The Innovation Research of Biodegradable Polymers for Sustainable Packaging

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Abstract. Packaging is an essential requirement for commodities to provide the requisite protection. Synthetic plastic polymers manufactured from fossil fuels are the most frequently packaging materials. However, their usage currently is being partially restricted because they are not totally degradable and thus lead to serious environmental problem. Therefore, biodegradable polymers for sustainable packaging have drawn more attention nowadays. The innovation research and recent advances of biodegradable polymers such as polylactic acid (PLA), polysaccharides, and proteins are reviewed in this paper.

Introduction

Packaging could provide commodities protection from physical damage and contamination, and increase shelf life [1]. Almost all commodities in everyday life could not come without packaging [2]. During the past 50 years, synthetic polymers, such as polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET) or polystyrene (PS), are the most frequently packaging because of their advantages of low density, low cost, inertness, thermoplasticity, transparency, and resistance to microbial growth [1]. However, conventional polymers packaging, manufactured from fossil fuels such as oil or natural gas, not only consume non-renewable and finite resources, but also contribute to the generation of municipal solid waste (MSW) [3,4]. The concept of sustainability is one of the most highly discussed in the area of packaging nowadays [1]. Much attention has been paid to developing naturally degradable materials to replace the traditional polymers packaging [5-7]. The development of degradable polymers has become a highlight of the sustainable development and circular economy movements [7,8]. Biodegradable polymers are promising degradable materials, because they produce only water, carbon dioxide and inorganic compounds without leaving any toxic residues during biological degradation [7,9]. Biopolymers produced from various natural resources have been considered attractive alternatives for non-biodegradable petroleum-based plastics since they are renewable, environmentally friendly and biodegradable [4]. Sustainability is a huge driver for innovation. The recent advances and new aspects of biodegradable polymers for sustainable packaging, such as polylactic acid (PLA), polysaccharides, and proteins are reviewed in this paper.

PLA

Polylactic acid (PLA) is currently one of the most extensively researched and utilized biodegradable polymers [10]. PLA is biodegradable thermoplastic polyester made from natural resources such as corn and sugarcane either chemically or by fermentation [11]. Generally, it is produced through condensation polymerization of lactic acid monomers or ring-opening polymerization of lactides [12]. It is considered as an environmentally benign substitute of low and high-density polyethylene (LDPE and HDPE), PET, and PS, owing to its beneficial properties of biocompatibility, high mechanical strength, outstanding transparency, easy processability, ability to be easily composted, and monomer renewability [11,13,14].

Unfortunately, although most physical properties are equivalent to the engineering plastics, the current incorporation of PLA into large-scale commercial applications has been greatly restricted by
several disadvantages, such as its low thermal resistance, inherent brittleness, and poor water-vapor barrier properties [13,14,15]. The use of natural fibers as fillers or reinforcing agents and other biodegradable polymers (for example, starch and protein) in PLA blends or composites is expected to considerably lower the price of the products as well as to improve the properties without compromising the biodegradability [13,16].

Richard Eungkee Lee et al. [12] manufactured PLA foam sheets with two-layered structures by laminating a solid high heat deflection temperature (HDT) PLA film onto a low density PLA foam sheet, which is able to increase the thermoformed foam products’ service temperatures and to satisfy the requirements in hot-fill food packaging applications.

Stephen Spinella et al. [10] employed a green manufacturing technique, reactive extrusion (REx), to improve the mechanical properties of PLA. A fully biosourced PLA based polymer blend was conceived by incorporating small quantities of poly(ω-hydroxytetradecanoic acid) (PC14), which showed good adhesion between dispersed PC14 phases within the continuous PLA phase as shown in Fig. 1.

![Figure 1. SEM images of cryofractured PLA/PC14 (90/10) prepared by melt blending [10].](image1)

Lan Xie et al. [17] used a confined flaking technique to establish the degradable nanolaminar poly(butylene succinate) (PBS) in PLA films, which created compact polymer with “nano-barrier walls” (shown in Fig. 2). A dramatic decrease in the oxygen permeability coefficient was observed for the film incorporated with well-organized PBS nanosheets compared to pure PLA and pure PBS.

![Figure 2. SEM images of section cryofracture surfaces of PLA/PBS composite films after etching the PLA matrix for PLA/PBS (80/20) [17].](image2)

Marta Musiol et al. [18] studied the biodegradation of commercial PLA/poly(3HB-co-4HB) (PLA/poly(3-hydroxybutyrate-co-4-hydroxybutyrate)) and their composites with 20%wt of jute fibres, and found that the presence of jute did increase the biodegradation rate. Natural materials as polymer composites have been drawn much attention as they exhibit attractive advantages such as
low cost, lower energy requirements for processing, and their renewable and degradable characteristics [19,20].

