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Zhang, C, Yang, Z, Shi, J et al. (7 more authors) (2021) Physical properties and bioactivities of chitosan/gelatin-based films loaded with tannic acid and its application on the preservation of fresh-cut apples. LWT: Food Science and Technology, 144. 111223. ISSN 0023-6438

https://doi.org/10.1016/j.lwt.2021.111223

Physical properties and bioactivities of chitosan/gelatin-based films loaded with tannic acid and its application on the preservation of fresh-cut apples

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Abstract

The aim of this research is to develop composite (CG-TA) films by using chitosan (CS)/gelatin (GL)/tannic acid (TA), for fresh-cut apples preservation. TA was used as the crossing-linking agent to improve the mechanical and barrier properties, and as the active substance to improve the anti-ultraviolet and antioxidant abilities of the chitosan/gelatin (CG) film. With the incorporation of TA from 0 to 1 wt%, the tensile strength of the CG film was increased while the water vapor and oxygen permeability of the film were decreased. The CG-TA films containing more than 0.5 wt% of TA could effectively block most of UV light. The DPPH radical scavenging assay significant increased to 89.3 % when the concentration of TA reached 2 wt%. Finally, CG and CG-TA films, as an active packaging material, were used to preserve the cold-stored fresh-cut apples. Compared with CG film, the CG-TA films obtained better performance on decreasing weight loss, delaying browning degree, inhibiting the lipid oxidase activity and decreasing the malondialdehyde content during 10 d of storage under 4 °C. Therefore, CG-TA films were expected to be a new active packaging material for preserving fresh-cut apples.

Keywords: Chitosan; Gelatin; Tannic acid; Active food packaging; Fresh-cut apples.

1. Introduction

The preservation for fresh-cut food products has become a critical issue, with the expanding market. This is because many fresh-cut food products are perishable. For example, fresh-cut fruit are susceptible to enzymatic browning and flavor loss during storage (Alves et al., 2017; Yousuf et al., 2018). Hence, to reduce their quality loss and therefore extend their shelf life is meaningful.

In recent decades, much attention has been paid to active food packaging with antioxidant and anti-ultraviolet properties, which goes beyond the traditional food packaging mainly with barrier abilities. Active packaging materials could help to maintain the postharvest quality and extend the shelf life of fresh-cut fruit by providing a suitable microenvironment. Gómez-Estaca et al. (2014) reported that antioxidant packages maintained an internal environment suitable for food storage by continuously releasing antioxidants or removing undesirable compounds. Vilela et al. (2018) found ultraviolet packages can protect packaged food by decreasing the photo-oxidation in the packaging. Therefore, active packaging is considered as a potential approach to improve the postharvest quality of the fresh-cut fruit.

For modern food industry, eco-friendly and non-toxic materials are always highly desirable, and active food packaging materials have no exception. Hence, many studies focused on developing active packaging materials by using polysaccharides (Salarbashi et al., 2016) and proteins (Chollakup et al., 2020). Gelatin (GL) is a product formed by partial hydrolysis of collagen. Due to its abundant source, relatively low cost and excellent functional property, GL has become one of the most commonly used biomaterials to prepare biodegradable packaging materials (Gómez-Estaca et al., 2009). However, GL film has poor water barrier and mechanical properties, which are the most detrimental problem in the application process. Chitosan (CS) is cationic polysaccharide isolated from deacetylation of chitin. CS film has been widely studied in food packaging, to their good biocompatibility, mechanical property and antibacterial capacity (Qiao et al., 2020). Usually, the mechanical property of the polymer films could be modified by using two or more polymers (Hedayatnia et al., 2019). For example, CS was usually used to combine with GL to develop the blend films with improved mechanical and barrier properties, because of their electrostatic interactions and intermolecular hydrogen bonding (Gómez-Estaca et al., 2010).

