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Characterization of a sodium alginate–carboxymethylcellulose film incorporated with cinnamaldehyde, and its application for preservation of almond paste

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RESEARCH ARTICLE

Abstract

Almond paste is a valuable traditional product that quickly deteriorates due to oxidative rancidity and microbiological spoilage. This study involved the enhancement of sodium alginate (SA)- and carboxymethylcellulose (CMC)-based edible films with varying concentrations of cinnamaldehyde (CN: 0.1%, 0.2%, and 0.3% w/v) to prolong the shelf life of almond paste. With the increase in CN concentration, the films showed higher opacity (1.64 ± 0.05 A/mm–29.58 ± 0.19 A/mm) and allowed a higher permeability to water vapor (0.18 ± 0.01 g·mm/(m^2·h·kPa)–0.44 ± 0.03 g·mm/(m^2·h·kPa)). Furthermore, CN enhanced the antibacterial efficacy of the films against food pathogens. On the other hand, sensory evaluations of almond paste samples showed that the coating with films containing 0.2% CN was preferable to the other coatings. The coated samples indicated an inhibition in total mesophilic aerobic count from 3.39 log colony-forming unit (CFU)/g to 2.87 log CFU/g and a decrease in peroxide value from 6.46 milli-equivalent (meq) O_2/kg to 5.76 meq O_2/kg after 15 days of storage at 25°C. The results suggest that SA–CMC film enriched with CN may be useful in preserving foods with high lipid content.

Keywords: cinnamaldehyde; sodium alginate; carboxymethylcellulose; coating; almond paste

Introduction

Cultivated almonds, designated as *Amygdalus communis*, are now classified in the *Prunus* genus of the *Rosaceae* family (Rahemi and Gradziel, 2024). Almond is an important plant grown in various regions of Türkiye (Çapanoğlu and Boyacıoğlu, 2008). Almond contains three different parts: kernel (nut), hull, and shell (Company *et al.*, 2017). Almond is advantageous to health because of its nutritious constituents. The composition primarily consists of lipids (44.7–54.1%) and is abundant in unsaturated fatty acids. In addition, it contains high amounts of protein (18.5–24.0%) and dietary fiber (7.9–16.0%). The vitamin and mineral composition mostly includes α-tocopherol and potassium, phosphorus, magnesium, and calcium (Yada *et al.*, 2013). Almonds, which are consumed in various ways, such as raw and roasted, are also processed in different forms, such as spread and paste (Dhankhar *et al.*, 2022). Almond paste, also known as "marzipan," is a mixture prepared

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with ground almonds and glucose syrup. The proportion of these two ingredients may change, and coloring agents, flavoring agents, etc. are added to the formulation according to the final product (Baiano and Del Nobile, 2005; Faid *et al.*, 1995). Traditional Turkish almond paste contains only almonds, powdered sugar, and water (Ciftci and Ozilgen, 2019). Almond paste has a short shelf life if stored at room temperature. Microbiological spoilage and oxidative rancidity are important spoilage factors of almond paste during storage (Baiano and Del Nobile, 2005).

In the literature, various studies have been conducted for increasing the shelf life of almond paste. For this purpose, studies are conducted on the addition of some ingredients, such as stabilizers, antioxidants, and maltose (Çapanoğlu and Boyacıoğlu, 2008) as well as utilization of black carrot juice as a natural antioxidant (Ciftci and Ozilgen, 2019). Additionally, research has been conducted on packaging methods to enhance the shelf life of almond paste, including the use of flexible films (such as nylon or ethylene–vinyl alcohol copolymer layers) combined with nitrogen or oxygen scavengers (Baiano and Del Nobile, 2005), polyvinyl chloride film, aluminum foil, and polyamide–polyethylene film materials (Romeo *et al.*, 2010) as well as storage in glass jars and polyethylene bags (Takma *et al.*, 2018). However, to the best of our knowledge, no attempt has been made to coat almond paste with an edible active film formulation.

Carboxymethylcellulose (CMC) is a polymer formed by an alkaline-catalyzed chemical reaction between cellulose and chloroacetic acid. CMC is used as a stabilizer and thickener in food products because of its excellent water-binding ability and hydrophilic properties (Abdin *et al.*, 2023). CMC is also used in edible film formulations because of its biodegradable, tasteless, odorless, and nontoxic quality and gelling properties (Akhtar *et al.*, 2018); however, when used independently, it produces films with low mechanical strength, poor water vapor barrier, and opacity (Akhtar *et al.*, 2024). Sodium alginate (SA) is a polymer derived from marine algae, and is composed of two hexuronic acid residues connected by 1,4-glycosidic bonds. SA is water soluble, nontoxic, exhibits high viscosity in water, and possesses acid resistance. SA shows excellent film-forming capabilities (Eltabakh *et al.*, 2021), but if used alone, it has poor mechanical properties, a strong hydrophilic character, and low thermal stability (Akhtar *et al.*, 2024). In recent years, the production of SA and CMC composite films has been studied by many researchers (Akhtar *et al.*, 2024; Sun *et al.*, 2023; Yang *et al.*, 2022).

The combination of CMC and SA creates strong interactions through hydrogen bonds. When these materials are blended, the physical, thermal, and mechanical properties of the films are improved due to intermolecular interactions (Das *et al.*, 2023). However, the lack of active ingredients in these films, such as antimicrobial and antioxidant agents, limits their use in food products (Thivya *et al.*, 2021). Cinnamon spice is made from the inner bark of trees belonging to the *Cinnamomum genus*. Cinnamon species contain cinnamaldehyde (CN) in varying amounts of up to 90.5% (Shreaz *et al.*, 2016). CN is the main constituent of cinnamon essential oil (Yan *et al.*, 2024). CN is a yellow and oily liquid with a strong cinnamon odor and taste (Haddi *et al.*, 2017). CN inhibits several microorganisms, such as bacteria, mold, and yeast, and the toxins produced by these microorganisms (Doyle and Stephens, 2019).

