



Barrier coating of bananas using plant cellulose and edible lemon peel fibrils to extend shelf-life

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ABSTRACT

Barrier coating of fruits can decrease postharvest loss and improve food security. This study demonstrated the performance of barrier coating using pure plant-based carbohydrate polymers to improve the shelf life of bananas. The coating formulation was made of only two carbohydrate polymers: microfibrillated cellulose (MFC) from commercial bleached hardwood pulp fibers and lemon peel (LP) fibrils from milling commercial dry lemon peels. Bananas were coated by dipping into different MFC/LP suspensions with varied LP loadings. The measured banana peel color browning, fruit weight loss, firmness, and total soluble solids were used to compare coating performance with the control (without coating). The substitution of MFC with LP fibrils decreased ethylene production and the respiration of oxygen and carbon dioxide, due to the improved adhesion of the coating film on banana peels. Compared with control (no coating), substituting MFC with LP delayed banana peel browning by 2 days, decreased weight loss by up to 20 % and browning fraction by up to 33 %, compared with 1 day, 12 %, and 7 %, respectively, using pure MFC coating. Considering typical banana shelf life in supermarket is approximately 7 days, a 2-day shelf-life extension is significant.

1. Introduction

Bananas are rich in dietary fibers, vitamins, minerals, phenolic compounds, and a favorite among consumers due to their affordability and high nutritional value. According to the Food and Agriculture Organization of the United Nations, global import and export values and quantities rank at the top among fresh fruits, reaching USD 29.0 billion and 43.7 million tons in 2023 [62]. It is well-known that bananas have a very short shelf life. Due to their climacteric nature, bananas exhibit a surge in ethylene production during ripening. In addition, their physicochemical properties and flavor also undergo rapid changes during this period [1]. Peel color changes quickly from green to yellow, accompanied by the softening of fruit pulp with a rise in sweetness, and finally to brown in the form of dark spots spreading progressively. As a result, postharvest waste is very significant between 25 and 50 % [2,3]. The visual appearance of bananas without browning, sweet taste, and proper softness are important factors consumers use in making purchasing decisions. Therefore, decreasing banana peel browning without

significantly slowing down the banana ripening meets consumer purchasing preference, resulting in decreasing waste. This in turn improves the economic bottom line for growers and grocers.

Much research indicated that barrier coatings are very effective in extending the shelf life of bananas [4–7]. Cellulose, the most abundant natural carbohydrate polymer on earth, has been shown to have excellent gas barrier and film-forming properties as a result of hydrogen bonding, and can be used to prolong the shelf life of fruits via coating [8,9]. Cellulose nanofibrils (CNF) have been proven capable of forming an effective barrier film on bananas peels to extend shelf life [4,61]. Unfortunately, cellulose nanomaterials (CNM) such as CNF are not only cost-ineffective but also has health and environmental concerns through inhalation due to the potential release of air-born cellulosic nanoparticles [10]. To address these two issues, our earlier study demonstrated the effectiveness of pure microfibrillated cellulose (MFC), also called cellulosic microfibrils (CMF), for the barrier coating of bananas [7]. MFC, a much coarser material than CNF decreases production costs and eliminates issues associated with inhalation safety and

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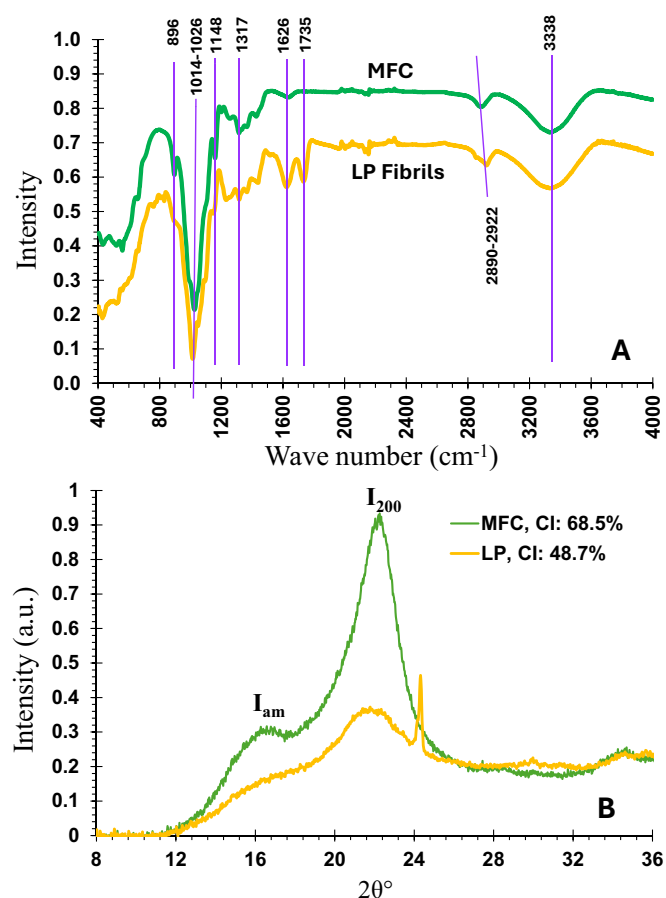


Fig. 1. FTIR (A) and XRD (B) spectra of MFC and LP fibrils. CI is the Segal crystallinity index.

environmental and human health regulations. However, as a coarser material, the coating film formed by pure MFC is not sufficiently dense, inadequate for best performance in banana preservation. Thus, MFC barrier coating formulation needs improvement for commercial banana preservation.

Citrus fruits, as one of the most popular and widely cultivated fruits, have an annual global production of 169.4 million tons in 2023 according to the Food and Agriculture Organization of the United Nations [62]. Nearly half of citrus fruit crops are used for industrial production, which results in a significant amount of biowaste particularly from the peels that accounts for 7–40 % of the fresh fruit weight [11,12]. Citrus fruit peels can be a valuable bioresource, containing primarily carbohydrate polymers such as pectin and fibers, phenolic compounds, essential oils [13,14]. The pectin in citrus peels can serve as a thickener to improve the processability of the coating solution and the adhesion properties of the resultant coating film. The hemicelluloses in citrus peels also facilitate crosslinking, especially when adding citrus peels into MFC to form a dense film to improve barrier performance and optical properties while weakening the film's waterproof and mechanical performance [15], which is advantageous for water washing of coated fruits. Citrus extracts and essential oils are rich in bioactive components that could enhance the coating film's antioxidant and antimicrobial properties [16–18], especially for applications in edible barrier coatings of nano formulation with nanoparticles, nano emulsions, or nanofibers [19]. Because of these benefits [15], the pectin and cellulose derived from citrus fruit peels have been used as additives in barrier coating [20,21], which requires, however, extra processing to extract pectin and cellulose from citrus peels. Using whole citrus peels in barrier coating can eliminate this extra processing step and associated costs. Studies

have been carried out mixing powders from whole citrus peels in various coating formulations [22] with gelatin [23], PVA [24], starch [25], and polyethylene [26] as the main constituent material. Most formulations use synthetic or petroleum-derived chemicals such as Tween 80, PVA, polyethylene, or expensive plant or animal-derived components such as glycerol, xanthan gum, and gelatin. Furthermore, commercial powders of citrus peels are sold as micro-particles with very low aspect ratios close to unity which results in brittle films with poor film-forming performance.

In this study, we incorporated whole lemon peels into MFC fruit barrier coating formulation. Our recent study found that substituting a portion of MFC with carboxymethyl cellulose (CMC), a cellulose derivative food additive, improved the effectiveness of MFC for banana barrier coating [27], especially in decreasing banana peel browning. These improvements are due to (1) CMC with its small physical dimension can fill the voids in the MFC network through crosslinking to result in a dense film that decreases gas permeation, and (2) CMC has natural antioxidant property [28,29]. We hypothesized that lemon peels can play similar roles as CMC because it can be mechanically processed into smaller physical dimensions with the short chain length of pectin and hemicelluloses, and lemon peels also have natural antioxidant and antimicrobial properties derived from its flavonoids, polyphenols, extractives and essential oils [16–18]. The novelties of this study are: (1) Using only renewable and abundant plant-based carbohydrate polymeric materials, i.e., MFC and edible lemon peel fibrils, for fruit barrier coating formulation without synthetic or petroleum-derived materials; (2) Novel processing of lemon peels to avoid pulverizing into powders to obtain lemon peel microfibrils with great aspect ratios to enhance film-forming performance; (3) Achieving significant reduction in banana peel browning without substantially slowing down banana ripening to meet consumer purchase preference and improve shelf life. This study aims to develop an effective coating formulation using only plant-based natural carbohydrate polymers to decrease food waste, improve food security, and improve edible whole citrus fruit peel utilization without additional processing through extraction and/or separation.

2. Materials and methods

2.1. Materials

The virgin bleached kraft hardwood pulp (BKHP) and lemon peels (LP, Fig. S1) were provided by WestRock Company (Atlanta, GA, USA) and Fiberstar, Inc. (River Falls, WI, USA), respectively. Fresh Chiquita bananas were purchased from a local supermarket in Madison, Wisconsin. Bananas were yellowish green without noticeable visible scars and were of similar maturity at the time of purchase. To minimize the effects of the variations in the initial ripen stage of the bananas on the coating performance, each coating parametric variation study used bananas from the same hand. Inhouse water purified by reverse osmosis was used throughout the experiments.

2.2. Characterization of the kraft bleached hardwood pulp fibers and lemon peels

The chemical composition of the as received BKHP fibers and the LP were analyzed using a procedure developed by the National Renewable Energy Laboratory (NREL) [30]. The BKHP and LP were first hydrolyzed using two-step acid hydrolysis. Klason lignin was determined gravimetrically. The soluble lignin was measured using the absorbance at 205 nm following TAPPI Useful Method UM-250 [63]. The chromatographic procedure [31] was used to resolve the wood sugars. Filtered hydrolysates were then diluted 23-fold and injected into an ion chromatography system (Dionex ICS 6000, Thermo-Fisher Scientific) with a pulsed amperometric detector and a CarboPac PA1 (4 × 250 mm) column. Quantifications of monomeric sugars, galacturonic, and glucuronic acids in LP were achieved by producing standard curves from pure

