REVIEW

Recent advances in polylactic acid biopolymer films used in food packaging systems

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Summary

The main purpose of food packaging is to maintain food quality and safety. For this purpose, packaging materials harmless to environment and health should prevent or reduce food spoilage. The packaging industry tends to natural and biodegradable materials due to migration of harmful components and causing environmental pollution of synthetic materials. The use of biodegradable materials as food packaging is one of the most studied approaches in recent years. Biodegradable polymers can be categorized in three groups as extracted from the biomass, produced by microorganisms and obtained from chemical synthesis. Polylactic acid (PLA) has unique properties as good transparency, availability in the market and low environmental impact, unlike fossil-derived materials. Compared to synthetic and other biodegradable polymers, PLA films have good tensile strength, as well as good barrier properties against flavour and smell. However, in order to solve disadvantages such as fragility and low thermal stability, plasticizers, stabilizers, fillers and antioxidants are used in the structure. In this review, it was aimed to present the latest advances in the functional properties of PLA important for its use in food packaging, namely, barrier, mechanical, thermal and optical properties, together with degradability.

Keywords

polylactic acid; biopolymer; food packaging; bio-based packaging

Packaging is one of the most important elements in marketing of a food product. This is very important not only because it contains products, but also it protects products from pollution, environmental damage and makes it easier to transport and store them. Currently, traditional plastic materials (like petroleum-derived polymers polyethylene (PE), polyethylene terephthalate (PET), polypropylene (PP) and polystyrene (PS) represent the main food packaging materials. The use of plastics is increasing every year and is estimated to reach 33 gigatonnes in 2050. These materials show useful properties such as flexibility, mechanical durability, stability, low weight, excellent barrier properties and low price. However, non-biodegradable materials cause environmental pollution. Recently, the preference for natural and biodegradable materials in food packaging products is increasing [1, 2]. Application of renewable polymer sources as food packaging are also increasing. According to the Sustainable packaging alliance (SPA, Melbourne, Australia), for a packaging to be sustainable, it should be impressive in terms of cost and functionality for users, should be resource- and energy-efficient, should be turned continuously through natural or industrial systems so that material degradation can be minimized, should be recyclable, should be environmentally friendly and non-toxic, must not pose any risk to humans and the ecosystem [3]. In addition, in order to make a logical choice of food packaging between bio-based and fossil-based materials, the environmental effects of these polymer materials should be compared and, for this purpose, life-cycle assessment (LCA) is one of the tools available. LCA is a specially developed system for assessing the environmental load of a packaging material and the cycle used for the production, use and disposal of this material [4].

In this review, the main emphasis is put on the fact that biodegradable polymers attract scientific and technological attention around the world with

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their reduction of the waste problem, are used in renewable packaging and do not harm the environment. Especially in the family of biopolymers, polylactic acid seems to be one of the most notable in use as a food packaging material. The fact that it can be obtained from cheap and natural sources, has high mechanical strength compared to other biopolymers and is easy to process, were the main research topics of this review.

Classification of biopolymers

Biodegradable polymers are classified in three basic groups according to their origin and production [5, 6] (Fig. 1).

The first group are biopolymers that are extracted from the biomass, such as some polysaccharides (wheat starch, potato starch), cellulose derivatives (straws, wood) or proteins (casein, whey protein, gelatin, gluten), pectins (chitosan or chitin, gums) or lipids (fatty acids, waxes).

Starch exhibits several disadvantages like stronger hydrophilic behaviour together with weaker barrier and mechanical features than other bioplastics [7]. This limits its utilization in industrial applications. However, starch-based polymers are used together with other polymers to create a wide variety of structures.

Cellulose is one of the most commonly found biopolymers in nature but, because it is not thermoplastic, it is difficult to use it in machine process applications [8]. Compared to plant cellulose, bacterial cellulose is purer and has higher crystallinity, better barrier and mechanical properties, together with higher water-holding capacity but its usage is still limited due to high price [9].

A natural amino polysaccharide chitosan, chemical or biologically derived from chitin and used as food packaging, has various advantages such as being non-toxic, biodegradable, film-forming and antibacterial. Despite all these desired properties, due to its low thermal stability and mechanical strength as well as high water vapour and gas permeability, chitosan applications are limited [10].

Pectins are a class of polysaccharides found in cell walls and intercellular parts of plants and fruits. Pectins are anionic, amorphous, non-toxic and easily soluble in water. The barrier and mechanical properties of pectin-derived films are good but show weak resistance to moisture, low elongation and a highly fragile structure [11].

Alginate, derived from brown algae, is a low-cost, non-toxic, biodegradable polymer. It is used in film production due to its advantageous features such as gel generation, stabilizing and thickening properties. However, these biofilms have a fragile structure, so plasticizing material needs to be added to improve the flexibility of the films [12].

Second group of polymers are produced by microorganisms such as polyhydroxyalkanoates (PHA), polyhydroxybutyrate (PHB) or polyhydroxybutyrate-*co*-hydroxyvalerate (PHBV), and others such as xanthan, bacterial cellulose or pullulan.