Cross-linking agents can further improve the mechanical and barrier properties of biological films by building a dense cross-linking network in film matrix (Choi et al., 2018). Tannic acid (TA) is an abundant plant-derived polyphenol, which is recognized as a natural substitute for chemical cross-linking agents. The previous studies have reported that GL film could be cross-linked by the TA to improve the mechanical performance of film (Anvari and Chung, 2016). The cross-linking capacity of TA in CS film was also evaluated by Rivero et al. (2010). Besides, the phenolic compound, as an active agent, can also improve the bioactivities of the biological films. The antioxidant, anti-ultraviolet and antitumour capacities of the TA have recently received considerable attention. Zhang et al. (2020) developed a novel cellulose/tannic acid composite films, and the addition of TA endowed the films with strong anti-biofilm activity. Li et al. (2019) found that the antioxidant and UV-absorbing activities of cellulose nanofibrils were increased with the incorporation of TA. Therefore, TA as an active substance can incorporate with film matrix to develop active packaging. However, to our best knowledge, there is no relevant report that using CS /GL-based film loaded with TA as an active packaging material to preserve the fresh-cut fruit.

Hence, the aim of this work was to develop the composite CG-TA films with improved physical properties and bioactivities and then to investigate its ability to improve the postharvest quality of a typical fresh-cut fruit (apple). The intermolecular interactions, microstructure, optical, mechanical and barrier properties of the films were characterized. Moreover, the functional properties including anti-ultraviolet and antioxidant capacities were investigated. Finally, the composite films were used to preserve the fresh-cut apples, and the relevant quality indexes including weight loss, browning index, lipoxygenase (LOX) activity and malondialdehyde (MDA) content, were determined.

2. Materials and methods

2.1. Materials and reagents

TA was purchased from Sangon Biotechnology Co., Ltd. (Shanghai, China). Other materials and reagents including CS powder (Mw: 117.000 g mol^{-1}, deacetylation degree: 80 %), gelatin (bloom 260, molecular weight of 5.2×10^4 Da, type B), 2,2-diphenyl-1-picrylhydrazyl (DPPH), trichloroacetic acid ($C_2HCl_3O_2$) and acetic acid (CH_3COOH) were all purchased from Sinopharm Chemical Reagent Co., Ltd (Shanghai, China). All the reagents used were in analytical grade. “Fuji” apples of uniform size, color, maturity and no physical damages were purchased from the local market (Zhenjiang, China).

2.2. Preparation of CG-TA composite films

In this study, a two-step method of mixing and drying was used to prepare the films according to the method reported by Zhang et al. (2019) with slight modifications. Briefly, 2 % (*w/v*) of CS solution was prepared by dissolving CS power into distilled water containing 2 % (*v/v*) acetic acid with stirring for 3 h at 60 °C. 2 % (*w/v*) of GL solution was prepared by dissolving GL into distilled water with stirring for 2 h at 50 °C. Afterward, the blend solution was prepared by mixing the CS and GL solution at a ratio of 1:1 (*v/v*) and continuous stirred at 50 °C. The TA with different quality (0, 0.5, 1 and 2 wt% based on the total weight of the chitosan and gelatin) was added into the blend solution to prepare the final composite solution. After thoroughly stirring and degassing, the composite solution was distributed into a plastic petri dish and dried in an incubator at 25 °C with 75 % relative humidity (RH). The composite film containing 0, 0.5, 1 and 2 wt% of TA was named as CG, CG-TA I, CG-TA II and CG-TA III film, respectively. Before testing, all films were conditioned in a desiccator with 75 % RH at 25 °C for 48 h.

2.3. Characterization of the composite films

2.3.1. Fourier transform infrared (FT-IR) spectroscopy

FT-IR spectrum of the film was measured using a Nicoletis50 infrared spectrometer (Perkine Elmer 16 PC spectrometer, Boston, USA), equipped with an attenuated total reflection (ATR) accessory. The spectrum was analyzed at a resolution of 2 cm^{-1} in the wave number range of 1000–4000 cm^{-1}.