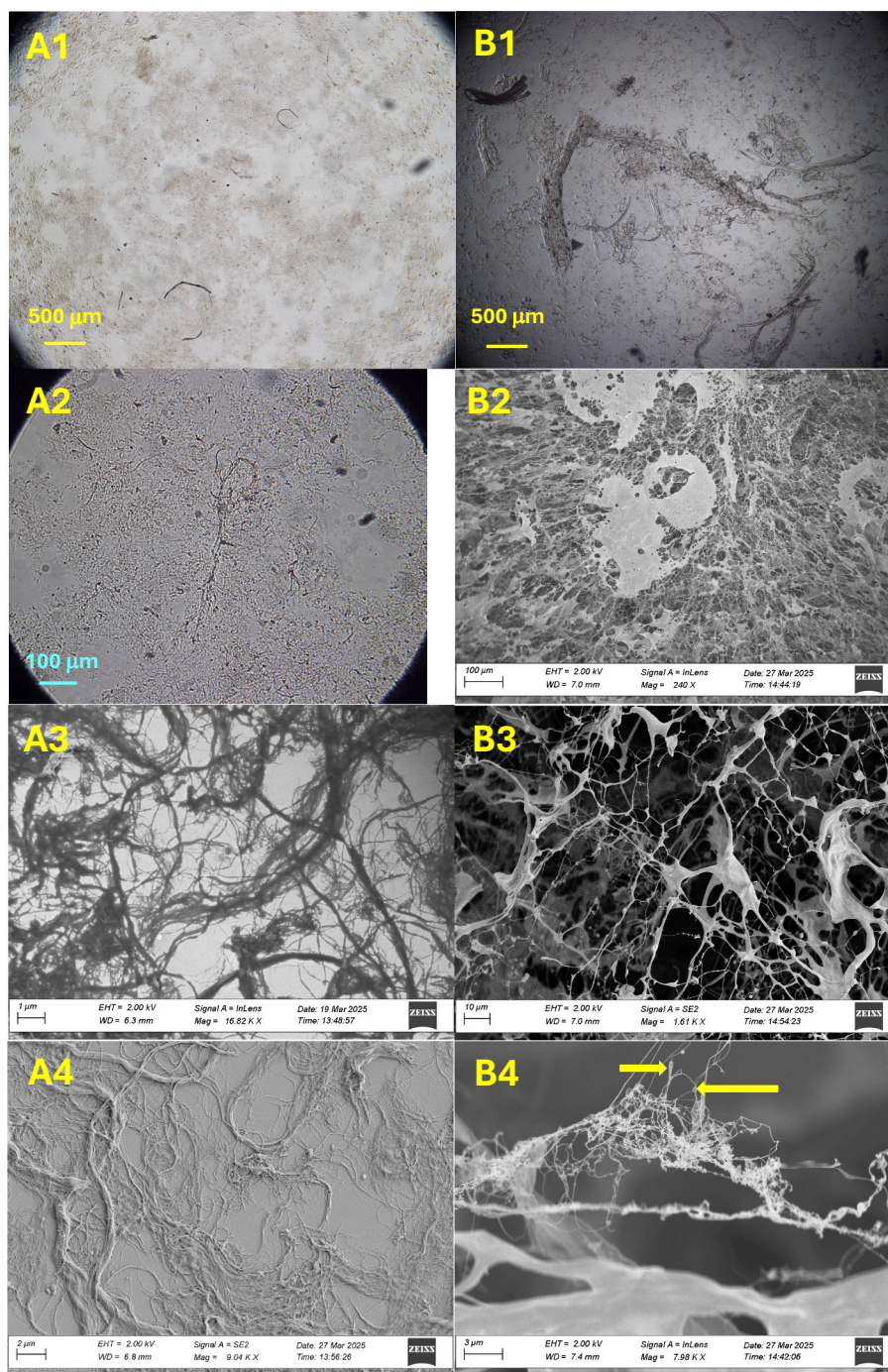


Fig. 2. Light and scanning electron microscopy (LM and SEM) images of microfibrillated cellulose (MFC) and milled lemon peel (LP) fibrils. A1-A4: MFC; B1-B4: LP fibrils. Before imaging, all SEM samples were freeze-dried on carbon tape except for A4 which was vacuum-dried on a Si wafer. Arrows in B4 indicate nano-size agglomerated materials containing water-soluble pectin.

sources of all the desired analytes. The results are presented in Tables S1 and S2.

The BKHP and LP were further characterized using Fourier transform infrared spectroscopy (FTIR) and X-ray diffraction (XRD) analyses. The oven-dried BKHP and freeze-dried LP were ground and passed through a 60-mesh sieve to obtain BKHP and LP powders. FTIR analysis was performed on a spectrometer (Spectrum two, PerkinElmer) in the wavenumber range from 4000 to 400 cm^{-1} . For XRD analysis, BKHP and LP powders were placed inside the 25 mm cavity of a zero-diffraction powder holder made of Si (MTIxl, Richmond, CA). Samples were measured using a Bruker D8 Discovery X-ray diffractometer with Cu—K

α radiation, and a 0.5 mm aperture at the Wisconsin Center for Nano-scale Technology. Data were collected for 120 s using a Bragg-Brentano configuration at the incoming beam angle of 12° . The Segal crystallinity index (CI) was calculated using the 200 diffraction peak intensity ($I_{200} \sim 22.3^\circ$) after subtracting the amorphous intensity ($I_{am} = 18^\circ$) according to Eq. (1).

$$CI (\%) = \frac{I_{200} - I_{am}}{I_{200}} \times 100 \quad (1)$$

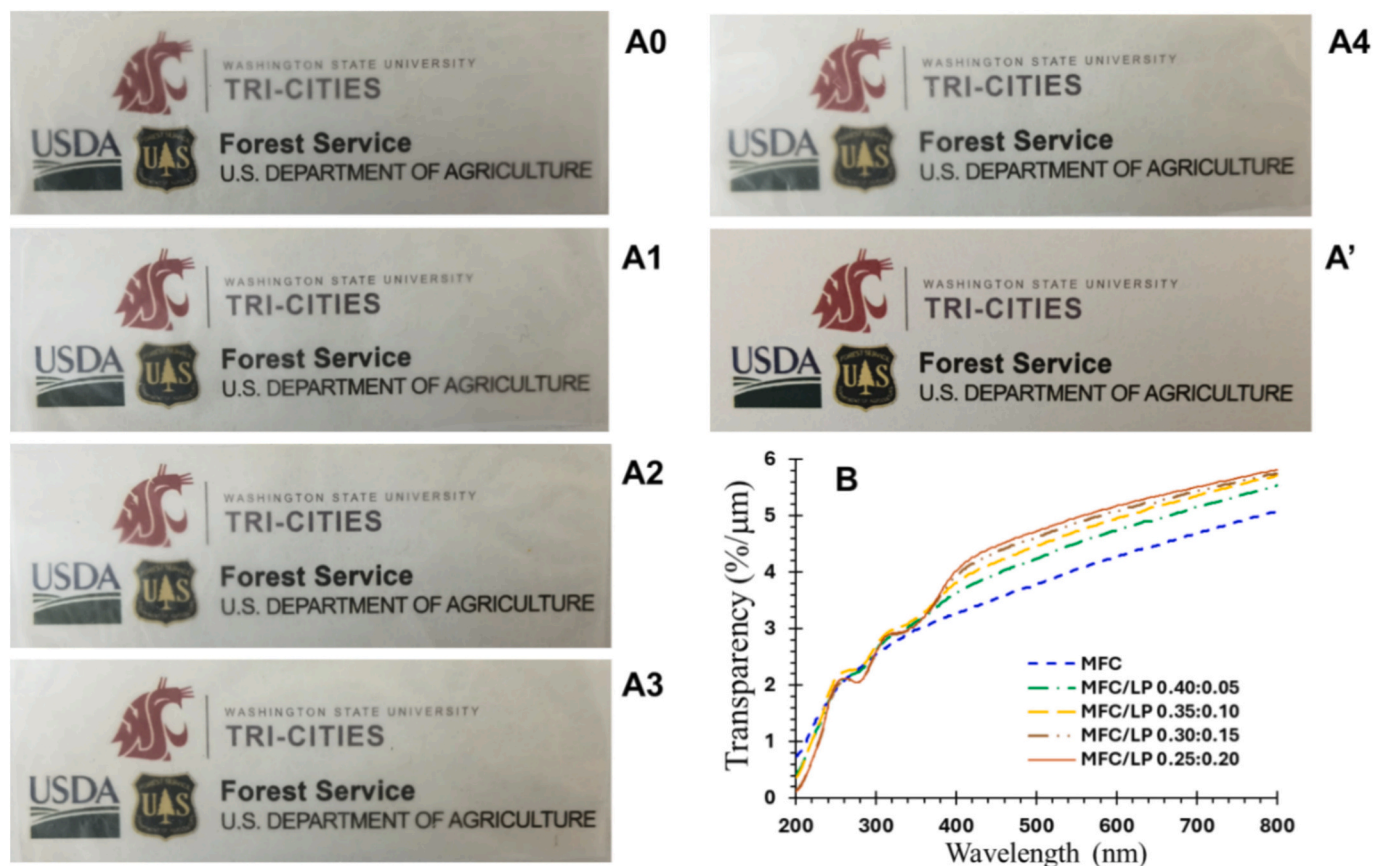


Fig. 3. Transparency of MFC/LP casting films made using varied MFC/LP suspension concentration (%) ratios. Visual observation of logos covered by different films: (A0) MFC = 0.45; (A1) MFC/LP = 0.40:0.05; (A2) MFC/LP = 0.35:0.10; (A3) MFC/LP = 0.30:0.15; (A4) MFC/LP = 0.25:0.20; (A') No film cover. (B): Wavelength-dependent film optical transparencies.

2.3. Preparation of microfibrillated cellulose/lemon peel (MFC/LP) suspensions

Wet BKHP was dispersed in water, then disintegrated in a disintegrator (Model 73-06-01, TMI, Ronkonkoma, New York, USA) for 10,000 revolutions. Subsequently, the pulp suspension was microfibrillated using a stone disk mill – SuperMassColloider (SMC) (Model MKZCA 6-2 J, Disk MDGA 6–80#, Masuko Sangyo Co., Ltd., Japan). BKHP suspension at 2 wt% solids prepared using 60 g OD (oven dry weight) BKHP was directly fed into the SMC, as described previously [7]. The 2 wt% solids loading was based on an optimization study on MFC production using the same SMC reported earlier [32]. The MFC was obtained after four passes through the SMC at 1500 rpm.

Similarly, 40 g of dried lemon peels were soaked in water and disintegrated for 10,000 revolutions, and then microfibrillated in the SMC at 2 wt% solids but only through one pass. LP fibers are primarily formed by the thin primary cell wall which is less rigid compared with wood fibers such as BKHP reported in this study which are formed mainly by thick and rigid secondary walls to withstand the compressive load of trees. Therefore, fibrillation of LP needs much less energy than that to fibrillate BKHP as listed in Tables S3 and S4.

MFC/LP coating formulations with the same mass concentration of 0.45 wt% were prepared by mixing the suspensions of MFC and LP fibrils. The concentration ratios of MFC (wt%) over LP (wt%) were 0.45:0.00, 0.40:0.05, 0.35:0.10, 0.30:0.15, and 0.25:0.20, or LP substitutions of 0, 11 %, 22 %, 33 %, and 44 %. These suspension samples were kept in a refrigerator until use.

2.4. Wettability of MFC/LP suspension on banana peel surface

The wettability of MFC/LP suspensions on banana peel surfaces was evaluated using an optical tensiometer (TL100, Theta Lite, Biolin Scientific Oy, Finland). The contact angle (θ) of MFC/LP suspensions on the banana peel surface was measured using the sessile drop method. The surface tension (γ_{LV}) of MFC/LP suspensions was measured via the pendant-drop method.

2.5. Preparation of MFC/LP casting films

Self-standing films of MFC/LP mixtures were made by casting. A desired volume of MFC/LP suspension was poured into a petri-dish with a diameter of 90 mm to result in a film grammage of 10 g/m². The mixture was evenly spread in the dish and dried at ambient conditions (22 ± 2 °C, 17 ± 2 % RH).