Cinnamaldehyde has antifungal and antibacterial properties against pathogenic bacteria, such as *E. coli* (He *et al.*, 2019; Wang *et al.*, 2005). In addition, CN compounds have a reducing effect on lipid peroxidation (Keshvari *et al.*, 2013). Moreover, CN is classified as “Generally Recognized as Safe” (GRAS) by the United States Food and Drug Administration (FDA). This identification is also held by the Flavor and Extract Manufacturer’s Association (FEMA). It is also designated as “Status A” (may be used in foodstuffs) product by the European Council (Friedman, 2017). CN is used in fruit juices, fruits and vegetables, meat and chicken products, and seafood, or in the processes applied to these products, especially because of its inhibitory effect on food pathogens. CN also is added to animal feeds to investigate antimicrobial effects (Friedman, 2017). In addition, many researchers added CN to food packaging to develop antimicrobial packaging (Gao *et al.*, 2024; Ghiasi and Golmakani, 2023; Zhao *et al.*, 2021) and preserve foods, such as cheese (Gao *et al.*, 2024), strawberries (Zhou *et al.*, 2024), and beef (Qiu *et al.*, 2025). In this study, CN was added to the film produced to cover almond paste. Besides its antimicrobial properties, the aroma it would give to the coating material was thought to be pleasant to the consumer when used with the almond paste product. Therefore, this study aimed to develop an active composite film that would increase the shelf life of almond paste by protecting it against both microbial and oxidative spoilage. For this purpose, SA and CMC were used as film constituents and CN was used as an active ingredient.

Materials and Methods

Materials

The films were obtained from food-grade and powder form of SA and CMC ($C_8H_{16}O_8$; molecular weight: 240.21 g/mol) (Yasin Teknik, Istanbul, Turkiye). Cinnamaldehyde (98%) was purchased from Carl Roth GmbH (Karlsruhe, Germany). The chemicals used in films and analysis were of analytical grade and included: glycerol (84–88%) Tween 80, nutrient agar, plate count agar (PCA), potato dextrose agar (PDA), peptone water, ethanol, diethyl ether, sodium hydroxide, acetic acid, chloroform, starch, potassium iodide, and sodium thiosulfate. Traditional almond paste containing granulated sugar, almonds, and drinking water was purchased from a local market (Istanbul, Turkiye) on the day it was produced.

Preparation of composite films

Sodium alginate (0.75 g) and CMC (0.25 g) were added to distilled water (100 mL) and heated in a water bath (60°C) with continuous stirring with a mechanical stirrer at 400 rpm (RW20, IKA, Germany) for 30 min (Han *et al.*, 2018). As a plasticizer, glycerol (50% of the total weight of SA+CMC) was mixed, and the solution was degassed in an ultrasonic bath (PMUY4LD, Protech, Turkiye). Solution (40 mL) was poured into polystyrene Petri dishes (14-cm diameter) and dried at 50°C (ED53, Binder, Germany) for 24 h. The films were conditioned in a desiccator containing saturated solutions of $Mg(NO_3)_2$ at 25°C and 55±2% relative humidity (RH).

In the films containing CN, following the heat treatment, the film-forming solution was allowed to cool to 25°C; then, 0.1 g, 0.2 g, and 0.3 g of CN, and Tween 80 (CN/Tween 80: 1/1 w/w) were incorporated and homogenized by mixing at 10,000 rpm (Yellowline D125, IKA, Germany) for 5 min. CN content was determined through preliminary tests based on the films' aroma. The same protocol was followed afterwards (Figure 1). The film containing SA–CMC and glycerol was called control (C) and the films containing 0.1%, 0.2%, and 0.3% (w/v) CN were called 0.1 CN, 0.2 CN, and 0.3 CN, respectively.

Characterization of films

Thickness and density

The thickness of the films was measured from random regions of the film by a digital micrometer (Insize, 3109-25A, Germany) with 0.001-mm precision. To measure the density, the films were cut into squares of 39 × 39 mm and their mass, area, and thickness were measured. The density results were expressed as g/cm^3.

Opacity

The opacity of the films was measured by a spectrophotometer (T60UV, PG Instruments, UK). The films were cut into 3 × 0.4 cm and attached to the surface of the spectrophotometer cuvette. Opacity was calculated by dividing the absorbance at 600-nm wavelength by film thickness (Park and Zhao, 2004).

Water vapor permeability (WVP)

The WVP of the films was determined according to the standard water method E96/E96M (American Standard Testing Methods [ASTM], 2015) with some modifications defined by McHugh *et al.* (1993). The aluminum test cups (opening: 29.7 cm^2) were filled with 20 mL of distilled water and covered with circular films. The cups were stored in a desiccator with saturated $Mg(NO_3)_2$ solution at 25°C and 55±2% RH. Weight loss of the cups was noted, and WVP was calculated as described by Yıldırım-Yalçın *et al.* (2019). The results were expressed as g mm/m^2 h kPa.

Mechanical properties

The percentage of elongation at break (EAB, %) and tensile strength (TS, N/mm^2) of the films were determined according to the ASTM (2012) standard method D882. The films were cut into strips of 15 × 70 mm and placed at an initial length of 40 mm in a texture analyzer (LAB3-2512 A, Mesdan LAB, Italy). The load cell of the device was 100 N and the cross-head speed was 25 mm/min.

Fourier transform infrared (FT-IR) spectroscopy

Chemical interactions between film components were determined using a Bruker Invenio S Spectrometer (Bruker Co., Ettlingen, Germany). The transmittance spectral regions were obtained between 4000-400 cm^{-1} with 4 cm^{-1} resolution.

Antimicrobial activity

The antimicrobial activity of films was determined using the well diffusion method. Film solution (100 μL, before drying step of film preparation) was poured into wells (6-mm depth) on nutrient agar plates previously inoculated with 0.1 mL culture of *Bacillus cereus*, *Escherichia coli*, *Listeria monocytogenes*, *Salmonella typhi*, and *Staphylococcus aureus*. The plates were incubated at 37°C for 24 h to analyze inhibition zone (mm).

Preparation of wrapped almond paste

All surfaces of almond paste samples (approximately 10 g), except the control sample (C), were wrapped by one layer of the prepared film.

