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Review

Advancing Sustainability: Utilizing Bacterial Polyhydroxyalkanoate for Food Packaging

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Abstract: Polyhydroxyalkanoates (PHAs) are promising biodegradable polymers known for their biodegradability and eco-friendly properties. Recent studies indicate that PHAs can reduce the environmental impact by up to 50% compared to petroleum-based plastics. This comprehensive review evaluates the application of PHAs in sustainable food packaging, covering over 100 studies published between 2018 and 2023. The review highlights advancements in PHA production, with a focus on submerged and solid-state fermentation methods, achieving up to a 60% improvement in production efficiency through optimized culture selection. Sustainable extraction and purification methods have been identified, reducing energy consumption by 30%. Blending PHAs with other biodegradable polymers like polylactic acid, starch, and cellulose enhances material performance, with up to a 40% improvement in mechanical properties. The incorporation of antimicrobial agents and essential oils has been shown to extend the shelf life by 25% while maintaining food safety standards. This review underscores the potential of active PHA-based packaging in improving the barrier properties by 35% when combined with coatings, positioning PHA as a key material for the future of environmentally responsible and safe food packaging.

Keywords: polyhydroxyalkanoate; biodegradable polymers; sustainable food packaging waste biomass; active packaging

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1. Introduction

Biobased and biodegradable food packaging films, derived from renewable sources like plant-based materials or agricultural waste, offer a sustainable alternative to conventional fossil fuel-derived packaging materials [1]. These films break down naturally into harmless by-products, reducing pollution and waste accumulation [2]. As sustainability gains importance among consumers and industries, the adoption of biobased food packaging films or biocomposites, such as those reinforced with natural fibers like cellulose, hemp, or flax, is also gaining traction [3], as eco-friendly packaging solutions continue to grow [2]. They represent an innovative approach at the intersection of sustainability and packaging technology.

Annual production of food waste from the 28 European Union (EU) countries has been on the rise, with an estimated production of 116 million tons in 2020 [4]. This waste includes not only food that is discarded but also by-products generated during food processing. Additionally, agro-industrial wastes globally are estimated to make up more than 30% of agricultural production each year, emphasizing the significant volume of waste generated by these sectors [5]. Efforts are being made to valorize these waste streams. Research has explored various methods to convert agro-industrial waste into value-added products such as biofuels, bioplastics, and bioactive compounds [6–8].

Biodegradable plastics have gained attention as environmentally friendly alternatives to conventional plastics due to their natural decomposition ability. Types like polylactic

acid (PLA), polycaprolactone (PCL), starch blends, and polybutylene succinate (PBS) offer diverse applications and environmental benefits [9]. Among these, PHAs stand out for their microbial origin, versatility, and superior biodegradability. PHAs are synthesized by bacteria as intracellular storage compounds under nutrient-limited conditions with an excess of carbon sources (sugars or lipids). This natural production process involves feeding bacteria with organic substrates, which they convert into PHAs that accumulate within their cells. After fermentation, the PHAs are extracted and purified for various applications. These include food packaging, agricultural films, disposable containers, medical devices, sutures, drug delivery systems, and tissue-engineering scaffolds due to their biocompatibility and non-toxicity. PHAs are poised to play a crucial role in sustainable plastic alternatives, helping to address global plastic pollution [10]. However, the development of biodegradable polymers, including PHAs, for food packaging brings with it certain drawbacks that merit consideration. Firstly, the production cost of PHAs is currently higher than that of conventional plastics, making them less economically viable for widespread use. While advancements in microbial engineering and fermentation processes are gradually reducing these costs, significant investment and research are still needed to make PHAs more affordable. Secondly, the cultivation of raw materials for PHA production, such as corn or sugarcane, can have environmental impacts. Large-scale farming of these crops may lead to land-use changes, deforestation, and increased water consumption. Sustainable agricultural practices must be implemented to mitigate these effects. Thirdly, the biodegradation process of PHAs, although environmentally beneficial, may require specific conditions to occur efficiently. Industrial composting facilities are often necessary to ensure complete and rapid decomposition, which may not be available in all regions. This limitation can reduce the effectiveness of PHAs in reducing plastic waste in environments where proper composting infrastructure is lacking [11]. Lastly, the performance characteristics, i.e., the limitation of the processability and material properties of biodegradable polymers, including PHAs, may not always match those of conventional plastics [12]. Biodegradable polymers often demonstrate shortcomings in terms of their ability to act as diffusion barriers for small molecules (oxygen, carbon dioxide, water vapor), which is crucial for food-contact applications [12]. Another drawback is the mechanical strength of biodegradable polymers. Biobased alternatives may lack the tensile strength and flexibility of conventional plastics, affecting their effectiveness as packaging materials [13]. Careful optimization of the mechanical properties is essential to ensure these films can withstand handling, transportation, and storage without compromising the packaging integrity or food safety. Moreover, the availability and cost of biodegradable polymers can pose significant challenges. Presently, the production capacity for biobased polymers remains limited in comparison to that for synthetic polymers from petrochemicals [14]. Restricted production capacity can lead to elevated costs for biodegradable packaging materials, rendering them less economically viable for widespread adoption [5].

Despite these challenges, the potential environmental benefits of PHAs make them a promising option for reducing plastic pollution. The importance of utilizing PHAs for food packaging is underscored by several key factors: environmental impact, plastic pollution, biocompatibility, sustainability and regulatory compliance. Continued research, technological advancements, and sustainable practices are essential to overcoming the current drawbacks of PHAs, such as the high production costs and performance limitations. By addressing these challenges, we can fully realize the potential of PHAs in creating a more sustainable future for food packaging, ultimately helping to reduce the environmental footprint of the packaging industry.

The novelty of the present study lies in its holistic approach to evaluating the potential of PHAs for food packaging, addressing both the material's capabilities and its broader environmental impact. This study will comprehensively assess the essential characteristics required for food packaging films—such as the barrier properties, mechanical strength, transparency, and biodegradability—while also providing a detailed overview of various biobased and biodegradable polymers used in the industry. It will delve into the microbial

production process of PHAs through the fermentation of renewable resources, offering insights into the scalability and sustainability of this approach. Additionally, this study will explore the concept of active packaging, investigating how PHAs can be utilized to enhance the functionality and safety of food packaging. Finally, it will conduct a life cycle assessment (LCA) of biobased polymers in food packaging, providing a thorough evaluation of their environmental impacts. This integrated methodology aims to present a comprehensive understanding of the potential and challenges of using PHAs in the food packaging industry.

2. Important Properties of Food Packaging Films

In order to preserve food quality and safety, food packaging films need to offer certain properties contributing to their efficacy (Table 1). Ensuring resistance to tearing or puncturing during handling, transportation, and storage, tensile strength and toughness are indispensable attributes [15]. Food packaging films typically exhibit tensile strength in the range from 6 to 170 MPa, while the toughness values range from 50 to 100 kJ/m^2 [16]. High elongation at break is also desirable, usually from 100 to 900%, indicating that the films can withstand stretching and deformation without breaking or tearing [15,17]. Another important property is thermal stability, which ensures that films will withstand temperature variations without compromising the mechanical and barrier properties [15]. The thermal stability of packaging films is typically observed with a decomposition temperature of 350–420 °C [18]. The oxygen and water vapor barrier properties are also evaluated, preventing oxidation and spoilage of the packing food [19]. Low oxygen and water vapor permeability extend the shelf life and maintain food freshness [19,20]. The optimal values for water vapor transmission rates typically range from 0.001 to 0.3 g/(m^2·day) and 1×10^{-4} to 44.76×10^{-4} cm^3/(m^2·day) for oxygen permeability [21].

Table 1. Most important properties of food packaging films [22–24].

Property	Petroleum-Based Polymers	Biodegradable Polymers
Barrier Properties	High moisture, oxygen, and gas barriers (WVTR: 6.9 g/m^2 day PET, 1.5 g/m^2 day LDPE; OTR: 36 mLO_2/m^2 day bar PET, 1624 mLO_2/m^2 day bar LDPE)	Moderate to good, but generally lower than petroleum-based (e.g., PLA has moderate/low oxygen barrier properties (180 mLO_2/m^2 day bar), and low moisture barrier (35.5 g/m^2 day); PHB has good oxygen barrier properties (22 mLO_2/m^2 day bar), but moderate to low moisture barrier (5.5 g/m^2 day)
Mechanical Strength	High tensile strength (PET 59.4 MPa), good impact resistance	Varies; PHB has good strength (43.9 MPa), while PLA and starch blends are more brittle
Thermal Stability	High (PETs have high melting points, 245 °C)	Lower; PCL 70 °C, PLA and TPS melts around 140–160 °C, while PHB is more thermally stable (180 °C) but still lower than PET
Transparency	Excellent (especially PET (glass-like visibility))	Good; PLA is highly transparent, PHA is less so, starch blends can be cloudy
Flexibility	High (especially LDPE)	Varies; PHA and some blends are flexible, but PLA is more rigid
Biodegradability	Non-biodegradable	Biodegradable under industrial composting conditions (PHA, PLA, starch blends)
Compostability	Not compostable	Compostable under specific conditions (e.g., industrial composting for PLA, PHA)
Recyclability	Recyclable (but limited recycling rates in practice)	Limited recyclability, typically more focused on composting or biodegradation

