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Eco-friendly additives for biodegradable polyesters: Recent progress in performance optimization and environmental impact reduction

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ABSTRACT

Biodegradable polymers have emerged as principal alternatives to petroleum-based polymers, which exert a detrimental impact on the environment. Among these, biodegradable polyesters stand out as particularly noteworthy, renowned for their exceptional properties that find application across diverse industries, including food packaging, biomedical devices, and agriculture. This comprehensive analysis delves into the impact of various additives, with a focus on eco-friendly ones, on biodegradable polyesters, as evidenced by recent research endeavors. The additives are systematically categorized to provide a nuanced understanding within distinct industrial contexts. In the exploration of these additives, a meticulous classification is employed, ensuring a focused analysis of each industry. Furthermore, this study is the first inclusive approach to categorizing additives for biodegradable polyesters. Noteworthy categories encompass hydrolysis stabilizers, oxidative and Ultraviolet (UV) stabilizers, nucleation agents, and crosslinking agents, each examined in detail within the scope of this review paper. The biodegradable polyesters under scrutiny in this review encompass PHA, PBS, and PBAT. The

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Abbreviations: DPPH, (2,2-diphenyl-1-picrylhydrazyl); ATBC, Acetyl Tributyl Citrate; ASL, Acetylated Soda Lignin; ACR, Acrylic Impact Modifier; ABS, Acrylonitrile-Butadiene-Styrene; AMPs, Antimicrobial peptides; ZIKA, Bis(2,6- Diisopropylphenyl) Carbodiimide; BDICDI, Bis(2,6-diisopropyl phenol) Carbodiimide; BS, Butadiene-Styrene; BA, Butyl Acrylate; CAPE, Caffeic Acid Phenethyl Ester; CNC, Cellulose Nanocrystalline; CAGR, Compound Annual Growth Rate; DBP, Dibutyl Phthalate; DCP, Dicumyl Peroxide; DMA, Dynamic Mechanical Analysis; ECSO, Epoxidized Cottonseed Oil; EOs, Essential Oils; EG, Ethoxylated; FDA, Food and Drug Administrator; GRAS, Generally Recognized As Safe; Tg),, Glass-Transition Temperature; g-CNC, Grafted Cellulose Nanocrystals; HALS, Hindered Amine Light Stabilizers; IR, Infrared Spectroscopy; KL, Kraft Lignin; LDHs, Layered Double Hydroxides; MA, Maleic Anhydride; MCSO, Melanized Cottonseed Oil; MOFs, Metal-Organic Frameworks; MC, Methyl Cinnamate; MIC, Minimum Inhibitory Concentrations; MDR), Multi-Drug-Resistant; NBR, Nitrile rubber; ODA, Octadecyl Amine; OMMT, Organic Montmorillonite; OA, Orotic Acid; ε-PL, ε-Poly-L-lysine; PHBV, Poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate); PALs, Poly(aspartic acid-*co*-lactide); PBAT, Poly(butylene adipate-co-terephthalate); PBS, Poly(butylene succinate); PHA, Polyhydroxyalkanoates; PHB, Polyhydroxybutyrate; PLA, Polylactic acid; PCL, Polycaprolactone; PE, Polyethylene; PP, Polypropylene; PVC, Polyvinyl Chloride; PLLA, Poly(L-lactic acid); PEG, Poly(ethylene glycol); PEG, Polyethylene Glycol; PMBS, Poly(methyl methacrylate)-poly(butadiene-styrene); PMDA, Pyromellitic Dianhydride; PMMA, Polymethyl Methacrylate; RSA, Radical Scavenging Activity; ROS, Reactive Oxygen Species; SAEO, Salvia Abrotanoides Essential Oil; SEM, Scanning Electron Microscope; AgNPs, Silver Nanoparticles; SCNC, Silylated Cellulose nanocrystals; SL, Soda Lignin; NaOH, Sodium Hydroxide; Na2S, Sodium Sulfide; TGA, Thermogravimetric Analysis; TPS, Thermoplastic Starch; TEM), Transmission Electron Microscopy; TAIC, Triallyl Isocyanurate; TAM, Triallyl Trimesate; TMC, Trimethylene Carbonate; TMPTA, Trimethylolpropane Triacrylate; UV, Ultra Violette; WoS, Web of Science; ZnO NPs, Zinc Oxide Nanoparticles; ZnS NPs, Zinc Sulfide Nanoparticles; ZrO2, Zirconiumdioxid.

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examination of these specific polymers allows for a focused and comparative analysis, shedding light on the nuanced responses to various additives within each polymer category. This review provides an in-depth discussion of the interplay between additives and biodegradable polyesters, contributing to the development of environmentally friendly materials across various industries.

1. Introduction

The widespread production of commodity plastics has led to unparalleled pollution on land and in oceans, posing a significant challenge, particularly in developing countries with inadequate waste management systems [1]. In recent years, there has been a notable surge in plastic production, driven by its cost-effectiveness, versatility, and advantageous physical properties, which have broadened its range of applications. The increasing use and slow degradation rate of plastics, however, have led to accumulation in the environment, which is recognized as a major drawback [2,3]. Annual production of plastics has increased dramatically from 1.5 million in 1950 to 348 million in 2015 [4] and almost 400 million in 2021 [5]. In 2023, global plastic production reached approximately 413.8 million metric tons and is expected to reach 1231 million metric tons annually by 2060 [6]. Approximately 50 % of plastic waste is still disposed of in landfills, with additional plastic leaking into the environment during different stages of its life cycle [7]. However, some countries such as Austria, Germany, and the Netherlands have achieved 80–100 % energy and material recovery of waste plastics [3]. Starting on July 3, 2021, the European Union (EU) implemented a ban on the market availability of single-use plastic items such as plates, cutlery, straws, balloon sticks, and cotton buds [8].

Asian nations contribute approximately 81 % of the total global oceanic plastic pollution by weight. Several efforts have been made to address the environmental issues associated with waste plastics; however, biodegradable plastics are still known as a viable and the most effective solution. Biodegradation is the process of completely breaking down a substance into simple molecules, such as water, nitrate, and ammonium (if the polymer contains organically bound nitrogen in its structure), through reduction or oxidation [9,10]. The search for a naturally biodegradable substitute for conventional plastics was a compelling subject that attracted scholars from diverse fields of study [11–13]. In recent years, biodegradable polyesters (Table 1) have attracted several industries, including food packaging, automotive, biomedical, and many others. Polylactic acid (PLA) is the most widely used at the industrial and academic levels among biodegradable polyesters. Fig. 1 shows the distribution of publications in different biodegradable polyesters in the last 8 years generated by VOS Viewer based on a database of the Web of Science (WoS). As can be seen, PLA, which is becoming more advanced in 3D printing, has been studied more by researchers in the last few years.

Over the past 50 years, production volumes of polyesters have been on the rise and are projected to continue increasing due to their versatility for numerous applications and environmentally friendly nature. Among these polyesters, Polyhydroxyalkanoates (PHA) and PLA are

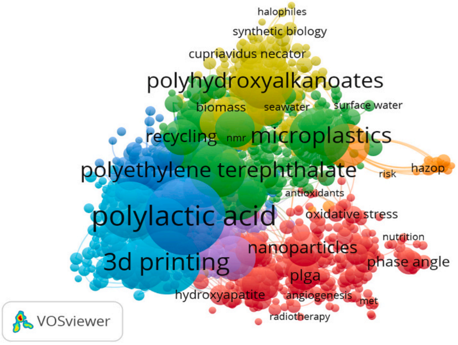
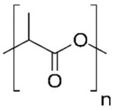
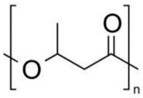
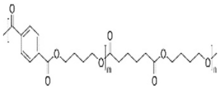
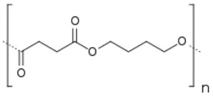
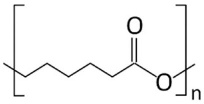


Fig. 1. Co-occurrence of the main authors' keywords, with the minimum number of occurrences of 5 for the recent publications in biodegradable polyesters.

Table 1
The main properties of the most used biodegradable polyesters.

Biodegradable polyester	Molecular formula	Structure	Key applications	T_g (°C)	T_m (°C)	σ_B(MPa)	ε_B(%)	E(MPa)
PLA	$(C_3H_4O_2)_n$		3D printing, biomedical implants, food packaging	50–80	170–180	26	64	4107
PHB	$[COCH_2CH(CH_3)O]_n$		Biomedical, biodegradable film	2–5	175–180	25	5	1800
PBAT	$H(O(CH_2)_4OCO(CH_2)_4CO)_n(O(CH_2)_4OCOC_6H_4(O)_mH$		Compostable packaging, mulch film	−28	100–120	21	350–600	87
PBS	$HO(CO(CH_2)_2COO(CH_2)_4O)_nH$		Foot packaging, textile, and agriculture films	−37	110–116	35	400	550–700
PCL	$(C_6H_{10}O_2)_n$		Drug delivery, Tissue engineering	−60	60	10–16	530	250–300

more attractive to researchers due to their biodegradability and performance. PHAs are biodegradable polymers synthesized in microbial cells as an energy store in the form of intracellular granules. Although the majority of PHA research has been conducted on polyhydroxybutyrate (PHB) and its *co*polymer with valerate Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), several varieties of monomers have been identified [14,15]. Thus, PHA is a fully biobased and biosynthesized polymer and is readily and fully biodegradable in compost, soil, and water environments.

The most prominent biodegradable polyester is PLA, which is a thermoplastic linear aliphatic polyester produced by a chemical process from lactic acid and has a high market value in the world, especially in the U.S. Lactic acid is primarily produced through the process of lactic acid fermentation using saccharide-rich plant materials, with corn and sugarcane being the predominant sources [16]. PLA is manufactured at an industrial scale, enhancing its appeal within the bioplastics sector due to its cost-effectiveness and widespread availability.

Unlike PHB, PLA is readily biodegradable only under controlled industrial composting conditions. PLA biodegradation in soil or water environments is relatively slow and does not comply with the related international standards [17,18]. Studies indicate that PLA microbial mineralization must be preceded by the chemical hydrolysis of ester bonds in the polymer, which occurs rapidly at temperatures above 55 °C [19]. Recent claims suggest that PLA can be manufactured to be biodegradable in soil; however, this topic remains unresolved [20].

The properties of pure PHA, PLA, and other biodegradable polyesters are far from perfect. As they are intended for various applications, improving and optimizing their properties is essential to meet specific application requirements. In polymer science and engineering, this is usually accomplished using additives. Still, over time and with the help of different techniques such as additive development, they were transformed into versatile and high-performance materials as we know them now.

Although additives have been used in this area for several years, another priority has been added in recent years. Initially, the focus was solely on improving the target polyester; however, due to stringent regulations in different industries, the selection of appropriate additives also emphasizes their eco-friendly properties. Recent studies have tried to review these additives from different views and categories. However, a comprehensive review categorizing additives based on their intended function, with a particular focus on eco-friendly options, is still lacking.

Table 3 summarizes the latest review in this area. As indicated by the publication date and the number of recent studies in that review, the need for further research is strongly emphasized. This review is unique in its attempt to provide a holistic analysis of additives and highlight their effects on the properties of polyesters. This is important because adding an additive can sometimes introduce new issues that need to be resolved. This review is the first to systematically investigate different types of additives, particularly eco-friendly ones, in polyesters across various industries. By doing so, this study enables researchers to determine the optimal use of each additive type and develop more effective strategies for enhancing the performance of biodegradable polyesters. Moreover, each industry has its own standards, making it different from other industries in terms of the use of additives. In this regard, this comprehensive review is further important to look at this difference at the end, making this study important not only for academic researchers but also for those who research on an industrial scale.

Conducting this latest systematic review is crucial in light of the rapid advancements in polyester additives. As outlined in Table 2, the focus is on examining the potential of these additives to address those issues while being eco-friendly. The primary objective of this study is to thoroughly examine the most recent research advances in the use of eco-friendly additives to improve the performance of commonly used and advantageous polyesters.

Table 4 summarizes additives used by researchers to address the issues of different biodegradable polyesters. This comprehensive table divides the additives by their category and the results of the try trigger. The subsequent sections will provide a detailed discussion of each category. As can be seen, PLA attracts most research studies based on their application in various industries. This table shows the most used additives in each category. For instance, lignin is commonly used as an antioxidant due to its high performance, environmental impact, and low price and Joncryl ® ADR-4368 as chain extender in biodegradable polyesters. This table can help further show the additives that can be used in different categories. For instance, essential oils can be used as natural antimicrobial agents and plasticizers. In addition, these additives will be discussed in more detail in the following sections.

Table 2
Research questions and motivations of the systematic review.

Research questions	Explanation
RQ1: What are the main drawbacks of biodegradable polyesters that reduce their application?	Biodegradable polyesters are among the highly interesting polymers; however, the mechanical and thermal properties of most of them, along with the high degradability of some, need to be improved to make them more widely used.
RQ2: Can the blending of additives improve their performance?	Depending on the purpose of each blending for improving one property, other properties could be influenced negatively or positively. The final product and its properties determine whether or not the use of additives can improve their application. It is crucial to determine if blending additives is a suitable way to reduce their drawbacks.
RQ3: Which type of additive from which category can improve the intended function yet is eco-friendly?	Each biodegradable polyester has its own drawbacks in specific industries. First, the industry in which a specific polyester is intended to be used should be identified. Then, the drawbacks of polyester should be studied for further improvement, making it suitable for that industry. These steps cause an increase the application of biodegradable polyesters in different industries

Fig. 2 depicts various additives used in polyesters to enhance their performance. Antimicrobial additives such as essential oils, peptides, and antibiotics are utilized to inhibit microbial growth in applications such as food packaging and biomedical devices. Antioxidants such as lignin and CAPE prevent oxidation, thereby increasing the durability and resistance of the material, particularly in environments prone to degradation [150]. Plasticizers like ATBC, citrate derivatives, and PEG improve flexibility and reduce brittleness, both of which are crucial in packaging and consumer goods. Conversely, crosslinkers such as TMPTA and DCP strengthen the molecular structure to enhance the mechanical properties of polyester. Similarly, chain extenders such as PMDA and Joncryl-type compounds restore or reinforce polymer chains, thereby enhancing their resilience [151,152]. Impact modifiers, such as acrylate rubber particles, enhance toughness and reduce brittleness in products. Enhanced mechanical strength is required for various applications in the construction and automotive industries. Nucleation agents, such as talc and sodium stearate, regulate crystallization to optimize thermal and mechanical stability. Lastly, hydrolysis stabilizers protect the material from degradation caused by moisture, extending its lifespan in environments such as agriculture or food storage. These additives together allow polyesters to be tailored for diverse applications, from biodegradable bags to biomedical devices and textiles.

Fig. 3 shows the paper that was already published in this research study in the last 10 years, and additives are more used for these biodegradable polyesters. The database for this figure was generated using VOS Viewer, based on the WoS database with keywords such as PHB, PLA, PBS, additive, and industry. As shown, mechanical properties are the primary focus of researchers, as they are one of the most crucial

Table 3
Constraints of previous review studies.

