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Development and characterization of sugar palm starch and poly(lactic acid) bilayer films

M.L. Sanyang[a], S.M. Sapuan[a,b,c,*], M. Jawaid[c], M.R. Ishak[d], J. Sahari[e]

[a] Green Engineering, Institute of Advanced Technology, Universiti Putra Malaysia, 43400 Serdang UPM, Selangor, Malaysia
[b] Department of Mechanical and Manufacturing Engineering, Universiti Putra Malaysia, 43400 Serdang UPM, Selangor, Malaysia
[c] Laboratory of Biocomposite Technology, Institute of Tropical Forestry and Forest Products, Universiti Putra Malaysia, 43400 Serdang UPM, Selangor, Malaysia
[d] Department of Aerospace Engineering, Universiti Putra Malaysia, 43400 Serdang UPM, Selangor, Malaysia
[e] Faculty of Science and Natural Resources, Universiti Malaysia Sabah, Jalan UMS, Kota Kinabalu, 88400 Sabah, Malaysia

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ABSTRACT

The development and characterization of environmentally friendly bilayer films from sugar palm starch (SPS) and poly(lactic acid) (PLA) were conducted in this study. The SPS-PLA bilayer films and their individual components were characterized for their physical, mechanical, thermal and water barrier properties. Addition of 50% PLA layer onto 50% SPS layer (SPS50-PLA50) increased the tensile strength of neat SPS film from 7.74 to 13.65 MPa but reduced their elongation at break from 46.66 to 15.53%. The incorporation of PLA layer significantly reduced the water vapor permeability as well as the water uptake and solubility of bilayer films which was attributed to the hydrophobic characteristic of the PLA layer. Furthermore, scanning electron microscopy (SEM) image of SPS50-PLA50 revealed lack of strong interfacial adhesion between the SPS and PLA. Overall, the incorporation of PLA layer onto SPS films enhances the suitability of SPS based films for food packaging.

1. Introduction

Packaging films developed from petroleum based plastics are widely used in the food industry to preserve and extend the shelf-life of foodstuffs. However, the negative environmental impacts which emanate from the excessive usage of non-biodegradable plastics drew the attention of material scientists and engineers towards renewable and biodegradable food packaging materials. Over the past decades, intensive research efforts have been dedicated to substitute petroleum based plastics with more environmentally friendly materials for food packaging applications (Bonilla, Fortunati, Atarés, Chiralt, & Kenny, 2014; Sun, Sun, & Xiong, 2013). Hence, biopolymers have drastically emerged as alternative replacements to resolve environmental problems caused by conventional packaging plastic wastes.

Among many biopolymers, starches have attracted exclusive attention for the preparation of biodegradable packaging films. Starch based films have demonstrated several advantages such as renewability, sustainability, biodegradability, availability and affordability. They also exhibit physical characteristics which are similar to the conventional packaging plastics in terms of transparent, odorless and tasteless (Cano, Jiménez, Cháfer, Gónzalez, & Chiralt, 2014). Moreover, starch based films are reported to be nontoxic which has also contributed to their growing acceptance as potential packaging alternative (Kurek, Galus, & Debeaufort, 2014). On this basis, sugar palm starch like most other starches has received considerable research interest in producing starch based films for food packaging purposes. Sugar palm starch is extracted from sugar palm (*Arenga pinnata*) which is a multipurpose tree within the *palmae family* and mostly found in tropical countries (Ishak et al., 2013). This novel biopolymer has been reported to be a good film-forming material due to its high amylose content (∼37%) as compared to many commercial starch polymers (Sahari, Sapuan, Zainudin, & Maleque, 2012). Recently, the authors (Sanyang, Sapuan, Jawaid, Ishak, Sahari, 2015a; 2015b; 2015c) reported the functional properties such as physical, mechanical, thermal and barrier properties of sugar palm starch based films for food packaging.

It has been well documented in literature that starch based products have inherent drawbacks such as high brittleness, poor water vapor barrier and high moisture sensitivity which in turn affects

* Corresponding author at: Green Engineering, Institute of Advanced Technology, Universiti Putra Malaysia, Serdang, 43400 UPM Selangor, Malaysia.
E-mail addresses: sapuan@upm.edu.my, drsapuan@yahoo.com (S.M. Sapuan).

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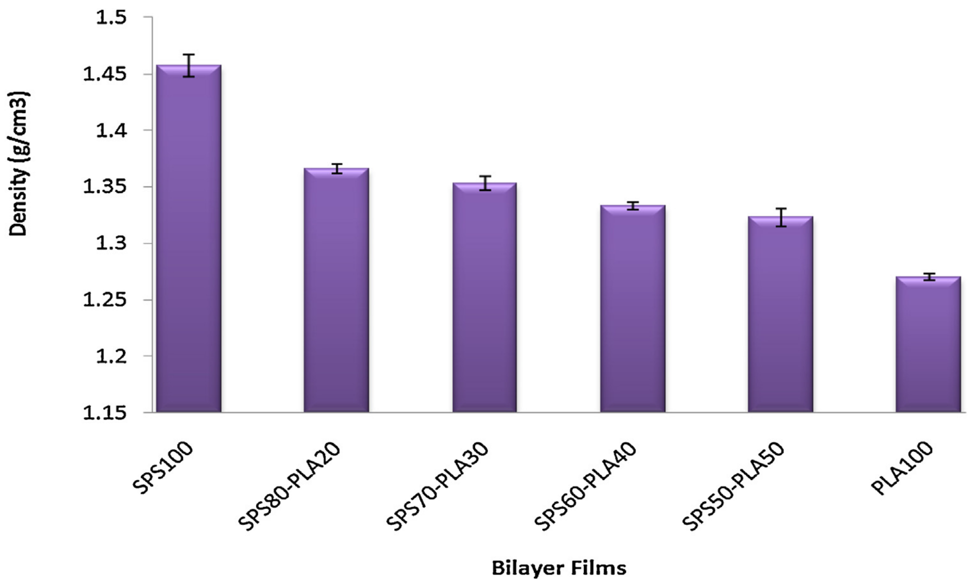