2.3.2. Scanning electron microscopy (SEM)

The cross-section of the film was characterized by a scanning electron microscopy (S-4800, Hitachi High-Technologies Corporation, Japan), and its accelerating voltage was 5 kV. All film samples were pasted on the specimen holder with double-side adhesive tape and then sprayed with gold in a vacuum evaporator.

2.3.3. Color measurement

The color of the film was analyzed using CR-400 Minolta colorimeter (Minolta Camera, Co., Ltd., Osaka, Japan) with a white plate (L^* (99.46), a^* (-0.08) and b^* (-0.10)) as the standard background. The color parameters (L^*, a^* and b^*) were determined by calculating the average value of five random points on the film sample.

2.3.4. Mechanical property

According to ASTM D882-00 standard method, tensile strength (TS) and elongation at break (EB) were measured using an Instron Universal Testing Machine (Model 4500, Instron Corporation, Canton, MA, USA). Before test, all films were cut into strips of 60 mm length and 20 mm width. Film sample was fixed between tensile grip and the initial grip. The initial grip separation and cross-head speed was set at 40 mm and 0.06 mm s^{-1}, respectively. Each film sample was analyzed by ten replicates, and final measurement was represented by the average value.

2.3.5. Water vapor permeability (WVP)

The WVP of the film was determined according to the ASTM E96-95 standard method. The opening of centrifuge tube (50 mL) containing 20 mL distilled water was tightly sealed by film samples. The centrifuge tube was placed in a desiccator containing entirely dried silica gel. The weight of the tube was measured every 2 h for 48 h. The WVP of each film sample was calculated by the following equation:

$$\mathrm{WVP} = \frac{M \times d}{S \times t \times \Delta P} \qquad (1)$$

where M was the changed weight of centrifuge tube (g), d was the average film thickness (mm), S was the transfer area (m^2), t was time (s), and ΔP was the partial water vapor pressure difference between the distilled water and dry atmosphere.

2.3.6. Oxygen permeability (OP)

The oxygen permeability (OP) of film was determined using a Model GTR-7001 automated oxygen permeability testing machine (Systester Instruments Co., Ltd., Jinan, China) following the standard method (ASTM D3985-05, 2005). One side of the film sample was oxygen and the other side was nitrogen. Oxygen transmission rate (OTR) was measured and OP was calculated according to the equation:

$$\mathrm{OP} = \mathrm{OTR} \times \frac{d}{\Delta P^*} \qquad (2)$$

Where, d was film thickness (mm), and ΔP^* was the partial pressure of oxygen.

2.3.7. UV-vis barrier property

UV-vis spectroscopy of the film was measured using a UV-vis spectrophotometer (Agilent CARY 100, Varian Corporation, USA) at selected wavelengths (200–800 nm). Before test, film samples were cut into rectangular strip (4 cm × 1.5 cm) and fixed in the quartz spectrophotometer cells. The spectroscopy of each film sample was done in triplicate.

2.3.8. Antioxidant property

The antioxidant property was determined by evaluating the DPPH radical scavenging activity according to the method of Lee et al. (2019) with some modification. Sample strip (20 mg) was placed in DPPH solution in methanol (4 mL, 0.25 μM) and the mixture system reacted in the dark at 20 °C for 1 h. The supernatant was extracted and then its absorbance was measured at 517 nm. DPPH radical scavenging activity was determined based on the following equation:

$$\text{DPPH radical scavenging rate (\%)} = \frac{A_x - A_y}{A_x} \times 100 \qquad (3)$$

Where A_x was the absorbance of the blank solution and A_y was the absorbance of solution after reaction.

