to research conducted by Dutta et al. [65] on the biodegradation of epoxy and MF-modified polyurethane films in three distinct soils, the tensile strength of blend films was found to decrease with increases in the soil burial exposure period for 60:40 blends of PUE:Epoxy, with the maximum degradation being observed in *Solmora* soil. The other blends used in this study had constant mechanical properties for the first 60 days, which provided additional information about the best microorganism consortia for each blend's biodegradation. From a different angle, the mechanical property characterization of various polyethylenes, including degradable polyethylene, low density polyethylene, and high density polyethylene, in soil mixed with 50% (*w/w*) mature municipal solid waste compost demonstrated the evolution of biodegradation of these plastics over a fifteen-month period in municipal burial soil [75].

5. Standardized Protocols

Currently available test procedures for evaluating polymer degradability have been established based on simulating the conditions encountered by the materials in the environment (e.g., marine, soil, UV exposure, etc.) in a laboratory environment, which gives them a limited adaptability. Table 1 lists some of the most significant American Society for Testing and Materials (ASTM) standards according to the objective, key feature microorganism, and monitoring parameter. The major advantage of this series of methods is the high reproducibility of the testing environmental conditions; however, it must be highlighted that these tests only involve a small number of fungal and bacterial species under simulated conditions, while finding the exact same collection of microorganism species in diverse geographical situations is extremely rare in natural deterioration. Moreover, the selection of the microorganisms, their importance in deterioration, and their relevance to the local environmental circumstances are some of the key biases of current available standards. For example, tropical and subtropical climates will have dominant microflora that differs greatly from cold and dry places, which influences biodegradation dramatically.

Table 1. Standard methods for testing the biodegradation of polymers.

Standards	Scope	Condition	Analyzed Parameters	Ref.
D5526-18	Anaerobic biodegradability of plastic materials under accelerated landfill conditions	30–300 days in anaerobic condition Mesophilic temperatures (35 ± 2 °C) pH between 7.5 and 8.5 Decomposition under dry (more than 30% total solids) and static non-mixed conditions Pretreated household waste exposed to a methanogenic inoculum derived from anaerobic digesters operating	CH_4 evolution CO_2 evolution Wet-weight loss	[76]
D5988-18	Aerobic biodegradation of plastic materials in soil	Equivalent to ISO 17556 120–180 days in aerobic condition Mesophilic temperatures: 20 to 28 °C ± 2 °C pH between 6 and 8 Natural, fertile soil collected from the surface layers of fields and forests (at least three diverse locations)	CO_2 evolution O_2 consumption	[77]
D5511-18	Anaerobic biodegradation of plastic materials under high solids	Equivalent to ISO 15985 15–30 days in anaerobic condition Temperature: 37 ± 2 °C or 52 ± 2 °C pH between 7.5 and 8.5 Methanogenic inoculum derived from anaerobic digesters operating only on pretreated household waste Decomposition under high solids (more than 30% total solids) and static non-mixed conditions.	CH_4 evolution CO_2 evolution	[78]
D7991-22	Aerobic biodegradation of plastics buried in sandy marine sediment			[79]
D5338-15 (2021)	Aerobic biodegradation of plastic materials under controlled composting conditions	Equivalent to ISO 14855 45 days in aerobic condition Thermophilic temperatures (58 ± 2 °C) pH between 7 and 8.2 Inoculum compost from municipal solid waste	CO_2 evolution Visual assessment Weight loss	[80]
D6954-18	Plastics that degrade in the environment by a combination of oxidation and biodegradation	Decomposition in soil, landfill, and compost in which thermal oxidation occurs Degree of physical property losses by thermal and photo-oxidation processes and biodegradation Temperatures for decomposition in soil (20 to 30 °C), landfill (20 to 35 °C), and composting facilities (30 to 65 °C).	DSC (T_g) Molar weight loss Polydispersity index Tensile strength loss Weight loss CO_2 evolution	[81]
D7475-20	Aerobic degradation and anaerobic biodegradation of plastic materials under accelerated bioreactor landfill conditions	Simulate change from aerobic to anaerobic condition over time as landfill depth increases Material is mixed with household waste, then pretreated and stabilized aerobically in the presence of air; exposed to a methanogenic inoculum derived from anaerobic digesters operating only on pretreated household waste Aerobic incubation 30 ± 10 °C for 4 weeks Anaerobic incubation 35 ± 2 °C for 4 months	O_2 consumption CO_2 evolution CH_4 evolution Tensile strength loss Molar weight loss	[82]
D6400-23	Plastics designed to be aerobically composted in municipal or industrial facilities	Equivalent to ISO 17088 180 days in aerobic condition Thermophilic temperatures Municipal and industrial aerobic composting facilities	CO_2 evolution Weight loss	[83]
D6868-21	Biodegradation of polymers as coatings to be aerobically composted in municipal or industrial facilities	Thermophilic temperatures (58 ± 2 °C) 180 days in aerobic conditions Municipal and industrial composting facilities	CO_2 evolution Weight loss	[84]

6. Recent Advances in the Biodegradation of Polymers

During the last three decades, a large body of research has been devoted to the study of numerous bacterial strains and fungal degraders of polymers, as well as the biodeterioration of polymers by these microorganisms in various environments and under optimal physiological conditions, which cannot be covered in the context of a brief review. Hence, for the sake of brevity, this section focuses on polymers that are particularly susceptible to biodeterioration by microorganisms, highlighting the literature's findings by parallel comparative methodologies and systematic examination and assessments.

