International Journal of Biological Macromolecules 59 (2013) 282–289

Contents lists available at SciVerse ScienceDirect

International Journal of Biological Macromolecules

journal homepage: www.elsevier.com/locate/ijbiomac

Development of tea extracts and chitosan composite films for active packaging materials

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ARTICLE INFO

Article history:
Received 27 February 2013
Received in revised form 19 March 2013
Accepted 9 April 2013
Available online 18 April 2013

Keywords:
Tea extracts
Chitosan film
Antioxidant ability
Food simulants
Fourier transform infrared spectrometry

ABSTRACT

The effects of 0.5%, 1% and 2% green tea extracts (GTE) and black tea extracts (BTE) on the physical, structural and antioxidant properties of chitosan films were investigated. Results showed that the addition of tea extracts significantly decreased water vapour permeability and increased the antioxidant ability of films. The DPPH radical scavenging ability of GTE films was stronger than that of BTE films in all food simulants (0%, 20%, 75% and 95% ethanol). The equilibration time in different food simulants decreased with the increased ethanol concentration. DSC and FTIR spectra analysis indicated that there was strong interaction in film matrix, which could be reflected by the physical and mechanical properties of composite films. This study revealed that an active chitosan film could be obtained by incorporation of tea extracts, which may provide new formulation options for developing an antioxidant active packaging.

1. Introduction

Recently, one of the major challenges in food preservation is the design of active food packaging. Active food packaging may carry antioxidants and antimicrobial agents in packaging systems [1], which could be more effective than antioxidant and antibacterial substances directly applied to the food surface due to providing continuous release of active additives from packaging materials. Moreover, with the increasing health concerns of the consumers, current packaging researches pay more attention to the use of natural additives in edible packaging materials [2–5].

Chitosan, a linear polysaccharide consisting of β-1,4-linked D-glucosamine and N-acetyl-D-glucosamine [6,7], is considered to be a biobased environmentally friendly material which is found to be nontoxic, biodegradable and with antioxidant characteristics [8]. In addition, featured by being able to incorporate functional substances and plant essential oil, chitosan has the biocompatible advantage over other biobased packaging materials [3,8,9]. Therefore, chitosan has the potential to be used as substitute resource of active food packaging and may have bright application prospect in food processing industry [1].

Tea is one of the most consumed beverages in the world for its bioactive compounds associated with numerous health benefits [10,11]. And tea extracts has been generally demonstrated to be powerful antioxidants. In some studies, epigallocatechin gallate (EGCG, the major catechin in tea extracts) is 20 times more active than vitamin C and 30 times more than vitamin E [11]. Depending on the manufacturing process, tea is classified into three major types: non-fermented green tea (produced by drying and steaming the fresh leaves), semi-fermented oolong tea (produced after undergoing a partial fermentation stage prior to drying), and fermented black tea (produced after undergoing a post-harvest fermentation stage prior to drying and steaming) [12]. Studies showed that both green tea and black tea extracts had strong antimicrobial and antioxidant ability [12–14]. The oxidation degree of tea extracts depends on the fermentation process of tea leaves, which is reflected by the capacity of scavenging reactive oxygen and nitrogen species, [12,15]. In food processing industry, tea extracts were reported to delay the oxidation in various foods including soybean oil [16], vegetable oil [14] and dry-fermented sausage [17]. However, little information on the effects of tea extracts on the physical and antioxidant properties of chitosan-based film has been reported.

Being able to determine the antioxidant capacity in different food simulants is important to the design of active packaging film [18]. However, little release kinetics of antioxidant substances from biodegradable films to food products has been explored.

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http://dx.doi.org/10.1016/j.ijbiomac.2013.04.019

Antioxidant release is dependent on the food characteristics, thus it is interesting to evaluate substance migration in different food-simulating solvents. European Food Safety Authority (EFSA) recommendations in terms of food simulants are contained in the European Commission Directive 97/48/EC [19]. Aqueous and acidic foodstuffs are well simulated by distilled water and 10–50% ethanol, while fatty foodstuffs is simulated by substitute fatty simulants such as isooctane or 95% ethanol [4,20].

The purpose of this work was to evaluate the effects of green tea and black tea extracts on the physical, structural and antioxidant properties of chitosan-based films. Moreover, in order to develop an active packaging film, the antioxidant capacity of tea extracts in five different food simulants (aqueous solutions containing 0%, 20%, 75%, 95% ethanol) were also evaluated by analyzing 1,1-diphenyl-2-picrylhydrazyl (DPPH) radical scavenging ability.

2. Materials and methods

2.1. Materials

Crab chitosan (deacetylated degree: 90.2%, viscosity: 62cps at 25 °C) was purchased from AK Biotech Ltd. (Shandong, China). Glycerol, acetic acid, DPPH (1,1-diphenyl-2-picrylhydrazyl), magnesium nitrate ($Mg(NO_3)_2$) and anhydrous calcium chloride ($CaCl_2$) were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Green tea extract (green tea polyphenols: 99.9%, total catechins: 81.2%, EGCG: 51.7%) and black tea extract (Theaflavins: 61.8%, total tea polyphenols: 22.3%, total catechins: 10.2%) used in this study was provided by Fuzhou Corona Science & Technology Development Co., Ltd. (Fujian, China).

