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Antioxidant-Incorporated Poly(vinyl alcohol) Coating: Preparation, Characterization, and Influence on Ripening of Green Bananas

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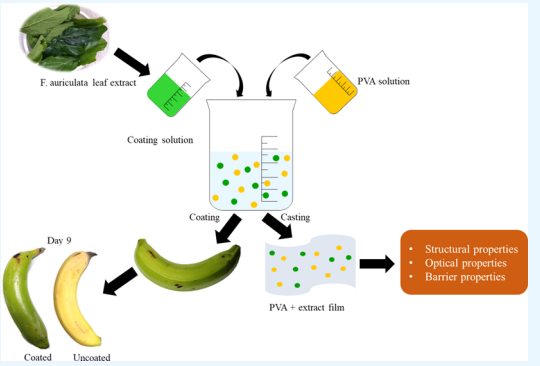
ABSTRACT: In this study, the gallic acid (antioxidant)-rich leaf extract of *Ficus auriculata* was incorporated into poly(vinyl alcohol) (PVA) and utilized as a coating to delay the ripening of green bananas. The films exhibited low opacity of 0.86 ± 0.014 for pure PVA (PP) and 0.92 ± 0.019, 0.99 ± 0.020, and 1.18 ± 0.029 for PVA + 1% extract (PE1), PVA + 5% extract (PE5), and PVA + 10% extract (PE10), respectively, indicating excellent transparency. The weight loss was higher in the uncoated group than in any coated fruits. The reduction in titratable acidity and the increase in total soluble sugars were slower in all of the coated samples as compared to the uncoated ones. The fruits without any treatment attained complete maturity on the ninth day where the ion leakage was 85.61 ± 2.33% while that of PP was 56.36 ± 2.95% and those of PE1, PE5, and PE10 remained below 30%. The coated samples showed better retention and consequently slower degradation of chlorophyll. The fruits coated with pure PVA as well as 10% extract-incorporated PVA remained acceptable till day 15, while the ones with 1 and 5% of extract reached full ripeness on day 18. Results of the present investigation suggest that safe, low-cost, and environmentally friendly coatings can improve the shelf life of perishable produces like bananas.



1. INTRODUCTION

Banana is among the most widely consumed and popular fruits in the world that act as a staple food for many communities owing to its nutritional content and taste.[1] Banana is a climacteric fruit with a high moisture content, which makes it highly perishable due to rapid postharvest physicochemical changes.[2] Despite the large popularity, the low shelf life of bananas poses a significant commercial loss, especially where provisions for low-temperature storage are absent or scarce.[3] The commonly practiced postharvest treatment of bananas includes controlled atmosphere storage, low-temperature storage, and application of ethylene inhibitors.[4,5] However, their application may be limited by chilling injury resulting from cold storage, nonuniform ripening, and high processing cost, among others, which make them infeasible for the majority of the banana-producing population.[5] This necessitates the development of a simple and economical yet effective technique for the preservation of bananas and other perishable products.

The application of a coating to extend the commercial shelf stability of various types of foods has gained substantial research interest in the recent past. They are particularly successful in the preservation of fresh fruits. Coatings aid in improving the shelf stability of fruits by creating a semipermeable barrier and a modified atmosphere with a controlled exchange of moisture and metabolic gases such as oxygen, carbon dioxide, and ethylene. This consequently prolongs the shelf life by delaying the ripening and inhibiting the activity of spoilage-causing microorganisms.[6] Hence, coating using different safe and biocompatible materials has been successfully explored to improve the shelf stability and bioactivity of different fruits like pears,[7] bananas,[6,8] avocados,[9] plums,[10] and strawberries.[11] Similarly, to prolong the shelf life of bananas, several types of coatings such as carrageenan, starch, and chitosan have been studied.[12,13] The use of PVA coating that incorporates *Sonneratia ovata* extract and is based on a heterojunction catalyst composite has also been reported for bananas.[14,15] The nature-based coatings are safer compared to their chemical counterparts and are reported to maintain better freshness and keep the quality of agricultural produces.[10]

Poly(vinyl alcohol) (PVA) is a biocompatible and biodegradable polymer with outstanding physical, optical, mechanical, and film-forming properties in addition to excellent chemical resistance.[15] PVA and its composites have successfully been studied for their potential applications in food packaging systems.[16] Considering its nontoxic and safe profile, it has earned the generally recognized as safe (GRAS) status, promising its use for the production of edible films.[17]