Analysis of wrapped almond paste

Sensory analysis

The acceptability values of coated almond paste samples were determined on the day of wrapping. The Social and Human Sciences Ethical Committee of Istanbul Aydin University approved methods for the sensory testing in this study (No.: 2024/11). Untrained 50 panelists,

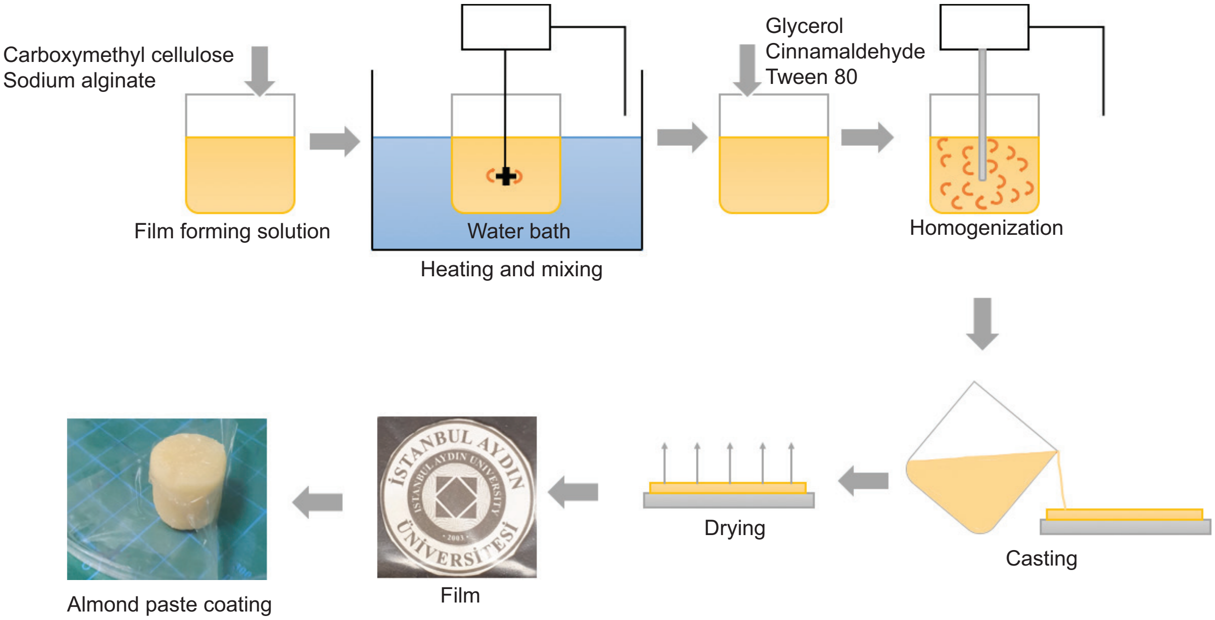


Figure 1. Schematic representation of production of film and coating on almond paste.

aged 30–60 years evaluated the appearance, odor, texture, taste, and the overall acceptability of almond paste samples based on the 7-point hedonic scale from 1 to 7 (1: very unpleasant to 7: very pleasant). Panelists were also asked to indicate their intention to eat and purchase the samples.

Storage study

For storage study, the most preferred (0.2 CN) among the coated almond paste samples according to the results of sensory analysis was used together with the uncoated control sample. The control and 0.2 CN samples were stored at 25°C for 15 days, and the microbial spoilage and oxidative degradation of the samples were monitored on day 1, 4, 7, 10, and 15 of storage.

For microbial analyses, 10 g of each sample was weighed under aseptic conditions and homogenized in a stomacher with previously sterilized peptone water to prepare the first dilution. Serial dilutions (10^{-1}–10^{-3}) were spread on PCA for total viable counts and incubated at 37°C for 48 h, and on PDA for yeast–mold counts, and incubated at 25°C for 5 days. The microbiological count was expressed as $\log_{10}$ colony forming units (CFU)/g sample.

For oxidation analysis, oil was extracted from each almond paste sample before analysis. The samples were extracted with hexane in a Soxhlet extractor, and then the solvent was removed with the help of a rotary evaporator (Faid *et al.*, 1995). The free fatty acid content of samples was analyzed according to the Association of Official Analytical Chemists (AOAC, 2000) method No. 940.28. The oil, mixed with ethanol and diethyl ether (1:1, v:v), was titrated with 0.1-N NaOH in the presence of phenolphthalein indicator until a permanent pink color was obtained. The results were expressed as %oleic acid. The peroxide value of samples was analyzed according to the AOAC (2000) method No. 965.33. The oil was mixed with acetic acid chloroform (1.5:1, v:v). Saturated potassium iodide solution was added to the mixture and kept in the dark, followed by the addition of distilled water and starch solution. The mixture was titrated with sodium thiosulfate until the purple color disappeared. The results were expressed as milli-equivalent of oxygen/kg (meq O_2/kg) of lipids.

Statistical analysis

The experimental data were given as mean ± standard deviation. Analysis of variance (ANOVA) and Duncan's multiple comparison test were performed using the SPSS software (IBM SPSS Statistics 19, SPSS Inc., Chicago, USA) and 95% confidence interval (95% CI) was used for all analyses.

Results

Characterization of films

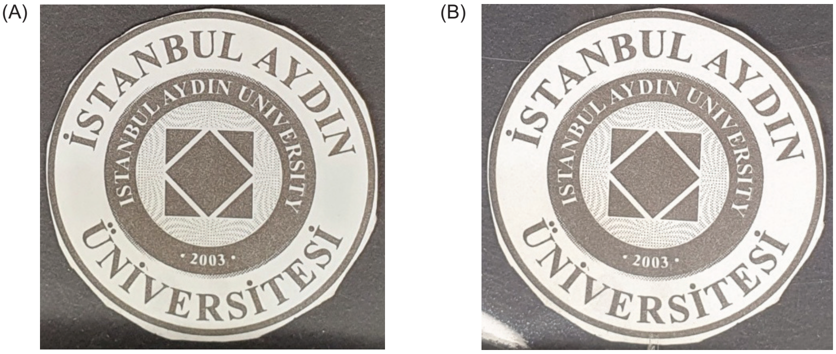
Thickness, density, and opacity

The thickness (mm), density (g/cm^3), and opacity parameters of films are presented in Table 1. To produce films