2.2. Film preparation

Chitosan (CH) solution (2%, w/v) was prepared by dispersing chitosan in an aqueous solution of glacial acetic acid (1%, w/w) at 25 °C and stirring with a magnetic stirrer (Shanghai Huxi Analysis Instrument Co., Ltd., Shanghai, China) for 4 h. After chitosan was completely dissolved, 30% glycerol (w/w chitosan) was added as the plasticizer and the solution was stirred for 1 h. Then green tea extract (GTE) and black tea extract (BTE) were added into polymer solutions to reach a final concentration of 0.5%, 1% and 2% (g/100 ml film solutions). All the solutions were homogenized by using a rotor–stator homogenizer (IKA T25-Digital Ultra-Turrax, Staufen, Germany) at 13,500 rpm for 2 min. Finally, these film-forming solutions were vacuum degasified at room temperature (25 °C) with a vacuum pump for 1 h to remove air bubbles. Sample nomenclature was CH-nGTE or CH-nBTE, the n value being the addition content of tea extract in 100 ml film solutions.

A casting method was used to obtain films. Film-forming solutions (200 g) were casted over the leveled glass plates (25 cm × 25 cm) and dried at 25 ± 1 °C and 60 ± 2% relative humidity for 48 h. Then, the dried films were carefully peeled from the plates and stored for 48 h in desiccators containing $Mg(NO_3)_2$ saturated solutions (53% relative humidity) at 25 °C before further tests.

2.3. Rheological behavior

The rheological behavior of film-forming solutions was analyzed according to the method described by Sánchez-González et al. [21] by using a R/S plus rotational rheometer (Brookfield Engineering Laboratories, Inc., Middleboro, MA, USA) with a fixed outer cylinder and rotating measuring bob. The radius of rotating cylinder was 12.50 mm, the length of the cylinder and the gap width was 50.00 mm and 2.00 mm, respectively. Rheological curves were obtained after a stabilization time of 5 min at 25 °C. The shear stress (σ) was measured as a function of shear rate (γ) from 0 to 512 s^{-1}, taking 5 min to reach the maximum shear rate and another 5 min to attain zero shear rate. Experiment data were fitted to the Ostwald de Waale model (Eq. (1)) in order to determine the consistency (k) and the flow behavior indexes (n). Apparent viscosities were calculated at 100 s^{-1}. The measurements were performed in triplicate at 25 °C.

$$\sigma = k\gamma^n \quad (1)$$

2.4. Film thickness and water content

A hand-held micrometer (with an accuracy of 0.01 mm) was used to measure the thickness of the film. Six replications were conducted for each sample treatment at random position.

Films were weighed before and after drying in an oven at 105 °C for 24 h. Water content was calculated as follows:

$$\text{Water content} = \frac{M_0 - M}{M_0} \quad (2)$$

where M_0 was the initial film mass (g) and M was the bone-dry mass (g). Water content was expressed as g H_2O/g wet base.

2.5. Water vapour permeability

Water vapour permeability (WVP) was determined gravimetrically based on the method described by Talja et al. [22] with some modifications. The films (60 mm × 60 mm) were sealed onto permeation cells (rigid plastic with wall thickness: 1.8 mm, inner diameter: 42 mm, height: 25 mm) filled with granular (Φ < 2 mm) anhydrous calcium chloride. The permeation cell was covered with a film sample attached with tape to ensure there is no leakage. The stagnant air gap under the films was less than 6 mm. The weight of calcium chloride was maintained at 20.5 ± 0.2 g for each treatment to ensure the consistent air gap. The permeation cells were then placed in desiccators containing saturated NaCl solutions, providing RH gradients of 0/75% at 25 °C. The permeation cells were weighed until changes in the weight were recorded to be the nearest 0.001 g. WVP was calculated as follows:

$$\text{WVP} = \frac{mL}{At\Delta p} \quad (3)$$

where m was the weight of water permeated through the film (g), L was the thickness of the film (m), A was the permeation area (m^2), t was the time of permeation (s), and Δp was water vapour pressure difference across the film (Pa). Five repetitions were performed for each film sample.

2.6. Film solubility and swelling degree

The solubility and swelling degree of the films were determined according to the methods described by Silva et al. [23] and Zhong et al. [24] with some modifications. Film pieces (25 mm × 25 mm) were dried at 70 °C for 24 h in a vacuum oven (Shanghai Yiheng Technology Co., Ltd., Shanghai, China) to get the initial dry mass (M_1). Then the films were placed in 50 ml beakers with 30 ml distilled water. The beakers were covered with plastic wraps and stored at 25 °C for 24 h. Water remaining in the beakers was discarded and the residual film pieces were dried superficially with filter paper. The residual film pieces (M_2) were again dried at 70 °C for 24 h in a vacuum oven to determine the final dry mass (M_3). Three measurements were taken for each film sample. Film solubility and swelling degree were calculated by using the following equations respectively:

$$\text{Film solubility} = \frac{(M_1 - M_3) \times 100}{M_1} \quad (4)$$