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Incorporating carboxymethyl cellulose into microfibrillated cellulose for barrier coating to delay banana browning and extend shelf life

Jing Geng[a], Nicole Stark[b], Peter Kitin[b], Xiao Zhang[a,*], Nayomi Z. Plaza[b], J.Y. Zhu[b,*]

[a] Voiland School of Chemical Engineering and Bioengineering, Washington State University, Richland, WA 99354, USA
[b] USDA Forest Service, Forest Products Laboratory, Madison, WI 53726, USA

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ABSTRACT

Postharvest fruit losses can be 25–50 %. This study is to develop a microfibrillated cellulose (MFC)-based low-cost barrier coating technology to extend banana shelf life and decrease postharvest losses. The coating performance of MFC adding carboxymethyl cellulose (MFC/CMC) on banana was evaluated by monitoring banana peel appearance/color browning, weight loss, firmness, and total soluble solids (TSS) at the ambient condition. The results showed that MFC/CMC coating with the addition of a proper amount of CMC could effectively delay banana browning by 2 days and decrease weight loss over 30 % compared with control. When using aqueous suspensions of MFC/CMC with concentration (wt%) ratio of 0.55:0.05 for coating, banana ripening process was not slowed with respect to banana browning, in other words, MFC/CMC (0.55:0.05) coating delayed peel browning by 2 days, it also simultaneously decreased softening. This work has potential for developing a low-cost and plant-based coating material to extend fruit shelf life.

1. Introduction

Banana, as one of the world's major fruit crops, is mainly cultivated in the tropic and subtropic regions. Its global production reached 135.1 million tons with import and export value up to USD 28.1 billion in 2022 according to Food and Agriculture Organization of the United Nations (FAOSTA, 2022). As a typical climacteric fruit, banana is harvested in the mature green state to minimize postharvest loss (Anyasi et al., 2013). Rapid respiration and a surge in ethylene production triggers the rapid banana ripening (Brat et al., 2020; Domínguez & Vendrell, 1994). As a result, bananas go through several irreversible biochemical changes in their appearance, weight, firmness, taste, odor toward the end of shelf life (Maduwanthi & Marapana, 2019). Among these characteristics, banana's appearance or peel color is mostly used by consumers as the criteria to determine banana quality in purchasing, partly because banana peel color is a qualitative indicator of ripening (Symmank et al., 2018). Hence, for retailers, delaying banana peel browning is an important key to boost profits in addition to decreasing banana weight loss.

Although a complete understanding of banana peel color browning is not available, it is generally understood that the brown pigments of benzo-quinone and melanin are formed through the enzymatic reactions of natural phenols with oxygen (Ali et al., 2022; Joslyn & Ponting, 1951). Furthermore, the formation of brown spots occurs with the increase in local oxygen (Mosera et al., 2009; Steinbock et al., 2022). This suggests that controlling the availability of oxygen especially locally is critical to control banana peel browning. It is generally understood that barrier coating is an effective approach to prolong the shelf life of bananas (Amoroso et al., 2022; Geng et al., 2024; Hu et al., 2016; Shinga & Fawole, 2023). The coating film forms a physical barrier not only protecting bananas from physical impacts and microbiological attacks but also preventing the transport of various gases in and out of bananas to significantly slowdown the biochemical reactions during ripening (Ali et al., 2022; Firdous et al., 2023). The regulation of ethylene, water vapor, O_2, CO_2 and other gases inside the banana can delay banana weight loss, browning, and fruit deterioration (Ali et al., 2022; Chavan et al., 2023; Deng et al., 2017; Kim et al., 2022). An ideal coating should be capable of stopping peel browning in addition to slowing but not stop ripening from consumer preference point of view because consumers prefer a ripe banana for taste and nutrients. This is certainly difficult to achieve. Studies have been reported in using optimized storage in cooled shipping containers and modified atmosphere packaging (Jedermann et al., 2013; Nguyen et al., 2004), as well as adding natural antioxidant to inhibit phenol enzymatic reactions (Kim et al., 2022; Paudel et al.,

* Corresponding authors.
E-mail addresses: x.zhang@wsu.edu (X. Zhang), Junyong.zhu@usda.gov (J.Y. Zhu).

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2020) to slow down browning. Using some natural compounds such as chitosan that contains oxalic acid also showed effective in decreasing banana browning during storage (Wantat et al., 2021). However, comparative studies are lacking on evaluating the effectiveness of barrier coatings for both browning prevention and ripening control.

Among various coating materials, naturally derived compounds such as protein, lipid, and polysaccharides attracted great interest as safe and preferred alternatives to petrochemical-based biodegradable materials (Wang et al., 2024; Wu et al., 2023). Much progress has been made in using these eco-friendly barrier coating materials for fruit preservation (Ali et al., 2022; Deng et al., 2017; Pham et al., 2023; Wantat et al., 2022). The key barriers to move these technologies to commercial market are cost of the materials (Wang et al., 2024). Among naturally derived and biodegradable materials, cellulose is a low-cost alternative (Bahmid et al., 2023). However, most existing studies require extensive and cost prohibitive chemically or mechanically processing cellulose into cellulose derivatives, cellulose nanomaterials, and/or making cellulose-based composite formulations with expensive or petrochemical materials (Deng et al., 2017; Deng et al., 2018; Zheng et al., 2022; Zhou et al., 2023). Microfibrillated cellulose (MFC) as a renewable material can be sustainably produced from low-cost plant biomass through moderate mechanical processing (Wang et al., 2012; Zhang et al., 2023). MFC film can decrease the permeation of water vapor, O_2, CO_2 (Belbekhouche et al., 2011), implying the potential of MFC barrier coating for banana preservation. Our previous study demonstrated that MFC suspension forms a film on banana surfaces to slow banana ripening process (Geng et al., 2024). However, the study did not show substantial slow down banana peel browning using MFC coating.

Carboxymethyl cellulose (CMC), a cellulose derivative and food additive, has a strong film-forming capability (Shahbazi et al., 2016) and is also capable of preventing the permeation of water vapor, O_2 and CO_2 (Bifani et al., 2007). CMC has been shown effective for barrier coating of chicken breast with the addition of ethanolic propolis extract (El Sheikha et al., 2022). Furthermore, some studies reported that CMC coating can delay the decay of fruits and vegetables such as apples, strawberries, eggplants, tomatoes, et al., during storage (Dey et al., 2023; Panahirad et al., 2021). Recent studies have shown that CMC alone can slow down banana browning. For example a barrier coating containing 1.5 % CMC and 2 % surfactant (Tween 80) significantly delayed the banana peel browning (Kim et al., 2022). CMC coating with 0.25 % Tween and 1 % glycerol not only delayed banana ripening but also inhibited banana color change (Ali et al., 2022). Due to the brittleness and high hydrophilicity of pure CMC film, and the poor adhesion of pure CMC solution onto banana surface, CMC alone lacks sufficient adhesiveness and flexibility, making it unsuitable for banana coating (Yildirim-Yalcin et al., 2022). The objective of the present study is aimed at using MFC with the addition of CMC to develop a low-cost cellulose-based barrier coating to decrease banana browning and weight loss without significantly slow down banana ripening. The novelties of the present study are: (1) using low-cost and sustainable MFC, (2) simple formulation by simply mixing aqueous MFC suspension with CMC, and (3) decreasing banana browning while maintaining freshness and taste. The significance of this work is its potential for developing a plant-based simple barrier coating formulation for climacteric fruits without using cost prohibitive materials and/or unsafe chemicals.

