# Can Sustainable Packaging Help to Reduce Food Waste? A Status Quo Focusing Plant-Derived Polymers and Additives

Imke Korte [1], Judith Kreyenschmidt [1,2,*], Joana Wensing [3], Stefanie Bröring [4], Jan Niklas Frase [5], Ralf Pude [5], Christopher Konow [6], Thomas Havelt [7], Jessica Rumpf [7], Michaela Schmitz [5,7,*] and Margit Schulze [7,*]

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Featured Application: Exploitation of renewable resources and plant extracts for the development of sustainable materials used for active food packaging.

Abstract: The promotion of sustainable packaging is part of the European Green Deal and plays a key role in the EU's social and political strategy. One option is the use of renewable resources and biomass waste as raw materials for polymer production. Lignocellulose biomass from annual and perennial industrial crops and agricultural residues are a major source of polysaccharides, proteins, and lignin and can also be used to obtain plant-based extracts and essential oils. Therefore, these biomasses are considered as potential substitute for fossil-based resources. Here, the status quo of bio-based polymers is discussed and evaluated in terms of properties related to packaging applications such as gas and water vapor permeability as well as mechanical properties. So far, their practical use is still restricted due to lower performance in fundamental packaging functions that directly influence food quality and safety, the length of shelf life, and thus the amount of food waste. Besides bio-based polymers, this review focuses on plant extracts as active packaging agents. Incorporating extracts of herbs, flowers, trees, and their fruits is inevitable to achieve desired material properties that are capable to prolong the food shelf life. Finally, the adoption potential of packaging based on polymers from renewable resources is discussed from a bioeconomy perspective.

Keywords: active packaging; bio-based polymers; bioeconomy; essential oil; food waste; natural additives; permeability; plant extracts; shelf life; sustainable packaging

1. Introduction

The main functions of food packaging are protection/preservation, containment, communication/marketing, and convenience. Thereby, food safety and quality related properties as well as reducing food waste are targeted [1–3]. With an appropriate packaging solution, a high quality, safe (extended) shelf life along the entire supply chain can be

ensured. Thus, possible waste of the food product prior to final consumption can be prevented [1,3–6].

There are different packaging strategies to achieve a longer shelf life. One of the main strategies is the application of materials with certain barrier functions like gas and water vapor permeability (WVP) guaranteed by various fossil-based materials [7,8] (for definition of terms such as fossil-based, bio-based, etc., see glossary at the end of the main body text). Many of these fossil-based foils are complex multi-layer materials that represent a large group of individual fossil-based polymers with different chemical and technical characteristics due to diverse requirements regarding food safety and waste reduction [8–10]. Another strategy is the use of active packaging [2,11]. Based on the European Union (EU) Guidance to the Commission Regulation No 450/2009, active materials are defined as: "[. . .] materials [. . .] that are intended to extend the shelf life or to maintain or improve the condition of packaged food; they are designed to deliberately incorporated components that would release or absorb substances into or from the packaged food or the environment surrounding the food." [12].

Today, around 40% of all plastics circulating are applied for packaging [13], of which approximately 60% are used for food and beverages while the rest covers non-food applications [14]. In Germany, the packaging consumption of glass reached 35.0 kg/head, for paper 98.5 kg/head and for plastic 39.0 kg/head in 2018 [15]. Plastic waste represents one of the most complex material mixtures from a recycling perspective [16]. Moreover, there are increasing issues concerning the harm caused to the environment mainly due to the manufacturing phase (i.e., oil refinery and material production), problematic end-of-life strategies, and adverse effects on human health [5,14]. Despite the negative environmental and health-related effects, fossil-based plastics are favored because of their lightweight nature and low costs [8,9]. As a result, the use of plastic packaging is growing [17] caused by the need to reduce food waste due to the steadily growing population and market expansion [18]. To deal with the issue of adverse impact of fossil-based plastics on human health and environment, the design and production of plastics and plastic products must take account of the end-of-use strategy such as reuse, repair, and recycling needs [14]. This leads to a paradigm shift from linear to circular economy. The core principles "take, make, dispose" of a linear economy are replaced by "take, make, reuse" in a circular economy. "Reuse" involves circular criteria like repair, refurbish, and recycle, as recently reported by Taleb et al. [19].

Within the last decade, the development and promotion of more sustainable materials became key roles on social and political levels in the EU [14]. Pursuing these strategies, the European Commission adopted a Circular Economy Action Plan in 2015 [20]. This action plan lays the foundation for a new plastics economy addressing the environmental issues concerning plastics and forcing the EU to move towards a more sustainable model for economic development [21]. The EU launched and developed the action plan in 2018 with the "European Strategy for Plastics in a Circular Economy", the so called plastics strategy [14]. The plastics strategy forces the industry to rethink plastics design and their usage, disposal, and recycling within the entire value chain. The main goal is to achieve improvements in sustainability [8].

To reduce environmental impacts, one key requirement is the complete reusability and/or recyclability of all plastic packaging placed on the EU market by 2030 [14]. Moreover, Matthews et al. stated that innovations in food packaging have to focus on maintaining food safety and shelf life and reducing food waste [8].

Besides current political requirements, sustainable packaging is also an important aspect for consumers [14,22]. As consumer preferences shifted to high quality and safe products with enhanced shelf life, the development of various new trends in packaging systems has arisen [1]. There are different strategies for the development of sustainable packaging such as reducing packaging materials by decreasing the thickness and/or number of layers, applying bio-based and/or biodegradable materials, reducing the amount of layers, and using easy recyclable materials [22]. For example, material properties of simple

mono-layer materials can be improved by design of packaging itself such as introducing packaging conditions through a modified atmosphere or by using active packaging solutions [23]. Recently, Pauer et al. reported that the environmental benefit of weight reduction is greater than the benefit from improved recyclability in terms of meat packaging [24].

Therefore, bio-based and biodegradable polymers represent a growing field in creating environmentally friendly materials [25]. The continued use of agricultural and industrial by-products and waste flows such as corn stover, wheat straw, and whey constitutes from dairy and cheese industries as raw materials would provide a significant ecological advantage and would reduce pressure on land use [26–28]. Plant-based (waste) materials such as wood and lignocellulosic residues from agriculture and forestry are a major source of polysaccharides; therefore, they are considered as sustainable alternatives. They have the potential to be used instead of fossil resources [29,30]. Concerns about greenhouse gas (GHG) emissions and the security of industrial feedstock supplies promote substituting conventional fossil-based feedstock in the production of synthetic materials with biomass [31]. Several studies considering cradle-to-grave life cycle analysis (LCA) of various bio- and fossil-based plastics showed that the production and use of plastics produced from renewable resources is generally advantageous in terms of saving fossil resources and reducing GHG emissions [28,32,33]. Biodegradable or compostable plastics can reduce the amount of waste sent to landfills [1,8]. Natural bio-based polymers such as agar, chitosan, cellulose, and starch represent the group of bio-based polymers in food packaging applications [34].

Although significant effort is currently being made to develop novel, sustainable materials, there are currently no competitive alternatives which offer the same level of protection to fossil-based multi-layer plastic packaging, especially for fresh products like meat [8]. Although research focuses on improving the bio-based film characteristics of packaging materials, their mechanical, thermal, and physical properties are still non-satisfactory, and their use in industrial applications is often restricted [2]. To counteract these disadvantages by replacing fossil-based plastic packaging, research, and developments include active packaging based on bioactive polymers and composites obtained from renewable resources [1,35]. There are several developments in this field, but up to now, a widespread use in the market is still missing. Today, a limited number of bio-based plastics with food packaging applications are commercially available mainly based on the following polymers: poly(lactic acid) (PLA), poly(hydroxyalkanoates) (PHA), pol(yethylene furanoate) (PEF), poly(butylene succinate) (PBS), thermoplastic cellulose derivatives, and starch-based films [1].

This review focuses on most recent developments of packaging materials (polymers and composites) that are produced from renewable resources and considered as promising alternatives for fossil-based plastics. Annual and perennial crops, herbs, flowers, lignocellulosic and agricultural residues are a major source of polysaccharides, proteins, and lignin. In addition, they can also be used to obtain plant extracts and essential oils (EO). Therefore, we consider their application in terms of sustainable packaging that contribute to the reduction of food waste. First, challenges along food supply chains related to packaging characteristics are discussed: advantages and disadvantages related to the characteristics of the most prominent fossil-based plastics packaging are compared. They are evaluated regarding food waste, recyclability, and sustainability issues (Section 2). Second, characteristics of selected packaging plastics (mainly plant-derived polymers) in terms of mechanical, thermal, and physical properties are highlighted. Biodegradability will not be addressed in detail but mainly properties, such as gas permeability with influence on the food quality, safety, and shelf life (Section 3). Third, natural additives with focus on plant extracts in the context of active food packaging are presented (Section 4). Incorporating extracts of herbs, flowers, trees, and their fruits is inevitable to achieve desired material properties that are capable to prolong shelf life, resulting in reduced food waste. Finally, in Section 5, the potential of renewable resources is evaluated from a bioeconomy perspective of a packaging core matrix as well as a source for natural additives for active packaging.

2. Challenges in Food Supply Chains Related to Packaging Characteristics

2.1. Current Packaging Characteristics

While bio-based systems are the focus of the review, we begin by discussing general issues of packaging and the reasons behind the current ubiquitousness of fossil-based food packaging. Intensive research and development efforts over a long period of time resulted in packaging solutions which are optimized with regard to various important parameters that guarantee their functionality including material weight per unit of packed volume [8,36], mechanical characteristics (impact and tear strength, stiffness, flexibility), durability, and many others [2,5,9,17,37,38]. Moreover, fossil-based plastics are less expensive per weight unit compared to most of the bio-based materials. Therefore, currently, alternative bio-based materials are more expensive because of poor commercial availability and lack of efficient production processes. In the future, the prices for fossil resources will rise due to the limited availability, and on the other hand, costs for bio-based plastics may drop due to improvements in production process efficiency [39]. The current focus of research activities is the reduction of packaging waste especially fossil-based material [5]. LCA showed that a reduction in environmental impact of the packaging itself can be best achieved by minimization of used materials (thinner layers) that retain mechanical and barrier properties rather than emphasizing end-of-life issues (such as recycling or disposal) [7].

Today, multi-layer materials are widely used for food packaging throughout the food industry. As multi-layers of plastic can be easily adjusted to the various requirements of different food types, this kind of packaging can offer effective solutions to maintain food safety and quality, achieve optimal shelf life, and minimize food waste [8]. Although from an environmental, sustainability, and biocompatibility perspective, the use of multi-layered packaging materials has to be reduced, their global use for food applications is growing. This development is based on the mechanical and barrier properties of multi-layer materials (higher resistance to water, gas, and aroma transfer) [1,7,8]. Especially, the barrier against oxygen is a key factor for food quality and safety [7]. According to McMillin, appearance, color, lipid stability, nutritive value, and palatability (texture, flavor, aroma) are significant factors that must be considered when choosing a packaging solution [40]. Changing to biobased packaging materials, these factors have to be considered to ensure the shelf life and quality of the food [41,42].

So far, a broad variety of bio-based materials have been investigated to meet the purpose and achieve the properties of commercial packaging plastics. Although many new materials are used, companies worry about their physical inferiority compared to conventional polymers. Currently, European legislation and regulations are forcing companies to eliminate or reduce conventional plastic in packaging (COM/2018/028 final) [8]. Even if there is a desire to change, there is a conflicting pressure that prohibits changes in packaging because of different attitudes along the supply chain. The companies are faced with a challenge of alternatives offering higher costs and lower functionality, existing infrastructure, and inconsistent legislation [43,44].

