

Environmentally Benign Food Packaging Material based on Polylactic acid - A Brief Review

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Abstract - Polymer based materials have almost replaced the conventional materials like paper, paper board, metal and glass in the food packaging applications due to their convenience, versatility and tailor-made properties. In spite of their widespread acceptance as better option due to light weight, easy processability, low cost, long term retention of food quality and impact resistance, they are accused for their nonbiodegradability and long term landfilling. To address these problems, polylactic acid (PLA) can be a suitable candidate. The advantage of PLA is that it can be derived from renewable resources rather than depending on fossil fuels, it can break down into natural elements upon composting and it does not produce any toxic fumes if burned for energy recovery. The major disadvantage of PLA is its brittleness which can be overcome by blending with other FDA approved polymers like polyethylene glycol (PEG) or adding bioplasticizer like glycerine or by making nanocomposites with suitable fillers. Hence PLA can be designed to be environmental friendly food packaging material with all the desired set of functional properties. This review focuses on recent developments of PLA based blends and nanocomposites for advanced food packaging applications.

Key Words: Food packaging, Polymer, PLA, Biodegradable, Shelf-life, Smart packaging.

1. INTRODUCTION

Packaging helps in protecting the stored food product from external factors like dust, micro-organisms, moisture, oxygen, insects and other deteriorates and provides shelf-life, nutritional, cooking as well as ingredients information to the customers. It acts as an interface between food and consumer. In recent times, due to rise in consumer awareness, there is a demand for environmental friendly packaging material along with safety, convenience and extended shelf life at affordable cost. In this context polymer based materials scores high over metal or glass for food packaging applications [1]. Several biopolymers like cellulose, chitosan, starch, PLA, PVP, PCL, PHB etc. are used for food packaging purpose. But these polymers have their own inherent limitations when used alone. The current trend is to blend different biopolymers like starch/PLA, starch/PCL, PLA/PEG, PLA/PHA, PLA/PCL, PLA/PBS etc. and/or to make composites with suitable fillers, so that the overall performance can be improved at the same time their limitations can be reduced.

Food packaging is designed depending on the texture of food, amount to be handled (bulk or retail packs) and it comes in various forms like drums, barrels, bottles, jars, cans, pouches, flexible films, rigid containers, disposable bags, boxes, baskets etc. Customized design is necessary to attract the consumers, which is much easier with polymers than with metals and glass. Also processing of polymers consumes less energy than metals and glass. Many a times food packaging materials are of single use, so their discarding frequency is also more. To decrease the environmental footprint of such packaging materials, use of biodegradable, sustainable and recyclable materials is very important in efficient food supply chain management. In this regard, PLA based packaging materials has a great role to play in the upcoming years.

1.1 Materials used for food packaging

The shelf-life of packed food is mostly determined by packaging material and its design. The conventional materials used for packaging include paper-based materials, glass, metals like aluminium, steel, tin and synthetic polymers like polyethylene, polypropylene, polystyrene and polyethylene terephthalate etc. The main advantages of paper based packaging is that they are light weight, low cost, renewable resource based and environmental friendly but they are not suitable for liquid foods, not heat sealable, poor barrier and not durable. On the other hand, glass is an absolute barrier to moisture, oxygen and other environmental agents, so retains freshness of food for long period of time without altering the organoleptic properties of food. It is sterilizable, reusable, recyclable and transparent to see the packed product but it is highly brittle, heavy and demands energy-intensive production [1].

Metals offer a combination of excellent physical protection, impact resistance, barrier properties, decorative potential but they are of high cost and may be corrosive towards acidic and alkaline food items. Keeping all these limitations, food industries have started extensive use of polymers derived from fossil-fuels as they are cheaper, ductile, light in weight, good barrier, heat sealable, printable and can be easily processed. But they are non-biodegradable and tend

to remain as litter for long time if disposed, causing other environmental problems. Hence there is a need for development of biodegradable food packaging material which can give almost similar properties as synthetic plastic/polymer packages [2]. Among which, PLA has great potential to be processed into films for packaging applications, which can replace heavier bottles and containers for a range of food and beverage products. The key to successful food safeguarding is to select the suitable packaging material and design it to satisfy competing needs with regard to product characteristics, environmental issues, marketing considerations and cost.