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The incorporation of bioactive components such as phenolic compounds has substantial potential for applications in food packaging.[18] Gallic acid is a water-soluble phenolic compound that has been ascribed with potent antioxidant and antimicrobial properties. The enhancement of the antioxidant properties has been reported in gallic acid-incorporated PVA films.[19] Thus, from the available literature, it is envisaged that the incorporation of gallic acid-rich extract in PVA coatings can be beneficial for delaying the ripening of bananas. The leaf extract of *Ficus auriculata* is found to be rich in gallic acid and shows high antioxidant activity.[20] This shows that the extract could potentially be used as an additive to enhance the shelf life of fresh produces. However, no work has been reported regarding the use of *F. auriculata* leaf extract for extending the shelf life of fruits.

Despite its desirable properties and safety, the application of PVA for the direct coating of bananas is scant. In this work, we propose to use PVA as a surface coating for enhancing the shelf life of bananas by delaying the ripening process. Gallic acid-rich extract of *F. auriculata* leaves was incorporated as an additive to augment the functional properties of the coating. The coatings were characterized by various characteristics that affect the efficiency of their use as coating for bananas. Insights into the effect of different types of coatings on the change in physicochemical attributes of bananas during storage were critically analyzed. We envision that this work will be helpful in the design and application of low-cost and easy coating systems to boost the shelf life of perishable fruits.

2. MATERIALS AND METHODS

2.1. Materials. *F. auriculata* leaves were collected from the IIT Guwahati campus. Poly(vinyl alcohol) (PVA) (M.W. 1,15,000) was obtained from Loba Chemie (Mumbai, India). Sodium hydroxide (NaOH) was obtained from Merck, India. Water from the Millipore Milli-Q purification system was used in all of the experiments. Green banana samples were collected from a local market. They were thoroughly washed to remove any adhered dust and dirt particles and air-dried.

2.2. Preparation of Extract and Coating Solution. The gallic acid-rich extract from *F. auriculata* leaves was prepared using an optimized protocol from our previous work.[20] Briefly, leaf powder and slightly alkaline water at pH 8 were mixed in a ratio of 1:10 g mL^{-1}. The mixture was subjected to ultrasound-assisted extraction at 50 °C and 50% sonication power for 30 min using a sonication bath (P 30H, Elmasonic). The concentration of gallic in the extract from *F. auriculata* leaf was found to be 312.9 mg L^{-1}. The antioxidant activity of the gallic acid-enriched extract was 91.7%.[20] The coating solution was prepared by mechanical mixing of PVA solution and the extract at different concentrations and denoted as PP (pure PVA), PE1 (PVA + 1% extract), PE5 (PVA + 5% extract), and PE10 (PVA + 10% extract) (Table S1).

The 10% PVA solution was first prepared by heating the mixture at 90 °C under stirring for 6 h. Following the complete dissolution, the solution temperature was brought down to 50 °C and the extract of *F. auriculata* leaves was added to the PVA solution on the basis of the total coating solution. The solution was mixed for another 30 min at 50 °C. The final blend was centrifuged at 10,000 rpm for 10 min to remove any undissolved particles or gas bubbles. The green banana samples with intact peel were then coated with the prepared solution by dipping the fruits in the solution for 30 s. The coated samples were separately grouped according to the composition of the coating solution. The bananas were then removed and placed on a metal grid until the solution stops dripping. The samples were then transferred to a chamber maintained at 25 ± 1 °C where the study continues. Three samples were removed from each group every third day to measure various physicochemical properties until full ripening. To allow the characterization of the coating surface, the solution was cast using a casting knife on clean glass plates to obtain films containing different extract concentrations. The films were then dried overnight under ambient conditions and dried through assisted airflow in a laminar hood.

