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Functional chitosan-based grapefruit seed extract composite films for applications in food packaging technology

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ABSTRACT

Chitosan-based composite films with different amounts of grapefruit seed extract (GFSE) (0.5, 1.0 and 1.5% v/v) were fabricated via solution casting technique. Experimental results showed that GFSE was uniformly dispersed within all chitosan film matrices. The presence of GFSE made the films more amorphous and tensile strength decreased, while elongation at break values increased as GFSE content increased. Results from the measurement of light transmission revealed that increasing amounts of GFSE (from 0.5 to 1.5% v/v) did not affect transparency of the films. Furthermore, packaging of bread samples with chitosan-based GFSE composite films inhibited the proliferation of fungal growth as compared to control samples. Hence, chitosan-based GFSE composite films have the potential to be a useful material in the area of food technology.

1. Introduction

In recent years, increasing attention has been directed to the development of films with antimicrobial and antifungal properties in order to improve food safety, extend shelf-life and to minimize the use of chemical preservatives [1]. Consumers are also demanding the packaging materials to be formulated from natural materials, environmentally friendly and biodegradable while improving food preservation [2–7]. Chitosan, a natural biopolymer has immense potential for applications in food technology, owing to its biocompatibility, non-toxicity, short time biodegradability and excellent film forming ability. Besides, chitosan has inherent antimicrobial and antifungal properties against various groups of pathogenic and spoilage microorganisms [8].

To enhance the antibacterial and antioxidative properties of chitosan, antimicrobials could be incorporated into it to achieve a synergistic effect. Among the antimicrobial agents used in packaging films, grapefruit seed extract (GFSE) is antioxidant and exhibits a wide spectrum of microbial growth inhibition against both Gram-positive and Gram-negative bacteria [9–11]. Furthermore, GFSE possesses strong antiseptic, germicidal, antibacterial, fungicidal and anti-viral properties.

In a study by Shin et al. [12], GFSE was incorporated into red algae film (RA) to be used as a film for the wrapping of cheese and bacon. RA film containing 1% GFSE was determined to be the optimal concentration as the film had the best tensile strength and highest elongation at break values. In another study, based on both mechanical properties and antimicrobial activity, the optimal GFSE concentration recorded for use in barley bran-gelatin (BBG) composite film containing GFSE was also 1% [13]. Above 1% GFSE, tensile strength values were found to decrease. Moreover, the zone of inhibition for *Escherichia coli* O157:H7 extended over a larger area with an increasing concentration of GFSE up to 1%. Though it has been reported that increasing the concentration of GFSE increases the antimicrobial activity of films, Jang et al. reported that the inhibition zone of films with more than 1.5% GFSE decreased [14].

Therefore, to investigate the effects of the incorporation of GFSE on the morphological, optical, mechanical, crystallinity and antifungal properties of chitosan-based composite films, variation in the amounts of GFSE was chosen to be 0.5, 1.0 and 1.5% v/v. In this study, the incorporation of antimicrobials derived from grapefruit seed extract (GFSE), a natural antimicrobial and antifungal agent, was examined in the fabrication of biodegradable chitosan-based composite films.

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2. Materials and methods

2.1. Materials

Powder chitosan of medium molecular weight (MW: 190–310 kDa) with a deacetylation degree of 75–85% was purchased from Sigma–Aldrich. Acetic acid of reagent grade used as a solvent for dissolving chitosan was purchased from Sigma–Aldrich. Grapefruit seed extract (GFSE) as an antimicrobial additive to be incorporated into chitosan-based composite films was purchased from ABC Techno Inc. (Tokyo, Japan).

2.2. Fabrication of films

Chitosan-GFSE film forming solutions were prepared by dissolving chitosan powder (1.5% w/v) in acetic acid solution (2.5% v/v). Varying amounts of GFSE (0, 0.5, 1.0 and 1.5% v/v) was added immediately drop-wise into each film-forming solution. The film forming solutions were stirred at 700 rpm at 24 °C to attain complete dispersion of chitosan and GFSE. The film forming solutions were then filtered to remove any undissolved impurities. A total of four solutions each were prepared with the following compositions: pure chitosan (CG0), chitosan-0.5% v/v GFSE (CG1), chitosan-1.0% v/v GFSE (CG2) and chitosan-1.5% v/v GFSE (CG3).

The prepared clear solution was cast into petri dishes and dried at 50 °C under vacuum in a convection oven. The dried films were peeled off and further conditioned in a desiccator at 50 ± 5% relative humidity and 23 ± 2 °C before they were sealed and stored at 4 °C until further characterization and analysis.