1.2 Properties and requirements of polymeric food packaging materials

The physical, chemical, mechanical, optical, thermal and barrier properties mainly determine the suitability of a polymer for food packaging application and are mainly dependent on the structure, type and composition of polymer. It is estimated that the crystallinity of polymer shows a strong effect on the strength, transparency and barrier properties. In general, synthetic polymers are semi-crystalline, PLA can exhibit different level of crystallinity depending on its synthesis condition. Shelf life extension can be achieved with proper combination of polymers or by suitable modifications like blending, compounding or by making composites. Transparent packaging can give customers a preview of inside product before it is opened, PLA being transparent can serve the purpose. Many polymers are transparent and can be readily colored to any shades, also printable, which helps in customer attraction. Figure 1 shows the requirements of an ideal food packaging material.

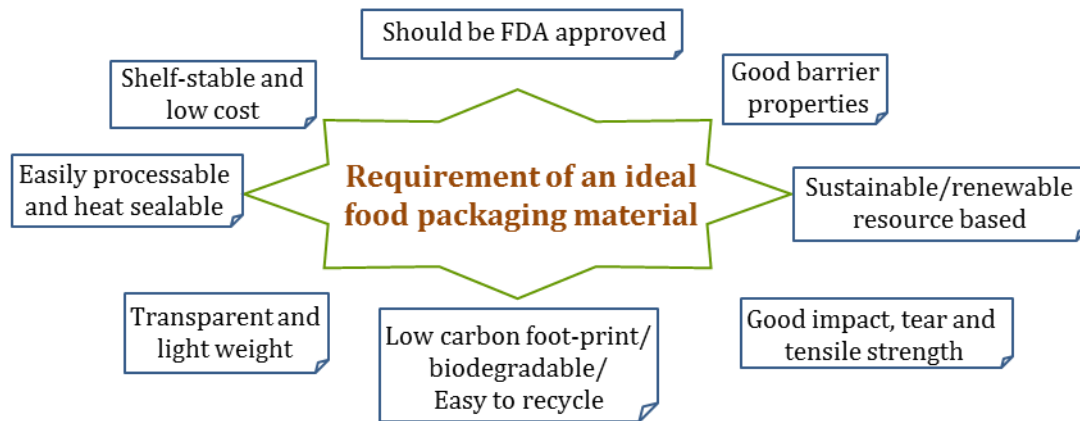


Figure 1. Requirements of an ideal food packaging material

The food packaging polymer must be FDA approved and should be as inert as possible towards the content packed, there should not be leaching and migration of packing material into the food. Chemical resistance at different pH, thermal stability (boil-in-bag packages), flavour/aroma retention, resistance towards oils, fats and other food additives are expected out of an ideal packaging material. Some food products like fruits, vegetables and red meat needs selective permeation in packaging, that allows the flow of carbon dioxide and oxygen in and out of the package because they respire to conserve their freshness. Excellent CO₂ sealing property is offered by PET and hence preferably used in the carbonated drink industry. To enhance the gas/flavour/aroma barrier property, multiple layers are being utilized in thermoformed containers and packaging films (coextruded). Mechanical properties like tear strength, tensile strength, flexibility, puncture resistance/impact resistance are required for packaging material especially for bulk packages and for delicate food items like egg, chips and other crispy products [1]. Heat sealability, microwave resistance (ready to eat cooked foods), autoclave and sterilizing temperature tolerance (like retort packing) are essential for some special category of food packaging materials. After serving the main role (containment, protection, preservation, convenience, marketing, communication) as food packaging, it must be biodegradable or easily recyclable so that it meets eco-safety. All these functions can be served by the polymers if they are properly formulated, processed and designed to meet the customised requirement. This is also evident from recent developments taken in food sector replacing the conventional materials.