2.4. Application for preserving the fresh-cut apples

Apples were pre-washed by chlorinated water (500 ppm) for 3 minutes and then rinsed thoroughly by distilled water, removing impurities and killing surface microorganisms. The apple peel was removed and the apple without peel was cut into slices of the same size and shape (4 × 2 × 1 cm) with a sterile sharp knife. The fresh-cut apples were randomly placed in a polyethylene terephthalate packages box, and then sealed by CG, CG-TA I, CG-TA II and CG-TA III films, respectively (without film packaged as control). All groups were immediately stored in a vertical display freezer at 4 °C and 75 % RH and monitored at 0, 2, 4, 6, 8, 10 d. The experiment was repeated in triplicates.

2.4.1. Weight loss

The weight loss of fresh-cut apples was represented by the value of the difference between the weight before and after storage. The weight of fresh-cut apples was measured by an electronic balance at 0, 2, 4, 6, 8, 10 d. The result of weight loss was expressed as the percentage of mass loss during storage to the initial weight.

$$\text{Weight loss (\%)} = \frac{W_0 - W_z}{W_0} \times 100$$

(4)

Where W_0 was the initial weight of fresh-cut apples and W_Z was the weight of fresh-cut apples during storage period (z = 2, 4, 6, 8, and 10 d).

2.4.2. Browning index (BI)

The degree of browning of fresh-cut apples was quantized by BI during the storage period. The color parameters (L^*, a^* and b^*) of each sample were determined by CR-400 Minolta colorimeter. The BI was calculated by the formula as follows (Zambrano-Zaragoza et al., 2014):

$$\text{BI (\%)} = \frac{x - 0.31}{0.172} \times 100 \qquad (5)$$

$$\text{Where, } x = \frac{a^* + 1.75L^*}{5.645L^* + a^* - 3.012b^*} \times 100$$

2.4.3. Determination of LOX activity and MDA content

LOX activity was measured according to the method described by Zhang et al. (2003) with slight modification. Sample tissues (2.0 g) were homogenized in 10 mL phosphate buffer (0.05 mol L^{-1}, pH 6.8) and then centrifuged (15000 × g, 15 min at 4 °C). The substrate solution was made by 2.750 mL of 50 mM potassium phosphate buffer (pH 6.8) and 50 μL of sodium linoleate. 0.2 mL of supernatant of sample tissues was added into the substrate solution to react at room temperature. The change of absorption of reaction system was measured at 234 nm and recorded every 30 s for 3 minutes. The amount of the enzyme that caused a change of 0.01 in absorbance per minute was defined as one unit of LOX activity.

MDA content of fresh-cut apples was determined according to the method of Zhong et al. (2018). Sample (2.0 g) was ground with 10 mL of 10 % (*w/v*) trichloroacetic acid (TCA) and then centrifuged (4000 × g, 10 min at 4 °C). The 2 mL of supernatant was extracted and then mixed with 2 mL of 10 %

(*w/v*) TCA containing 0.6 % (*w/v*) thiobarbituric acid (TBA). The mixture was placed in boiling water bath to react for 15 min. Afterwards, the mixture was cooled rapidly with cold water and centrifuged at 4000 × g for 10 min. The supernatant was extracted and its absorbance was measured at 450 nm, 532 nm and 600 nm.

2.5. Statistical analyses

Data of this study were analyzed with SPSS 13.0 statistic program (SPSS Inc., IL, USA). The statistical significance between means was evaluated using an analysis of variance (one-way ANOVA). Differences were considered as statistically significant when $p < 0.05$.