6.1. Aliphatic Polyesters

To address the issues of plastic waste accumulation, aliphatic polyesters are among the most promising materials for applications such as packaging and mulch films. For a wide range of applications, Polylactic acid, polycaprolactone, poly(3-hydroxybutyrate) (PHB), and their copolymers are the most researched aliphatic polyesters. Copolymerization and blending of these materials yield a wide range of characteristics and degradation behaviors. Various polymers and fillers, including as PVC, PET, polyvinyl alcohol (PVAl), and PE, have been blended with polycaprolactone; nevertheless, the mechanical qualities or the degradability were diminished in comparison to the homopolymers [85]. In a comparative evaluation of the biodegradation of PHB, PCL, and PLA over the course of more than 10 months at 25, 37, and 50 °C in soil and compost, PCL showed faster deterioration than PLA and PHB [86].

Enzymatic biodeterioration of other aliphatic polyesters, such as poly (ethylene adipate) (PEA), poly (β-propiolactone) (PPL), polybutylene succinate (PBS), and polyethylene succinate (PES), has been studied to some extent. Although lipases from several other organisms, including *R. arrizus*, *R. delemar*, *Achromobacter* sp., *Candida cylindracea*, and hog liver esterase, demonstrated activity on PEA [87], *Penicillium* sp. strain 14-3 was the most potent degrader of PEA. *Acidovorax* sp., *Variovorax paradoxus*, and *Sphingomonas paucimobilis* were found to be microorganism strains that are able to degrade PPL [88]. On agar plates containing emulsified PBS, *Microbispora rosea*, *Excellospora japonica*, and *E. viridilutea* developed clear zones. After eight days of incubation in liquid media containing *Microbispora rosea*, 50% (*w*/*v*) of the PBS films were degraded [89].

To some extent, different aliphatic polyesters share the family of degrader microorganisms. *Aspergillus fumigatus* strain 76T-3 can degrade PHB, PES, PBS, PCL, and PLA [90]. A thermophilic *Bacillus* sp. TT96 may generate clear zones on PES, PCL, and PBS and deteriorate them, but it cannot degrade PHB [91]. *Bacillus pumilus* strain KT102, which is one of the fastest degraders of PES, can also degrade PCL but not PBS, PHB, or PLA [92].

6.1.1. Polycaprolactone (PCL)

Polycaprolactone is a biodegradable synthetic semicrystalline aliphatic polyester with a low melting point (T_m = 60 °C) that is used as an implantable biomaterial in numerous food, drug, and biomedical applications [93]. It has been demonstrated that PCL can be degraded by the activity of aerobic and anaerobic microorganisms, both of which are common in a wide variety of environments [16,94], including in soil medium [95] and sea water [96]. Lipase has been proven to deteriorate PCL in general [97].

Castilla-Cortázar et al. [98] compared the rates of hydrolytic and enzymatic (*Pseudomonas* lipase) degradation of PCL and concluded that enzymatic degradation (14 weeks) was quicker than hydrolytic degradation (60 weeks), producing the same level of degradability and having a different mechanism. The results of the morphology and swelling measurements suggested that the hydrolytic degradation affected the entire sample through a bulk erosion mechanism, whereas the enzymatic degradation appeared to follow a superficial erosion mechanism [98]. Two comparative studies on the efficacy of the bacterial [99] and fungal [100] biodegradation of PCL with varying molecular weights (7130, 18,600, and 35,000) were conducted using gel permeation chromatography, differential scanning calorimetry, and ASTM G21-70. The most effective fungal degraders for PCL were verified according to the following order: *Aspergillus Fischeri*, *Fusarium* sp., *Chaetomium Globosum*, *Aspergillus Flavus*, *Penicillium Funiculosum*, and finally *Aspergillus Niger* [100].

A comprehensive study on the biodegradation of PCL by three different lipases derived from *Lactobacillus brevis* and *Lactobacillus plantarum* and their co-cultures, with different bacterial concentrations varying from 0.5 to 5 mg/mL, highlighted that polymer degradation is dependent on both the concentration and type of the enzyme, and increasing enzyme concentration alone is not necessarily indicative of higher biodegradation [101]. The effect of molecular weight (Mw = 33,000, 57,000, and 76,000) on the biodeterioration of medical PCL by *Candida antarctica lipase* revealed no evidence of the molecular weights'

influence on the biodegradability of the PCL films; however, the molecular weights did have some influence on the surface pore diameters of the films [102]. Table 2 lists some of the systematic studies on bacterial and fungal degraders of polycaprolactone.

Table 2. Bacterial and fungal degraders of PCL.

	Organism	Characterization										Time (Days)	Degrad. %	Ref.
		SEM	Grav. *	CO_2	DSC/ TGA	NMR	pH	XRD.	Mech	M_w *	FTIR			
Bacterial	*Pseudomonas*	X	X		X			X	X			98	20	[98]
	Lactobacillus brevis	X	X		X						X	10	10	[101]
	Lactobacillus plantarum	X	X		X						X	10	60	[101]
	Amano Lipase P. Cepacia,		X	X						X		47	90	[103]
	Household refuse (strain 2.2)	X	X	X							X	18	100	[104]
	Ralstonia sp. strain *MRL-TL*	X	X								X	40	64	[105]
Fungal	*Candida antarctica Lipase*	X	X		X	X	X	X		X	X	1	85	[102]
		X	X		X			X			X	3	87.6	[106]
	Fusarium solani cutinase	X	X		X			X			X	3	80.8	[106]
	Pullularia pullulans		X						X	X		42		[107]
	Penicillium lilacinus D218		X									10	10	[30]
	Aspergillus sp. strain *ST-01*	X	X				X					6	100	[108]
	Cryptococcus laurentii	X	X		X			X		X		30	100	[109]
	Fusarium	X	X		X			X		X		30	100	[109]

* Grav.: Gravimetry, M_w: Molecular weight characterization.