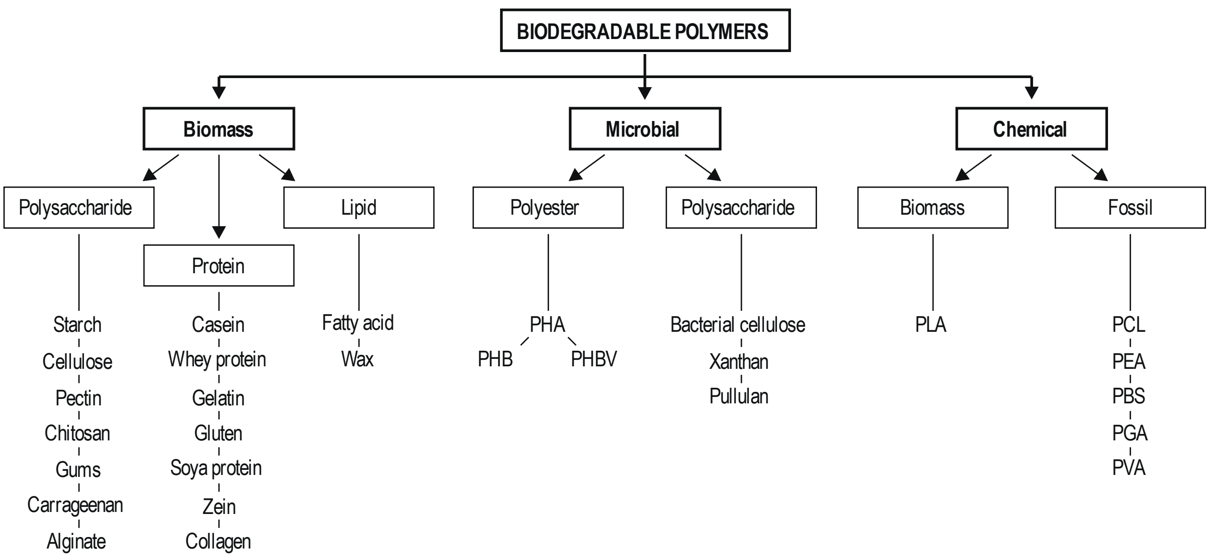

Fig. 1. Grouping of biodegradable polymers.

PHA – polyhydroxyalkanoate, PHB – polyhydroxybutyrate, PHBV – polyhydroxybutyrate-*co*-hydroxyvalerate, PLA – polylactic acid, PCL – poly-ε-caprolactone, PEA – polyesteramides, PBS – polybutylene succinate, PGA – polyglycolic acid, PVA – poly(vinyl) alcohol.

Among these polymers, the PHA polymer is obtained by microorganisms [13]. Short-chain PHA show semi-crystalline, hard and brittle properties, on the other hand, those with a medium chain length show elastomeric properties [14]. The most commonly used PHA polymer is poly-3-hydroxybutyrate (PHB). This polymer has similar properties as polyethylene and polypropylene, making it a suitable alternative to petroleum-derived biodegradable polymers [15].

Biopolymers from the last group are obtained by chemical synthesis from compounds such as polylactic acid (PLA) and from fossil resources such as poly-ε-caprolactone (PCL), polyesteramides (PEA), polybutylene succinate (PBS), polyglycolic acid (PGA) and poly(vinyl) alcohol (PVA). PCL is a biodegradable packaging material, having good barrier and antimicrobial properties. However, it shows a weak structural and functional stability, which impedes its commercial value [16]. PGA is similar to PLA except the methyl side group. It shows mechanical hardness, good gas barrier properties and high thermal stability. PGA is often used in copolymers like poly(lactic-*co*-glycolic acid (PLGA) [17]. Pullulan is a polysaccharide that can be dissolved in water. Pullulan-derived films are odourless, tasteless, highly transparent and have great oxygen barrier properties. The functional properties of these films can be improved by using biopolymers like gums [18]. Another bioplastic is poly(vinyl) alcohol (PVA), which shows good oxygen barrier, physical and optical properties. However, low water vapour permeability, biodegradation and high price limit its use as a food packaging material [19].

The biopolymers most studied by researchers as biodegradable food packaging materials are PLA nanocomposites. PLA is safe for all food packaging applications and is declared to be generally recognized as safe (GRAS) by the United States Food and Drug Administration (FDA) [20]. PLA is an aliphatic thermoplastic (100% bio-based) and it is obtained from lactic acid (LA) with an optical purity of approximately 98–99 % [21]. The crystallization behaviour of PLA is one of the most important parameters determining the rate its biodegradation [22]. In the first stage for the production of PLA, maize or other carbohydrate-derived food is converted to glucose and then converted into lactic acid by fermentation. In the later stage, PLA is produced through the ring-opening polymerization of lactide, the dimer of lactic acid (Fig. 2). PLA has polymers of various molecular weight and only high molecular weight ones can be used in the packaging field. PLA is one of the most widely used because it provides many positive properties to the packaging materials. One of the most desirable features of PLA compared to other hydrocarbon-containing polymers is the reduction of CO_2 emission [23, 24]. High transparency, good printing property, no leakage of the packaging material at low temperature, availability in the market and low price make PLA advantageous. PLA films have good tensile strength compared to other polymers on the market, as well as good barrier properties against flavour and smell [25]. PLA has a high modulus and strength, thermoplastic-type processability as wll as grease and oil resistance. PLA has a good potential to obtain microparticles and nanoparticles, because the size and shape of particles can be changed as desired [26]. PLA is used as a functional biodegradable material in some food packaging applications, but some features are insufficient. The weaknesses of PLA are weak gas-barrier features, low flexibility, low toughness and weak thermal stability. Many researchers are investigating methods of overcoming these limitations by modifying PLA with various polymers. Its brittle structure may also limit applications of PLA. However, with the use of nanotechnology and the production of safer PLA nanocomposites, most of its weaknesses will potentially be solved compared to petrochemical-derived polymers [24]. It makes PLA usable for flexible packaging applications, thus increasing the ductility. These conditions should use low molecular weight plasticizers, like poly(ethylene glycol) (PEG) [27]. PLA is used commercially in food packaging applications but perhaps the most important limitation of this polymer is current price, which is still more expensive than traditional plastic materials. Because of this, it can be used in high-priced foods.

