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Effect of corn oil on physical, thermal, and antifungal properties of gelatin-based nanocomposite films containing nano chitin

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ABSTRACT

Effects of corn oil on physical and antifungal properties of gelatin nanocomposite films containing chitin nano fibers (N-chitin) were investigated in this study. Different concentrations of corn oil (0.10, 0.20, and 0.30; g/g of dry gelatin) and (0.05 g/g of dry gelatin) N-chitin were used to prepare an emulsion nanocomposite gelatin film. Subsequently, characteristics of films were assessed by analyzing moisture content, solubility, hydrophobicity of the surface, water vapor permeability (WVP), as well as mechanical, thermal, and antifungal properties. The results obtained showed that films containing N-chitin had better mechanical properties than net gelatin film. In addition, N-chitin decreased film tendency to water absorption. However, because both gelatin and N-chitin have hydrophilic nature, nanocomposite films were still wettable. In this sense, incorporation of oil into film formulation improved barrier properties more than nanoparticles alone. In spite of the fact that gelatin films containing just N-chitin had antifungal properties, emulsion nanocomposite films did not show any antimicrobial activity. In addition, improving effect of oil on film properties was observed for concentrations up to 0.20 g/g, because films containing 0.30 g/g oil possessed weaker mechanical and thermal stability. Therefore, around 0.20 g/g of oil in gelatin nanocomposite films can lead to create biodegradable polymers with more acceptable properties for food packaging.

1. Introduction

In recent years, a vast array of studies have concentrated on substituting synthetic polyolefin polymers with degradable packaging, due to undesirable environmental effects of the first and also present concerns about lack of fossil energies (Ge et al., 2015). In this regard, it has been attempted to produce edible packaging for foods, provided from carbohydrates, proteins or composition of these materials with lipids (Kanmani & Rhim, 2013). However, because of some deficiencies such as poor mechanical, barrier and thermal properties of biodegradable packaging, they still couldn't be utilized in industrial scale (Carvalho & Grosso, 2014). In order to remove these unpleasant properties of degradable packaging, a number of approaches have been studied such as making different compositions of substances to introduce synergistic effect on their barrier properties (Limpisophon, Tanaka, & Osako, 2010), applying plasticizers, cross linking agents and different fillers to elevate packaging properties (Martucci, Accareddu, & Ruseckaite, 2012), and as the recent and the most interesting approach, using nanoparticles which can not only improve physical properties of biopolymers but also may add antibacterial, antioxidant and nutraceutical capacities to them (Nafchi, Moradpour, Saeidi, & Alias, 2014). Incorporation of nanoparticles into polymer formulation makes nanocomposite films identified as solid materials including at least one component at nano size. Generally, nano particles are classified as both organic and inorganic materials that can be made of existing natural or synthetic substances.

One of the most interesting organic nano materials is nano chitin which is considerably known for its high availability and profitable capacities such as antibacterial effect, low cost, and non-toxicity (Rubentheren, Ward, Chee, & Tang, 2015). So far, nano chitin has been studied as a filler and antibacterial agent in

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chitosan, starch, carrageenan, and soy protein degradable polymers (Chang, Jian, Yu, & Ma, 2010; Li, Li, Ke, Shi, & Du, 2011; Lu, Weng, & Zhang, 2004; Rubentheren et al., 2015; & Shankar, Reddy, Rhima, & Kim, 2015). Regarding the fact that chitin has hydrophilic nature compatible to the most of polysaccharide and protein polymers and can be dispersed homogenously in their matrices (Chang et al., 2010; Lu et al., 2004), it can be proper nano filler for gelatin biopolymer to alleviate its poor thermal, mechanical and barrier properties. However, the effect of chitin nanofiller on water vapor barrier properties is not considerable, since N-chitin itself is a hydrophilic material whose improving effect on water vapor resistance of films is limited to creating a tortuous pass way across the polymer (Rubentheren et al., 2015; Shankar et al., 2015).

The most important reason for including lipids to films' composition is their hydrophobic nature which can decrease biopolymers' water absorption and permeability. Concerning this, different studies have focused on preparation of emulsion films with high barrier properties against water vapor and gases. For example, Limpisophon et al. (2010) studied improving effect of stearic acid and oleic acid on water vapor barrier properties of gelatin films. In another study, Chiumarelli and Hubinger (2014) produced biodegradable films based on cassava starch containing carnauba wax and stearic acid and assessed optimum formulation for better barrier properties. In addition, incorporation of lipids to edible films can improve water vapor barrier properties however; they decrease thermal and mechanical quality of the films (Bourlieu, Guillard, Vallès-Pamiès, Guilbert, & Gontard, 2009). Considering mentioned reasons, it is necessary to apply different reagents together to improve the thermal, mechanical and barrier properties of films.

Therefore, gelatin film containing N-chitin and different concentrations of corn oil have been prepared in order to study the effects of nanoparticle and oil on packaging properties of gelatin biopolymer.