Today, many companies are trying to use materials that are more recyclable instead of using bio-based materials. According to the adopted "European strategy for plastics in a circular economy" (COM/2018/028 final) where sustainability is the underlying motivation, recycling of plastic packaging is a key factor [8]. Recycling is viewed as the primary mechanism to reduce the environmental and waste management issues that are related especially to the use of conventional plastic [45]. Therefore, sorting and recycling capacities have to be expanded and modernized. Industries are investing in research and innovation activities to develop new technologies that support and increase the recovery of plastic packaging material [14,38,46].

As recently reported, in a circular economy, resources, materials, and products remain as long as possible on the market, minimizing waste and resources. This results in major economic benefits, innovation, and growth. However, safe disposal and recycling of

materials often remain challenging. Reasons are poor management and enforcement, regulatory disparities, lack of infrastructure, and high cost of waste recycle systems [19].

Although many polymers are recyclable, due to additives and related quality issues, recycling rates remain low [38]. Currently, the recycling rate of packaging waste in the EU reaches 67% in total [47], 42% of plastic (packaging waste) [48], and 72% of paper and cardboard [49], respectively. Until 2025, at least 65% by weight of all packaging waste has to be recycled. Regarding specific materials contained in packaging waste, 50% of plastics and 75% of paper and cardboard are the target rates for recycling. By 2030 the recycling rate of all packaging waste rises to 70%, and for plastic and paper and cardboard the targets are 55% and 85%, respectively [50].

Moreover, the multi-layer structure makes plastic waste one of the most complex material mixture from a recycling perspective [16]. Recycling of these materials is accompanied by either high costs, technical difficulties regarding the separation process of the different polymers, or the inability to recycle mixed polymers [8,45]. Current recycling technologies for processing and handling solid plastic waste streams include gasification as a thermo-chemical conversion process for the recycling of polymeric composites, pyrolysis, fluid catalytic cracking, hydrogen technologies (hydrocracking and IH^2 process), and the catalytic pressure-less depolymerization process [16,51–55].

The purpose of future redesign and waste management is the reduction of the amount of plastic that is accumulated in the environment and disposed on landfills, especially in developing countries [56,57]. So, further innovations in both recyclable packaging designs and corresponding cost-effective technologies are needed—independent of the material origin (natural or artificial polymers) [8].

2.2. Food Waste and the Meaning of Packaging

Food wastage and loss describe a major concern in the food supply chain that takes all of the involved stakeholders into consideration [58]. There is an increasing concern about the amount of food waste in Europe as wasted food has a significant impact on the use of natural resources and the environment [59]. The United Nations set a target of halving the actual amount of global food waste per capita at retail and consumer levels and reducing food losses along production and food supply chains as part of their sustainable development goals for 2030 (goal 12.3 of the UN General Assembly) [59].

Differentiating between avoidable (edible) food waste and unavoidable (non-edible) food waste, proper waste management, and recycling strategies is required to reduce unavoidable food waste. Various chemical and biological processes can be used to convert food waste into bio-commodity chemicals and bio-energy [60].

Packaging prevents avoidable food waste and has the potential to further decrease it [42]. According to a study of Bruckner et al., the shelf life of poultry under aerobe packaging conditions at 4 °C accounts for 98.6 h [61]. At 4 °C, the shelf life of poultry packed under modified atmosphere packaging (70% oxygen (O_2)/30% carbon dioxide (CO_2)) is prolonged to 228 h [62]. The kind of packaging has a high impact on the shelf life of poultry and can more than double it.

Caldeira et al. focused on food waste generated in the EU for the major food groups: sugar beets, oil crops, potatoes, vegetables, fruit, cereals, meat, fish, dairy, and eggs. The food waste generated at each stage of the food supply chain was quantified. In total around 638 mega tons (Mt) primary foods result in approximately 129 Mt of food waste generated along the food supply chain. Fruit and vegetables were the food groups presenting the highest amount of food waste overall, with similar amounts generated at the primary production and consumption stages [63]. Products of food categories with a relatively short shelf life, like fresh meat, tend to be the most wasteful products [64,65].

The amount of wasted food means that not only the products themselves are lost but also a high amount of primary resources of fuel, land, and water, including resources needed for breeding and fattening of animals, for cultivation of plants, and raw materials for processing and packaging during production as well as along the entire supply

chain [65,66]. Therefore, for a packaging system, it is important to find a balance between the environmental impact of the package itself, on the one hand, and the impact originating from the potential loss of the packaged product, on the other [66,67].

Considering the environmental impact, this is much higher for producing the food itself than the (multi-layer) plastic packaging. Therefore, if food waste occurs, the negative overall environmental impact rises with every step in the supply chain due to more used resources [8,66,68]. An analysis of the food supply chain and the points where food waste is generated showed that reducing packaging is important, but it must still fulfil its duty of protection as the main criteria for sustainability. Otherwise, the supply chain overall will be less sustainable [24,68,69].

Steinbuchel reported that the production of starch plastic granules requires 25–75% less energy, and GHG emissions are reduced about 20–80% compared to poly(ethylene) (PE) [70]. Weiss et al. reviewed the environmental impacts of bio-based materials in a meta-analysis of LCA data. Therefore, one metric ton (t) of bio-based materials saves 55 ± 34 gigajoules (GJ) of primary energy and 3 ± 1 t carbon dioxide equivalents of GHG emissions relative to conventional materials [31].

Conte et al. assessed the environmental impact of single-layer and multi-layer conventional packaging. The results show that multi-layer surpass single-layer materials by environmental impact when food waste is included in the system boundaries [71].

Pettersen et al. studied the possibility of packaging chicken fillets in recyclable mono-layer materials (high-density polyethylene (HDPE)) instead of complex multi-layered materials (amorphous polyethylene terephthalate (APET)/(PE)) as a replacement for more sustainable packaging system without decreasing the quality of fresh chicken fillets. The results show that a competitive quality and shelf life can be obtained [72].

Zhang et al. focused on a case study, based on LCA data, where the ability of active packaging to minimize food losses by using thymol/carvacrol-enabled active packaging for fresh beef was investigated. Different scenarios have been considered in terms of overall environmental performance of the food and packaging system, including the effect of food loss reduction by using active packaging. It was shown that a breakeven point can be achieved considering the evaluated impact categories in the scenario using the best-performance active packaging whereas differences were observed between the impact categories. The breakeven point can be achieved as early as 0.1% food loss elimination occurs, whereas in the case of cumulative energy demand (fossil), it required more effort to reach the breakeven point. In this case, the active packaging performance needs to reduce food losses at least by 0.6% [73].

3. Plastics Used for Food Packaging

Within the last decades, a broad variety of polymers prepared from renewable resources have been studied as potential substitute for conventional packaging plastics [44]. The European Committee of the Regions stated that further research on the relation between packaging and food preservation on a life cycle basis is needed, and that possible alternative approaches to prevent food waste without the use of fossil-based (complex) plastics has to be investigated [74]. Increasing the exploitation efficiency of natural resources plays an important part on the way to a circular economy [75]. Next to circularity, sustainable packaging should be safe for the environment and humans [76]. The idea of biodegradable polymers, particularly obtained from renewable sources, stems from the need to close the natural cycle of matter [77]. Bio-based applications can be a useful replacement considering the biodegradability, biocompatibility, and recyclability [45,78,79].

Different studies show a limited but growing number of natural polymers used as films and coatings applied for food packaging [80,81]. So far, their practical use depending on the material is restricted due to lower performance in fundamental packaging functions. Challenges such as relatively poor thermal, mechanical, and rheological properties; higher costs; lack of compatibility with the processing and recycling systems currently available; or perceived environmental issues of natural polymers must be overcome. In addition,

the barrier properties of natural polymers, especially the moisture barrier properties due to the hydrophilic nature of these polymers, are detrimental to existing packaging materials [3,82–84].

The WVP is important for fresh foods where dehydration and absorption of moisture should be avoided [85,86]. In general, the water vapor permeability is affected by several factors: chemical structure of macromolecules, degree of cross-linking, crystallinity and porosity, comparative humidity, and the addition of a plasticizer [87]. Oxygen permeability is another fundamental parameter of food packaging material. Low values in oxygen permeability are aimed to prevent deterioration in food quality [88].

To contribute to the reduction of packaging waste by preservation of fresh foods and to enhance their applications, currently, most natural polymers are mixed or blended with synthetic compounds such as PLA, poly(caprolactone) (PCL), and poly(hydroxybutyrate) (PHB). Furthermore, lightweight polysaccharide-based nanomaterials that could replace traditional plastic packaging are shown to improve antimicrobial activity, thermal, mechanical, and gas barrier properties while retaining the biodegradable and non-toxic characteristics of polysaccharides such as chitosan, carboxymethyl cellulose (CMC), and starch [37,83,89–93].

3.1. Classification of Plastics

Currently, the food packaging industry depends on fossil-based plastics which in turn originate from a finite raw material feedstock [8]. The finite resources issue induces a movement towards reducing the usage of virgin plastics towards a plastic production based on alternative raw materials such as renewable resources and biomass waste that have the potential to become plastic alternatives [44,94].

Next to conventional plastics that are fossil-based and non-biodegradable (e.g., PE, PP, poly(ethylene terephthalate) (PET), "bioplastics" were developed. According to the European Bioplastics association, "a plastic material is defined as a bioplastic if it is either bio-based, biodegradable, or features both properties" [95]. Thus, bioplastics involve a range of materials that show different properties and applications [1]. Figure 1 illustrates the categories of the plastics used for food packaging applications.

Thus, biodegradability (and even more compostability) is considered as a useful characteristic providing one option to reduce plastic waste. Biodegradation occurs when a product undergoes a significant change in chemical structure under specific environmental conditions. Biodegradable polymers can, for example, be decomposed to natural substances (CO_2 or methane (CH_4) and water (H_2O)) by microorganisms that are found in the environment like algae, fungi, and bacteria [1,8,89,96]. As biodegradation depends on the chemical structure of the material compound rather than on its origin, the basis of biodegradable plastics are not necessarily renewable resources [1,97,98].

Biopolymers such as proteins, polysaccharides, and lignin are natural polymers produced by the cells of living organisms (e.g., forestry and agricultural crops, terrestrial and marine animals) [29,30]. These biopolymers can be used for the manufacturing of packaging materials and therefore have a high potential to replace synthetic plastics [99,100]. Most common biopolymers currently used for food packaging applications are synthesized [34,83,101,102].

In the following subchapters, the review focuses on different groups of bio-based polymers and their characteristics regarding food packaging applications starting with synthetically manufactured polymers using natural monomers (Section 3.2.) followed by polymers isolated from renewable resources (Section 3.3).
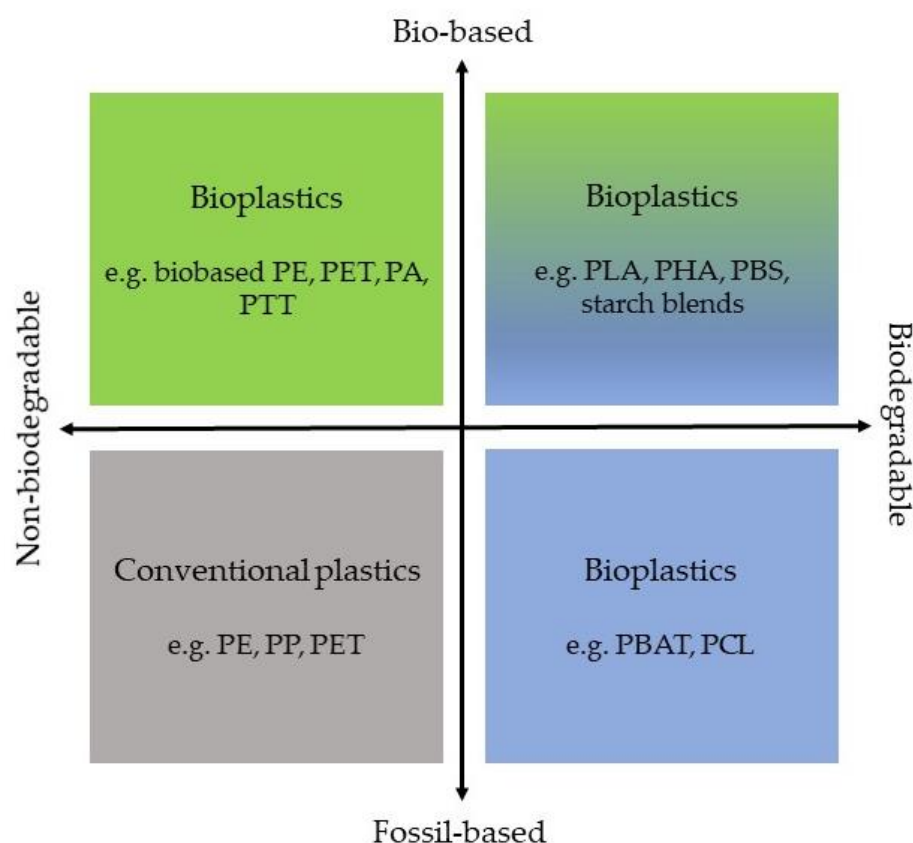
Figure 1. Classification of plastics used for packaging applications. Adapted from [95]. Poly(amide) (PA), poly(butylene adipate terephthalate) (PBAT), poly(butylene succinate) (PBS), poly(butylene succinate) (PBS), poly(caprolactone) (PCL), poly(ethylene) (PE), poly(ethylene terephthalate) (PET), poly(hydroxyalkanoate) (PHA), poly(lactic acid) (PLA), poly(propylene) (PP), poly(trimethylene terephthalate) (PTT).