To overcome these drawbacks, a number of additives must be added to the structure. When selecting additives used in the enhancement of biodegradable food packaging materials, environmental compatibility as well as safety should be considered. Incompatibility may also occur due to the different polarities of the polymers with the additives used. Therefore, various methods are preferred to ensure that the additives to be used are distributed uniformly in the structure and form a strong bond with the polymer chain. Additives based on maleic anhydride (MA), called harmonizers, are used widely due to their good performance. Materials prepared in this way guarantee that additives remain permanently in the polymer for a long time [28].

Plasticizers

Various plasticizers are mainly added to biode-

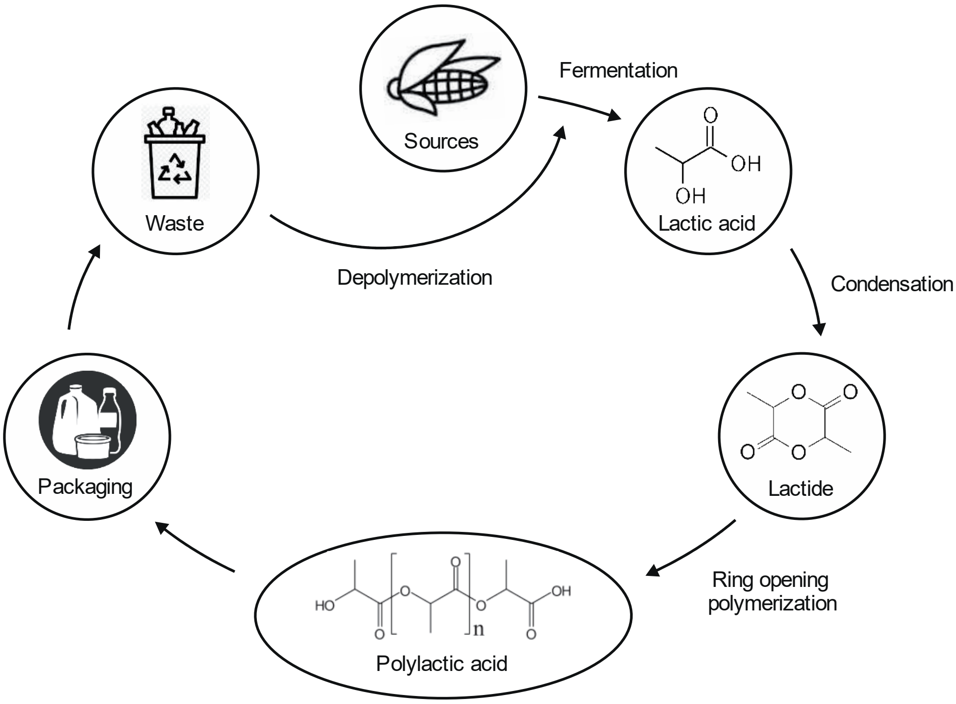

Fig. 2. Synthetic pathway of polylactic acid.

gradable films, as films prepared only with bioplastics become difficult to process if they are brittle and have poor mechanical or barrier features. Plasticizers are small molecules that react with the chains of biopolymers, weakening the internal hydrogen bond, increasing the flexibility of the produced films and having low volatility. They also make it easier to remove the film from the forming mold. They also influence the sorption and barrier properties of films, supporting their packaging properties [29]. Adding plasticizers to the structure increases the gas permeability of PLA. D-limonene, a natural terpene, improved crystallinity of PLA when added to the structure as a plasticizer [30]. Besides, adding plasticizer to structure increases volume of PLA matrix, resulting in a decrease in oxygen barrier features [30]. In a study by Liu et al. [31], poly-1,4-butylene adipate (PBA), used as a plasticizer, improved mechanical features of PLA. Before the addition of plasticizer, the breakpoint value of pure PLA at elongation at break was 7.9 %, while addition of PBA increased it to 74.4 %. Thus, the elongation capacity of the material increased.

Stabilizers

The ultraviolet (UV) region of light corresponds to a wavelength area ranging from 200 nm to 380 nm. UV light has a higher energy than visible light (380–780 nm). UV rays have more energy than visible light because they have a shorter wavelength, which causes higher photochemical oxidation than other wavelength light fields. When PLA is exposed to UV light, it easily undergoes oxidative degradation, causing the backbone of PLA to weaken, resulting in differences in the colour of the polymer. Effective stabilizers absorb UV light to eliminate damage to light-sensitive packaged foods [32]. Examples of UV stabilizers can be given as follows: it may require additives to block UV light transmission in dairy products. With the use of PLA, it is possible to preserve the taste and appearance, extend the shelf life and improve the quality of the product [31]. By coating Ricotta cheeses with methylcellulose, PEG, chitosan based nanoparticles in chitosan matrix, high UV protection activity was detected [33].