6.1.2. Polylactic Acid (PLA)

Polylactic acid and its composites are some of the most studied biodegradable polymers [4,9,26,35,36,48,52,60–64,110–114]. PLA is an aliphatic (linear) polyester composed of ester-bonded polymerized lactic acid monomers. Depending on its enantiomers' structural conformations and thermal histories, PLA can be either amorphous or semicrystalline in its solid form [4], which influences its biodegradability. PLA made from 50 to 93% L-lactic acid is strictly amorphous, while PLA made from more than 93% L-lactic acid is considered semicrystalline. Both meso- and D-lactide create twists in the usually extremely regular molecular structure of poly(L-lactide) [115]. Poly(L-lactide) and poly(D-lactide) are known to combine in an equimolar stereocomplex crystalline structure that has a substantially greater melting temperature (230 °C) than the respective homopolymers [115], and hence has an inferior biodegradability.

In addition to common parameters such as molecular weight, crystallinity, purity, temperature, and pH, biodegradation of PLA has also been found to be dependent on the presence of terminal carboxyl or hydroxyl groups, water permeability, and additives acting catalytically, which may include inorganic fillers. The impact of hydroxyl and carboxyl groups, catalysts, and solvents on polymerization and the ultimate molecular weight of polylactic acid were reviewed in detail by Madhavan Nampoothiri et al. [5].

In research on the biodeterioration of PLA over the past two decades, researchers have primarily concentrated on bacterial or fungal degraders, with a propensity for bacterial research [4]. In order to identify families and subclasses of efficient microorganisms for PLA biodegradation, Sangwan and Wu [116,117] studied the gene sequences of both bacterial and fungal degraders on buried PLA. In one of their studies, they showed that *Paecilomyces*, *Thermomonospora*, and *Thermopolyspora* were the genera that were most prevalent in the compost samples [116]. In another study, they found that the most prevalent groups of microorganisms during the biodegradation of PLA/OMLS nanocomposites were represented by cloned gene sequences from individuals in the *phyla Actinobacteria* and *Ascomycota* [117].

A study on thirteen poly(L-lactide)-degrading microorganisms isolated from forest soils indicated that several families of bacteria, including *Thermomonosporaceae*, *Micromonosporaceae*, *Streptosporangiaceae*, *Bacillaceae*, and *Thermoactinomyceine taceae*, are able to

degrade PLA [28]. In a comprehensive investigation of 25 *Amycolatopsis* strains by Pranamuda and Tokiwa, 15 produced clear zones on agar plates emulsified with poly(L-lactide), indicating that this genus has a wide variety of PLA degraders [118]. Regarding fungal degraders, one of the most extensive studies was undertaken by Torres et al. [119], concerning 14 fungal strains commonly found in natural soils, including families such as *Aspergillus*, *Rhizopus*, *Penicillium*, and *Trichoderma*, which brought to light the effectiveness of *Fusarium moniliforme* and *Penicillium roqueforti* in the biodeterioration of PLA. Table 3 lists some of the bacterial and fungal degraders that have been researched in the literature, along with commonly used experimental methods for determining the characterization, time, and degradability percentage.

Table 3. Bacterial and fungal degraders of PLA.

	Organism	Characterization										Time (Days)	Degrad. %	Ref.
		Clear Zone	SEM	Grav. *	CO_2	DSC/ TGA	NMR	pH	Mech.	M_w *	TOC *			
Bacterial	*Bordetella petrii PLA-3*		X	X	X	X			X	X		40	4	[44]
	Amycolatopsis sp. *HT 32*	X	X	X				X			X	14	60	[120]
	Amycolatopsis sp. *KT-s-9*	X	X	X				X			X	37	86.1	[113]
	Amycolatopsis sp. *3118*			X								14	100	[114]
	Amycolatopsis sp. *K104-1*	X	X	X				X				8	>90	[29]
	Amycolatopsis sp. *41*	X						X		X	X			[121]
	Amycolatopsis orientalis subsp. *orientalis IFO 12362*	X	X	X						X	X	14	46	[122]
	Saccharothrix waywayandensis JCM 9114	X	X	X						X	X	14	44	[122]
	Saccharothrix waywayandensis		X	X				X			X	7	15	[123]
			X	X				X			X	7	95	[123]
	Kibdelosporangium aridum	X	X	X				X			X	14	97	[124]
	Bacillus brevis			X						X	X	20	≈20	[125]
	Bacillus stearothermophilus			X		X				X	X	20	30	[126]
	Geobacillus thermocatenulatus			X		X				X	X	20	≈85	[127]
	Thermomonospora sp.							X		X		28		[128]
	Stenotrophomonas maltophilia LB 2-3.		X				X		X	X		40	50	[129]
	Thermopolyspora flexuosa				X	X				X		100		[130]
	Pseudonocardia sp. *RM423*		X	X	X							28	70.9	[131]
Fungal	*Fusarium moniliforme*		X		X			X		X		7	100	[119]
	Penicillium roqueforti		X		X			X		X		7	100	[119]
	Tritirachium album ATCC 22563		X	X				X			X	14	76	[132]
	Eurotiomycetes species		X	X	X					X		60	21–27	[117]
	Aspergillus fumigatus		X						X			56	100	[133]
	Thermomyces lanuginosus		X						X			56	100	[133]
	Trichoderma viride		X	X		X				X		21	≈18	[134]

* Grav.: Gravimetry, M_w: Molecular weight characterization, TOC: Total organic carbon.