2. Materials and methods

2.1. Materials

A virgin bleached kraft hardwood pulp (BKHP) was supplied by WestRock Company. Its chemical composition and moisture content can be referred to our previous work (Geng et al., 2024). Sodium carboxymethyl cellulose (average Mw ~700,000 Da) was purchased from Sigma-Aldrich Corporation. Fresh Chiquita bananas from a local market in Madison, Wisconsin, were used as purchased for the study without applying any ethylene treatments. Bananas chosen have a light yellow and greenish color with typical length of approximately 16 cm and 3 cm in diameter.

2.2. Preparation of microfibrillated cellulose/carboxymethyl cellulose (MFC/CMC) suspensions

Oven-dried weight of 60 g BKHP were dispersed in water at 2 wt%, then disintegrated in a disintegrator (Model 73–06-01, TMI, Ronkonkoma, New York, USA) for 10,000 revolutions, subsequently mechanical fibrillated in a stone disk mill – SuperMassColloider (SMC) (Model MKZCA 6–2 J, Disk MDGA 6–80#, Masuko Sangyo Co., Ltd., Japan) as described previously (Geng et al., 2024; Hu et al., 2015). The BKHP suspension underwent four passes through the SMC to obtain MFC. The fibrillation energy consumption was recorded and listed in Table S1.

Sodium carboxymethyl cellulose (CMC) was added into water and then stirred thoroughly until dissolution. MFC suspension was then poured into the CMC solution and mixed thoroughly to obtain a MFC/CMC suspension. Various CMC:MFC concentration (wt%) ratios were used to obtain mixtures with the concentration of the combined CMC and MFC maintained at 0.60 wt%. Specifically, the concentration ratios of MFC (wt%):CMC(wt%) = 0.60:0.00, 0.55:0.05, 0.50:0.10, 0.45:0.15, and 0.40:0.20. These CMC loadings are substantially lower than those in literature studies using CMC as the main ingredient with minimum concentration of 0.5 % (Ali et al., 2022; Kim et al., 2022). The mixture suspensions were stored in a refrigerator until use.

2.3. Characterization of MFC/CMC suspensions

The morphology of the BKHP and MFC samples was observed using light microscopy (LM) and scanning electron microscopy (SEM) as described previously (Zhang et al., 2023) to observe the fiber diameters and fibril structure. For SEM analysis, suspension drops of the BKHP and MFC suspensions were frozen, then freeze dried and sputter-coated with 5 nm of gold. The SEM images were obtained using a Zeiss Evo 15 (LaB6) SEM system or a Zeiss Gemini 450 FE-SEM (Carl Zeiss AG, Oberkochen, Germany). SEM and light microscopic images of original fibers and the MFC were supplied in Fig. S1.

The wettability of MFC/CMC suspensions on banana peel surface was evaluated using contact angle measurements by an optical tensiometer (TL100, Theta Lite, Biolin Scientific Oy, Finland). The contact angle was obtained by imaging immediately after a drop of MFC/CMC suspension impinging onto the banana peel surface.

2.4. Preparation of MFC/CMC films through casting

MFC/CMC films were produced using solution casting in petri-dishes. The amount of MFC/CMC suspension mixture was pre-determined to ensure the resultant films have a grammage of 10 g/m^2.

2.5. Characterization of MFC/CMC films

Visual appearance and gas permeation are the two most important properties of coating film relevant to banana preservation. The optical transparency of the MFC/CMC films as a measure of visual appearance was evaluated by photography and UV–vis measurements (Model 8453, Agilent Technologies; Palo Alto, California, USA) in the wavelength range of 200–800 nm. The surface microstructure of the top and the bottom sides of MFC/CMC casting films were observed by SEM. In addition, small pieces (about 10 mm^2) of MFC/CMC films were gently peeled off coated banana surface, lyophilized, and then observed by SEM, as described earlier. Water vapor permeation (WVP) of the films was measured according to ASTM E96/E96M-21 (doi:https://doi.org/10.1520/E0096_E0096M-21). Briefly, a 68–3000 EX-Cup vapometer assemblies (Thwing-Albert Instrument Company, West Berlin, New Jersey, USA) was used to gravimetrically determining the amount of water permeated through an MFC/CMC film and adsorbed on 150 g anhydrous calcium chloride contained in a cup with an opening diameter of 6.35 cm. The cup was sealed by the MFC/CMC film after being cut into a circular shape of approximately 7.0 cm in diameter, placed on the cup opening and sandwiched between two neoprene gaskets. The assemblies were placed under at 21 °C, 50 % RH, and a water vapor saturation pressure of 2493 Pa. The test cup was weighed every 24 h until constant weight was reached.

The hydrophilia of MFC/CMC casting films was determined also using the optical tensiometer (TL100, Theta Lite, Biolin Scientific Oy, Finland). The real-time contact angles of the water drop on MFC/CMC casting films were recorded at the time interval of 10 s for 60 s.

2.6. Barrier coating bananas

Bananas were first washed with tap water to remove stains and residual chemicals and wipe-dried with paper tower before coating, then immersed into various MFC/CMC suspensions for approximately 3 min, and dried under the ambient condition (23 ± 3 °C, 38 ± 3 % RH).

2.7. Banana appearance and browning analysis

The appearance of uncoated and coated bananas was observed by photographing the two opposite sides of each banana during storage. Consistent lighting and camera position were maintained for all photos taken.

The two photos of the opposite sides of each banana formed a group. Each image group was processed using Python 3.8. Background subtraction was first applied to each image. Each image was then converted into a color data set in RBG space. With thresholding, the RBG data set was then divided into either yellow or black (brown) pixels. The ratio of the numbers of yellow pixels over the total numbers of yellow and black pixels was calculated as the browning fraction. Thresholding in the color space was accomplished using a cylindrical cone as the threshold boundary as described in the Support Information. The validity of this thresholding approach in RBG space was demonstrated by comparing the browning fraction with that calculated using gray level thresholding of the same banana image (Fig. S2). It was further verified by comparing the browning fractions with those calculated using a set of selected RBG values (color patterns) thresholding (Fig. S3). Both comparisons showed the results agree well from all three methods.

The banana browning fraction data was fitted to a logistic model as shown by Eq. (1) to describe and predict banana browning process:

$$y = A_2 + (A_1 - A_2)/(1 + (x/x_0)^q) \quad (1)$$

$$EC_{20} = 10^{(log(x_0)+log(0.25)/q)} \quad (2)$$

where A_1 is the banana browning fraction at the beginning of the experiments (set as 0.09); A_2 is the banana browning fraction at the end of the experiments (set as 1.00); q (day^{-1}) is the maximum slope of banana browning rate over time; x_0 (day) is the storage time corresponding to the maximum slope (q), and EC_{20} (day) is the start time of rapid browning process.

2.8. Weight loss and firmness

Banana weight loss was determined by weighing the bananas daily.

The firmness of a banana was measured upon peeling off the banana peel using a high accuracy handheld fruit firmness tester (FR-5105, Ace Instruments, Lutron electronic enterprise Co., Ltd., Taiwan) which was equipped with an 11-mm penetrometer tip (FRTP-11). Peeled banana fruit was hand punctured to a depth of approximately 8 mm. The force reading from the tester was recorded as the firmness of the banana. Each banana was tested on its top, middle and bottom (stem end) sections (Fig. S4).