2.6. Characterization of MFC/LP films

The casting films were characterized for their optical transparency using a UV–vis spectrophotometer (Model 8453, Agilent Technologies, Palo Alto, California, USA). The hydrophilia of the films was also evaluated by the same optical tensiometer described earlier (TL100, Theta Lite, Biolin Scientific Oy, Finland). Water droplets falling on the MFC/LP casting film were photographed in real time for 60 s. The morphologies and microstructures of the MFC/LP casting films as well as the MFC/LP films peeled from the coated banana surfaces were examined by scanning electron microscopy (SEM) as described previously [27]. The morphology of MFC/LP micro- and nanofibrils were examined similar to an earlier study [33] with the following modifications. Droplets of 0.01

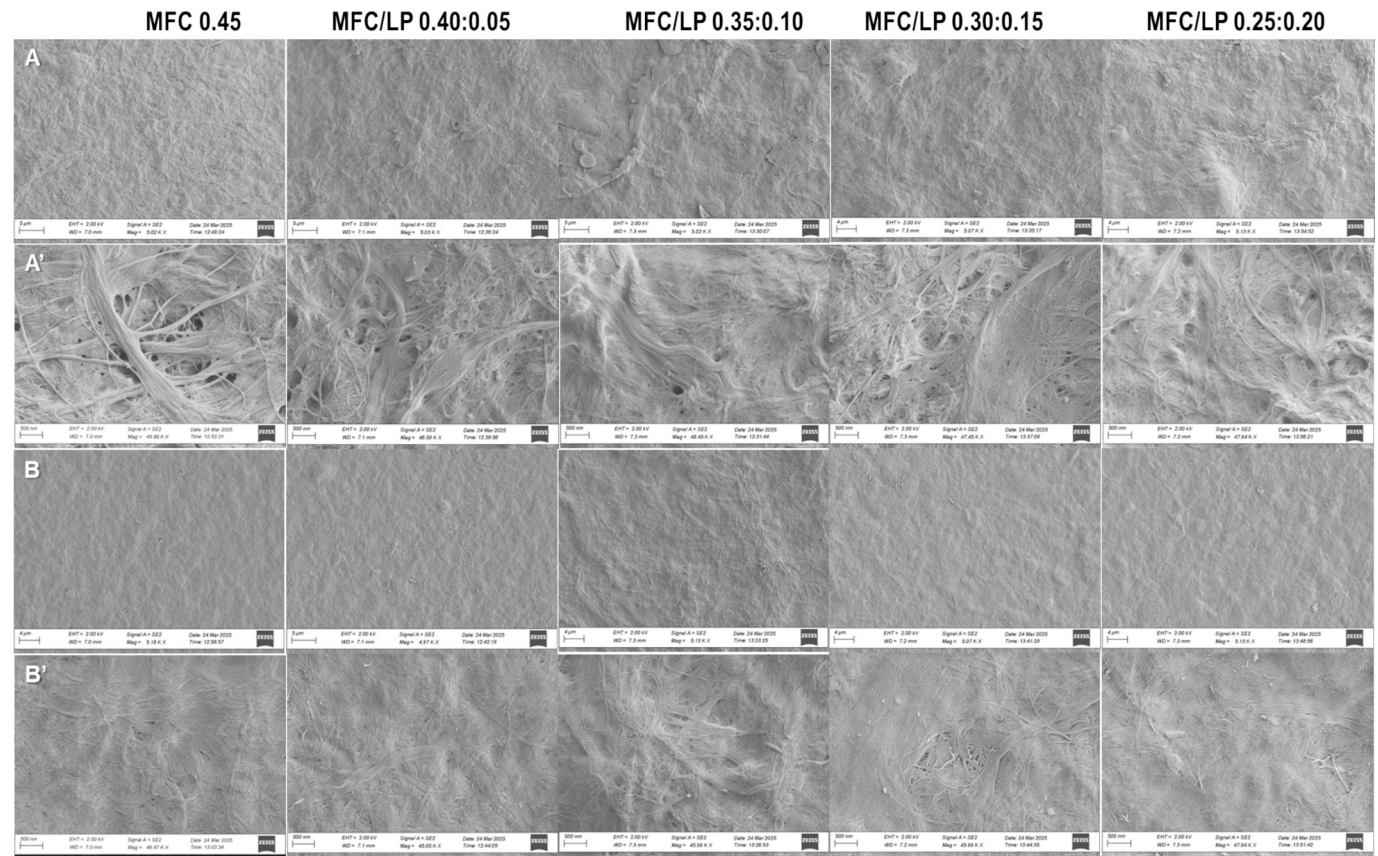


Fig. 4. SEM images of MFC/LP casting films made with varied MFC/LP suspension concentration (%) ratios. A and A': Top surface; B and B': Bottom surface.

Table 1
Thickness, density, grammage and WVP of MFC/LP films along with results from ANOVA and Tukey test ($\alpha = 0.05$).

	Group label	Grammage (g/m ²)	Thickness (μ m)	Density (g/cm ³)	WVTR (g/h/m ²)	WVP ($\times 10^{-11}$ g/m/s/Pa)
MFC 0.45	X1	10.02 $\pm$ 0.08	7.68 $\pm$ 0.77	1.32 $\pm$ 0.15	14.541 $\pm$ 0.579	2.50 $\pm$ 0.33
MFC/LP 0.40:0.05	X2	9.94 $\pm$ 0.08	8.32 $\pm$ 0.33	1.20 $\pm$ 0.04	14.491 $\pm$ 0.195	2.71 $\pm$ 0.10
MFC/LP 0.35:0.10	X3	9.94 $\pm$ 0.08	6.80 $\pm$ 0.57	1.47 $\pm$ 0.11	14.915 $\pm$ 0.321	2.30 $\pm$ 0.24
MFC/LP 0.30:0.15	X4	10.16 $\pm$ 0.00	8.16 $\pm$ 0.67	1.25 $\pm$ 0.10	15.284 $\pm$ 0.398	2.82 $\pm$ 0.17
MFC/LP 0.25:0.20	X5	9.94 $\pm$ 0.08	8.40 $\pm$ 0.85	1.29 $\pm$ 0.12	15.891 $\pm$ 0.593	2.87 $\pm$ 0.25
F value		8.50	4.99	5.35	6.85	4.40
p value		0.00035	0.00594	0.00429	0.00241	0.01500
Tukey critical difference		0.14	1.253	0.21	0.969	0.50
Paired groups with p < 0.05		X1-X4, X2-X4, X3-X4, X4-X5	X2-X3, X3-X4, X3-X5	X2-X3, X3-X4, X3-X5	X1-X5, X2-X5, X3-X5	X3-X4, X3-X5

wt% MFC/LP suspension were put on a carbon tape that was mounted on an aluminum stub, and the samples were lyophilized. Similarly, 2 μ L droplets of 0.001 wt% MFC/LP were placed on a Si wafer (100-oriented), then dried overnight in a vacuum oven at 50 $^{\circ}$ C and pressure 7 psi. Then the samples were sputter-coated with 5 nm Pt and observed at 2–3 kV accelerating voltage with a Zeiss Gemini 450 FE-SEM microscope (Carl Zeiss AG, Germany).

The water vapor permeation (WVP) through the MFC/LP casting films was measured according to ASTM E96/E96M-21 as described in our prior work [7].

2.7. Evaluation of the performance of MFC/LP barrier coating on bananas

Each banana was washed and wiped dry using a paper towel before coating, then completely submerged into a MFC/LP suspension for 3 min. The coated bananas were hung out to dry and stored for 15 days at ambient (22 $\pm$ 2 $^{\circ}$ C, 17 $\pm$ 2 % RH). The ambient condition was used to

better simulate the practical banana storage conditions in supermarkets.

The two opposite sides of each banana were photographed daily to study the changes in the banana's appearance and peel color.

The banana browning fraction was calculated according to a method we developed previously [27]. Briefly, the two photos of the opposite sides of each banana formed a group. Each image was imported into Python 3.8 to convert the image into an 8-bit color data set in RGB space, i.e., x(R), y(B), z(G). A cylindrical cone with its vertex at the white point (255, 255, 255), base radius of 70, and height being the line between the black (0, 0, 0) and white point (255, 255, 255) was used to threshold the image in the RGB space as either yellow (outside of the cone) or black (brown, inside the cone). Then the ratio of the numbers of black pixels over the total numbers of black and yellow pixels is calculated as the banana browning fraction.

To quantify the banana browning process, the banana browning fraction data were fitted to a logistic model showing below:

$$y = A_2 + (A_1 - A_2)/(1 + (x/x_0)^q) \tag{2}$$

Table 2

Water droplet images and contact angles on banana peel surface, and surface tension γ_{LV} of the MFC/LP suspensions along with results from ANOVA and Tukey test ($\alpha = 0.05$).

Coating formulation	Group label	CA (°)	γ_{LV} (mN/m)
MFC 0.45	X1	76.6 ± 1.6	46.3 ± 2.2
			
MFC/LP 0.40:0.05	X2	79.1 ± 0.9	40.0 ± 2.9
			
MFC/LP 0.35:0.10	X3	78.5 ± 1.2	36.0 ± 2.6
			
MFC/LP 0.30:0.15	X4	75.7 ± 1.8	47.0 ± 2.5
			
MFC/LP 0.25:0.20	X5	82.5 ± 1.0	48.3 ± 1.4
			
F value		16.8	37.7
p value		3.4206 ⁻⁶	2.6576 ⁻¹¹
Tukey critical difference		2.6	3.5
Paired groups with p < 0.05		X1-X5, X2-X4, X2-X5, X3-X4, X3-X5, X4-X5	X1-X2, X1-X3, X2-X3, X2-X4, X2-X5, X3-X4, X3-X5

$$EC_{10} = 10^{(\log(x_0) - \log(9)/q)} \quad (3)$$

where A_1 is the browning fraction of bananas on Day 0 (set as 0.10); A_2 is set at 1.00, which means that bananas are completely browned; q (day⁻¹) is the maximum slope of banana browning fraction over time; x_0 (day) is the storage time corresponding to the maximum slope (q), and EC_{10} (day) is the time when rapid banana browning takes place.

All testing bananas were weighed daily. Banana weight loss was determined from the banana's initial (day 0) weight and the measured daily weight W_t (t is the number of days stored) using Eq. (4).

$$\text{Weight loss (\%)} = (W_t - W_0)/W_0 * 100 \quad (4)$$

The firmness and total soluble solids (TSS) were measured every three days for all banana fruits. Bananas were peeled off to perform firmness test as described previously [27] using a FR-5105 high-accuracy fruit hardness tester (Ace Instruments, Lutron Electronic Enterprise Co., Ltd., Taiwan). An 11 mm penetrometer tip (FRTP-11) was used in all firmness measurements.

Five grams of banana fruit was used for TSS measurements. The fruit was ground thoroughly in 10 mL of distilled water. The pulp was centrifuged for 5 min at 10,000 rpm. The supernatant was measured to determine TSS using a digital handheld pocket refractometer (3810 PAL-1, Cole-Parmer, Atago Co., Ltd., Tokyo, Japan).

Banana respiration and ethylene release were measured daily. Before each measurement, each banana was weighed and placed in an 86 oz. (2.5 L) plastic container that was covered by plastic wrap, then sealed with a perforated lid and stored at ambient. The concentrations of ethylene (C_2H_4), oxygen (O_2), and carbon dioxide (CO_2) in the container were measured through the hole on the lid using a three-gas analyzer (F-950, Felix Instruments, Camas, WA, USA). The banana was taken out of the container after each measurement and placed back to the same position before the measurement under the same storage condition. Ambient concentrations of these three gases were also measured using

the same gas analyzer. Ethylene release, oxygen and carbon dioxide respiration in a given time period were calculated by subtracting the measured ambient C_2H_4 , O_2 , CO_2 concentrations from the measured C_2H_4 , O_2 , CO_2 concentrations, respectively, in the banana container, then times the empty volume of the banana container and divided by the banana weight and the measurement duration, i.e., 1 h, as described in detail in the Support Information. For oxygen respiration, positive values were reported.