6.1.3. Poly(3-hydroxybutyrate) (PHB)

Poly(3-hydroxybutyrate) is a polyhydroxyalkanoate (PHA) and is a naturally produced polyester manufactured by many bacteria as an intracellular carbon or energy reserve [85]. PHAs are often produced by microbes from the *Alcaligenes*, *Azobacter*, *Bacillus*, and *Pseudomonas* genera [135]. *Alcaligenes eutrophus* is the most extensively utilized organism due to its high reproductive rate and substantial accumulation (up to 80% dry weight) [136,137]. In intact cells, PHB is entirely amorphous, but after extraction, it crystallizes. The copolymerization of 3-hydroxyvaleric acid with PHB frequently modifies the characteristics of PHB (3HV). Poly(3-hydroxybutyrate/3-hydroxyvalerate) copolymers

(PHBV) have been created with HV contents ranging from 0 to 90% [85]. The rate of in vitro degradation of PHB and PHBV under physiological conditions is relatively slow [138], but the rate of enzymatic degradation was demonstrated to be two to three orders of magnitude higher than the rate of simple hydrolytic degradation [139]. *Pseudomonas* [138], *Actinomadura* [140], *Penicillium lilacinus* [30], and *Streptomyces* [141] are just a few of the bacteria and fungi [142] that can both aerobically and anaerobically deteriorate PHAs. Altaee et al. [143] investigated the biodegradation of PHB films and nanofiber films derived from *Rhodococcus equi* in fertile garden soil with pH 7.30 and humidity of 80% at 30 °C for 6 weeks, concluding that all types of polymeric films were degraded to monomers and oligomers of R-3-hydroxybutyrate, which were then assimilated by microorganisms and their enzymatic activities. Table 4 highlights some of the systematic studies on the bacterial degradation of poly(3-hydroxybutyrate).

Table 4. Bacterial degraders of PHBs.

	Organism	Characterization										Time (Days)	Degrad. %	Ref.
		Clear Zone	SEM	Grav. *	DSC/ TGA	pH	XRD.	Mech	M_w *	FTIR	TOC			
Bacterial	*Alcaligenes faecalis*		X	X					X			1	68	[139]
	Microbulbifer sp. *SOL66*		X	X	X		X	X	X			2	95	[144]
		X	X	X					X	X		7	100	[145]
	Streptomyces sp. strain *MG*	X	X			X					X	3	100	[146]
	Actinomadura sp. *AF-555*	X	X							X		30		[140]

* Grav.: Gravimetry, M_w: Molecular weight characterization, TOC: Total organic carbon.

6.2. Polyolefins

Compared to aliphatic polyesters, polyolefins are less likely to deteriorate when exposed to microorganisms due to their backbone chemical structure, which consists of only long carbon chains (C–C and C–H bonds) that are more resistant to degradation than ester bonds. Polyolefins are composed of several repeating units: methylene units (HDPE), methylene and methyne units (LDPE), and a methylene and methyne group per repeating unit in the case of PP, which has an exceptionally high molecular weight (several hundreds or thousands of Daltons) [147]. Moreover, branching enhances the compaction of the chains, which in turn prevents bacteria from approaching the chains. During degradation, the C–C and C–H bonds oxidize and biodeterioration reduces the quantity of carbonyl groups and converts them into carboxylic acids, hence facilitating the oxidation of polyolefins. The polymer carbon chains are hydrolyzed into fragments during the biofragmentation phase, releasing intermediate products that include long-chain aliphatic groups including alkanes and alkenes. In the bioassimilation phase, microorganisms take up 10 to 50 carbon hydrocarbon fragments produced by biofragmentation and digest them [148]. Several different types of microorganisms capable of degrading polyolefins, from bacteria and fungi to microbial consortiums, have been isolated from places such as soil containing plastic trash, the ocean, and the digestive systems of plastic-eating worms [47,147–149]. Specifically, waxworm [47], yellow mealworm [150], and superworm [150,151] larvae have been observed to be able to digest PE, PP, and PS foams.

6.2.1. Polyethylene (PE)

Similar in significance to PLA among aliphatic polyesters, PE is the most investigated polyolefin in the literature [12,152,153]. Polyethylene is a synthetic polymer with a high hydrophobicity level and molecular weight, making it difficult for microorganisms to adhere to its surface, reducing the rate of biodeterioration. There have been a few long-term studies dedicated to tracking and characterizing the biodeterioration of the same polyethylene sample by fungal and bacterial degraders over the course of several years [154,155]. The molecular

weight of polyethylene is an important factor in its susceptibility to biodeterioration by microorganisms. Generally, PE is not biodegradable naturally, and to make it biodegradable, its degree of crystallinity, molecular weight, and mechanical properties, which are responsible for its resistance to degradation, must be modified, which can be accomplished by increasing its hydrophilicity and/or shortening its polymer chain by oxidation so that it is more amenable to microbial degradation [10].

A comprehensive study on the degradability of high density polyethylene (HDPE), low density polyethylene (LDPE), and linear low density polyethylene (LLDPE) films by abiotic and biotic factors (*Rhodococcus rhodochrous* bacteria) revealed that, regardless of abiotic or biotic approach, the average molecular weight and molecular weight distribution (measured by SEC) are directly proportional to the solubility of the PE film in the incubation media (detected by NMR) and its ability to deteriorate [156]. This is because HDPE has fewer unsaturated terminals, which makes it more resistant to the action of microbial enzymes.

Yang et al. [47] provided evidence of PE biodeterioration by two bacteria isolated from the gut of Indian mealmoths (*Plodia interpunctella* larvae) by different characterization techniques, including electrospray ionization mass spectrometry (ESI-MS) and X-ray photoelectron spectroscopy (XPS) characterization. The ATR/FTIR analysis of PE films incubated for 90 days with bacterial strains from the genera *Comamonas*, *Delftia*, and *Stenotrophomonas* revealed that the metabolic activity of these bacteria can biodegrade PE films, inducing oxidation, vinylene formation, and chain scission, among other chemical changes [157].