3.2. Selected Biodegradable Synthetically Manufactured Polymers

3.2.1. Biomass-derived Chemically Manufactured Polymers

One commercially manufactured example should be discussed here: poly (lactic acid) (PLA) and related products. PLA is a thermoplastic and biodegradable aliphatic polyester (Figure 2). As it has its roots in the aliphatic class of polyesters, PLA can be created either by chemical processing of lactic acid monomer or by fermentation of a carbohydrate [102].
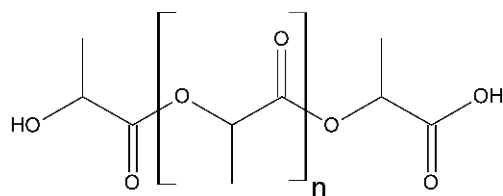
Figure 2. PLA structure.

PLA is the first polymer synthesized from bio-based monomers commercialized on a large scale and can be shaped into injection molded objects, films, coatings, and 3D printed materials [103]. Next to PHAs, starch, and PCL, PLA is the primary biodegradable polymer used for monolayer and some multilayer applications [83,104]. So, PLA films are applied as thermoformed trays, cups, bowls, bags, or jars for packaging of fresh salads, ready-to-eat meals, deli products, beverages, potato chips, and yoghurt among other uses [105,106]. Renewability, biodegradability, and biocompatibility are attributes that make PLA one of

the best polymeric substitutes for various fossil-based polymers [83]. However, the PLA synthesis—and in turn the corresponding products—are still rather expensive [39].

So far, PLA and corresponding copolymers are used to substitute polyolefins as high-density poly(ethylene) (HDPE), low-density poly(ethylene) (LDPE), poly(propylene) (PP), PET, and poly(styrene) (PS) as packaging materials [37,83] due to comparable mechanical properties like stiffness and tensile strength, gas permeability, and transparency (Table 1) [107–109].

Table 1. Barrier and mechanical properties of selected synthetic polymers and biomass-derived chemically synthesized materials.

Film Composition	Permeability			Mechanical Properties		References
	O_2	CO_2	H_2O Vapor	Tensile Strength	Elongation at Break	
	$[cm^3 \cdot mm/(m^2 \cdot d \cdot atm)]$		$[g \cdot mm/(m^2 \cdot d \cdot atm)]$	[MPa]	[%]	
Synthetic Polymers						
HDPE	44–91 (20 °C, 65% RH)	100 (23 °C, 0% RH)	0.15 (38 °C, 90% RH)	32	150	[110,111]
LPDE	98–183 (23 °C, 50% RH)	NR	0.44 (38 °C, 90% RH)	10	400	[110,111]
PE	50–200 (23 °C, 50% or 0% RH)	100–1000 (23-25 °C, unknown RH)	0.5–2 (23 °C, 85% RH)	18	350	[111–113]
PET	1–5 (23 °C, 50% or 0% RH)	3–7 (23 °C, 75% RH)	0.5–2 (23 °C, 85% RH)	55	300	[111–113]
PP	50–100 (23 °C, 50% or 0% RH)	200–900 (unknown conditions)	0.2–0.4 (23 °C, 85% RH)	26	80	[111–113]
Biomass-derived chemically synthesized materials						
PLA	3.5–15 (23 °C, 50% or 0% RH)	32.9–72 (23 °C, 0% RH)	1.6–3.6 (38 °C, 85% RH)	50.45	3	[110,114,115]

NR: Data not reported.

The production of composites by adding nanofillers is a way to extend and improve the properties of PLA [83,104,109]. The addition of many nanofillers (three-dimensional spherical and polyhedral, two-dimensional nanofibers or one-dimensional sheet-like nanoparticles) has been studied and lead to satisfactory achievements in the design of PLA nanocomposites [116].

Panseri et al. studied the effectiveness of PLA-based packaging solutions compared to a conventional reference package consisting of APET/PET trays wrapped in plastic films of poly(vinyl chloride) (PVC) to store red fresh meat during its refrigerated shelf life. By using PLA packaging in combination with a gas mixture of 66% O_2, 25% CO_2, and 9% N_2, it was possible to maintain an optimum red color together with a reduced content of volatile compounds associated to off-flavors of meat samples [117].

Marra et al. investigated biocomposite films of PLA with zinc oxide regarding mechanical, barrier, and antimicrobial properties. The results showed that PLA films with 5 wt% of zinc oxide exhibit good mechanical properties related to a high modulus and stress at yielding, decrease of permeability to CO_2 and O_2 carbon dioxide and oxygen, and a slight increase of water vapor permeability. Furthermore, the incorporation of 5% zinc oxide leads to an antimicrobial activity against *E. coli* after 24 h with a reduction value of 99.99% [37]. Vanitha and Kavitha incorporated cellulose natural fibers from palm sprouts in a PLA matrix. The results showed that the mechanical resistance increased, and the water absorption rate decreased significantly with the optimum concentration of palm

sprouts fiber in the PLA-film. Interactions between palm sprout and PLA restrict the water infiltration [118].

The incorporation of lignin in PLA films via simple blending results in a small but significant increase of the oxygen barrier properties, as well as an improved antiradical efficiency that increases with the severity of the heat treatment of the blends [119]. Moreover, the water sorption capacity decreased with an increase of lignin loading from 7 wt% to 15 wt% while tensile strength increased, as shown in a study of Spiridon et al. [120]. Gordobil et al. used commercial alkaline lignin and organosolv lignin from almond shells as PLA filler, which greatly improved the thermal stability and increased the elongation at break. Low percentages up to 1% unmodified lignin did not affect the maximum strain, while it was decreased with increasing lignin content at percentages greater than 5% [121]. In addition, kraft and organosolv lignin were examined as nucleating agents, showing that both lignins induce heterogeneous nucleation and increase the crystallization rate in PLA by shortening the crystallization half time and increasing the degree of crystallinity in PLA, while not affecting the processing window of the polymer [122]. One problem when incorporating lignin is its compatibility with PLA, which can be overcome with the addition of triallyl isocyanurate (TAIC), leading to the formation of PLA-TAIC-lignin crosslinked structures as interface, improving the compatibility in the blend and thus the mechanical, thermal, and hydrolytic degradation properties [123]. However, using TAIC, the biocompatibility has to be studied before applying it for food packaging. Another possibility is the introduction of lignin nanoparticles (LNP) into PLA using a Pickering emulsion template method where lignin acts as stabilizer. According to the results of this study, lignin could increase the decomposition temperature by approx. 10%, reduce the light transmission in the UV region, and increase the Young's modulus but also decrease the tensile strength and elongation at break. Moreover, the crystallinity of PLA could be improved with the addition of lignin [124]. In another approach, LNP in PLA films could inhibit the growth of bacterial plant pathogens and showed a high antioxidant activity, while migration values remained below the legislative limits, suggesting the exploitation of LNP-PLA films as food packaging material [125].

The combination of LNPs with another lignocellulosic nanofiller, namely, cellulose nanocrystals (CNC), in PLA films can in fact improve UV light blocking capability and strength and modulus values compared to neat PLA or PLA binary systems, confirming a synergic effect of LNP and CNC [126]. This was also reported in another study, where Young's modulus, elongation at break, and toughness of neat PLA films were improved by 14%, 77%, and 30%, respectively, by incorporation of high lignin-containing cellulose nanocrystals. In contrast, commercial lignin-coated CNCs showed inferior crystallinity, smaller surface area, and a higher degree of agglomeration, concluding that the presence of LNPs is important for the compatibility between the PLA polymer matrix and CNCs [127].

3.2.2. Polymers Produced by Microorganisms

Here, we discuss the most prominent representatives of polymers produced by microorganisms: PHA and PHB and corresponding copolymers or composites. In the group of PHAs, more than 100 known bio-derived polymers exist. The most common ones are PHB and corresponding copolymers such as poly(3-hydroxybutyrate-co-3-hydroxy-valerate) (PHBV) and poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBHHx) (Figure 3) [110,128].
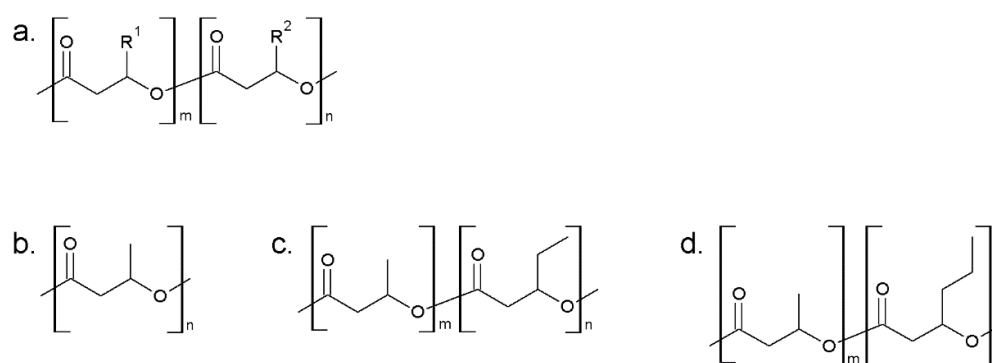
Figure 3. Overview of several poly(hydroxyl alkanoate)s (PHA) used for food packaging applications: (**a**) general structure of PHAs with residues R1/R2 as alkyl chains ranging from 1-13 carbons, (**b**) poly(hydroxyl butyrate) (PHB), (**c**) Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), and (**d**) poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBHHx).

In general, PHAs can be used to coat paper or paperboard to produce water-resistant surfaces, making the coated material completely biodegradable. PHB-coated paperboard has been used for packaging of ready meals, while PHBV-coated paperboard has been used for dry products, dairy products, and beverages [129]. In addition to functionalizing the surface of fiber-based materials, PHAs can also functionalize paper and board's grease resistance and sealability [110]. PHAs involve a range of biodegradable thermoplastic polymers that are produced through fermentation by different microorganisms [99]. These polymers are characterized by thermomechanical properties that are similar to synthetic polymers such as PP [130]. PHAs can be processed into different products including films, trays, and coatings on other bio-based materials (e.g., paperboard) [110].

Initially, PHAs were used to make everyday articles like shampoo bottles. Moreover, they are used to produce carrier bags, containers and paper coatings, biodegradable bags, and lids. Currently, their use in terms of packaging applications is restricted as they are not transparent [106,110]. Their gas and water vapor permeability offer opportunities to be applied as food packaging materials. Copolymerization as well as blending are used to improve physical-mechanical properties of PHAs [110].