2.9. Total soluble solids (TSS)

5 g of banana fruit were mashed, then fully dispersed in 5 mL distilled water, and centrifuged for 2 min at 10,000 rpm. The collected supernatant was used to determine banana TSS using a digital handheld pocket refractometer (3810 PAL-1, Cole-Parmer, Atago Co., Ltd., Tokyo, Japan).

2.10. Statistical analysis

Measurements of optical transparency, contact angles, banana weight, firmness, and total soluble sugars were carried out at least 2 times, and the collected data was represented as the mean value ± standard deviation. The differences among means of the trials were detected by a significant p-value ≤0.05. Banana weight loss and color (imaging) were measured every day while banana fruit firmness and total soluble sugars every 3 days from day 1.

3. Results

To understand the effects of MFC/CMC barrier coating on slowing banana peel browning and ripening process, the physical and optical properties of the films made of the five barrier coating formulations through casting in petri-dishes were first examined.

3.1. Hydrophilia and water vapor permeation of casting films

Surface of cellulosic films was hydrophilic as expected. Increasing

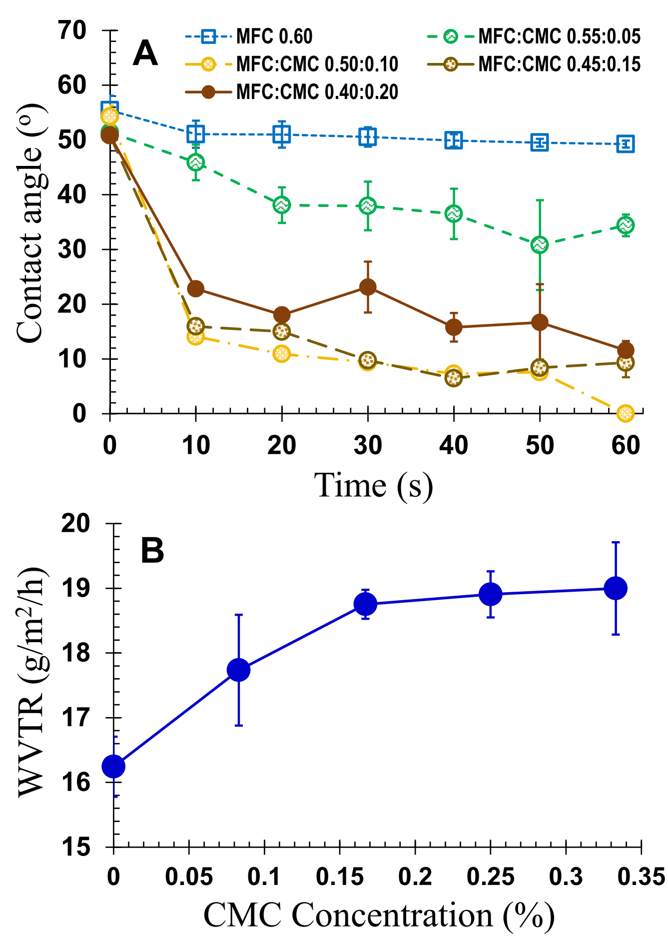

Fig. 1. Physical properties of MFC/CMC casting films. (A) Time-dependent water contact angle; (B) Effect of CMC loading on film water vapor transmittance rate measured at 21 °C and 50 % RH.

CMC loading decreased the measured water contact angle in general (Fig. 1A), therefore increasing the hydrophilic nature of the film. The water contact angles on the pure MFC casting film decreased slightly with time within the first 60s. In contrast, the contact angle of the films with higher CMC loadings decreased more rapidly with time, and the water drop was pretty much completely absorbed by the film in 60 s. This indicates that the barrier coating, especially those with higher CMC content, is highly wettable and potentially washable when applied to banana peels, in agreement with our earlier study (Geng et al., 2024). It was observed that water droplets caused the CMC-containing casting films to shrink and wrinkle (Fig. S5). The measured final contact angle after 60 s of approximately 10° from the casting film MFC/CMC (0.40:0.20) might be a result of strong film shrinkage and surface tension induced by CMC solubilization in water.

Water vapor permeation directly affect banana weight loss, respiration, and therefore ripening. Permeation of MFC/CMC casting made films was measured at 21 °C and 50 % RH. The variations of water vapor permeability (WVP) among films made of different MFC/CMC formulations were within measurement standard deviations (Table S2), suggesting that substituting CMC with MFC may not significantly affect casting film WVP. However, variations in water vapor transfer rate (WVTR) among MFC/CMC casting made films with different CMC loadings were observed as shown in Fig. 1B. The WVTR values of pure MFC and pure CMC films were 16.2 and 27.1 $g \cdot h^{-1} \cdot m^{-2}$, respectively (Table S2). The WVTR of the MFC/CMC films increased first rapidly with increasing CMC loading and then plateaued (Fig. 1B). The greater WVTR of films with higher CMC substitution may be contributed by their thinner film thickness (Table S2). The greater WVTR of films with a greater amount CMC such as the pure CMC film indicated that CMC bonds to water molecules more easily than MFC in addition to their thinner thickness, in agreement with the film contact angle measurements (Fig. 1B).

3.2. Casting film microstructure

The microstructure of MFC/CMC films affects gas permeation. SEM images of both the top and the bottom (the surface contacting the petri-dish) of films are shown in Fig. 2. Comparing the bottom surfaces of the pure MFC (0.60) film (Fig. 2A) and the MFC/CMC (0.50:0.10) film (Fig. 2B), the pure MFC film has holes and gaps on the bottom surface. The addition of CMC resulted in a flatter surface and denser microstructure because CMC particles are much smaller than the fibrils of MFCs and can fill the voids formed within the MFC fibril networks, therefore leading to a better film-forming performance than MFC. Due to gravity, CMC tends to settle down to the bottom of the petri-dish through the voids in the MFC networks to form a flatter and smoother surface on the bottom than on the top surface, as shown in Fig. 2. Because of the CMC settling phenomenon, the top surfaces of the films of MFC/CMC (0.55:0.05), MFC/CMC (0.45:0.15), and MFC/CMC (0.40:0.20), are all similar to the top surface morphology of the pure MFC film. In contrast, the top surface of the pure CMC film is very smooth and flat.

3.3. Optical transparency of MFC/CMC casting films

Barrier coating film optical transparency can affect fruits appealing to customers. Therefore, the optical transparency of the casting films of MFC/CMC suspensions were measured. Substituting MFC with CMC increased the optical transparency of the resultant MFC/CMC casting films as shown by the optical spectra of the MFC/CMC casting films (Fig. 3). However, this change in optical transparency was not obvious by visual observation. These MFC/CMC casting films in general had very good optical transparency in the visible wavelength as shown, suggesting that the appealing color of banana peels to consumers would not be affected by the MFC/CMC barrier coating.