Table 1. *Cont.*

Property	Petroleum-Based Polymers	Biodegradable Polymers
Cost	Relatively low and well-established (USD 800–USD 1600 per metric ton) Factors influencing cost: LDPE isLDPE is generally cost-effective, but prices can fluctuate based on crude oil prices, supply-demand dynamics, and regional production capabilities.	Higher than petroleum-based polymers, though costs are decreasing with scale (USD 2000–USD 6000 per metric ton)
Environmental Impact	High carbon footprint, non-renewable.	Lower carbon footprint, derived from renewable resources, fully biodegradable Due to natural conversion, PLA emits 2.8 kg CO_2 kg^{-1} during its life cycle. PLA saves ~66% of the energy required to produce conventional plastics Starch utilization in bioplastics production causes a reduction in GHG emissions (>80%) and fossil fuel consumption (>60%). When compared to synthetic plastics, starch might cause an increase in eutrophication potential and land usage
Application Suitability	Widely suitable across various applications	Suitable for specific applications like food packaging where biodegradability is prioritized

* Water vapor transmission rate (WVTR) [g/m^2day] (23 °C, 85% ΔRH); oxygen transmission rate (OTR) [mLO_2/m^2day bar] (25 °C, 0% RH); PET (polyethylene terephthalate); LDPE (low-density polyethylene); PLA (polylactic acid); PHB (polyhydroxybutyrate); TPS (thermoplastic starch).

Moreover, inhibiting microbial growth by encompassing antimicrobial additives is an important step as well. The strong antimicrobial properties of the films inhibit the growth of bacteria and fungus on the surface of packaging film, thus reducing the risk of contamination and spoilage of the packaged product [20,25]. Commonly used antimicrobial agents include chitosan, essential oils, silver and metal oxide nanoparticles [26]. The minimal inhibitory concentration (MIC) values highlight the effectiveness of these antimicrobial additives in inhibiting the growth of specific microorganisms. In Table 2, the MIC values of common antimicrobial agents are shown.

Table 2. Sample MIC values of commonly used antimicrobial agents.

Antimicrobial Agent	MIC	Microorganism Type	References
Chitosan	0.1–1.0 mg/mL	Gram-positive bacteria (*Staphylococcus aureus*, lactic acid bacteria, *Listeria innocua*)	[27]
Chitosan	0.5–2.0 mg/mL	Gram-negative bacteria (*Escherichia coli*, *Pseudomonas* spp., *Salmonella* spp.)	[27]
Silver nanoparticles	0.001 and 0.1 μg/mL	*Staphylococcus aureus*, *Escherichia coli*	[28]
Essential oils	0.25 to 2.0 μg/mL	Gram-positive (*Listeria monocitogenes*, *Listeria innocua*, *Brochothrix thermosphacta*) and Gram-negative bacteria (*Escherichia coli*, *Salmonella* spp.)	[29]
SiO_2 nanoparticles	0.156 mM	*Streptococcus mutans*	[30]
ZnO nanoparticles	31.25 μg/mL	*Escherichia coli*	[31]

Not least, the packaging material serves to label the product with mandatory and voluntary information. The mandatory information for prepacked foods includes the product name, list of ingredients, net weight, nutritional information, manufacturer, country of origin, and instructions for use, while for beverages containing more than 1.2% by volume

of alcohol, the actual alcoholic strength by volume, which is prescribed by Regulation (EU) No. 1169/2011. The voluntary information important for consumers could also include nutrition or health claims (Regulation (EC) No. 1924/2006). Applying different colors and bar codes also conveys important technical details of the declaration.

Plastic films such as polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET), and polystyrene (PS) offer numerous advantages for food packaging due to their versatility, functionality, convenience, and cost-effectiveness. However, the preservation of food properties over an extended period may require more than single-layer films can provide, leading to the adoption of high-barrier multilayer films. These advanced films are manufactured using technologies like thermal lamination and co-extrusion. Despite their excellent properties, non-biodegradable plastic films derived from fossil fuels pose significant environmental challenges. To tackle this issue, there is a growing emphasis on developing new polymers sourced from biological origins, such as biobased multilayer barrier films, to meet the stringent barrier requirements of fresh food packaging while promoting sustainable packaging solutions [32,33].

To ensure that packaging films are biodegradable and compostable, they must comply with the EN 13432:2003 standard [34]. This standard specifies that polymers must undergo at least 90% conversion to CO_2 within 180 days under specific temperature, humidity, and oxygen conditions to be considered compostable [35]. Additionally, the polymer should be intrinsically biodegradable, disintegrate without posing ecotoxicity risks, and fully degrade into CO_2, water, and biomass without leaving distinguishable residues, aligning with the ASTM D6400 and ISO 17088 guidelines [36]. Compliance with these standards ensures that polymers used in composting applications undergo comprehensive biodegradation, supporting environmentally friendly waste management and promoting circular economy principles by closing the materials' life cycle.

The European Union has implemented several key regulations that align with sustainability development goals, aiming to reduce environmental impacts and promote sustainable practices in packaging and chemical usage, such as Directive (EU) 2018/852 [37]—Amendment to Directive 94/62/EC [38] on Packaging and Packaging Waste, Directive (EU) 2019/904 [39]—Single-Use Plastics Directive, REACH Regulation (EC) No. 1907/2006 [40]—Registration, Evaluation, Authorisation, and Restriction of Chemicals, and EU Ecolabel Regulation (EC) No. 66/2010 [41]. These regulations collectively support the EU's sustainability objectives by promoting the reduction of waste, the responsible use of chemicals, and the development of environmentally friendly products. They are critical in driving the transition toward a more sustainable and circular economy within the EU.

3. Biobased and Biodegradable Polymers

Biobased polymers, derived from primary biomass such as plants or organic materials like starch from corn or potatoes, represent a significant avenue for biopolymer production [15,17]. However, recent research has shifted toward utilizing waste biomass and by-products, termed second-generation biomass, to create biopolymers [25]. This approach holds promise in adding value to resources currently considered waste. The classification of biobased polymers based on the raw material used for production is shown in Figure 1 and the properties, advantages, disadvantages, limitations, and real-time applications of some biobased polymers are shown in Table 3.

PHAs, derived from second-generation biomass, offer a compelling alternative to petroleum-based polymers, particularly in food packaging applications, due to their biodegradability and versatile properties. Polymers like polylactide (PLA) and chitosan have also gained attention for their eco-friendly characteristics, demonstrating potential in food packaging [27]. These advancements in biodegradable polymers for packaging underscore a shift toward sustainable solutions, addressing both environmental concerns and the need for effective food preservation [42].

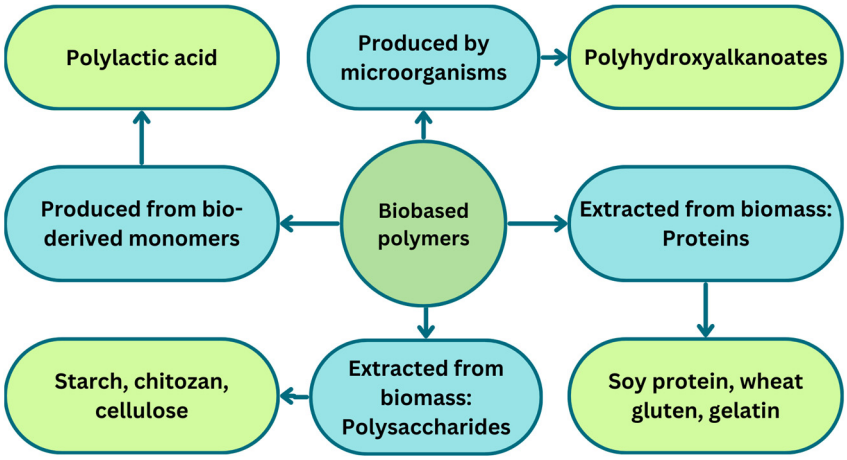

Figure 1. Classification of biopolymers based on the raw material used for production.

Table 3. Biobased polymers: properties, advantages, disadvantages, limitations, and real-time applications [22–24].

Polymer	Properties	Advantages	Disadvantages	Limitations	Real-Time Applications
PLA	Transparent, high strength, biodegradable, thermoplastic	Renewable source, compostable, good clarity, easy processing	Brittle, poor thermal stability, low impact resistance	Limited use in high-temperature applications	Food packaging, disposable cutlery, 3D printing, medical implants
PHA	Biodegradable, good barrier properties, thermoplastic	Biocompatible, high biodegradability, versatile mechanical properties	High production cost, brittle	High cost and limited commercial availability	Biodegradable packaging, medical sutures, agricultural films
TPS	Biodegradable, flexible, thermoplastic	Low cost, compostable, easily blended with other polymers	Poor moisture resistance, low mechanical strength	Limited durability, needs blending with other polymers for improved properties	Biodegradable bags, packaging films, disposable items
PBS	Biodegradable, good thermal stability, flexible	Good mechanical properties, heat resistant, compostable	Higher cost compared to other biobased polymers	Limited commercial availability	Packaging materials, agricultural films, biodegradable tableware
PBAT	Biodegradable, flexible, good impact resistance	Flexible, good mechanical properties, suitable for blending	Derived partially from fossil fuels	Still partly reliant on petrochemical sources	Biodegradable films, agricultural mulch, compostable bags
PCL	Biodegradable, low melting point, easy to process	Biocompatible, good flexibility, blends well with other polymers	Slow biodegradation rate, low melting point	Limited use in high-temperature applications	Medical devices, drug delivery systems, biodegradable packaging
Cellulose-based polymers	Biodegradable, good mechanical properties, moisture-sensitive	Abundant, renewable, good film-forming ability	Sensitive to moisture, difficult to process	Limited water resistance, requires additives for improved durability	Food packaging, films, textiles, coatings

* PBS (polybutylene succinate), PBAT (polybutylene adipate terephthalate), PCL (polycaprolactone).