2.8. Statistical analysis

All measurements were performed at least three times, and the results are presented as mean with standard deviation (SD). t -Test was applied to evaluate statistical significance of the measured barrier coating performance parameters relative to the control set (no coating) in terms of banana weight loss, browning fraction, firmness, and TSS, as well as ethylene release and oxygen and carbon dioxide respiration between the coating runs and the control. Details of t -Test analysis were provided in the Support Information. One-way analysis of variance (ANOVA) was also used to assess the differences in the measured casting film physical properties and the contact angle and surface tension of different coating formulations on banana peel surface. Both the F values and p values were reported to evaluate the significance of the effect of LP substitution on these parameters. Tukey-test was then subsequently applied to evaluate the significance of the observed differences between any paired groups from measurements.

3. Results and discussion

3.1. The chemical and crystal structures of the MFC and LP fibrils

FTIR analysis reveals the chemical structures of MFC and LP fibrils. The MFC FTIR spectrum exhibited typical cellulose and hemicellulose structures (Table S5) [34] as shown in Fig. 1A. Specifically, OH stretching at 3335 cm⁻¹, C—H stretching around 2900 cm⁻¹, C—H bending at 1317 cm⁻¹, C—O—C antisymmetric bridge ring stretching at 1148 cm⁻¹, C—O stretching between 1014 and 1026 cm⁻¹, and β -glycosidic linkages of cellulose at 896 cm⁻¹. The main feature of the LP FTIR spectrum (Fig. 1A) is the distinct carbonyl (C=O) stretching peak at 1735 cm⁻¹ due to the presence of pectin in the lemon peels [35]. The galacturonic acid in LP pectin is represented by the O—H and C—H stretching at 3335 cm⁻¹ and around 2900 cm⁻¹, respectively. The β -glycosidic linkages peak at 896 cm⁻¹ is much weaker for LP than for MFC due to the significantly lower cellulose content (Tables S1 and S2). The peak at 1626 cm⁻¹ represents water in the MFC and LP fibrils.

X-ray diffraction (XRD) analysis was used to evaluate cellulose crystallinity [36]. XRD spectrum from the MFC exhibited the typical diffraction peak associated with crystalline cellulose at 22.3° (Fig. 1B). In contrast, the LP had a less intense signal with two main peaks: a broad peak at 22.3° and a sharp peak at 24.8°. This indicates that the LP has less crystalline cellulose than the MFC (CI of 48.7 % vs 68.5 % for MFC). Chemical composition analysis indicated that the LP had significantly lower cellulose content than the MFC (comparing Tables S1 with S2). Additionally, scattering from pectin in the LP can also contribute to the low crystallinity as pectin typically exhibits an amorphous large peak between 15 and 25° [37–39]. The sharp peak at 24.8° is attributable to the minerals in LP such as Ca, K or Fe [40]. This agrees with previously observed narrow peaks that were attributed to syngenite [40].

3.2. Morphologies of MFC and LP fibrils

The MFC produced from the commercial virgin BKHP (mixed birch and maple) shows a typical entangled fibril network structure as revealed by the light and scanning electron microscopic images (Fig. 2A1–A4). The MFC is relatively uniform in physical structure with a few large fibers remaining in the field of view (Fig. 2A1). SEM of freeze-

Table 3

Time-dependent images of water droplets on MFC/LP casting films.

	MFC 0.45	MFC/LP 0.40:0.05	MFC/LP 0.35:10	MFC/LP 0.30:0.15	MFC/LP 0.25:0.20
0 s					
1 s					
5 s					
10 s					
30 s					
60 s					

dried materials shows that the fibril networks are highly entangled and aggregated due to hydrogen bonding induced by drying for SEM (Fig. 2A2, A3). Submicron fibril aggregates/bundles can be clearly seen (Fig. 2A3). Individual fibrils and fibril aggregates can be better viewed in vacuum-dried samples (Fig. 2A4) with typical fibril diameters ranging from tens of nanometers to sub-micrometers. This morphology of MFC ensures the formation of a mechanically strong coating film [41,42].

Lemon peels are easier to fibrillate than wood fibers because they are formed mainly by the primary cell wall with weaker physical integrity. However, the fibrillated LP appears coarser and less uniform with a great extent of fibril aggregation than the MFC based on low magnification light microscopy (comparing Fig. 2B1 with A1, B2 with A2). LP fibrils contain a large amount of pectin, rich in hydroxyl and carboxy groups. Freeze drying for SEM imaging induced a great extent of hornification or fibril aggregation because of the hydroxyl and carboxyl groups. SEM imaging showed that the unaggregated fibrils in LP fibrils are finer than the unaggregated fibrils in MFC (comparing Fig. 2B3 with A3, B4 with A4) even though the LP fibrils were obtained with only one pass through the SMC compared with four passes for producing MFC. LP fibrils contain large amounts of pectin fibers and hemicelluloses. Mechanical treatments such as ultrasound can extract pectin from citrus peels [43]. The release of water-soluble pectin fibers by SMC milling in this study was evidenced by (1) the increase in the viscosity of the LP fibril suspension due to gelation by pectin based on visual observation, and (2) the formation of nano-size agglomerated pectin matters along the LP fibrils (Fig. 2B3 and B4). This type of agglomerated structures also called “grains” [44] were formed by the formation of ice crystals in freezing drying (for SEM) which squeezed the soluble pectin out of the crystals to be concentrated along the LP fibrils. This mechanism was well explained in literature and was used to produce complex hydrogel/cryogel

composites with interesting structures [45,46] including pectin hydrogels [47]. This “grain” structure was also clearly observed by atomic force microscopy in pectin/xyloglucan composite gel images [47].

3.3. Physical properties of MFC/LP casting films

Understanding the relevant physical properties of MFC/LP casting films can help to understand the performance of barrier coating using MFC/LP. The visual appearance of fruits is an important criterion that customers use in making purchasing decisions. The casting films made of different MFC/LP formulations all exhibited excellent transparency because of the microscale dimensions of the MFC and LP fibrils. All the logos covered with casting films (Fig. 3A0-A4) of different MFC/LP formulations can be clearly seen without noticeable differences, compared with the logos without film cover in Fig. 3A'. UV-VIS spectrophotometric measurements indicated that the film with the most LP fibril loading has the highest optical transparency in the visible range of 400–700 nm (Fig. 3B). It was reported that the application of pectin to polymethyl methacrylate (PMMA) improved pectin/PMMA film transparency [48]. The films containing LP fibrils also exhibited two absorption peaks at wavelengths around 283 nm and 324 nm, corresponding to the benzoyl ring and cinnamoyl ring, respectively, in the flavonoids of phenolic compounds [49,50] in the LP.

SEM imaging can reveal the physical microstructures of MFC/LP casting films. The top surfaces of all films (Fig. 4A and A') exhibited a more porous structure and were not as smooth as the bottom surface of the corresponding film (Fig. 4B and B', contacting the petri dish). The voids on the film's top surfaces reveal the porous nature of the films made primarily with a long aspect ratio MFC. The smooth bottom surface resulted from the settling effect of nanoscale cellulose fibrils in

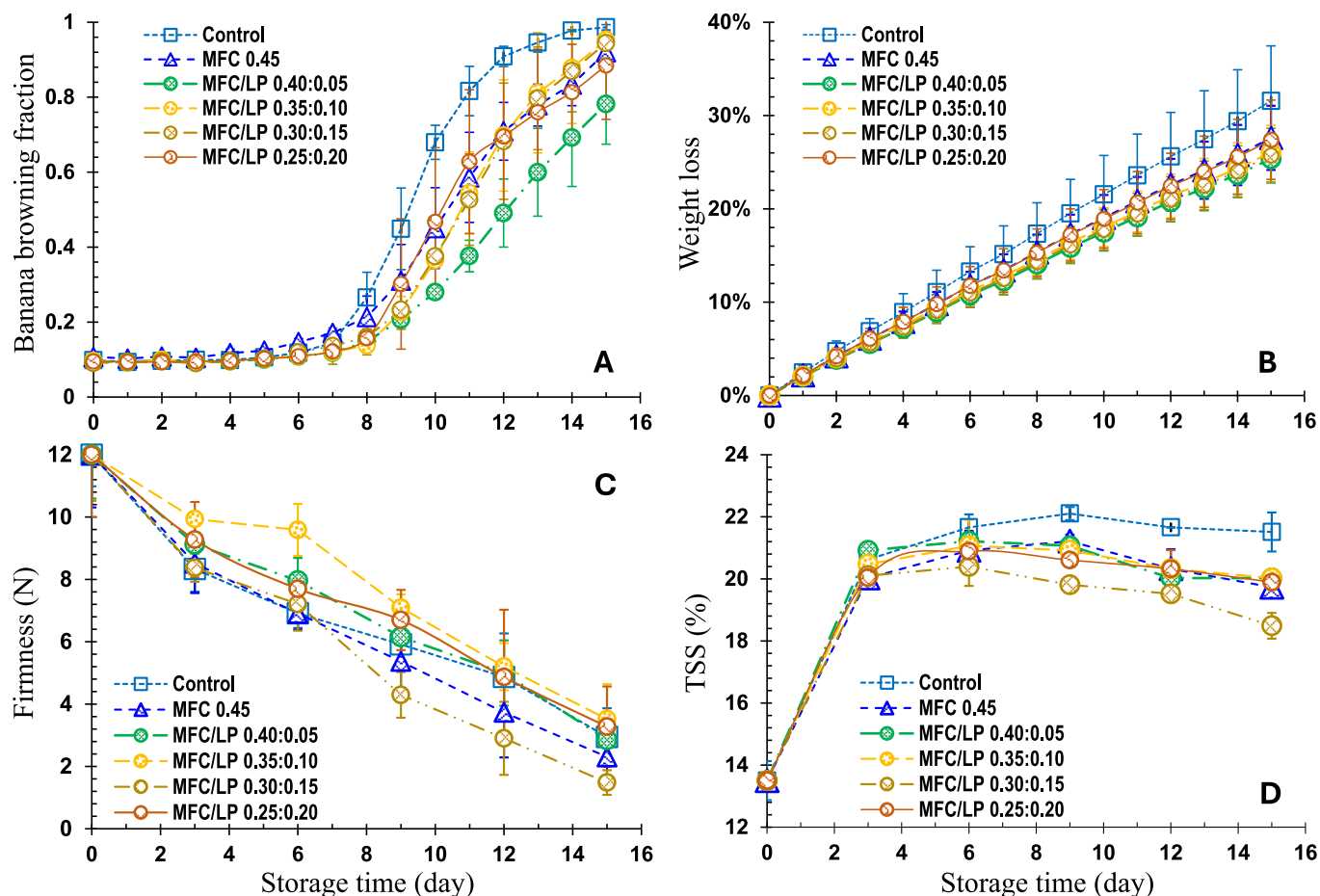


Fig. 5. Effects of MFC/LP barrier coating on time-dependent banana physical and chemical properties: (A) Peel browning fraction; (B) weight loss; (C) firmness; (D) total soluble sugars (TSS).

MFC, low aspect ratio LP fibrils, and the water-soluble viscous pectin fibers during film casting and drying. This is corroborated by the fact that the film made of pure MFC without short aspect ratio LP fibrils and pectin exhibited the largest void size on the order of 0.5–1 μm compared with other films (Fig. 4A'). Furthermore, the voids appear to decrease in both size and depth with increasing LP fibril substitution.