2. PLA AS FOOD PACKAGING MATERIAL

Food is a very complex, delicate and diverse entity to be addressed in terms of overall quality. Which requires holistic approach of identifying a suitable material that can satisfy each stakeholders (supplier, processor, logistic operators, retailers, consumers, regulatory agencies, environmentalist) participating in food supply chain management. At present, PLA is the most exploited biopolymer in food packaging sector. This is mostly due to its resemblance in many mechanical properties with other petrochemical based plastics and its ability to adopt customised modifications. There is a high potential for PLA to be used in short lifecycle applications where biodegradability is highly favourable such as in

disposable water bottle or as a container for perishable items like milk, fruit juice, vegetables and meat. Despite its ability to degrade when exposed to environmental condition, PLA is extremely strong in any other normal applications. D- or L-lactic acid monomer forms the basic unit of PLA. It is an aliphatic polyester, mostly produced by ring-opening polymerization of lactide and/or polycondensation of lactic acid. It can also be produced commercially by fermentation of corn, sugars, tapioca or sugarcane, even by some agricultural waste [3]. The advantage of PLA as food packaging material is shown in Figure 2.

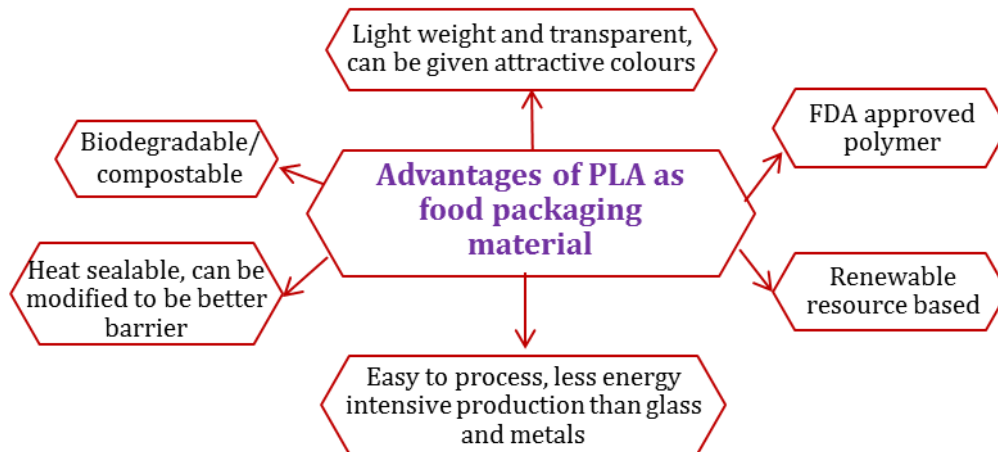


Figure 2. Advantages of PLA over other food packaging material

PLA can be easily processed by conventional industrialized methods already in use like blown film extrusion, thermoforming, compression and injection molding. To overcome the inherent limitations of PLA and to expand its scope of application, it is blended with other polymers or nanofillers. Degradation rate of PLA in the environment depends on the nature of soil. Degradation is stimulated by hydrolytic and enzymatic pathways in the environment [3]. Food spoilage is a complex process that could be originated by a range of physical, chemical, microbiological, biochemical or enzymatic actions. Hence a single material may not able to give complete protection against all possible factors for extended period. There comes the material blending, coextruded sheets, laminates and composites role. The sections below discusses about the PLA based blends and composites designed for food packaging applications.

2.1 PLA based blends for food packaging

The main shortcomings of PLA for its direct use as food packaging material is its brittleness, low melt strength, poor processability and formability, however this can be very well addressed by blending with other FDA approved tougher/ductile polymer. PLA can be blended with biopolymers like PHBV (polyhydroxybutyrate valerate) or other PHAs, which have properties similar to ABS and can reduce brittleness. Besides, PHBV also improves PLAs heat resistance. Although both PLA and PHBV are brittle polymers, blends containing even a small amount of PHBV shows a significant ductile deformation. PLA/PHB forms immiscible blend system, to improve the miscibility, different compatibilizers like ethylene oxide/propylene oxide block copolymer and a mixture of liquid surfactants with a variable hydrophilic-lipophilic index was suggested by D’Anna et al. [4]. A notable increase in elastic modulus was reported for the compatibilized blends as compared to the pure counterparts, with a significant improvement of the HDT. The capability of PHB to act as a nucleating agent for PLA crystallization, increases its barrier performance and mechanical strength. PLA/PHB flexible films plasticized with d-limonene is reported by Arrieta et al. [5]. Presence of d-limonene improved the processing and interaction between PLA and PHB and eases the film disintegration during composting. PLA blending with polyethylene or copolyesters can also reduce brittleness and increase tear strength.