Characterization studies on the biodegradability of biobased packaging materials are crucial for evaluating their environmental impact and overall sustainability. These studies employ various analytical techniques to monitor and understand the degradation process, which can vary depending on the material composition, environmental conditions, and exposure duration. The key techniques used in these studies include Fourier-transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), differential scanning calorimetry (DSC), thermogravimetric analysis (TGA), mechanical property testing, water absorption tests, nuclear magnetic resonance (NMR) spectroscopy, and microbial analysis [43]. FTIR is utilized to detect changes in the chemical structure of materials such as PLA during degradation. SEM provides detailed images of surface erosion and morphological changes in packaging materials. DSC measures thermal property changes, such as the melting temperature and crystallinity, to assess how materials evolve during degradation. TGA offers insights into the thermal stability and weight loss of materials like TPS throughout the degradation process. NMR spectroscopy analyzes chemical structure modifications and identifies degradation products. Mechanical property testing assesses changes in tensile strength, elasticity, and brittleness as materials degrade. Lastly, microbial analysis examines the microbial communities involved in the degradation of materials such as PHA and identifies the by-products formed. These characterization studies provide a comprehensive understanding of how biobased packaging materials perform under various environmental conditions, which is essential for determining their suitability for sustainable packaging applications.

3.1. Polyhydroxyalkanoates

PHAs represent a class of biodegradable polyesters that have garnered notable attention in the domain of food packaging, primarily due to their mechanical, thermal, and barrier properties [44]. PHAs' molecular architecture is characterized by a helical structure, with the PHA helix maintained through hydrogen bonds formed between the carbonyl groups of individual monomers [45]. These monomers, identified as hydroxyalkanoates, consist of carbon chains ranging from three to five atoms, categorizing them as "short chain length PHA" (*scl*-PHA), or six or more atoms, designating them as "medium chain length PHA" (*mcl*-PHA), as shown in Figure 2 [45]. *Scl*-PHAs include monomers such as 3-hydroxybutyrate (3HB), 4-hydroxybutyrate (4HB), or 3-hydroxyvalerate (3HV) [46]. On the other hand, *mcl*-PHAs consist of monomers such as poly-3-hydroxyhexanoate (PHHx), poly-3-hydroxyoctanoate (PHO), poly-3-hydroxydecanoate (3HD) [46].

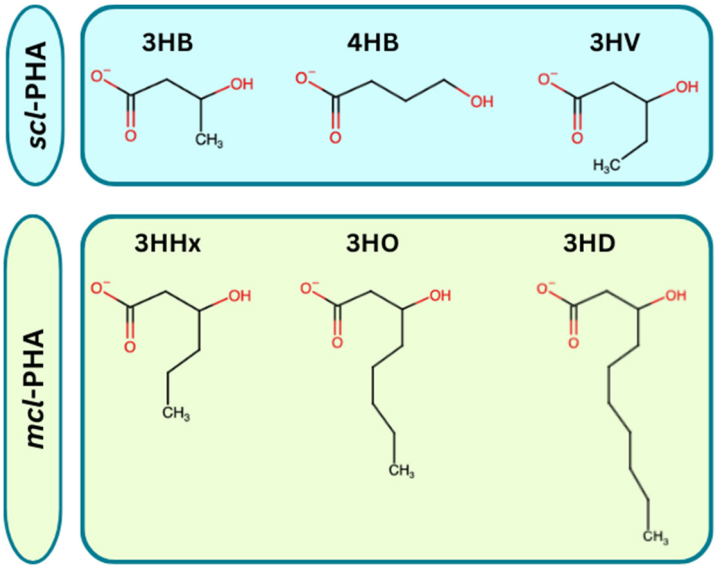

Figure 2. Classification of PHAs.

PHAs are distinguished by their high crystallinity, thermoplastic characteristics, and pronounced brittleness [47]. The most extensively researched PHA, the homopolyester poly(3-hydroxybutyrate) (PHB), stands as the quintessential representative of *scl*-PHA materials. PHB is a partially crystalline and biocompatible homopolymer with mechanical properties similar to PP, except for its lower flexibility. The main limitation of PHB is its high brittleness [47]. Another extensively researched PHA is poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV). PHBV is a copolymer of PHB and 3-hydroxyvalerate (3HV). PHBV, known for its improved mechanical properties compared to PHB, finds versatile applications. The incorporation of 3HV units into the polymer chain increases its flexibility and impact resistance while still maintaining its biodegradability [48].

PHAs are synthesized by specific bacteria, wherein they store PHAs as a form of energy reserve [48,49]. Bacteria capable of producing PHAs include species such as *Cupriavidus necator*, *Bacillius* sp., *Pseudomonas* sp., *Ensifer* sp. and others [50–53]. Accumulated under unbalanced nutritional states, PHAs utilize diverse substrates for fermentative production, ranging from food-based sources like fats and oils to renewable waste-derived materials (agro-industrial, etc.). As of 2022, estimates suggest that the industrial production of PHA has not exceeded 10,000 tons annually; however, PHA production capacities are expected to quadruple in the next five years [54].

In recent times, PHAs have gained traction across diverse sectors, spanning aquaculture, human health, and biofuels, showcasing their versatility and potential for multifaceted applications [55]. Particularly noteworthy are advancements in biomedical applications, with FDA-approved suture materials and scaffolds for bone tissue engineering [44]. While PHA-based hybrid materials show promise for tissue engineering, further research is warranted [56]. Research efforts concerning PHA-based materials extend to exploring composites, blends, thin films, carriers for bioactive substances, and self-assembled carriers for therapeutic applications [56–59].

The widespread application of PHAs is undeniable; however, their mass production remains costly. PHAs produced through the conventional fermentation process cost USD 4–6/kg, which is 5 to 6 times more expensive than polymers derived from fossil sources [60]. The substrate significantly influences the cost of PHAs; therefore, kitchen waste, by-products, cellulose, or activated sludge hydrolysates pose inexpensive alternatives. In addition, for industrial production, consistent PHA quality is crucial—a challenge when produced conventionally. Recently, in the pursuit of cost reduction strategies, extremophilic bacteria and their recombinants have emerged as highly applicable in next-generation industrial biotechnology (NGIB), offering PHAs with both lower production costs and stable properties.

PHBV has been investigated for its suitability in food packaging applications, owing to its well-balanced mechanical properties, including stiffness and tensile strength [47,60–62]. Concerning the barrier properties, PHBV-based films have exhibited promising barrier properties against gases and water vapor, rendering them attractive for use in food packaging applications [63]. Additionally, PHBV films display robust thermal stability, making them well suited for scenarios requiring heat resistance [63]. Furthermore, PHBV is non-toxic, biocompatible and completely biodegradable [64].

However, PHBV films exhibit certain limitations, such as brittleness at lower temperatures, reduced flexibility, and a comparatively higher cost when juxtaposed with conventional plastics, as previously noted [64]. A common approach in improving the limitations of PHBV films is to create blends and composites with other biodegradable polymers and fillers. In the following, we will briefly highlight the most important biodegradable polymer materials that show significant potential in combination with PHA.

The biodegradability and compostability of PHBV are influenced by various factors, including its chemical composition, environmental conditions, and the presence of additives. Research has shown that PHBV can achieve high levels of biodegradation, with some studies reporting up to 87% biodegradation after 75 days under thermophilic composting conditions [65]. To assess PHBV's biodegradability, experimental methods often involve

placing samples in controlled composting environments where the temperature, humidity, and aeration are carefully monitored. For instance, one study found that PHBV membranes fully disintegrated within 7 days under optimal composting conditions of 55 °C and 60% relative humidity [66]. Additional research has explored the degradation kinetics of PHBV in soil and marine environments, indicating that while biodegradation is slower in solid substrates compared to liquid environments, substantial degradation can still occur over extended periods, with complete degradation observed within 600 days in marine conditions [67].

Among the biodegradable polymers, PHBV is one of the fastest polymers to biodegrade under composting conditions. PLA, for example, while biodegradable, exhibits a slower degradation rate compared to PHBV, primarily due to its higher crystallinity and the formation of a biomass layer that hinders microbial access [68]. PCL is characterized by a relatively low biodegradation rate, as its hydrolysis process is slower and often requires specific environmental conditions to facilitate microbial action [69]. On the other hand, TPS demonstrates a rapid biodegradation process, particularly under composting conditions, but its degradation can be impeded by the presence of additives that affect moisture absorption and microbial penetration [70]. Overall, while PHBV stands out for its rapid biodegradability, TPS follows closely, with PLA and PCL exhibiting slower degradation rates under similar conditions.