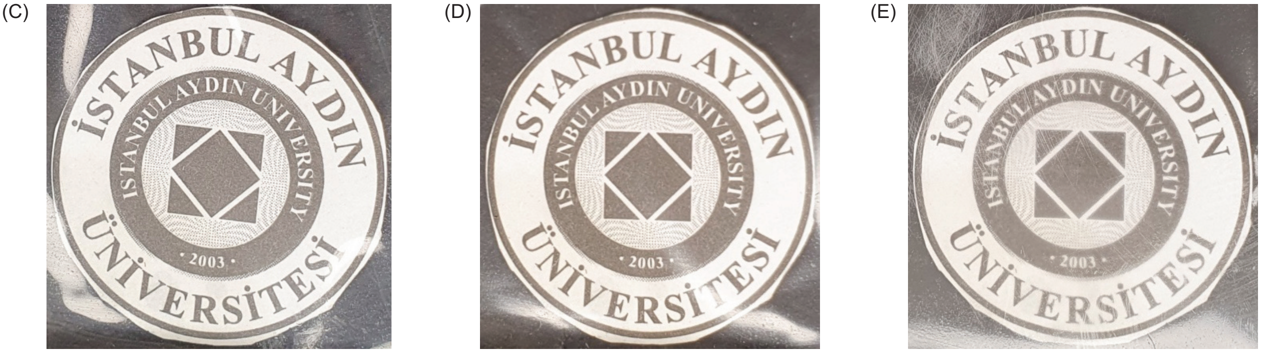
Table 1. **Thickness (mm), density (g/cm^3), water vapor permeability coefficient (WVP) values of film samples at 25°C (C: control; 0.1 CN, 0.2 CN, and 0.3 CN films containing 0.1%, 0.2%, and 0.3% [w/v] cinnamaldehyde [CN], respectively).**

Film	Thickness (mm)	Density (g/cm^3)	Opacity (A/mm)	WVP (g·mm/(m^2·h·kPa))
C	$0.025 \pm 0.003^{a,b}$	$1.36 \pm 0.10^{a,b}$	1.64 ± 0.05^{d}	0.18 ± 0.01^{c}
0.1 CN	0.022 ± 0.005^{b}	1.57 ± 0.14^{a}	6.55 ± 0.45^{c}	0.26 ± 0.02^{b}
0.2 CN	0.028 ± 0.005^{a}	$1.39 \pm 0.06^{a,b}$	7.75 ± 0.32^{b}	$0.24 \pm 0.01^{b,c}$
0.3 CN	0.032 ± 0.006^{a}	1.34 ± 0.10^{b}	29.58 ± 0.19^{a}	0.44 ± 0.03^{a}

Note: Different superscript alphabets in the same column indicate significant differences between different samples ($P < 0.05$).



No film C film



0.1 CN film 0.2 CN film 0.3 CN film

Figure 2. Visual differences in the appearance of films on the same visual background: (A) image without film, (B) C (control), (C) 0.1 CN (0.1% w/v), (D) 0.2 CN (0.2% w/v), and (E) 0.3 CN (0.3% w/v) cinnamaldehyde films.

with similar thickness after drying, the same volume of film solution was poured into containers. Film thickness directly affects water vapor barrier and mechanical strength properties as it was used in calculations. The thickness of the films varied between 0.022 mm and 0.032 mm. In comparison to the control, the addition of CN did not cause a significant change in thickness of the films ($P > 0.05$). Guo *et al.* (2020) also reported no significant difference between the thickness of CMC- and CN-containing films. Differences in film formulations and polymeric structure formed by molecular interactions in films change their density values (Pelissari *et al.*, 2013). Density of the films produced in this study varied between 1.34 g/cm^3 and 1.57 g/cm^3. Values showed that addition of CN did not cause a significant change in the density values of the films ($P > 0.05$).

The food packaging films are required to have high transparency and must exhibit the food product they cover. Therefore, the opacity values of food packaging films are crucial. The opacity values of films are given in Table 1, and the appearance of the same is demonstrated in Figure 2. The pure SA–CMC film (control film) had the lowest (1.64 ± 0.05) opacity value. Addition of CN and the increasing concentration of CN enhanced the opacity of SA–CMC film ($P < 0.05$). CN dispersed in the film with its characteristic yellow color increased light absorption of the film (Yan *et al.*, 2024). Similarly, in the literature, the opacity value of various films increased with addition of essential oil (Biswas *et al.*, 2018; Yan *et al.*, 2024). Biswas *et al.* (2018) stated that due to incompatibility between various essential oils added to CMC film, essential oil droplets created a separate phase and increased opacity. Visual comparison of the films in Figure 2 shows that opacity of the films increases with increased concentration of CN. It should also be emphasized that all images behind the films are still visible.

Water vapor permeability

Table 1 also shows the WVP results of films. WVP values of the films vary between 0.18 ± 0.01 g·mm/(m^2·h·kPa) and 0.44 ± 0.03 g·mm/(m^2·h·kPa). Addition of CN (0.1%) to the film significantly increased the WVP ($P < 0.05$), while 0.2% CN did not create a significant difference, compared to the control film ($P > 0.05$). A higher amount of CN (0.3%) caused a significant increase ($P < 0.05$) in WVP. Despite its hydrophobic structure, CN increased the WVP values of the films. While some studies in the literature reported that the WVP values of films decreased with the addition of hydrophobic essential oils and components (Amjadi *et al.* 2022), few studies stated that there were no significant changes or increase in WVP (Ayala-Zavala *et al.*, 2013). Dashipour *et al.* (2015) found that the addition of *Zataria multiflora* essential oil to CMC films increased the WVP values of the films, which was associated with the reducing effect of essential oil microdroplets on the cohesion forces of the films. In addition, Tween 80, used as a surfactant, may decrease the barrier feature against water vapor by increasing the volume between molecules by acting as a plasticizer (Brandelero *et al.*, 2010). In conclusion, CN and Tween 80 added to the SA–CMC film matrix increased WVP values due to structural modifications and plasticizing effects in film matrix.