In PLA/PMMA blends, PMMA helps to increase the glass-transition temperature while retaining clarity. PLA blending with chemically modified thermoplastic starch is reported by Shin et al. [6]. it forms thermodynamically immiscible (yet compatible) system. However, the interfacial adhesion can be further improved through transesterification reaction by the PLA-g-starch copolymers. The extent of biodegradability of the blends can be increased with increasing starch content. PLA blending with LDPE in different weight ratios using single screw extruder is studied by Hamad et al. [7]. The mechanical results showed incompatibility between PLA and LDPE but stress at break and Young’s modulus of the blend increased with increasing PLA content. In a study by Bijarimi et al. [8], PLA was melt blended with polypropylene and liquid natural rubber (LNR) in the Haake Rheomix internal mixer. It was reported that the elongation at break, notched impact and flexural strength increased considerably for the LNR compatibilized PLA/PP blend. Blending of PLA with petro-based plastics helps in cost reduction, as PLA is costlier than conventional polymers. PLA/PCL blends preparation

and characterization was reported by Matta et al. [9]. When compared to neat PLA, the blend system exhibited an increase in toughness and percentage elongation. PLA/PEG melt-blending at various ratios and extrusion into films is studied by Li et al. [10]. It was noticed that blends form partially miscible to miscible system depending on the composition. PEG plasticized the PLA, yielding higher elongation and lower modulus. Above 50 wt% of PEG content, the blend showed increasing crystallinity and modulus and a decrease in elongation at break. Enzymatic degradation results showed significantly greater weight loss for all the blends than that of neat PLA.

Polybutylene succinate (PBS) blending with PLA improves processability and makes easy conveying of the blend through processing machinery [11]. PBS is also biodegradable aliphatic polyester with properties similar to that of polypropylene. PBS can be easily processed into bags, films, boxes for both cosmetic and food packaging. PLA/polypropylene carbonate (PPC) blend with improved impact strength and elongation at break was studied by Zou et al. [12]. When the content of PPC is 40 wt%, the elongation at break increased by nearly 13 times and the impact strength was nearly 49% higher than that of pure PLA. The study showed that PPC content of 30 wt% exhibited higher tensile strength and good elongation at break. Table 1 show the PLA based blends used in food packaging application and their significant advantages over neat PLA. Though PLA is commercialized for single use disposal packaging applications such as cold drink cups, bottles, thermoformed lids and trays, containers, blister packages, overwrap as well as flexible films. Still suffer from certain limitations, thus blending can be used as a better strategy to minimise the inherent drawback of PLA and to widen the scope of PLA application in food packaging.

Table 1. PLA based blends for food packaging applications

PLA based blends	Advantages over neat PLA
PLA/PBS	<ul style="list-style-type: none"> Crystallization rate of PLA is accelerated by PBS. Accelerates biodegradation rate.
PLA/PHBV	<ul style="list-style-type: none"> Shows significant ductile deformation. Improves dimensional stability.
PLA/PHA	<ul style="list-style-type: none"> Higher elongation and tensile strength. Improved biodegradability.
PLA/PEG	<ul style="list-style-type: none"> Increases the toughness of PLA. PEG reduces the T_g as well as T_c and plasticizes PLA.
PLA/Starch	<ul style="list-style-type: none"> The biodegradability rate of PLA increases. Cost of starch is less compared to PLA, hence overall cost of blend reduces.
PLA/PHB	<ul style="list-style-type: none"> Improved mechanical strength and barrier performance. Improved thermal stability. Elongation at break increases without significant reduction of Young's modulus and tensile strength.
PLA/PCL	<ul style="list-style-type: none"> Improved mechanical stiffness and biocompatibility. Improved processability.
PLA/PPC	<ul style="list-style-type: none"> The elongation at break and impact strength of the blends are higher. The blends maintained high tensile strength.
PLA/PBT	<ul style="list-style-type: none"> Provides high elongation and better impact resistance. PBT adds high tensile strength to blend.
PLA/PE	<ul style="list-style-type: none"> Gives better surface finish/glossiness. Improved barrier property and cost reduction.
PLA/PP	<ul style="list-style-type: none"> Flexural and notched impact strength increases. Improved elongation at break and cost reduction.