Several studies have shown that PHB, PHBV, and PHBHHx films are promising materials for food packaging due to their good barrier properties. The oxygen permeability of PHAs is comparable to PET and PLA. The values are much lower compared to conventional polymers such as PE and PP. The water vapor permeability of PHAs is similar to materials such as PET and PLA but slightly higher than more apolar polymers such as PE and PP. The carbon dioxide permeability of PHAs is higher compared to PET but substantially lower than for common packaging materials such as PP and PE (Table 2) [131–136].

Table 2. Barrier and mechanical properties of bio-based polymers produced by microorganisms.

Film Composition	Permeability			Mechanical Properties		References
	O_2	CO_2	H_2O Vapor	Tensile Strength	Elongation at Break	
	$[cm^3 \cdot mm/(m^2 \cdot d \cdot atm)]$		$[g \cdot mm/(m^2 \cdot d\ atm)]$	[MPa]	[%]	
PHB (P3HB)	2–11.4 (23 °C, 0% RH)	3–28.9 (23 °C, 0% RH)	1–5 (unknown conditions)	35–40	3–8	[110,137]
PHBV (P(3HB-co-3HV))	4.9–16.7 (25 °C, 0% RH)	146 (25 °C, 0% RH)	1.5 (38 °C, 90% RH)	38	NR	[110,137]
PHBHHx	8.3 (23 °C, 0% RH)	54 (23 °C, 0% RH)	1.42 (23 °C, 0% RH)	20	850	[132,137]

NR—Data not reported.

Dilkes-Hoffman et al. summarized that a combination of PHA with thermoplastic starch (TPS), which provides one of the best oxygen barriers of all polymeric materials, seems to have the potential to lower food spoilage rates compared to conventional packaging materials according to good barrier properties [45].

PHB shows a high crystallinity and a high melting point. Therefore, PHB is often blended with PLA. This results in materials of improved mechanical, thermal, and physical properties compared to neat PLA [128]. Arrieta et al. figured out that blending PLA with 25% (*w/w*) PHB resulted in improved oxygen and barrier properties, whilst the inherent transparency of PLA was reduced [138].

Kovalcik et al. studied the melting and crystallization behavior, thermo-oxidative stability, mechanical and viscoelastic properties, and permeability for oxygen and carbon dioxide of composite materials of microbial PHBHV with methanol fractionated kraft lignin. The results showed that a concentration of already 1 wt% of methanol-extracted kraft lignin can act as an active agent for decreasing the oxygen as well as carbon dioxide permeability of PHBHV films. The gas permeability was decreased for oxygen by 77% and by 91% for carbon dioxide, respectively, compared to the native PHBHV film. The low thermo-oxidative stability of pure PHBHV was increased for the lignin-containing films. Based on this results, methanol-extracted kraft lignin is suggested as a suitable active additive in PHBHV films for applications, especially in the field of food packaging [133].

Although, high production costs limit the competitiveness in commercial applications, PHAs might have high potential as bio-based and biodegradable plastic packaging materials in the transition towards a circular economy [110].

3.3. Selected Plant-derived Polymers

3.3.1. Lignocellulosic Biomass and Lignin

Lignocellulosic biomass is the major structural component of plants, mainly consisting of cellulose (40–60%), hemicellulose (10–40%), and lignin (15–30%), whereby the latter one is the most complex constituent [139]. Lignin is a randomly crosslinked macromolecule composed of the three monolignols *p*-coumaryl alcohol, coniferyl alcohol, and sinapyl alcohol, which form the residues *p*-hydroxyphenyl (H), guaiacyl (G), and syringyl (S), respectively, as shown in Figure 4 [140]. It can be obtained from woody biomass (e.g., pine, poplar, birch), annual plants (e.g., wheat straw, miscanthus, switchgrass), or agricultural residues (e.g., sugarcane bagasse) by various extraction processes. The molecular structure of lignins strongly depends on the botanical origin but also on the growing site, season, and isolation process [141,142]. There are different types of technical lignins that can either be classified as sulfur-containing or sulfur-free. The most common ones are lignosulfonates, kraft lignin, organosolv lignin, and soda lignin [139]. Most industrial lignins originate from the pulp and paper industry with up to 90 million tons of kraft lignin released per year worldwide, though only 2% of it are used commercially for value-added products [143]. One reason for that might be the deficient quality or missing specifications of technical lignins, as they are rather undefined products with a complex composition and impurities from the pulping process (such as remaining sugars or thiol groups). So far, this restricts their industrial exploitation, as resulting products have varying properties, which are inferior to fossil-based products.
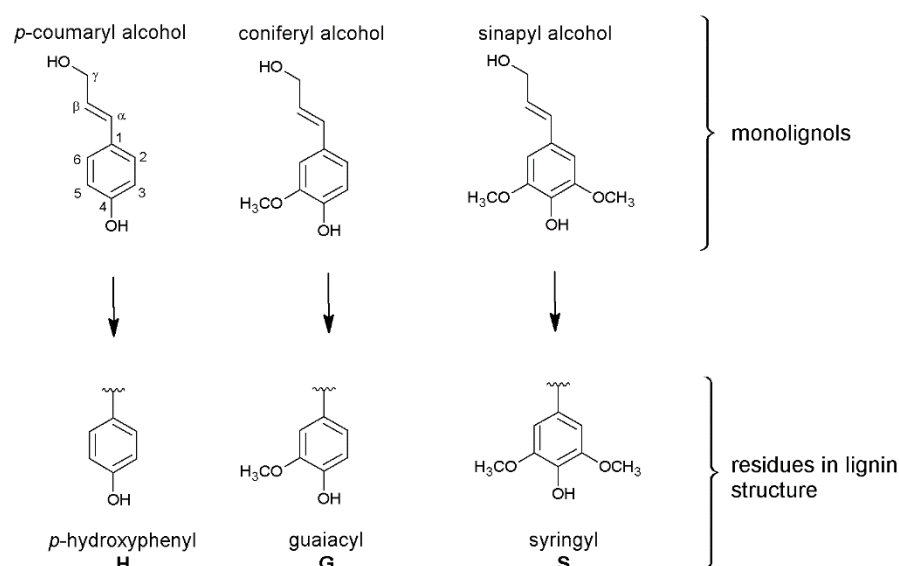
Figure 4. Lignin monolignol structures: p-coumaryl alcohol, coniferyl alcohol, and sinapyl alcohol forming the specific residues p-hydroxylphenyl (H), guaiacyl (G), and syringyl (S) [140].

Nevertheless, in consideration of the global energy crisis and the depletion of fossil fuels and petrochemicals, the potential of lignin has been a key topic in biorefinery research [139,144–146]. As polyphenols, lignins possess numerous interesting functional properties, such as antioxidant activity, and thus, they are investigated as active packaging materials for the protection of light- or oxygen-sensitive goods [119]. Antioxidant polymers are a field of great interest, as the use of macromolecular antioxidants is related to the possibility to produce materials with long-term stability. Due to its cross-linked 3D structure and enzymatic resilience, lignin possesses a higher thermal and biological stability compared to low molecular weight compounds and thus could be used in special fields where the exploitation of low-molecular antioxidant substances would be inefficient due to their higher diffusion rates. Moreover, carcinogenic effects have been observed for synthetic antioxidants: butylated hydroxyanisole (BHA) and butylated hydroxytoluene (BHT), for example, are able to cause cytotoxicity and carcinogenesis as shown in in vivo studies [147]. Azadfar et al. have already shown that lignin has the potential to serve as raw material for antioxidants like guaiacol and 4-vinylguaiacol, whereby their antioxidant activity is comparable to that of commercial antioxidants [148].

Consequently, lignin has gained increasing interest as environmentally benign antioxidant and its ability to improve mechanical, thermal, and barrier properties when incorporated in conventional packaging films. In general, a reduction in WVP of lignin-based films is explained by the hydrophobicity of lignin. It acts as a barrier in the polymeric matrix and increases the path for the diffusion of water vapor, resulting in lower permeation of water molecules through the polymer film [149]. Next to the utilization of lignin as additive/blend in different polymer matrices, it could also be used as raw material for the development of polymeric packaging materials, e.g., as polyol substitute in polyurethanes or polyesters or as phenol substitute in resins. Hult et al. investigated softwood lignin esterified with tall oil fatty acid as coating on paper board. The thermoplastic properties of lignin were enhanced, and the water vapor and oxygen transmission rate decreased while tensile strength was not affected [150]. Polyurethanes of high transperency and flexibility for construction or packaging applications were prepared by Klein et al., where petroleum based polyols could be substituted with kraft lignin up to 80 wt% [151]. In addition, demethylated lignins were also used to enhance the reaction selectivity towards polyurethane formation [152]. In a recently published study by Hao et al. thermoset coatings with integrated self-healing and removal properties were investigated. They investigated a kraft lignin functionalized with carboxylic acid groups as curing agent

with poly(ethylene glycol) diglycidyl ether, which resulted in a crosslinked structure [153]. Hambardzumyan et al. designed novel nanocomposite films of lignin and CNC that could be self-supported or used as coatings. This combination of the antibacterial properties of lignin and oxygen barrier properties of CNC films are promising as food packaging material [154].

Rastogi et al. summarized different possibilities for bio-based paper coatings. Next to polyester and polysaccharides discussed so far, also lipids and proteins could be used [155]. Nevertheless, a full exploitation at industrial scale is not possible, due to different crystallization behaviour, brittleness, or melt instabilities that lead to difficulties in processing of these biopolymers. Blending the lipids and proteins with other biopolymers, such as lignin, may provide a route to overcoming this obstacle.

Lignin cannot only be used in paper coatings but also can be used to improve the strength of paperboard. Flory et al. developed a green binder system with *Salix* lignin that was equal to the wet tear strength of the commercial vinyl acetate binder [156]. Inspired by the reinforcement principle of lignin and cellulose in wood, Jiang et al. developed a cellulose fiber scaffold with lignin as reinforced matrix via successive infiltration and mechanical hot-pressing treatments. The resulting composite shows a high isotropic tensile strength of 200 MPa, compared to 40 MPa of conventional cellulose paper, and Young's modulus of 10 GPa, which is even higher than many fossil-based plastics. In addition, also the thermostability and UV-blocking performance is enhanced due to the lignin addition [157].

In conclusion, lignins are promising candidates for environmentally benign antioxidants with a great abundance [158–160]. Regarding the studies summarized in this review, lignin use as an additive seems more favorable than its copolymerization, as copolymerization usually requires at least one functionalization (such as demethylation or carboxylation). Thus, further effort is necessary to get a deeper understanding of the lignin structure and the processing conditions required to maintain and enhance the antioxidant and antimicrobial properties.

3.3.2. Protein-Based Polymers

Research in food packaging has also focused on protein-based films due to their good film-forming properties, low cost, and biodegradable nature [83]. Materials synthesized from proteins exhibit desirable film-forming and barrier properties, which are often comparable to fossil-based products [90]. Proteins from different sources have been investigated for the synthesis of bioplastic films for packaging applications including collagen, gelatin, caseins, soy/whey/quinoa protein, egg white protein, myofibrillar protein, corn zein, wheat gluten, and keratin [83,161,162].

Among all the protein sources, soy proteins got great attention as a potential source for bio-based packaging materials. This development is based on excellent film-forming and oxygen barrier properties of films produced from soy protein isolate. However, these materials cannot meet the requirements of a film with mechanical and water barrier properties guaranteed by conventional plastics [83,163]. Compared to films from other proteins, soy protein-based films are characterized by transparency, flexibility and cost-effectiveness [162]. Furthermore, they show good oxygen barrier properties under low moisture conditions [164]. A disadvantage that limits their use beside low mechanical strength is a lack of heat stability compared to LDPE [165–168].