3.2. Polylactic Acid

Polylactic acid (PLA) stands out as a well-studied biobased and biodegradable polymer, primarily synthesized through industrial polycondensation of lactic acid or ring-opening polymerization of lactide, which is derived from renewable sources such as sugar cane, corn starch, or tapioca [71]. The leading manufacturers of biodegradable PLA films include Ingeo (NatureWorks, Plymouth, MN, USA), PURAC (PURAC Co., Rayong, Thailand), BIOFRONT (Teijin, Tokyo, Japan), HiSun (Revoda, Stoney Creek, ON, Canada), and Pyramid (Tate and Lyle, Pinckneyville, Denmark). These biopolymers are widely utilized across various industries. They are found in disposable household items such as drinking cups, cutlery, trays, food plates, and containers; in food packaging; waste bags; shopping bags; and agricultural applications, including soil retention sheeting and films. Additionally, PLA is used in drug delivery systems, biomedical devices, disposable garments, feminine hygiene products, and diapers. In commercial food packaging, PLA is employed in manufacturing caps (often as PLA blends), coffee capsules and pouches (PLA/PHB), shopping and waste bags (blends of PLA/PHA/PBAT), clear films for fruits and vegetables (PLA or PLA/Bio-PET blends), and teabags (PLA blends). The global production capacity of PLA, according to the European Bioplastics Association, reached 457,000 tons in 2021, reflecting a significant increase from 394,800 tons in 2020 [72].

PLA boasts mechanical properties comparable to PET, including high tensile strength ranging from 50 to 100 MPa [73]. It exhibits a glass transition temperature of around 60–65 °C and a melting temperature of between 150 and 160 °C, which contribute to its versatility in processing and end-use applications. PLA is also transparent and can be processed into films, fibers, and rigid plastics, making it suitable for a variety of applications from packaging to textiles [74]. Additionally, PLA's biocompatibility makes it suitable for biomedical applications like surgical implants and drug delivery systems [75]. Unlike conventional plastics that take centuries to degrade, PLA typically biodegrades within six months to two years under industrial composting [76]. Studies indicate a reduction in PLA's composite crystallinity with extended biodegradation time, dropping from 90.59% to 86.85% over 50–70 days [77].

Despite its advantages, PLA does have limitations, notably a low glass transition temperature, inherent brittleness, and poor barrier properties [78]. Nevertheless, PLA finds extensive use in biomedical equipment, food packaging, disposable tableware, and as a material for 3D-printed scaffolds in tissue engineering [78–81]. Its applications further

extend to drug delivery systems and medical implants, and as a potential replacement for conventional plastics [82].

3.3. Starch

Following cellulose, starch is the second most abundant organic compound on Earth [83]. Produced by plants as an energy reserve, starch consists of two glucose polymers: linear amylose and highly branched amylopectin. Commercially, starch is predominantly sourced from crops such as wheat, rice, corn, potato, casava, and barley, where it constitutes 60–90% of the plants' dry weight. As a semi-crystalline polysaccharide, starch is both widely available and cost-effective [83,84]. Starch is highly regarded as one of the most promising biodegradable polymers for food packaging due to its many advantageous properties. These include biodegradability, low cost, abundance, transparency, colorlessness, flavorlessness, tastelessness, reduced water sensitivity, excellent oxygen barrier properties, renewability, edibility, and its effectiveness as a film-forming biopolymer. Despite these benefits, starch alone is not ideal for food packaging because it lacks key attributes such as a sufficient vapor barrier, mechanical strength, and thermal stability [85]. Additionally, starch tends to be brittle due to the extensive inter- and intra-molecular interactions between starch chains and has a hydrophilic nature [86].

Some limitations of using starch as a packaging material can be overcome by chemical or physical modification of the native starch. One of them is the production of thermoplastic starch (TPS). Starch is typically plasticized with additives like glycerol, glycol, sorbitol, or sugars and processed using techniques like extrusion or casting. Such a method yields a homogeneous material characterized by coexisting rigid–elastic and amorphous domains, collectively known as TPS [87]. Global TPS production is estimated to be around 800,000 tons per year [43]. TPS typically exhibits moderate mechanical properties with a tensile strength ranging from 20 to 30 MPa [88]. Biodegradation studies indicate that TPS can degrade within 6 to 12 weeks under composting conditions [70]. However, challenges such as low flexibility (2–10% elongation at break), poor barrier properties and high hydrophilicity limit its use as a standalone packaging film [83,84,88,89].

Despite its challenges, TPS has significant potential as a raw material due to its natural origin, biodegradability, and low environmental impact, making it a strong candidate for further innovation. Unlocking TPS's full potential hinges on effective modification. By blending TPS with other biodegradable polymers like PLA or PCL, or incorporating nanomaterials such as clay, cellulose nanocrystals, or graphene, researchers can greatly improve its mechanical properties, flexibility, and barrier performance. For example, adding plasticizers or cross-linking agents enhances TPS's elongation at break and reduces its hydrophilicity, making it more suitable for flexible and moisture-resistant packaging. The inclusion of cellulose nanocrystals, for instance, decreased the water vapor permeability of starch films from 7.5 ± 0.35 g $\times$ h·m·Pa^{-1} to 4.25 (1% cellulose nanocrystals) and 4.55×10^{-7} g $\times$ h·m·Pa^{-1} (2% cellulose nanocrystals) [90]. Furthermore, innovative methods such as reactive extrusion, chemical modification, and the use of biobased fillers can extend TPS's applications across various industries. These enhancements not only refine TPS's physical properties but also allow for tailored degradation rates, creating materials that degrade over specific periods according to application needs.

3.4. Chitosan

Chitosan, a biodegradable polymer derived from chitin found in crustacean exoskeletons and fungal cell walls, can be produced through chitin deacetylation using enzymatic or chemical methods [91]. As a biodegradable, biocompatible, and non-toxic material, chitosan has garnered significant attention for its potential applications in various fields, including pharmaceuticals, agriculture, and environmental management. The global production of chitosan totals approximately 6–8 million tons per year [92].

Chitosan is distinguished by notable thermal stability, with a degradation point exceeding 200 °C [93]. Its most notable attribute lies in its antimicrobial efficacy, demonstrating

wide-spectrum action against bacteria, fungi, and certain viruses [93]. The antimicrobial potency stems from its physicochemical characteristics, particularly its positive charge, which facilitates interaction with negatively charged microbial cell walls, leading to structural disruption. Chitosan can be processed into various forms, including solutions, hydrogels, nanofibers, porous scaffolds, microspheres, nanoparticles, and membranes, enabling diverse applications [94]. Presently, chitosan is widely used in the medical field for drug delivery systems, wound healing, regenerative medicine, and food preservation [94–97].

Biodegradability is another key advantage of chitosan-based materials, with studies showing that they can degrade within a few days to several weeks. For instance, chitosan-based sponge materials were observed to biodegrade within a 3-week period [98], while chitosan-based scaffolds have been shown to biodegrade within several days in vitro [99].

Despite its many advantages, raw chitosan does have certain limitations, such as poor mechanical strength, low solubility in neutral pH conditions, and limited processability. To maximize its potential, various modifications can be applied. Blending chitosan with other polymers or incorporating nanoparticles like silver or graphene can significantly improve its mechanical properties, electrical conductivity, and thermal stability. For instance, the non-covalent bond formation between chitosan and alginate makes them highly compatible for food packaging, while blends like starch–chitosan also show promise as film materials [100]. Additionally, combining chitosan with pectin results in transparent packaging materials with enhanced mechanical properties. To address the shortcomings of pure chitosan, natural plant extracts such as olive pomace, purple-fleshed sweet potato extract, apple peel, black soybean seed coat extract, and Chinese chive root extract have been incorporated, exhibiting strong antioxidant and antimicrobial activities [100–102]. Some of these extracts, like soybean seed coat and purple-fleshed sweet potato, have also been explored for their pH-sensing capabilities, leading to the development of smart food packaging materials. Moreover, chemical modifications, such as acetylation, quaternization, or grafting, can be introduced to enhance chitosan's solubility and functionality. These modifications improve its solubility in different pH environments, increase its binding capacity for various drugs, and enhance its compatibility with other materials in composite systems. Additionally, the development of chitosan-based nanocomposites and hydrogels represents a cutting-edge approach in material science. Chitosan nanocomposites can be engineered to create highly porous structures with controlled release properties, making them ideal for environmental remediation and controlled drug delivery applications. Chitosan hydrogels, with their exceptional water absorption and retention capabilities, are particularly suited for use in agricultural soil conditioners and medical dressings.

3.5. Cellulose

Cellulose, a renewable and biodegradable polymer, has garnered significant attention in the development of biodegradable packaging films sourced from natural materials such as wood, cotton, jute, kenaf, and even waste materials like old corrugated cartons and used disposable paper cups [103–105]. The production of cellulose-based polymers involves extracting cellulose from these sources and processing it into films. Cellulose can be chemically treated and dissolved in solvents such as lithium bromide molten salt hydrate or ionic liquids to form a solution, which is then cast into films [106]. Alternatively, cellulose nanofibers can be isolated from cellulose sources and used to reinforce other biopolymers, like starch, PHBV, PHB, or polyvinyl alcohol, to create composite films [85].