3. Results and discussion

3.1. FT-IR spectra of composite films

The functional groups and inter-molecular interactions in the films can be determined by FT-IR spectral analysis (Fig. 1). In the spectrum of TA, the wider bands at 3600–3000 cm^{-1} were due to −OH stretching. The peak at 1730 cm^{-1} correspond to the presence of carboxyl carbonyl group, while the bands at 1612, 1521 and 1447 cm^{-1} were attributed to aromatic C═C bonds (Erdem et al., 2013). The bands of GL and CS at 3297/3256 cm^{-1} was attributed to N − H and O − H stretching (amides A), at 2935/2925 cm^{-1} to C–H stretching (amides B). Furthermore, the spectra of GL and CS showed characteristic peaks at 1636/1650 cm^{-1} and 1529/1552 cm^{-1}, which assigned to amide I (C═O stretching) and amide II (N − H bending and C − N stretching), respectively. In addition, the typical band at 1235 cm^{-1} of GL represented C–N and N–H stretching (Haghighi et al., 2019; Liu et al., 2020). When CS and GL were blended, a broadening and a slight shift (from 3297 to 3285 cm^{-1}) of the amide-A band of GL were described as the interaction of the carboxyl groups from GL and the amino groups from CS (Staroszczyk et al., 2014). In addition, the regions of amide I, amide II and amide III bands of CG films occurred obvious changes compared to pure films. Previous studies have confirmed that the phenomenon was caused by intermolecular hydrogen bonds and electrostatic interactions between CS and GL (Qiao et al., 2017).

With the addition of TA, the FT-IR spectrum of CG film showed some changes in the intensity and the position of bands. The amide-A band was slightly broadened and shifted from 3285 to 3281/3280 cm^{-1}, indicating the formation of hydrogen bonding between the hydroxyl groups of polyphenols in the TA and the amino/hydroxyl groups in CS/GL (Yao et al., 2017). After TA addition, the amide II (1546 cm^{-1}) and amide III (1252 cm^{-1}) of CG film shifted to 1536/1535 cm^{-1} and 1244/1242 cm^{-1}, respectively. The shifts in the position of amides groups confirmed the interactions existed between functional groups of biopolymers with functional groups of TA. Benbettaïeb et al. (2015) also found a similar change when natural antioxidants (ferulic acid, quercetin and tyrosol) incorporated chitosan-gelatin blend films. They suggested that the shifting of amides groups might be due to the presence of interaction between the amide groups of chitosan/gelatin and hydroxyl groups of antioxidants. When the concentration of TA above 0.5 wt%, a new peak was observed at 1450/1449 cm^{-1} in CG-TA films. It may be attributed to the C=C ring stretching absorptions of TA in the composite films (Hong, 2016). The results indicated that TA has successfully cross-linked to the CG film. In addition, the intensity of peaks decreased slightly with the further increased of TA concentration, indicating cross-linking strength increased.

Fig. 1 goes here.

3.2. Microstructure of composite films

The cross-section of CG and CG-TA films was observed in Fig. 2. The cross-section of CG film (Fig. 2A) exhibited a uniform, smooth and without any pores structure, indicating that the presence of excellent compatibility between CS and GL. This was accord with the fact that the polyanion-cation complexes formed between two biopolymers via associative interactions (Mohammadi et al., 2018). Regarding to the CG-TA films, the CG-TA I film had a homogeneous cross-section (Fig. 2B), and no obvious difference was observed between CG film and CG-TA I film. This may be due to that low concentration of TA was not enough to change the microstructure of film. The cross-section of CG-TA II film became dense and tight (Fig. 2C) when the concentration of TA increased to 1 wt%, which played an important role in improving the mechanical and barrier properties of film. It might be due to cross-linking network established within polymer matrix of films by the interactions between biopolymers and TA. Similar result was also reported by Cui et al. (2014), who developed a interpenetrating polymer networks hydrogels composed of CS/GL cross-linked by genipin. However, the cross-section of CG-TA III film (Fig. 2D) became rough and porous when the concentration of TA increased to 2 wt%. This result was similar to those reported by Mathew and Abraham (2008), who found that high concentration of ferulic acid could cause phase contrast and slight phase separation in film matrix.

Fig. 2 goes here.