Fillers

The filler to be used in food packaging must be compatible with food in storage conditions and be non-toxic. It is expected that inorganic fillers that are able to join the structure for use in packaging materials will not lead to agglomeration, reduction of transmittance in the visible light region, discrepancy with the polymer structure and acceleration of degradation of the polymer under UV exposure [19]. Bio-nanocomposites are bio-derived polymers consisting of two basic elements: biopolymers and nanofiller agents (such as montmorillonite, TiO_2, ZnO, AgO, SiO_2, in the particle range of 1–100 nm). The nanofillers used in conjunction with the biopolymer matrix and their functional properties are shown in Fig. 3 [34]. For improvement of mechanical, barrier and thermal features of biodegradable packaging materials (reinforcing) fillers like calcium sulphate, cellulose nanoparticles, lignocellulosic tissues, calcium phosphates, graphite and silica nanoparticles are added to the structure. It was stated that integrating a small amount of layered silicate (nanoclay) as a natural filler to polymers enhances these properties. All these develpoments are a result of the nanometric size of the clay layers used to increase the surface of the filler. Thus, the contact between the filler and the matrix increases and the elements of the material to be improved become more harmonic with each other. The addition of nanofillers to the biopolymer provides a very good interface with the hydrogen bond between the nanofillers and the polymer. The packaging material prepared by adding montmorillonite nanocomposite to the PLA film has good barrier properties [35]. Natural fibres can also be used as fillers to increase the barrier and mechanical features of bioplastic materials.

Antioxidants

One of the main causes of spoilage of food products, which affects both sensory and nutritional value, is autooxidation of lipids. By adding natural antioxidant substances to packaging materials, the effects of oxidation reaction are reduced and stability of oxidation-sensitive food products is increased. The antioxidant activities of these components are due to their redox properties, which play a role in neutralizing free radicals, extinguishing single and triple oxygen or decomposing peroxides. As a result of this, an increase in storage stability is observed as antioxidant capacity increases [36, 37]. In one study, angelica essential oil (it is a plant essential oil which is an effective antioxidant to reduce lipid oxidation) was added to the PLA structure in order to develop a biodegradable packaging film with antioxidant features. The films containing this oil were found to show significant antioxidant activity by 49.4 % inhibiting the 2,2'-diphenyl-1-picrylhydrazyl (DPPH) radical [36]. In a study in which PLA-curcumin film was prepared for use as a food packaging material, it was noted that the films showed good antioxidant activity in proportion to curcumin content. The scavenging activity of pure PLA film by DPPH and 2,2'-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid (ABTS) was found to be of 1.8 % and 3.1 %, respectively, while with the addition of curcumin (1.5 %, w/w) increased it to 76.6 % and 94.7 %, respectively. The antioxidant capacity of PLA-curcumin layer was lower than curcumin, which is thought to be due to the limited release of curcumin by the polymer [38].

Functional properties of polylactic acid films

Results of some studies on the use of PLA as a food packaging material are summarized in Tab. 1 [25, 27, 32, 39–56].

Barrier properties

Plastics in general show a relatively permeable structure for small and suspended molecules such as water vapour, gas, organic liquid and

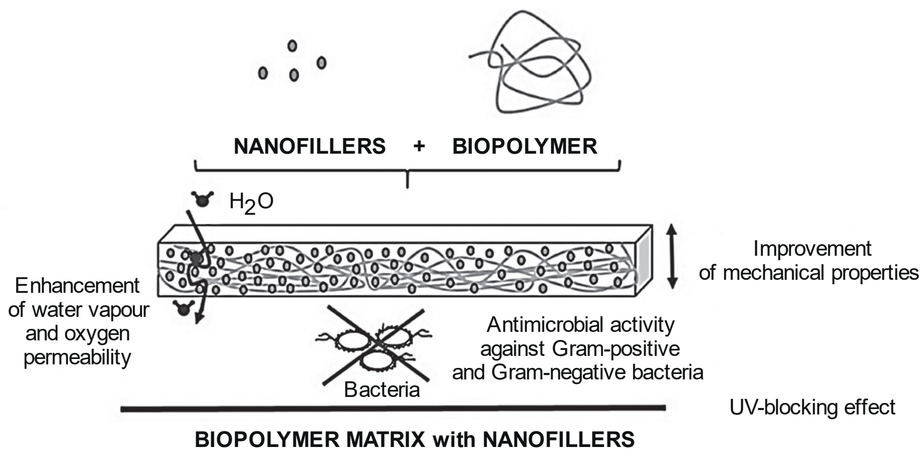

Fig. 3. Illustration of nanocomposite films and their functional properties [34].

Tab. 1. Studies on the use of polylactic acid for food packaging.