3.4. Banana peel surface wettability by MFC/CMC suspensions

The wettability of MFC/CMC suspensions on banana surface is critical to formulating a uniform coating layer on banana surface. The wettability was determined based on contact angle measurements of a suspension drop upon impacting banana surface. As shown in Fig. 4, increasing CMC loading increased the contact angle, i.e., decreased the wettability of the suspension. The banana surface is hydrophobic due to the wax and cutin in banana peel cell walls (Freeman & Turner, 1985). CMC with a much smaller molecular weight and more hydroxyl groups, had a better hydrophilia than MFC, leading to a decrease in the interaction force between the suspension and the hydrophobic banana surface. Overall, all the contact angles of MFC/CMC suspensions at total carbohydrates concentration of 0.6 % were lower than 90°, indicating a good wettability of the MFC/CMC suspensions on banana surface.

3.5. Microstructures of barrier coated films on banana surface

SEM images of MFC/CMC barrier coating films peeled off from banana surface are shown in Fig. 5. It was noticed that the overall fibril distribution and structure in these films are similar to those found in casting films (Fig. 2). However, these films contain dimples (positive prints of the epicarp surface) that were not present in casting films with a petri-dish. Increasing CMC loading in coating increased the visibility of these dimples (middle column in Fig. 5). These hemispherical dimples have a rather uniform diameter of approximately 10 μm and were formed on the coating film by protrusions of the banana epidermal cells on the coating films (Brat et al., 2016; Moser et al., 2009). With the increase in CMC addition, the adhesion effect of coating films on the banana surface was improved and the protrusions were more pronounced and deeper. When the CMC addition amount was too high such as MFC/CMC (0.40:0.20), the coating film could not be peeled off (not shown) from banana surface due to the strong adhesion. A previous SEM study of banana fruit coatings also demonstrated that addition of cellulose nanofibrils (CNF) and nanocrystals (CNC) improves the adhesion of the coating films (Deng et al., 2017).

The images with greatest magnification in Fig. 5 (right column) also indicate that increasing CMC loading decreased the porosity of the films coated on banana surface, similar to adding CNC found in literature (Deng et al., 2017). The voids or gaps between fibrils in the pure MFC can allow gas permeation for banana breathing/respiration. The MFC/CMC (0.55:0.05) coating film also contain voids in favor of banana respiration. The MFC/CMC (0.50:0.10) coating film contains much smaller pores which may significantly hinder banana respiration. Overall, increasing CMC concentration decreased film porosity. As a result, too high a CMC loading can near completely shut down banana respiration as will be discussed later.

3.6. Effect of MFC/CMC barrier coating on bananas peel browning

Banana peel visual appearance in terms of color will change from green, yellow to dark brown as they went through ripening and decaying. The initial browning percentages of the peel surface area of the bananas with and without MFC/CMC barrier coating were approximately the same of 9 % as shown in Fig. 6A. The browning fractions were obtained from image analysis of photographic pictures of the bananas (Fig. 7) as described earlier and Support Information. The tested bananas were selected from the same hand with similar maturity. Based on the browning data, the control bananas and the bananas coated with pure MFC film began to brown at Day 4, and the bananas coated with MFC/CMC films began to brown at Day 6 (Fig. 7). This indicated that pure MFC barrier coating had minimal effect on preventing banana peel browning, in agreement with our earlier study (Geng et al., 2024), though it delayed the banana ripen process as shown by the same earlier study based on banana firmness and total soluble solids (TSS). However, with the addition of CMC, the MFC/CMC barrier coating significantly

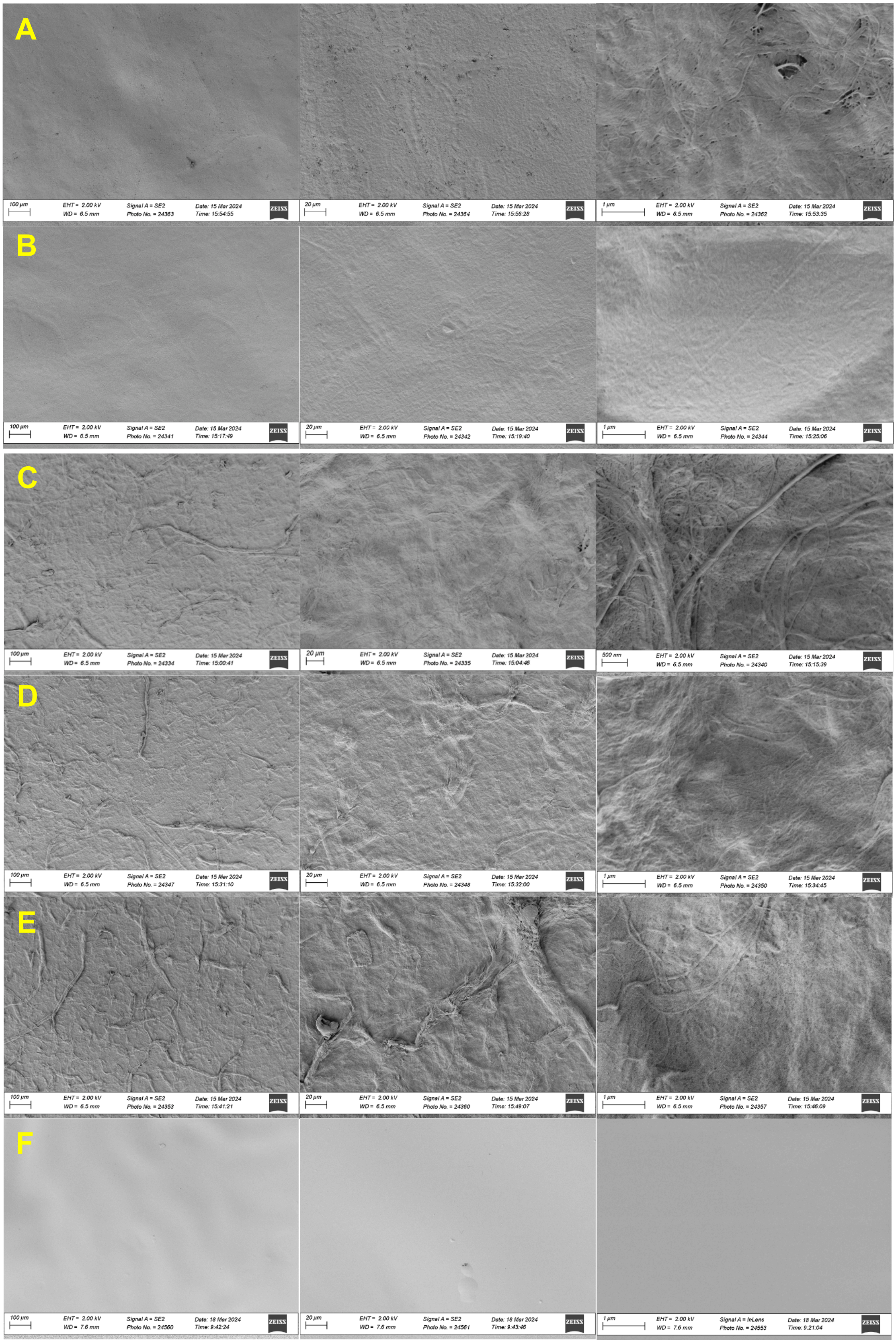

Fig. 2. SEM images of MFC/CMC casting films made with varied MFC/CMC suspension concentration (%) ratios. (A) MFC = 0.60 (bottom surface); (B) MFC/CMC =0.50:0.10 (bottom surface); (C) MFC/CMC =0.55:0.05 (top surface); (D) MFC/CMC =0.45:0.15 (top surface); (E) MFC/CMC =0.40:0.20 (top surface); (F) CMC =0.60 (top surface).