Whey proteins are able to form elastic films [169], which are transparent, flexible, and exhibit good oil and oxygen barrier properties at low humidity. A disadvantage is a moderate moisture permeability. Nevertheless, whey proteins have been intensively studied as raw material for biodegradable packaging [170].

Collagen and gelatin are proteins originating from animal sources acquired by a controlled hydrolysis reaction. In nature, collagen is the most abundant occurring protein [171]. Collagen-based bioplastic films are characterized by good mechanical properties, [172] and therefore, are suitable for various applications [173]. In contrast to collagen-based

films, gelatin films show poor mechanical and barrier properties according to their hydrophilic nature [174]. Biscarat et al. determined functional properties of gelatin-based films. Compared to synthetic polymers, good gas barrier properties were reached by gelatin films cross-linked with ferulic acid. Gelatin films with poly(ethylene glycol) (PEG) 200 showed high gas barrier properties and high permselectivity towards carbon dioxide and oxygen [80].

Gelatin has been introduced in the manufacturing of packaging films due to its low cost and abundance [175]. Furthermore, gelatin is used to produce biodegradable packaging materials due to its good properties, such as low melting and gelling points, good capacity of oxygen barrier, biodegradability, and excellent film formation [176]. The use of gelatin-based composite films incorporating other materials like chitosan, sunflower oil, and corn oil to enhance the barrier and mechanical properties of these films was studied by different authors [177]. In addition to gelatin, gluten is used to prepare films of high homogeneity, excellent gas barrier properties, and mechanical strength [178].

Kanatt developed a new intelligent-active food packaging film using poly(vinyl alcohol) (PVA) and gelatin incorporated with Amaranthus leaf extract to monitor freshness and increase the shelf life of fish and chicken meat. Incorporation of Amaranthus leaf extract improved its mechanical and water vapor barrier properties next to active functions. The decrease in solubility enables the use for packaging of flesh foods. Samples packed in neat films had a shelf life of 3 days while those in active films spoiled after 12 days. The results of the study suggest the application of Amaranthus leaf extract containing PVA-gelatin films being both active and intelligent ensuring quality and safety of flesh foods [179].

For comparative purposes, Table 3 shows the discussed barrier as well as mechanical properties of protein-based polymers.

Table 3. Barrier and mechanical properties of protein-based polymers.

Film Composition	Permeability			Mechanical Properties		References
	O_2	CO_2	H_2O Vapor	Tensile Strength	Elongation at Break	
			[g·mm/(m²·d·atm)] *	[MPa]	[%]	
Soy protein isolate film with glycerol (casting method)	NR	NR	1710 (23 °C, 50% RH)	6.97	113.94	[180]
Whey protein isolate film with glycerol	92.448 g/(m²·d) (25 °C, 90% RH)	NR	1680 (25 °C, 75% RH)	3	13	[181]
Pumpkin oil cake protein isolate film with glycerol	16.06 cm³/(m²·d·atm) (23 °C; unknown RH)	21.15 cm³/(m²·d·atm) * (23 °C; unknown RH)	NR	0.86–6.56	22.2–196.61	[165]
Fish gelatin film with glycerol	NR	NR	1040 (24 °C, 50% RH)	9.08	44.93	[182]
Beef skin gelatin films with corn oil (extrusion)	$0.8\text{–}4.7 \times 10^{-4}$ cm³·mm/(m²·d·atm) (23 °C, 50% RH)	NR	$4.05\text{–}8.61 \times 10^6$ (23 °C, 50% RH)	1.43–5.37	1.68–2.60	[183]

* In some data, the units were normalized. NR—Data not reported.

3.3.3. Polysaccharides

Packaging films based on carbohydrate sources are generally transparent and homogeneous films with effective oxygen barriers at intermediate to low humidity and good mechanical properties. According to their hydrophilic character, they have poor water vapor barrier qualities, and they are relatively sensitive to moisture. Furthermore, the films obtained from several polysaccharides are brittle usually due to interactions between the polymer chains. This leads to limited applications of polysaccharide- and protein-based coatings and films [2,106,184]. To meet the required properties, a pretreatment such as plasticization with small molecular weight–compatible constituents, blending, or chemical modification is needed [130]. While considering mechanical properties, the tensile strength

of polysaccharide-based films are similar to those of synthetic polymers; differences are observed in elongation at break (Table 4) [82].

For several polysaccharides, the film-forming properties and especially their potential for edible packaging has been studied, including starch, cellulose, and its derivatives, alginate and chitosan (Figure 5) [161,185–187].
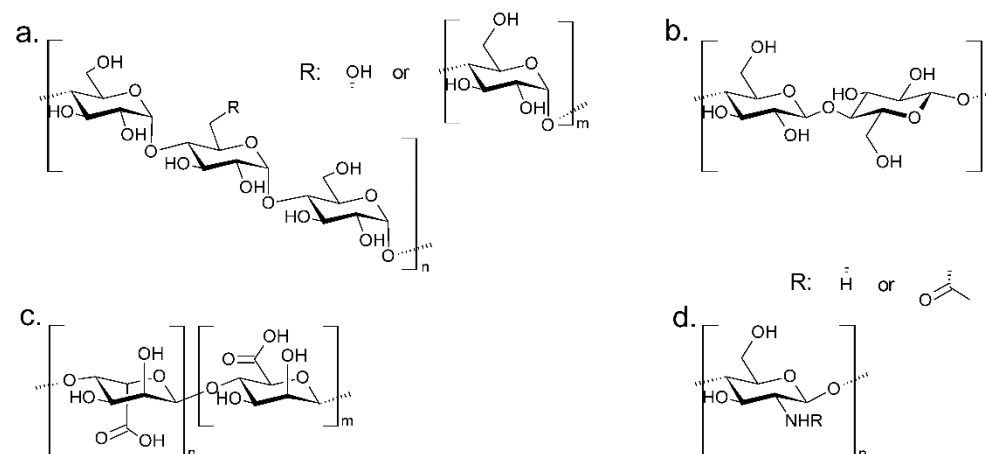

Figure 5. Overview of several polysaccharides used for food packaging (edible) films: (**a**) starch, (**b**) cellulose, (**c**) alginate, and (**d**) chitosan [188].

Table 4. Barrier and mechanical properties of polysaccharide-based polymers.

Film Composition	Permeability		Mechanical Properties		References
	O$_2$	H$_2$O Vapor	Tensile Strength	Elongation at Break	
	[g·mm/(m^2·d·atm)] *		[MPa]	[%]	
High amylose cornstarch films without plasticizer (amylose:amylopectin ratio 80:20)	NR	1260 (20 °C; 52.9% RH)	34.32	1.41	[189]
Low amylose cornstarch films without plasticizer (amylose:amylopectin ratio 25:75)	NR	1430 (20 °C; 52.9% RH)	44.38	2.40	[189]
Thermoplastic (cassava) starch (extrusion)	0.182 (Ambient temp., 0% RH)	36.8 (25 °C, 50% RH)	5.8	78	[190,191]
Methylcellulose mixtures in ethanol	NR	446–945 (25 °C; 52% RH)	25–33	29–14	[192]
Hydroxypropyl methylcellulose without plasticizer	NR	974,000 (23 °C, 50% RH)	61.04	29.51	[193]
Carboxymethyl cellulose film with glycerol (casting method)	NR	683 (25 °C, 52.8% RH)	6.10	201.73	[194]
CMC-film with 50 wt% ethanol organosolv lignin with glycerol (casting method)	NR	2570 (20 °C, 0% RH)	20	5.92	[149]
Agar/10% lignin composite film with glycerol	NR	13,400 (25 °C, 50% RH)	51.8	22.1	[195]
(Sodium) alginate film with glycerol (casting method)	NR	13,600 (25 °C, 50% RH)	41.1	8.5	[196]
Chitosan film with glycerol	0.188 × 10^{-2} (25 °C, <50% RH)	210–3020 (25 °C, 0–100% RH)	8.9	38.5	[197]

* In some data, the units were normalized. NR: Data not reported.

Starch

Starch is a natural polysaccharide easily available on an industrial scale. Many plant-based polysaccharides and cost-effective starch-based materials have been extensively investigated as an alternative material for fossil-based food packaging applications due to their environmental compatibility and biodegradability [198]. Although starch-based packaging, which has good film-forming properties and excellent oxygen barrier, is already in wide use, this material still has some disadvantages such as the poor vapor and oxygen moisture barrier and poor mechanical properties compared to conventional non-biodegradable plastics used in food packaging industry (Tables 1 and 4) [198–201].

The oxygen barrier properties are correlated to a high-ordered hydrogen-bonded network structure. The barrier properties can be improved by increasing the crystallinity or a higher content of amylopectin [199]. The poor moisture barrier is caused by a strong hydrophilic behavior [200]. A higher crystalline structure in starch-based films leads to less sensitivity of moisture and to environmental relative humidity. Considering the poor mechanical properties, starch-based films show a relatively high tensile strength while the elongation percentage is low [199]. The high tensile strength is attributed to the extensive intra-molecular hydrogen bonds between amylose, amylopectin, and amylose-amylopectin molecules. Amorphous regions in starch-based films formed by amylose cause brittleness and thereby influence the poor mechanical properties [189].

As starch-based films are odorless, colorless, and tasteless, starch is used in either pure or blended form as a biodegradable coating or packaging film. The coatings of edible starch are also applied for other kinds of foods to maintain quality and to extend the shelf life of products [200,202–204]. However, as starch blends may contain additives like compatibilizers and plasticizers that can migrate out of the matrix, only some starch blends are suitable for food packaging applications [106].

Starch has already been combined with lignin to improve its poor thermo-mechanical properties while simultaneously decreasing its water vapor permeability significantly [205]. Miranda et al. confirmed these findings in their studies, showing that the presence of lignin in combination with CNC increased maximum stress and modulus of elasticity, barrier properties, and the thermal stability of the material [206,207]. Javed et al. published a study concerning starch-based coatings for paper packaging materials with lignin, investigating the self-supporting films regarding their mechanical properties and chemical stability in water as well as their barrier properties when used as coating on paper board. When lignin is added, the dissolution of starch from the composites could be significantly decreased. The addition of ammonium zirconium carbonate (AZC) leads to further improvement of the storage modulus, indicating that crosslinking had occurred [208].

Cellulose and Derivatives

Cellulose represents the most abundant renewable polymer source available in nature. Biodegradable films made out of this raw material are characterized by renewability, low cost, non-toxicity, biocompatibility, biodegradability, and chemical stability [209]. For example, cellulose films (known as Cellophane®) are used for wrapping fruits in bio-based trays [105].

Films made of cellulose exhibit good toughness, tensile strength, high surface gloss, and good transparency [210]. Whereas hemicellulose-based films are brittle, the flexibility, toughness, and oxygen permeability can be improved by addition of plasticizers [211,212]. A disadvantage of cellulose films is their poor water vapor barrier. This is caused by the underlying hydrophilic nature of polysaccharides [83]. Soaking cellulose with alkali to swell the structure followed by different derivatization reactions, CMC, methylcellulose (MC), hydroxypropyl cellulose (HPC), and hydroxypropylmethyl cellulose (HPMC) are available. They are used as raw materials to prepare biodegradable films, which are transparent, water soluble, odorless, tasteless, and flexible and have moderate strength and resistance to lipid compounds (Table 4) [213].

CMC is most often used for biodegradable film production. It is highly soluble (in water) and crystalline and can build solid and flexible films [214,215]. Beneficial characteristics include film-forming properties, good mechanical and gas barrier properties, transparency, ease of processing, and low price [216]. Next to its good film-forming properties, CMC has been studied as antibacterial food packaging in composites with chitosan [217,218] and pectin [219]. Michelin et al. investigated the incorporation of organosolv lignin from corncob in CMC-based films, which leads to an improved water resistance of approx. 60% and reduction in the water vapor permeability of 20%, while also enhancing the thermal stability and antioxidant activity [149].