Parameters	[21]	[22]	[23]	[24]	[25]	[26]	[27]	[28]	[29]	This Paper
Studying biodegradable polyesters or some of them	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓
Crosslinkers and chain extenders for biodegradable polyesters	×	✓	×	×	×	✓	×	✓	✓	✓
Antimicrobial additives in biodegradable polyesters	×	×	✓	×	×	×	×	✓	×	✓
Plasticizers in biodegradable polyesters	×	×	✓	×	✓	✓	×	✓	✓	✓
Impact modifiers in biodegradable polyesters	×	×	✓	×	×	×	×	✓	×	✓
Oxidative and hydrolysis stabilizers in biodegradable polyesters	×	✓	×	×	✓	✓	×	×	✓	✓
Nucleation agents in biodegradable polyesters	×	×	×	×	×	×	×	✓	✓	✓
Influence of additives in different industries	×	✓	×	✓	×	×	×	×	✓	✓
Comprehensive overview of the influence of additives on each other	×	×	✓	×	×	×	×	×	×	✓
The most recent research studies (from the last two years based on the current date and the date that the review accepted)	✓	×	✓	✓	×	×	×	×	×	✓

aspects requiring improvement in these polymers. Additionally, bio-based components were among the most frequently used keywords in this field. Furthermore, biomedical applications and bio-composites are among the most frequently mentioned topics. However, Fig. 1 indicates that 3D printing, which heavily relies on PLA, also requires further advancements in this field. In conclusion, a comprehensive review on eco-friendly and bio-based additives in these polyesters holds significant value.

2. Hydrolysis stabilizers

Polymer hydrolytic degradation is defined as the scission of chemical bonds in the polymer structure by water absorption, a process that ultimately breaks down the polymer chains into smaller oligomers and, eventually, individual monomers [153,154]. In this type of degradation, water molecules first attack the water-labile bonds by either imbibition into the polymer matrix, followed by bond hydrolysis, or direct access to the polymer surface [155]. Hydrolysis can also be catalyzed via water nucleophilic attack (neutral hydrolysis) and by an enzyme, acid, or base that could have a significant influence [156]. The ability of ester linkages to go through hydrolysis processes has made polyesters an exciting alternative to petroleum-based polymers. Among all the available degradation processes, hydrolytic is an interesting step that allows closed-loop recycling processes and is considered one of the best options to eliminate plastic linkages to chemical hydrolysis and microbial/enzymatic chain cleavage reactions. Crystallinity, molecular weight, chemical structure, and other factors mentioned in Fig. 4 are the most important properties that should be considered in studying hydrolytic degradation.

Their hydrolysis degradation should be controlled to make the biodegradable polyesters able to be used in packaging for food, beverages, and agricultural films. The extent of hydrolysis can be reduced by decreasing the hydrolytic reaction rate, mitigating the effects of autocatalysis, or preventing water penetration. Several attempts have been made to control polyester hydrolysis, and it has been suggested that the rate of PLA hydrolysis is influenced mainly by the degree of crystallinity and molecular weight [157,158].

Biodegradable polyesters such as PLA and PBS are prone to hydrolytic chain scission when exposed to moisture, which is autocatalyzed by carboxylic end-groups generated during degradation [159]. Hydrolysis stabilizers mitigate (i) scavenging or deactivating water in the polymer, and (ii) neutralizing acidic chain ends. Different additives are used in this regard to enhance the hydrolysis stabilization of PLA and PHB. Polyester hydrolysis stabilizers are additives widely known as water scavengers containing functional groups. Table 4 highlights the most used additives in this area. One of those types of additives is Carbodiimide, which is eco-friendly [30]. The key advantage of carbodiimides is that they not only bind carboxyls but also directly consume some of the infiltrating moisture (forming urea derivatives). Therefore, they attack both aspects of hydrolysis [159].This additive showed an efficient stabilizing of PLA, suppressing chain scissions of ester bonds during abiotic hydrolysis, especially at concentrations above 1.5 % *w*/w [30]. Previous researchers have shown that carbodiimide could react with substances with low molecular weight and the moisture-stabilizing carboxyl or hydroxyl groups when it blends with polyesters and retard the hydrolysis degradation of PLA [160]. In this process, BDICDI or ZIKA reacts with water and carboxylic acids, leading to the conversion of imine bonds into amide bonds [157]. The incorporation of carbodiimides has been shown to prolong the lifetime of biodegradable polyesters, such as PLA, by suppressing the initiation of hydrolytic degradation during melt processing. Carbodiimides have also been observed to support the crosslinking of PLA chains, further enhancing the polymer's resistance to degradation. Furthermore, the addition of carbodiimides can extend the lag phase preceding the onset of polyester hydrolysis [49]. This is attributed to the ability of carbodiimides to react with the small-molecule carboxylic acids formed due to ester bond scission at the polymer chain ends during the initial stages of degradation. As carbodiimides are highly reactive towards carboxylic acids, they can effectively suppress the autocatalytic effect associated with the formation of these degradations by-products in PLA. The inhibition of the autocatalytic degradation mechanism by carbodiimides plays a crucial role in extending the lifespan of biodegradable polyesters [161].

Despite their advantages, carbodiimides are sacrificial additives. They are consumed as they react with water and acids, and their stabilizing effect diminishes over time. High loadings (>1–2 wt%) are often required to observe long-term benefits [30] [], which can be an expensive option given the relatively high price of these additives. Moreover, if the moisture ingress is high enough, carbodiimides may be depleted before the intended service life ends. Another limitation is that additives that react only with carboxyl end-groups but not with water (such as epoxy-based chain extenders) show little to no improvement in long-term hydrolysis stability [162]. This is because water can still freely attack the polymer, so capping the acid ends alone is insufficient.

Blending polyesters with each other has been tried by several researchers to eliminate some of their drawbacks, including the hydrophobicity of some of them, like PHBV [159,163]. For example, PBAT has been employed to address this issue in PHBV, as it is known for its hydrophilic properties among biodegradable polyesters [164].

In the quest for sustainable hydrolysis stabilizers for biodegradable polyesters, this comprehensive review evaluates and compares the efficacy of diverse additives presented in Table 5. Among the additives, PAL stands out for its economic viability and its unique ability to modulate the biodegradation rate in PLLA/PAL blends, particularly in aqueous environments. On the other hand, PMMA demonstrates high transparency, Ultra Violette (UV) resistance, and intrinsic mechanical properties, effectively retarding degradation, especially on the material surface. TMC proves intriguing for PCL, showcasing a suitable hydrolytic degradation rate and a low melting point, though the inferior hydrolysis capacity of the carbonate linkage is noted. TMPTA emerges as a versatile option, negligibly affecting other properties while enhancing hydrolysis time and overall hardness in polyester formulations. Another important factor to consider is their environmental friendliness, with PAL being the

Table 4
An overview of the additives used by researchers in different categories.

Base polyester	Type of additive	Name of additive	Blending Purpose	Results	Author (s) [Reference]
PLA	Anti-Hydrolysis	Bis(2,6-diisopropyl phenol) Carbodiimide (BDICDI)	Blending of carbodiimide additive with PLA to control hydrolytic stability and biodegradability.	BDICDI was a suitable stabilizer of PLA-suppressing chain scissions of ester bonds through abiotic hydrolysis.	Stloukal et al. [30]
	Nucleation agent	Talc, sodium stearate, and calcium lactate	Study the nucleating ability of three different agents (talc, sodium stearate, and calcium lactate) and find the most optimal one.	Talc was an effective nucleating agent, and in comparison, with other agents, it is the most optimal one, while sodium stearate and calcium lactate had little or negligible nucleating ability in the blend.	Li and Huneault [31]
	Nucleation agent	Cellulose nanocrystalline (CNS) and Silylated Cellulose nanocrystals (SCNC)	Study of the influence of nucleation agent on morphology, non-isothermal and isothermal crystallization behaviours.	The crystallinity increased by a low amount of SCNC, but CNC did not have a high influence. PLLA/SCNC composites nanostructured with a highly dispersed nanocrystal phase more than PLLA/CNC. Moreover, high crystallinity causes to enhance tensile modulus and nanocomposite strength.	Peri et al. [32]
	Plasticizer	Polyethylene Glycol (PEG)	Study of the influence of the addition of PEG plasticizer on the PLA mechanical properties	Miscibility range of the PLA with PEG reduced by enhancing the Mw of plasticizer (PEG). PEG plasticizing efficiency enhanced by reducing PEG Mw. It caused to enhance the PLA elongation at break significantly.	Baiardo et al. [33]
	Plasticizer	Melanized Cottonseed Oil (MCSO)	Investigation of the effect of MCSO on thermal, mechanical, barrier, and morphology properties of the polymer	The results showed that although this plasticizer had a low plasticizing effect on the polymer, it enhanced the mechanical properties. For example, enhanced elongation at break from 4 to 16 %. It caused a small reduction in Tg.	Carbonell-Verdu et al. [34]
	Impact modifier	ultrafine acrylate rubber particles	Investigation of the application of ultrafine acrylate rubber particles to reduce the problem of brittleness in PLA and the influence of this rubber material on toughness	The results showed that although in the previous research the high content of the toughness agent caused a good enhancement in toughness, just 1 % of their additives triggered a significant enhancement in the toughness of the PLA.	Petchwattana et al. [35]
	Impact modifier	Acrylic Impact Modifier (ACR), is one kind of methyl methacrylate–butyl acrylate (BA) copolymer with a core–shell structure	Study the influence of using ACR as an impact modifier and PMMA as an additive to improve polymer properties in PLA.	ACR copolymers with various core–shell ratios blended with PLA showed a significant influence on toughening PLA. Moreover, their results showed that blending PMMA with high content with ACR caused better dispersion on the PLA matrix.	Li et al. [36]
	Crosslinking agent Chain Extender	Triallyl Isocyanurate (TAIC) SR533 Joncryl® ADR 4368	Study of the effects of two different types of additives (crosslinking agent and chain extender) on the polyesters on thermal, rheological, and tensile properties of gamma-irradiated PLA.	The rheological results exhibited that irradiated PLA had a lower shear viscosity than that of neat PLA. The blending of the Cross-linking Agent or Chain Extender into PLA sharply increased the shear viscosities of neat and irradiated PLA. Moreover, the influence of the Crosslinking Agent on the enhancement of tensile properties was shown to be higher than that of the chain extenders	Hachana et al. [37]
	Antimicrobial Agent	Spruce resin, ZnO, and essential oils (thyme oil and clove oil)	To improve the antimicrobial property of PLA and enhance physical characteristics	Essential oils had an effective influence on the inhibition of *E. coli* and *S. aureus* bacteria while increasing the water vapor properties of the composite. Spruce resin and ZnO, separately and in combination, showed great antimicrobial activity, and PLA film with essential oils showed the highest antimicrobial properties. All films showed high UV stabilization. ZnO increased tensile strength, and essential oils could act as plasticizers, increasing the flexibility	Yaman et al. [38]
	Antioxidant	Acetylated soda lignin (ASL) and non-acetylated soda lignin (SL)	To impose antioxidant activity to PLA for potential biomedical applications	After lignin acetylation, high compatibility of lignin and PLA was achieved. Pure PLA showed the highest tensile strength, and 5 % of SL	Mearaj et al. [39]

(*continued on next page*)

Table 4 (*continued*)

Base polyester	Type of additive	Name of additive	Blending Purpose	Results	Author (s) [Reference]
				showed the highest elongation at break compare to higher percentage (up t 20 %) and all contents of ASL. SL showed the highest radical scavenging activity (RSA), increasing the content results in higher RSA. 80 % RSA resulted from blending 20 % of SL.	
PHB	Branching crosslinking	DCP initiator and Triallyl Trimesate (TAM) provided	Investigation of modifying the chain architecture of PHB and study of the properties, modulus, and thermal stability of the PHB	The rheology results revealed that these additives showed an effective branching/crosslinking agent for PHB. The results showed a considerable enhancement in the crystallization temperature, finer spherulitic structure, faster crystallization kinetics, and higher thermal stability.	Kolahchi and Kontopoulou [40]
	Anti-Oxidant	Caffeic Acid Phenethyl Ester (CAPE)	Study of the influence of the Oxygenate stabilizer and antimicrobial additives on PHB application and its performance	The result was that CAPE incorporated in PHB was in an amorphous state. This state is favorable for use in drug dosage. Moreover, the sample with CAPE showed better anti-oxidant activity than that without CAPE.	Ignatova et al. [41]
PBS	Crosslinking agent	Dicumyl Peroxide (DCP)	Blending a crosslinker into PBS, to improve its properties, especially elasticity and mechanical strength, by blending an organic peroxide, DCP	By blending DCP with PBS, the PBS crosslinked sufficiently, and the gel content was enhanced by increasing the DCP percentage. Although the addition of DCP did not significantly affect tear strength, Crosslinked PBS increased tensile properties.	Kim et al. [42]
	Anti-Oxidant	bis/trisphenols that derived from ferulic acid	Study to improve the stability of the polyester against oxidation stabilizer.	Stability of molecular mass average, but there is no differences in the structure of the PBS. Moreover, their results showed that the activity of this anti-oxidant is slightly more than a petrochemical anti-oxidant named Irganox 1010.	Reano et al. [43]
	Nucleation agent	Orotic Acid (OA)	Study of the influence of OA as a nucleation agent on PBS crystallinity	Adding OA to the composites can enhance the crystallization of PBS in both isothermal and non-isothermal conditions, and it is known as a sufficient nucleation agent for PBS.	Lee et al. [44]
Poly(L-lactic acid) (PLLA)	Hydrolytic degradation controller	Poly(aspartic acid-co-lactide) (PALs)	Study of the effects of the PAL composition on the blend structure and the kinetics of hydrolysis	PAL did not accelerate the PLLA hydrolysis in air. Blending PAL in water significantly increased the degradation of PLLA; moreover, with increasing PAL concentration, the degradation rate constant increased linearly.	Oyama et al. [45]
Poly(ethylene terephthalate) (PET)	Anti-Hydrolysis Anti-Oxidants	Polycarbodiimide (Stabaxol® P100) IrganoxB561® Pentaerythritol tetrakis(3-(3,5-ditert- butyl-4-hydroxyphenyl) propionate	Investigation of the influence of different additives on thermo-oxidative and hydrolytic stabilization of recycled post-consumer poly (ethylene terephthalate)	Antioxidant additives cause an increase in the intrinsic viscosity of recycled PET for dry material. The result showed that the anti-hydrolysis agent had a predominant influence on enhancing intrinsic viscosity when combined with other variables. Although this additive is known as an anti-hydrolysis, the polycarbodiimide is considered a chain extender.	Freitas et al. [46]
Poly(butylene adipate-co-terephthalate) (PBAT)	Anti-Oxidants	Kraft Lignin (KL)	Evaluate the thermostability of kraft lignin as a natural antioxidant when incorporated into the PBAT matrix	Thermogravimetric Analysis (TGA) confirmed lignin antioxidant activity in an oxidative condition. Blending Lignin enhanced the T10% of pure PBAT, and it can be concluded that KL incorporation inhibits oxidation. On the other hand, there is no big difference in biodegradation rate between samples at higher temperatures.	Tavares and Rosa [47]
	Antimicrobial Agent	Clove essential oil	Study the influence of essential oil on the antimicrobial activity of PBAT for strawberry preservation	The blending of this essential oil provided significant inhibition against *E. coli and S. aureus.* The influence on *S. aureus* was greater than *E. coli*. This natural essential oil, which has a high antimicrobial capacity, can be used in antimicrobial food packaging.	Xue et al. [48]

(*continued on next page*)

Table 4 (*continued*)