Biopolymers	Modifiers	Application area	Functional properties	Ref.
PLA		Red meat (beef steak)	Decrease amount of volatile compounds associated to off-flavours Maintain quality of meat along shelf-life Improvement of optimum red colour	[25]
PLA	Polyethylene glycol Poly(lactide-ethylene glycol-lactide) (as triblock copolymer)		Reduction of glass transition temperature Free volume fraction significantly improved for 15% polyethylene glycol	[27]
PLA	Diatomaceous earth (as reinforcing filler) Coffee ground extract (as oxygen scavenger)		Improvement of oxygen barrier and mechanical properties The use of diatomite and coffee grounds extract improved oxygen barrier properties and reduced value of oxygen transmission rate	[39]
PLA	ZnO nanoparticles (as nanofiller)	Minced fish cake	Improvement of water vapour permeability (by up to ~30.5 %), tensile strength (by up to 37.5 %) and UV-light barrier properties Improvement of antibacterial activity against *Escherichia coli* and *Listeria monocytogenes* Reduction of thermal persistence of composite films compared with PLA film	[40]
PLA	MgO nanoparticles (as nanofiller) Polyethylene glycol		Improvement of elongation at break (by up to 760 %) Improvement of antibacterial activity (death of ~47 % bacteria after 24 h) and optical properties	[41]
PLA	Maleic anhydride and dicumyl peroxide (as an initiator) Polyvinylchloride (matrix)		Improvement of processability by decreasing the temperature of mixing Improvement of mechanical properties (high level of compatibility) PLA exerted a stabilizing effect on thermal degradation	[42]
PLA	Thymol (as antibacterial)	Apple pieces	Improvement of storage stability and smooth surface over a wide range of pH Increasing of antimicrobial properties against *Escherichia coli* Enhancement of encapsulation efficiency (60.3 ± 8.0 %)	[43]
PLA	Organo-modified montmorillonite		Improvement of barrier properties preserving the materials ductility Reduction in oxygen transmission rate (~ 25 %) Improvement of thermal stability	[44]
PLA	ZnO:Cu Ag nanoparticles	Food simulant	Increase of the degree of crystallinity Improvement of thermal and mechanical properties Suitable barrier features against water vapour, UV light, carbon dioxide and oxygen Improvement of antibacterial activity against *Pseudomonas aeruginosa* but not efficient against *Staphylococcus aureus*	[45]
PLA	ZnO nanoparticles		Improvement of mechanical properties (elongation at break 74.9 ± 0.6 %, tensile strength 6.85 ± 0.19 MPa) Improvement of antimicrobial activity against *Escherichia coli* and *Staphylococcus aureus* Improvement of thermal and UV barrier properties of the composite membrane	[46]

Biopolymers	Modifiers	Application area	Functional properties	Ref.
PLA blends*			Improvement of processability and ductility Reduction of the glass transition temperature Improvement of plasticizing effect Enhancement of free-standing, homogenous and amorphous morphology Efficient biodegradation rates over a short period of time under marine conditions	[47]
PLA	Silicon oxide (as nanofiller)	Food simulant	Reducing the corrosion and contaminant migration (the final migration was reduced by 23.8–36.9 %) Increased the water contact angle from 85.5 ± 0.4° to 101.8 ± 1.2° Decreased of oxygen transmission rate 48.5 % and water vapour transmission rate 28.5 % Preserving the transparency and shape of the films	[48]
PLA banana fibre		Coffee jar lid	40 % banana fibre lid performed ecologically better than PLA and better regarding photo-chemical oxidant formation, climate change and fossil depletion	[49]
PLA	Chlorine dioxide (as disinfectant) Glucono delta-lactone (as acidifier)	Fresh tomatoes	Conservation of chlorine dioxide release and sensory properties of tomato (after 4 weeks of storage) Improvement of antimicrobial activity against *Salmonella* and *Escherichia coli* O157:H7	[50]
PLA	Halloysite (as nanofiller)	Cherry tomatoes	Extended shelf-life Improvement of mechanical, thermal, and barrier properties	[51]
PLA	Selenium (as nanofiller)		Exhibited higher antimicrobial activity against *Escherichia coli* and *Staphylococcus aureus* Improvement of mechanical properties (elongation at break 181.7 ± 20.5 %, tensile strength 11.5 ± 0.8 MPa) Improvement of UV barrier properties	[52]
PLA-P3,4HB	Angelica essential oil (as antioxidant)	Peach	Improvement of gas and water vapour permeability Enhanced the shelf-life more than 15 days Improvement of CO_2 and O_2 transmission ratio and of water vapour permeability	[36]
PLA-PHB			Improvement of barrier properties (water vapour transmission rate 4.30×10^{-8} g m·m^{-2}s^{-1}Pa^{-1}, oxygen transmission rate 6.51 cm^{3}·m^{-2}d^{-1} 0.1 MPa)	[53]
PLA			Improvement of barrier properties (water vapour transmission rate 28.95×10^{-12} kg m·m^{-2}s^{-1}, oxygen permeability $(1.96–2.94) \times 10^{-18}$ kg m·m^{-2}s^{-1}Pa^{-1})	[54]
PLA			Improvement of mechanical properties (tensile strength 21 MPa, elongation at break 2.5 %, Young's modulus 0.35 GPa)	[55]
PLA			Improvement of thermal properties (melting temperature 130–180 °C, glass transition temperature 40–70 °C)	[56]
PLA			Improvement of optical properties Light transmission rate: < 5 % at 190–220 nm, 85 % at 225–250 nm, 95 % at > 300 nm Colour values: *L** 90.64 ± 0.21, *a** –0.99 ± 0.01, *b** –0.50 ± 0.04	[32]