As mentioned before, mechanical and barrier properties of cellulose-based films can be improved by the production of nanocomposites. Moura et al. proposed nanocomposites using chitosan as nanofiller in HPMC to enhance mechanical and film barrier attributes. HPMC films containing different concentrations of chitosan as nanoparticles were analyzed for mechanical properties, water vapor permeability, and oxygen permeability. They realized that chitosan nanoparticles tended to fill up poriferous spaces in the HPMC-matrix. This improves film tensile properties and water vapor permeability, concluding that a HPMC-chitson nanocomposite could be a possible material for food-packaging applications to extend the shelf life of food [158,220].

Another approach is the preparation of HPMC/chitosan films [221]. Alzagameem et al. examined the incorporation of different lignins in HPMC and HPMC/chitosan films [158,160]. Results show that lignins are in general more active against Gram-positive bacteria than against Gram-negative bacteria and that films with organosolv lignin possess a higher activity against *S. aureus* than films with kraft lignin. It was shown that biomass as well extraction process influence the properties of the films and that the antioxidant activity of lignins correlates with different parameters such as genotype and phenotype of biomass, pulping and purification of lignin and the resulting heterogeneity [159].

Cellulose acetate (CA), cellulose acetate propionate (CAP), and cellulose acetate butyrate (CAB) are thermoplastic cellulosic-derived materials developed through esterification of cellulose. CA gets great attention because of its biodegradable nature, excellent optical clarity, and greater toughness [83].

Alginates

Alginate and corresponding derivatives are one of the most promising carbohydrates for packaging applications, especially for foods that are sensitive to gas permeation [222]. Alginates are naturally occurring indigestible polysaccharides. They are commonly produced from various genera of brown algae [223]. A lot of research on alginate has focused on edible coatings or to improve color and flavors [224,225].

Ahmed et al. evaluated the barrier, mechanical, and oil resistance of paper sheets coated with a novel cost-saving coating prepared at different temperatures to be used in packaging purposes. The coating is based on sodium alginate (SA) with new core-shell inorganic particles composed of waste silica fume core covered with cobalt(II) oxide/zinc oxide (CoO.ZnO) oxides. SA is broadly used due to high oil resistance and enhanced greaseproof properties.

Very recently, a binary oxide CoO.ZnO on the surface of silica was shown to decrease the penetration of oil and grease through the paper pores leading to lower oil absorption and enhanced mechanical properties. Tensile strength was decreased whereas stiffness showed slight increase in case of paper sheets coated with SA-CoO.ZnO/SiO_2. Incorporation of SA-SiO_2 and SA-CoO.ZnO/SiO_2 in the fiber matrices improved the tear and burst indices properties. The network created by SA film blended with (CoO.ZnO/SiO_2) pigments on paper sheet substrate drastically changed their mechanical and barrier properties. They are highly dense and organized, causing the creation of smooth distribution of nanoparticles and a strong surface that fills the pores in the paper matrices and increases the air, water vapor, and oil resistance of the samples coated with SA-CoO.ZnO/SiO_2 pigments compared with uncoated paper [223].

Chitosan

Chitosan is the second most widely found amino-polysaccharide in nature [226]. It is a biopolymer with antibacterial properties that can be found in fungi, insects, crabs, and shrimps and makes up a significant part of the crustacean waste that is discarded every year [227]. Chitosan is intensively investigated for biomedical applications [228] and bioactive packaging solutions [229]. Properties like being microbe resistant, biocompatible, and biodegradable make chitosan attractive for research and use in various applications [230]. In food packaging, potential applications of chitosan blend-based films are for fresh products (vegetable, meat, and fish) and foods with short to medium shelf life [231]. Moreover, chitosan is widely used as a material for nanofibers production due to its ability to form films with antibacterial properties [232,233]. Moreover, chitosan-based materials exhibit good mechanical properties and a selective permeability to carbon dioxide and oxygen [101,234]. The tensile strength and elongation at break values are comparable to appropriate values of HDPE and LDPE (Table 4).

However, the use of chitosan to produce flexible packaging is still limited due to high sensitivity to humidity and moisture and high oxygen permeability. As chitosan films are highly permeable to water vapor, their use in food packaging is restricted [234–236]. To overcome these limitations of this otherwise excellent polysaccharide biomaterial, several attempts have been made such as blending the films with other natural or synthetic polymers and the addition of several active and functional substances like fillers, plasticizers, crosslinkers, and natural oils [234].

Several studies are published that focus on improving the barrier and mechanical properties of chitosan-based films. Kamdem et al. developed a composite flexible film using chitosan as base polymer matrix, xylan to improve mechanical properties, and carvacrol (a monoterpenoid phenol) to control microbial protection. The results show that adding xylan significantly increases the elongation at break of the composite films and exhibits higher tensile strength and Young's modulus. The incorporation of carvacrol and xylan in the composite films was not effective in terms of antimicrobial activity [101].

Chitosan is another potential packaging material that is investigated with lignin as additive. The addition of lignin to chitosan improves the tensile strength, storage modulus, glass transition temperature, and degradation temperature, compared to pure chitosan films [237]. Moreover, lignin confers the scavenging properties to the chitosan films: this antioxidant activity is highly dependent on the film structure, functional properties, and surface activity, which is, in turn, dependent on moisture [238] and the homogenization process of the film-forming suspension [239].

Despite favorable properties of chitosan like its antioxidant and antimicrobial activity as well as a good biocompatibility and biodegradability, there are some drawbacks such as its dissolution in acidic media and poor thermal properties. Thus, the combination of chitosan with other polymers in binary or ternary films is intensively studied for various systems. Yang et al. investigated films based on PVA, chitosan, and LNPs for food packaging applications and found that lignin improved tensile strength and Young's modulus as well as the thermal stability of the systems [240].

In conclusion, Table 5 summarizes advantages and disadvantages of the polymers discussed in Section 3.

Table 5. Advantages and disadvantages of different bio-based polymers discussed in Section 3.

Material	Advantage	Disadvantage	Reference
PLA-based	Renewable, biodegradable, biocompatible Usable for mono- and multilayer applications Desirable mechanical properties (stiffness, tensile strength) Good gas permeability Transparent	Expensive (synthesis) Limited to rigid packaging	[39,83,104,107–109]
PHA-based	Water-resistant surfaces by coating Functionalize grease resistance and sealability Good thermomechanical properties Desirable gas permeability and WVP	Not transparent	[106,110,129,130]
Lignocellulose biomass and/or lignin-based	Abundance in nature Antioxidant activity with long-term stability Improves mechanical, thermal, and barrier properties Reduction in WVP Biodegradable	Deficient quality (technical lignins) Copolymerization requires functionalization	[147,149,158–160]
Protein-based	Abundance in nature Good film-forming properties Desirable barrier properties Transparent Low cost/cost-effective Environmentally compatible, biodegradable	Low mechanical strength Lack of heat stability	[83,90,165–168]
Polysaccharide-based	Abundance in nature Effective oxygen barriers (intermediate to low humidity) Good mechanical properties Transparent Potential for edible packaging	Poor water vapor barrier Sensitive to moisture	[2,82,106,184,187,198]

4. Natural Additives in the Context of Active Food Packaging

In the packaging and food industry, active packaging that prolongs shelf life and reduces food losses has already been widely used [67,241]. Currently, these packaging types are still mainly based on polymers from fossil-based resources [35]. However, as described previously, several other strategies in the field of bio-based materials are under development.

Currently, active packaging solutions follow different approaches [1,242]:

- Addition of absorbers and scavengers of gases, off flavors, moisture, taints, UV light;
- Removal of catalyzing undesired food components;
- Addition of emitters/generators of gases and flavors;
- Release of antioxidant and/or antimicrobial compounds;
- Temperature controlled systems (insulting materials; self-heating or cooling).

Thus, basic barrier properties (such as oxygen or moisture) can be improved by adding active ingredients in the packaging system and/or using functionalized polymers. Polymers used in films and coatings that are inherently antimicrobial are chitosan, poly(L-lysine), calcium alginate, acrylic polymers, and sustainable active microbiocidal (SAM) polymers [11,243].

A promising technology that is presented and discussed in this chapter is the integration of antimicrobial and antioxidative substances [244,245]. Antimicrobial systems target the control or reduction of microbial growth which often results in the extension of the lag

phase or in a reduced growth rate in the exponential phase [246,247]. Different antimicrobial strategies using plant extracts are based on absorption, release, and immobilization systems [244].

There are different opportunities to implement antimicrobials in packaging materials. In terms of time-releasing killing, either a volatile or non-volatile antimicrobial agent is temporarily trapped within the backbone material and released from the polymer to the environment. Volatile antimicrobial agents are released through evaporation or diffusion into the headspace of food in most cases without direct contact. Non-volatile antimicrobial agents are released by direct contact through diffusion into the food surface [243,248]. Another approach is the permanent immobilization of a non-volatile antimicrobial agent to a polymer backbone. The integration of antimicrobial agents in packaging can be realized by direct incorporation of the antimicrobial into the packaging material or by coating the packaging material with antimicrobial agents [243,244,248].

Natural antimicrobial, antioxidative, and photostabilizing agents used for the preservation of food are bacteriocins and extracts from biomass of animals, plants, and microorganisms. These include enzymes, EOs, and natural extracts from different plant sources [249,250]. Phenolic compounds and terpenoids are antioxidative and antimicrobial agents that occur in EOs extracted from different plants. Eos are the most abundant source of bioactive compounds [251]. The effect of EOs on the microbial cells depends on different mechanisms such as disrupting the enzyme structures, damaging the phospholipid bilayer of cell membrane, and compromising the genetic makeup of microbes [98]. Antioxidants are compounds that react with free radicals, neutralizing them and thereby preventing or reducing their damaging effects. Aromatic plants are a source of natural antioxidants because of the activity of secondary metabolites such as phenylpropanoids and EOs [252]. The antioxidant capacity of plant extracts is strongly related to the phenolic content [253,254]. Due to their redox property, they can act as reducing agents, hydrogen donors, singlet oxygen quenchers, metal chelating agents, and suppressors of free radicals [159,255]. The antioxidant activity is not a property of a single phenolic compound, but it is widely distributed among the phenolic phytochemical constituents. So, anthocyans, flavonoids, phenolic acids, phenolic terpenes, and volatile oils are particularly interesting as antioxidants in food packaging [159,256].

The search for appropriate substitutional bio-based core materials is important for tackling environmental issues. Currently, those materials include, but are not limited to, chitosan, starch, CMC, PLA, whey proteins, and combinations thereof. As they represent a comparably small amount of the packaging itself when compared to core materials, additives tend to be neglected. However, they are often critical to achieve the desired properties in packaging materials and can be capable of prolonging the shelf life of both packaging materials and packed goods, leading to reduced food loss.

In the following subchapters, the review summarizes applications of plant EOs and plant extracts in the context of active food packaging.

4.1. Plant Essential Oils

Plant-based active stabilizers can be isolated by extracting the appropriate biomasses by steam distillation using various solvents. Generally, EOs are complex mixtures containing over 300 different polar and non-polar volatile organic compounds, usually of low molecular weight (below 300) at quite different concentrations [257]. Often, two or three major components exist at relatively high concentrations (20–70%) while others are present in trace amounts. The major constituents of EOs are terpenoids and phenylpropanoids which provide the characteristic aroma and biological properties. Both families are comprised of phenolic compounds. The sufficiently high vapor pressure of EOs, in general, at atmospheric pressure and room temperature causes them to be found partially in the vapor state [257–259].

In comparison to producing plant extracts, a higher amount of plant biomass must be processed to obtain EOs. However, this can prove worthwhile as EOs contain active

components of biomasses in particularly high concentrations and are therefore typically highly effective in different applications. This allows manufacturers to obtain great effects with applying only small amounts of oil. Prominent examples of EO applications for active food packaging are listed in Table 6. Results obtained so far show that plant EOs are interesting components for active food packaging. However, the corresponding materials have to be specified regarding water resistance; water vapor permeability; mechanical properties; and enhancement of antimicrobial, antioxidant, photostabilizing, and light-absorbing properties. Particular research interest lies on EOs obtained from food sources such as cinnamon, thyme, and rosemary or food production byproducts as apricot kernels and banana leaves.