2. Materials and methods

2.1. Materials

Bovine gelatin was provided by Merck Chemical Co. (Darmstadt, Germany), which has the bloom number of 200 and density of 1358 kg/m^3. A commercial brand of corn oil, containing 100% lipids (11.42% saturated and 88.57% unsaturated) was purchased from Famila Co. (Iran). Glycerol and 50% glutaraldehyde were supplied by Sigma Chemical Co. (St. Louis, MO, USA). Chitin nano particles were obtained from Nano-shimi Yakhte Co. (Mazandaran, Iran) which had particle size of 50–70 nm. *Aspergillus niger* strain was received from Microbial collection of Drug (Applied Research Center, Tabriz university of medical science).

2.2. Preparation of gelatin/N-chitin film emulsions

The procedure of gelatin emulsion nanocomposite film preparation has been shown in Fig. 1. Chitin nanoparticles (0.05 g/g of dry gelatin) were dispersed in 100 ml distilled water with a magnetic stirrer for one hour. Then, the solution was sonicated in an ultrasonic bath (Parsonic 30S, Pars Nahand engineering Co. Iran) for further 30 min. Four gram dry gelatin was added to the 100 ml distilled water and held for 30 min for swelling and solving in water. Later, corn oil with concentrations of 0.10, 0.20 and 0.30 g/g of dry gelatin and Tween-80 (0.02 g/g of oil) were added to the solution and heated up to 55 °C while stirring for 30 min. After decreasing the mixture temperature to 35 °C, glycerol and 50% glutaraldehyde were added to the solution by concentrations of 0.30 and 0.01 g/g (based on dry gelatin), respectively, and mixed for

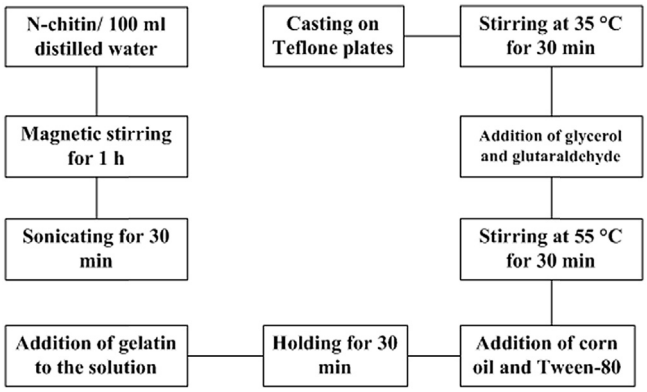

Fig. 1. Schematic diagram of gelatin emulsion nanocomposite film preparation.

30 min. Finally, emulsions were cast on Teflon plates of diameter of 16 cm and dried for 48 h at room temperature. All the film samples were prepared in triplicate and conditioned at 57 ± 2% RH for 48 h before testing (Mohammadi Nafchi et al., 2014; Seyedi, Koocheki, Mohebbi, & Zahedi, 2015).

2.3. Characterization of emulsion nanocomposite films

2.3.1. Moisture content and solubility

Moisture content of the films were measured using the method described by Rangel-Marron, Montalvo-Paquini, Palou, and Lopez-Malo (2013) in which 2 × 2 cm^2 pieces of the films were weighed before and after oven-drying at 105 °C for 24 h and weight loss percentage was reported as moisture content.

In order to determine water solubility of the films, oven-dried pieces of films were immersed in 50 ml distilled water and held for 24 h with gentle shaking by magnetic stirrer at 200 rpm. Then, films were strained and dried at 105 °C for 24 h. Subsequently, the solubility of the films was calculated as follows:

$$\%S = \frac{(m_i - m_d)}{m_i} \times 100 \quad (1)$$

where S is solubility of the film, m_i is the initial weight of dry film before immersing in water and m_d is the final weight of dried films. All of the experiments were done in triplicate.

2.3.2. Hydrophobicity of the film surface

To investigate the surface hydrophobicity of the films, water drop contact angle on the film surface was measured according to the method reported by Liu, Antoniou, Li, Ma, and Zhong (2015); First, Flat pieces of the films (2 × 2 cm^2) were placed on a smooth dark surface of a black box. Then, 10 μl distilled water was placed by a syringe as a drop on the film surface. After that, images of both sides of the water drop were recorded by digital camera (Canon, IXY, 12.1 megapixels). Finally, the contact angle of drop was assessed by Adobe Acrobat 9.0 professional and results were stated as an average of three measurements.

2.3.3. Water vapor permeability (WVP)

The WVP of the films were determined according to the ASTM E96-95 standard method, in which glass vials with average diameter of 1.5 cm and depth of 4 cm were equipped with perforated doors with average hole diameter of 4 mm. In order to create a conditioned atmosphere inside and outside of the vials dry $CaSO_4$ salt (RH ~ 0%) and saturated K_2SO_4 solution (RH ~ 97% at 25 °C) were applied, respectively. Film samples were fixed on top of the vials by their doors and the whole vial was weighed at 24 h intervals during