The density of the MFC/LP casting film was in general first increased with LP fibril substitution and then decreased, which is supported by ANOVA along with subsequent Tukey test (Table 1). The density of the casting film largely depends on the porosity of the film which increases with increasing LP substitution as discussed above (Fig. 4). It also depends on the densities of the constituent materials of the film, i.e., MFC and LP fibrils. Because SMC milling broke the micropores, such as the lumens and vessels in wood fibers and sacs and pores in the flavedo and albedo of lemon [51], as shown in Fig. 2A2, A3, B2, B3, the density of MFC can be assumed as the same as its constituents, i.e., cellulose and hemicelluloses of approximately 1.5 g/cm^3 , and the density of LP can be assumed as the same as its constituents, i.e., a lower amounts of cellulose and hemicelluloses and a larger amount of low density pectin of only 0.7 g/cm^3 . Therefore, increasing LP substitution decreased the density of the film constituent material, in addition to decreasing film porosity (Fig. 4) as discussed above. These two opposite contributing factors dictated the increasing and decreasing trend in the densities of the casting films with increasing LP substitution. The results in Table 1 indicate that the maximal density (lowest thickness) was achieved with an intermediate LP substitution of 22 % in formulation MFC/LP 0.35:0.10.

Film water vapor permeation depends on several factors such as the hydrophilicity of the films, the porosity or the density of the films, as well as the tortuosity of the pore channels within the films. It is generally

understood that a less hydrophilic film with greater density and pore channel tortuosity has a lower water vapor permeation. Increasing LP fibril substitution increased the resulting film hydrophilicity because LP contains a significant amount of pectin that is highly hydrophilic, as pectin is rich in hydroxyl and carboxy groups [52]. However, increasing LP substitution resulted in an increasing and decreasing trend in film density as discussed above. Consequently, the combined effects of film hydrophilicity and film density dictated the role of LP substitution on casting film water vapor permeation. The measured results indicate that in general the film water vapor transmittance rate (WVTR) was increased with increasing LP substitution (Table 1). This is also true for water vapor permeation (WVP) except for the film made of MFC/LP 0.035:10 with the greatest density. ANOVA supported both conclusions (Table 1). Subsequent Tukey test ($\alpha = 0.05$) indicates that the effect of LP substitution on WVTR observed is statistically significant for paired groups between each of the three low LP substitutions (X1, X2, X3) and the highest LP substitution (X5), affirmed the general trend of increasing LP increases WVTR. Similarly, the effect of LP substitution on WVP observed is statistically significant for paired groups between the highest density film (X3) and the films with each of the high LP substitutions (X4 and X5), which affirmed that the maximal WVP was achieved at a moderate LP substitution of 22 % (X3) MFC/LP 0.035:10 with greatest casting film density. Therefore, the trade-off for achieving a great casting film transparency by increasing LP substitution is increasing water vapor permeation.

In summary, the optical and physical properties of the casting MFC/LP films with constant grammage provided information on the effect of LP substitution on the film gas permeation. These properties are directly related to the film structure as well as the coating functionality.

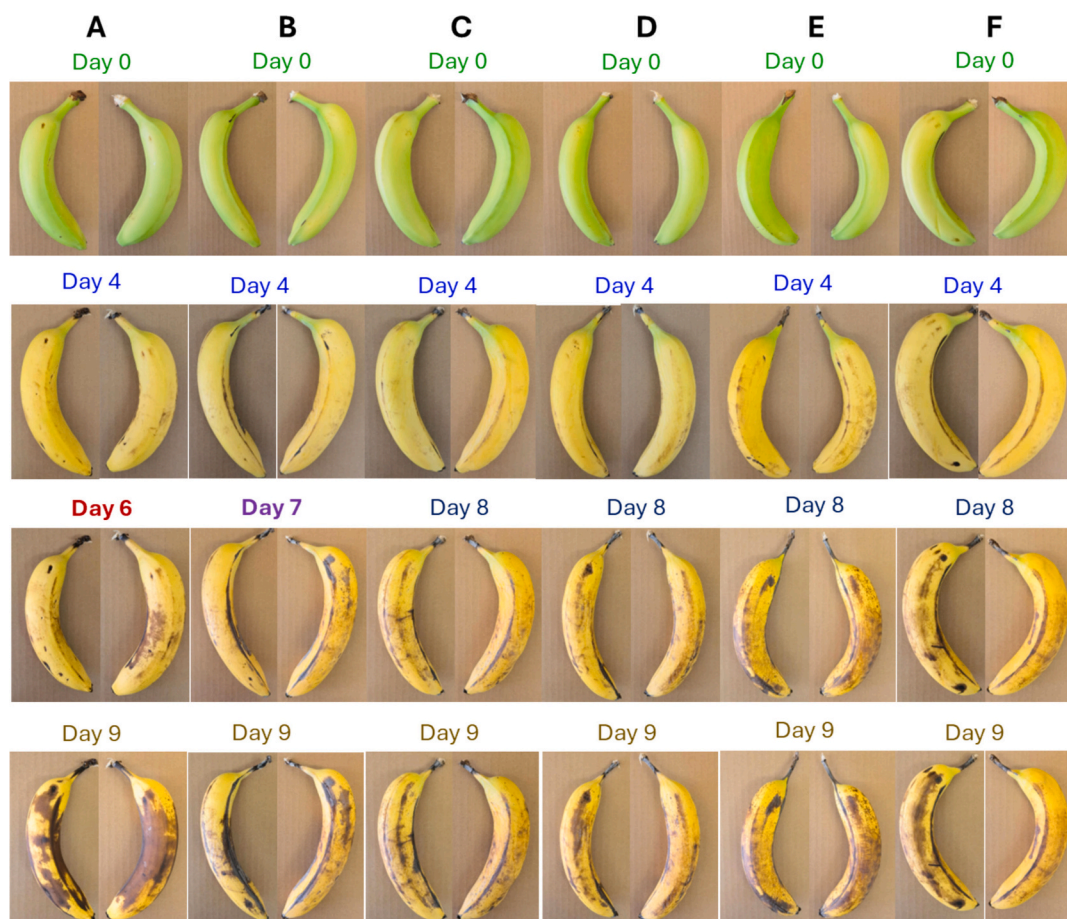


Fig. 6. Comparisons of the visual appearance of bananas without and with different coating formulations. A: Control (no coating); B: MFC 0.45; C: MFC/LP 0.40:0.05; D: MFC/LP 0.35:0.10; E: MFC/LP 0.30:0.15; F: MFC/LP 0.25:0.20.

Specifically, increasing LP substitution increased the optical transparency in the visible range of the barrier coating film, which ensures the bright color of fresh fruits without negatively affecting fruit appealing to customers. It also increases the content of flavonoids of phenolic compounds to result in a film with improved antioxidant and antimicrobial capabilities as the benzoyl and cinnamoyl rings in flavonoids have natural antioxidant and antimicrobial properties. Casting film structural porosity or density is directly related to film water vapor permeation and therefore affects barrier coating performance such as preventing fruit moisture or weight losses, regulate fruit respiration and ripening as will be discussed later in the Sections 3.5–3.6. The coating formulation of MFC/LP 0.35:0.10 with moderate LP substitution of 22 % resulted in the greatest casting film density (lowest porosity) and lowest WVP. This suggests that moderate LP substitution levels are preferred to achieve best barrier performance. This guideline is validated by banana barrier coating experiments, i.e., either high LP substitutions, or zero LP substitution (MFC 0.45) did not achieve the best performance for banana preservation, rather with the substitution of 11 % using formulation MFC/LP 0.40:0.05, as will be discussed later in the Sections 3.5–3.7. The slight deviation in the optimal LP substitution level from the casting film study is attributable to several factors such as coating film thickness and pore channel tortuosity, as well as film adhesion on banana peel surface.

3.4. Banana surface wettability

The wettability of MFC/LP suspensions on the banana peel surface was evaluated to predict the coating behavior for producing good and uniform barrier. The surface tensions of MFC/LP suspensions γ_{LV} first decreased with increasing LP substitution, reaching the lowest value of

36.0 mN/m at LP substitution of 22 %, i.e., MFC/LP 0.35:0.10, and then increased, as seen in Table 2. The suspension contact angle was not much affected with increasing LP substitution except at the highest LP substitution MFC/LP 0.25:0.20, which is affirmed by Tukey test. These data suggest that suspensions with lower LP substitutions of 11 % and 22 %, i.e., MFC/LP 0.40:0.05 and MFC/LP 0.35:0.10, had a better wettability and coating efficiency on the banana surface [53].

Good water washability of the coating layer is attractive to consumers even though bananas are always consumed after peeling. The hydrophilicity of MFC/LP casting films was evaluated as a measure of coating washability. It can be seen from Table 3 that the pure MFC casting film completely absorbed the water droplet in 10 s. Moreover, the water droplets on all MFC/LP casting films can be thoroughly penetrated and absorbed in 30 s. This fast water absorption suggests that all MFC/LP barrier coatings have great hydrophilicity and should be easily washable using water.

3.5. Performance of MFC/LP barrier coating on bananas

Banana peel browning is an important factor consumers use in making purchasing decisions and is often correlated to the freshness of the bananas. The banana browning fraction was measured using image analysis. As expected, banana browning fraction increased with storage time as shown in Fig. 5A. All MFC/LP barrier coating formulations slowed down the browning of the banana peel. The MFC/LP 0.40:0.05 formulation had the most significant effect on slowing down the banana browning process. Rapid peel browning started at about 7 days of storage. This can be directly visualized from the photographic images of the bananas in Fig. 6. The banana peel color changed from greenish to

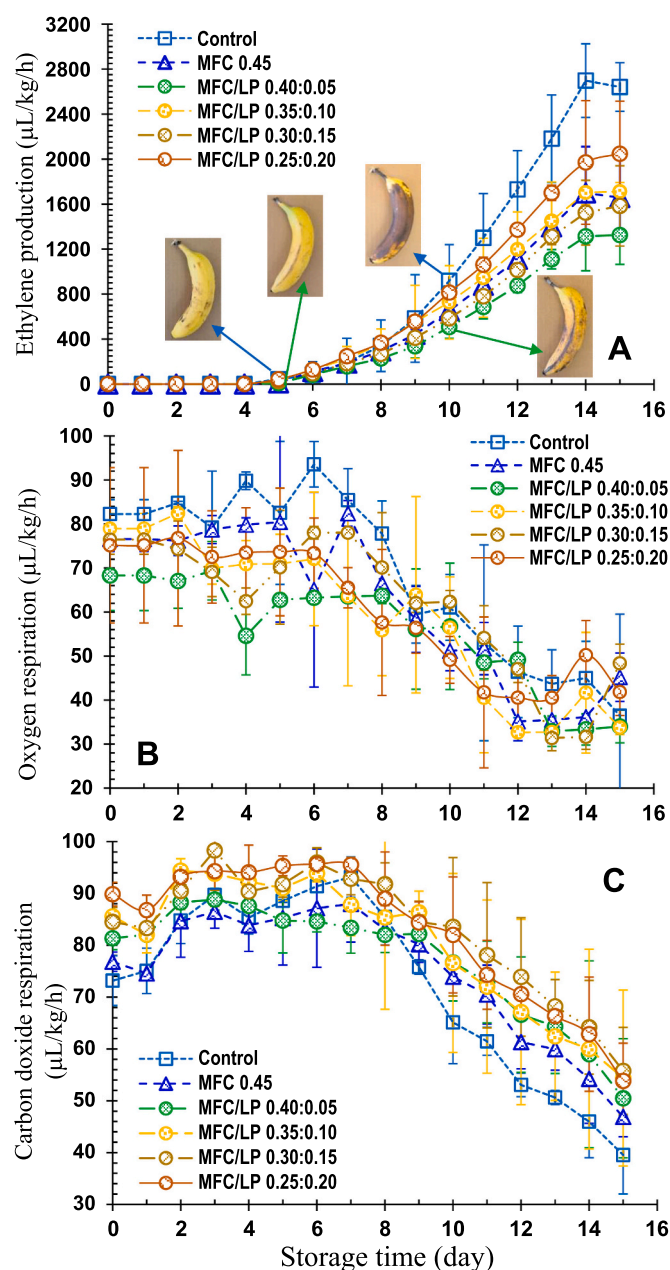


Fig. 7. Effects of MFC/LP barrier coating on time-dependent banana respiration. (A): Ethylene; (B): oxygen; (C): carbon dioxide.