Base polyester		Type of additive	Name of additive	Blending Purpose	Results	Author (s) [Reference]
Composite	PLA/ Wood flour	Anti-Hydrolysis	Bis(2,6- Diisopropylphenyl) Carbodiimide (ZIKA)	Study of the blending effects of ZIKA in the PLA/WF composites to investigate the influence of this additive on the hydrolysis stabilization of the polymer	Thermal and mechanical analysis, as well as experiments under accelerated hydrolytic conditions, revealed ZIKA's anti-hydrolysis influence in the PLA/ WF blend. Moreover, the results showed that ZIKA was capable of delaying the onset of polymer chain cleavage in the PLA/WF-ZIKA composite.	Holcapkova et al. [49]
	PLA/ PBAT	Chain Extender	Joncryl ® ADR-4368	Investigation of the biodegradation process of PLA/PBAT blends compatibilized with an epoxy (Joncryl® ADR-4368) as a chain extender in the soil under laboratory conditions.	The experiment showed a significant complexity of the biodegradation process of PLA /PBAT when compatibilized with a chain extender. The functional groups of chain extender reacted with the groups generated through the biodegradation in a competitive effect with molecular weight reduction, triggering retardation in biodegradation.	Palsikowski et al. [50]
		Nucleation agent	Talc	Study of the effect of Talc as a nucleation agent on thermal and morphological properties, especially the crystallization rate of PBAT/PLA blend films.	Increasing in density of nucleation and spherulitic growth with blending PBAT and talc, showed that talc was a sufficient nucleating agent for PLA and its influence is even more by adding PBAT.	Phetwarotai and Aht-Ong [51]
	PLA/ PBS	Plasticizer	PEG	Investigating the influence of PEG as a plasticizer on thermal, rheological and mechanical properties of the PLA/PBS blends	PEG into PLA/PBS caused a significant reduction in the Tg. The results showed that PEG prefers to migrate to the PLA phase more than PBS. The PEG increased the molecules' mobility of the mixture. Moreover, they concluded that this additive reduced the Young's modulus and tensile strength	Pivsa-Art et al. [52]
	PLA/ PHB	Plasticizer	Acetyl Tributyl Citrate (ATBC) & n-Butyryl tri-n-hexyl Citrate Molecular	Study of the influence of different types of plasticizers on morphological and thero-mechanical and properties of PHB/PLA	Plasticizers enhanced the elongation at break. However, the influence of ATBC is extremely higher than n-Butyryl tri-n-hexyl Citrate. They also concluded that a low molecular plasticizer in the blend improved the macromolecular chains mobility. As a result, the melting temperatures and crystallization reduced in comparison with PHB/PLA.	Mencik et al. [53]
	PLA/ Clay	Impact modifier	Ethoxylated (EG) terpolymer	Investigation the influence of impact modifier on properties of PLA/Clay composite and compare the properties with the pure polyester	Blending EG terpolymer with PLA/ clay bio composites caused a considerable enhancement in the impact strength in comparison with PLA/Clay without this agent. They reported that this influence can attributed to the chemical reactions between either carboxyl or hydroxyl groups of PLA and the epoxy groups of the used impact modifier to form a copolymer at the interface.	Cavalcanti et al. [54]

most eco-friendly among those mentioned. However, PMMA is still widely used due to its performance and desirable properties, which have motivated researchers to make it more eco-friendly [165]. For this reason, PMMA is widely used as a stabilizer for PCL and PLA in the 3D printing industry, rather than in biomedical applications.

The comparative analysis delves into each stabilizer's specific advantages, drawbacks, and applications, offering insights to guide the development of next-generation hydrolysis stabilizers for biodegradable polyesters. This exploration aims to inform researchers and industry professionals in pursuing sustainable innovations, providing a nuanced understanding of the diverse stabilizer options available and their potential impact on the performance of biodegradable polyester materials. A promising direction involves the development of hybrid stabilizers or the incorporation of naturally derived compounds with inherent water-repelling properties. Novel multi-functional additives such as aziridine derivatives have been shown to not only bind moisture but do so repeatedly by ring-opening reactions, offering prolonged protection compared to conventional carbodiimides [159].There is also interest in bio-based stabilizers – for instance, polyfunctional natural extracts or polymers that sequester water or neutralize acids without leaving persistent residues. The development of such stabilizers aims to balance improved shelf-life with the need for eventual biodegradation, which enables "tunable" lifespans for biodegradable polyesters by adjusting additive type and concentration.

3. Oxidation and UV stabilizers

Polymers, especially polyesters, are typically exposed to oxidative degradation during their life, affecting their performance and thermal and mechanical stability, especially for long-term applications

Table 5
An overview of the additives used by researchers as hydrolysis stabilizers.

Hydrolysis stabilizers	Common target polyesters	Advantages	Further information	Ref
PAL	PLA, PBAT, PBS	To make it, we don't need any catalysts or solvents, which makes it an economical additive.	Although PAL has a negligible influence on the hydrolysis of PLLA in air, hydrolysis suddenly starts when PLLA/PAL is immersed in water. PAL's ability to tune the rate of biodegradation in PLLA/PAL made this polymer even more applicable in medicine	[,55][45]
Polymethyl Methacrylate (PMMA)	PLA	High transparency, UV resistance, and intrinsic mechanical properties.	This additive can retard the PLA and PLLA degradation, especially on the surface, due to its higher crystallinity than PLLA, making the blend more resistant. It can create a material with different morphology and high mechanical entanglement of molecular chains	[56,57]
TMC	Polycaprolactone (PCL)	Its suitable hydrolytic degradation rate and low melting point make it an interesting additive for incorporation into the polymer matrix.	To be more specific, it can be noted that the hydrolysis capacity of the carbonate linkage is inferior to that of the ester linkage. But, the crystallization capability of PCL) is significantly superior to that of Trimethylene Carbonate (TMC).	[58,59]
Trimethylolpropane Triacrylate (TMPTA)	PLA, PBS, PHB	A suitable additive as it has negligible influence on other properties.	Enhancing hydrolysis time causes general hardness and can increase the plastic hardness of blended and pure polyester	[46,60,61]

Table 6
An overview of the additives used by researchers as nucleation agent.

Nucleation agents	Common target polyester	Advantages	Further information	Ref
OMMT platelets	PLA, PHB, PBS	Large specific surface area, excellent compatibility with polymer matrices during melt blending, strong thermal stability that ensures it remains in a solid phase during polymer crystallization, and a large interface area that facilitates crystal growth.	It offers more nucleation sites and encourage heterogeneous nucleation in the higher temperature range, which is favorable for polyester during the extrusion.	[62,63,64]
Thermoplastic starch (TPS)	PBAT, PLA, PHB, PBS	High tensile strength, elongation at break, and secant modulus	It causes reduction in tensile strength, secant modulus, and elongation at break. It is more efficient to be used in a small content.	[65–67]
inorganic nucleators, such as clay	PLA, PHB	The majority of them are not only non-toxic and improve biodegradability, but they are also cheaper than the majority of nucleators	They may increase energy consumption due to the enhancement in the polymerization temperature, which also increases the degradation risk. Organic agents lead to lower polymerization temperatures, which is more required, especially for PBS, due to its lower melting point, which can reduce the thermal degradation rate.	[68–72]
Layered Double Hydroxides (LDHs)	PLA, PHB	Simple to synthesize, their ability to increase the crystallinity, reduce the crystallization time, enhance the composite crystallization temperature, reduce the polymer nucleation free energy and the surface free energy	An inorganic chemical class with a layered structure made up of negatively-charged hydroxide anions and positive-charged metal cations. Weak electrostatic interactions hold the layers together, while interlayer anions or water molecules normally separate them.	[73,74]
Cellulose nanocrystalline (CNC)	PBAT, PLA, PHB, PBS	Excellent crystallinity, and tiny size of CNC, their low price with high performance regarding the mechanical properties	Natural cellulose, which is present in plants, is the source of CNC. Their price made them interesting choice to be used as filler for polyesters.	[75–77]

[166,167]. A number of variables, including chemical structure, oxygen penetration, morphology, and molecular weight, affect the oxidative degradation of polyesters. The main factors that influence oxidative degradation are mentioned in Fig. 5. In this regard, various techniques, such as oxidative stabilizers and UV stabilizers that can serve as protective materials for polymers, have been devised by researchers to mitigate polymers' deterioration, which is discussed here.

3.1. Oxidative stabilizers

The oxidative degradation mechanism of polyesters occurs in two main steps: 1) reduction in molecular mass due to random chain scission, and 2) predominant production of anhydrides [168]. Three mechanisms are associated with oxidative degradation in biodegradable polyesters, starting with initiation, which starts with the generation of free radicals, and in polyesters, ester bonds are susceptible to hydroperoxide (ROOH) formation that will decompose into reactive radicals (R•). This step follows the reaction of those free radicals with oxygen to form peroxy radicals (ROO•) that further cause hydrogen abstraction, forming hydroperoxides (ROOH) and new alkyl radicals (R•) and hydroperoxide decompose later into alkoxy (RO•) and hydroxyl (HO•) radicals. The continuity of this cycle triggers chain scission and oxidative degradation, and chain reaction effectively ends by the combination of these radicals in the termination mechanism [169,170]. Antioxidants can retard oxidation degradation by two primary steps; radical scavenging, where they donate hydrogen to free radicals to make them stable and decompose hydroperoxide into non-radical products [171,172].

To eliminate this step of degradation or retard this process, blending even a low concentration of antioxidants or oxidation stabilizer is required [173,174]. For biodegradable polyesters to be used in industries like agriculture (e.g., mulch films), outdoor furniture, and automotive parts, the use of oxidative stabilizers is crucial. These stabilizers can be classified into various types.

Several researchers have used lignin as an eco-friendly, inexpensive natural antioxidant for polyester modification [175,176]. They incorporated organosolv, soda, and KL, with various concentrations, into the polyesters. Soda lignin is obtained by treating wood chips with sodium hydroxide (NaOH) and sodium sulfide (Na_2S), while organosolv lignin is extracted using organic solvents like ethanol or methanol. Kraft lignin is a type of lignin that is obtained through the kraft pulping process. These

Table 7
An overview of the additives used by researchers as plasticizer.

Plasticizer	Common target polyester	Advantages	Further information	Ref
Glycerol	PLA, PHA, and PBS	Enhancing the space between chains and reducing intermolecular forces, low toxicity and volatility	The majority of them such as PEG are biodegradable, widely used specially for PLA.	[78–83]
Dibutyl Phthalate (DBP)	PHB	Good molecular structure, molecular weight, and chemical and physical properties, such as flexibility and durability	DBP helps to increase the distance between the polymer chains, making the plastic more flexible and less brittle	[84–88]
TPS	PLA and PBAT	It is biodegradable and renewable, its high processability and good in mechanical properties improvement	The characteristics of mixes of PBAT and TPS were discovered to be impacted by the discovery that TPS with urea displays considerably better tensile strength and modulus than TPS with starch plasticized by glycerol.	[89,90]
ATBC	PHB and PLA	Low toxicity, high compatibility with polyesters, and its biodegradability that make it applicable in biomedical and food packaging industry	It has the potential to reduce the meting point of PHB and PLA and increase the crystallinity of PHB at longer annealing time due to the positive effect of plasticizer on the molecular mobility and crystallinity	[91–94]
Epoxidized natural oils	PLA, PHB, PBAT	Environmentally friendly, their flexible properties, processability, high compatibility, etc. The epoxide groups on the plasticizer molecules can create hydrogen bonds with the carbonyl groups on the polymer chains, allowing the polymer chains to move more freely.	Epoxidized Chia seed oil, soybean oil, Karanja oil, are just a few examples of used plasticizers of this type. The other benefit of this additives is to use them as a compatibilizers for mixing two polyesters by using a reaction between hydroxyl and epoxy groups.	[95–98]

additives enhance oxidative degradation and matrix thermal degradation temperatures and, hence, improve the stability of polymers. These additives could be interesting for researchers as they can have an antimicrobial impact even at low concentrations [175,177]. It is worth noting that there is no significant difference between the effects of low and high concentrations of certain lignin; nevertheless, researchers aim to use high concentrations of lignin to enhance the mechanical properties despite its cost [178,179]. Lignin is a complex, hydrophobic due to aromatic rings and hydrophobic functional groups (like methoxy and phenolic groups), and structurally resistant polymer, which makes it inherently less susceptible to microbial attack [180,181]. For PBSA, which degrades mainly through hydrolysis followed by microbial action, lignin's hydrophobicity can help reduce water absorption, resulting in hydrolysis stabilization of the polymer matrix. For PBS, Zhang et al. [182] showed that by adding lignin to PBS, tensile strength and Young's modulus increased when lignin content didn't exceed 20 % based on the compatibility of lignin with PBS.

The lower concentration of lignin is considered optimal since there is no significant disparity in the anti-bacterial and anti-oxidant efficacy of the additive between high and low lignin concentrations. For instance, Wang et al. [183] and Shan et al. [177] concluded that 5 % and 3 % lignin in PLA-based composite can result in a high degree of antimicrobial activity (based on phenolic compounds in their structure) by inhibition of different bacteria such as *Escherichia coli* and *Staphylococcus aureus*. However, regarding thermal and mechanical properties such as tensile strength, a high concentration of lignin would be more beneficial [184]; owing to the low aspect ratio of this aromatic polyester, it can act as a filler. Mousavioun et el. [185] research results showed that the thermal properties of PHB can be improved by blending lignin into the polymer matrix, and based on these results, up to 40 % of soda lignin is compatible with PHB and compared to the neat PHB, this blend is more thermal stable in a wide range of temperature. This research group in another study [186] revealed that up to 30 % of lignin blended with PHB can act as a plasticizer, and more than 60 % of lignin could result in phase separation. Moreover, Xing et al. [187] showed that tensile and thermal properties of PBAT can be improved with high compatibility with up to 20 % of lignin and high UV protection with 10 %. Several research studies have rejected that idea by claiming that a high concentration of lignin is not a good solution due to several problems regarding its compatibility with some biodegradable polyesters such as PHB. For instance, Kai et al. [188] after adding 1 % to 30 % lignin into PHB showed that Young's modulus, elongation at break and tensile strength increased compared to neat PHB; however, by increasing the content of lignin more than 2 % higher content of lignin result in reduction in these properties, although all contents showed better properties in comparison with neat PHB. Even though it is biocompatible for most polymers, while creating particular applications, factors like lignin availability, purity, and modification must be taken into account. It's important to carefully assess any unique immunological responses that certain lignins or lignin derivatives may cause [189]. The antimicrobial activity, as well as mechanical properties and UV barrier of lignin composite, was further improved by blending with ZnO, TiO_2, and silver oxide, as they are approved in different industries such as the food industry by being authorized by FDA [190,191].

There are several natural and synthesized antioxidant additives, such as phenol, polyphenol, and vitamins, which showed superior performance compared to petrochemical antioxidants. The mechanism of the antioxidant contains intermolecular dimerization and radical scavenging that trigger higher flexibility in molecular structure. Bio-based antioxidants offer a promising alternative to petrochemical antioxidants, as their performance can surpass that of Irganox 1010 (or other Irganox types, e.g., Irganox 1098) [177,178]. Reano et al. [43] concluded that biobased antioxidants with the same content had better performance than Irganox 1010 on PBS. Additionally, these bio-based antioxidants enhance the stability of molecular weight, with minimal impact on the structure.

As an important bio-based antioxidant, essential oils (EOs) are natural extracts that possess antibacterial properties and can reduce symptoms of chronic illnesses by preventing oxidative decay caused by reactive oxygen radicals [178,179]. EOs that are used almost as antimicrobial agents for polyesters can also be used as anti-oxidants [192]. Eugenol, carvacrol, citral, thymol, and cinnamaldehyde are essential oils with high potential as antioxidants [193,194]. The content of essential oils incorporated into biodegradable polyester is different, and it depends on the type of polyester, type of essential oil, and the purpose of blending. For instance, Kumari et al. [127] blended grapeseed oil, ginger

Table 8
An overview of the additives used by researchers as impact modifier.