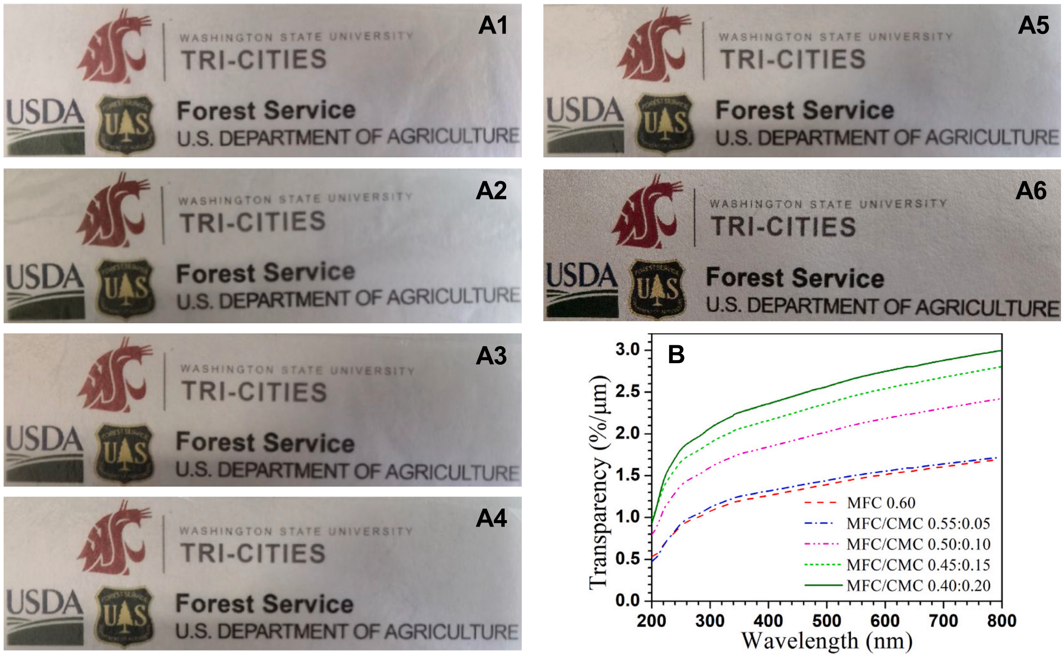

Fig. 3. Transparency of MFC/CMC casting films made with varied MFC/CMC suspension concentration (%) ratios. Visual observation of logos covered by different films: (A1) MFC = 0.60; (A2) MFC/CMC =0.55:0.05; (A3) MFC/CMC =0.50:0.10; (A4) MFC/CMC =0.45:0.15; (A5) MFC/CMC =0.40:0.20; (A6) No film. (B): Wavelength dependent film optical transparencies.

MFC 0.60	MFC/CMC 0.55:0.05	MFC/CMC 0.50:0.10	MFC/CMC 0.45:0.15	MFC/CMC 0.40:0.20
74.2±0.2°	78.8±0.1°	82.7±2.2°	85.9±2.0°	87.0±3.0°

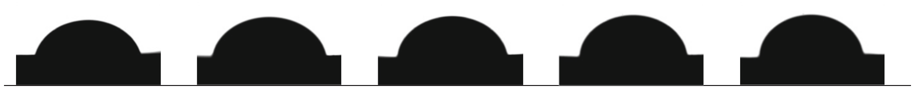

Fig. 4. Water contact angles and droplet images of MFC/CMC suspensions on banana surface.

slowed down the banana peel browning process, i.e., the bananas started to brown at day 6. This represents a 50 % improvement in shelf-life even with only 0.05 % CMC substitution compared with the pure MFC coating. This result is important to grocers as banana peel color is the most important measurer consumers use in making purchasing decision (Symmank et al., 2018).

The time-dependent browning fraction data shown in Fig. 6A were fitted to a logistic model (Eq. 1) with correlation coefficients (R^2) close to 0.99 (Table S3). In order to obtain a better understanding of MFC/CMC barrier coating formulation on banana browning, we plotted the starting time when rapid banana browning taking place *EC20* as well as the time at which banana browning rate was maximal against CMC loading as a fraction of total carbohydrate (MFC + CMC) loading as shown in Fig. 6B. The results clearly show that these two parameters were increased with increasing CMC loading in coating.

The effect of MFC/CMC barrier coating in preventing banana peel browning can be visually observed directly from the photographic images taken. As shown in Fig. 7, all bananas started with almost identical visual color at day 0. By day 4, the control and pure MFC-coated bananas showed brown spots/patches. In comparison, similar browning state of bananas coated with MFC/CMC formulations were observed in Day 6. This visually shows that CMC addition delayed the beginning of browning by 2 days. For further comparison, the browning state of bananas at day 7 and 9 were also shown in Fig. 7. At day 7, the appearance of bananas coated with MFC/CMC are still acceptable for sale, but obviously not salable for the bananas without coating or coated with pure MFC. Increasing CMC loading further improved the inhibition of banana browning as shown by the images of the bananas taken on day 9.

This is because CMC is a natural antioxidant that can inhibit enzymatic oxidation reactions of phenols in banana peel to slow down color change (Kim et al., 2022). In addition, CMC decreased water vapor permeation through the banana peel due to its moisture retention properties in real environment as will be discussed later.

3.7. Banana weight loss

The weight losses of bananas over time during storage were presented in Fig. 8A. Banana weight loss is linearly proportional to storage time for all bananas tested, in agreement with our earlier study (Geng et al., 2024). Comparing with the control banana, coating slowed down banana weight loss for all coating formulations as shown. The substitution of MFC with CMC further decreased banana weight loss. For example, weight loss was approximately 18 % for the control at day 6. Pure MFC coating decreased weight loss to approximately 15 %. With only 0.05 % CMC substituting MFC, weight loss was deceased to 12 % at day 6, or decreased by one third compared with the control without coating. The benefit of CMC substitution diminished at higher CMC loadings as shown. For example, increasing CMC substitution from 0.1 % to 0.2 % only resulted in marginal decrease in weight loss. This can be seen from the slope of the weight loss over time (Fig. 8A).