Mechanical properties

The TS and EAB of film samples are shown in Figure 3. The TS of film samples varied between 18.48 N/mm^2 and 26.94 N/mm^2. It was observed that the TS values were not statistically different between control, 0.1% CN, and 0.2% CN containing films ($P > 0.05$), while it decreased in films containing 0.3% CN ($P < 0.05$). The EAB of film samples varied between 6.75% and 13.50%. The EAB values increased with increasing CN concentration, but there was no significant change in the values of 0.1 CN and 0.2 CN films, compared to the control ($P > 0.05$). The EAB of films containing 0.3% CN was significantly higher than the control ($P < 0.05$). The microdroplets of hydrophobic component decreased cohesion forces in the film polymer matrix and increased porosity and flexibility of the films (Han *et al.*, 2018). Therefore, adding a high amount of CN reduced the TS and enhanced the EAB of SA–CMC films. Similar results were obtained by Noshirvani *et al.* (2017) by adding cinnamon essential oil to CMC–chitosan film. As the amount of essential oil increased, TS decreased and EAB increased. This result was related to the increasing number of polymer–essential oil interactions instead of polymer–polymer interactions that weakened the structure of the film and reduced TS. In addition, essential oil showed a plasticizing effect and caused an increase in EAB values. The higher standard deviation (SD) of the CN-added films, compared to the control sample, indicated that CN in the films increased their heterogeneity. Furthermore, the 0.3 CN film exhibiting the highest value of WVP also indicated the same.

Fourier transform infrared (FT-IR) spectroscopy

The FT-IR spectra of the films are presented in Figure 4. FT-IR spectra of the films show the possible interactions between SA–CMC and CN. The spectra of films containing CN were not significantly different from the spectrum of the control film. The broadband between 3,200 cm^{-1} and 3,500 cm^{-1} matched the O–H stretching of SA–CMC films (Shao *et al.*, 2015). The C–H stretching of SA–CMC

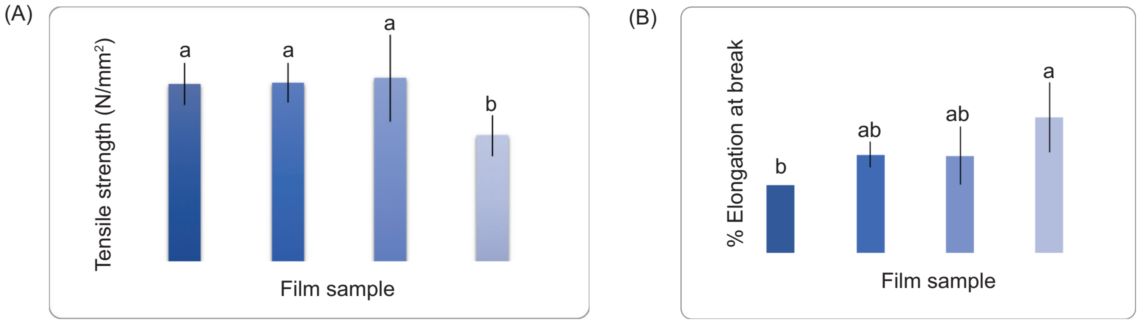


Figure 3. Mechanical properties of films (C: control; 0.1 CN, 0.2 CN, and 0.3 CN films containing 0.1%, 0.2%, and 0.3% (w/v) cinnamaldehyde, respectively) (A) tensile strength (N/mm^2) and (B) elongation at break (%).

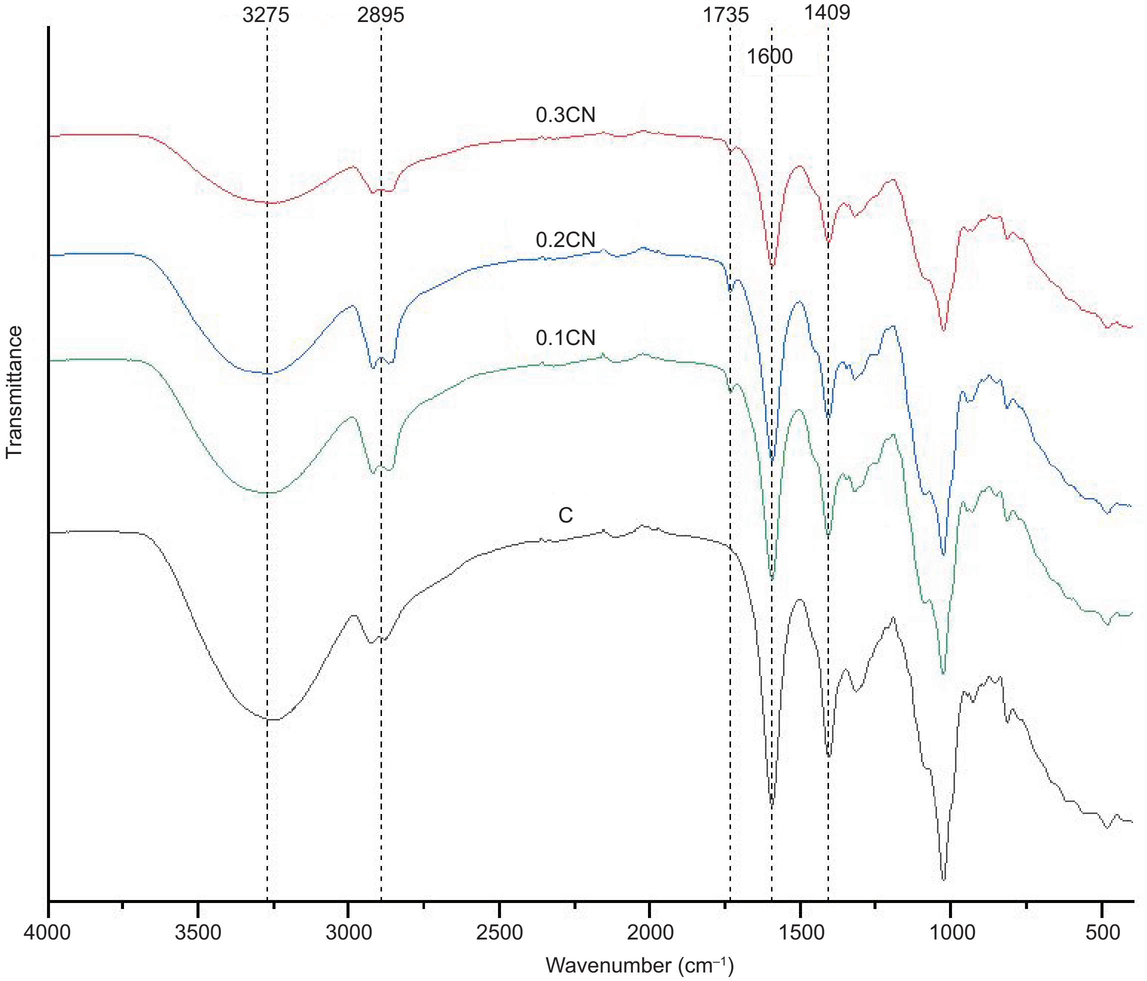


Figure 4. FT-IR spectra of films (C: control; 0.1 CN, 0.2 CN, and 0.3 CN films containing 0.1%, 0.2%, and 0.3% (w/v) cinnamaldehyde, respectively).