**Polysaccharides**

*Cellulose.* Cellulose is the most ubiquitous and abundant natural biodegradable polysaccharide and is derived by a delignification from wood pulp or cotton linters [11,21]. The packaging industry has always been using cellulose-based materials in large amount, which is experiencing a very innovative phase, driven by the introduction of cellulose in ‘nano’ form in various applications [21]. Nanocellulose has potential applications across several industrial sectors and allows the development of innovative materials, as well as the enhancement of conventional materials properties [21]. Safoura Ahmadzadeh et al. [22] developed cellulose nanocomposite foams incorporated with surface-modified montmorillonite (SM-MMT), and showed that the properties of cellulose matrix was improved as well as reducing the bubble sizes to prepare nanofoams. The results of thermal conductivity and morphology (Fig. 3) indicated that presence of SM-MMT improved thermal insulating properties due to reduction of average cell size [22]. Christian Aulin et al. [23] used the layer-by-layer (LbL) deposition method for the build-up of alternating layers of nanofibrillated cellulose (NFC) or carboxymethyl cellulose (CMC) with a branched, cationic polyelectrolyte, polyethyleneimine (PEI) on flexible PLA substrates, and formed optically transparent nanocellulosic films with tunable gas barrier properties. Commercially available paperboards are coated with thin layers of nanocellulose. According to ref [24], a moisture-protective layer of renewable alkyd resins is deposited on the nanocellulose precoated sheets using a lithographic printing or water-borne dispersion coating process, and found that the water vapor barrier properties of the nanocellulose precoated substrates were significantly improved by thin layers of renewable alkyd resins.

![Figure 3. Thermal conductivity values and morphology of cellulose/SM-MMT nanocomposite foams [22].](image)

*Chitin.* Chitin is the second most abundant biopolymer in nature and has tremendous potential in sustainable packaging, as its attractive properties, including biodegradability, antibacterial activity, and high strength. Jie Wu et al. [25] extracted chitin nanofibers (ChNFs) from crab α-chitin by a high pressure homogenization process under acidic, and reported that pure chitin films obtained by simply drying the ChNF dispersions exhibited high optical transparency and flexibility, and excellent gas barrier properties. Chitosan (CS) is a linear aminopolysaccharide derived from chitin, a major component of insects and crustacean shells [26]. Tao Zhang et al. [27] designed a novel CS based polyelectrolyte complex (PEC) containing phosphorus, nitrogen and carbon elements.

*Other polysaccharides.* Other polysaccharides, like starch, xylans, and mannans can also be used for the production of biopolymer [11]. Starch is a widely available and easy biodegradable natural resource, which exists out of amylose and amylopectine. High water content or plasticizers (glycerol, sorbitol) are necessary to produce both foams and solid starch-based molded articles. Unfortunately, starch plasticized with water has poor dimensional stability and becomes brittle as water is lost, and the properties of plasticized starch are poor at high humidity. The properties can be improved
significantly by blending with other polymers, fillers, and fibers [11,28]. The wood hydrolysates are rich in xylan and utilized in the production of fully renewable films that provide very good oxygen barrier function and mechanical integrity also at high relative humidity [29].

Proteins

Proteins are another natural material that can be used to produce biopolymer. There are animal-based proteins, like casein and whey proteins and plant-based proteins, like soy proteins, zein and wheat gluten [11]. Proteins have been evaluated for applications in the packaging field, since they present appropriate barrier properties to water and gas permeation [30]. Patrizia Cinelli et al. [30] coated a whey protein-based layer on PLA (as shown in Fig. 4), and found that oxygen barrier properties were significantly improved by the presence of the whey protein layer, which was not affecting the compostability of the final material. T. Garrido et al. [31] successfully prepared biodegradable films with soy protein extracted from soybeans used to obtain soy oil, and obtained transparent films with excellent UV barrier properties.

![Figure 4. SEM Micrograph of PLA film without (left) and with (right) whey layer coating [30].](image)

Summary

The most recent advances and the new aspects of biodegradable polymers, such as PLA, polysaccharides, and proteins, are discussed in this paper. In addition, new biodegradable polymers, the polyhydroxyalkanoates (PHA) family, which are biodegradable thermoplastic polymers produced in the microbial cells as an energy reserve through a fermentation process, has drawn more attention as a promising sustainable packaging materials. With the continuous development of new materials and new technologies, biodegradable polymer will gradually replace the position of traditional polymer in packaging applications.

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