Bacteria from the genera *Pseudomonas*, *Bacillus*, *Brevibacillus*, *Cellulosimicrobium*, and *Lysinibacillus*, as well as *Aspergillus* fungi, were shown to be polyethylene degraders after their 16S rDNA and 18S rDNA sequences were analyzed [158]. This research showed that fungi generally outperform bacteria in the biodeterioration of polyethylene. *Aspergillus oryzae* strain A5 had the maximum fungal degradation activity, which resulted in a mean weight reduction of about 36.4%. In a study of the biodeterioration of PE by eight Streptomyces family bacteria, *S. aburaviensis*, *S. parvullus*, *S. nigellus*, and *A. flavus* exhibited a moderate degree of degradation and weight loss in PE films [159]. Table 5 summarizes some of the systematic literature on bacterial and fungal degraders of polyethylene.

Table 5. Bacterial and fungal degraders of PE.

	Organism	Characterization										Time (Days)	Degrad. %	Ref.
		Clear Zone	SEM	Grav. *	Hyd. *	DSC/ TGA	CO_2	FTIR	Mech.	M_w *	BV *			
Bacterial	*Rhodococcus rhodochrous ATCC 29672*		X					X	X	X	X	180		[156]
	Rhodococcus ruber strain *C208*		X	X			X				X	60	7.5	[160]
			X	X	X			X			X	30	8	[161]
				X		X		X			X	30	2.5	[31]
	Staphylococcus arlettae	X		X				X				30	13.6	[162]
	Serratia marcescens		X	X		X		X				70	36	[163]
	Pseudomonas aeruginosa PAO1		X	X				X	X		X	120	20	[164]
	Pseudomonas aeruginosa												11	
	Pseudomonas putida												9	
	Pseudomonas syringae												11.3	
	Stenotrophomonas pavanii	X	X	X	X	X			X		X	56	25	[165]
	Bacillus amyloliquefaciens		X	X			X	X				60	16	[73]
	Bacillus cereus		X	X				X				40	7.4	[166]
	Bacillus gottheilii												5.8	
	Brevibacillus borstelensis			X				X				20	21	[46]

Table 5. *Cont.*

	Organism	Characterization										Time (Days)	Degrad. %	Ref.
		Clear Zone	SEM	Grav. *	Hyd. *	DSC/ TGA	CO_2	FTIR	Mech.	M_w *	BV *			
Larvae	*Plodia interpunctella Enterobacter asburiae YT1*	X	X	X	X			X	X		X	60	6.1	[47]
	Plodia interpunctella Bacillus sp. *YP1*												10.7	
	Zophobas atratus			X		X		X		X	X	90	73	[151]
Fungal	*Aspergillus niger*		X			X		X				140		[59]
			X	X		X						30	8	[167]
			X			X	X	X				270		[168]
		X	X	X						X		180	40	[169]
	Aspergillus japonicus		X	X		X		X				30	12	[167]
	Penicillium simplicissimum	X	X					X		X		90		[170]
	Penicillium pinophilum													
	Gliocladium virens		X			X	X	X				270		[168]
	Phanerochaete chrysosporium													
	Aspergillus tubingensis		X	X	X			X			X	30	8.5	[45]
	Aspergillus clavatus JASK1		X	X				X				90	35	[171]

* Grav.: Gravimetry, Hyd.: Hydrophobicity, M_w: Molecular weight characterization, BV: Bacterial viability and growth.

6.2.2. Polypropylene (PP)

Polypropylene and its blends are extensively used for engineering pipelines and containers. Both PP's tensile strength and molecular weight decrease as a result of degradation. Hydroperoxides may be produced as part of the reaction, destabilizing the polymeric carbon chain to generate a carbonyl group [172]. While the biodegradability of pure and high-molecular-weight PP is still up for debate [22], the biodegradability of low molecular weight PP has been extensively researched. Jeon and Kim [173] identified *S. panacihumi* PA3-2 as an effective degrading microorganism for polypropylene (PP) with a molecular weight as high as 228,000, investigating the real biodegradability of the PPs with no additional parameters, including heat and UV stabilizers. Table 6 covers part of the systematic literature on bacterial, larval (or bacteria from the intestines of larvae), and fungal polypropylene degraders.

Table 6. Bacterial and fungal degraders of PP.

	Organism	Characterization										Time (Days)	Degrad. %	Ref.
		Clear Zone	SEM	Grav. *	Hyd. *	DSC/ TGA	NMR	FTIR	Mech.	M_w *	BV *			
Bacterial	*Bacillus flexus*		X	X	X	X		X	X		X	365	10.7	[51]
	Pseudomonas azotoformans		X	X	X			X			X	365	0.55	[174]
	Pseudomonas stutzeri												1.2	
	Bacillus subtilis												1.5	
	Bacillus flexus												2.57	
	Bacillus gottheilii		X	X				X				40	3.6	[166]
Larvae	*Tenebrio molitor Kluyvera* sp.				X	X	X	X		X	X	35		[150]
	Zophobas atratus Citrobacter sp.													
	Zophobas atratus Enterobacter sp.													

Table 6. *Cont.*

	Organism	Characterization										Time (Days)	Degrad. %	Ref.
		Clear Zone	SEM	Grav. *	Hyd. *	DSC/ TGA	NMR	FTIR	Mech.	M_w *	BV *			
Fungal	*Aspergillus niger*	X	X	X						X		180	76	[169]
	Phanerochaete chrysosporium NCIM 1170 (F1)			X		X		X		X		365	10	[175]
	Engyodontium album MTP091 (F2)												9	

* Grav.: Gravimetry, Hyd.: Hydrophobicity, M_w: Molecular weight characterization, BV: Bacterial viability and growth.