PLA – polylactic acid, P3,4HB – poly(3-hydroxybutyrate-4-hydroxybutyrate), PHB – polyhydroxybutyrate.
* – blend of poly(L-lactic acid) and star-poly(DL-lactic acid)

steam. Therefore, the mass transfer range also varies according to the barrier capability of the packaging material. Permeation of vapour and gas from the surroundings to packaging materials and therefore to foods reduces the shelf life due to oxidation or microbial growth [57]. An ideal food packaging material should have a positive effect on protecting the food and also it should show water resistance and oxygen barrier properties. Insufficient barrier properties cause packaged food product to be impervious to outer factors such as temperature, water, air and humidity. Barrier properties to be considered in packaging studies regard oxygen and water humidity, because these components can move from the indoor or outdoor surroundings to the packaging tissue, resulting in adverse changes in product quality during shelf life [58]. The protective capacity is directly connected to the oxygen transmission rate (*OTR*) of the material. According to the character of the packaging material, there may be oxygen permeability from food to the environment or from the environment to food. This can lead to degradation reactions leading to oil and fat rancidity in foods, oxidation-induced vitamin loss or enzymatic browning [29]. Oxygen diffusion can be expressed by detection of *OTR* per packing material thickness. *OTR* is defined as the amount of oxygen gas that passes through a substance over a specific period (*OTR·e*, where *e* is thickness, in millimetres). In the polymeric structure, permeating oxygen molecules are pushed to move randomly around particles, resulting in a circuitous path. In this way, *OTR* is reduced and gas barrier property is provided [59].

Another expression describing the oxyen barrier property is the oxygen permeability coefficient (*OPC*). *OPC* refers to the amount of oxygen that acts on a unit area of material in one second (expressed as kilograms per square meter per second per pascals) [60]. When *OPC* of a packaging film is low, the oxygen pressure inside the packaging substance and the shelf life of the product is long. In a study comparing PLA, polystyrene (PS) and polyethylene terephthalate (PET) plastics, it was found that the *OPC* value of PS was higher than PLA, while PET showed the lowest value [61]. In a pilot study, beef steaks were packed with PLA film, while amorphous polyethylene terephthalate/polyethylene (APET/PET) plates and polyvinyl chloride (PVC) plastic film were used as reference packages for comparison. Using the packaging material combined with PLA, it was possible to maintain the optimum red colour longer than reference packaged and, with this, the amount of volatile ingredients responsible for off-flavour formation linked to oxidation was reduced [25]. Two ways to provide a physical barrier to oxygen permeability in food packaging and thus maintain freshness of foods are ranked: preference of antioxidant substances with the role of oxygen scavenger or addition of particles, fibres and plates [62]. Furthermore, polymer-based materials used in food studies are flexible or semi-rigid systems. More complex multilayer systems have been introduced to increase the barrier properties of these systems [63].

It is necessary to keep the moisture content of the package that is removed during the transfer of water vapour at the lowest level. The moisture barrier property is usually defined by water vapour transmission rate (*WVTR*), which is the amount of water vapour penetrating per unit of area and time through the packaging. The water vapour permeability coefficient (*WVPC*) and transmission rate are related concepts. *WVPC* is calculated as in Eq. 1. [57].

$$WVPC = WVTR \times \frac{x}{\Delta p} \quad (1)$$

where *WVPC* is expressed in kilograms per square milimeter per pascal, *WVTR* is expressed in gram per square meter per 24 h, x is thickness of film in millimetres and Δp is difference in partial pressure expressed in pascals.

Standard methods adopted industrially (such as ASTM E987 [64]) are used to determine the *WVTR*. Determination of *WVTR* values is an important property in calculating shelf life of packaged foods because the chemical and physical degradation is directly influenced by the moisture amount in the environment [65]. In packaging used to transport fresh food products exposed to moist conditions, it is essential to improve the water vapour permeability (*WVP*) property of the material. In a study investigating applicability of soya protein-PLA bilayer film as food packaging, it was found that the PLA layer greatly increased the barrier features of the films compared to pure soya protein. Because the PLA structure reduces the affinity of the material to water, a significant decrease in *WVP* was noticed along with the increase in the PLA content [66]. In another study on multilayer film containing PLA and gelatin, it was reported that PLA layer had a favourable effect on barrier features and reduced *WVP* of the multilayer film [67].

In addition to *WVTR* and *OTR*, another essential property in determining the barrier features of biodegradable packagings is the rate of carbon dioxide transmission (CO_2TR). CO_2TR defines the proportion of carbon dioxide that passes from food to the packaging material or from the packaging material to food per unit of area per second. This is indicated by kilogram per square milimeter per second per pascals units. However, PLA films show selective permeability against three gases, namely O_2, CO_2 and H_2O, which are important for fresh product packaging such as fruits and vegetables [68]. Foods containing excess CO_2, such as fermented foods and beverages, can impair the integrity of packaging and the quality of packaged foods. However, presence of CO_2 in other foods is important because it inhibits microbial growth in the food contained in the package [30]. With improved biodegradable equilibrium modified atmosphere packaging (EMAP) for fresh fruits and vegetables using PLA films, it was found that *WVP* ($1.4 \pm 0.3 \times 10^{-9}$ kg m·m^{-2}s^{-1}) was much higher than that of CO_2 ($4.4 \pm 0.1 \times 10^{-12}$ kg m·m^{-2}s^{-1}) and O_2 ($5.3 \pm 0.4 \times 10^{-12}$ kg m·m^{-2}s^{-1}), 1 000-fold higher, due to PLA being hydrophilic [69]. In addition, some features of plastic films like transparency, oxygen permeability and *WVP* can be improved by processes called stretching or orientation. Controlled stretching of the films at certain temperatures is provided by the orientation process, thus increasing the resistance of the films to tear and puncture [70].