Banana weight loss is primary contributed by water loss due to water vapor permeation through banana peel. To explore the relationship between the banana weight loss and water vapor permeation, we plotted the slopes of time-dependent banana weight losses against WVTR of the casting-made MFC/CMC films of 10 g/m^2 with different CMC loadings. The results show inverse correlation between the slopes of banana

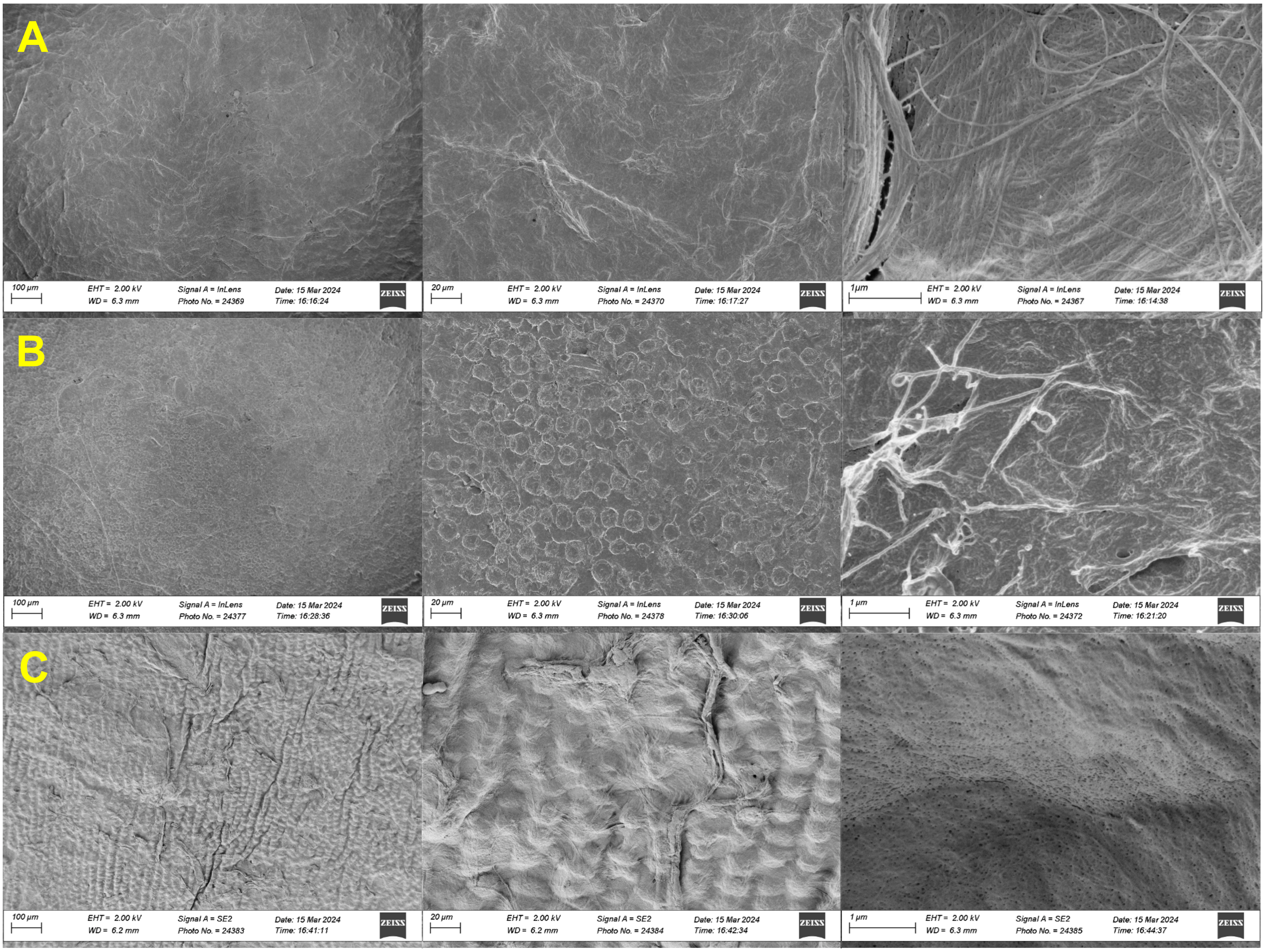

Fig. 5. SEM images of MFC/CMC barrier coating films peeled from banana surface. (A): MFC = 0.60; (B) MFC/CMC = 0.55:0.5; (C): MFC/CMC = 0.50:0.10.

weight losses over time and WVTR of the MFC/CMC casting films (Fig. S6), contradictory to intuition. Perhaps, banana weight losses are also contributed by other factors, such as the production and permeation (loss) of ethylene and CO_2. Moreover, the WVTR of the casting films were measured under standard method required testing conditions which are certainly different from those during storing coated bananas. The water-retaining property of CMC (Ramakrishnan & Kulandhaivelu, 2021) can play a role in reducing actual water permeation through banana peels by trapping moisture in ambient onto the coating films on banana peel surface to result in low WVTR values through banana peel in storage. This suggests that permeation study using standard casting films under standard test conditions may not be suitable to explain respiration of fruits with film barrier coating.

3.8. Banana firmness

The banana firmness measured in the bottom (stem end, Fig. S4), middle, and top sections are averaged and shown in Fig. 8B. The pure MFC coating appears to slightly delayed banana softening. MFC/CMC (0.50:0.10) and MFC/CMC (0.40:0.20) coatings accelerated banana softening, perhaps due to the anaerobic reactions under oxygen deficient condition caused by the low porosity (high density Table S2) of these two coatings as will be discussion in next section. A dense coating film packaging was found to reduce the firmness of tomatoes (Tas et al., 2017). As for the banana coated with the lowest CMC loading MFC/CMC (0.55:0.05), the banana firmness was approximately the same as the control banana.

3.9. Banana fruit total soluble solids (TSS)

TSS of bananas over storage time are shown in Fig. 8C. Comparing with the control banana, four coatings all decreased the TSS in bananas with pure MFC coating reduced the least (except day 15 with a large error bar) and the MFC/CMC (0.50:0.10) the most. TSS was mainly produced from bioconversion of starch that is up to 70 % to 80 % in green bananas on a dry weight basis (Hill, & ap Rees, T., 1993; Marriott et al., 1981; Zhang et al., 2005). It was found that low oxygen level in bananas could inhibit the starch breakdown into monosaccharides (Hill, & ap Rees, T., 1993). Because the four coating films have different porosities (Fig. 5), their abilities to block oxygen are different to result in varied starch saccharification levels and therefore TSS. Among them, the pure MFC coating delayed the conversion of starch to monosaccharides without affecting the final amount TSS in banana (except day 15 with a large error bar). As for the banana coated with MFC/CMC (0.55:0.05) film, its TSS at day 6 and day 9 were approximately 17.9 %, comparable to the TSS of the control banana of 18.0 % and 18.2 % at day 3, and 6, respectively. It should be mentioned that barrier coating with MFC/CMC (0.55:0.05) delayed banana browning by 2 days. In other words, the comparison of TSS should be made between the day 4 control banana and the day 6 banana coated with MFC/CMC (0.55:0.05) as these bananas have the same browning state. Under such a comparison, the TSS between these bananas were approximately the same. This suggests that