6.2.3. Polystyrene (PS)

Polystyrene (PS) is a high-molecular-weight hydrophobic polyolefin that is recyclable. Due to its light weight, rigidity, and excellent thermal insulation, PS is utilized in the production of disposable cups, packing materials, and laboratory ware. However, due to the same characteristics, it has a low biodegradability. The hydrophobicity of PS, in particular, makes it difficult for microorganisms to attach to its surface, and if adherence does occur, the high molecular weight restricts the biodeterioration ability of microorganisms.

PS is prone to oxidation at high temperatures or degradation by UV radiation from sunlight. When PS is degraded through thermal or chemical processes, it produces by-products such as styrene, benzene, toluene, and acrolein, all of which have a greater possibility of being degraded by microorganisms. Hence, in order to boost the level of degradation caused by microorganisms, PS should go through abiotic degradation first. There are few publications on polystyrene biodegradation; however, microbial (*Alcaligenes* sp. 559) deterioration of its monomer, styrene, has been reported [176].

Polystyrene is likely the material that can best demonstrate the impact of physical form on biodegradation (Section 3.2). Although this material demonstrates a relatively low degree of degradation when it is exposed to microorganisms, when it is in the form of a foam it can be easily degraded by a large number of larvae as a digestive meal, as stated in a number of published articles [58,177,178]. Regarding the filamentous fungi that degrade polystyrene, microscopic examination has demonstrated that *Curvularia* species can adhere, colonize, and penetrate the polymer structures of oxidized samples in nine weeks [179]. Some of the systematic literature on bacterial and larval degraders of polystyrene is summarized in Table 7.

Table 7. Bacterial and fungal degraders of PS.

	Organism	Characterization										Time (Days)	Degrad. %	Ref.
		Clear Zone	SEM	Grav. *	Hyd. *	DSC/ TGA	CO_2	FTIR	NMR	M_w *	BV *			
Bacterial	*Rhodococcus ruber* strain *C208*	X	X				X				X	30	0.8	[180]
	Bacillus cereus		X	X				X				40	7.4	[166]
	Bacillus gottheilii												5.8	
	Pseudomonas spp.		X			X		X	X			30	10	[181]
	Bacillus												23	
	Enterobacter sp.		X		X			X	X		X	30	12.4	[182]
Larvae	*Zophobas atratus*			X		X		X		X	X	90	38	[151]
	Tenebrio obscurus			X		X		X		X	X	31	55.4	[177]
	Tenebrio molitor												41.5	
	Tenebrio molitor Linnaeus Exiguobacterium sp. *YT2*	X	X		X		X			X	X	60	7.4	[178]
	Tenebrio molitor Linnaeus			X		X	X	X	X	X	X	16	97.4	[58]

* Grav.: Gravimetry, Hyd.: Hydrophobicity, M_w: Molecular weight characterization, BV: Bacterial viability and growth.

6.3. Polymeric Blends and Composites

Copolymerization and blending of biodegradable polymers, as well as employing these materials as components of composites, result in a wide range of properties and degradation behaviors. Blending is a significantly less complicated and time-consuming technique for producing the desired properties than the copolymerization procedure. It should be noted that the primary distinction between a polymer blend and a composite is that a polymer blend is created by combining two or more polymers to create a single phase, whereas a composite is created by combining two or more elements to create a multiphase or multicomponent system where each element exhibits its unique identity and properties, which in turn affect how the material degrades when microorganisms are present. In this section, three basic classes of composites and blends will be discussed: blends of two polymers, blends of one polymer and a natural material, and fiber-reinforced composites with a polymeric matrix.

6.3.1. Blends of Two Polymers

Since PCL and PLA already exhibit high degrees of enzymatic biodegradation in the context of two-polymeric-material blends, they are the most investigated polymers in blends with lower status biodegradable polymers in the literature.

Iwamoto and Tokiwa evaluated the biodeterioration of PCL in a blend with conventional plastics (LDPE, PP, PA6, PS, PET, PHB) by *Rhizopus arrhizus* lipase [183]. The high biodegradability of PCL was sustained in blends of PCL/LDPE and PCL/PP, but it significantly decreased in blends of PCL/PS, PCL/PET, and PCL/PHB, while blends of PCL/PA6 and PCL/PS did not show a significant change. They concluded that, in general, the higher the miscibility of PCL and conventional plastics, the more difficult the degradation of PCL in their blends by lipase. They also discovered that the enzymatic degradability of PCL/LDPE can be successfully controlled designing the composition and melt viscosity ratio of the blend to generate a continuous phase structure in the biodegradable plastic [184].

One could expect that the biodeterioration behavior of a blend of two biodegradable polymers in response to microorganisms falls somewhere between the results of the two independent polymers. However, an investigation of the enzymatic degradation of PLLA and PCL blend films by two different enzymes, namely *proteinase K* (degrader of PLA) and *Pseudomonas* lipase (degrader of PLA and PCL), revealed that the degradation by proteinase K of blends PLLA/PCL 50/50 and 75/25 was even higher than that of PLLA itself [62]. The explanation for this surprising outcome is the influence of blending on the crystallinity of the blend, which affects its biodegradability.

In some cases, when two polymers are immiscible with each other, a third component must be introduced as a compatibilizer to assist blending. For instance, the large polarity difference between polypropylene and PLLA causes them to produce an immiscible blend, and maleic anhydride-grafted polypropylene (MAPP), an efficient compatibilizer for polyolefin-based blends, is employed to improve the blending [185]. The use of compatibilizers is a two-edged sword; while they can aid with mechanical properties and biodegradability by increasing the miscibility of the two polymers, excessive usage of them can promote heterogeneity and decrease mechanical performance. As a result, the amount of compatibilizers in a two-polymer mix should be tailored to meet the mechanical and degradability criteria of the final blend.