2.3. Characterization of the Films. *2.3.1. Structural Properties.* The morphology of the films was visualized using a field emission scanning electron microscope (FESEM) (Zeiss, Sigma). The images of the surface of the films were recorded at 2000−3000 V using an Inlens detector. The surface roughness was examined with the help of an atomic force microscope (AFM) (Innova, Bruker). The microscopic images were obtained at a 1 Hz scan rate. The functional groups present on the different films were analyzed using attenuated total reflectance−Fourier transform infrared (ATR−FTIR) spectroscopy (IRAffinity-1, M/s Shimadzu, Japan). The spectra were obtained by scanning the films over a range of 4000−400 cm^{-1} wavenumber with an average of 30 scans per sample. The crystallography of the films was studied with the help of an X-ray diffractometer (Bruker D8 Advance) provided with Cu Kα radiation, and the diffractogram was recorded in the 2θ range between 5 and 60°. The surface chemistry of the films was further elucidated using X-ray photoelectron spectroscopy (XPS). The spectrometer (ESCALAB Xi+, Thermo-Scientific) was operated in the constant analyzer energy (CAE) mode using a monochromatic Al Kα X-ray source with a spherical energy analyzer.

2.3.2. Optical Properties. The optical property of the film in terms of opacity was analyzed using the technique adopted by Cazón et al.[21] with minor changes. The absorbance of the films at 500 nm was recorded by a UV−vis spectrophotometer (UV-2600, Shimadzu, Singapore). The opacity (A mm^{-1}) was calculated using eq 1

$$\text{opacity} = \frac{\text{Abs}_{500}}{x} \tag{1}$$

where Abs_{500} is the absorbance at 500 nm (A) and x is the film thickness (mm).

2.3.3. Thermal Properties. The thermal properties of the films were investigated utilizing a thermogravimetric analyzer (TGA) (TG 209 F1 Libra, Netzsch, Germany). Around 7 mg of films was used for each analysis and the films were subjected to heat treatment by heating from 20 to 800 °C at a rate of 10 °C min^{-1} under a nitrogen environment to maintain an inert condition. The graph indicating the change in mass with an increase in temperature was plotted.

2.3.4. Water Solubility (WS) and Water Vapor Permeability (WVP). The water solubility of the films was measured using the protocol adopted in a previous study[22] with minor modifications. For this, films of 2 × 2 cm^2 were cut and dried at 50 °C for 24 h to evaporate any residual moisture. The initial weight of the films (W_1) was noted, and they were placed in a 100 mL beaker containing 50 mL of distilled water and allowed to stand for 24 h at ambient temperature. After 24 h, the films were dried again at 50 °C for 24 h to remove the absorbed moisture and the final weight was recorded (W_2). The water solubility of the films was deliberated using eq 2.