Impact modifier	Common target polyester	Advantages	Further information	Ref
Ethylene-epoxy copolymers	PLA	Its processability and thermal stability, high compatibility with PLA, etc.	Increase the impact resistance and toughness of polyester resins. They can enhance the low-temperature impact characteristics of polyester resins when used in conjunction with other impact modifiers	[99,100]
Nitrile rubber (NBR)	PLA and PBAT	Good mechanical properties, such as tensile strength.	Reduce the crack in polyesters, which results in higher toughness. The processing conditions and the particle size are two main factors influence its performance.	[101,102]
BS	PLA, PBS and PBAT	Compatibility with the polymer matrix, affordability, and processing simplicity	Styrene offers the copolymer exceptional stiffness and dimensional stability, while butadiene, gives the copolymer great toughness and elasticity. It increases the impact resistance and toughness of the final material even if with their low content	[103,104]

Table 9
Cross-linking strategies for biodegradable polyester, including key mechanisms, initiators, polymer types, and simplified reaction schemes.

Mechanism	Agents/energy	Target polyester	Reaction scheme (compact form)	Advantages/disadvantages	Ref.
Cross-linking with multifunctional agents	TMPTA, multifunctional acrylates, epoxides	PLA, PHA and PBAT	Multifunctional group ($R–X_n$) + HO–Polymer / COOH–Polymer → Polymer–X–Polymer (cross-linked network)	Improves mechanical and chemical resistance; may reduce biodegradability	[37,105]
Radiation-induced cross-linking	UV, γ-rays	PLA, PHA and PBS	Polymer + hν → Polymer• → cross-linked polymer	No chemical additives needed; homogeneous cross-linking; high equipment cost	[,106]
Free radical cross-linking	Peroxides (e.g., DCP, BPO), vinyl monomers	PLA, PHB, PBS and PBAT	Initiator → 2 R• R• + Monomer → Monomer• Monomer• + Monomer → Cross-linked network	Versatile method; potential for degradation or unwanted branching	[107–109 [42,110]]

Table 10
An overview of the additives used by researchers as chain extenders or crosslinking agent.

Chain extender and crosslinking agents	Common target polyester	Advantages	Further information	Ref
Joncryls	PLA and PET	Significantly increase the PLA and PET molecular weight. Each Joncryl molecule has its own different epoxide functional groups that are stable under normal conditions but react irreversibly and quickly with carboxylates	Joncryl can increase the thermal stability, owing to an enhancement in energy of activation of thermal decomposition. It improves rheological behavior by increasing the complex viscosity	[111,112,113]
Pyromellitic dianhydride (PMDA)	PET, PLA, and PHB	Improve the rheological, thermal and mechanical characteristics of PLA and PHB. Its high compatibility with PLA and PHB	In a process known as imidization, PMDA creates a very stable imide link that aids in preventing the dissolution of the polymer chains.	[114,115]
Maleic Anhydride (MA)	PLA and PHB	It can improve the mechanical properties of PLA and PHB. Processability, biodegradability, low volatility, etc.	The hydroxyl groups of polyesters react with the dicarboxylic anhydride maleic anhydride to generate ester linkage. Introducing MA onto polyester chains could disturb PHB chains' regularity, then control the morphological structures and improve its properties	[116,117–122]
Silans such as methacryloxy silanes, vinyl silanes, amino silanes, etc.	PLA and PBAT	Resistant to the fast degradation, compatibility, adhesion, processability, etc.	It can create covalent connections between the chains of polymers. The hydroxyl groups on the polymer chains can react with silane to generate siloxane bonds, which form a crosslinked network that enhances the material's mechanical characteristics.	[123–126]

oil, and bergamot oil and studied their influence as antioxidant and antimicrobial agents. They reported that 5 % of these additives significantly improved the oxidation resistance of the polymer along with thermal and mechanical properties. They also showed plasticizing properties, improving the flexibility of PHB. Garrido-Miranda [195] studied the influence of eugenol in the PHB-TPS matrix as an antioxidant additive. His study revealed that only 3 % of eugenol showed 92 % radical scavenging. The reason is based on the ability of eugenol to reduce multiple (2,2-diphenyl-1-picrylhydrazyl) (DPPH) radicals by dimerization and reaction with free radicals. This reaction results in the formation of dehydrodieugenol, which enhances its radical scavenging activity.

Due to their low toxicity and high biodegradability, essential oils are ecologically favorable for use in polymers [196]. Another advantage of essential oils is that they have the ability to protect the polymeric material matrix from degradation during extrusion despite the possibility of their own degradation [197]. The downside of using essential oils is their high thermal degradability at elevated temperatures, which limits their applicability and can only be mitigated by using higher concentrations [198]. For PLA and PHB with melting points around 170–185 °C, a significant percentage of essential oils may be lost during extrusion (depending on the oil type) [185]; however, the remaining oils would still be effective, albeit with a concentration penalty. Another disadvantage of using Eos is their high volatility at high temperatures, reducing their efficacy; hence, nanoencapsulation can overcome this issue [199].

Catechin, a polyphenolic flavonoid, is a naturally occurring substance found in many plant species with anti-inflammatory properties. It can be found in various plant-based foods and drinks such as tea, chocolate, and fruits. Catechin has known as an interesting natural antioxidant for the polyesters industry to retard degradation during thermal processing and also to develop antioxidant materials [197].

Table 11
Literature on eco-friendly antimicrobial agents in biodegradable polyesters.

Research group	Polyester	Target industry	Antimicrobial agent	Purpose	Outcome
[127]	PHB	Food packaging applications	Grapeseed oil, ginger oil, and bergamot oil	Improve the flexibility and antimicrobial efficiency	Water vapor permeability reduced, higher antimicrobial and antioxidant actions. Suitable for fruitcakes, buns, sandwiches, and bread
[128]	PLA	Electrospun fibers for potential biomedical app[310]lications	Antimicrobial Peptides (AMP)	Create multifunctional biobased fibers with antimicrobial activity	Against most of the microorganisms, it didn't improve antibacterial action. However, blending both AMP and cationic polymer into PLA improved antibacterial action
[129]	Starch/ PBAT	Food packaging applications	AMP, ε-Poly-ʟ-lysine (ε-PL)	Extend the shelf life of peaches, and the antimicrobial actions of the film	More flexibility of film by adding ε-PL. High antimicrobial action against microorganisms that are prevalent in food
[130]	PLA	An active packaging for almond and beef	Green tea and rosemary polyphenolic	Extend foods shelf-life	High antioxidant and antimicrobial activities, inhibiting almond's and beef's (shorter than almond) lipid oxidation
[131]	PLA/ PBAT	Food industry	Cinnamaldehyde and tea polyphenols	Prevention of bacteria growth in meat	Inhibiting *E. coli* and *S. aureus* at specific temperatures, maintain the quality of meat by reducing water permeability
[132]	PLA-AgPalm	3D printing for biomedical applications	Chitosan	Antimicrobial, antiviral, and cytotoxic activity	Antimicrobial activity against the two most known bacteria, *E. coli* and *S. aureus*. Silver caused a high antiviral activity by inhibiting cytopathic effect of influenza virus and adenovirus.
[133]	PCL	Drug delivery	Zinc oxide nanoparticles (ZnO NPs) and Salvia abrotanoides essential oil (SAEO)	Antibacterial and anti-inflammatory functionalities	A low concentration of SAEO had almost zero antibacterial effect against *S. aureus*, but ZnO had a high performance against *S. aureus*. They minimized oxidative tissue damage caused by inflammatory cells

Catechin (even as low as 1 %) can provide a sufficient influence to retard polymer thermal degradation owing to its high antioxidant capacity. However, in Arrieta et al. [200] study, increasing the content of catechin from 1 % to 3 % increased the radical scavenging effect significantly due to the higher hydroxyl group availability that makes it capable of neutralizing free radicals. Its small particle size and high surface area make Catechin suitable for developing a greater nucleation effect. [201].

In a nutshell, among oxidative stabilizers, lignin derivatives, such as organosolv, soda, and kraft lignin, exhibit versatility in improving oxidative and thermal degradation temperatures, with potential antimicrobial benefits at low concentrations. The content of these additives depends on the purpose and the target industry of biodegradable polyester. For instance, high lignin content would not be the best option for industries requiring superior mechanical properties. Having up to 10 % lignin increases the mechanical properties and enhances the RSA to an acceptable level. It is worth mentioning that although the cost of lignin is low compared with most antioxidants, increasing the content of lignin would result in phase separation. Although EOs, like eugenol and citral, serve as dual-functionality antimicrobial and antioxidant agents, with challenges addressed through nanoencapsulation, the potential of lignin derivatives outweighs not only cost but also due to their high effectiveness. Regarding the amount of EOs, it should be noted that EOs have several functions and can influence different properties. For example, more than 5 % of EOs could act as a plasticizer and increase flexibility, which won't be cost-effective. However, based on the research studies, up to 5 % could improve the RSA and antimicrobial activity in most situations.

3.2. UV stabilizers

Biodegradable polyesters such as PLA show poor UV resistance due to their aliphatic structure, which is less effective for UV absorption [184]. Two commonly known mechanisms are responsible largely for the reason for PLA UV degradation. First, the PLA backbone C—O link is broken by a photolysis process when exposed to UV light. Second, PLA photodegrades more quickly and produces a hydroperoxide radical as a result of increased exposure to UV light and environmental factors. As exposure increases, this radical is subsequently degraded, resulting in the creation of molecules having carboxyl end groups.

UV stabilizers are additives that are included in polymers to shield them from the harmful effects of UV light. These stabilizers function by blocking UV radiation from penetrating and disintegrating the polymer molecules by absorbing, reflecting, or dispersing them.

They are often employed in polymeric materials such as plastics, coatings, and others exposed to sun UV rays or other causes. UV triggers photooxidative degradation, breaking down polymer chains, changing mechanical characteristics, and rendering materials practically worthless. There are several types of UV stabilizers, such as UV absorbers (absorbing UV and converting it to heating energy) [202,203], hindered Amine Light Stabilizers (HALS) (impede the production of free radicals, which slows the degradative process) [204,205], antioxidants, pigments (absorbing UV on the polymer surface and prevent them from reaching the polymer), metal deactivators (stop metal ions from catalyzing the degradation of polymers). When choosing a UV stabilizer for polyester, it is crucial to evaluate various important properties, including the UV absorption range, migration potential to the polymer surface, thermal stability, weathering resistance, and solubility of the stabilizer [206,207]. Quercetin, a flavonol with five hydroxyl groups, showed exceptional antioxidant and UV protection activities compared to phenolic compounds when blended with biodegradable polyesters [208]. Deng and Zhou [206] studied the influence of quercetin in the presence of food-grade methyl cinnamate (MC) blended with PLA on UV protection and antioxidant activities. Their results showed that up to 10 % of quercetin blended with 1.75 and 3 g/L MC resulted in high UV stabilization of PLA due to their cinnamoyl-like structure in quercetin [209], and based on *trans–cis* isomerization of MC mediated by a triplet state under UV radiation, increasing the amount of MC had a higher influence on PLA UV stabilization than quercetin [206,210]. Some nanoparticles, such as zinc sulfide nanoparticles (ZnS NPs), were used by researchers as UV stabilizers. For instance, Ali Raza et al. [211] studied the influence of this additive with three contents (1, 2, and 3 wt%) on UV protection and on colony forming unit (CFU) against *Staphylococcus aureus* and *Escherichia coli.* They showed that even 1 % of this additive results in high UV protection, and enhancing its content has less influence on increasing UV protection. The biopolymer composite's nano ZnS may induce oxidative stress, which would degrade bacterial membranes and result in antibacterial activity [212].

Another technique that has some similarities with pigments is dyeing

Table 12
MIC of most used essential oils applied in different industries.

Research group	Essential oil	Additive's type	Microbe name	Microbe's type	MIC(µg/mL)
[134]	Anethole	Anti-Fungal	*Candida albicans IFO 1061*	Fungi	625
			Candida rugosa IFO 1152		625
			Aspergillus niger ATCC 6275		313
[135]	Anethole	Anti-Fungal	*Saccharomyces cerevisiae*	Fungi	100
[136]	Thymol	Anti-Fungal	*Aspergillus niger*	Fungi	200
			PTCC 5154 (available in soil)		
[137]	Thymol	Anti-Bacterial	*Staphylococcus aureus*	Bacteria	128
			Enterobacter cloacae		256
			P. rettigeri		256
[138]	Thymol	Anti-Bacterial	*Salmonella Typhimurium*	Bacteria	64
			LT2 DT104		
			Salmonella Typhimurium ATCC 1408		64
[139]	Thymol	Anti-Bacterial	*Staphylococcus aureus ATCC 25923*	Bacteria	256
			LT2 DT104		
			Staphylococcus aureus B234		64
[140]	Thymol	Anti-Bacterial	*Escherichia coli*	Fungi	351
[141]	Thymol	Anti-Bacterial	*Agrobacterium tumefaciens*	Bacteria	1000
			Erwinia carotovora		2000
[139]	Eugenol	Anti-Bacterial	*Staphylococcus aureus ATCC 25923*	Bacteria	256
			LT2 DT104		
			Staphylococcus aureus B234		256
[140]	Eugenol	Anti-Bacterial	*Escherichia coli*	Fungi	5625
[138]	Eugenol	Anti-Bacterial	*Salmonella Typhimurium*	Bacteria	512
			LT2 DT104		
			Salmonella Typhimurium ATCC 1408		512
[138]	Eugenol	Anti-Bacterial	*Salmonella Typhimurium*	Bacteria	512
			LT2 DT104		
			Salmonella Typhimurium ATCC 1408		128
[142]	Eugenol	Anti-Fungal	*Aspergillus flavus*	Fungi	>6250
			Neoformanc KCCM 0564		
			Aspergillus ochraceus		6250
[143]	Isoeugenol	Anti-Bacterial	*Staphylococcus mutans*	Bacteria	500
[144]	Eugenol	Anti-Fungi	*Penicillium italicum*	Fungi	2.5
[140]	Eugenol	Anti-Bacterial	*Escherichia coli*	Fungi	5625
[140]	Carvacrol	Anti-Bacterial	*Escherichia coli*	Fungi	703
[139]	Carvacrol	Anti-Bacterial	*Staphylococcus aureus ATCC 25923*	Bacteria	64
			LT2 DT104		
			Staphylococcus aureus B234		128
[138]	Carvacrol	Anti-Bacterial	*Salmonella Typhimurium*	Bacteria	512
			LT2 DT104		
			Salmonella Typhimurium ATCC 1408		128
[145]	Carvacrol	Anti-Bacterial	*Escherichia coli O157:H7*	Bacteria	500
			Escherichia coli O157:H7		1000
			Escherichia coli O157:H7		1000
[136]	Carvacrol	Anti-Fungal	*Aspergillus niger*	Fungi	50
			PTCC 5154 (available in soil)		
			Aspergillus versicolor		3
[144]	Carvacrol	Anti-Fungi	*Penicillium italicum*	Fungi	0.625
[140]	Carvacrol	Anti-Bacterial	*Escherichia coli*	Fungi	703
[141]	1,8 Cineole	Anti-Bacterial	*Agrobacterium tumefaciens*	Bacteria	4000
			Erwinia carotovora		>5000
[144]	Trans-Cinnamaldehyde	Anti-Fungi	*Penicillium italicum*	Fungi	0.313
[144]	Carvone	Anti-Fungi	*Penicillium italicum*	Fungi	0.75
[146]	Carvone	Anti-Bacterial	*Staphylococcus aureus.*	Fungi	2000
[147]	Carmphor	Anti-Fungal	*Cryptocouccus*	Fungi	780
			Neoformanc KCCM 0564		
			A. flavus KCCM 11453		780
[148]	Citral	Anti-Fungal	*Geotrichum citri-aurantii*	Fungi	50
[146]	Citral	Anti-Bacterial	*Staphylococcus aureus.*	Fungi	500
[149]	D-Limonene	Anti-Bacterial	*Staphylococcus aureus*	Bacteria	1
			Bacillus subtilis		1
			Escherichia coli		1

the base polymer. Both methods add color to the base polyester; however, dyes and pigments differ in their solubility and application. On the other hand, dyes are organic compounds almost soluble in the polymer matrix, while pigments are insoluble in the polymer and stay on the surface. Dyeing is widely used in the textile industry, but its application is subject to strict regulations due to its high energy consumption [213,214].