3.3. Color of composite films

The color of the film is essential for the practical application in the food packaging. The color parameters (*L**, *a** and *b**) of all films were shown in Table 1. With the TA concentration increased, the *L** value of films decreased from 90.28 to 80.48. Meanwhile, the *a** and *b** values of films increased from -1.36 to 4.76, and from 9.12 to 17.70, respectively. These results indicated that the CG-TA films became darker and the color tended to be redder and yellower than CG film. The color differences between CG and CG-TA films could also be directly observed in Fig. 3. With the concentration of TA increased to 2 wt%, CG-TA III film showed significant brightness reduction and color changes compared to other films. The results could be due to that the special chromophores existed in the TA had an absorption effect on visible light. Hager et al. (2012) also found that wheat gluten films incorporated with TA showed reddish brown appearance.

Table 1 goes here.

Fig. 3 goes here.

3.4. Mechanical and barrier properties of composite films

The thickness of the all films was presented in Table 2. CG film had the lowest value of thickness (0.030 mm). The film thickness gradually increased with the concentration of TA increased and reached maximum value in CG-TA III film (0.041mm). The enhancement of film thickness could be due to the TA increased the curing material content of film (Ramziia et al., 2018).

The mechanical properties (TS and EB) of all films were presented in Table 2. The TS of CG film was significantly increased ($p < 0.05$) after incorporation of the TA. The TS value of the CG-TA films further increased with the increasing concentration of TA. When the dosage of TA was 1 wt%, the TS reached the highest value (48.52 MPa). The higher TS of CG-TA films could be due to that cross-linking network formed in films increased the strength and malleability of the films. Peña et al. (2010) also found that the TS value of GL film was significant inflated ($p < 0.05$) with the incorporation of TA. However, the TS value decreased slightly with the further increased of TA concentration. This result was similar to those found by Evranos et al. (2019), who concluded that excess bone ash incorporated into chitosan/gelatin blend films caused the phase separation of film matrix, thereby decreased the strength of films. In contrast, the EB of CG-TA films was decreased significantly ($p < 0.05$) with incorporation of the TA, especially the EB value decreased to 2.31 % when TA concentration increased to 2 wt%. This result may be due to that the hydrogen bonds formed between TA and film matrix decreased the chain mobility in CG-TA films, resulting in the decrease of flexibility (Wu et al., 2016).

The WVP of the film was presented in Fig. 4A. The WVP of the CG film was further decreased ($p < 0.05$) with the increasing concentration of TA. The films showed the lowest WVP when TA concentration reach 1 wt%, which decreased by 45 % compared with CG film. This result was mainly due to that the cross-linking network formed in CG-TA II films reduced the binding sites for water molecules, compressed the interspace for water vapor diffusion and limited the escape of water vapor. As Aljawish et al. (2016) discussed, the decreased WVP value could be attribute to the dense cross-linking network formed by ferulic acid and ethyl ferulate incorporated CS film. The OP of CG film showed a similar change trend with WVP (Fig. 4B), and reached the lowest value (4.18×10^{-6} cc m^{-1} d^{-1} atm^{-1}) when the dosage of the TA was 1 wt%. This result might be attributed to the fact that TA molecular distributed on the surface of composite films occupied the perpendicularly diffusive pathway of oxygen, showing that penetrating molecules was traveling in a more tortuous diffusive pathway, which resulted in decreasing oxygen permeability. Halim et al. (2018) also found that OP value of the films was significantly decreased ($p < 0.05$) when the biopolymer matrix incorporated with TA. However, CG-TA III film obtained a higher WVP and OP value than CG-TA II film. This result might be related to the existence of pores in CG-TA III film (Fig. 2D), allowing more water vapor and oxygen to diffuse. Therefore, the above results indicated that the barrier properties of CG film were improved by the addition of TA, and the CG-TA II film showed the optimal property.

Table 2 goes here.

Fig. 4 goes here.