2.2 PLA based nanocomposites for food packaging

An ever-changing consumer preference about food freshness, safety and convenience has led to the newer developments in food packaging. To satisfy these demanding needs, materials used for food packaging should play a diverse role. Nanocomposites of PLA are widely used for food packaging applications due to their superiority over pristine PLA. The limitations of PLA such as slow crystallization rate, poor barrier property, low HDT and impact strength can be substantially addressed by adding reinforcements, additives and nanofillers. PLA based nanocomposites can serve three main functions, firstly improves barrier and mechanical properties, including elasticity and thermal stability. Secondly, can work as smart packaging for information feedback, real-time quality assessment and can serve as a guard to detect fake and fraud products and as an indicator of the product exposure history to certain adverse factors like insufficient temperature or high moisture levels. Thirdly, as an active packaging (eg. antimicrobial, ethylene scavenging, moisture absorbing etc.) offers extended protection and preservation.

The new developments in extended shelf life food packages have mostly focused on delaying oxidation, controlling moisture migration, avoiding microbial growth, monitoring respiration rate of fresh foods and retention of volatile flavours/aroma. In all these functions PLA based nanocomposites have their contributions. Fillers like talc can act as a nucleating agent in PLA crystallization, which can increase crystallization rate and reduce overall molding cycle time [13]. Addition of calcium sulfate (dehydrated gypsum) improves heat resistance of PLA. Aframehr et al. [14] investigated the effect of CaCO₃ nanoparticles on the barrier properties and biodegradability of PLA. The results showed enhanced CO₂ and O₂ barrier properties. Very fine-particle of silica can increase toughness while maintaining transparency. A special high-aspect-ratio precipitated calcium carbonate also reduces PLAs brittleness. Silver, CuO, MgO, ZnO, TiO₂ and Fe₃O₄ nanoparticle (NP) based composites have attracted great interest due to their strong foodborne pathogen inhibition capability. Li et al. [15] reported the influence of PLA/ZnO nanocomposite films on the quality of fresh-cut apples. It showed better retention of firmness, color, sensory quality and total phenolic content along with strong microbial inhibition.

Recently, the use of bio based cellulose nanoparticles such as cellulose nanocrystals (CNC), bacterial nanocellulose (BC) and cellulose nanofibers (CNF) have been proposed to manufacture PLA-based biocomposites with enhanced mechanical and barrier properties [16]. However, due to hydrophilic nature and presence of hydrogen bonding, cellulose nanoparticles are not compatible with hydrophobic PLA. Strong interactions between cellulose nanoparticles, high surface area and high surface energy cause the irreversible agglomeration during melt mixing of nanocomposites. The dispersion of these nanoparticles in PLA still remains as the main challenge to process/develop their nanocomposites. To enhance the dispersion within PLA, surface functionalization by acetylation, oxidation and polymer grafting, surfactant/compatibilizer addition and use of hybrid processing approaches is practiced. CNF is considered as a bio-reinforcement of low density, high specific strength, high modulus and large aspect ratio which can be added to PLA to enhance the mechanical properties. Table 2 summarizes the advantages of PLA based nanocomposites for food packaging applications.

Table 2. PLA based nanocomposites for food packaging applications

PLA based nanocomposites	Advantages over neat PLA
PLA/CNF	<ul style="list-style-type: none"> Fully bio-based and biodegradable nanocomposite. High modulus and large aspect ratio. Better thermal stability than PLA. Enhanced oxygen barrier property.
PLA/SiO ₂ NPs	<ul style="list-style-type: none"> Improved oxygen barrier property. Improved tensile and tear strength. Improved transparency and thermal stability.
PLA/Cu NPs	<ul style="list-style-type: none"> Exhibit antibacterial effects against foodborne pathogens. Antifungal properties as well. Increased thermal stability and better gas barrier property.
PLA/TiO ₂ NPs	<ul style="list-style-type: none"> Decreased water vapour permeability. Tensile strength of films increases significantly. Improved barrier property and antibacterial activity.
PLA/ZnO NPs	<ul style="list-style-type: none"> Films exhibit antibacterial activity against food-borne pathogens. Prolong the shelf-life of fresh food products.
PLA/MgO NPs	<ul style="list-style-type: none"> Improved oxygen and water vapour barrier properties. Anti-bacterial property. Improved tensile strength and plasticity.
PLA/Silver NPs	<ul style="list-style-type: none"> Antimicrobial, anti-fungal and anti-viral properties. Better thermal stability and barrier property.
PLA/Nano Clays	<ul style="list-style-type: none"> Shows satisfactory antimicrobial activity. Improved barrier property. Improved mechanical and thermal property.