Table 6. Spotlight literature for plant essential oils used in the context of active food packaging.

Biomass	Packaging Matrix	Results	Reference
Apricot kernel EO	Chitosan	Prepared films showed better water resistance and improved antioxidant, antimicrobial, and mechanical properties; fungal growth on packaged bread is inhibited	[260]
Banana leaf EO	Gelatin	Improved antimicrobial properties (against *E. coli* and *S. aureus*) of gelatin films enriched with banana leaf EO; improvements on mechanical properties observed	[261]
Bergamot, lemongrass, rosemary, and clove EOs	PLA	Enhanced mechanical and antimicrobial properties against *E. coli*	[262]
Cinnamon and ginger EOs	CMC and Chitosan	Decreased water vapor permeability (particularly for cinnamon EO), antifungal activity against *A. niger* with a higher efficacy of cinnamon EO	[263]
Cinnamon bark EO	Gelatin	Antioxidant and antimicrobial effects observed (against *S. typhimurium* and *L. monocytogenes*), water resistance is increased	[264]
Cinnamon bark EO	PLA and Sea squirt (*Halocynthia roretzi*) shell protein	Enhanced antioxidant activity and antimicrobial effects against *L. monocytogenes*, *S. aureus*, *E. coli* and *S. typhimurium*; change of mechanical properties, decrease of water solubility, and water permeability	[265]
Cinnamon EO	Chitosan and Gum arabic	Better water barrier properties with a decrease in mechanical properties; high antioxidant effect when applying appropriate ratios of chitosan, gum Arabic, and cinnamon EO	[266]
Cinnamon EO	CMC and PVA	Enhancement of antioxidant and photostabilizing properties; highly effective against *Penicillium digitatum*; shelf life of packaged bread was increased	[267]
Cinnamon EO	Gelatin	Water vapor permeability and light-absorbing properties of enriched films increase while water content and elongation at break decrease; antifungal and antimicrobial activity against *E. coli*, *S. aureus*, *A. niger*, *Rhizopus oryzae*, and *Paecilomyces varioti* observed	[268]
Cinnamon EO	Whey protein	Effect of enriched films against *S. aureus*, no effect against *E. coli* observed	[269]
Cinnamon leaf oil	Gelatin	Antimicrobial effect against foodborne pathogens reported (*E. coli*, *S. typhimurium*, *S. aureus*, *L. monocytogenes*)	[270]
Clove bud EO	Pectin	Antioxidant and antimicrobial effects observed (against *S. aureus*, *E. coli*, and *L. monocytogenes*); improved mechanical properties (flexibility, resistance to breakage, water barrier properties, and heat stability)	[271]
Clove EO	Starch	Antifungal activity against *Colletotrichum gloeosporioides* and *Colletotrichum musae*, but not against *Saccharomyces bourladii*; enhanced shelf life of packaged bananas	[272]
Eucalyptus and Cinnamon EOs	PLA and PBAT	Antimicrobial activities against *E. coli* and *S. aureus* observed for both EOs with cinnamon EO showing a higher antimicrobial effect, increased biofilm inhibition and decreased UV-light transmission	[273]

Table 6. *Cont.*

Biomass	Packaging Matrix	Results	Reference
Eucalyptus globulus EO	Chitosan	Antibacterial effects against *S. entertidis*, *E. coli*, *B. cereus*, and *S. aureus* observed (especially in liquid phase); lower antibacterial effect in vapor phase	[274]
Ginger EO and Eugenol	Gelatin and Chitosan	Antioxidant effect observed for both Eugenol and ginger EO (depending on film formulation); comparable water vapor permeability with increased elasticity	[275]
Lavender EO	Starch, Furcellaran, and Gelatin	Enhanced antioxidant and antimicrobial effects against *E. coli* and *S. aureus*; change of mechanical properties with addition of Lavender EO (decrease of tensile strength, water absorption, etc.)	[276]
Lemon EO	Starch	Optical and mechanical properties examined; antimicrobial effects against *S. aureus* and *E. coli*	[277]
Olive oil, corn oil, sunflower oil	Chitosan	Particularly olive oil enriched films showed better mechanical properties and a high antibacterial activity	[278]
Oregano EO	Whey protein	Higher amounts of EO resulted in higher water vapor permeability and film flexibility; antimicrobial activity against *Penicillium commune*	[279]
Origanum vulgare, O. majorana EOs	Chitosan	Antimicrobial effect against *S. aureus* and *B. cereus* observed with both EOs, significantly enhanced effects for *O. vulgare* EO	[280]
Pistacia atlantica EO	CMC and Gelatin	Antimicrobial effect against *E. coli*, *S. aureus*, *Clostridium sporogenes*, and particularly *Salmonella enterica*; reduction of e.g., water vapor permeability, film thickness, and tensile strength	[281]
Rosemary and mint EOs	Chitosan, Pectin, and Starch	Rosemary and mint EOs improved water barrier properties and inhibited *Bacillus subtilis*, *E. coli*, and *L. monocytogenes*; both EOs resulted in enhanced antioxidant effects	[282]
Rosmarinus officinalis, Artemisia herba-alba, Ocimum basilicum and *Mentha pulegium* EOs	Alginate	Strong antibacterial activity against *S. aureus*, *E. coli*, *Salmonella enterica*, *Enterococcus faecium*, *Klebsiella pneumoniae*, and *Enterococcus faecalis*; physical properties analyzed, antioxidant effect observed	[283]
Satureja Khuzestanica EO	Kefiran and CMC	Antimicrobial effects against *S. aureus* and *E. coli*, significant antioxidant properties, change in physical properties (e.g., decrease in water vapor permeability)	[284]
Satureja Khuzistanica Jamzad EO	Whey protein	Antimicrobial effect particularly against *S. aureus* with *Pseudomonas aeruginosa* showing the highest resistance of analyzed bacteria; increased elongation at break and water vapor permeability	[285]
Summer savory EO	CMC and Agar	Antimicrobial effects particularly against *S. aureus*, *B. cereus*, and *L. monocytogenes* with lower effects against *E. coli*; alteration of physical properties (increased water vapor permeability, improved mechanical flexibility)	[286]
Thyme and Clove EOs	PLA and PBAT	Positive properties observed for both EOs, but particularly for clove EO films, including UV-blocking and highly antimicrobial effects (inhibition of *E. coli*, complete killing of *S. aureus*)	[287]
Thyme, rosemary, and oregano EOs	PLA	Significant antioxidant effect on packaged minced fish with moderate alteration of mechanical properties	[288]

Abbreviations: Essential Oil—EO; *Escherichia coli*—*E. coli*; *Staphylococcus aureus*—*S. aureus*; *Aspergillus niger*—*A. niger*; *Salmonella typhimurium*—*S. typhimurium*; *Listeria monocytogenes*—*L. monocytogenes*; *Salmonella entertidis*—*S. entertidis*; *Bacillus cereus*—*B. cereus*.

4.2. Plant Extracts of Various Biomasses

Quality and quantity of plant extracts strongly depend on their biomass origin and type of extraction processes. The most common extracts have been obtained by conventional solvent extraction methods (infusion, decoction, digestion, maceration, and percolation) using solvents such as water, ethanol, methanol, chloroform, or dimethylsulfoxide [289–291].

Such extracts typically retard bacterial growth and can also introduce antioxidant and photoabsorbing effects. Those properties are reported for various different biomasses including (but not limited to) herbs, flowers, trees, and their fruits [292–298]. In contrast to

essential oils, the effects tend to be less extensive; however, less biomass must be processed to obtain such extracts. Furthermore, as high-concentrated essential oils provide lipophilic surroundings for active substances, the application of polar active substances is only possible by (hydrophilic) extraction, thus introducing a whole new group of active compounds. The specific characteristics observed in plant extracts can be utilized by incorporating them in food packaging materials to positively affect the packed food. Prominent examples for this approach are documented in Table 7. The studies using plant extracts show similar improvements in packaging characteristics to plant essential oils. Decreased water vapor permeability; enhanced moisture and oil resistance; improved mechanical properties; and enhanced antimicrobial, antioxidant, photoabsorbing, and UV-stability are reported. While it is challenging to directly compare the obtained data with each other due to a variety of tests and extraction methods used, most researchers claim a relevant potential exists for plant-based stabilizers in food packaging applications. Again, plant-based active packaging research is typically focused on biomasses that represents foodstuff.

Table 7. Spotlight literature for plant extracts used in the context of active food packaging.

Extracted biomass	Packaging matrix	Results	Reference
Coffee beans and de-fatted cocoa beans	Starch	Synergistic antioxidant effect, decreased water vapor permeability, increased shelf life of palm oil	[299]
Grapefruit seed extract	PCL and Chitosan	Films showed better mechanical properties and inhibited bacterial growth of *E. coli* and *P. aeruginosa* for up to 6 days; successful tests with packaged salmon and bread	[300]
Herba Lophatheri extract	Chitosan	Moisture and oil resistance are enhanced, both antioxidant and antimicrobial activities observed (*E. coli*, *S. aureus*)	[301]
Kombucha tea extract	Chitosan	Decreased water vapor permeability and improved antioxidant, photoabsorbing, and antimicrobial effects (against *E. coli* and *S. aureus*); 3 days extended shelf life for packaged minced beef	[302]
Grape seed extract	Chitosan	Enhanced antioxidant and antimicrobial activity (total mesophilic aerobic bacteria, coliforms, *E. coli*, *L. monocytogenes*, *S. aureus*, and *P. aeruginosa*; shelf-life extension of refrigerated, vacuum-packed chicken breast fillets)	[303]
Rosemary extract	Starch	Significant antioxidant effect, increased UV-stability	[304]
Propolis extract and *Zataria multiflora* EO	Chitosan	Antimicrobial effects measured for mesophilic total viable plate counts, lactic acid bacteria, psychotropic bacteria, and *Pseudomonas*; synergistic effects observed; lower microbial load on packaged chicken	[305]
Sumac extract and *Zataria multiflora* EO	Chitosan	Antioxidant effects and prolonged shelf life on packaged meat observed; antimicrobial activity against different bacteria (e.g., *Pseudomonas* spp.)	[306]
Propolis extract and *Zataria multiflora* EO	PLA	Increased shelf life of packaged sausages, antimicrobial effects against common food pathogens (*S. aureus*, *E. coli*, *Vibrio parahaemolyticus*, *L. monocytogenes*)	[307]

Abbreviations: *Escherichia coli*—*E. coli*; *Pseudomonas aeruginosa*—*P. aeruginosa*; *Staphylococcus aureus*—*S. aureus*; *Listeria monocytogenes*—*L. monocytogenes*.

4.3. Encapsulated Plant Essential Oils

Advanced methods use plant essential oils after encapsulation by a variety of different techniques, including formation of nanofibers, nanotubes, and nanoparticles [308]. This way, the essential oils are more resistant against thermal influences [309–311]. The incorporation of encapsulated essential oil typically also improves the mechanical properties of packaging materials [312,313]. Encapsulation furthermore facilitates gradual release of active ingredients, leading to a more durable protection of the packed foodstuff. Encapsulated essential oils are also under investigation in other fields and applications including bio-based insecticides and cleaning agents [314,315]. Recent studies utilizing the encapsulation of essential oils in the context of food packaging are presented in Table 8. The

results confirm that encapsulated plant oils are able to improve water vapor permeability, transparency, and tensile strength as well as antioxidant and antimicrobial effects.

Table 8. Spotlight literature for encapsulated plant oils used in the context of active food packaging.

