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Producing novel sago starch based food packaging films by incorporating lignin isolated from oil palm black liquor waste

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ABSTRACT

Poor mechanical and barrier properties of starch-based films can be improved by incorporating natural polymer such as lignin. In the present study, novel food packaging films were prepared by casting method from sago palm (*Metroxylon sagu*) starch (as film matrix with 30% w/w glycerol as plasticizer) by adding lignin isolated from oil palm black liquor waste (from empty fruit bunch), as a reinforcing material (1, 2, 3, 4 and 5% v/w). Results showed packaging films produced by incorporation of isolated lignin to improve selected thermo-mechanical and barrier properties with significant reduction in water vapor permeability, and improved water resistance and seal strength. It is concluded that lignin isolated from oil palm black liquor waste to have great potential to be explored for food packaging purposes. Moreover, this packaging film will be more economical and environmental friendly.

1. Introduction

Of late, tremendous increase has been witnessed (~up to 200 million tonnes) in the global consumption of petroleum-based, non-biodegradable plastics (Siracusa et al., 2008). These synthetic polymers (e.g. polyesters, polyvinylchloride, polystyrene) find extensive applications in food packaging industries used to preserve fresh agriculture produce, dairy and meat products (Tharanathan, 2003; Siracusa et al., 2008; Sanye et al., 2012). Even though, synthetic or petroleum based plastics are physically strong and are economical, with favorable mechanical properties such as tensile and tear strength, heat seal ability, good barrier properties to oxygen and aroma compounds; being non-biodegradable, they tend to cause severe environmental waste disposal problems (Farris et al., 2009; Voon et al., 2012; Yang et al., 2012). Additionally, recycling of petroleum based plastics used for food packaging purposes is difficult, as foodstuffs sticking to them often contaminates them.

Depletion of natural resources world-over has raised the cost of petroleum-derived commodities, which has made it mandatory to explore for economical and effective substitutes (Voon et al., 2012). Increasing awareness on these vital issues has led researchers to explore and develop potential replacement for synthetic plastics by using raw materials from natural, renewable resources. One approach to overcome these problems is to produce biodegradable, biopolymer based plastics from polysaccharides or proteins, which can be an excellent renewable source, especially to develop food-packaging materials (Voon et al., 2012). Even though, biopolymer based films are not the ultimate replacement to petroleum-based plastics, it is expected that the packaging material developed can be an efficient barrier to moisture, gas, and reduce lipid peroxidation along with retaining natural aroma of a food commodity. Green packaging materials or bio-plastics has been estimated to contribute 5–10% of the present plastic market, which is approximately 50,000 tonnes in Europe alone (Siracusa et al., 2008).

Starch is one of the naturally occurring biopolymers, which are economical biodegradable resource and has easy availability. Of late, starch has received extensive attention in packaging industries for producing commercial thermoplastic polymers (Zhang and Liu, 2008; Chang et al., 2010). In Malaysia, sago-starch is exclusively produced from sago palm plant (*Metroxylon sagu*) and has tremendous potential to be explored to develop biodegradable packaging materials. Due to high hydrophilic nature, packaging films produced from starch possesses poor barrier (against gas and moisture) and mechanical properties. This limits their potential to be used as a basic raw material for developing biodegradable packaging materials.

Palm oil production industry is one of the leading food industries offering not only employment, but also helping to earn good foreign exchequer. During oil extraction process, large volume of oil palm biomass wastes are produced (from oil palm trunks, oil

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palm fronds, empty fruit bunches, shell, fibers) (Chew and Bhatia, 2008; Bhat et al., 2009). Additionally, during oil extraction process, large volume of black liquor wastes (BLW) (an aqueous solution of lignin, cellulose, hemi-cellulose, and inorganic chemicals used during oil extraction process) is produced as by-product. This BLW if left untreated can cause severe environmental damages (such as fouling of environment) and health risks (Bhat et al., 2009).

Lignin is a useful "waste material" that possesses tremendous potential to be used as a base material for various materials applications (Stewart, 2008). Lignin, a natural renewable polymer is abundantly present in oil palm BLW. Generally, the lignin content in spent liquor wastes (such as in kraft, Sulfite or soda liquors) varies between 15% and 55%. However, the exact lignin content obtained from oil palm BLW cannot be estimated accurately as this can vary depending on the extraction or the pulping procedures employed. Plant derived lignin are non-toxic and are known to exhibit rich antioxidant, anti-fungal, anti-cancer, anti-viral activities (Bhat et al., 2009). Lignin has been reported to posses' certain environmental and industrially useful properties such as: high modification options, easy biodegradability, better adhesiveness, adsorption, and solution properties, along with good compatibility with various non-toxic chemicals (Ciobanu et al., 2004). Also, lignin isolated from oil palm BLW has been reported to possess comparable functional groups (based on FTIR) to those of commercial lignin (Bhat et al., 2009), which in turn provides a strong basis to be explored for food industry applications, such as in the development of packaging films. Our hypothesis of undertaking this study was that strong intermolecular interactions (e.g. between OH group of lignin and H_2 molecule of sago starch) can occur leading to three way interactions (starch + lignin + starch), which is expected to 'space out' the distance between individual starch chain, thus resulting in development of films with improved thermo-mechanical properties.