The global production of cellulose polymer varies across references, with the most commonly cited range being approximately 75 to 100 million tons annually [106–108]. Cellulose-based films exhibit high mechanical strength and toughness attributed to their nanostructured nature and strong intermolecular interactions [109]. Despite the excellent gas barrier properties, cellulose-based films are susceptible to moisture attacks due to their hydrophilic nature [110]. Cellulose-based polymers find applications in biomedical engineering, material sciences, electronics, and catalysis, highlighting their versatility [111]. Research is ongoing on the use of cellulose-based polymers in additive manufacturing

technology to expand their applications and address their limitations for future developments [112].

The biodegradation time of cellulose varies depending on the conditions and methods used. Studies indicate that cellulose can degrade at different rates under various circumstances. For instance, under aerobic conditions, approximately 60% of cellulose can be degraded in activated sludge within 4 to 5 weeks [113]. Under anaerobic conditions, cellulose shows promising biodegradability, with nearly 60% being biodegradable under mesophilic conditions with a solid retention time of 2 weeks [114].

The real potential of cellulose, however, lies in its capacity for modification and customization to suit specific applications. By chemically or physically altering cellulose, researchers can develop innovative materials with enhanced properties. For instance, processes such as acetylation, etherification, or esterification can make cellulose hydrophobic, improving its suitability for moisture-resistant applications like packaging and coatings. The advent of nanocellulose—derived from cellulose in the form of nanofibers or nanocrystals—has opened up new possibilities for high-performance materials [115]. Nanocellulose boasts impressive mechanical properties, with tensile strength exceeding that of steel while remaining lightweight and flexible. Its high surface area and distinctive optical properties make it ideal for advanced composites, transparent films, and even electronic devices. Further expanding cellulose's functionality, blending it with other biopolymers or incorporating nanoparticles can yield even more versatile materials. For example, combining cellulose with biodegradable polymers like PLA or PCL can produce composites that not only offer enhanced mechanical strength and thermal stability but are also biodegradable. Adding nanoparticles such as silver or graphene to cellulose matrices can introduce additional properties, such as antimicrobial activity or improved electrical conductivity, making these composites well suited for medical applications or smart packaging [116].

Additionally, cellulose can be transformed into hydrogels with exceptional water retention capabilities, making them highly useful in agricultural soil conditioning or biomedical applications like wound dressings. These hydrogels are also tunable, allowing for the creation of materials that respond to environmental stimuli, ideal for use in smart delivery systems.

3.6. Protein

Protein-based polymers are increasingly recognized as viable biodegradable materials for packaging films, which is attributed to their renewable and eco-friendly characteristics. Abundant in nature, proteins can be derived from diverse sources such as whey, gelatin, soy protein, etc. [117]. Their outstanding characteristics—such as high tensile strength, transparency, excellent gas barrier properties, strong film-forming ability, nutritional value, and elasticity—make them particularly suitable for food packaging applications [118]. Additionally, proteins exhibit excellent biocompatibility, making them particularly suitable for use in medical applications such as wound dressings, drug delivery systems, and tissue-engineering scaffolds [119].

The biodegradation rate of protein-based polymers varies depending on the specific protein type and environmental conditions. Research indicates that protein-based films often exhibit comparatively swift biodegradation rates when juxtaposed with other biobased polymers. For example, in a study conducted by Patil et al., gelatin films treated with glutaraldehyde demonstrated complete degradation within a brief timeframe ranging from 3 to 10 days [120].

The real potential of proteins lies in their ability to be modified and engineered to meet specific needs. By altering protein structures through techniques like cross-linking, denaturation, or the incorporation of functional groups, researchers can enhance their mechanical strength, thermal stability, and barrier properties. For instance, cross-linking proteins with agents such as glutaraldehyde or transglutaminase can significantly improve their durability and moisture resistance, making them more suitable for packaging appli-

cations [121]. Moreover, blending proteins with other biopolymers, like polysaccharides or synthetic polymers, can create composites with improved flexibility, strength, and resistance to environmental factors, broadening their application potential. For instance, protein–polysaccharide blends can be developed into edible films with excellent oxygen and moisture barrier properties, ideal for food packaging [122]. Advances in protein-based nanomaterials represent a cutting-edge approach in materials science. By manipulating proteins at the nanoscale, it is possible to create nanofibers, nanoparticles, and hydrogels with unique properties, such as high surface area, enhanced solubility, and controlled release capabilities [123]. These nanomaterials are especially promising in drug delivery, where they can be engineered for controlled release, or in environmental applications, such as water purification or biosensors. Chemical modifications of proteins further expand their functionality. Introducing hydrophobic or hydrophilic groups can adjust their solubility and interaction with other materials, enabling the creation of customized composite materials [124]. Additionally, enzymatic modifications can be employed for more controlled and sustainable alterations, leading to materials with properties tailored for specific applications.

3.7. Polycaprolactone

Polycaprolactone (PCL) is a biodegradable polyester characterized by a low melting point of 60 °C and a glass transition temperature of −60 °C, and it is produced through the ring-opening polymerization of ε-caprolactone. While PCL is recognized for its excellent blend-forming capability and high solubility, it does have limitations, such as its poor mechanical and thermal properties, which can restrict its use in high-temperature applications and its effectiveness as a barrier to gas and water vapor permeation [125]. Despite these challenges, PCL shows great promise for use in food packaging films due to its biodegradability and compatibility with other materials [126].

PCL's slow degradation rate allows it to be used in long-term applications, such as in drug delivery systems, where it can provide sustained release of therapeutic agents over extended periods.

The biodegradation rate of PCL varies depending on factors like the molecular weight, crystallinity, and specific environmental conditions, with the degradation times ranging from several months to years [127]. Research, such as the study by Cesur et al., has shown that PCL composite films can degrade within six months under certain conditions [128].

One of PCL's significant strengths is its ability to blend with other biodegradable polymers, such as PLA or PHA, to improve its mechanical properties, thermal stability, and degradation rates [129]. These blends can be tailored to enhance PCL's performance in specific applications. For example, increasing the PHA content in a PCL blend can boost the material strength and thermal resistance, making it suitable for more demanding applications like durable packaging or agricultural films. Conversely, a higher PCL content can improve the flexibility and elongation at break, which is advantageous for medical devices or flexible packaging solutions. The biodegradability of PCL/PHA blends is also customizable by adjusting the ratio of the two polymers. PHA's faster degradation rate can help accelerate the overall breakdown in composting or biological environments, while PCL's slower degradation ensures the material maintains its structural integrity over time. This tunable degradation is particularly beneficial in medical applications, such as drug delivery systems or tissue-engineering scaffolds, where the material's lifespan needs to align with healing processes or drug release timelines. Through these modifications and blends, the potential of PCL as a versatile biodegradable material is significantly expanded.

4. Production of PHAs from Biomass

PHAs are derived from biomass using acidogenic fermentation techniques. Microbiologically, there are two primary methods for PHA production. The first method involves using pure bacterial cultures, selecting a single strain of PHA-producing bacteria, which yields a higher PHA output but at a higher production cost. The second method uses

mixed microbial cultures (MMCs), resulting in a lower PHA yield but significantly lower production costs [130].

Furthermore, PHA can be produced via submerged and solid-state fermentation techniques. Submerged fermentation involves cultivating microorganisms in a liquid medium, whereas solid-state fermentation utilizes a solid substrate for microbial growth. These techniques have been extensively researched for PHA production, with solid-state fermentation showing promise due to its potential for more sustainable and economical PHA production [131].

4.1. Pure Cultures vs. Mixed Microbial Cultures

Production using pure cultures involves using specific bacteria like *Cupriavidus necator*, *Bacillius* sp., *Pseudomonas* sp., and others [132]. These pure cultures require aseptic conditions and high-purity carbon sources, leading to higher production costs [133]. On the other hand, mixed microbial cultures (MMCs) offer a more cost-effective solution by utilizing cheap feedstocks, such as waste glycerol, agro-industrial waste, wastewater and others, without the need for sterilization. MMCs can also utilize renewable volatile fatty acids (VFAs) from wastes and industrial effluents, making them environmentally friendly [134].

When comparing the two approaches, pure culture production is known for its efficiency in PHA production, as specific strains can be optimized for high yields [135]. However, the use of pure cultures for PHA production from complex substrates like whey can be challenging due to the limited ability of good PHA producers to grow on lactose [136]. In contrast, MMCs can adapt to a variety of waste streams as carbon sources, making them versatile and suitable for sustainable production [137]. Additionally, MMCs can be enriched through selective pressures like feast and famine regimes to enhance the culture with PHA-accumulating organisms [138].

4.2. Submerged Fermentation vs. Solid-State Fermentation

On the technological side, there are two main methods for PHA production: submerged and solid-state fermentation. In submerged fermentation, the production of PHAs occurs in batch reactors when microbes are cultured with nutrient-limiting concentrations of nitrogen, phosphorus, sulfur, or oxygen, and excess carbon sources [139]. This technique typically involves a three-stage process, as shown in Figure 3.