yellowish in the first 4 days. By day 4, the bananas remained appealing to consumers with minor peel browning. The effectiveness of different barrier formulations can be seen by comparing the number of days to reach a similar level of browning at which the banana is still acceptable to consumers, in other words, “the last day to sell”. As shown in Figs. 6 and S2, the last day to sell was day 6 for the control, day 7 for the pure MFC coating, and day 8 for the MFC/LP coatings. This comparison suggests that barrier coating with MFC/LP 0.40:0.05 slowed banana peel browning by 2 days, in agreement with our earlier banana barrier coating study using CMC substitution of MFC [27]. It should be emphasized that 2 days, though short, represent a 33 % increase in shelf life compared with control considering typical banana shelf life in supermarkets is only approximately 7 days. The bananas used in the study were purchased from a local market, which could be a week after harvest. This time elapses since harvest can significantly affect coating performance. The difference in the appearance of bananas without and

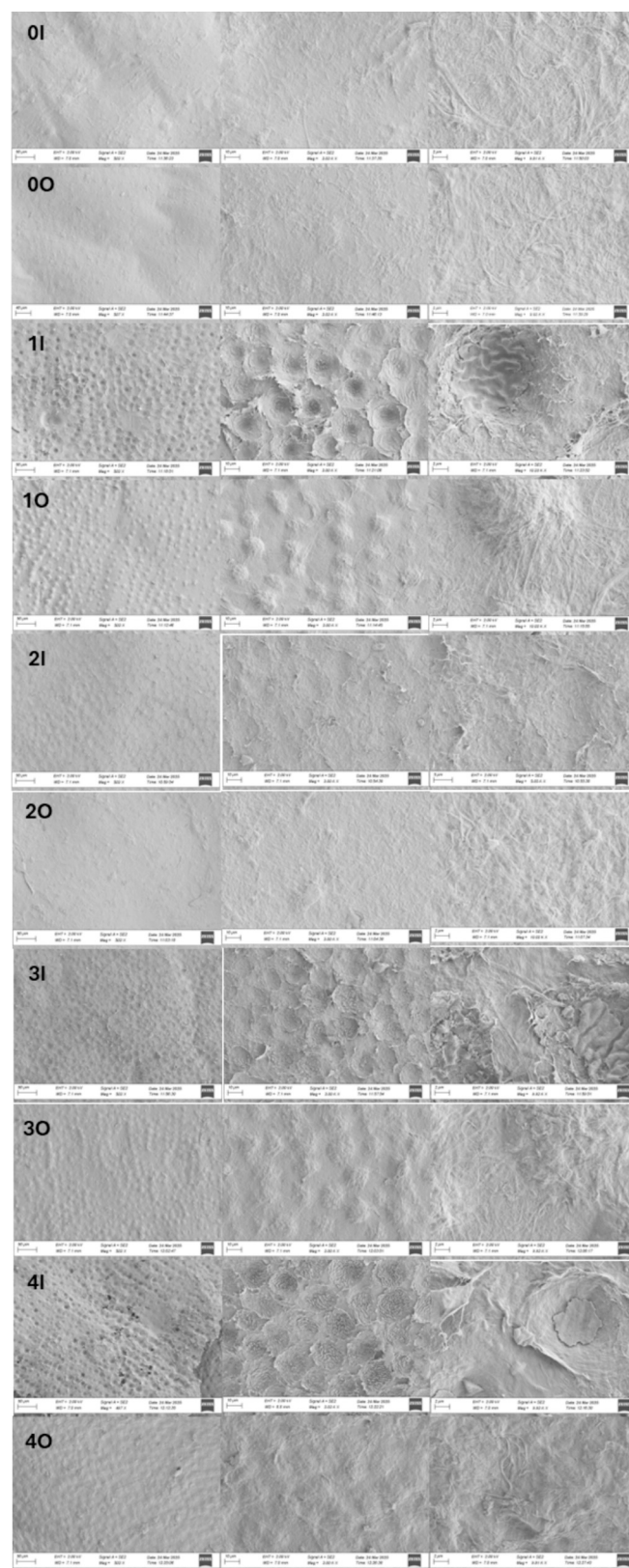


Fig. 8. SEM images of MFC/LP barrier coating films peeled off banana peel surface. O: MFC 0.45, 1: MFC/LP = 0.40:0.05, 2: MFC/LP = 0.35:0.10, 3: MFC/LP = 0.30:0.15, 4: MFC/LP = 0.25:0.20; I: Inner surface touching banana epicarp, O: outer surface facing ambient air.

with different coatings on day 9 further validated the effectiveness of MFC/LP coatings. Fitting the browning fraction data using a logistic model (Eqs. (2) and (3)) resulted in an EC_{10} (the onset of rapid browning) value of approximately 7 days for the control sample (Table S6). Comparing the EC_{10} values for different coatings, the effectiveness of MFC/LP coatings for slowing down browning is evident, and in agreement with the visual observations discussed above. *t*-Test indicates that the observed differences in browning fractions between each of the coating groups and the control group (no coating) were statistically significant with the probability of greater than 99 % (Table S7).

It is generally understood that the oxidation of the natural phenols in the banana peel through enzymatic reactions results in the brown pigments of benzo-quinone and melanin [54,55]. Barrier coating decreased oxygen supply as can be seen from the decreased oxygen respiration of bananas as will be discussed later, which decreased the formation of brown spots [2,56].

Fruit weight loss is another important measure of barrier coating performance because it directly affects the economic profits of growers and grocers as bananas are sold by weight. Different MFC/LP coating formulations all decreased banana weight loss as shown in Fig. 5B. Formulations with the lowest LP substitution of 11 %, e.g., MFC/FC = 0.40:0.05, had the lowest weight losses at approximately 14 % on day 8 (the last day to sell). Pure MFC and the formulation with the greatest LP substitution of 44 %, i.e., MFC/LP = 0.25:0.20, had slightly more weight losses but remained lower than the control. *t*-Test indicates that the measured differences in banana weight loss between each of the coating groups and the control group (no coating) were statistically significant with the probability of greater than 99.9 % (Table S7).

Barrier coating using MFC/LP had a limited effect on banana fruit firmness. The measured banana fruit firmness of bananas with and without coatings is within the error margin as shown in Fig. 5C, especially when comparing the firmness values on the last day to sell among the control banana (day 6) and the coated bananas (day 8). *t*-Test supported this (null) hypothesis with 95 % probability (Table S7). This also agrees with our earlier study using CMC substitution of MFC under normal banana ripening scenarios [27]. Barrier coating did not affect the rapid rise in the total dissolved solids (TSS) in the banana fruits in the first 3 days of storage as shown in Fig. 5D. TSS plateaued after 3 days. Coated banana fruits generally had a slightly lower TSS level supported by *t*-Test with 95 % probability (Table S7), which is favorable to grocers as most consumers prefer a not too firm banana once it ripens.

3.6. Effect of barrier coating on banana respiration

It is well known that a surge in ethylene production occurs during banana ripening [1]. Within the first week of storage, ethylene release was low as shown in Fig. 7A, similar to banana browning (Fig. 5A). The effect of LP loading in coating on ethylene release was not differentiable for some coating formulations due to the variations in their initial states among different bananas as well as the low amount of ethylene release. The rapid release of ethylene started on day 6 when the effect of the barrier coating on ethylene release and peel browning became noticeable as validated by *t*-Test with 95 % probability except for the coating group with MFC/LP 0.35:0.10 (Table S7). Ethylene is a plant hormone that triggers color changes and fruit ripening [57]. The measured ethylene release data indicated a direct correlation between ethylene release and banana peel browning (Fig. S3A). The data also showed that this correlation is universal for bananas with and without MFC/LP barrier coatings, i.e., all the data sets collapsed onto the same correlation curve. This indicated that suppressing ethylene release can decrease banana peel browning. As shown in Fig. 7A, barrier coating especially using the formulation MFC/LP 0.40:0.05 decreased ethylene release, as a result, it slowed down banana browning compared with control (no coating) as presented in Figs. 5A and 6.

The ethylene release data also had an excellent universal correlation with banana weight loss during ripening with or without barrier coating

(Fig. S3B). However, the correlation curve had an *y* intercept of approximately 10 %. This intercept occurs because weight loss started on day 1, long before the surge of ethylene production on day 6, and increased linearly with storage time as shown in Fig. 5B. This correlation suggests that banana weight loss in the first 6 days is primarily contributed by the loss of moisture during which the biological ripening process was slow. The barrier coating not only can prevent moisture loss but also slow down ethylene release to result in decreasing banana weight loss (Fig. 5B). A negative universal correlation was obtained between ethylene release and banana fruit firmness (Fig. S3C), which confirmed that the bananas became soft as a result of biochemical reactions in the ripening process that released ethylene. It was noticed that the total amount of dissolved solids had a very weak correlation with ethylene release (Fig. S3D), which suggests that enzymatic hydrolysis of starch and degradation of cell wall are not related to ethylene production.

Decreasing environment oxygen concentration can slow ethylene production, fruits ripening, and the deterioration of fruits [58]. Respiration data indicated that barrier coating decreased oxygen respiration, as shown in Fig. 7B. *t*-Test supported the observed differences in oxygen respiration between each of the coating groups and the control group (no coating) with 95 % probability (Table S7). Furthermore, the coating using MFC/LP 0.40:0.05 resulted in the lowest oxygen respiration with the control being the highest, which was affirmed by *t*-Test with 99.9 % probability (Table S7). The oxygen respiration data agreed with the ethylene release data reported in Fig. 7A and banana peel browning in Figs. 5A and 6. Because the oxidation of natural phenols in banana peels resulted in the peel browning, decreasing oxygen respiration can decrease oxygen supply to alleviate peel browning [2,56]. The data showed that oxygen respiration was relatively constant in the first week right before the surge of ethylene production, then decreased linearly with time after day 7 by approximately 50 %, and leveled off after day 12, consistent with the leveling-off of ethylene release shown in Fig. 7A. The measured carbon dioxide release data (Fig. 7C) showed a similar trend to that of oxygen respiration except for the pure MFC 0.45 coating group. *t*-Test affirmed the observed differences between each of the coating groups and the control (no coating) with a probability of 95 % (Table S7). Leveling off carbon dioxide concentration was not observed even after 15 days.