Based on these facts, the main objective to undertake the present study was to incorporate lignin isolated from BLW to develop novel, sago-starch based packaging films with good mechanical and thermal properties, which is envisaged to find wide applicability for food packaging purposes and also be useful to overcome relevant environmental issues.

2. Materials and methods

2.1. Material

Oil palm black liquor waste was collected from Division of Bioresource, Paper and Coatings Technology, University Sains Malaysia. Commercial lignin (lignin alkali with low sulfonate content, Batch, 04414PE, 4% sulfur, pH 10.5) was obtained from Sigma–Aldrich (Sigma, U.S.A.). Sago starch was purchased from Sim Company Sdn. Bhd. (Penang, Malaysia), while Di-methyl sulfoxide (DMSO) (90%) was purchased from Fisher Scientific Company, UK. Glycerol was obtained from R M Marketing Company, Essex, UK.

2.2. Lignin from black liquor wastes

A mixture of lignin and sodium hydroxide (NaOH) in solid form (~1 kg) was procured from Metro Knight (M) Sdn. Bhd. (Malaysia). This lignin was prepared after spray drying oil palm black liquor wastes and the protocol followed to obtain pure lignin (used in the present study) is provided as a schematic representation in Fig. 1.

2.3. Film preparation

The film forming solutions were prepared by addition of a known percentage of sago-starch with distilled water (4 g sago starch/100 ml water) at room temperature. Glycerol was added as plasticizer at a concentration of 30% (30 g glycerol/100 g sago starch). Lignin (isolated lignin and commercial lignin) was initially dissolved with 90% DMSO and this solution was incorporated into the film solution at different concentrations (1, 2, 3, 4 and 5% v/w of film forming solution). Subsequently, the solution was heated in a hot water bath (at 85 °C) under constant magnetic stirring (at 350 rpm for 45 min) and the resulting solution was cooled to room temperature.

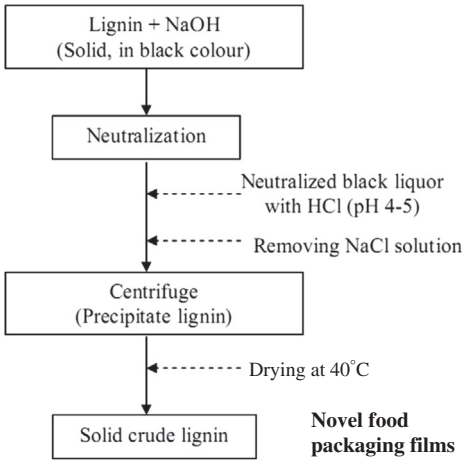

Fig. 1. Schematic representation of extracting lignin from oil palm black liquor waste.

The film thickness was controlled by casting same amount of film solution onto Poly-acrylic plates (16 × 16 cm) (90 g). After casting, films were air dried for 24 h (at 25 ± 1 °C) followed by oven drying at 40 °C for next 24 h. Further, the dried films were peeled manually and stored in desiccators maintained (at 25 ± 1 °C) at 58% relative humidity (RH) (saturated sodium bromide solution) until further evaluation. Prior to use, conditioning of samples were performed by keeping the desiccators maintained at 0% (silica gel) and at 58% RH. Films produced without addition of lignin served as control.

2.4. Film thickness

The thickness of each individual film produced was measured using a hand-held micrometer (dial thickness gauge 7305; Mitutoyo Co, Tokyo, Japan) to a nearest of 0.01 mm. The thickness was measured at randomly selected locations on each film (n = 10) and the mean values were recorded.

2.5. Mechanical properties of films

By employing standard methods (ASTM, 1980, 1980), the tensile strength (TS), elongation at break (EB) and elastic modulus (EM) of prepared films were measured using an TA-XT plus Texture Analyzer (Stable Micro System, UK). Prepared films were cut 10 × 2 cm and the strips were pre-conditioned as detailed earlier (Section 2.3). Before testing, thickness of the strips were measured at different points (app. 10), and the mean thickness was used to calculate the mechanical properties of the films.

In this experiment, a 5 'kg' load cell was used for the above elaborated measurements. Filmstrips were clamped between the tensile grips probe (A/TG) with initial distance between the grips set at 50 mm and cross head speed at 0.40 mm/s. From the initial slope of linear part of stress–strain diagram, Tensile strength (TS, MPa), elongation at break (EB, %) and elastic modulus (E, MPa%) were