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An overview of the preparation and characterization of biodegradable polylactic acid (PLA) films for food packaging

Nugarjuna P.S[1]

DR. Shivbrat Singh[2]

PhD Research scholar, Department of Chemistry, Sunrise University[1]

Professor, Department of Chemistry, Sunrise University[2]

Abstract:
Biodegradable PLA which is produced by polymerization of lactic acid derived from the starch of plants is a good candidate for replacing the current petroleum-based polymers such as polystyrene (PS) or Polypropylene (PP) in food packaging application. These petroleum-based polymers do contribute to large-scale environmental pollution as it is a non-degradable polymer. Being classified as Generally Recognized As Safe (GRAS) for the food contact applications, PLA film for food packaging application can be synthesized in lab-scale and mass production by using solvent casting and injection molding which are common for the thermoplastic fabrication with the fast production and low in cost. The characterization methods used for the PLA film analysis can be done by the Universal Tensile machine (UTS) in which the main characteristics of the polymer film compatibility and its performance with the combined materials have been discussed in comparison with those of pure polymer films.
Key Words: Biodegradable, Polypropylene, Tensile machine

Introduction :

Polylactic acid (PLA) is a biodegradable polymer derived from renewable resources such as corn starch tapioca roots, chips, or starch or sugarcane [1,2,4]. PLA is produced by chemical conversion [3] of corn and its carbohydrate sources into dextrose which is then further fermented into lactic acid and followed by polycondensation. PLA could be obtained by ring-opening polymerization [2,4] which is the process of breaking the cyclic lactide chain to develop a long and continuous PLA chain. With its classification being generally recognized as safe (GRAS) by the United State Food and Drug Administration (FDA) [2,5,6], the use of PLA has been approved to be safe for use in food packaging.

Of the plastic materials, petroleum-based plastics, such as low-density polyethylene (LDPE), high- density polyethylene (HDPE), polyethylene (PE), polypropylene (PP), polystyrene (PS), and polyamide (PA) [14]. The huge usage of these non-renewable plastic materials have developed consistent waste production issues which cause environmental pollution. The problem that arises on this issue is the disposal and nonbiodegradability of the polymer that hardly to degrade as they are produced from petroleum-based resources. Biodegradation of polymer can be defined as the ability of the polymers to disintegrate its body due to the natural environmental factors that lead to the loss of its main properties such as the mechanical and the chemical configuration, and the last products of the process could be the combination of water, carbon dioxide, minerals, and other by-products such as biomass and the organic compounds [6].

Polylactide (PLA) is one of the most promising bio-based polyesters [7] being studied extensively for its biodegradable properties for food packaging [8]. PLA being classified as biodegradable polymers that can disintegrate [9] and undergo deterioration when in contact with the moisture or the environment [13]. The degradation of PLA is influenced by several factors and two common degradation mechanisms involving the structure and hydrolysis media [10,11,12]. Guzman *et al*. (2011) reported that the degradation of PLA is initially due to the hydrolysis of the PLA structure into a compound that soluble in water followed by the bacterial attack [12] on the fragmented residues such as the moisture, carbon dioxide, and the other organic compounds [11]. Hydrolytic degradation in PLA can take place during melt processing [10] which can be activated by the presence of moisture at high temperatures and the hydrolytic reactions are particularly faster under alkaline conditions.

PLA has good mechanical strength and thermal plasticity that can be used for many applications. PLA has low in toughness with only 10% of elongation at breaking point [15], in other words, PLA is relatively a brittle polymeric material. The crystal structure of the PLA can be in amorphous or semi- crystalline in a solid-state [15], depending on its stereochemistry and thermal history. PLA stated to have a melting point around 151℃with the value of heat of fusion around 21.5 J/g [16]. Therefore, PLA is chosen as the material discussed in this research. The purpose of this research is to provide information related to the influence of several modifications to the mechanical properties of the PLA.

Preparation Methods of Polylactic Acid

PLA can be processed into varieties of products depending upon the material to be made. Due to its good thermal processability, PLA can be fit for different fabrication processes either injection molding, sheet extrusion, blow molding, thermoforming, film-forming, fiber spinning, or casting method [6,14,17]. The manufacturing routes show certain fundamental similarities [6], with the major differences depending on whether a thermoset or thermoplastic biopolymer is to be processed. The basic step involves melting the biopolymer mix [13,17] followed by casting, extrusion, or blow molding depending upon the final desired product. It was reported that the commonly used technique for PLA film processing is the extrusion and solvent casting method [5,11,14].

Extrusion allows PLA resins to be mixed homogeneously under high temperatures. It is the best method for the biopolymers made up of aliphatic esters as it is difficult to process them by methods as film-blowing and blow molding due to their low melting point [17]. The solvent casting method uses a solvent such as chloroform or dichloromethane to dissolve PLA resin [18,19,20] and cast to obtain films with high transparency and gloss [14]. The film solution allowed to dry to eliminate any humidity in the PLA pallet [18,19,20] to form a film. The drying process in vacuum conditions reported reducing the shrinkage rate of the film produced [5].

Comparing its mechanical and optical properties to those of conventional plastics, PLA could substitute LDPE and HDPE, PS or PE in several food packaging applications [14]. To extend PLA applications, the properties like flexibility, stiffness, barrier properties, thermal stability and production costs must be improved. Modifications have been carried out to make PLA a good fit for the food packaging applications. Generally, modifiers have been studied to improve polymer stiffness at higher temperatures, reduce cost, or increase the degradation rate of PLA [6].