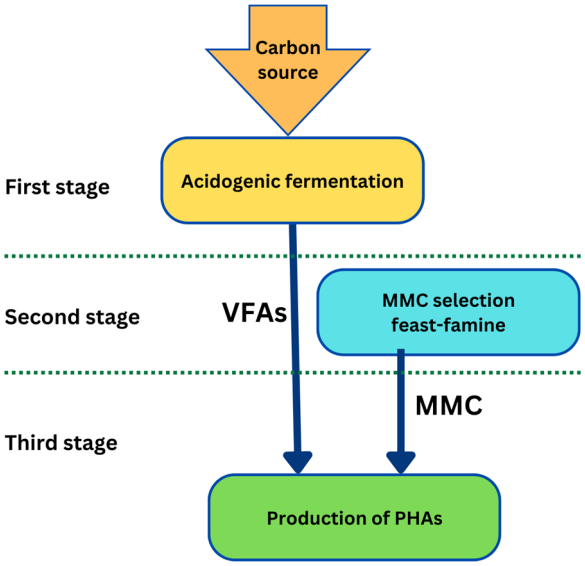

Figure 3. Schematic of the PHA production process.

First, acidogenic fermentation takes place, where organic matter is converted into VFAs, which serve as precursors for PHA production. The acidogenic fermentation process

can be carried out by MMCs, which consist of a diverse range of microorganisms that work together to convert the organic carbon into VFAs [138]. The production of VFAs through acidogenic fermentation is a crucial step in the PHA production process as it provides the necessary substrates for PHA biosynthesis [140]. The common VFAs mainly comprise acetic acid, propionic acid, butyric acid, lactic acid and valeric acid [141].

The second stage of PHA production is culture selection, which involves enriching the MMC with PHA-producing microorganisms [140]. This is achieved through a feast–famine regime where the culture is subjected to alternating periods of nutrient abundance (feast) and scarcity (famine). During the feast phase, PHA-accumulating bacteria store carbon and energy in the form of PHA, while non-PHA accumulators do not. This selective pressure allows for the enrichment of PHA-producing microorganisms within the culture. The goal of culture selection is to obtain a microbial community that is highly efficient in PHA production [47].

The third stage of PHA production is PHA production itself, where the selected PHA-accumulating MMC from the culture selection stage are fed with VFA-rich substrates. The PHA-accumulating microorganisms utilize the VFAs as carbon sources and convert them into PHAs, thus accumulating the biomass that yields PHAs. The PHAs can then be extracted and purified [47].

On the other hand, solid-state fermentation (SSF) is a promising alternative technique for producing PHAs due to its economic and sustainablity advantages [47]. SSF process uses significantly smaller quantities of water and other resources, thus making the production process more economical. Producing PHA via SSF involves the use of lignocellulosic-based residues as low-cost raw materials for PHA production. These residues, such as brewer's spent grain, agricultural waste and even household waste, can be utilized as a source of fermentable sugars for microbial fermentation [47]. SSF offers several benefits, including the utilization of abundant and low-cost raw materials and high productivity.

In SSF, microorganisms are grown on solid substrates with a small amount of water under controlled conditions. The solid substrate provides a suitable environment for microbial growth and PHA production. During fermentation, microorganisms metabolize the substrate and produce PHAs as intracellular carbon and energy reserves [46]. After fermentation, the cells containing PHAs can be separated from the culture broth using conventional methods such as centrifugation, filtration, or flocculation–centrifugation [49]. SSF offers advantages such as lower water and energy requirements, utilization of low-cost raw materials, and potential for valorization of waste streams; however, it presents challenges in terms of process control and scalability.

4.3. Carbon Sources for PHA Production

Regarding the carbon sources for the production of PHAs, several different materials were explored. Plant and animal oils have been demonstrated to be excellent carbon sources for high-yield production of PHAs [142]. The conventional carbon sources used include carbohydrates, fatty acids, sugars, and alkanes, but these can impose high production costs [143]. Attempts have been made to find sustainable and affordable sources of carbon to reduce production costs, such as lignocellulosic biomass and industrial wastes [143–145]. Other sources mentioned include wastewater [139], municipal solid waste [146] and even waste polystyrene fragments [147]. The choice of carbon source can impact the yield and properties of the PHAs produced. For example, vegetable oils have shown higher PHA yields compared to glucose as a sole carbon source [148]. In Table 4, there are examples of the reported PHA yields using different carbon sources.

Table 4. Examples of the reported PHA yields from different carbon sources.

Carbon Source	PHA Yield	Culture Type	Reference
Olive oil distillate	0.9 g PHA/g	*Cupriavidus necator*	[142]
Starchy waste stream	5.12 g PHA/L	*Cupriavidus necator* DSM 545	[149]
Cheese whey waste stream	0.12–0.20 g PHA/L	MMC	[150]
Cheese whey	0.35–0.52 g PHA/L	*Enterobacter cloacae*, *Raoultella ornithinolytica*, *Citrobacter freundii*, *Escherichia coli*, *Vibrio parahaemoliticus*, *Leuconostoc* spp.	[136]
Waste frying oil	0.19–0.34 g PHA/g	*R. eutropha*	[151]
Waste sludge and synthetic wastewater	0.648 kg/m^3	MMC	[152]

* MMC—mixed microbial cultures.

4.4. Extraction and Purification Methods

Extraction of PHAs from biomass after production is a crucial step in the process of obtaining these biodegradable polymers. Traditionally, chloroform has been a standard method for PHA extraction due to its effectiveness in solubilizing PHAs from biomass. However, alternative methods are being developed to address concerns regarding the toxicity and environmental impact of chloroform [153]. One such alternative method involves the use of sodium hydroxide and sodium hypochlorite for PHA extraction. This method offers the advantage of higher average molecular weights of extracted PHAs compared to other extraction methods, which can influence the properties of the polymer [154].

Another approach to PHA extraction involves the use of acids. Acid-based extraction methods have been explored for their ability to efficiently recover PHA from biomass. However, the use of acids can sometimes lead to degradation of the PHA polymer, affecting its quality and properties [154]. On the other hand, extraction using supercritical CO_2 has emerged as a novel method for PHA extraction. Supercritical CO_2 offers the advantage of being a green and environmentally friendly solvent, making it a promising alternative to traditional extraction methods. Studies have shown that supercritical CO_2 can effectively extract PHAs from biomass while maintaining the integrity of the polymer [155].

4.5. Identification and Analysis of PHAs

Following the extraction and purification of PHAs, various methods are utilized for their identification. Fourier-transform infrared spectroscopy (FTIR) is commonly employed for this purpose [156]. Additionally, differential scanning calorimetry (DSC), thermogravimetric analysis (TGA), and nuclear magnetic resonance (NMR) are used to characterize PHA [157].

DSC investigates the melting behavior, glass transition temperature, and crystallinity of PHAs, providing insights into their thermal stability and phase transitions. TGA analyzes the weight loss and thermal stability [158]. NMR studies the chemical makeup of intact PHA polymers, revealing the molecular structure, composition, and monomer units [159].

Scanning electron microscopy (SEM) is also valuable for identifying and characterizing PHAs post-extraction and purification. SEM allows observation of the PHAs' microstructure and surface morphology, providing insights into their physical characteristics at a microscale level [160]. SEM complements other analytical methods like FTIR, DSC, TGA, and NMR, offering a comprehensive understanding of PHAs' structural and surface properties.

5. Active Packaging Based on PHAs

Active packaging, as defined by the EU, involves integrating active components into packaging materials to preserve and extend the quality and shelf life of products. Innovative approaches aim to enhance food safety, reduce food waste, and improve sustainability

in the food industry. Active packaging utilizes materials that release active substances, such as antimicrobial nanoparticles or antioxidant agents, into the packaged food to inhibit microbial growth, delay oxidation, and maintain freshness [161]. By incorporating active elements, active packaging can significantly reduce the risk of foodborne illnesses, enhance product quality, and minimize food losses. Overall, active packaging represents a promising solution to prolonging food's shelf life, improving food safety, and advancing sustainable practices in the food industry.

5.1. Biodegradable PHA-Based Blends

Blending PLA with PHAs is a strategy used to enhance PLA's poor barrier properties, although the two polymers exhibit only partial miscibility. Kanda et al. demonstrated that a blend containing 10 wt.% PHBV with PLA exhibited significantly improved tensile strength and Young's modulus compared to neat polymers. However, other blend ratios resulted in inferior mechanical properties due to the low miscibility [162]. Compatibilization is essential for creating PLA/PHA polymer blends [163].

In a study by Ma et al. [164], the addition of the plasticizers mono-caprylin glycerate (GMC) and glycerol monolaureate (GML) improved the miscibility of PLA/PHB blends, enhancing the tensile strength and water vapor transmission rate (WVTR) with a minor negative impact on the oxygen barrier properties when adding 0.5 wt.% GMC and GML. Another study utilized small amounts (0.1–1.0 wt.%) of oligomer-like PLA (oLA) as a compatibilizer for PLA/PHBV blends, resulting in overall improvements in the mechanical and barrier properties, particularly with the addition of 1 wt.% oLA to the PLA/PHBV 90/10 blend [78].

Corn starch was blended with PLA in a study by Mangaraj et al. [165] to develop a biodegradable food packaging film. The authors blended 10–20 wt.% of corn starch with PLA in the presence of an organic peroxide and glycidyl methacrylate as a compatibilizer and created packaging films using the extrusion blown-molding method. Composite films were used to test the shelf life of capsicum, and their efficacy was compared to LDPE films. The study concluded that the developed PLA-corn starch films performed on a par with the LDPE films, having extended the shelf life of capsicum by 8 days and 15 days at 25 and 8 °C, respectively [165].