In the UV stabilizer realm, types such as UV absorbers, HALS, antioxidants, pigments, and metal deactivators play unique roles. When selecting a UV stabilizer, factors such as UV absorption range, migration tendency, thermal stability, weathering resistance, and solubility must be considered. Dyeing, a technique akin to pigmentation, introduces color to the polyester, with regulations and energy consumption challenges.

Despite their benefits, conventional UV/oxidative stabilizers have potential drawbacks. Many (HALS, benzotriazoles, etc.) are petroleum-derived and hard to biodegrade. Therefore, their use in "fully biodegradable" systems raises concerns. They may also move to the surface over time, reducing long-term efficacy and potentially contaminating food in packaging applications. Some stabilizers (such as aromatic

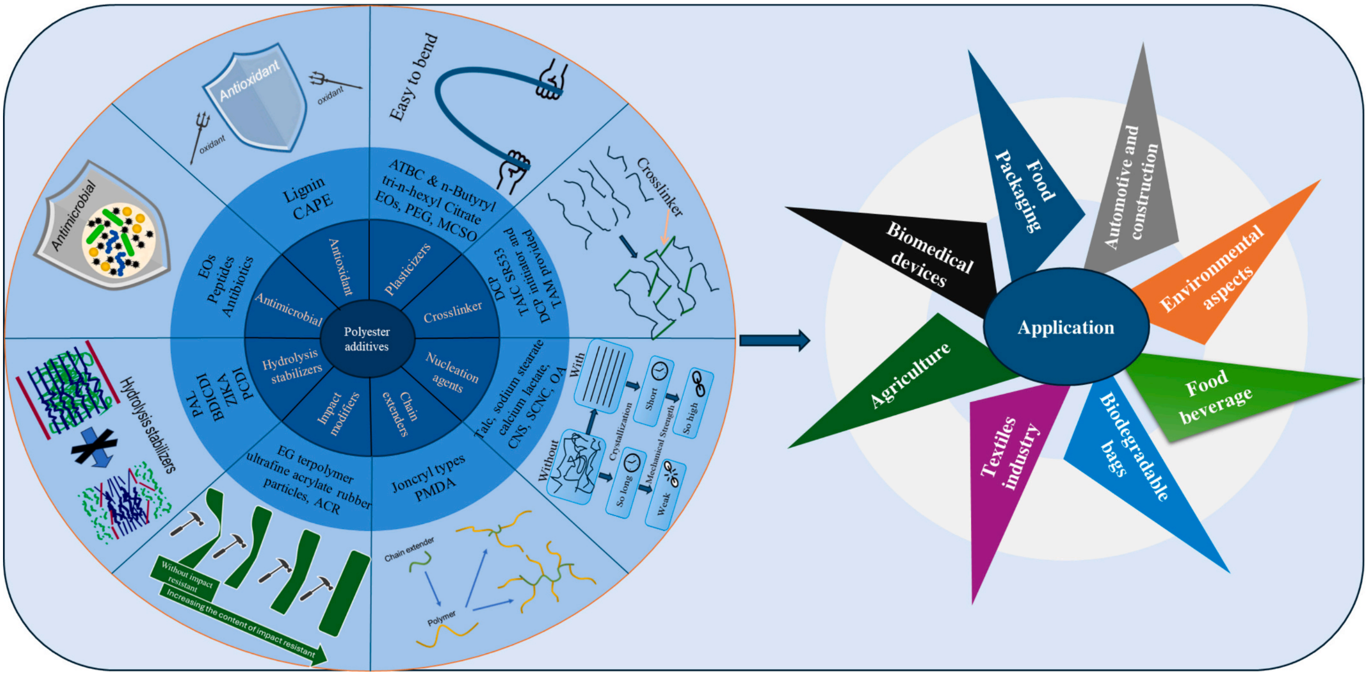


Fig. 2. Overview of various additives used in biodegradable polyesters.

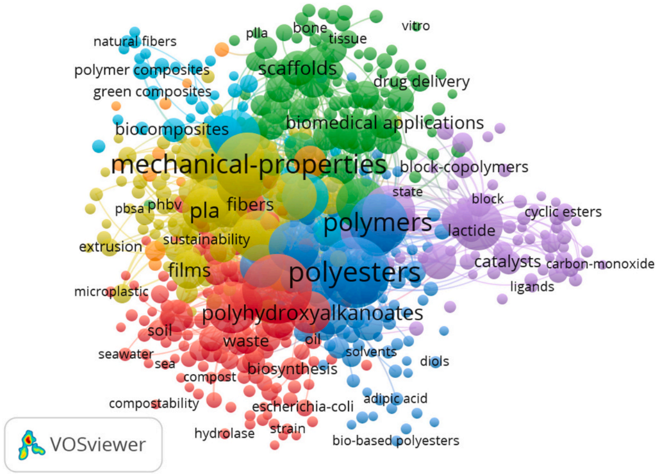


Fig. 3. Co-occurrence of the main authors' keywords, with the minimum number of occurrences of 5 for the recent publications in biodegradable polyesters' properties and application.

antioxidants) can even act as pro-oxidants if used improperly. They may also slightly reduce the polymer's initial mechanical strength or transparency. Moreover, UV absorbers for some biodegradable polyesters such as PBAT are required to be modified to reduce their migration from polymer matrix and get rid of their poor thermal stability. In food packaging, where PBAT is commonly used, this migration is even worse, as it can contaminate the packaged products [215,216]. Synthesizing UV absorbers with high molecular weight and grafting UV absorbers into the structure of polymer chains were two techniques used by researchers. Nowadays, some research studies immobilized UV absorbers on some inorganic nanoparticles or graphene-based materials [216,217]. There is also a trend towards green stabilizers that align with biodegradability. Research has demonstrated, for example, that biosourced polyphenols and flavonoids (like naringin, quercetin) can enhance PLA's UV and oxidative stability [218,219]. Likewise, lignin and its derivatives are being widely studied as low-cost, natural UV stabilizers to replace synthetic additives [220,221]. Another promising avenue is nano-additives such as ZnO or TiO_2 nanoparticles, which provide UV shielding and antioxidant effects. These inorganic particles are themselves non-degradable but can be used in very small amounts and are generally considered safe (e.g. ZnO will eventually mineralize in the environment). Overall, future development is focused on formulating stabilizer packages that maintain polymer performance during use but do not impede, and may even assist, the polymer's eventual environmental breakdown.

4. Nucleation agents

Some polyesters exhibit low crystallinity, leading to reduced thermal stability, poor mechanical properties, and suboptimal barrier and migration performance [222–224]. These drawbacks cause their applications to be reduced in various industries, such as packaging, particularly for food containers and bottles. To overcome this problem, researchers explored the use of nucleation agents to lower the surface-free energy barrier, increase the crystallization rate, and enhance polymer resistance [225–227]. The nucleation agents act as locations for crystal growth, boosting the number of nucleation sites and encouraging a more uniform crystal structure. This may result in enhanced mechanical qualities, including greater stiffness and strength, as well as enhanced thermal stability. Additionally, nucleation agents may be employed to regulate the crystal size and form, which improves the transparency and optical qualities of specific polymers. The melting and crystallization temperatures of suitable nucleators are often greater than those of the polymer matrix. Consequently, the molten polymer molecular chain can initiate and develop on the solidified nucleator crystal's surface acting as a heterogeneous nucleation site [228]. Shi et al. [224] incorporated octadecyl amine (ODA) grafted cellulose nanocrystals (g-CNC) in the polymer matrix with four different contents: 0.1, 0.2, 0.5, and 1 %. Their results showed that by enhancing the content of this additive, mechanical properties increased significantly, while based on possible aggregation, by using the higher content of 1 %, a slight reduction in tensile strength and strain was observed. Crystallization transmission of PLA improved significantly by adding g-CNC, and increasing the content of this additive increased this property

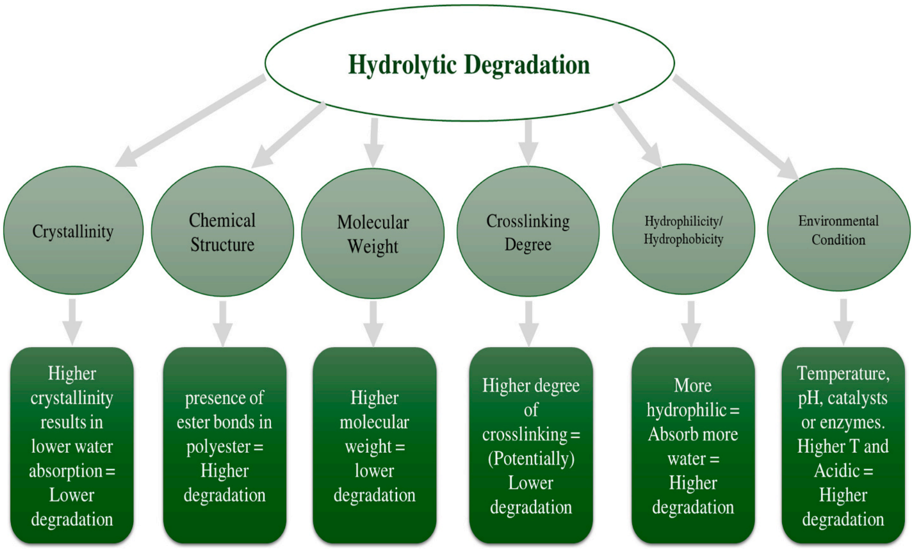


Fig. 4. The properties of polymers that influence hydrolytic degradation.

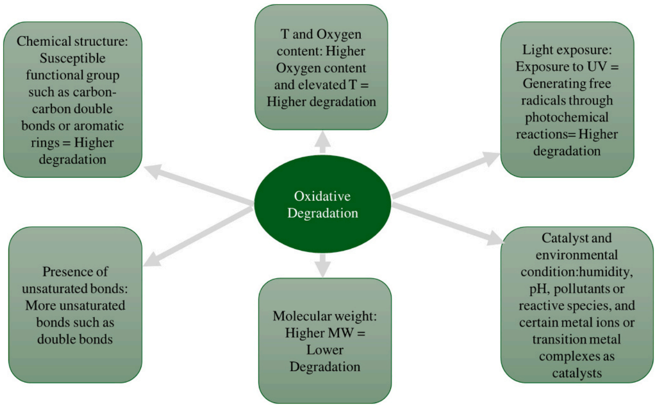


Fig. 5. The influence of different factors on the oxidative degradation

significantly. This additive was further used as it could enhance the transparency of PLA, making it more favorable in packaging by providing more crystal nuclei and reducing crystal size and light scattering [229]. Organic Montmorillonite (OMMT)-CNC system was used with various contents of 1–4 % in PLA to enhance the crystallization rate and mechanical properties [62]. In terms of mechanical properties, their results were the same as the previous one by concluding that higher content of this additive causes agglomeration, resulting in mechanical properties deterioration, while lower content of 1 and 2 % increase it. They also showed that CNC-OMMT system fillers dispersed in PLA more uniformly than CNC and OMMT, and higher content (4 %) increased the crystallization rate significantly. Several studies have used layered Double Hydroxides (LDHs) as nucleation agents in PLA. Geng et al. [73] studied 0.1 to 0.5 % amide ethylenediamine tetraacetic acid intercalation layered double hydroxides (AE-LDHs) and showed the increase of crystallinity by 30 % by adding 0.5 % as AE-LDHs provides abundant nucleation sites that causes faster and uniform crystallization. Moreover, as this additive facilitates high crystallization, it restricts cold crystallization.

Generally, heterogeneous nucleation agents involve the formation of nuclei on the surface of foreign particles or interfaces within the polymer melt. These foreign surfaces, provided by nucleation agents, serve as templates for the organization of polymer chains into ordered crystalline structures [230]. However, homogeneous nucleation occurs when crystallization initiates spontaneously within the bulk polymer melt, without the presence of any foreign particles or surfaces. In this process, small clusters of polymer chains organize themselves into ordered crystalline structures, forming nuclei from the homogeneous polymer matrix. Due to the process and the experiment situations, homogeneous nucleation is rare as it needs more precise environmental conditions and equipment [231].

Ostwal's step rule and classical nucleation theory (CNT) are the most famous theoretical models for nucleation process. Ostwal's step rule proposes that nucleation and growth are separate, sequential steps in the crystallization process. Nucleation agents can affect nucleation kinetics by accelerating or inhibiting the formation of nuclei without significantly altering subsequent crystal growth. Meanwhile, CNT—widely used in this field—describes nucleation as a process where critical nuclei form based on a specific size and energy barrier. Nucleating agents can modify CNT predictions by altering the critical nucleus size or reducing the energy barrier required for nucleation [232].

There are several types of nucleation agents available, including organic, inorganic, and high molecular nucleating substances. Examples of these substances include boron nitride, talc, orotic acid, uracil, compounds derived from oxalamides, as well as nanocellulose, among others [69,233].

Among organic nucleating agents for polyesters, different classes exist: amides and nitrogen-containing compounds such as *N*,*N*-dimethylformamide [224] and thymine [225]; acids like benzoic acid [226,227]; surfactants such as sodium stearate [215]; and esters. As an organic nucleator, thymine (a pyrimidine base in DNA) showed a sufficient influence as it has much higher T_m and T_c than the majority of polyesters.

Table 6 highlighted the most used nucleation agent used in biodegradable polyesters. Among the organic nucleators, thymine stands out for its remarkable impact, exhibiting higher melting and crystallization temperatures than most polyesters. In the category of inorganic nucleators, organomodified montmorillonite (OMMT) platelets offer a large specific surface area, excellent compatibility during melt blending, and strong thermal stability, facilitating heterogeneous nucleation. Cellulose nanocrystalline emerges as an attractive option due to its excellent crystallinity, small size, low cost, and notable mechanical performance. The selection of an optimal nucleating agent depends on specific application requirements, balancing factors such as mechanical enhancement, cost-effectiveness, and environmental impact.

A high loading of inorganic nucleant may act as a filler, potentially causing opacity and embrittlement if the particles are large or poorly compatibilized. Although crystallinity often improves strength and modulus in one hand, it can reduce extensibility on the other hand. For example, PLA with excessive talc showed increased crystallinity. However, a drop in elongation at break from 3.8 % to 2.0 %, accompanied by brittle fracture behavior was observed due to poor polymer-filler interfacial adhesion [234]. Many researchers are exploring biobased and nanoscale nucleating agents that not only speed crystallization but also reinforce the polymer. Nanoscale cellulose and chitin crystals, for example, can act as efficient nucleating sites while being renewable and biodegradable themselves [235,236]. Another promising strategy is using small amounts of stereocomplex PLA or PHA fibers to nucleate crystallization in PLA or PHB matrices – these not only accelerate crystallite formation but form a wholly biodegradable composite. Continued development of highly effective nucleants (such as zinc phenylphosphonate or novel amide derivatives [234]) will enable faster processing and improved properties, but the emphasis will be on those that maintain clarity (for films) or toughness, and do not leave harmful residues in compost.