Fig. 1. Density of SPS, PLA and SPS-PLA bilayer films in different proportions.

their mechanical behavior (Bie et al., 2013; Liu, Adhikari, Guo, & Adhikari, 2013; Yu, Dean, & Li, 2006). These drawbacks limit their wide applications and need to be addressed. Thus, the functional properties of SPS based films could be further enhanced by reinforcing SPS film with poly(lactic acid) (PLA) layer film in the form of bilayer films. González and Alvarez Igarzabal (2013) reported the preparation and characterization of biodegradable bilayer films from isolated soy protein (SPI) and PLA. According to these authors, the PLA layer improved the mechanical properties of the bilayer films, whereas it decreases the water vapor permeability and moisture uptake as compared to SPI films without PLA. In their study, Rhim and Ng (2007) and Rhim, Park, and Ha (2013) also developed multilayer films based on plasticized soybean protein isolate and PLA films. They reported that the multilayer films provided good water vapor barrier and improved mechanical properties as compared to those of SPI. In the case of Martucci and Ruseckaite (2010), they prepared a biodegradable three-layer sheet with PLA as outer layers and 30% glycerol plasticized gelatin as the inner layer. They also found that the addition of PLA layers to the plasticized gelatin film significantly reduced the water vapor permeability and increased the mechanical resistance of the native film.

PLA is considered as one of the most promising biodegradable polyesters with various outstanding properties. It is obtained from the fermentation of sugar feedstocks (i.e. corn, sugarcane, etc), which are renewable and are readily biodegradable (Siracusa, Rocculi, Romani, & Rosa, 2008). Apart from being versatile, recyclable and compostable, PLA has high transparency, high molecular weight, and highwater solubility resistance. In addition, it also has good processability as well as high strength and modulus. Moreover, PLA is viewed as the most potential synthetic biopolymer to replace regular conventional plastics in some applications (Xiong et al., 2013). Hence, it is among the most widely used bio-based polymers in numerous applications, including food packaging (Sapuan, Sahari, & Sanyang, 2014). Recently, analysis of a computer based expert system conducted by Sanyang and Sapuan (2015), revealed that PLA is more suitable for food packing than other bio-based materials.The combination of SPS with PLA film to form a bilayer film structure could be an attractive biodegradable food packaging material with improved functional properties.

Studies related to starch/PLA blend films are extensively reported but to the best of our knowledge, there are no studies conducted on the preparation of SPS-PLA bilayer films. Thus, the aim of the current study was to enhance the functional properties of SPS films by combining them with PLA layer. In addition, the effect of PLA layer at different proportions on the physical, mechanical, thermal and water vapor barrier properties of SPS films were investigated.

2. Materials and methods

2.1. Materials

The sugar palm starch (SPS) was extracted from sugar palm tree at Jempol, Negeri Sembilan in Malaysia. SPS used in this work constitute of 37.8% amylose and 62.2% amylopectin. Poly(lactic acid) resins from NatureWork 2000D were supplied by LGC Scientific. Reagents grade of glycerol, sorbitol and chloroform were utilized to help in processing SPS and PLA.

2.2. Preparation of SPS-PLA bilayer films

Sugar palm starch and PLA blend films were prepared using casting method. Both polymers were dissolved separately in two different solvents before finally incorporating them together. In brief, SPS film was prepared by adding 10 g of SPS to 125 mL of distilled water to form 8% (w/w) aqueous dispersion. Thereafter, the film forming solution was heated at $95 \pm 2\,^{\circ}$C for 15 min under constant stirring in a hot water bath. This step was observed to provide homogeneous dispersion of the solution by disintegrating the starch granules. Subsequently, 30% (w/w, starch basis) of glycerol and sorbitol combination (1:1) was added into the dispersions as plasticizer. After an additional 15 min heating process at $95 \pm 2\,^{\circ}$C, the film forming solutions were left to cool, and then casted in glass petri-dishes. The glass petri-dishes served as casting surface which gives the film smooth and flat surface. The freshly cast films were finally dried in an oven at 40 °C.

Prior to the SPS-PLA bilayer formation, PLA layer film was prepared by dissolving PLA resin in chloroform (10% by weight). This was performed by mechanically stirring the solution at a constant temperature of 60 °C for 1 h. SPS-PLA bilayer were produced in the form of thin films by pouring the PLA solution in petri dishes (10 cm diameter) containing the dried SPS films and the fresh films were left to dry overnight at room temperature ($25 \pm 2\,^{\circ}$C). The two layers were incorporated in different ratio (SPS-PLA): (100–0, 80–20,