3.7. Microstructures of barrier coating film on banana peel

Barrier coating performance depends not only on the basic film properties of the coating formulation, which can be evaluated using casting films, but also on the film's adhesion to the banana peel. Lose contact between the coating film and the banana peel surface with insufficient adhesion leaves space between the film and the banana peel. This space serves as a reservoir for supplying oxygen and other gases, accelerating ripening, and resulting in poor barrier performance. SEM imaging of the fragments of peeled films from banana surfaces coated with different MFC/LP formulations revealed the prints of the banana epicarp surface in the form of dimples formed on the coating films by protrusions of the banana epidermal cells as reported in the literature [56,59]. The dimples are hemispherical with a uniform diameter of approximately 10 μm [27] as shown in Fig. 8. The dimples are very shallow and almost invisible on the pure MFC 0.45 coating film fragment (Fig. 8OI and OO). The dimples are deeper and more distinct on the coating film fragments with MFC/LP formulations (Fig. 8). It should be noted that peeling off the MFC/LP coating films was considerably more difficult than peeling off the pure MFC coating film, suggesting MFC/LP coatings had better adhesion than pure MFC coating, and furthermore, Fig. 8 may not represent the regions with the most adhesion to the banana peels for the MFC/LP coating films, and therefore, it did not show deeper dimples for the formulations with greater LP fibril substitution. The greater adhesion of the coating to banana peels using formulations containing LP fibrils also supports improved barrier performance, in

agreement with the ethylene release and oxygen respiration data shown in Fig. 7.

3.8. Future work

This study shows promise in using plant-based carbohydrates for barrier coating of bananas to extend shelf life to reduce postharvest waste. However, more work is needed for commercialization. Issues to be addressed include: (1) demonstrating the concept using immediately harvested fruits, studies are underway in our laboratory currently by working with collaborators. (2) understanding the effect of barrier coating on fruit taste, nutrition, etc. It is known that a too dense coating can result in an anerobic ripening process to form alcohols due to lack of oxygen respiration [7,60]. Any disruption in normal fruit ripening can certainly change the taste of fruit and nutrition value. We will rely on collaborators with established protocols for this study using both standard instruments and human taste testing. (3) demonstrating the safety of the coating material and process with approval from regulatory agencies by using established in-vitro and in-vivo testing methods. (4) field trial studies at our collaborator fruit farm in large quantities using different fruits and commercial harvest process flow to obtain relevant process data for tech-economic and life cycle analyses.

4. Conclusions

This study developed an effective barrier coating formulation using pure plant-based carbohydrate polymers to extend the shelf life of bananas. The barrier coating formulation mixed two carbohydrate polymers: microfibrillated cellulose (MFC) from commercial bleached hardwood pulp fibers and fibrils of commercial lemon peels (LP). The formulation with the best performance had a MFC over LP concentration (wt%) ratio of 0.40:0.05. Compared with the control (no coating), barrier coating with this formulation decreased ethylene production and slowed down banana peel browning, resulting in a longer shelf life of 2 days, as well as decreased banana weight loss. Compared with barrier coating using pure MFC, the substitution of MFC with LP fibrils increased the adhesion of the coating film on the banana peel surface to form a tight coating layer as revealed by SEM imaging, which decreased ethylene release and oxygen respiration. The decreased oxygen respiration slowed down the enzymatic oxidation reactions responsible for banana peel color browning during banana ripening. The substitution of MFC with LP fibrils also improved the hydrophilia of the coating film, which improved the washability of the coating layer even though bananas are always consumed after peeling. This study demonstrated the utilization of whole LP without extraction/separation.

CRedit authorship contribution statement

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

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Declaration of competing interest

Authors Geng, Zhu, and Zhang are co-inventors of a provisional U.S. patent application.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ijbiomac.2025.149666>.

Data availability

Some data are available upon request.

References

- [1] A. Salvador, T. Sanz, S.M. Fiszman, Changes in colour and texture and their relationship with eating quality during storage of two different dessert bananas, *Postharvest Biol. Technol.* 43 (2007) 319–325, <https://doi.org/10.1016/j.postharvbio.2006.10.007>.
- [2] O. Steinbock, Q. Wang, P. Knoll, Front-like expansion and arrest of programmed cell death in brown banana spots, *Phys. Biol.* 19 (2022) 046003, <https://doi.org/10.1088/1478-3975/ac664d>.
- [3] M. Al-Dairi, P.B. Pathare, R. Al-Yahyai, H. Jayasuriya, Z. Al-Attabi, Postharvest quality, technologies, and strategies to reduce losses along the supply chain of banana: a review, *Trends Food Sci. Technol.* 134 (2023) 177–191, <https://doi.org/10.1016/j.tifs.2023.03.003>.
- [4] L. Amoroso, K.J. De France, C.I. Milz, G. Siqueira, T. Zimmermann, G. Nyström, Sustainable cellulose nanofiber films from carrot pomace as sprayable coatings for food packaging applications, *ACS Sustain. Chem. Eng.* 10 (2022) 342–352, <https://doi.org/10.1021/acssuschemeng.1c06345>.
- [5] M.H. Shinga, O.A. Fawole, *Opuntia ficus indica* mucilage coatings regulate cell wall softening enzymes and delay the ripening of banana fruit stored at retail conditions, *Int. J. Biol. Macromol.* 245 (2023) 125550, <https://doi.org/10.1016/j.ijbiomac.2023.125550>.
- [6] D. Hu, H. Wang, L. Wang, Physical properties and antibacterial activity of quaternized chitosan/carboxymethyl cellulose blend films, *LWT* 65 (2016) 398–405, <https://doi.org/10.1016/j.lwt.2015.08.033>.
- [7] J. Geng, J. O'Dell, N. Stark, P. Kitin, X. Zhang, J.Y. Zhu, Microfibrillated cellulose (MFC) barrier coating for extending banana shelf life, *Food Hydrocoll.* 150 (2024) 109671, <https://doi.org/10.1016/j.foodhyd.2023.109671>.
- [8] N. Lavoine, I. Desloges, A. Dufresne, J. Bras, Microfibrillated cellulose - its barrier properties and applications in cellulosic materials: a review, *Carbohydr. Polym.* 90 (2012) 735–764, <https://doi.org/10.1016/j.carbpol.2012.05.026>.
- [9] N.A. Bahmid, S.A. Siddiqui, H.D. Arianto, K.S. Sasmitaloka, N.B. Rathod, S. K. Wahono, et al., Cellulose-based coating for tropical fruits: method, characteristic and functionality, *Food Rev. Int.* 40 (2024) 1069–1092, <https://doi.org/10.1080/87559129.2023.2209800>.
- [10] J.D. Ede, K.J. Ong, M. Goergen, A. Rudie, C.A. Pomeroy-Carter, J.A. Shatkin, Risk analysis of cellulose nanomaterials by inhalation: current state of science, *Nanomaterials* 9 (2019) 337, <https://doi.org/10.3390/nano9030337>.
- [11] H. Kato-Noguchi, M. Kato, Pesticidal activity of citrus fruits for the development of sustainable fruit-processing waste management and agricultural production, *Plants* 14 (2025) 754, <https://doi.org/10.3390/plants14050754>.
- [12] N. Mahato, M. Sinha, K. Sharma, R. Koteswararao, M.H. Cho, Modern extraction and purification techniques for obtaining high purity food-grade bioactive compounds and value-added co-products from citrus wastes, *Foods* 8 (2019) 248, <https://doi.org/10.3390/foods8110523>.
- [13] H. Jiang, W. Zhang, Y. Xu, L. Chen, J. Cao, W. Jiang, An advance on nutritional profile, phytochemical profile, nutraceutical properties, and potential industrial applications of lemon peels: a comprehensive review, *Trends Food Sci. Technol.* 124 (2022) 219–236, <https://doi.org/10.1016/j.tifs.2022.04.019>.
- [14] V. Revathi, S. Bora, N. Afzia, T. Ghosh, Orange peel composition, biopolymer extraction, and applications in paper and packaging sector: a review, *Sustain. Chem. Pharm.* 43 (2025) 101908, <https://doi.org/10.1016/j.scp.2025.101908>.
- [15] M.R.V. Bertolo, T.S. Pereira, F.V. dos Santos, M.H.M. Facure, F. dos Santos, K.B. R. Teodoro, et al., Citrus wastes as sustainable materials for active and intelligent food packaging: current advances, *Compr. Rev. Food Sci. Food Saf.* 24 (2025) e70144, <https://doi.org/10.1111/1541-4337.70144>.