Mechanical properties

Polymeric packaging materials protect food during storage and transport making it resistant to mechanical stress. Mechanical properties are important to describe the mechanical behaviour and internal structure properties of packaging materials. To use biodegradable packaging materials in food packaging applications, sufficient strength, impact strength, hardness, ductility and other mechanical features need to be ensured. It is necessary to choose materials with good mechanical properties that can withstand the pressure of external factors [68].

Flexibility is an important feature of packaging materials for developing a flexible packaging structure, providing resistance to frequent bending. The mechanical features vary depending on the chain hardness, molecular symmetry and intermolecular forces of individual polymers used in the mixture [71]. The mechanical features of PLA change with the contents of L- and D-isomers in the structure and the processes applied to it [61]. Tensile tests are applied to determine the tensile feature of plastic materials. Conducting these tests is important for measuring the threshold force required to break the material. These tests show how much tensile stress a plastic substance can withstand when exposed to controlled conditions [30]. Towing the comprehensive analysis of tensile elongation (expressed in percent), tensile modulus (in gigapascals), tensile stress (in megapascals), tensile elongation at breakpoint (in percent), tensile strain (in megapascals) and tensile elongation at yield point (in percent) in ASTM D882 standard test is determined by using these parameters [68].

Tensile strength (*TS*) is a basic property to consider the strength of a packaging material. *TS* is an indicator of the capability of the substrate to endure loads in the plane of the layer. When considered in conjunction with elongation, it determines the capability of the substrate to absorb energy before failure. *TS* varies depending on the surface area and length, the bonding strength and strength of the fibres. Pure PLA has a very high module value (~1 855 MPa), *TS* of approximately 60 MPa and has a rigid structure. The addition of 7% coconut oil to PLA resulted in *TS* of the mixture to be reduced to approximately 41 MPa. This reduction resulted in less interfacial adhesion between the two phases [72]. In a study examining films containing propolis extract and PLA for meat packaging, it was observed that the best mechanical features had PLA film. It was been stated that *TS* of the control PLA film was 27.28 MPa and this value was the highest among the samples. The addition of PEG and $CaCO_3$ to the PLA film caused a serious decrease in the *TS* value (18.11 MPa) [73].

Elongation is defined as a measure of how much the substrate can stretch before breaking. The unit is specified by a percentage of the sample length. The total elongation of matter, or how much energy it can absorb (i. e. flexibility), is related to the natural structure of the material. Films prepared for use in food packaging are subjected to elongation during the packaging process, observing the elastic modulus and elongation characteristics of the film. Stressed films must be of minimum hardness to prevent puncture during transport and life cycle [30]. Elongation at break is a concept that refers to brittleness of a film. In a study that investigated the stability of packaging films containing PLA, the effect of moisture and temperature on elongation at break was found to be lower than tensile strength. No major change at 5 °C and only the highest moisture content (98% relative humidity) affected elongation at 25 °C. After 91 days of storage at 98% relative humidity, elongation rate decreased by approximately 40 % and with this limit, the material became more brittle [74]. Packaging films used in food industry must be durable so that they do not break during application. According to a study of PLA/ZnO films, *TS* value increased while the elongation at break value decreased with the increase in ZnO content. It was mentioned that the reason for this phenomenon may be related to uneven distribution and aggregation or may be that the nanoparticles limit the ductility of the PLA structure [75]. Young's modulus is a critical feature for rigid packaging applications. In the study by Holm et al. [74] mentioned above, Young's modulus dropped slightly under 25 °C storage conditions at the highest humidity, while no reduction was observed at 5 °C. Therefore, hardness of the film was found to be virtually unaffected by humidity and environment temperature. In this aspect, the properties of PLA appear to be similar to those of PS because PLA has a high Young's modulus of approximately 3 GPa and a low value of elongation at break of approximately 2–5 %. Some of the components added to develop the different characteristic of the PLA film have a positive effect on the mechanical properties, while others have a negative effect. For example, in a study in which a layer of SiO_X was added to the poly-L-lactic acid (PLLA) film to produce a layered packaging for chilled meat, it was found that tensile strength and Young's modulus values increased by 91.6 % and 119.2 %, respectively, compared to pure PLLA film [76]. On the other hand, a study conducted by Righetti et al. [77] described the mechanical properties of biocomposite obtained by adding potato pulp powder to the PLA layer and observed a drop in *TS*, elongation at break and elastic modulus with raising the percentage of potato pulp powder, and it was noted that the material suffered from a loss of mechanical quality compared to pure PLA. While PLA generally has higher modulus and lower elongation values, starch-derived biopolymers had higher elongation and lower modulus.