films occurred around 2,895 cm^{-1} as two peaks. The peaks around 1,600 cm^{-1} and 1,409 cm^{-1} matched antisymmetric and symmetric vibrations of COO^{-} groups of CMC and SA (Tong *et al.*, 2008). The spectra of CN-added films showed a new peak at 1,735 cm^{-1}, indicating the C=O stretching vibration from CN (Yang *et al.*, 2020). The intensity of the characteristic peaks of SA–CMC films at 3,275, 1,600, and 1,409 cm^{-1} decreased with increasing CN concentration. These changes suggested an interaction between CN components and functional groups in the SA–CMC film matrix. For this reason, it was opinioned that changes took place in the barrier and mechanical properties of the films.

Antimicrobial activity

The antimicrobial effect of CN is explained by the lethal effect it provides through the leakage of cell components because of its interaction with the membrane of bacterial cells (Alves *et al.*, 2020). The antimicrobial assays of CN-containing films were studied against *B. cereus*, *E. coli*, *L. monocytogenes*, *S. typhi*, and *S. aureus*. The average diameters of the inhibition zones (mm) for 0.1 CN, 0.2 CN, and 0.3 CN films are shown in Table 2, and the photographs of the zones are depicted in Figure 5. No zone formation was observed in the control film. CN interacts with the polymer structure, rather than the intact polymer–polymer interactions, by acting on the intimate bonding of CMC–SA polymer matrix (Noshirvani *et al.*, 2017). In the film samples containing CN, zones were formed against all tested bacteria. With increase in CN concentration, the zone diameter also increased ($P < 0.05$). Therefore, it was concluded that the CN concentration positively affected the antimicrobial activity against *B. cereus*, *E. coli*, *L. monocytogenes*, *S. typhi*, and *S. aureus*. As the amount of CN in the film content increased, its release from the film matrix and the inhibitory effect against microorganisms also increased. The antimicrobial effect of CN was attributed to the disruption of mitochondrial and plasma membranes, cell folding, and loss of cell wall regularity. Furthermore, CN

Table 2. Inhibition zone (mm) of the films (C: control; 0.1 CN, 0.2 CN and 0.3 CN films containing 0.1%, 0.2% and 0.3% [w/v] cinnamaldehyde, respectively).

Film	*B. cereus*	*E. coli*	*L. monocytogenes*	*S. typhi*	*S. aureus*
C	–	–	–	–	–
0.1 CN	3.96 ± 0.37^c	2.89 ± 0.17^c	3.04 ± 0.79^c	2.18 ± 0.20^c	2.33 ± 0.44^c
0.2 CN	9.63 ± 0.98^b	9.98 ± 2.74^b	9.56 ± 1.49^b	4.20 ± 1.36^b	5.29 ± 0.13^b
0.3 CN	14.31 ± 0.25^a	17.41 ± 0.82^a	12.82 ± 0.56^a	11.16 ± 0.38^a	8.04 ± 0.51^a

Note: Different superscript alphabets in the same column indicate significant differences between different samples ($P < 0.05$).

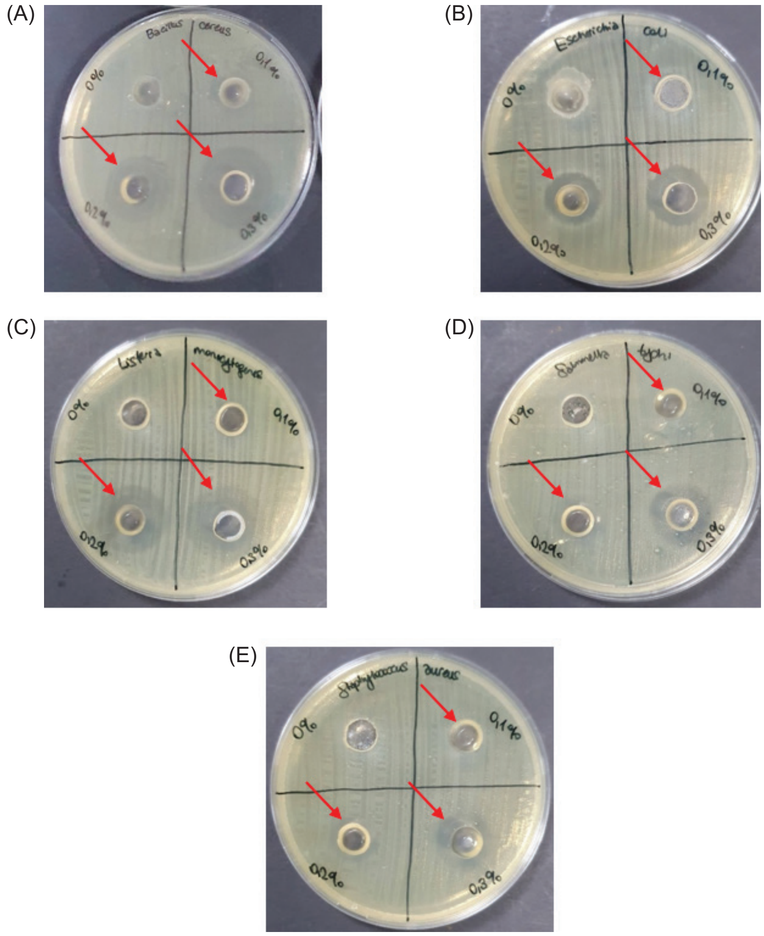


Figure 5. Images of well diffusion assay of films (0% for control; 0.1%; 0.2%, and 0.3% shows the films containing 0.1%, 0.2%, and 0.3% (w/v) cinnamaldehyde, respectively) for (A) *Bacillus cereus*, (B) *Escherichia coli*, (C) *Listeria monocytogenes*, (D) *Salmonella typhi*, and (E) *Staphylococcus aureus*.

demonstrated an inhibitory effect on enzymes responsible for synthesizing cell walls (Noshirvani *et al.*, 2017).