- [16] N. Durmus, Z. Gulsunoglu-Konuskan, M. Kilic-Akyilmaz, Recovery, bioactivity, and utilization of bioactive phenolic compounds in citrus peel, *Food Sci. Nutr.* 12 (2024) 9974–9997, <https://doi.org/10.1002/fsn3.4570>.
- [17] S. Grover, P. Aggarwal, A. Kumar, S. Kaur, R. Yadav, N. Babbar, Utilizing citrus peel waste: a review of essential oil extraction, characterization, and food-industry potential, *Biomass Convers. Biorefinery* 15 (2025) 5043–5064, <https://doi.org/10.1007/s13399-024-05382-y>.
- [18] D. Yun, Z. Wang, C. Li, D. Chen, J. Liu, Antioxidant and antimicrobial packaging films developed based on the peel powder of different citrus fruits: a comparative study, *Food Biosci.* 51 (2023) 102319, <https://doi.org/10.1016/j.fbio.2022.102319>.
- [19] N. Kumar, A. Upadhyay, S. Shukla, V.K. Bajpai, M. Kieliszek, A. Yadav, et al., Next generation edible nanoformulations for improving post-harvest shelf-life of citrus fruits, *J. Food Meas. Charact.* 18 (2024) 1825–1856, <https://doi.org/10.1007/s11694-023-02287-8>.
- [20] M.A.P. Cotrim, M.R.F. de Ferraz, E. Ayres, Pectin edible film obtained from orange peel: characterization and application to improve the quality of minimally processed apples, *Macromol. Symp.* 413 (2024) 2400088, <https://doi.org/10.1002/masy.202400088>.
- [21] P. Kalita, T. Kalita, N.S. Bora, P.K. Roy, L. Pachau, S. Roy, A review on natural polymer-based film/coating in postharvest mushroom preservation, *Food Control* 172 (2025) 111185, <https://doi.org/10.1016/j.foodcont.2025.111185>.
- [22] D. Yun, J. Liu, Recent advances on the development of food packaging films based on citrus processing wastes: a review, *J. Agric. Food Res.* 9 (2022) 100316, <https://doi.org/10.1016/j.jafr.2022.100316>.
- [23] H. Taghavi Kevij, M. Salami, M. Mohammadian, M. Khodadadi, Z. Emam-Djomeh, Mechanical, physical, and bio-functional properties of biopolymer films based on gelatin as affected by enriching with orange peel powder, *Polym. Bull.* 78 (2021) 4387–4402, <https://doi.org/10.1007/s00289-020-03319-9>.
- [24] P. Terzioğlu, F. Güney, F.N. Parin, İ. Şen, S. Tuna, Biowaste orange peel incorporated chitosan/polyvinyl alcohol composite films for food packaging applications, *Food Packag. Shelf Life* 30 (2021) 100742, <https://doi.org/10.1016/j.fpsl.2021.100742>.
- [25] H.F. Chhatariya, S. Srinivasan, P.M. Choudhary, S.S. Begum, Corn starch biofilm reinforced with orange peel powder: characterization of physicochemical and mechanical properties, *Mater. Today Proc.* 59 (2022) 884–892, <https://doi.org/10.1016/j.matpr.2022.01.339>.
- [26] N. Meydanju, S. Pirsia, J. Farzi, Biodegradable film based on lemon peel powder containing xanthan gum and TiO₂-Ag nanoparticles: investigation of physicochemical and antibacterial properties, *Polym. Test.* 106 (2022) 107445, <https://doi.org/10.1016/j.polymertesting.2021.107445>.
- [27] J. Geng, N. Stark, P. Kitin, X. Zhang, N. Plaza, J. Zhu, Incorporating carboxymethyl cellulose into microfibrillated cellulose for barrier coating to delay banana browning and extend shelf life, *Food Chem.* 486 (2025) 144664, <https://doi.org/10.1016/j.foodchem.2025.144664>.
- [28] P. Paudel, S.H. Seong, A. Wagle, B.S. Min, H.A. Jung, J.S. Choi, Antioxidant and anti-browning property of 2-arylbenzofuran derivatives from *Morus alba* Linn root bark, *Food Chem.* 309 (2020) 125739, <https://doi.org/10.1016/j.foodchem.2019.125739>.
- [29] J. Kim, J.Y. Choi, K.D. Moon, Effect of edible coating with *Morus alba* root extract and carboxymethyl cellulose for enhancing the quality and preventing the browning of banana (*Musa acuminata* Cavendish) during storage, *Food Packag. Shelf Life* 31 (2022) 100809, <https://doi.org/10.1016/j.fpsl.2022.100809>.
- [30] A. Sluiter, B. Hames, R. Ruiz, C. Scarlata, J. Sluiter, D. Templeton, et al., Determination of structural carbohydrates and lignin in biomass laboratory analytical procedure (LAP), in: NREL Technical Report. NREL/TP-510-42618 (Version 07-08-2011), 2011.
- [31] M.W. Davis, A rapid modified method for compositional carbohydrate analysis of lignocellulosics by high pH anion-exchange chromatography with pulsed amperometric detection (HPAEC/PAD), *J. Wood Chem. Technol.* 18 (1998) 235–352.
- [32] C.S. Hu, Y. Zhao, K.C. Li, J.Y. Zhu, R. Gleisner, Optimizing cellulose fibrillation for the production of cellulose nanofibrils by a disk grinder, *Holzforschung* 69 (2015) 993–1000, <https://doi.org/10.1515/hf-2014-0219>.
- [33] X. Zhang, P. Kitin, U.P. Agarwal, R. Gleisner, J.Y. Zhu, Characterizing lignin-containing microfibrillated cellulose based on water interactions, fibril properties, and imaging, *Carbohydr. Polym.* 316 (2023) 120996, <https://doi.org/10.1016/j.carbpol.2023.120996>.
- [34] Z. Chen, T.Q. Hu, H.F. Jang, E. Grant, Modification of xylan in alkaline treated bleached hardwood kraft pulps as classified by attenuated total-internal-reflection (ATR) FTIR spectroscopy, *Carbohydr. Polym.* 127 (2015) 418–426, <https://doi.org/10.1016/j.carbpol.2015.03.084>.
- [35] D.B. Pal, A. Kapoor, A.K. Arya, R.K. Arya, A.K. Tiwari, Characterization and sustainable utilization of *Punica granatum* and *Citrus limetta* peels: insights for biomass valorization, *Biomass Convers. Biorefinery* 15 (2025) 24403–24411, <https://doi.org/10.1007/s13399-024-05553-x>.
- [36] K.S. Salem, N.K. Kasera, M.A. Rahman, H. Jameel, Y. Habibi, S.J. Eichhorn, et al., Comparison and assessment of methods for cellulose crystallinity determination, *Chem. Soc. Rev.* 52 (2023) 6417–6446, <https://doi.org/10.1039/d2cs00569g>.
- [37] A.O. Nasser, S.L. Kareem, Removal of Congo red from aqueous solution using lemon peel-Fe₃O₄ nanocomposite adsorbent, *Biomass Conversion and Biorefinery*. 14 (2024) 23183–23193, <https://doi.org/10.1007/s13399-023-04496-z>.
- [38] R. Karim, K. Nahar, F.T. Zohora, M.M. Islam, R.H. Bhuiyan, M.S. Jahan, et al., Pectin from lemon and mango peel: extraction, characterisation and application in biodegradable film, *Carbohydr. Polym. Technol. Appl.* 4 (2022) 100258, <https://doi.org/10.1016/j.carpta.2022.100258>.
- [39] N. Wathoni, C. Yuan Shan, W. Yi Shan, T. Rostinawati, R.B. Indrari, R. Pratiwi, et al., Characterization and antioxidant activity of pectin from Indonesian mangosteen (*Garcinia mangostana* L.) rind, *Heliyon* 5 (2019) e02299, <https://doi.org/10.1016/j.heliyon.2019.e02299>.
- [40] A. Czech, E. Zarycka, D. Yanovych, Z. Zasada, I. Grzegorzczak, S. Klys, Mineral content of the pulp and peel of various citrus fruit cultivars, *Biol. Trace Elem. Res.* 193 (2020) 555–563, <https://doi.org/10.1007/s12011-019-01727-1>.
- [41] K. Syverud, P. Stenius, Strength and barrier properties of MFC films, *Cellulose* 16 (2009) 75–85.
- [42] S. Van Nguyen, B.K. Lee, Microfibrillated cellulose film with enhanced mechanical and water-resistant properties by glycerol and hot-pressing treatment, *Cellulose* 28 (2021) 5693–5705, <https://doi.org/10.1007/s10570-021-03894-8>.
- [43] M. Kumari, S. Singh, A.K. Chauhan, A comparative study of the extraction of pectin from Kinnow (*Citrus reticulata*) peel using different techniques, *Food Bioprocess Technol.* 16 (2023) 2272–2286, <https://doi.org/10.1007/s11947-023-03059-4>.
- [44] J. Grenier, H. Duval, F. Barou, P. Lv, B. David, D. Letourneur, Mechanisms of pore formation in hydrogel scaffolds textured by freeze-drying, *Acta Biomater.* 94 (2019) 195–203, <https://doi.org/10.1016/j.actbio.2019.05.070>.
- [45] S. Deville, E. Saiz, R.K. Nalla, A.P. Tomsia, Freezing as a path to build complex composites, *Science* 311 (2006) 515–518, <https://doi.org/10.1126/science.1120937>.
- [46] J. Grenier, H. Duval, P. Lv, F. Barou, C. Le Guilcher, R. Aid, et al., Interplay between crosslinking and ice nucleation controls the porous structure of freeze-dried hydrogel scaffolds, *Biomater. Adv.* 139 (2022) 212973, <https://doi.org/10.1016/j.bioadv.2022.212973>.
- [47] S. Feng, J. Bi, J. Yi, X. Li, J. Lyu, Y. Guo, et al., Modulation of ice crystal formation behavior in pectin cryogel by xyloglucan: effect on microstructural and mechanical properties, *Food Res. Int.* 159 (2023) 111555, <https://doi.org/10.1016/j.foodres.2022.111555>.
- [48] F. Zou, H. Li, Y. Dong, G.C. Tewari, J. Vapaavuori, Optically transparent pectin/poly(methyl methacrylate) composite with thermal insulation and UV blocking properties based on anisotropic pectin cryogel, *Chem. Eng. J.* 439 (2022) 135738, <https://doi.org/10.1016/j.cej.2022.135738>.
- [49] N. Jayaprakash, R. Suresh, S. Rajalakshmi, S. Raja, E. Sundaravadeivel, M. Gayathri, et al., One-step synthesis, characterisation, photocatalytic and bio-medical applications of ZnO nanoplates, *Mater. Technol.* 35 (2020) 112–124, <https://doi.org/10.1080/10667857.2019.1659533>.
- [50] W.J. Chien, D.S. Saputri, H.Y. Lin, Valorization of Taiwan's Citrus depressa Hayata peels as a source of nobletin and tangeretin using simple ultrasonic-assisted extraction, *Curr. Res. Food Sci.* 5 (2022) 278–287, <https://doi.org/10.1016/j.crf.2022.01.013>.
- [51] H. Wang, H. Xiao, Y. Wu, F. Zhou, C. Hua, L. Ba, et al., Characterization of volatile compounds and microstructure in different tissues of 'Eureka' lemon (*Citrus limon*), *Int. J. Food Prop.* 25 (2022) 404–421, <https://doi.org/10.1080/10942912.2022.2046600>.
- [52] H. Zhang, Z. Fan, D. Peng, C. Huang, X. Wu, F. Sun, Tuning the hydrophobic performance and thermal stability of pectin film by acetic anhydride esterification, *Int. J. Biol. Macromol.* 276 (2024) 133746, <https://doi.org/10.1016/j.ijbiomac.2024.133746>.
- [53] S. Soradech, J. Nunthanid, S. Limmatvapirat, M. Luangtana-anan, Utilization of shellac and gelatin composite film for coating to extend the shelf life of banana, *Food Control* 73 (2017) 1310–1317, <https://doi.org/10.1016/j.foodcont.2016.10.059>.
- [54] M.A. Joslyn, J.D. Ponting, Enzyme-catalyzed oxidative browning of fruit products, *Adv. Food Res.* 3 (1951) 1–44, [https://doi.org/10.1016/S0065-2628\(08\)60258-X](https://doi.org/10.1016/S0065-2628(08)60258-X).
- [55] S. Ali, M.A. Ullah, A. Nawaz, S. Naz, A.A. Shah, G. Gohari, et al., Carboxymethyl cellulose coating regulates cell wall polysaccharides disassembly and delays ripening of harvested banana fruit, *Postharvest Biol. Technol.* 191 (2022) 111978, <https://doi.org/10.1016/j.postharvbio.2022.111978>.
- [56] S. Moser, T. Müller, A. Holzinger, C. Lütz, S. Jockusch, N. Turro, et al., Fluorescent chlorophyll catabolites in bananas light up blue halos of cell death, *PNAS* 106 (2009) 15538–15543, <https://doi.org/10.1073/pnas.0908060106>.
- [57] M. Chen, H. Gu, L. Wang, Y. Shao, R. Li, W. Li, Exogenous ethylene promotes peel color transformation by regulating the degradation of chlorophyll and synthesis of anthocyanin in postharvest mango fruit, *Front. Nutr.* 9 (2022) 911542, <https://doi.org/10.3389/fnut.2022.911542>.
- [58] A.A. Kader, D. Zagory, E.L. Kerbel, Modified atmosphere packaging of fruits and vegetables, *Crit. Rev. Food Sci. Nutr.* 28 (1989) 1–30, <https://doi.org/10.1080/1040398909527490>.
- [59] P. Brat, M. Lechaudel, L. Segret, R. Morillon, O. Hubert, O. Gros, et al., Post-harvest banana peel splitting as a function of relative humidity storage conditions, *Acta Physiol. Plant.* 38 (2016) 234, <https://doi.org/10.1007/s11738-016-2253-0>.
- [60] M. Duca, Plant Respiration. *Plant Physiology. Biological and Medical Physics Biomedical Engineering*, 2015, pp. 123–148.
- [61] Z. Deng, J. Jung, J. Simonsen, Y. Zhao, Cellulose nanomaterials emulsion coatings for controlling physiological activity, modifying surface morphology, and enhancing storability of postharvest bananas (*Musa acuminata*), *Food Chem.* 232 (2017) 359–368, <https://doi.org/10.1016/j.foodchem.2017.04.028>.
- [62] FAOSTAT, Food and Agriculture Organization of United Nations, FAOSTAT Data 2023, <http://www.fao.org/faostat/en/#data>.
- [63] TAPPI, Technical Association of the Pulp and Paper Industry Standard Method, *UM 250, Acid-Soluble Lignin in Wood and Pulp*, TAPPI Press, Atlanta, GA, 1991.