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Development and characterization of a novel biodegradable edible film obtained from psyllium seed (*Plantago ovata* Forsk)

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ABSTRACT

In this study, the physical, thermal and mechanical properties of a novel edible film based on psyllium hydrocolloid (PH) were investigated. PH films were prepared by incorporation of three levels of glycerol (15%, 25%, and 35% w/w). As glycerol concentration increased, water vapor permeability (WVP), percent of elongation (*E*%) and water solubility of PH films increased whilst, tensile strength (TS), surface hydrophobicity and glass transition point (T_g) decreased significantly. At the level of 15% (W/W) of glycerol, PH films showed the lowest WVP values (1.16×10^{-10} g H_2O m^{-2} s^{-1} MPa^{-1}), *E*% (24.57%) and water solubility (47.69%) and the highest values for TS (14.31 MPa), water contact angle (84.47°) and T_g (175.2 °C). By increasing glycerol concentration, PH films became slightly greenish and yellowish in color but still transparent in appearance. This study revealed that the psyllium hydrocolloid had a good potential to be used in producing edible films with interesting specifications.

1. Introduction

Due to the continuous growing of global population and decreasing rate of food production, protection and maintenance of food becomes more and more important and vital. One of the primary and important ways is to use healthy packaging materials (Mariniello et al., 2003). In recent years, synthetic petroleum-based polymers have been widely used in food packaging, but these materials are not biodegradable and increase environmental pollution (Pérez et al., 2009) and some of them generate carcinogenic compounds. On the other hand, consumers' interest in high quality food has accompanied with environmental concerns (Moreira et al., 2009). In recent years, many researchers focused on producing edible films based on biopolymers like polysaccharides, proteins, and fats (Andreuccetti et al., 2011; Ghanbarzadeh et al., 2011). Because biopolymers are biodegradable and environmentally friendly materials, they will reduce the amount of chemical hazards and domestic wastes (Arvanitoyannis, 1999; Avila-Sosa et al., 2010).

Although all of edible films are not good barriers against water vapor, they could be used as a carrier of active compounds, antimicrobial agents or preservatives, which protect food quality (Campos et al., 2011; Pérez et al., 2009). In addition to good mechanical properties, most of edible films are potentially used to extend the shelf-life and improve the quality of almost any food systems by serving as mass transfer barriers to gases, lipid, flavor or aroma (Jiménez et al., 2010).

Various polysaccharides have been used for the preparation of edible films including starch (Osés et al., 2009), tapioca (Vásconez et al., 2009), corn (Psomiadou et al., 1996), cellulose and cellulose derivatives such as HPMC (hydroxypropyl-methylcellulose), CMC (carboxymethylcellulose), and MC (methylcellulose) (Pérez et al., 2009; Sánchez-González et al., 2009). Pullulan (Shih et al., 2011), alginate, carrageenan (Yang and Paulson, 2000), and policaju gum (Carneiro-da-Cunha et al., 2009) are also used in preparing edible films. Moreover, pectin, gellan (Pérez et al., 2009) and chitosan [poly-b-(1 → 4)-*N*-acetyl-D-glucosamine] with different deacetylation degrees and molecular weights are also used for making edible films (Vásconez et al., 2009). Sometimes the blend of these biopolymers with other biopolymers, glycerol (as a plasticizer), hydrophobic substances and/or antimicrobial compounds have been widely made to improve the physical, mechanical, functional, organoleptic, and nutritional properties of prepared edible films (Gaudin et al., 1999; Myllärinen et al., 2002; Vásconez et al., 2009). Plasticizers decrease intermolecular attractions between nearby polymeric chains. As a result, they generate more flexibility and possibly significant changes in barrier properties of the final film (García et al., 2000).

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Psyllium is an annual plant from the *Plantago* genus (Craeyveld et al., 2009). Around 200 species of this genus broadly distributed all over the moderate regions of the world (Guo et al., 2009). Species from the *Plantago* in the name of *Plantago ovata* Forsk are widely grown in India and Iran. Common names of *P. ovata* Forsk include Blond psyllium, Indian plantago, Ispaghula (meaning "horse ear" in Indian) and Spagel (Guo et al., 2009). The psyllium (*P. ovata* Forsk) seed husk which is a well-known source for the production of psyllium hydrocolloid (PH) (Craeyveld et al., 2009) is widely utilized in pharmaceutical and food industries (Singh, 2007; Yu et al., 2003). Recently, psyllium has been imaginatively used in the domestic and industrial wastewater treatment – as it is effective in removing solid waste and dye from tannery, domestic and textile wastewater – and landscape industry (Guo et al., 2008, 2009; Mishra and Bajpai, 2005). The psyllium is utilized in pharmaceutical industries as a medicinally bioactive polysaccharide and used for the medical treatment of constipation, colon cancer, diarrhea, high cholesterol, diabetes and inflammation bowel diseases – ulcerative colitis (Singh, 2007). Additionally, it is also used in food industries as constituting the gel and enhancing the consistency and stability (Bemiller and Whister, 1996). According to the above mentioned properties, all of these specifications mostly attribute to hydrocolloid nature of psyllium husk. The PH is a complex polysaccharide which is composed of a highly branched non-starch polysaccharide with primarily a main chain contains $(1 \rightarrow 4)$ and $(1 \rightarrow 3)$ linkages, where the side chains are connected to the main chain via O-3 and/or O-2 linkage. The main chain of this polysaccharide composes of xylopyranose residues. Likewise, its side chains including single arabinofuranose and xylopyranose residues or short side chains consisting of these monosaccharides (Craeyveld et al., 2009; Guo et al., 2008; Singh, 2007).