Analysis of wrapped almond paste

Sensory analysis

Table 3 shows the appearance, odor, texture, taste, and the overall acceptability scores of coated almond paste samples on the day of wrapping. Panelists were also evaluated on their willingness to eat and buy almond paste samples. The sensory parameters of the coated almond paste samples were found not to be different ($P > 0.05$) in terms of the CN content of the coating material. However, when the willingness to eat and the willingness to buy were evaluated, the 0.2 CN sample reached higher scores than others. Because of sensory analyses, the storage study was carried out with almond paste wrapped in 0.2 CN film.

Different opacity values of the films may have affected the appearance scores of almond paste samples wrapped with these films. When the results were examined, although no statistically significant difference was observed, the 0.2 CN sample had a higher appearance score. The willingness to eat and buy samples also confirmed this result. This result concluded that consumers considered the 0.2 CN product; in this product, both coating and wrapped product were visible, compared to the coatings with low and high opacity.

However, the most important distinguishable criterion for consumers here is probably the taste criterion. The low taste added by CN was not perceived by consumers, whereas the high taste because of CN was described as slightly bitter. Therefore, consumers tended for 0.2 CN product, which had moderate level of added CN. The SD of sensory scores was high because the tests were applied to untrained panelists. In addition, this situation clearly established that some panelists liked the products and others did not like them at all. Based on this result, it was interpreted that these products were acceptable to all consumer groups but appealed to a certain group of consumers.

Table 3. Sensory acceptance scores for appearance, odor, texture, taste, and the overall acceptability of coated almond paste samples with films (0.1, 0.2, and 0.3 cinnamaldehyde [CN] films containing 0.1%, 0.2%, and 0.3% [w/v] CN, respectively).

Sensory parameter	0.1 CN	0.2 CN	0.3 CN
Appearance	4.56 ± 1.46^a	4.84 ± 1.46^a	4.68 ± 1.67^a
Odor	4.08 ± 1.71^a	4.60 ± 1.67^a	4.14 ± 1.62^a
Texture	4.18 ± 1.84^a	4.48 ± 1.75^a	4.34 ± 1.73^a
Taste	4.12 ± 2.04^a	4.52 ± 1.81^a	4.26 ± 1.99^a
Overall acceptability	4.28 ± 1.67^a	4.70 ± 1.46^a	4.46 ± 1.49^a
Willingness to eat			
Yes	21	24	20
Maybe	8	14	12
No	21	12	18
Willingness to buy			
Yes	14	15	12
Maybe	12	19	19
No	24	16	19

Note: Different superscript alphabets in the same column indicate significant differences between different samples ($P < 0.05$).

Storage study of almond paste

Figure 6 shows the images of almond paste samples during wrapping as well as wrapped with 0.2 CN film. The films maintained their structural integrity over the 15-day storage period. The total mesophilic aerobic counts of uncoated and coated almond paste samples varied between 2.46 log CFU/g and 3.39 log CFU/g during 15 days of storage at 25°C (Table 4). Its low microbial counts in almond paste are associated with its low water activity (Faid *et al.*, 1995). Takma *et al.* (2018) investigated the shelf life of almond paste in glass jars and polyethylene bags at room temperature and refrigerator temperature, and reported that the total mesophilic aerobic counts increased depending on storage under all conditions, and they preferred to store almond paste in glass jars at room temperature because of the high storage cost at refrigerator temperature. The total mesophilic aerobic counts of uncoated almond paste increased until the seventh day of storage and reached its maximum level in all samples on the last day of storage. On the first day of storage, the total mesophilic aerobic counts of coated almond paste were at their lowest level. It increased up to the fourth day of storage and gradually decreased in subsequent days. The counts of coated samples on the 15th day were lower than that on the fourth day ($P < 0.05$). It was intended that this happened due to the fact that the analyses were conducted using different almond paste samples, or that the CN effect became apparent on almond paste with the passage of time. The total mesophilic aerobic counts were found to be lower in coated almond paste than in uncoated almond paste except on

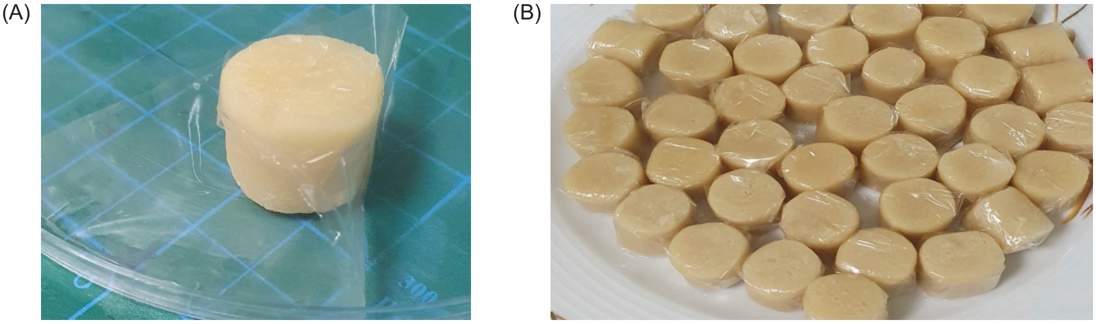


Figure 6. Images of almond paste samples (A) during wrapping and (B) after wrapping with 0.2 CN film containing 0.2% (w/v) cinnamaldehyde.

the fourth day of storage ($P < 0.05$). Because the films were produced in a laboratory environment and the coating was done manually, it was intended that there was an increase in microbial load of coated samples during first days. Decrease in total mesophilic aerobic bacteria count following an initial increase could be due to CN activity, or it could also be due to the use of different samples during the storage experiment. On the final day of storage, the results showed a decrease in the total mesophilic aerobic count because of CN transfer that occurred when the CN-enriched film contacted almond paste samples.