6.3.2. Blends of Polymeric and Natural Materials

The inclusion of natural or organic materials (e.g., cellulose-based compounds) in a blend with synthetic polymers has been demonstrated to speed up and increase the likelihood of blend biodeterioration [52,68,111,186–188]. As starch offers cost performance advantages due to its renewable nature, low cost, and year-round availability, blends of synthetic polymers and starch have received the most attention in the literature. Since starch is hydrophilic and does not blend well with hydrophobic synthetic polymers such as polyesters and polyolefins, numerous solutions have been developed and proposed to

address this issue [20]. Blended starches can take the shape of granules, gelatinized starch, or even starch that has undergone chemical modification to become a thermoplastic [6]. It should be noted that including natural materials may have the drawback of reducing the physical and mechanical properties of the blend. As reported by Pranamuda et al. [189], biodegradability by lipase was found to rise with starch content in PCL/granular tropical starch blends; however, this came at the expense of a significant reduction in the blends' tensile strength and elongation.

The biodegradability of blends made with low-density polyethylene (LDPE) and polylactic acid (PLA) was greatly improved by the addition of thermoplastic starch (TPS) that contained a high glycerol content [52]. In a study on the end-of-life evaluation of extruded fibers from a blend of PLA, PCL, and microcrystalline cellulose (MCC) in different compositions under simulated composting conditions following the standard ASTM International D5338-15 protocol, it was discovered that higher MCC and PCL components and lower percentages of PLA in the test polyblends accelerated biodegradation [190]. Zhao et al. [95] investigated the biodegradation behavior of polycaprolactone and a natural lignocellulosic material (rice husk) blend in simulated soil medium, focusing on the effect of the rice husk component on composite biodegradability in terms of polymer matrix crystallinity, improved hydrophilicity, and substrate depolymerase-binding capacity. They concluded that the incorporation of rice husk can affect the crystallization of the PCL phase, as both melting temperature (T_m) and crystallinity (x_c) decrease with increasing rice husk content, resulting in significantly higher biodegradability after 57 days of incubation in comparison to the unmodified PCL. The primary driver behind this improvement is that organic materials are naturally highly degradable and easily deteriorate in the presence of microorganisms, which leads to the development of pores in the blend, increasing the surface area where the microorganisms can come into contact with the polymeric component of the blend, thereby increasing the degradation rate [52,95]. Figures 6 and 7 depict the scanning electron microscopy of biodegraded poly(lactic acid)/thermoplastic starch and low-density polyethylene/thermoplastic starch blends with varying compositions (80/20, 70/30, 60/40, and 50/50) after 14 weeks in garden compost soil investigated by Li et al. [52]. This study shows that blending TPS with LDPE and PLA in a continuous shape at a 50/50 composition significantly enhances the surface area of TPS, thus increasing the biodegradation rate of the blends relative to pure TPS.

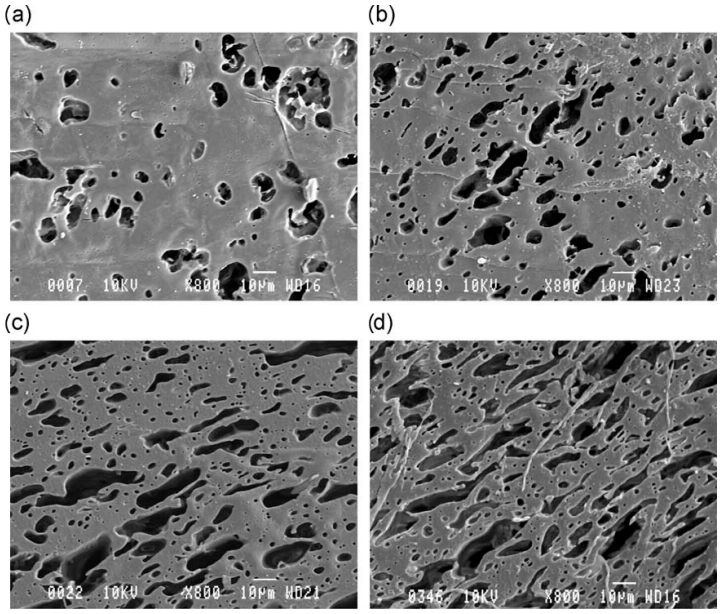


Figure 6. SEM image of PLA/TPS blends after 14 weeks incubation: (**a**) 80/20, (**b**) 70/30, (**c**) 60/40, and (**d**) 50/50. Adapted with permission from [52] John Wiley and Sons, copyright 2011.

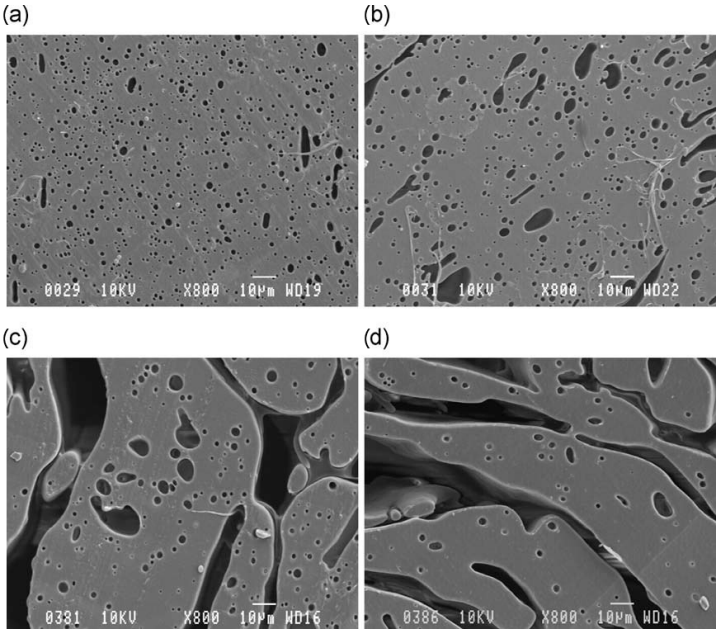


Figure 7. SEM image of LDPE/TPS blends after 14 weeks incubation: (**a**) 80/20, (**b**) 70/30, (**c**) 60/40, and (**d**) 50/50. Adapted with permission from [52] John Wiley and Sons, copyright 2011.