Considering all mentioned beneficial characteristics of PH in addition to the low production cost (in comparison with most biopolymers), the aim of this study was to investigate the possibility of producing a novel biodegradable edible film from PH with glycerol as plasticizer in the different concentrations, and study its physical and mechanical properties.

2. Materials and methods

2.1. Materials

The psyllium seeds and ethanol (with 96% purity) were obtained from the local medical market in Tehran and Razi Corporation (Tehran, Iran), respectively. Other materials including glycerol, calcium nitrate and calcium chloride were purchased from Merck Corporation (Whitehouse Station, NJ 08889-0100 USA).

2.2. Extraction of psyllium hydrocolloid and preparation of edible film solution

Sequential processes employed to extract PH from psyllium seeds. About 10 g psyllium seeds sieved and washed with its triple weight of ethanol (96% w/v) for 15 min under constant stirring. This procedure repeated three times to remove all foreign matter such as dust, dirt, stones, and chaff. After removing ethanol and drying the seeds in an oven at 70 °C, to extract the PH, an electronic Stirrer (RZR 2102 control, Heidolph, Germany) was used to disperse cleaned seeds in 100 mL distilled water at 80 ± 3 °C for 1 h under stirring at 600 rpm constantly. To prevent the water loss as a result of evaporation, water (80 °C) was added at different intervals to the system. The insoluble non-carbohydrate fractions of psyllium seeds removed by filtration and the dry matter of filtrate (or dry hydrocolloid) was determined (28%). Enough seed used for extracting hydrocolloid needed for preparing film-forming solution containing 1.2% (w/v) hydrocolloid concentration. Mixtures containing filtrate and glycerol in 15%, 25%, and 35% (w/w) concentration were prepared under constant stirring (750 rpm) at 80 ± 3 °C for 5 min. Then a vacuum-oven applied for 15 min at 80 ± 3 °C to remove air bubbles. The prepared solution was casted onto a Teflon plate and dried at room temperature (23 ± 2 °C) and room relative humidity for 48 h. Finally, the obtained PH films were peeled from plates and conditioned at 23 ± 2 °C in desiccators containing saturated solutions of Ca $(NO_3)_2$, $6H_2O$ (50 ± 2% relative humidity, RH) for at least 48 h prior to tests.

2.3. Edible film physical properties

2.3.1. Moisture content

The prepared film samples (3×3 cm^2) were dried at 103 °C in a laboratory oven (Blue M Electric Co., Blue Island, IL) and their moisture content (MC) was determined gravimetrically in different time intervals until their weights become constant. Three replications of each film sample were used for calculation of moisture content.

2.3.2. Film thickness

A digital micrometer (Mitutoyo Corp. MDC-1 SB digital micrometer, Japan) with accuracy of 0.001 mm used at least on 10 random positions of the film sample to determine the thickness. The mean values of film thickness were the basic data for calculation of mechanical and physical properties.

2.3.3. Total soluble matter (TSM)

PH films were dried at 103 ± 2 °C for 24 h in a laboratory oven (Blue M Electric Co., Blue Island, IL), and weighed to determine the initial dry weight. Dried pieces of film samples were directly immersed in 50 mL of distilled water (25 °C for 3 h) under stable stirring (600 rpm) using a magnetic mixer (Hanna, HI 190 M). The obtained solution then filtered through a (Whatman #1) qualitative filter paper and dried in an oven (103 °C for 24 h) to separate the insoluble portion of dried film. Then the following formula (Eq. (1)) used to calculate the dissolved portion of dried film in water:

$$\%\mathrm{TSM} = [(\text{initial dried film} - \text{insoluble dried film})/\text{initial dried film}] \times 100 \quad (1)$$

All the experiments repeated three times, and their arithmetic averages were reported.

2.3.4. Surface hydrophobicity

The surface hydrophobicity of the PH films was calculated from contact angle measurement using a goniometer (Kruss G10, Germany) with a 5 μL drop of distillated water on the surface of a film sample with dimensions of 4.0×4.0 cm^2 at 25 °C. The angle of the tangent to the basis of the droplet (contact angle) was measured and expressed in degrees. To obtain a reliable contact angle for each film sample, this test repeated at least six times and the mean values were calculated.

2.4. Color measurement

A CIE colorimeter (Minolta CR 300 Series, Minolta Camera Co. Ltd., Osaka, Japan) was used to determine the color of PH films. The colorimeter was calibrated using a standard white plate ($L_* = 97.10$, $a_* = +0.13$, $b_* = +1.88$, $c_* = 1.88$). Later, the color measurements were performed by placing the film sample over CIE colorimeter. Instrumental color readings were lightness or L (0 = black, 100 = white) and chromaticity parameters or a (−60 = greenness to +60 = redness), and b (−60 = blueness to +60 = yellowness). At least three points of each sample were