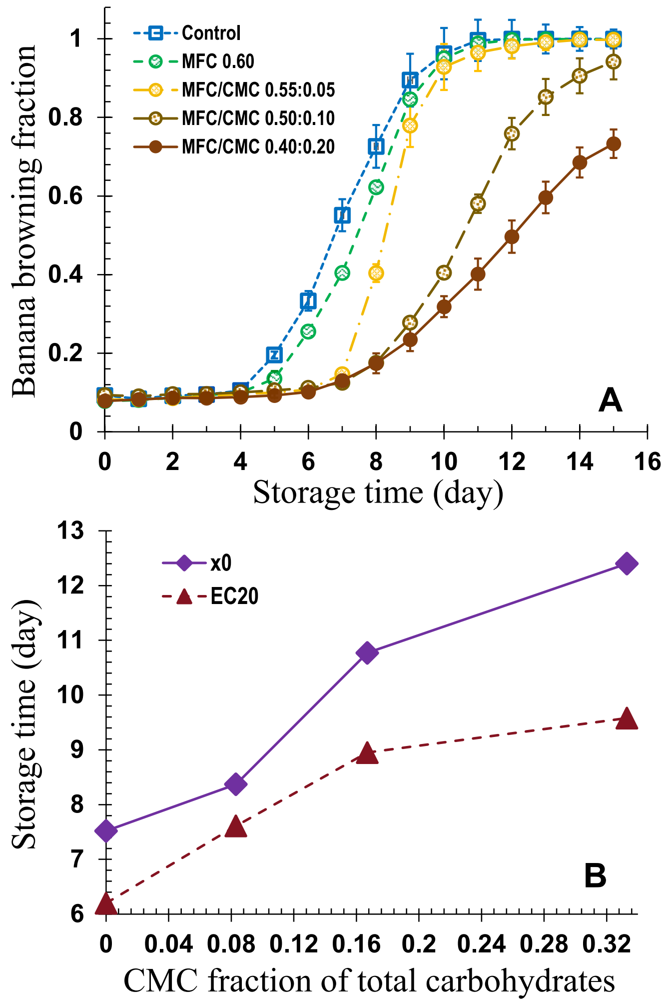

Fig. 6. (A): Time-dependent browning fraction of bananas with and without MFC/CMC barrier coating. (B): Correlation of CMC loading with *x*0 (storage time corresponding to the maximum browning slope) and *EC20* (the start time of rapid browning process).

barrier coating with MFC/CMC (0.55:0.05) simply extended banana shelf life by 2 days without affecting banana ripening and consumer preference in terms of TSS and peel appearance.

It is interesting to notice in Fig. 8C that the banana coated using MFC/CMC (0.40:0.20) with the greatest CMC loading did not result in a lower TSS than that from the banana coated with MFC/CMC (0.50:0.10). Barrier coating using MFC/CMC (0.40:0.20) resulted in the least porous film as reflected by the densities of the casting films (Table S2). Therefore, this film should permeate the least amount of oxygen and inhibit starch saccharification the most (Hill, & ap Rees, T., 1993) to result in the lowest TSS, contradictory to the results shown in Fig. 8C. It is understood that the porosity of a MFC/CMC barrier coating film also affects banana respiration such as oxygen intake, which can disrupt banana ripening and degradation process for complete energy release. Banana fruits respiration during storage can be categorized into aerobic (with oxygen) and anaerobic (no-oxygen) respirations, resulting in complete energy release or normal ripening and incomplete energy release or abnormal ripening, respectively. The biochemical reactions of monosaccharides (represented by glucose here) associated with these two respirations can be described (Duca, 2015):

$$C_6H_{12}O_6 + 6O_2 \rightarrow 6CO_2 + 6H_2O + 2867\ kJ/mol \quad \text{(aerobic)} \quad (3)$$

$$C_6H_{12}O_6 \rightarrow 2C_2H_5OH + 2CO_2 + 200\ kJ/mol \quad \text{(anaerobic)} \quad (4)$$

These equations suggest that aerobic reaction mainly produces CO_2 and water with greater heat (energy) release while anaerobic reaction results in alcohol and smaller amounts of CO_2 and heat release than aerobic reactions. For those coating films with good porosities such as the pure MFC (0.6 %) coating and the coating with a low amount of CMC (MFC/CMC = 0.55 %:0.05 %), oxygen can be easily permeated, so bananas went through aerobic reactions and the resultant CO_2 and water vapor can be permeated out of coating films. Nothing extraordinary was noticed during peeling bananas in experiments, in agreement with experimental observations, i.e., the denser the film, the lower the TSS.

The coating with MFC/CMC (0.40:0.20) and MFC/CMC (0.50:0.10) both with a higher CMC loading resulted in much less porous films with lower or minimal oxygen permeation, which facilitated anaerobic reactions to produce ethanol, in addition to CO_2. The decreased permeation of CO_2 and water vapor out of the bananas with barrier coating, consistent with the decreased weight loss reported in Fig. 8A, resulted in a gas pressure build-up inside bananas as visually observed in our experiments (slightly swollen bananas after day 4 in Fig. 7). The accumulation of water vapor resulted in the formation of water-soaked tissue (Imahori et al., 2005; Wendakoon et al., 2006a; Wendakoon et al., 2006b). It was observed that a fair amount of water came out of the bananas coated with MFC/CMC (0.50:0.10) along with a large amount of gas and some alcohol smell during peeling these bananas starting day 3. The gas and the water were from the trapped CO_2 and water-soaked tissues due primarily to the aerobic reactions, notice that one mole of glucose can produce 6 mol of CO_2 and 6 mol of water in Eq. (3), while the alcohol was from the anaerobic reactions. The release of gas and alcohol smell was also observed during peeling bananas coated with the greatest amount of CMC, MFC/CMC (0.40:0.20), starting day 3. The gas and the alcohol were both primarily from the anaerobic reactions (Eq. (4)) and trapped inside of bananas coated with the least porous film using the greatest amount of CMC. Comparing with the bananas coated with MFC/CMC(0.50:0.10), the lower O_2 and higher CO_2 inside the bananas coated with MFC/CMC (0.40:0.20) slowed down normal ripening due to decreasing the respiration of bananas (Heydari et al., 2010; Ullah et al., 2009), in addition to the stronger inhibition of starch saccharification due to the lower amount of O_2, to result in a lower TSS. This is also consistent with the slightly greater firmness of the bananas coated with MFC/CMC (0.40:0.20) than the firmness of the bananas coated with MFC/CMC (0.50:0.10) as shown in Fig. 8B.

4. Conclusions

Barrier coating of bananas using MFC suspension with CMC substitution decreased the porosity of the coating films to result in decreased banana weight loss, slowed banana softening and peel browning, therefore extending banana shelf life. Specifically, banana color browning can be delayed by 2 days and weight loss decreased by 1/3 with an only small amount of CMC substitution (0.05 % CMC out of 0.6 % MFC). The delayed peel browning and decreased weight loss and total soluble solids can be attributed to decreased permeation of gasses or banana respiration through banana peel surface. The delayed browning is also due to the antioxidant properties of CMC. It is important to notice, banana ripening process was not slowed with respect to banana browning measured by total soluble solids (TSS), in other words, MFC/CMC (0.55:0.05) coating delayed peel browning by 2 days, it simultaneously decreased TSS formation by approximately 2 days, favorable to grocers and consumers. A high CMC loading in barrier coating formulation facilitated anaerobic biochemistry reactions in banana ripening process to result in the production of ethanol from the saccharified starch in banana. This study provides a pure polysaccharide coating formulation for effective preventing banana weight loss, browning, softening, without affecting ripening to meet consumer needs and preferences. Future work on the subject should focus on large scale trial runs and techno-economic and life cycle analyses for moving the technology to commercialization.