As mentioned before (Section 6.2), polyethylene has a lower likelihood of being degraded by microorganisms due to its high molecular weight and hydrophobicity. Therefore, adding highly biodegradable substances, such as starch, to a low-density polyethylene matrix may improve carbon–carbon backbone decomposition [191]. The starch enhances the hydrophilicity of polyethylene, allowing it to be catalyzed by amylase enzymes, which are easily degraded by microorganisms. In the presence of lignin-degrading bacteria of the species *Streptomyces* and also in the presence of the white-rot fungus *Phanerochaete chrysosporium*, biodegradation of starch polyethylene films containing a prooxidant and 6% starch also demonstrated polyethylene degradation [192]. The rate of degradation of starch-filled polyethylene was shown to be highly sensitive to environmental conditions and other chemicals in the formulation [193], as well as the oxidation of contaminants such as fats and oils [194].

Similar to the blending of two synthetic polymers, the blending of a natural material with a synthetic polymer may necessitate the inclusion of a compatibilizer as well. In an investigation on the effect of compatibilizers on the mechanical properties and biodegradability of PLA/starch blends, maleic anhydride (MA) and maleated thermoplastic starch (MATPS) were used to improve interfacial adhesion in preparing PLA/starch blends [195]. The morphological (SEM) and thermal (DSC) analysis of the blends demonstrated that MA is a good compatibilizer and boosts mechanical properties by increasing the crystallinity of the blend, whereas MATPS is not as effective for this system. Furthermore, at the same PLA/starch ratio, MA compatibilized blends demonstrated greater biodegradability than plain PLA/starch blends. In another study on PCL/tapioca starch (granular and gelatinized) blends using poly(dioxolane) as a compatibilizer, the effect of PDXL's molecular weight (M_n = 10,000 and 200,000) on the PCL/TS blends showed that the mechanical properties of PCL/TS/PDXL blends were dependent on starch content rather than the compatibilizer [196]. Using α-amylase, the enzymatic degradability of PCL/TS/PDXL blends improved as the TS concentration rose, but it was not dependent on how evenly the starch was distributed inside the PCL matrix.

6.3.3. Fiber-Reinforced Composites

Regarding the biodegradability of composites, particularly fiber-reinforced composites, significant attention has been given to composites with biodegradable polymer as their matrix and organic fiber, e.g., flax [110,196] or okra [61], as their reinforcing structure rather than durable materials such as carbon fibers. The enzymatic degradation of a flax fiber-reinforced polylactide composite by four distinct types of enzymes—lipase, protease, esterase, and proteinase K—showed that PLA dominates the biodegradability of the composite in terms of the most effective enzyme [110]. It was also confirmed that by increasing the flax–fiber content in the composite by 30%, the biodegradability of the composite by proteinase K increased from 0.7% to 11.9% after one day and from 20.9% to 51.9% after nine weeks. Another study went a step further and investigated the influence of fiber architecture on the biodegradability of FLAX/PLA composites. The comparison of morphological analyses and gravimetric measurements of composites with three different fiber architectures (quasi-isotropic, random in the x–y plane, and unidirectional) and three different fiber contents (10, 20, and 30%) confirmed that the presence of fibers and the architecture of the fiber in the composite can dramatically influence its degradation behavior, highlighting that fibers act as channels for water and the microorganisms that promote the formation of cracks and crazes [196].

7. Conclusions

This review discussed the mechanism of biodeterioration of polymers by microorganisms, as well as the environmental parameters and polymer properties that influence this phenomenon. We discussed the qualitative and quantitative methodologies and accessible standard protocols that can be used to assess the biodegradability of polymers. After going over the most recent research on the biodegradation of the two primary categories of polymeric materials (aliphatic polyesters and polyolefins), as well as their blends and composites with one another or natural materials, some key points must be brought to light.

The complex nature of the biodegradation of polymeric materials triggered by microorganisms is dependent on a wide variety of parameters, any one of which can alter the outcome. The best possible results can be obtained by first choosing the appropriate microorganism for the polymer and then creating the optimal environmental conditions (temperature, humidity, and pH) in order to maximize the activity of the microorganism. Since this phenomenon is a surface activity, increasing the adhesion of the organisms to the resistant polymer surface, which can be accomplished by applying surface-active chemicals or triggering the production of surfactant by the microorganism, will increase the probability of biodeterioration. Additionally, to promote biodegradability, it is necessary to engineer polymers with active functional groups that facilitate the first step of interaction with microorganisms.

Blending low-biodegradability polymers with other high-biodegradability polymers or natural materials that are more easily degraded is another way to boost these materials' biodegradability. To achieve the best outcome, one must choose two polymers that are degraded by the same genus or species of bacteria or fungi and engineer a continuous phase between the two substances. In this manner, under optimal environmental conditions, the bacteria or fungi will degrade one of the polymers in the blend first. This will result in greater surface contact between the microorganisms and the second polymer, which will result in an overall rise in the degradation rate.

This review emphasizes the significance of the continuous study of this subject in order to discover sustainable and environmentally acceptable alternatives to non-degradable polymers, particularly stressing the fact that very few studies have been devoted to the biodegradability of composites with polymers as their matrix, as well as the effect of fiber architecture in composites, which plays a crucial role in the biodegradability of composites.

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