2.3. Field emission scanning electron microscopy

Surface morphology of the fabricated films was examined using field emission scanning electron microscopy (FE-SEM Hitachi S-4300 Scanning Electron Microscope) at an accelerating voltage of 15 kV. Prior to analysis, the films were coated using gold sputtering.

2.4. Film transparency

The transparency of the films was determined by measuring the percentage of transmittance (%T) using a UV–vis spectrophotometer (UV-3101PC, Shimadzu, Japan) according to ASTM standard D1746. Transmittance spectrum ranging from 220 to 800 nm was recorded for each sample. Three replicates of each film composition were measured and the average value was recorded.

2.5. Film thickness

Film thickness was measured using a hand-held digital micrometer (Mitutoyo, Mitutoyo Corporation, Japan) with an accuracy of 0.001 mm. Measurements of film thickness were taken randomly at ten different locations of the film and the mean thickness was calculated.

2.6. Mechanical properties

Tensile properties of the films were measured according to ASTM D882. Film samples 8 × 2.5 cm rectangular strips were prepared and pre-conditioned at 50 ± 10% relative humidity and 23 ± 2 °C for at least 48 h prior to test. Tensile tests were conducted in an Instron universal testing machine (Instron 3345 Tester, Instron, Norwood, MA). Initial grip distance and cross-head speed were set at 5 cm and 5 mm/min, respectively. The tensile strength (TS) and elongation at break (EAB) were determined. TS was calculated by dividing the maximum load of a sample by the initial cross-sectional area and the EAB was expressed as a percentage of the change in the initial gauge length of a sample at the point of failure. Five samples of each film composition were prepared and assessed.

2.7. X-ray diffraction

X-ray diffraction (XRD) patterns of chitosan powder, pure chitosan and chitosan-based composite films were obtained using a LabX XRD-7000 Shimadzu X-ray diffractometer. The diffraction spectra were analysed over a 2θ range from 5 to 40°, using CuKα (30.0 mA, 40.0 kV) at a scan rate of 0.15°/min with a sampling pitch of 0.05°.

2.8. Antifungal activity evaluation

The antifungal activity of the chitosan-based composite films was evaluated by direct contact with bread samples. Bread slices were purchased from a local confectionery store. Uniform bread samples (4 × 4 cm) were singly packed and sealed in direct contact with the pure chitosan or chitosan-based composite films with 0.5, 1.0 and 1.5% v/v GFSE. Samples packed in low-density polyethylene (LDPE) served as the control. All the samples were stored at 24 °C for 30 days. The experiment was repeated twice.

3. Results and discussion

3.1. Surface morphology of films

FE-SEM micrographs of chitosan-based composite films with different GFSE contents and pure chitosan film (CG0) are shown in Fig. 1. The micrographs revealed significant differences in surface morphology at different amounts of GFSE incorporated in the fabricated films. With increasing GFSE contents from 1.0% v/v GFSE (CG2) to 1.5% v/v GFSE (CG3), larger areas of heterogeneity with a more rugous surface was observed (Fig. 1c and d). The significantly larger rugous surface for CG3 as compared to CG2 could possibly be due to the hydrophilicity of GFSE. More moisture might be absorbed when a larger amount of GFSE is incorporated within the film matrix [15]. It was also noted that though both CG2 and CG3 were heterogeneous, the surface morphology of the films was uniform, suggesting that GFSE was well dispersed within all chitosan film matrices (CG1, CG2 and CG3).

3.2. Light transmission and film transparency

The transparency level of pure chitosan film (CG0) and all chitosan-based composite films (CG1, CG2 and CG3) were measured quantitatively, as shown in Table 1. As can be observed, the incorporation of GFSE did not affect the transparency of the films. There were no significant differences in the light transmission values with increment of GFSE amounts added in the chitosan-based composite films, and all chitosan-based composite films showed remarkable transparency at visible range. Therefore, it could be concluded that the transmittance of all chitosan-based films was not affected by the incorporation of GFSE.

Moreover, at 600 nm, which is a wavelength typically used for film transparency [16], the light transmission values of chitosan-based composite films were comparable to commonly used synthetic films such as low-density polyethylene (LDPE) (86.9%), oriented polypropylene (OPP) (89.1%), polyester (83.5%) and polyvinylidene chloride (PVDC) (90.0%) [17]. Hence, the results suggest that chitosan-based composite films have adequate transparency and clarity for use as see-through packaging materials.