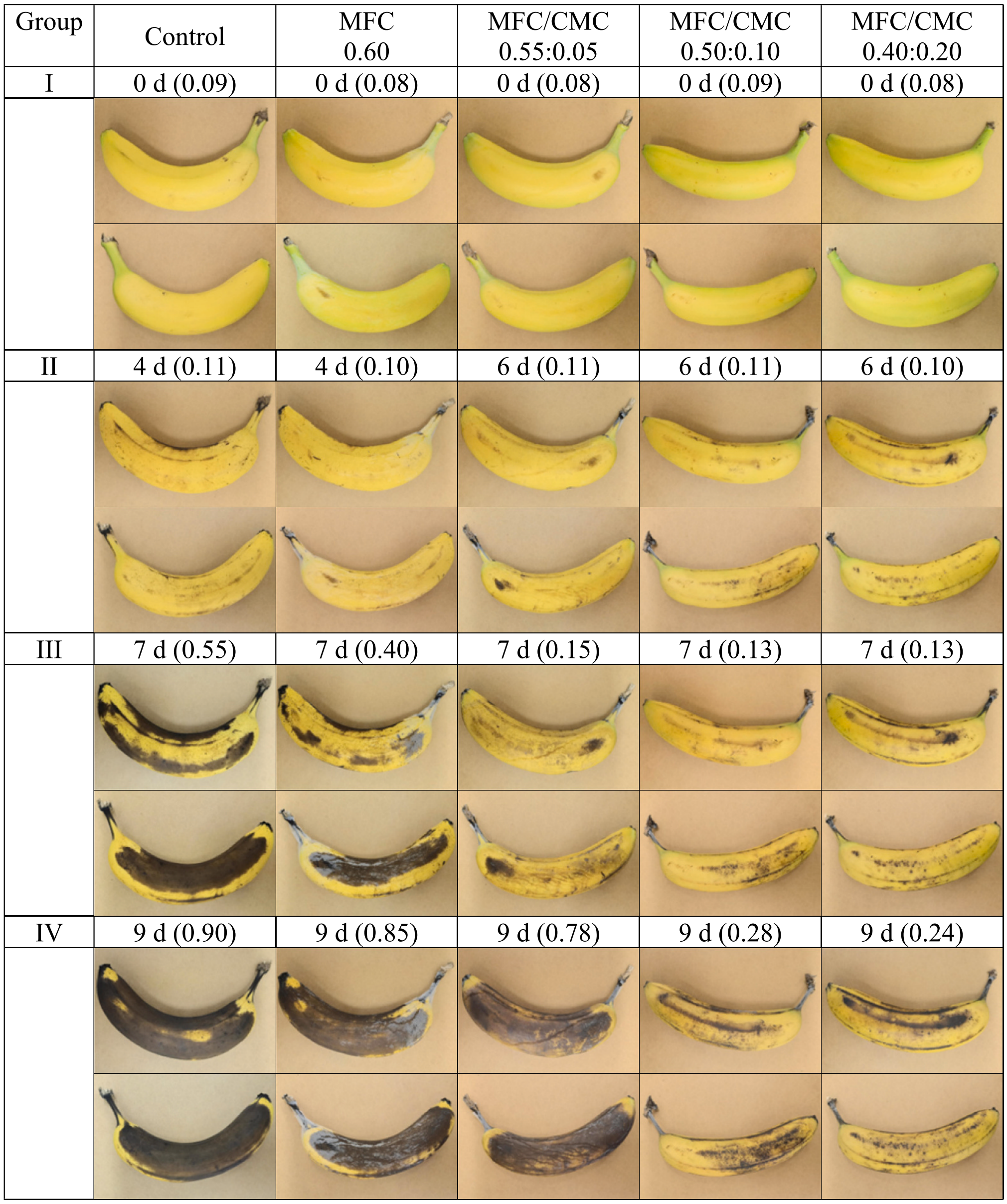

Fig. 7. The two opposite-side images of each banana with and without MFC/CMC barrier coating during storage. The numbers in the parentheses are the browning fractions based on image color pixels.

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CRediT authorship contribution statement

Jing Geng: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Nicole Stark:** Writing – review & editing, Supervision, Resources, Methodology. **Peter Kitin:** Writing – review & editing, Visualization, Methodology, Investigation, Formal analysis, Data curation. **Xiao Zhang:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition. **Nayomi Z. Plaza:** Data curation, Methodology, Visualization, Writing – review & editing. **J.Y. Zhu:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Funding acquisition, Formal analysis, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Authors Geng, Stark, Zhu and Zhang are co-inventors of a provisional U. S. patent application.

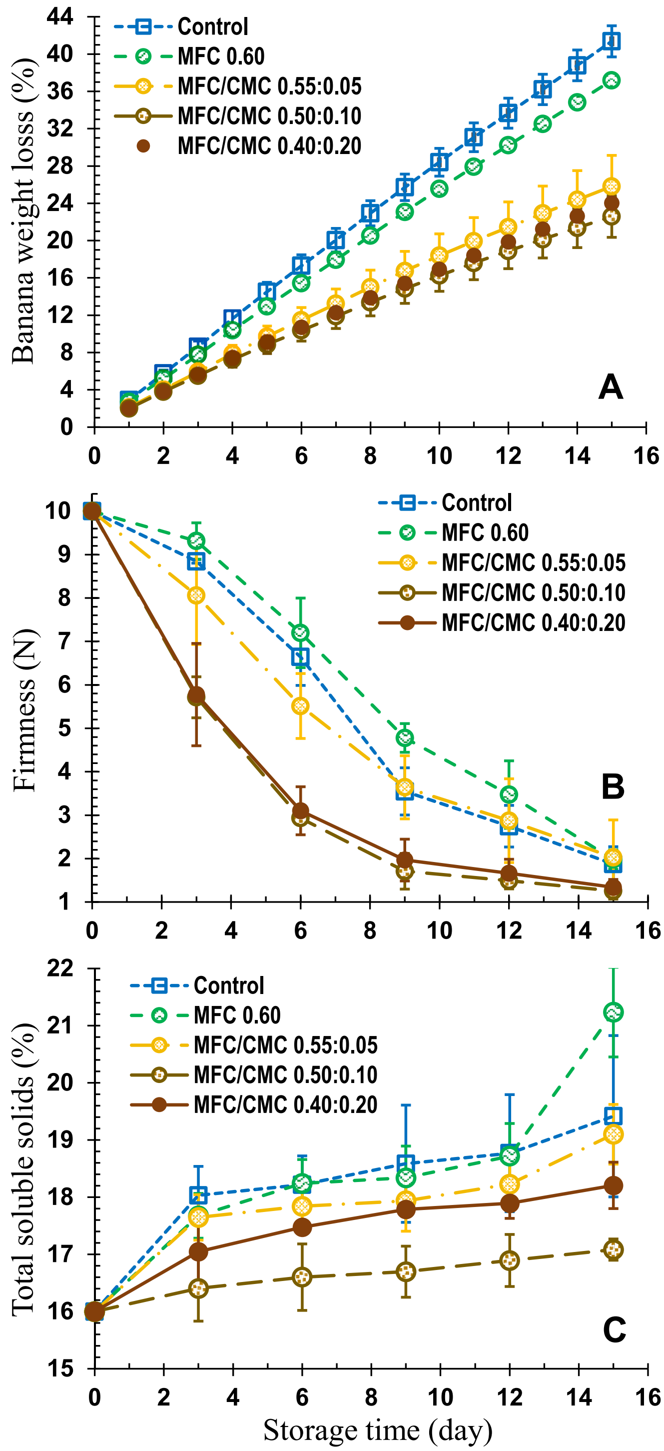

Fig. 8. (A): Time dependent weight loss of bananas with and without MFC/CMC barrier coating; (B): effects of different MFC/CMC coatings on time-dependent firmness of banana fruits; (C): time-dependent total soluble solids (TSS) of bananas with and without MFC/CMC coating.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodchem.2025.144664.

Data availability

Data will be made available on request.

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