Being the growing alternatives as ‘green’ food packaging polymer [6], the choice of polymers or plasticizers and blending treatment of PLA with various polymers to be used as modifiers for PLA is important as it has been a facile way to improve the properties of PLA. Some efforts of PLA modifications in the field of packaging reported by different authors are presented in Table 1.

Table 1. Summary of PLA film modifications for packaging applications by different authors

Approaches		Treatment	Observation	References
Addition	Additives	Emulsifiers, Stabilizers	Improve physical properties (flow behavior and particle size) and microstructure of oil-in-water (o/w) emulsions, improve plasticizing properties	[6], [36], [44]
	Modifiers	Polyglycerol esters	Improve the elongation at break	[6]
	Fillers	Nanofillers (clays, silica, metals), natural fibers/bio- fibers (kenaf, hemp, flax)	Improve film properties with antimicrobial & UV light screening properties, improve elongation at break, improve mechanical properties & reduced matrix's molecular weight	[5], [7], [23], [26], [27], [33], [35], [37]
	Plasticizers	Glycerol, Polyethylene glycol (PEG)	Overcome the brittleness and widen PLA's applications, increases the transparency of film, improve the mechanical, thermal & barrier properties of the film, enhance the crystallinity and biodegradability of PLA	[6], [30], [31], [39]
	Compatibilizer	PLLA–PBS block copolymer	Improve the adhesion between the blended components	[8], [14]
Blending with polymers		Polyhydroxy butyrate (PHB), Hydroxyapatite	Improved mechanical strength, oxygen barrier, water resistant of the polymer as compared with pure PLA	[4], [6], [15], [17],
		Natural biopolymers (starch, cellulose) with different plasticizers	Lowering the price, decreasing Tg, increasing crystallinity and biodegradability, increase the transparency of packaging films	[28], [29], [38], [41], [43]
Incorporation of active compounds		Essential oils (tea tree, bergamot & lemongrass), Nisin, Thymol	Improve functional (mechanical & antibacterial) properties, addition of essential oil reduces water vapor permeability, moisture absorption and increases transparency of film	[14], [18], [19], [30], [32], [42]
Fabrication of bilayer and/or trilayer films		Biopolymers (starch, cellulose)	Good transparency, improved mechanical performance, better water barrier properties, enhance the physical performance of films	[10], [14], [16]
Physical Treatment		Orientation	Significant improvement in tensile and impact properties	[6], [12],

Annealing	Increase toughness	[34]
Aging	Increase Tg	

Characterization methods of the Polylactic acid

The polymer manner plays an important role in the mechanical properties and the process utilized to prepare the final product. Changes may occur in the mechanical and physical properties of many packaging materials after the incorporation of active compounds, additives, modifiers, fillers, and

compatibilizers. Many packaging containers are commercially used below room temperature, so it is important to assess the mechanical performance under these conditions.

For tensile modulus and flexural modulus, PLA has the highest value in comparison to PS, PP, and HDPE [6]. A relatively wide range of tensile strength (14–70 MPa) and deformation at break (1–8%) can be found depending on the type of PLA and process it involves in [14]. Since PLA-based materials are rigid and brittle, plasticizers and fillers have been added to enhance the mechanical performance of the PLA films. The incorporation of the reinforcing component to polymer has recently proposed to improve the poor properties of this biopolymer film.

Getme *et al.* (2019) reported that the development of PLA film reinforced with fibers as filler gives a higher modulus of biocomposites compared to virgin PLA. Several authors also reported the PLA plasticization with polyethylene glycol (PEG) of different molecular weights increases its crystallinity and biodegradability [6]. As plasticizers decreased the glass-transition temperature (Tg), lower stress at yield and higher elongation at break were observed in the different studies [14,22].

A study done by Vartiainen *et al.* (2014) found that the formed copolymer of PLA and biodegradable polymer (polybutylene adipate terephthalate (PBAT) improved the compatibility between both polymers. The toughness of the compatibilized blend was increased significantly without severe loss in tensile strength. The improved toughness of the compatibilized blend was due to the shear yielding mechanism [24]. Yang *et al.* (2016) have compatibilized PLA/starch blends with biobased epoxy- functional compatibilizer, and the result showed a significant increases in impact strength, tensile strength, and elongation at break. The improved mechanical performances and reduced surface tension suggest that the compatibility between the starch and PLA has been improved after the incorporation of compatibilizer [25,26]. A comparison of the values of the tensile properties of neat PLA films reported by different authors is described in Table 2.

Table 2: Tensile strength (MPa) and elongation at break (%) of neat PLA films reported by different authors (Muller et al., 2017)

PLA	Processing	Tensile Strength (MPa)	Elongation at Break (%)
4042D	Melt-blending Compression molding	70.2	7.4
2002D	Melt-blending Compression molding	55.0	4.5
2002D	Extrusion Injection molding	60.0	2.0
4042D	Melt-blending Compression molding	56.3	3.6
2002D	Extrusion Blown molding	34.6	2.1
2000D	Casting (Chloroform)	24.8	7.9
4032D	Extrusion Injection molding	65.0	5.0
4042D	Extrusion Injection molding	38.0	1.0

Conclusion

PLA is one of the best materials that can be further study as a substitute for the common petroleum- based food packaging. The preparation of the PLA thin film using extrusion and solvent casting method need to comply with a suitable scale for further research purposes. Although biopolymers are environmentally friendly and the most attractive packaging materials, the industrial applications are limited due to some factors. Hence, the methods of modifying PLA focusing on the incorporation of renewable reinforcement can be of great interest to enhance some limitations of PLA while maintaining their transparency.

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