2.3 Recent developments in PLA based food packaging

A package is often the only product exposure customers experience before buying. Consequently, innovative/smart and attractive packaging can boost sales in a competitive era. Regulations enforced add an extra challenge to food supply chain that operates across international borders. The package itself is designed to convey information about the product such as net weight, ingredient declaration, nutritional value, cooking instructions, manufacturer details, brand identification, storage instruction, the date up to which it is safe, for whom it is intended, pricing, genetically modified ingredients and

allergens alert etc. Renewable and biodegradable polymers like PLA have attracted much consideration due to sustainability issues and environmental concerns connected with petrochemical based polymers. Consumer market drive towards greener packaging and waste reduction made food industries to look upon renewable packaging materials. Film is the second largest application for PLA. Flexible food packaging films are made thinner to minimize resource utilization and reduce waste generation. Apart from serving the basic functions, food packages are also expected to play active and smart/intelligent roles like conveying the safety status of food, freshness indication, previous thermal storage history tracing (frozen foods, ice creams), hotness/coldness indication of the product (coffee, tea, beverage) without opening the package, detection of possible adulteration and active/control release of antimicrobial agents/scavengers etc. [17].

There is a compelling need for innovative technologies to guarantee food security. Market globalization, industrialization, urbanization, consumer awareness, improved economic status, time-bound life style management, zero-preservative, convenience, easy dispensing, consumer appealing and regulatory requirement have led to worldwide changes in lifestyle and nutritional habits. PLA based smart packaging is a broad concept, which includes both the active and intelligent packaging which can monitor the external and internal changes that happens in a food product (intelligent) and further respond (active) by communicating with an external interface [18]. The purpose of smart packaging is to prolong the shelf-life, retain freshness, exchange quality status of the product with consumers, boost product's safety and improve traceability of the product while moving across the supply chain. Active packaging as an alternative to traditional packaging, contains diverse components embedded into the system which are capable of absorbing/releasing substances from/into the packaged food to avoid decay. PLA based intelligent packaging is used to track and monitor conditions of packed foods, to capture and provide product information during the storage and transportation. Thus, intelligent packaging systems customarily comprise hardware elements like integrity indicators, food spoilage indicators, rancidity indicators, gas sensors, ripeness and freshness indicators, time-temperature indicators, responsive coating, litmus paper, optical label, colorimetric sensor/chromogenic chemosensors, conductivity or pH electrodes and sensor-enabled smart RFID tags to sense the quality status of food [19]. PLA based food packaging systems have evolved smarter/intelligent with integration of wireless communication, emerging electronics and cloud data solutions. Innovative smart packaging solutions are useful to assess the inclusive safety and quality of food supply by enhancing product traceability and reducing the amount of food waste.

3. CONCLUSION

Global awareness of material sustainability has opened up the demand for bio-based polymers like PLA. The food industry has witnessed great advances in packaging sector with most active and intelligent innovations happened during the preceding years. These advances have led to enhanced food quality and safety with fewer burden on environment. Role of industry and consumers on taking up environmental responsibility is constantly questioned in food supply chain. Biopolymers like PLA represent the ideal solution to combat the massive plastic waste evils/litter that is faced in recent times. Since packed foods are highly popularized/consumed among current generations, packaging material disposal frequency is also more, which has to be suitably addressed to lower their ecological footprints. In this direction biodegradable PLA based blends and nanocomposites have great role to play. The mechanical properties offered by PLA are similar to petro-based plastics, if not it can be matched with certain modifications; hence it can be a better alternate to conventional plastics for food packaging applications. Functional fillers can be added to PLA to expand its role as active and/or smart/intelligent packaging. Customer driven food market may excite the researchers to do more and more innovations in upcoming years, wherein PLA have its strong essence and become an inseparable part of sustainable food packaging.

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