The total yeast–mold counts of uncoated and coated almond paste samples varied between 2.80 log CFU/g and 3.56 log CFU/g during 15 days of storage (Table 4). The total yeast–mold counts of uncoated and coated almond paste samples increased gradually until the seventh day of storage ($P < 0.05$), and on the last storage day, there was no significant change compared to the seventh day ($P > 0.05$). While the total number of yeast and mold in uncoated almond paste was lower than in coated almond paste in the first two days of storage because of the effect of coating film or the initial load of almond paste samples, while no statistically significant difference was observed between the samples on other days of storage ($P > 0.05$).

The free fatty acid of uncoated and coated almond paste samples varied between 0.55% and 1.12% during 15 days of storage (Table 4). Almond paste samples are susceptible to oxidation because of the high content of unsaturated fatty acids in almonds, and the amount of free fatty acid is expected to increase during storage. The content of free fatty acid in uncoated and coated almond paste samples increased significantly ($P < 0.05$) with storage time. Lin *et al.* (2012) also reported that there was an increase in free fatty acid in almond samples during storage. Moreover, Çapanoğlu and Boyacıoğlu (2008) investigated the quality and shelf life of almond paste using stabilizers, maltose syrup, and antioxidants, and reported that the free fatty acid in the samples increased significantly after the second week of storage. When uncoated and coated almond paste samples were compared, no statistically significant difference was found between the samples on daily basis ($P > 0.05$). The coating had no significant effect on the content of free fatty acid of almond samples during storage because the content was related to the hydrolysis of oils, which occur due to moisture and enzyme activity.

The peroxide value of uncoated and coated almond paste samples varied between 0.00 meq O_2/kg and 6.46 meq O_2/kg during 15 days of storage (Table 4). Dhankhar *et al.* (2022) investigated the storage stability of almond paste enriched with almond peel as an antioxidant and reported that peroxide value in the samples was between 0.03 meq O_2/kg and 4.39 meq O_2/kg and it increased with storage. Peroxide values were below the detection level on day 1 and day 4 for both uncoated and coated samples. Peroxide value of the samples could be detected from the seventh day of storage, and as storage progressed, the peroxide value increased gradually ($P < 0.05$). Although coated almond paste showed lower peroxide value than uncoated almond paste on all days, the difference between the samples was found to be statistically significant only on the last day of storage ($P < 0.05$). The coating reduced peroxide value in almond paste samples during 15 days of storage. CN inhibits chain reactions during lipid peroxidation by scavenging free radicals with its high antioxidant activity (Keshvari *et al.*, 2013).

Table 4. Effect of film containing cinnamaldehyde (CN) on total mesophilic aerobic counts, total yeast–mold counts, free fatty acid, and peroxide values of almond paste during storage.

Analysis	Day 1	Day 4	Day 7	Day 10	Day 15
Total mesophilic aerobic count (log CFU/g)					
Uncoated	2.86 ± 0.03^{d}	$3.03 \pm 0.17^{c,d}$	$3.28 \pm 0.02^{a,b}$	$2.99 \pm 0.03^{c,d}$	3.39 ± 0.01^{a}
Coated	2.46 ± 0.15^{e}	$3.14 \pm 0.13^{b,c}$	$3.08 \pm 0.17^{b–d}$	$2.95 \pm 0.22^{c,d}$	2.87 ± 0.03^{d}
Total yeast–mold counts (log CFU/g)					
Uncoated	2.80 ± 0.04^{d}	3.05 ± 0.21^{c}	$3.50 \pm 0.10^{a,b}$	$3.29 \pm 0.07^{b,c}$	$3.38 \pm 0.03^{a,b}$
Coated	$3.28 \pm 0.06^{b,c}$	$3.41 \pm 0.19^{a,b}$	3.56 ± 0.16^{a}	3.09 ± 0.29^{c}	$3.40 \pm 0.03^{a,b}$
Free fatty acid (%)					
Uncoated	0.55 ± 0.02^{c}	0.82 ± 0.00^{b}	0.79 ± 0.23^{b}	$0.70 \pm 0.05^{b,c}$	1.03 ± 0.15^{a}
Coated	$0.68 \pm 0.00^{b,c}$	$0.74 \pm 0.00^{b,c}$	0.82 ± 0.05^{b}	$0.65 \pm 0.06^{b,c}$	1.12 ± 0.05^{a}
Peroxide value (meq O_2/kg)					
Uncoated	–	–	2.77 ± 0.02^{d}	4.07 ± 0.08^{c}	6.46 ± 0.19^{a}
Coated	–	–	2.58 ± 0.01^{d}	3.80 ± 0.15^{c}	5.76 ± 0.04^{b}

Coated and uncoated samples within the same analysis were statistically evaluated at different storage periods. The values with different superscript alphabets in the same analysis group are significantly different ($P < 0.05$).

Conclusions

In this study, edible films based on grade SA and CMC were enriched with CN and their thickness, density, opacity, water vapor permeability, tensile strength, maximum elongation at break, and chemical bands with FT-IR were investigated. The effectiveness of CN-containing film was tested to protect almond paste from microbiological spoilage and oxidative degradation during storage at 25°C. Increased amounts of CN resulted in more opaque films. Film samples, including the highest level of CN (0.3%) had the lowest tensile strength and the highest elongation at break than other CN-added and control films. With increasing CN concentration, the antimicrobial activity of films against pathogenic bacteria also increased. However, sensory analysis of wrapped almond paste showed that the highest concentration of CN (0.3%) reduced consumer willingness to purchase and eat, while film containing 0.2% CN (0.2 CN) found an appropriate concentration without any negative effect on consumer desirability. The storage study of coated almond paste with 0.2 CN film yielded positive findings regarding total mesophilic aerobic count and peroxide value. Based on these findings, it was concluded that SA–CMC film produced with CN was applicable for preserving food products, such as almond paste. Despite the fact that this coating is expensive, it is important to consider the value of almond paste. It must be emphasized that the amount of CN added to the film is the most important factor. Although antimicrobial activity increases as the amount of CN increases, a certain level must remain for food-specific sensory characteristics. Therefore, the future studies should continue to investigate the effects of coatings on different food products.

Data Availability Statement

The data are available from the corresponding author upon request.

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Author Contributions

Orhun Macit: formal analysis, and writing – original draft. Meral Yildirim-Yalcin: conceptualization, investigation, and writing – review and editing. Hatice Sena Olcay: methodology, and writing – original draft. All authors had read and agreed to the published version of the manuscript.

Conflicts of Interest

The authors declared no conflict of interest.

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