Magalhães et al. [166] investigated the mechanical and biodegradation properties of 1:1 blends of TPS with PHBV. TPS was plasticized with glycerol (25 wt.%) and melt-blended with PHBV in the presence of organically modified montmorillonite (2.5–10%) as a compatibilizer. The results showed that the incorporation of hydrophobic PHBV and organoclay was able to significantly reduce the humidity adsorption of the blends. Additionally, higher concentrations of the organoclay improved the tensile strength and Young's modulus but slightly decreased the flexibility of the blend. Similar results were obtained in the research of Garrido-Miranda et al. [167], who produced PHB/TPS blends with organoclay as a compatibilizer, confirming its compatibilization efficiency. However, further research is warranted on this topic, as there are currently no recent articles published that investigate PHA/TPS blends for food packaging films. Chitosan is incompatible with most biobased thermoplastic polymers; therefore, compatibilization is also a necessary step. Vernandez et al. [168] grafted maleic anhydride (MA) and glycidyl methacrylate (GMA) to PHBV and PLA in order to improve the compatibility with chitosan. The authors used one-step reactive compatibilization using organic peroxide as an initiator. Subsequent analysis confirmed the improved compatibility of the PHBV/PLA polymer blends.

Zhang et al. [169] used cellulose nanocrystals and cellulose nanofibers to improve the mechanical and barrier properties of PHB film using the solvent-casting method. The study found that the improvements to the mechanical properties as well as the barrier properties were the highest with the addition of 1 wt.% of nanocellulose. Similarly, another study by Popa et al. [170] produced a material using PHB and microfibrillated cellulose grafted with methacrylic acid. The authors produced materials with more balanced stiffness–toughness properties compared to pure PHB.

Protein-based films have attracted research interest for their excellent film-forming ability and strong barriers against oxygen and aromas [117]. However, due to their high moisture absorption, the films have limited water vapor barrier properties. Researchers have worked on enhancing the barrier and mechanical properties of these films for broader applications in active and smart packaging [117].

Blending PHBV with PCL offers a promising route toward enhancing the flexibility and toughness of the resulting material, which are crucial properties for packaging films. However, the inherent immiscibility between PHBV and PCL underscores the pivotal role of a compatibilizer in ensuring the uniform dispersion and compatibility of the blend components. Compatibilization can be achieved using a crosslinking agent with peroxide to create a PHBV–PCL copolymer, improving the blend compatibility and mechanical properties [171]. A similar approach can be applied to PHB–PCL blends. Further investigation is required to explore non-reactive compatibilization strategies; however, the existing literature suggests that PCL or PHBV grafted with maleic anhydride could serve as a suitable non-reactive compatibilizer [172].

In their study, Zhao et al. [173] explored the incorporation of natural rubber into PHBV to enhance its flexibility and toughness, facilitated by the use of organic peroxide and a co-agent. The resulting materials, containing 15 wt.% of NR, exhibited mechanical properties comparable to polypropylene (PP) while offering an optimal balance of toughness and strength. However, it is noteworthy that the inclusion of NR led to a deterioration in the barrier properties of neat PHBV. Rodriguez-Uribe et al. [174] investigated the use of biobased polybutylene succinate (BioPBS) and PHBV composites reinforced with talc and starch as potential alternatives to single-use plastic packaging. To improve the compatibility, the authors synthesized compatibilizers by grafting maleic acid (MA) to BioPBS and BioPBS/PHBV (80–20 wt.%), thus creating MA-g-BioPBS and MA-g-BioPBS/PHBV compatibilizers. The barrier properties of BioPBS were greatly improved by blending with PHBV and talc. Maleic anhydride-based compatibilizers improved the oxygen barrier but slightly reduced the water vapor barrier because of the hydrophilicity of MA groups. The use of starch further decreased the barrier properties of the composite, although it improved the flexibility of the material.

5.2. Antimicrobial Active Packaging PHA-Based Materials

Antimicrobial properties are crucial for food packaging films due to their ability to inhibit the growth of bacteria and fungi, thereby extending the shelf life of food products and preventing foodborne illnesses. Prominent properties are especially important in the context of food packaging, as they help to maintain the quality and safety of the packaged food [175]. Antimicrobial active agents are commonly added to polymer blends used in food packaging films to enhance their antimicrobial properties. Some commonly used antimicrobial active agents include essential oils, chitosan, and silver nanoparticles [175].

As discussed before, chitosan is used as an additive in polymer blends to improve the antimicrobial properties of the composite. One study by Cavalli et al. [176] focused on the development of biodegradable active packaging with PLA, polyethylene-co-vinyl acetate (PEVA), polyethylene glycol (PEG) and chitosan blends. The antimicrobial efficacy of the blends was assessed using bread slices, wherein the chitosan-containing blends exhibited a reduction of approximately 35% in the count of molds and yeasts present in the bread slices. This finding underscores the effectiveness of chitosan as a natural antifungal agent. Chitosan is also researched as a component of edible films and coatings, as discussed in a recent review article by Muñoz-Tebar et al. [91]. Although various studies have investigated PHBV–chitosan composites, primarily for biomedical applications, there remains a notable gap in the research concerning their potential application in food packaging films. Further research is needed to explore and optimize the suitability of these composites for such applications.

Essential oils (EOs) are highly concentrated plant extracts that are known for their aromatic properties and various therapeutic benefits. They are derived from different parts

of plants, including leaves, flowers, stems, and roots, through processes such as steam distillation or cold pressing. Essential oils are composed of volatile compounds, primarily terpenes, which give them their characteristic aroma and flavor [177]. There are numerous types of essential oils, each with its own unique composition and properties. Some of the most common types include lavender, peppermint, lemon, eucalyptus, tea tree, and rosemary oil. Featured oils have been widely studied for their antimicrobial properties and are commonly used in food packaging to improve the shelf life and safety of food products [178]. The antimicrobial properties of essential oils make them effective additives in food packaging films. Essential oils contain bioactive compounds that have been shown to inhibit the growth of various microorganisms, including bacteria, fungi, and molds. Bioactive compounds can disrupt the cell membranes of microorganisms, interfere with their metabolic processes, and inhibit their growth and reproduction [178]. Incorporating essential oils into packaging films offers a means to establish a protective barrier against microbial contamination. The essential oils can migrate from the film into the food product, creating a zone of inhibition that prevents the growth of spoilage and pathogenic microorganisms. This can help extend the shelf life of perishable foods and reduce the risk of foodborne illnesses [150]. In addition to their antimicrobial properties, essential oils also possess antioxidant activity. They can scavenge free radicals and inhibit oxidative reactions that can lead to food spoilage and degradation. By incorporating essential oils into packaging films, the oxidative deterioration of food products can be minimized, preserving their quality and sensory attributes [178]. The incorporation of essential oils into various biodegradable polymers has also been explored for applications in food preservation. Figueroa-Lopez et al. [179] investigated the effect of the incorporation of oregano essential oil (OEO), rosemary extract, and green tea extract into PHBV films. They confirmed the antimicrobial activity of EO-infused PHBV films against *S. aureus* and *E. coli*, as well as the antioxidant activity of the composite. Similarly, Melendez-Rodriguez et al. [179] developed eugenol-containing PHBV film with long-term antimicrobial activity by encapsulating eugenol essential oil in the pores of mesoporous silica nanoparticles. In another study, Da Costa et al. [180] melt-blended different clays with PHBV and OEO. The resulting composites containing OEO showed an inhibitory effect on selected Gram-positive and Gram-negative bacteria. Additionally, essential oils have been evaluated as antimicrobial agents in the coating of Edam cheese, where they have been combined with lactic acid to enhance their antimicrobial properties [181]. Furthermore, thymol, when incorporated into starch/cellulose nanofiber films, has been shown to improve antimicrobial effectiveness while maintaining essential physical and mechanical properties. Films loaded with thymol exhibit strong antibacterial effects, rapidly eliminating pathogens like *Escherichia coli*, which is crucial for preventing food spoilage and contamination. Similarly, gelatin-based films infused with nisin, a natural antimicrobial peptide, and ethylenediaminetetraacetic acid (EDTA) have demonstrated effective inhibition of *Escherichia coli* growth, highlighting the potential of biodegradable gelatin films as active packaging solutions [181–183].

The integration of metallic nanoparticles, such as silver, SiO_2, and ZnO, into packaging materials is a promising approach for food preservation [184]. Silver nanoparticles, synthesized using plant extracts, exhibit antioxidant properties, while ZnO nanoparticles in packaging films demonstrate antibacterial activity against pathogens like *Bacillus subtilis* and *Escherichia coli*, extending food's shelf life [185]. SiO_2 nanoparticles, combined with ZnO, further enhance the antimicrobial properties and improve the mechanical and barrier properties of packaging [186]. Research has shown that incorporating SiO_2 into PHBV creates a biodegradable nanocomposite with enhanced thermal, mechanical, and antibacterial properties, making it suitable for food packaging [187]. Silver nanoparticles on PHBV films have also shown increased antimicrobial effectiveness, particularly in apple juice samples [184]. Additionally, the incorporation of Ag-ZnO nanoparticles into PHBV has demonstrated potent antimicrobial activity against *S. aureus* and *E. coli*, with improvements in the mechanical and barrier properties observed at a 3% nanoparticle concentration [185].