3.5. Antioxidant activity of composite films

The antioxidant capacity of film was evaluated by measuring the DPPH radical scavenging activity, and the results were shown in Fig. 4C. The DPPH scavenging ability of the CG film (14.6 %) was significantly improved ($p < 0.05$) when TA was added into CG film. The scavenging ability further increased with the increasing concentration of TA. The scavenging ability of the CG-TA III film reached the highest value (89.3 %), with around 6.11- fold compared to CG film. This result indicated that the composite films obtained excellent antioxidant activity due to the addition of TA. This was because polyphenols could trap free radicals and block the free radical chain reaction (Leopoldini et al., 2011). Rui et al. (2017) also reported that the antioxidant capacity of films was improved when gallic acid incorporated the chitosan-gelatin blend films.

3.6. UV-vis barrier property of composite films

The UV-vis barrier property plays an important role in retarding lipid oxidation and maintaining the sensory property of packaged food. The transmittance of CG and CG-TA films in the wavelength range of 200–800 nm was presented in Fig. 4D. The results showed that CG film had a certain barrier capacity for UV light, which mainly due to the presence of aromatic amino acids residues of gelatin (Bonilla and Sobral, 2016). The transmittance rate of CG film was kept at an relatively high level (> 75 %) when the wavelength exceeded 400 nm, indicating it was very transparent. Bi et al. (2020) also obtained a similar result. CG-TA films showed better UV barrier property than CG film. With the increasing concentration of TA, the barrier property for UV light was stronger than before. When the dosage of TA exceeded 0.5 wt%, almost all of UV-C and UV-B and most of UV-A could be blocked effectively. This could be due the fact that the specific phenolic hydroxyl groups and chromophores in TA could absorb a certain degree of UV light. Notably, the presence of chromophores in TA also influenced the transmittance of CG-TA films for visible light. Zhai et al. (2017) also found a similar phenomenon when roselle anthocyanins were added into biopolymer films. In summary, TA could be used as a UV blocking agent to enhance the UV-blocking property of the CG film.

3.7 Application for preserving the fresh-cut apples

3.7.1. Weight loss

As shown in Fig. 5A, the quality of fresh-cut apples of all groups showed a downward trend during storage period. This result may due to the fact that the stomata transpiration and dry matter consumption caused by respiration. The weight loss rate of unpackaged fruits reached 29.6 % at the end of storage period, which was much higher ($p < 0.05$) than that of packaged groups (11.6 % to 15.2 %). A similar result was also reported by Lan et al. (2020), who found that unpackaged mangoes had the highest weight loss rate after 10 d storage. Among packaged groups, fresh-cut apples packaged by CG-TA films obtained lower weight loss rate than CG film, especially CG-TA II film (11.6 %). This may be related to the fact that the formation of cross-linking network improved water vapor barrier property (Fig. 4A) of CG-TA films, thereby greatly inhibiting water vapor to escape from the packaging box.

3.7.2. Browning index (BI)

As we all know, browning index is an essential quality parameter for fresh-cut fruits. The BI of the fresh-cut apples packaged with different films was presented in Fig. 5B. The BI of all fruits increased rapidly in the initial 2 d of storage and rose smoothly thereafter. This was because reactive oxygen species (ROS) triggered by physical stress damaged the original compartmentalization functions of cell membrane, allowing polyphenol substrates to contact with the polyphenol oxidase and phenol peroxidases, causing rapid browning of fresh-cut apples (Gao et al., 2018). After 10 d storage, the fresh-cut apples packaged with CG film obtained significantly ($p < 0.05$) higher browning index than CG-TA II and CG-TA III films (62.8, 56.8 and 56 for CG, CG-TA II and CG-TA III, respectively). The browning of the fruits packaged with CG-TA films was delayed effectively. This result might be due to follow several reasons: first, the packaging box sealed by CG-TA films maintained a relatively low oxygen content due to CG-TA films have better oxygen barrier property (Fig. 4B) than CG film, which reduced the chance of oxygen participating in the enzymatic reaction. Second, strong UV-barrier (Fig. 4D) of composite films protected cells from the harm of lipids peroxidation, maintaining the integrity of cell membrane, restraining the enzymatic browning of fresh-cut apples. Similar result was also reported by Toivonen and Brummell (2008), who found that the integrity of membrane could affect the browning rate of fresh-cut fruits and vegetables. The above results indicated that CG-TA composite films could improve the browning of fresh-cut apples.