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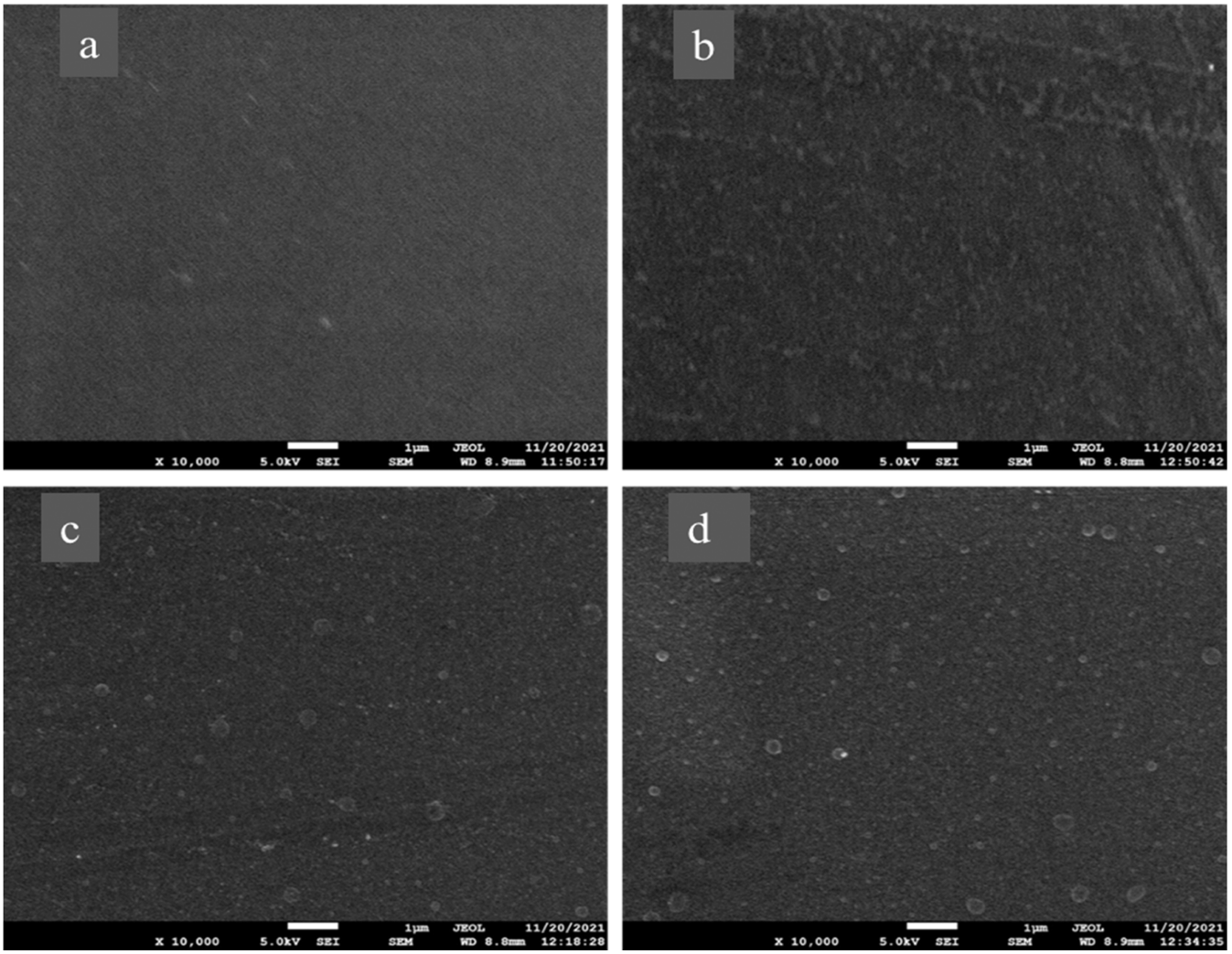

Figure 1. FESEM images of films: (a) PP, (b) PE1, (c) PE5, and (d) PE10.

$$\text{solubility}\ (\%) = \frac{W_1 - W_2}{W_1} \times 100 \tag{2}$$

The barrier property measured through water vapor transmission rate and permeability (WVTR and WVP) was determined using the protocol followed in a previous study[23] with slight modifications. Briefly, glass cups half-filled with calcium chloride were covered using the pure and blended films and placed inside a desiccator. A relative humidity (RH) of 75.2 ± 0.17% was maintained inside the desiccator using a saturated sodium chloride solution.[24] The transport of water vapor through the films was obtained by measuring the weight gain of the cups at an interval of 24 h for 7 days, and the weight gain was plotted against time. The WVTR was determined from the slope obtained from the linear regression by dividing the slope by the area of vapor transfer (in m^2). The WVP (g $m\ h^{-1}\ Pa^{-1}$) was estimated using eq 3.

$$\text{WVP} = \frac{\text{WVTR} \times x}{P(R_1 - R_2)} \tag{3}$$

where P is the saturation vapor pressure of water at ambient temperature, R_1 and R_2 are the relative humidity values in the desiccator and the cup, respectively, and x is the thickness of the film (m). $[P(R_1 - R_2)]$ is the driving force for the moisture transfer and is 2376.30 Pa under these conditions.

2.4. Physicochemical Analyses of Fruit. 2.4.1. Weight Loss. Weight loss is brought by moisture loss from the banana samples, which in turn influences the appearance and acceptability of the fruits. The weight of the samples from different sample groups was recorded every day using an analytical weighing balance. The average values of the weight change are presented as follows

$$\text{WL}\ (\%) = \frac{W_0 - W_i}{W_0} \times 100 \tag{4}$$

where W_0 and W_i are the initial and weights at sampling time, respectively.

2.4.2. Titratable Acidity (TA). For the measurement of TA, the banana pulp was first blended in a mixer grinder to get a homogeneous paste. This was centrifuged at 10,000 rpm for 10 min to get a clear juice free from the pulp.