Thermal properties

Conventionally in the process of melting polymers, biopolymers are subjected to high temperatures. Therefore, biopolymers must have thermal stability in order to avoid degradation and maintain their properties. Glass transition temperature (*Tg*) is one of the properties that best shows the task of water in food. *Tg* of bioplastics significantly affects the thermomechanical features and the chemical and physical stability of foods. In addition, the plasticizing effect of a component is also associated with *Tg*. The plasticizing effect is generally defined by the function of *Tg* depending on the volume part or weight of water. The flexibility of amorphous biopolymers is significantly decreased when cooled below *Tg*. At temperatures under *Tg*, segmental motion does not appear and a change in its size in the polymer leads to temporary deterioration of the primary valence bonds. Amorphous plastics (like PLA) show best performance below *Tg*, but elastomers should be used under the brittle point [78].

Melting point (*Tm*) is another parameter that determines thermal properties of biopolymers. *Tm* is estimated as the temperature at endothermic peak. Polymers with a lower *Tm* have higher melting stability, which expands the processing capacity of the polymer. Because *Tm* of PLA is low, it is not appropriate for use in heat-treated packaging [24]. In a study conducted by Auras et al. [79], two types of PLA films (resins nominally containing 98 % L-lactic acid and 94 % L-lactic acid) were used and the produced films were compared with PET and PS. Both forms of PLA were found to have lower *Tg* and *Tm* than PS and PET. This indicated that PLA was a suitable packaging material for heat sealing and heat treatment.

Optical properties

Optical properties of food-covering materials are critical in terms of acceptance by the consumer, general appearance and visualization of the product. The transmission rates of UV and visible light are two major elements in food packaging design to keep products throughout their shelf life. UV-Vis spectroscopy is used to be able to characterize optical charasteristic like transparency. Film samples are scanned from 200 nm to 800 nm with the help of a spectrophotometer to determine UV and visible light barrier properties. Opacity or transparency (*O*) is a critical physical feature for packaging materials regarding light transmission [80]. *O* is determined by the following equation:

$$O = \frac{A_{600}}{T} \qquad (2)$$

where A_{600} is absorbance at 600 nm and *T* is thickness (expressed in milimetres).

In a study investigating the effect of gamma irradiation at different doses at room temperature on commercial PLA crust used as tomato packaging, the *T* value of the PLA sample, which was never irradiated, was found to be 0.350. It was observed that this value increased until the absorbed dose reached 300 kGy (0.825 opacity), but a dose of 600 kGy (0.452 opacity) caused a serious decrease [80].

The CIELAB scale is related to colour, as colour distribution is done with a smoother and more precise sorting it is one of the favourites colour measurement methods in research. Digital colorimetry devices are low-cost for evaluating film colour. Colour parametres L^*, a^* and b^* are calculated with this method. L^* value is an expression of the lightness (brightness), distance varies between 0 (black) to 100 (white); a^* value is an expression of the redness, distance varies between –60 (green) to +60 (red); b^* value is an expression of the yellowness, distance varies between –60 (blue) to +60 (yellow). The total colour difference (ΔE) is calculated by the equation:

$$\Delta E = \sqrt{\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2}} \quad (3)$$

where ΔE is the total colour difference between the sample films, ΔL^*, Δa^* and Δb^* are the changes between the white standard plate and the colour values of the films [81].

Colour difference threshold that can be distinguished by the human eye is 2.0 and the difference can be detected at ΔE values higher than this value. The consumer often pays attention to the image when choosing a packaged food product and, in this case, transparency is an important parameter. In a study with PLA films, they were found to be colourless and transparent [30]. While the pure PLA sample had no colour and was transparent, samples exposed to a higher dose of irradiation (>150 kGy) were found to be less transparent with a slightly yellowish colour [80].

Degradation properties

Many of the packaging materials used in various industries are designed to be used for many years, while food packaging is used in shorter-term applications due to the shelf life of foods. For this reason, it is highly desirable that the packing components are renewable. In the biodegradation process, using the metabolic activities of microorganisms, polymeric materials are converted into completely or largely useful small-molecule structures. However, some tests are required to calculate how bio-based a bioplastic is. A set of national, international and European standardized tests has been defined to improve the biodegradable materials or packaging from these materials. Various standards are applied, such as ISO 17088, ISO 14855, ASTM D6400, ISO 21644 [82–85], which determine methods for testing biodegradability. Biodegradation can be tested based on properties such as weight loss, changes in tensile strength, bacterial activity in soil, production of carbon dioxide or changes in molecular weight ratio. Important biodegradability tests can be listed as follows: soil burial method, pure culture method, compost method and anaerobic spoilage in case of sewage sludge [86]. PLA can be biodegradable in degradation conditions such as composting. Subjected to compost conditions, PLA becomes hydrolysis-sensitive and begins to decompose. In a study investigating degradability of PLA containers under ambient exposure and composting conditions, PLA bottles made of 96% L-lactide were found to show lower degradation than PLA bottles made of 94% L-lactide, and it was emphasized that this was mainly due to their higher crystallinity. Factors such as crystallinity at beginning and L-lactite content of the material, relative humidity, temperature and pH of the compost pile are important in the degradation rate of the packaging materials [87].

CONCLUSIONS

The legislation and environmental campaigns of countries proposing the production and use of polymers that can be completely biodegradable justify the research and production of new packaging materials including PLA. Given its weaknesses, further research needs to be done to improve the quality and performance of films made from PLA, thereby expanding their use and application in the food sector.

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