5. Plasticizers

The polymers' properties depend on the molecular weight and molecular weight distribution. High molecular weight polymers have high strength, but poor processability and high melt viscosity [237,238]. Conversely, while low molecular weight polymers offer superior toughness and more favorable rheological properties, they tend to have lower rigidity [239]. Plasticizers, being non-volatile low molecular weight substances, possess significant potential as an additive for polymer blending. They can effectively address the aforementioned drawbacks of polyesters, while also enhancing their ductility and toughness [240,241]. By incorporating plasticizers, certain biodegradable polyesters can be further optimized for applications in flexible packaging, medical devices such as tubing, and biodegradable bags. A schematic representation of the plasticizer's influence on the polymer matrix is shown in Fig. 6.

Plasticizers also increase the free volume in polymer blends, leading to an increased distance between polymer chains and a reduction in their interactions [242,243].

There are many different kinds of plasticizers. However, traditional petroleum phthalate plasticizers are used widely over the world and are in great demand due to their price and performance [244,245]. Phosphates (for polymers that require flame-retardant properties) [246,247], adipates that are almost usable for PBAT and PET that require flexibility at low temperatures [248], and epoxidized vegetable oils such as linseed and soybean oils [249,250] are among the most used plasticizers for polyesters.

Epoxidized natural oils as the eco-friendliest plasticizers for biodegradable polyesters, have gained an increasing attention not only due to their performance in reducing T_g, but also for some industries such as food packaging they are important due to their antimicrobial properties. Besides, some epoxidized natural oils such as Epoxidized Cottonseed Oil (ECSO) can causes an enhancement in the polymer stability because oxirane rings can scavenge acid groups by catalytic degradation and this can affect the light and thermal stabilization positively. For this reason, in the industry, epoxidized vegetable oils are used as a thermal and light stabilizers [251]. In this case, finding a suitable oil for needs to be studied and consider several factors such as the type of polymer, oils' properties and the final product.

Table 7 represents the most effective additives as plasticizers used for biodegradable polyesters. The selection of an ideal plasticizer involves considering factors such as plasticizing efficiency, resistance to migration, compatibility with the base polymer, low volatility or non-volatility, and sustained plasticizing properties across temperature ranges. Among the various plasticizers explored for biodegradable polyesters, epoxidized natural oils stand out as a highly effective and environmentally friendly choice. Their flexible properties, processability, and compatibility with polyesters contribute to improved mechanical and thermal performance. Notably, the epoxide groups on these plasticizer molecules create hydrogen bonds with carbonyl groups on polymer chains, facilitating increased molecular mobility. This unique characteristic allows polymer chains to move more freely, reducing intermolecular forces and, consequently, lowering melt viscosity. Additionally, epoxidized natural oils, such as Chia seed oil, soybean oil, and Karanja oil, exhibit the added benefit of serving as compatibilizers in blending different polyesters. Through reactions between hydroxyl and epoxy groups, these additives contribute to enhanced compatibility, making them a versatile and promising choice for optimizing the properties of biodegradable polyesters in diverse applications.

Utilizing plasticizers as additives may cause potential decay of mechanical strength and the risk of additive migration. While 5–15 % plasticizer yields a more flexible material, it also lowers tensile strength and stiffness [252]. Phase separation or "oiling out" can also occur when plasticizers are used above a certain concentration. An excess amount of low-molecular-weight additives like glycerol in PLA leads to a sticky, heterogeneous material that cannot be properly processed. Over time, many plasticizers tend to leach to the surface (volatilizing or extracting out), which not only diminishes the plasticizing effect but could also pose safety issues in food contact uses. To address these drawbacks, future research is focused on non-migrating and bio-based plasticizers. One approach is using polymeric or reactive plasticizers that bond into the polyester matrix (for instance, epoxidized soybean oil can both

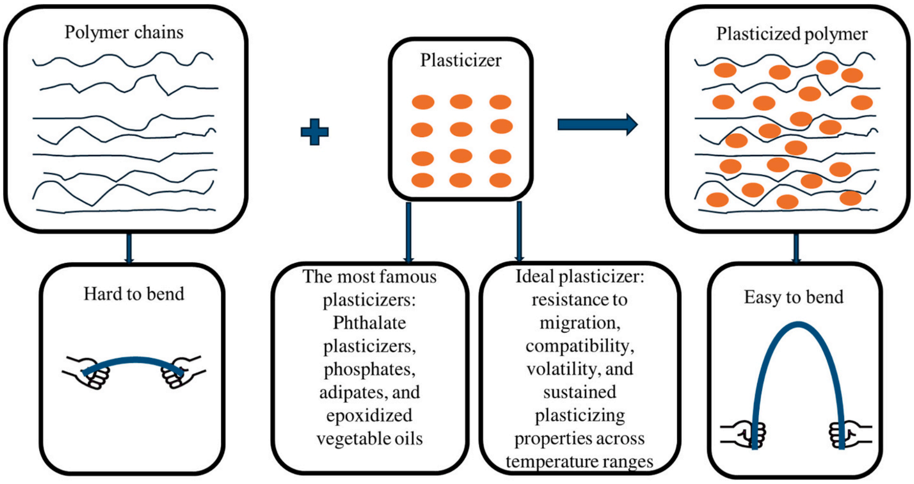


Fig. 6. The schematic of plasticizer influence on the polymer matrix.

plasticize and lightly crosslink PLA, reducing migration) [253]. Another approach uses naturally derived plasticizers such as fatty-acid esters, glycerides, or deep-eutectic mixtures which are inherently compatible and biodegradable. Ongoing developments aim to achieve long-term flexibility (retaining ductility over the product's life) while ensuring the additives themselves do not introduce toxicity and will break down harmlessly in compost environments. Blends of biodegradable polyesters (e.g. PLA/PBAT blends) are also viewed as a way to "internalize" plasticization without conventional additives, effectively creating an all-polymer system with enhanced flexibility.

6. Impact modifiers

Impact resistance is a factor to measure the material strength ability to endure the sudden stress, that is one of the most important mechanical properties that attracting the researchers' attention due to its relation to service life, liability, and the safety of the plastic product [254,255]. To make the biodegradable polyesters suitable for products that need to withstand mechanical stress, such as automotive components, electronic housings, and sporting goods, impact resistant additives should be used.

Several research works have been done using impact modifiers to improve the toughness of the polyesters especially PLA [256–258]. From the previous research, it's known that impact toughness of polymers can be enhanced by blending impact modifiers. Common impact modifiers are rubbery compounds that cause sudden loads efficiently because of their flexible chain backbone. [259]. However, there are several types of impact modifier that can be used in polyesters, such as glass fibers [260,261], and butadiene-styrene (BS) copolymers (such as acrylonitrile-Butadiene-Styrene (ABS) and poly(methyl methacrylate)-poly(butadiene-styrene) (PMBS)) [35,262,263], grafted copolymers [264,265], etc. Grafted copolymers can be relatively eco-friendlier due to the polymer selection.

Impact modifiers are typically added to polymers in amounts up to 10–15 %; however, increasing the impact modifier more than that may lead to a reduction in the impact strength because of the presence of large agglomeration and the decreasing of modulus in the most rubber-based materials [266].

Another technique researchers used for impact modification of polyesters is blending other polymers or polyesters with the base polyester. Several factors and properties should be considered, including the thermal stability, cost, processability, impact resistance, and density of the impact modifier polymer [267]. Moreover, the selected polymer should exhibit higher toughness, greater impact resistance, and at least comparable or superior dimensional stability to the base polymer [268,269]. For example, when a PBAT/PBS mixture is used to modify the impact of PLA, a significant improvement in the impact strength can be achieved [255,270]. Moreover, it is worth mentioning that PBAT almost causes deformation in the matrix to absorb the impact energy and make a super tough blend [271,272]. Another polyester to be mentioned used as an impact modifier is PCL, which has the potential to enhance the toughness of several polyesters [273–276]. PCL has a low melting point as well as higher flexibility when in comparison with the majority of polyesters such as PET and PLA. Besides, PCL is a biodegradable polyester, making the polymer biodegradable, and in case it is used as a biodegradable polymer, it makes a biodegradable plastic with higher flexibility due to its high flexibility (like PBAT) in comparison with the regular biodegradable polymers [277,278].

Table 8 exhibits the additives used by researchers as impact modifier. Among the impact modifiers explored for biodegradable polyester, ethylene-epoxy copolymers stand out as a promising choice, especially in the context of PLA. Their remarkable processability, thermal stability, and high compatibility with PLA make them a preferred additive. Ethylene-epoxy copolymers not only enhance the impact resistance and toughness of polyester resins but also exhibit a notable capability to improve low-temperature impact characteristics when used in conjunction with other impact modifiers. This makes them a versatile and effective option for addressing the brittleness of polyesters, particularly PLA, offering a well-balanced enhancement of mechanical properties essential for prolonged service life and safety in plastic products.

Utilizing impact modifiers may lead to lower tensile strength, modulus, and sometimes thermal resistance [279]. Adding a soft phase usually lowers the composite's stiffness and yield strength because the load-bearing cross-section is effectively reduced by the presence of the rubbery phase. For example, while PLA/PBAT blends are much tougher, the tensile modulus and strength can drop in proportion to PBAT content [279]. This means designers must sometimes compensate by increasing thickness or accept a lower rigidity. Another issue is phase separation and compatibility. If the impact modifier is not compatible, the blend will have poor interfacial adhesion, leading to voids and actually lower strength. Impact modifiers can also affect processing behavior. A high elastomer content can make the melt more elastic which might cause die swell or difficulties in extrusion. Some modifiers (like certain polyurethanes or EVA) could also degrade at processing temperatures or cause bubble formation if they contain moisture. Changing the aesthetic aspect is recognized as another issue with impact modifiers, especially for packaging applications. Adding a second phase can turn a transparent polymer opaque. For packaging that requires clarity, impact modifiers that cause haze are not suitable.

The future research on impact modifiers lies in developing novel bio-based elastomeric additives and optimizing multi-component systems. An example is the creation of fully biobased impact modifiers – for example, plant-oil-derived thermoplastic elastomers or polyesters from biosynthesized monomers (like polyitaconates or bio-polyisoprene) that can blend with PLA. There is active research into block copolymers that contain segments miscible with PLA and segments that are rubbery; these can act as self-compatibilizing impact modifiers that form nano-scale domains. Such block or graft copolymers (PLA-co-PBSA, or PEG-PCL-PLA triblocks, etc.) can provide massive toughness improvements while minimizing stiffness loss [280]. Another promising strategy is dual modification, i.e., combining a plasticizer or chain extender with an impact modifier to get the benefits of both improved ductility and maintained strength. For instance, adding a small amount of chain extender to PLA/PBAT blends can create in situ compatibilizers (PLA-PBAT copolymers) that allow effective toughening at lower PBAT content – achieving high toughness with only minimal loss in modulus [281]. With the surge in additive manufacturing, there's interest in toughening 3D-printed PLA using modifiers that can be mixed into filaments or resins; for example, incorporating short elastomeric fibers in the filament or core-shell particles in UV-curable PLA resins for 3D printing. Furthermore, machine learning and formulation design are being applied to predict optimal combinations of multiple modifiers (impact modifier + nucleating agent + plasticizer) to simultaneously improve toughness, processing, and other properties. As research progresses, we can expect new impact modifiers that are renewable, compatibilized at the molecular level, and efficient at low concentrations.

7. Cross-linkers and chain extenders

Chain extenders and crosslinking agents are comparable because they both increase the number of chemical links between polymer chains, which increases a polymer's strength and stiffness. These additives make biodegradable polyesters particularly appealing for industries such as automotive parts manufacturing, construction materials, and durable goods.

7.1. Crosslinking agents

In polymers, cross-linking usually refers to the application of cross-links to make a change in the polymers' physical properties [282,283]. Some studies have shown that blending cross-linking agents with polyesters can produce promising elastomeric materials [284].

Generally, the main properties of crosslinking agents that should be taken into account are their cost, compatibility, thermal resistance, reactivity, toxicity, etc. [285,286].

As an initiator is usually employed to initiate crosslinking, finding a good initiator is of actual importance. The initiator acts as a catalyst and starts and accelerates the mechanism, and without an initiator, the crosslinking process might no longer be placed or will be so slow [287]. Azo compounds (decomposed by light or heat to produce free radicals) [288,289], peroxides (decompose at high temperatures) [110,290], and redox systems (free radicals provided by electron transformation) [291,292] are three types of the most used initiators. Sometimes, it is valuable to study the influence of crosslinkers and initiators on the crystallinity and glass transition temperature to see whether or not they influence the solidity of the polymer matrix, as they can be used as hardeners [116].

There are different types of crosslinking agents for biodegradable polyesters, such as silane [293,294], isocyanates, epoxy resins, acrylics, melamine formaldehyde resins, peroxides, etc. Epoxy resins are usable as they contain an epoxy group, which can react with the hydroxyl group of polyesters to make a crosslinked material with boosted tensile strength, toughness, and temperature resistance [295,296]. Moreover, this type of crosslinker can improve chemical resistance and the electrical properties of polymers [297,298].

Sometimes, even adding a low amount of an initiator such as dicumyl peroxide or radiation could cause unreferred chain branching or chain scission predominantly. The chain branching in the polymer blends triggers difficulty in polymer chain crystallization [299,300]. Consequently, crystals of smaller sizes are formed and lead to blends with decreased melting temperatures. In this case, the temperature of extrusion is very important when choosing a suitable crosslinker and initiator.

Table 9 summarizes the main cross-linking mechanisms used in biodegradable polyesters with representative reaction schemes illustrating the chemical principles behind cross-linking strategies. The table also summarizes the advantages and disadvantages of different mechanisms.

7.2. *Chain extenders*

Chain extenders are an additive class applied to modulate polyester's mechanical properties. They are characterized as being reactive, with functional groups forming the ends of polyester chains such as carboxylic acids and alcohols [301]. Moreover, they have at least two reactive sites. Chain extenders are known as one of two general categories: activating-type chain extenders and addition-type chain extenders [302]. Activating-type chain extenders are composed of an electrophilic site connected to a suitable substituent. They act as intermediates for a substitution reaction linking the ends of two polymer chains but don't become part of the extended chain. On the other hand, chain extenders, which are known as addition-type, contain at least two electrophilic sites and are intended to form a permanent bridge between at least two polymer chains. There are different types of chain extenders for polyesters, and the use of each is dependent on the application, the base polymer, and the conditions in which the base polymer is processed.

The mechanisms by which chain extenders interact with polyester chains typically involve nucleophilic addition or ring-opening reactions. For example, epoxy-functionalized oligomers (such as Joncryl®) can react with hydroxyl or carboxyl terminal groups through ring-opening of the epoxy groups, forming ether or ester linkages, improving flexibility and reducing brittleness [111–113], as reported in Fig. 2:

- Epoxy–R + HO–Polyester → R–O–CH_2–CH(OH)–Polyester
- Epoxy–R + COOH–Polyester → R–O–CO–Polyester

Similarly, carbodiimide-type extenders form urea linkages with carboxyl end groups, stabilizing PLA against hydrolysis [49](Table 4):

- R–N=C=N–R' + COOH–Polyester → R–NH–CO–O–Polyester

Anhydride-based compounds such as PMDA react with hydroxyl termini to form half-ester structures, and at higher ratios can induce branching or even cross-linking, enhancing resilience [114,115]:

- Anhydride + HO–Polyester → Half-ester linkage + COOH

Several factors should be taken into account when finding the best possible chain extender for polyesters. The first and foremost is the compatibility with the base polyester, molecular weight, processing condition, thermal stability, availability, etc.