TA was determined using the protocol followed in a previous study.[25] Briefly, 1 mL of juice was mixed with 9 mL of distilled water and the diluted juice was titrated against a 0.1 M NaOH solution to a final pH of 8.1. Three drops of phenolphthalein indicator were added to identify the endpoint. Malic acid is the predominant organic acid present in bananas and hence calculations for titratable acidity were made based on this acid.

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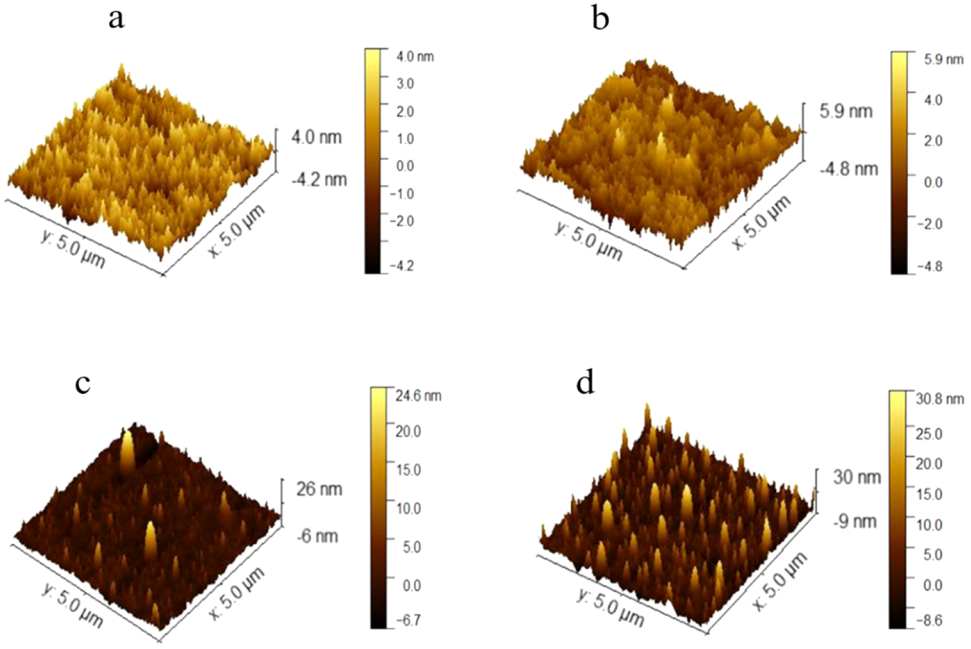

Figure 2. AFM images of films: (a) PP, (b) PE1, (c) PE5, and (d) PE10.

The TA denoted as grams of malic acid per 100 g of fruit juice was calculated using eq 5.

$$\text{TA}\ (\%) = \frac{V_{\text{NaOH}} \times 0.1 \times 0.067}{V_{\text{sample}}} \times 100 \quad (5)$$

where V_{NaOH} is the volume of NaOH used to bring the pH to 8.1, 0.1 is the molarity of the NaOH solution, V_{sample} is the volume of the juice taken, and 0.067 is the conversion factor for malic acid.

2.4.3. Total Soluble Solids (TSSs). TSS is a vital parameter for the determination of the quality of bananas since it indicates the level of sweetness. The TSS of the sample on each collection day was measured using a portable refractometer (Erma, Japan). One to two drops of the homogenized juice were placed on the sampler and the reading was noted, and the TSS was presented as °Brix.

2.4.4. Chlorophyll Content. The chlorophyll content of the coated and uncoated banana peels was measured in intervals of 3 days until the completion of the study. For this, 0.1 g of peel free from pulp and fibers was mixed with 80% acetone using a mortar and pestle for 3−5 min. The chlorophyll solution was filtered with Whatman 1 filter paper and the absorbance of the filtrate was measured with a UV−vis spectrophotometer (UV-2600, Shimadzu, Singapore). The chlorophyll content of the peels was determined using eqs 6−9.[8]

$$\text{chlorophyll}\ a = 12.7 \times A_{663} - 2.995 \times A_{645} \quad (6)$$

$$\text{chlorophyll}\ b = 22.95 \times A_{645} - 4.67 \times A_{663} \quad (7)$$

$$\text{total chlorophyll}\ (\text{mg L}^{-1}) = \text{chlorophyll}\ a + \text{chlorophyll}\ b \quad (8)$$

$$\text{total chlorophyll}\ (\text{mg g}^{-1}) = \frac{\text{total chlorophyll}\ (\text{mg L}^{-1}) \times 25\ \text{mL}}{\text{sample weight} \times 1000} \quad (9)$$