Biomass	Packaging Matrix	Encapsulation Details	Results	Reference
Chrysanthemum EO	-	Chitosan nanofibers	Antioxidant and antimicrobial effect against *L. monocytogenes* observed e.g., on packaged beef, prolongation of shelf life possible	[316]
Cinnamon EO	PLA	Nanofibers	Better antimicrobial effect against *S. aureus* and *E. coli* observed for encapsulated EO; encapsulation process is more suitable formulation method to maintain EO properties; shelf life of packaged pork was prolonged	[310]
Clove EO	Alginate	Inclusion complex	Successful incorporation of clove EO complexes; resulting in less transparent and flexible films, decreased elasticity, increased water vapor permeability	[317]
Cumin seed oil	-	Nanoemulsion (Whey protein, Guar gum)	Antimicrobial effect of encapsulated oil against *S. aureus*, *E. coli*, and *A. flavus*	[318]
Cuminum cyminum EO	-	Chitosan nanoparticles	Significant antioxidant effect in packaged white button mushrooms observed, resulting in presumed shelf-life prolongation	[319]
Laurel EO and silver nanoparticles	PE	Liposomes in Chitosan	Antioxidant properties observed during 7 days of storage with only about 30% of EO released from liposomes; antimicrobial effect against *S. aureus* and *E. coli* results in 6 days prolonged shelf life of packaged pork	[320]
Lavandula angustofolia EO	-	Nanoemulsion (Whey protein)	Encapsulation enhanced thermal stability of EO; antibacterial effect is observed	[309]
Menthone, Oregano, Cinnamon, Lavender and Citral EOs	-	Starch nanoparticles	Enhanced stability of antioxidants against thermal influence after encapsulation; antimicrobial effects against *E. coli* and *S. aureus* are prolonged	[311]
Moringa oil	Gelatin nanofibers	Chitosan nanoparticles	High antimicrobial activity of encapsulated Moringa oil against *L. monocytogenes* and *S. aureus* for 10 days without affecting the sensory properties of packaged cheese	[321]
Oregano EO	-	PCL nanocapsules	High retention of encapsulated Rosemary EO (determined via carvacrol content) observed, suitability for long-term delivery of carvacrol can be assumed	[322]
Oregano EO	Soy protein	Microencapsulation by ionic gelation	Strong antioxidant and antimicrobial properties against *E. coli* and *S. aureus*; enhanced effects and mechanical properties with microencapsulated EO in contrast to free EOs	[312]
Rosemary EO	Starch and CMC	Chitosan nanogel	Films with encapsulated EO show higher water vapor permeability, higher transparency, and tensile strength; immediate (free EO) and gradual (encapsulated EO) antimicrobial effects against *S. aureus* were observed	[313]
Thymbra capitata EO	-	Zein nanoparticles	Both free and encapsulated EO are effective against *E. coli* and *L. monocytogenes*; presumably due to controlled release, encapsulated EO showed lower antimicrobial efficacy compared to free EO	[323]

Table 8. *Cont.*

Biomass	Packaging Matrix	Encapsulation Details	Results	Reference
Thyme EO	-	Nanofibers (Chitosan, Gelatin)	Both free and encapsulated thyme EO has antioxidant and antimicrobial effects against *Clostridium perfringens*; tests show that such nanofibers could be used to substitute nitrite in meat products	[324]
Thyme EO	Gelatin	Nanofibers	Antimicrobial effect against *Campylobacter jejuni* in packaged chicken observed	[325]
Thyme EO	Ink (for paper packaging)	Halloysite nanotubes	Strong antibacterial activity against *E. coli*, mesophilic aerobic bacteria, molds, and yeasts for up to 10 days after encapsulation in Halloysite nanotubes	[326,327]
Zataria multiflora EO	PVA	Nanofibers (Chitosan, PVA, Gelatin)	Encapsulated *Zataria multiflora* EO completely inhibited growth of *S. aureus*, *P. aeruginosa*, and *Candida albicans* for 24 h; tested material is developed for use as wound dressing	[328]

Abbreviations: Essential Oil—EO; *Listeria monocytogenes*—*L. monocytogenes*; *Staphylococcus aureus*—*S. aureus*; *Escherichia coli*—*E. coli*; *Aspergillus flavus*—*A. flavus*; *Pseudomonas aeruginosa*—*P. aeruginosa*.

5. Adoption Potential of Bio-Based (Active) Packaging along the Value Chain

Active packaging based on biopolymers is identified as the more sustainable alternative compared to conventional packaging. In addition, the integration of natural additives has positive effects on the quality and shelf life of the packaged product [39,329,330]. However, in order to be successful in the market, this innovative concept needs to be adopted along the whole agricultural food value chain [331,332].

More specifically, farmers need to collect, process, and deliver raw materials such as annual plants (e.g., miscanthus) or residues from agricultural production (e.g., sugarcane bagasse) [333]. The packaging industry must adopt renewable resources as raw materials and might also need to adjust their production processes for the application of bio-based and/or biodegradable polymers as a packaging core matrix and to integrate natural additives into the packaging materials. Food companies need to be willing to pay more for the material to pack their products, and the consumer needs to accept the concept of bio-based active packaging [334,335]. As the implementation of active packaging based on bio-based polymers entails several changes for farmers, industry, and consumers, the remaining section reviews extant studies exploring the adoption decisions of these value chain actors.

Existing literature looking at the adoption behavior of farmers finds that these value chain actors are generally skeptical towards innovations related to the bioeconomy. Therefore, monetary incentives and assistance with the novel practices and processes might be necessary [336]. In addition to farmers, food processing companies might also serve as the provider of by-products as raw materials to produce active bio-based packaging. However, there is currently a lack of research regarding the adoption decisions of managers in those companies. Today, a growing number of farmers are interested in adopting practices to valorize by-products [337]. Therefore, these farmers need to be targeted by policy initiatives and could then serve as opinion leaders to positively influence the adoption decisions' of their communities [338,339].

Focusing on industry representatives such as packaging producers and food companies, exploratory studies identify several factors driving their adoption decision. First of all, the market prices and the availability of renewable resources for the production of bio-based polymers and natural additives are relevant for the adoption decisions of packaging producers [334]. Moreover, relevant policy instruments need to be implemented to foster research and development of bio-based polymers with natural additives (e.g., subsidies) or even to ban conventional (multi-layer) plastics [14,334,340]. This would increase the competitiveness of bio-based and/or biodegradable packaging with conventional plastics [334].

However, even when policy instruments are in place, the level of consumer demand is the most important factor driving the adoption decision of industry representatives [340].

Consumer studies indicate that the final actors in the value chain have both positive and negative associations with bio-based products. They may misunderstand the concept of 'bio-based' [341–343]. However, the majority of consumers seem to believe that sustainable packaging is important and useful [344]. Moreover, results of two studies provide evidence that bio-based packaging seems to increase the preferences for the packaged product [345,346]. In fact, empirical results from a discrete choice experiment indicate that consumers are willing to pay a price premium for bio-based plastic packaging [335]. Considering the calculations by van den Oever et al., this price premium even covers the additional costs for bio-based and/or biodegradable plastics compared to conventional materials [39]. Food companies could therefore switch to bioplastic packaging without expecting any lost profits [335]. Moreover, especially those consumers with high levels of environmental awareness and innovativeness seem to prefer bio-based plastics over conventional plastic products [347–349]. Products packaged with bio-based materials thus need to be presented in retail locations which are preferred by this type of consumers such as organic stores [335]. However, as bio- and fossil-based plastic packaging are not easy to be distinguished by consumers, the packaging needs to be labelled accordingly [350].

After its use as packaging material, end-of-life solutions also need to be considered for active packaging derived from bio-based and/or biodegradable polymers from renewable resources [335]. Depending on consumers' disposal behaviors, bio-based bioplastics can be decomposed given the right conditions (in case of biodegradable and/or compostable compounds) or the material can be used to generate renewable energy. Thus, it is very important that the disposal options are clearly communicated to the consumers [351]. In fact, consumers are even willing to pay a price premium for biodegradable and recyclable packaging [352,353].

6. Conclusions

Besides current political requirements that aim to improve sustainability aspects, the development and promotion of more sustainable materials have gained more importance due to consumer interests. As customers preferences shifted to high quality and safe products with enhanced shelf life, the development of various new trends in packaging systems has arisen. Research focuses on improving the characteristics of bio-based packaging materials, in particular mechanical, thermal, and physical properties. Although bio-based polymers provide significant opportunities in terms of sustainability and biocompatibility, their use in industrial applications is often restricted due to lower performance in fundamental packaging functions. Companies are faced with a challenge of alternatives offering higher costs, limited functionality, existing infrastructure, and inconsistent legislation. Furthermore, a lack of compatibility with conventional processing technologies has to be overcome. Food companies need to be willing to pay more for the material to pack their products and the consumer needs to accept the novel concept of (active) packaging. The *proof-of-concept* is shown by a few commercially available biopolymers with food applications such as PLA, PHAs, PEF, PBS, and thermoplastic cellulose or starch-based films. In the future, the market for sustained active packaging will certainly increase due to enhanced efforts and innovations in material development and processing technologies.

List of Abbreviations

APET	Amorphous poly(ethylene terephthalate)
AZC	Ammonium zirconium carbonate
CA	Cellulose acetate
CAB	Cellulose acetate butyrate
CAP	Cellulose acetate propionate
CMC	Carboxymethyl cellulose
CoO.ZnO	Cobalt(II) oxide/zinc oxide
EO	Essential oil
GHG	Greenhouse gas
HDPE	High density poly(ethylene)
HLCNC	High lignin-containing cellulose nanocrystals
HPMC	Hydroxypropylmethyl cellulose
LCA	Life cycle analysis
LDPE	Low density poly(ethelene)
PA	Poly(amide)
PBAT	Poly(butylene adipate terephthalate)
PBS	Poly(butylene succinate)
PCL	Poly(caprolactone)
PE	Poly(ethylene)
PHA	Poly(hydroxyalkanoate)
PHB	Poly(3-hydroxybutyrate)
PEF	Poly(ethylene furanoate)
PET	Poly(ethylene terephthalate)
PHBHHx	Poly(3-hydroxybutyrate-co-3-hydroxyhexanoate)
PHBV	Poly(3-hydroxybutyrate-co-3-hydroxyvalerate)
PLA	Poly(lactic acid)
PP	Poly(propylene)
PS	Poly(styrene)
PTT	Poly(trimethylene terephthalate)
PVA	Poly(vinyl alcohol)
PVC	Poly(vinyl chloride) (PVC)
SA	Sodium alginate
SAM	Sustainable active microbiocidal (SAM)
SiO_2	Silicon dioxide
TAIC	Triallyl isocyanurate
WVP	Water vapor permeability

Glossary

Active packaging	Materials designed to deliberately incorporated components that would release or absorb substances into or from the packaged food or the environment surrounding the food.
Bioactive	Compound that has an effect on a living organism, tissue/cell.
Bio-based	Compound that is composed (in whole or in significant part) of biological products or renewable domestic agricultural or forestry materials (including plant, animal, and marine materials).
Biodegradable	Degradability achieved via microorganisms.

Bioplastics	Plastics that are either bio-base, biodegradable, or features both properties.
Biopolymers	Natural polymers produced by the cells of living organisms (e.g., forestry and agricultural crops, terrestrial and marine animals), examples are polysaccharides, proteins, and lignin.
Compostable	Compounds approved to be degradable by microorganisms at defined conditions (e.g., temperature, humidity, time).
Edible packing	Compounds approved to be metabolized by humans.
End-of-life options	Including re-use, recycling, recovery, disposal, and others (such as littering, ingestion).
European Green Deal	Action plan to boost the efficient use of resources by moving to a clean, circular economy, restore biodiversity, and cut pollution.
Fossil-based	Compounds obtained from crude oil, natural gas, brown or hard coal.
Renewable resource	Resource which will replenish to replace the portion depleted by usage and consumption, either through natural reproduction or other recurring processes in a finite amount of time in a human time scale.

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