Diols (substances with two hydroxyl groups that can interact with the carboxyl groups of polyester chains to extend the chain), glycolurils (which contain hydroxyl and urea groups that can react with the carboxyl groups of polyester), carboxylic acids (which have carboxyl groups that can react with the hydroxyl groups of polyester), and triamines (which are triamine compounds that can elongate polyester chains by reacting with the acid anhydride groups at their ends), are just a few examples.

For the coupling reaction, when anhydride functionalities of one PMDA molecule react with terminal hydroxyl end groups of two PLA, two carbonyl groups form per blended PMDA moiety (Fig. 7) [303]. When more than two PLA chains and one PMDA molecule react with each other, branching takes place. When more than one PMDA reacts with more than two chains of PLA, complex branching occurs, which causes crosslinking generation. [304]. Besides, the interaction of the −OH and –C=O of PLA groups with reactive PMDA functional groups has been found to improve elastic modulus, hardness, elongation, and

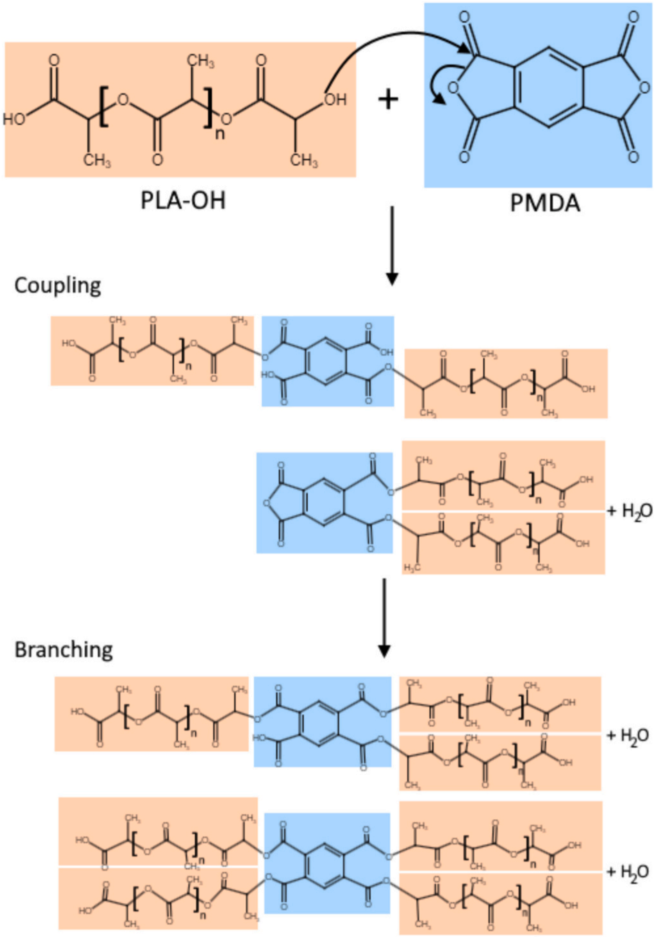


Fig. 7. Reaction mechanisms for PLA and PMDA chain extender [305].

creep behavior.

Table 10 highlights the most used additives for biodegradable polyesters as chain extenders or cross-linkers. When selecting the optimal cross-linker or chain extender for polyesters, essential factors to consider include compatibility with the base polyester, molecular weight, processing conditions, thermal stability, and availability. The choice of the additive should align with the specific application, ensuring integration with the base polymer and achieving the desired enhancements in mechanical and thermal properties. In terms of selecting the best additive, every mentioned additive has its own benefit and should be studied along with the purpose of using it. For instance, Joncryls demonstrate remarkable efficacy in significantly increasing the molecular weight of PLA and PET, offering improved thermal stability and rheological behavior. PMDA proves advantageous for PET, PLA, and PHB by enhancing their rheological, thermal, and mechanical characteristics through stable imide link formation. Maleic anhydride MA exhibits promise in enhancing the mechanical properties of PLA and PHB while contributing to their processability and biodegradability. Silans, such as methacryloxy silanes, vinyl silanes, and amino silanes, present resistance to fast degradation, compatibility, adhesion, and processability, creating covalent connections between polymer chains and forming a crosslinked network. In terms of environmentally friendly aspects, several additives can be chosen, such as tartaric acid and citric acid (non-toxic and mechanical properties improver of PLA and PHB), epoxidized vegetable oils and lignin derivatives (as functionalized biopolymers as chain extenders), and Glycerol Diglycidyl Ether (GDE) and MA (if they are synthesized in green synthetic way) are among the most interesting additives in this category [306,307].

Despite their advantages, extensive crosslinking can impede biodegradability by making polyester less accessible to hydrolysis or microbial attack. This is because a tightly crosslinked network cannot be enzymatically broken down easily. Excessive use of chain extenders may also cause gel formation or excessive branching, which has a deteriorating effect on mechanical properties. Moreover, many conventional chain extenders (e.g. epoxy oligomers like Joncryl or aromatic diisocyanates) are fossil-based and not biodegradable [308,309]. The development of biobased chain extenders and dynamically crosslinkable systems can be a solution to this issue. Researchers have reported success using epoxidized vegetable oils (such as epoxidized soybean oil) together with organic acids (e.g. citric, malic acid) as a renewable chain-extension system for PLA and PBS [253]. Another potential concept is using reversible crosslinking (via Diels–Alder bonds or other thermally reversible links) to allow reprocessing and eventual degradation. By tailoring the type and amount of chain extender, it is now possible to fine-tune the rheology of biodegradable polyesters for advanced applications (like foamable PLA and high-impact blends) while ensuring the materials remain compostable at end-of-life. However, the ongoing challenge is to maximize thermal stability and mechanical strength without introducing additives that linger in the environment. That's the main motivation to develop fully biodegradable reactive modifiers.

8. Antimicrobial agents

An antimicrobial agent is an agent that prevents the activity of microorganisms. Several additives have been added to the polymeric matrix, especially in PHB and PLA matrix, in recent years, including natural additives (such as essential oils), enzymes, chelating agents, antibiotics, peptides, and metals to provide antimicrobial activity [310–312]. The algorithm of selecting a suitable antimicrobial agent to its influence on the polymer matrix is summarized in Fig. 8. When biodegradable polyesters want to be used in applications where hygiene is crucial, such as medical devices, food packaging, and agricultural products. Table 11 exhibits several eco-friendly antimicrobial additives used by researchers in biodegradable polyesters in different industries. As can be seen, EOs are among the top-used ones; however, in some cases, bio-based metal oxides result in higher performance against some microorganisms. The factor that should be considered here is the concentration of the additives, which should be based on standards and regulations defined by each industry.

8.1. Essential oils

Essential oils (EOs) are oily aromatic substances extracted from plants using various techniques such as fermentation, extraction, and expression [293,294]. They are known as complex compounds containing a mixture of volatile and non-volatile components, many of which have been found to be effective in combating multi-drug-resistant (MDR) bacteria. EOs can also be used in combination with antibiotics to enhance efficacy synergistically. However, their applications are limited by factors such as water insolubility, susceptibility to oxidation, and volatility [295]. The effectiveness of each EO can be assessed by parameters such as the minimum inhibitory concentration (MIC), which indicates the lowest concentration of a chemical substance that inhibits visible microbial growth [296]. Table 12 summarizes the MIC of the most used essential oils for the inhibition of some specific microorganisms. Essential oils are used in polyesters for various purposes, including as plasticizers, antioxidants, antimicrobial agents, and barrier agents [297,298]. EOs are gaining increasing attention in the food industry, particularly in polymers for food packaging. In fact, because the active components in essential oils migrate, their addition to packaging films and coatings has resulted in products with improved optical and barrier properties, as well as antioxidant and antibacterial activity [313]. Alkyl polymers such as PLA and PHA and starch-based polymers are examples of commercially used biopolymers in the production of sustainable food packaging films. However, the challenge associated with the use of these

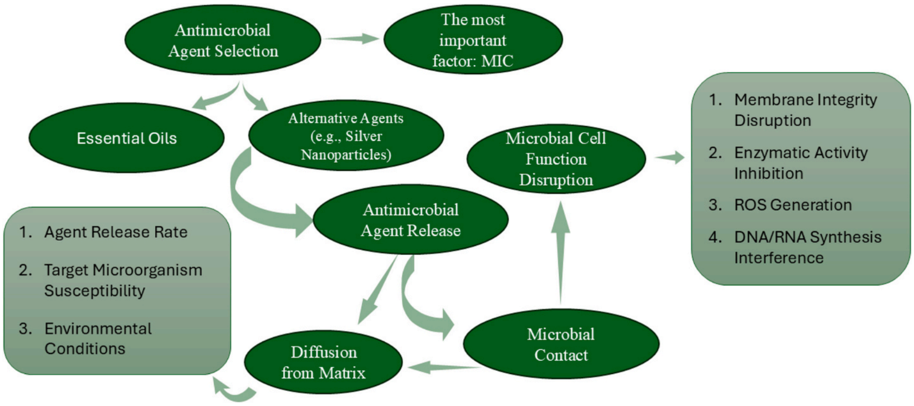


Fig. 8. The algorithm for selecting the mechanism of antimicrobial agent.

polymeric materials is to meet the requirements of improved mechanical and barrier performance, as well as market competitiveness. The addition of active ingredients such as EOs allows not only the improvement of the physicochemical properties of biopolymer-based films but also the increase in food safety and quality [313,314]. The use of additives in polymers in contact with food is restricted due to safety and the environment. EOs are natural and also approved by the Food and Drug Administrator (FDA) as Generally Recognized As Safe (GRAS) GRAS additives [315] without any harmful influence on food [316]. For this reason, active food packaging has become increasingly popular in recent years as a way of extending the shelf life of food products while improving consumer protection. [317].

Plant essential oils' active ingredients inhibit microorganisms by inhibiting protein synthesis, destroying the cytoplasmic membrane, and disrupting the proton motive force, electron flow, and active transport. [318] Carvone [318], oregano [319], carvacrol [320,321], and thymol [322,323] are the most interesting EOs for the food packaging industry due to their abundance and performance in antimicrobial and antioxidant activities. They include phenolic compounds that have antimicrobial effects against bacteria and fungi that are present in food.

To some extent, some types of EOs can influence the biodegradation of polymers by their influence on crystallinity. In this case, the degradation could be enhanced or retarded, as well as the antioxygenic activity of some EOs, such as thymol and eucalyptol, which made them more attractive, especially in the food packaging industry [317].

Furthermore, the barrier properties of the films are enhanced by the inclusion of EOs, with EOs playing a significant role in improving water barrier properties. Specifically, water molecules predominantly diffuse through the continuous polymer phase, where EO lipid droplets are situated. These discontinuities contribute to an increased tortuosity factor for water transfer within the matrix [324].

In particular, films incorporating citrus essential oil exhibited a more significant reduction in water barrier properties compared to films with tea tree essential oil. This observation can be explained by the higher hydrophobicity of the primary constituent of citrus essential oil, limonene, compared to the primary constituent of tea tree oil, terpinen-4-ol [112]. Numerous studies suggest that this effect depends on various structural factors, including the matrix type, the composition and quantity of the added oil, and their interactions with the matrix.

The incorporation of essential oils (EOs) into the films resulted in a notable decrease in oxygen permeability values. This reduction can be attributed to the increased solubility of the gas in the lipid phase, with the addition of lipids generally improving oxygen barrier properties of the films [113]. The positive effect of EOs on the oxygen barrier properties of films is further attributed to the lower oxygen solubility in more polar oils, such as thyme and basil EOs, and the presence of antioxidant compounds that play a role in oxygen scavenging.

When selecting antimicrobial agents, the effectiveness of the additive is determined by the specific requirements of the application. Various options have been explored in recent years for polymeric matrices, particularly in PHB and PLA. Among the most promising candidates, Eos, especially thymol and carvacrol, stand out for their demonstrated antimicrobial and antioxidant activities, making them versatile choices for enhancing polymer properties. Not only is their antimicrobial potential significant, but the level of their influence, as determined by their minimum inhibitory concentration (MIC), also highlights their practical value. The ability of EOs and other additives to contribute to active food packaging, alongside their FDA-recognized status as Generally Recognized As Safe (GRAS) additives, further boosts their appeal. Additionally, the growing market demand for essential oils emphasizes their potential as a preferred additive. The selection of the best additive is influenced by factors such as solubility, volatility, and its impact on biodegradation, underscoring the need for a careful, application-specific selection process.

8.2. Other types of antimicrobial agents

There are several types of antimicrobial agents, such as peptides, metals, antibiotics, etc. A peptide is an amino acid chain that is relatively short in length, and its bonds link the amino acids in a peptide's sequence. Antimicrobial peptides (AMPs) are small molecular peptides and types of biomaterial that play a critical role in the innate immunity of hosts against a wide range of microorganisms, including bacteria, fungi, and viruses [325]. They are produced from both natural and synthetic sources, and to date, the AMP database has documented 6105 generic AMPs (including natural and synthetic AMPs). Natural AMPs originate from six kingdoms: bacteria, archaea, protozoa, fungi, plants, and animals. Due to the distinctive membrane rupture mechanism of AMPs, they have emerged as potent candidates for antimicrobial agents [326]. These agents interact with the membrane of the microbes by electrostatic interaction and cause damage to the physical morphology of the bacteria. The carpet mechanism, disordered toroidal pore, toroidal pore, and barrel-stave pore have all been postulated as mechanisms for membrane retardation of bacteria by antimicrobial peptides [327]. Moreover, due to the fungi's eukaryotic cellular structure, peptides can provide higher antifungal activity than traditional antibiotics [328]. The first AMP isolated from bacteria is nisin, which is produced by *Lactococcus lactisis* and demonstrates cytotoxicity against other types of bacteria, enabling it to compete for nutrients in the environment [329]. Nisin can destroy the bacterial membrane, and disrupting the genomic DNA of the bacteria can inhibit cell function [330]. In recent decades, nisin has been known as a good antimicrobial additive for polyesters, especially for PLA, as it has a high bactericidal activity and is approved as a safe food preservation [331]. It has been reported that nisin could inhibit *Staphylococcus aureus* when it is blended with phosphorylated soybean protein isolate /poly(l-lactic acid)/ Zirconiumdioxid (ZrO_2) and has proved to be a promising additive in wound dressing, food packaging, and medicine [332].