2.4.5. Ion Leakage. The leakage of the electrolytes from the banana peels represented by the membrane stability index (MSI) was measured using the protocol followed in a previous study[8] with a slight alteration. Briefly, 3 g of sliced banana peel free from the pulp was taken in a 50 mL beaker containing 30 mL of deionized water. The mixture of peel and water was kept covered at room temperature and stirred at 150 rpm for 4 h. The conductivity of the solution (k_1) was measured at the end of 4 h using a conductivity meter (CON 2700, Eutech Instruments). Then, the beaker was held in a water bath at 100 °C for 30 min to facilitate the release of the electrolytes. Then, the mixture was cooled to room temperature, followed by the conductivity measurement (k_2). The MSI was determined using the following relation

$$\text{ion leakage}\ (\%) = \frac{k_1}{k_2} \quad (10)$$

2.5. Statistical Analysis. The results were presented as mean ± standard deviation (SD) obtained from three replicates. The one-way analysis of variance (ANOVA) and Tukey's test were performed to determine the statistical difference between different study groups for each sampling day. The test was carried out using OriginPro 9, and the results were considered to be significant at $p < 0.05$.

3. RESULTS AND DISCUSSION

3.1. Structural and Functional Analysis of Films. It can be seen from the FESEM images (Figure 1) that the film prepared by PP results in a uniform and homogeneous surface with no significant defect. This signifies the good film-forming property of the polymer during casting. The addition of the leaf extract affected the homogeneity of the polymer solution at all concentrations. However, the extract seems to be well dispersed in the solution owing to the good hydrophilicity of the phytochemicals present in the extract. Increasing the concentration of the extract enhances the formation of porous-like structures that spread throughout the surface, resulting in a rough surface. This may be ascribed to the rearrangement of

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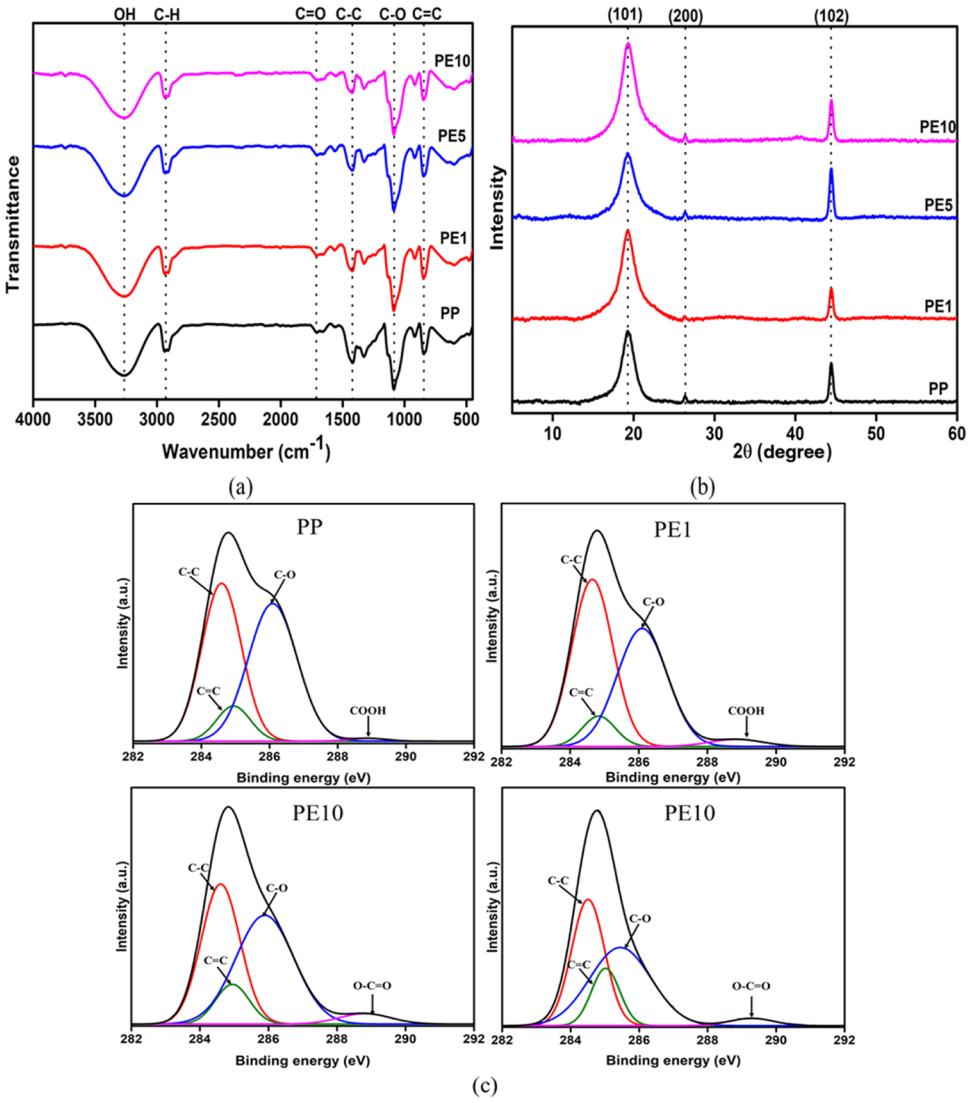