3.7.3. LOX activity and MDA content

As shown in Fig. 5C, the LOX activity of unpackaged and packaged fruits all showed a wave-like change during the storage period. After 10 d storage, the LOX activity of fresh-cut apples packaged with CG-TA II and CG-TA III films was 114 and 118 × 10^{-3} U kg^{-1}, respectively, which was obvious ($p < 0.05$) lower than other treatment groups. Correspondingly, the MDA content (Fig. 5D) of the above two packaged groups also showed relatively low values (3.56 and 3.67 μmol kg^{-1}, respectively). This was because MDA accumulation was related to non-enzymatic processes, especially LOX (Lo’ay and Dawood, 2017). The decreased in LOX activity and MDA content may be attributed to the fact that the oxygen barrier property (Fig. 4B) and UV blocking property (Fig. 4D) of CG films were improved by the addition of TA, thereby reducing the damage of oxygen and ultraviolet rays to fresh-cut apples. Due to MDA was an indicator of membrane disruption, the results could indicate that CG-TA films were beneficial to alleviate cell oxidative damages. Previous research also found that MDA content indirectly reflected the existence of correlation between the integrity of the cell membrane and browning degree (Dokhanieh and Aghdam, 2016). The finding was consistent with the results of BI (Fig. 5B) of fruit.

Fig. 5 goes here.

3.7.4. Physical appearance

The effect of packaging on appearance quality of fresh-cut apples during storage was shown in Fig. 6. The surface of unpackaged fruit appeared wrinkled skin at the end of storage period, while the packaged fruit was relatively full. The results might be attributed to the fact that the packaging films inhibited the water loss of fruit during storage. Meindrawan et al. (2018) also found that there was a linear correlation between weight loss and visual quality of fruit. In addition, the color gradually changed during storage in both unpackaged and packaged fruit. Notably, the color of fresh-cut apples packaged by CG-TA films changed more slowly than CG film. Among them, CG-TA II and III films packaged fruit showed better color appearance. The BI value (Fig. 5B) of fresh-cut apple also supported the results. Therefore, the browning of fruit could be inhibited effectively by the CG films incorporated with TA, thereby improving the appearance quality of fruit.

Fig. 6 goes here.

4. Conclusion

In our research, the composite films were developed successfully by incorporating TA into chitosan-gelatin film matrix. The physical properties and bioactivities of the composite films were improved due to the addition of TA. The dense cross-linking microstructures have been established in the films by the interaction between TA and chitosan/gelatin groups. As a result, the mechanical and barrier properties of the films were strengthened. The addition of TA improved significantly the anti-ultraviolet and antioxidant capacities of CG film. In general, the CG film modified by 1 wt% of TA exhibited better mechanical and barrier properties, but the CG film modified by 2 wt% of TA showed better anti-ultraviolet and antioxidant abilities. Moreover, the fresh-cut apples packaged with CG-TA films had better freshness degree and postharvest quality as compared with that without packaging or packaged with CG film. There was no significant difference in preservation effect between the CG film containing 1 wt% and 2 wt% of TA. Considering to the results of comprehensive characterization and application experiments, the CG film incorporating 1 wt% of TA was more suitable as an active packaging material to improve the postharvest quality of fresh-cut apples.

Acknowledgments

This work was supported by the National Key Research and Development Program of China [grant numbers 2017YFC1600805]; the National Natural Science Foundation of China [grant numbers 31772073, 31671844]; the Natural Science Foundation of Jiangsu Province [grant numbers BE2019359]; and Priority Academic Program Development of Jiangsu Higher Education Institutions (PAPD).

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