Biometallic materials are used in implants and various biomedical applications due to their precise mechanical performance [333]. Using different metal-based additives, particularly metal-organic frameworks (MOFs), as antimicrobial agents is another possibility that has attracted high attention in the last years, such as zinc, copper, titanium, silver, etc. [334–336]. Silver, particularly silver nanoparticles (AgNPs), has recently received much attention due to its antimicrobial properties [337,338]. For a long time, these nanostructures have been used as antibacterial agents in the health and hygiene industry [339], cosmetics [340], and food storage [341]. The effectiveness of nanosilver-based biomaterials as antimicrobial agents has been evaluated against a wide range of pathogenic microorganisms, including bacteria, fungi, viruses, and yeasts [342,343]. Although the mechanism of action of nanostructures is not yet clearly known, they probably exert their antimicrobial effects through one of the following methods: (a) Microbial membrane damage and structural changes due to AgNPs binding to the cell surface (cavity formation, membrane perforation, and cytoplasm leakage), (b) cell structure damage result from the release of free Ag ions, and the generation of reactive oxygen species (ROS) or deactivation of proteins enzymes, and nucleic acids [344,345]. All these things have made AgNP to be considered a good alternative to antibiotics. [346,347]. Moreover, silver ions and silver nanoparticles (even at low content) have been reported as good ingredients for coating polyester, such as PLA and PHBV, to inhibit the activity of feline calicivirus in vegetables, which is a highly contagious virus and results in a serious respiratory illness [348]. The other antimicrobial group to be mentioned is antibiotics, which are known as the first natural antimicrobial agent and are produced mainly by microorganisms [349]. There are several antibiotics, such as gentamicin sulfate, daptomycin, imipenem, etc. used to inhibit microbes in polymer industries with various categories such as food packaging and drug delivery [350–352].

One issue in incorporating antimicrobial additives into biodegradable polymers is the potential impact on the polymer's properties. Some additives (especially organic oils or acids) can plasticize the polymer or react during processing. Therefore, they can change mechanical or thermal performance [353]. Besides, the antimicrobial effect may diminish as the active compound is depleted or loses potency (e.g. volatilization of essential oils). Moreover, metal nanoparticles and certain biocides can be toxic to humans or ecosystems if they migrate out; for example, silver ions are effective antimicrobials but must be used in low concentrations to avoid cytotoxicity. Another limitation in using antimicrobial additives is processing stability. Many natural antimicrobial agents (like herbal extracts, enzymes, or bacteriocins) are sensitive to the high temperatures of melt processing and can degrade, which complicates their integration into plastics [353,354]. For instance, incorporating thyme or oregano oil directly into PLA can lead to significant losses of volatile components during extrusion, reducing efficacy.

To address these issues, current research is focusing on controlled-release and nano-encapsulated antimicrobials. By encapsulating essential oils or bioactive compounds in nanocarriers (nanoclays, cyclodextrins, or polymer microcapsules), their volatility can be reduced and a sustained release achieved, maintaining the antimicrobial activity over a longer duration. There is also a push towards multi-functional additives that provide antimicrobial action alongside other benefits; for example, lignin or certain plant extracts can concurrently improve UV stability and impart antimicrobial properties [189,355]. Another promising research direction is developing inherently antimicrobial biodegradable polymers or coatings. For instance, modifying polyester chains to include quaternary ammonium groups or peptides that are antimicrobial by nature can eliminate the need for migrant additives. Researchers are also examining the possibility of combining different antimicrobials (e.g. a metal nanoparticle with an essential oil or chitosan) to achieve broad-spectrum activity at lower individual concentrations.

9. Influence of additives on various industries

Their application in various industries has been highly developed by using additives that change and improve particular characteristics of the biodegradable polyesters. This chapter discusses how various additives that have been reviewed will affect the applicability and performances of biodegradable polyesters within major industries such as food packaging, agriculture, biomedical, automotive, and 3D printing industries. Fig. 9 illustrates the key applications of additives in various industries.

The food packaging industry requires materials with excellent barrier properties, antimicrobial activity, and mechanical load resistance, while also being ecologically friendly. Additives can modify biodegradable polyesters like PLA and PBAT to address industry challenges, including antimicrobial agents, hydrolysis stabilizers, and plasticizers. Plasticizers play the main role in making food packaging materials suitable for wrapping materials or containers by making PLA or PBAT more flexible [356,357]. Besides, antimicrobial agents are used as a solution to provide food packages with the ability to inhibit microbial growth. For both industries, essential oils show a promising potential, and they can be studied with their microbial inhibition ability before being incorporated into the polyester [358,359]. Essential oils can further be more beneficial because of their antioxidant activity, which protects packaged goods from oxidation. To maintain its integrity, avoiding packaged goods from humidity is of actual importance, where hydrolysis stabilizers such as carbodiimides can be among the best solutions [359], but the amount of this additive in the composite should be controlled as it is a type of coupling agent. However, PMMA is another option used by researchers in this industry as a hydrolytic stabilizer. The question that might be raised here is about the eco-friendliness of PMMA and how it can meet stringent regulations in food packaging. Despite the fact that it is petroleum-based, PMMA in the PMMA-PBAT blend is still compostable if PMMA content is low. In this context, as PMMA is usually used in low content in PBAT for the food industry, it can meet EN 13432 and ASTM D6400, requiring 90 % of material biodegradable in industrial composting for less than 6 months. Besides, this additive is transparent and non-toxic, which makes it safe to be used in the food industry.

In agriculture, crosslinkers and chain extenders, nucleation agents, and antimicrobial agents are among the most required additives for biodegradable polyesters. There are several applications of biodegradable polyesters in this industry, such as coating for slow-releasing fertilizers and coating and mulching films [360,361]. Mechanical strength is one of the factors that should be considered for coating, where crosslinkers can be used to ensure the standard of this property [362]. For some of the biodegradable polyesters, such as PBS and PHB, there would be other minuses that should be addressed, such as their high biodegradability in soil. To be more specific, the seed inside the soil needs to survive before germination from fungi, slow-releasing fertilizers need to stay in good shape for almost 3–4 months, and mulching films for 1–3 months. As those polyesters are highly degradable, they need to be resistant to microorganisms, resulting in the need to blend antimicrobial agents such as essential oils [310]. However, their allelopathic properties, their influence on the activity of soil enzymes that are crucial for organic matter breakdown and nutrient cycling, and the influence of these additives on soil fauna, such as earthworms, should be studied before using them. For further improvement, nucleating agents such as talc and cellulose nanocrystals can be implemented to improve mulch films' thermal and mechanical properties by promoting their crystallization [363].

The biomedical industry is another industry in which biodegradable polyesters can be implemented. Elongation in the material used in medical devices and flexibility are two factors that should be considered, increasing the need for plasticizers in this area. It is worth noting that there are plenty of additives in each category that can be used in one

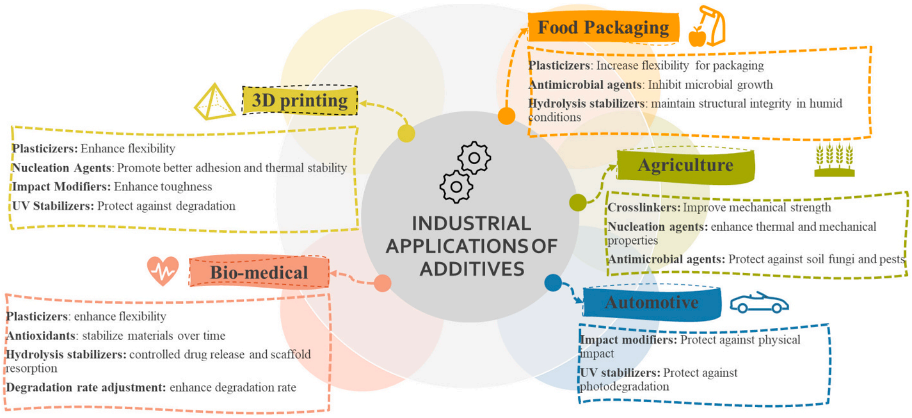


Fig. 9. Industrial applications of additives on polyesters.

industry, while they can be used in another one. For instance, acetyl tributyl citrate (ATBC) is a good option as a plasticizer when it comes to food packaging, while it wouldn't be a good option for the biomedical industry. The reason is that in food packaging, standards such as FDA and EFSA, low toxicity, and high compatibility with polyesters like PLA should be considered. On the other hand, in the biomedical industry, biocompatibility and being biodegradable in a controlled manner are the main factors that should be considered. Lignin as an antioxidant is another additive that has wide usage in biomedical industries, specifically in PLA, ensuring that the material can be stable over a timeframe [364]. Moreover, essential oils have recently been based on their antioxidant activity and their biocompatibility. Hydrolysis stabilizers such as PMMA are used in this industry to ensure controlled drug release or scaffold resorption in tissue engineering applications [365,366]. Although carbodiimide is mentioned as a suitable hydrolysis stabilizer in food packaging, PMMA is preferable. In the food packaging industry, materials usually need to maintain their structure in wet and humid conditions, such as in refrigerated storage, where carbodiimide could be preferable. On the other hand, PMMA have been studied extensively, and their compatibility meets the stringent standards [365,367]. This industry requires predictable degradation rates, such as biodegradable implants or drug delivery systems.

Impact modifiers and UV stabilizers are among the most important that have been used in the automotive industry. Biodegradable polyesters are used for components like interior trims and panels, where butadiene-styrene and ethylene-epoxy copolymers can be implemented into them, improving their impact resistance and roughness. HALS and UV absorbers are two additives that are used in this industry to protect the materials in this industry from photodegradation, which is among the most important issues in this industry. Moreover, to make biodegradable polyesters suitable for coating and seals in this industry, plasticizers such as epoxidized polyesters can be used as well.

In the 3D printing industry, several types of additives can be used to improve the properties of biodegradable polyesters, especially PLA. Within the 3D printing industry, there is extensive interest in such biodegradable polyesters, especially PLA, for reasons of processability, renewability, and ecological acceptability. To enable better adhesion and faster cooling during printing, nucleation agents have been incorporated into PLA to enhance their crystallinity and thermal stability [234,368]. Improving the elongation of PLA and its flexibility and reducing its brittleness are among the top requirements of using this polymer in the 3D printing industry, where plasticizers such as ATBC have been confirmed by researchers for their performance in this area [368,369]. Furthermore, objects that are 3D-printed and are exposed to sunlight require UV stabilizers to reduce their photodegradation. In this area, HALS are widely used in the automotive industry. Moreover, for 3D-printed materials that require high-quality mechanical properties, impact modifiers have been used to improve the toughness. Acrylic copolymers are among the most used impact resistors in this industry due to several benefits. One of the main factors is its compatibility with most of polymers in this industry, especially PLA. Moreover, if a hydrolysis stabilizer such as PMMA is used as an additive for PLA, acrylic copolymers have a high level of compatibility with PMMA and PLA [365,370]. Furthermore, for this matrix, they can add clarity and transparency [371]. In case the 3D-printed materials are used in medical or food-contact applications, antimicrobial agents, mostly nano additives such as zinc oxide or silver nanoparticles, would be ideal [372,373]. The main reasons behind the less applicability of essential oils antimicrobial agents, apart from the encapsulated ones or those that are chemically-stabilized, in this industry are the high level of thermal degradation during extrusion (when it comes to industry) and its lower compatibility with PLA in comparison with other additives [374,375]. The in vivo application of 3D-printed biomedical polymers requires controlled degradation within the body [376]. However, polymers with high molecular weight and crystallinity, such as PLA, tend to degrade slowly. One effective approach to adjust their degradation rates is the incorporation of additives like bioactive glass particles [377], cellulose nanocrystals [378], cellulose nanofibers [379], bioglass [380], and tricalcium phosphate, which can accelerate degradation.

The other industries that are prone to use modified biodegradable polyesters with additives are the construction, textile, electronic, and consumer products industries [381]. Plasticizers are highly used in the textile, construction, and consumer products industries [382,383]. The only factor that should be considered in construction is that the plasticizers are phosphate-based, which is also flame-retardant, ensuring the safety of construction materials. Consumer products and electronic industries widely use impact modifiers for biodegradable polyesters. Consumer products are usually use this type of additive for reusable bottles and toys, and in the electronic industry, impact modifiers used to enhance the durability and shock resistance of biodegradable polyesters.

10. Future perspectives

Although natural additives are becoming more attractive than polymer-based ones, significant investment and research are still required to reduce the use of polymer-based additives. It is clear from the current state of research on biodegradable polyester additives that much remains to be discovered regarding their effectiveness, environmental impact, and scalability. Therefore, future studies in this field should focus on investigating innovative additives, refining processing techniques, and developing more accurate models for predicting biodegradation rates. Furthermore, research efforts must aim to gain a comprehensive understanding of their characteristics, applications, and limitations, given the potential of biodegradable polyesters to address urgent environmental issues such as plastic pollution. This will ensure their successful integration into existing waste management systems. Future research could focus on exploring new natural additives that are cost-effective and derived from waste, such as lignin, which has great potential to address various issues related to polymer degradation, as well as mechanical and thermal challenges. Moreover, improving human health and protecting the environment are key considerations when transitioning to biodegradable plastics and modifying the use of conventional additives, whether through technological advancements or economic penalties. As with other recommendations, studying different types of additives to be used simultaneously would be valuable in order to have highly applicable and environmentally friendly polymers. Nowadays, most research studies focus on the environmental aspects, while the mechanical and thermal properties of polymers are the factors that should be considered for making a highly applicable material. In this regard, studying different aspects of additives can result in additives that are not only environmentally friendly and cheap, as mentioned above, but also more useful in terms of thermal and mechanical properties.

Last but not least, studying the influence of these additives on the soil, compost, or water after biodegradation is crucial. Researchers reported that the degradation of biodegradable polyesters, such as PHB, can lead to nutrient Limitations, reduction in pH, and altered microbial community composition. This can change the soil ecosystem in the long term. In this regard, the influence of each additive should be studied in terms of the soil ecosystem before using it. For example, sometimes, a high percentage of essential oils might negatively affect important soil microorganisms. Therefore, studying this influence will help us select the appropriate additive and determine whether the optimal concentration is of actual value. A review of the influence of biodegradable polyester biodegradation on soil in the future would be beneficial in this research area.

11. Conclusions

In conclusion, it should be noted that polymer additives are essential for improving the qualities of plastics and other polymeric materials. These additives are appropriate for a variety of applications since they can increase the finished product's strength, durability, and aesthetic appeal. The use of polymer additives is anticipated to rise in the upcoming years due to ongoing technological and scientific developments, which will result in the creation of new and enhanced goods. Based on the unique needs of each application, producers must carefully choose suitable additives.

One of the most important factors that should be considered is the side influence of each additive on the properties of a polymer. To be more specific, when an additive is added to a polymer for a specific purpose, it might have a negative or positive effect on other properties, which should be considered. For instance, when essential oils are used to enhance a polymer's antimicrobial properties, they sometimes have a negative effect on its mechanical or thermal properties. In this case, conducting comprehensive research on all influencing properties would be of actual importance.

Due to the severe regulations regarding the transition to environmentally friendly materials, it is not only important to use biodegradable polymers but also to choose environmentally friendly additives. This transition also needs to be expanded by exploring new types that can address the current problems regarding the high price of these additives. This can be divided into several research areas, such as chemistry, biochemistry, microbiology, and genetics.

CRediT authorship contribution statement

Ahmad Fayyazbakhsh: Writing – review & editing, Writing – original draft, Methodology, Funding acquisition, Conceptualization. **Nima Hajinajaf:** Writing – review & editing, Writing – original draft. **Hamed Bakhtiari:** Writing – review & editing, Writing – original draft. **Michael Feuchter:** Writing – review & editing, Writing – original draft. **Ilaria Improta:** Writing – review & editing, Writing – original draft. **Ehsan Salehi:** Writing – review & editing. **Sara Kamal Shahsavar:** Writing – review & editing. **Marketa Julinova:** Writing – review & editing, Writing – original draft. **Amirehsan Ghasemi:** Writing – review & editing, Writing – original draft. **Bita Ghasemi:** Writing – review & editing, Writing – original draft. **Reza Afsharnia:** Writing – review & editing. **Marek Koutný:** Writing – review & editing. Young-Cheol Chang: Writing - Review & Editing, Visualization, Supervision.

Declaration of competing interest

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

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Data availability

All data analysed through this study are included in this manuscript.

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