Figure 3. (a) FTIR spectra, (b) X-ray diffractogram, and (c) deconvoluted C 1s XPS spectra of pure and blend films.

the water-soluble components like gallic acid during the evaporation of water during film preparation.[26]

It was observed from AFM images (Figure 2) that the addition of extract increased the surface roughness consistent with the concentration. At a lower concentration of 1% (PE1), the average roughness increased just slightly to 1.286 nm from 1.073 nm of the PP film. This indicates a uniform mixing of the extract components with the polymer matrix at the molecular level in low concentrations. However, as the extract increased from 5% (PE5) and 10% (PE10), the average surface roughness increased to 2.475 and 4.535 nm, respectively. This is presumably due to the dispersion and aggregation of the compounds present in the extract leading to an increased roughness.[27] In addition, the evaporation of water from the nonhomogeneous regions may lead to increased surface roughness. The findings corroborate the FESEM results.

The functional groups responsible for the chemical interactions in the films determined through FTIR (Figure 3a) show a peak at 3265 cm^{-1}, which is allotted to the O−H stretching of the alcohol and carboxylic acid. However, this peak shifts slightly toward the left with a marginal increase in the width with the addition of the extract. This may be elucidated by the establishment of hydrogen bonds subsequent to the interaction between the hydroxyl groups of PVA and the phenolic such as gallic acid that are present in the extract.[28] The peaks at 2924 and 1710 cm^{-1} may correspond to the C−H stretching as well as the C═O stretching of gallic acid ester, respectively.[28] The peaks at 1420, 1080, and 840 cm^{-1} can be attributed to C−H vibration, C−O stretching, and C═C bending, respectively.[29,30]

The diffractogram of the films obtained from the XRD analysis (Figure 3b) shows the crystal structure. The peaks at 2θ = 19.28, 26.38, and 44.4° are characteristic of PVA and correspond to the crystal planes of (101), (200), and (102), respectively.[31]

The peak position corresponding to the (101) plane is attributed to the hydrogen bonding resulting from the molecular chains within or between PVA. The addition of extract did not cause any shift in the peak position or any new peak, indicating that the crystal structure remains unaffected. There is an intensification of the peak with the addition of the extract. This may be explicated by the hydrogen bonding between PVA and the constituents of the extract that improves the ordering arrangement of PVA molecules.[32] The position of the PP peaks remains unchanged in all of the blend films