5.3. Multilayer PHA-Based Packaging

Multilayer packaging materials offer a versatile solution by combining various biobased polymers to achieve enhanced functionality and performance in terms of packaging applications. Incorporating multiple layers with specific properties enables multilayer packaging to deliver improved barrier properties, mechanical strength, and tailored functionalities to meet diverse product requirements (Figure 4) [188].

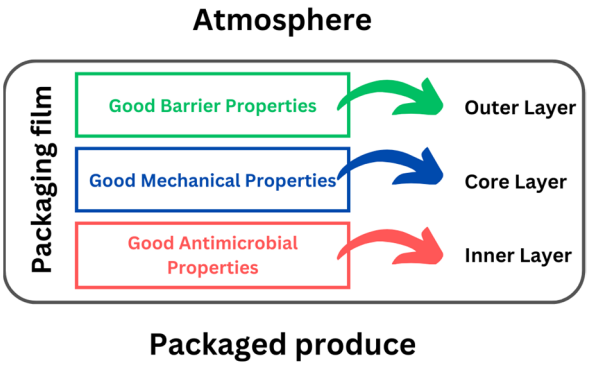

Figure 4. Multiple functionalities in a multilayer films.

Arieta et al. [61] developed a biodegradable bilayer with antioxidant properties using PHBV, PHB and PLA. The outer layer was composed of PHBV and the inner active layer was formed from PLA with 25 wt.% of PHB. The inner layer was loaded with 1 wt.% to 3 wt.% of catechin, which is a natural antioxidant. The study found that this biocomposite was effective as an antioxidant for food preservation, but the mechanical properties were not significantly affected.

Figueroa-Lopez et al. [32] developed an antimicrobial multilayer packaging film composed of PHB, PHBV and eugenol as an antimicrobial active agent. The outer layers were formed from PHB and PHBV (on the food contact side), while the inner layer was a PHBV with 15 wt.% of eugenol. The resultant multilayer film had improved water vapor and aroma barrier properties, high hydrophobicity and showed high inhibition against *S. aureus* and *E. coli*.

Dilkes-Hoffman et al. [189] developed a multilayer film composed of TPS with a PHBV coating to improve TPS's barrier properties. The PHBV-coated TPS films significantly reduced the water uptake into the TPS films, thus improving the barrier properties of the materials. Similar results were previously obtained by the lamination of PLA with sugar palm starch [190] and PCL with TPS [191].

PHA coatings were also utilized to improve the moisture resistance of cellulose-based films and papers. Cherpinski et al. [192] created a multilayer composite using double-sided coatings of PHB and PHBV to cellulose-based films. The multilayer composite significantly improved the water resistance and water vapor barrier performance of the film. Mendelez-Rodriguez et al. [193] developed a multilayer film composed of PHBV and PHB outer layers and cellulose nanocrystals (CNCs) as the inner layer. The resulting multilayer films had increased flexibility due to the incorporation of CNCs and excellent oxygen and water vapor barrier properties. Furthermore, Hernandez-Garcia et al. [194] coated PHBV on paper to develop a multilayer film for packaging applications. Their findings demonstrated that applying dual layers of PHBV effectively enhanced the paper's mechanical strength and flexibility while offering protection against moisture and simultaneously lowering the aroma and oxygen permeation. These improvements had only a minimal impact on the paper's optical and thermal characteristics, making PHBV paper a promising option.

6. Life Cycle Assessment of the Biobased Polymers in Food Packaging

Packaging plays a crucial role in advancing sustainable food supply systems, but it also poses several environmental challenges, particularly when it comes to plastic packag-

ing. In 2019, approximately 40% of plastics in the EU-28 (plus Norway and Switzerland) were used for packaging, with a significant portion designated for food [195]. Despite their advantages—light weight and high performance—plastics used in food packaging have a very short lifespan, leading to a considerable amount of post-use waste. This situation exacerbates issues related to fossil fuel consumption, waste management, and the persistence of non-degradable plastic in the environment. In 2015, global greenhouse gas emissions from conventional plastics amounted to 1.7 gigatons of CO_2-equivalent. If current trends continue, this figure could rise to 6.5 gigatons by 2050. Waste management practices in 2018 showed that in the EU-28 (+Norway and Switzerland), 42% of post-consumer plastic packaging waste was recycled, nearly 40% was incinerated with energy recovery, and the remainder was landfilled [196]. These concerns have prompted a search for alternative materials, with biodegradable biobased plastics emerging as a promising solution. These alternatives could reduce reliance on fossil fuels and help mitigate climate change. However, their effectiveness is heavily influenced by the end-of-life (EoL) management practices, which vary by region and technological advancements, and they also place pressure on land use due to their reliance on food resources.

Biodegradable plastics offer a potential solution to plastic litter, although their effectiveness can vary based on the material properties and environmental conditions. This aligns with EU policies aiming for more sustainable packaging solutions. To determine if these new materials are indeed more environmentally sustainable, a comprehensive life cycle assessment (LCA) is necessary. A LCA helps compare the environmental impacts of different polymers and assess the benefits of bioplastics. Generally, bioplastics show lower impacts on climate change and fossil fuel dependence but may have higher impacts in areas like eutrophication and toxicity. A LCA faces challenges, such as incorporating indirect impacts like food loss and weight (FLW) and assessing long-term effects of mismanaged plastic waste. Recent studies have introduced new indicators and models to address these issues, but challenges remain. For instance, research by Boone et al. (2023) compared the environmental impacts of organic falafel packaged in fossil-based plastic versus a biobased, biodegradable plastic (PHBV) [197]. The study found that conventional plastic had a lower overall environmental impact, largely due to the higher mass and impact of the PHBV packaging. Future advancements in PHBV production technology and better recycling methods could improve its environmental footprint. The study also highlighted that conventional packaging had advantages in terms of transportation and utility consumption due to differences in the mass and volume. While no difference in shelf life was observed between the two packaging types for falafel, other factors, like protection during transport, remain to be explored. Moreover, evaluating plastic packaging's impact on marine ecosystems through indicators like "lifetime costs on marine ecosystem services" suggests that while PHBV has a higher potential for marine litter, its lower persistence reduces the overall impact on marine ecosystems.

To enhance the environmental benefits of bioplastics and reduce greenhouse gas emissions, further research is needed on LCA's environmental, economic, and social impacts. Investigating the biodegradation of these materials in various environments as an EoL option and improving the accessibility of bioplastic disposal methods are crucial steps. Policymakers should focus on these areas to reduce plastic waste mismanagement and promote more sustainable packaging solutions.

7. Conclusions

The development and introduction of biobased and biodegradable food packaging films offers a promising avenue for sustainable packaging solutions that address the pressing environmental issues associated with traditional fossil fuel-derived materials. Biobased and biodegradable food packaging films derived from renewable sources such as plant-based materials and agricultural waste offer an innovative approach at the interface between sustainability and packaging technology. Efforts to reduce the environmental impact of food waste, which is steadily increasing worldwide, underline the importance of

environmentally friendly packaging solutions. The valorization of agro-industrial waste and the exploration of different conversion methods into value-added products help to reduce waste and promote the principles of the circular economy. However, the transition to biodegradable polymers for food packaging poses some challenges, including limitations in processability, mechanical strength and cost-efficiency compared to conventional plastics. Optimizing the material properties and overcoming the production capacity constraints are critical to improving the viability and competitiveness of biodegradable packaging materials. Key properties such as the tensile strength, toughness, thermal stability and barrier properties are critical to the effectiveness of food packaging films in maintaining food quality and safety. In addition, the addition of antimicrobial additives and the provision of the necessary labeling information improve the functionality and consumer acceptance of these packaging materials. Biobased polymers, particularly PHAs, are a promising alternative to petroleum-based plastics due to their biodegradability and versatility. Recent advancements have expanded their use into food packaging, aquaculture, health, and biofuels, showcasing their diverse applications. To harness their full potential and advance sustainable packaging, continuous innovation and cross-industry collaboration are crucial. Using agricultural waste, organic by-products, or renewable feedstocks for PHA production offers a cost-effective, eco-friendly solution, reducing reliance on fossil fuels and lowering carbon emissions. Microbial fermentation with specialized bacteria or engineered microorganisms is an efficient, scalable method that produces high-quality PHA with minimal environmental impact, supporting large-scale, sustainable production.

In conclusion, while the existing body of research on PHA-based packaging has made significant strides in areas such as production techniques, material blending, and short-term functionality, there are critical gaps that need to be addressed to fully realize the potential of PHAs as a sustainable alternative in food packaging. Specifically, there is a pressing need for more comprehensive LCAs that encompass the entire environmental footprint of PHA packaging, from production through to disposal. Additionally, further investigation is required to explore the scalability of PHA production, particularly in terms of cost-effectiveness and the sourcing of sustainable carbon materials. Moreover, long-term studies are essential to assess the real-world performance of PHA-based packaging across various applications and conditions, and to compare it with other biodegradable materials. By addressing these gaps, future research can contribute to a more complete and practical understanding of the role of PHAs in advancing sustainable food packaging solutions.

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