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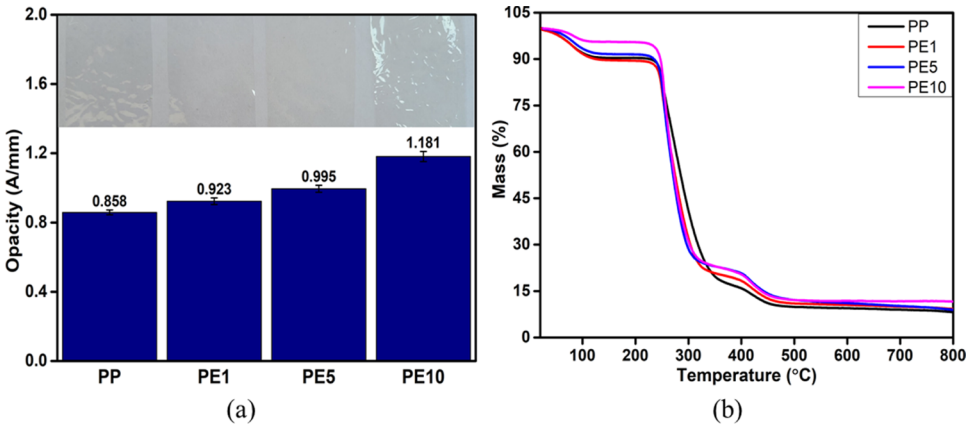

Figure 4. (a) Opacity of the films. (b) TGA curve of the films. Opacity values are mean ± SD obtained from three replications.

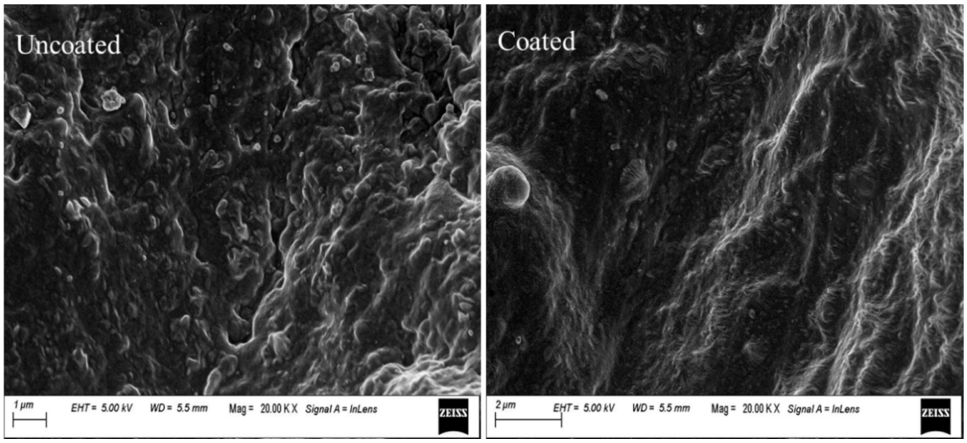

Figure 5. FESEM images of control and coated banana peels on day 9.

extract, which indicates that there was no significant change in the lattice distance resulting from the blend.[33]

XPS was used to further comprehend the surface chemistry of the pure and extract-incorporated PVA films (Figure 3c). The deconvoluted C1s spectra depicted four major surface components corresponding to binding energies of 284.5, 284.9, 286.09, and 288.8 eV that can be attributed to C−C, C═C, C−O, and O−C═O, respectively.[34] The binding energies shift positively with the increasing concentration of the extract, which might be due to the modification of the PVA surface. This could be ascribed to the strong interaction between the increasing concentration of extract and PVA.[35] The addition of extract resulted in a change in the carbon−oxygen ratios as observed from the survey plots (Figure S1). The atomic percentage of carbon increased from 62.3% in PP to 62.6, 65.4, and 71% in PE1, PE5, and PE10, respectively (Table S2). This may be attributed to the increase in carbon moieties present in the constituents of the extract.

3.2. Optical and Thermal Properties of Films. The transparency and opacity of edible films is an important parameter as it allows the visual observation of the foods being treated at any instance. The films obtained from all of the treatment groups resulted in low opacity, indicating excellent transparency (Figure 4a). The addition of extracts caused a slight increase in the opacity of the films with 0.86 ± 0.014 for PP and 0.92 ± 0.019, 0.99 ± 0.020, and 1.18 ± 0.029 for PE1, PE5, and PE10, respectively. Despite this, the final films resulted in good transparency as can be seen from the images taken with a white background.

The thermogravimetric analyzer was used to observe the thermal property of the PVA-based films under inert conditions (Figure 4b). The first degradation was observed in the range of 60−120 °C, which may be attributed to the removal of weakly bound surface moisture. This is followed by a sharp degradation (230−345 °C), which corresponds to the starting of the degradation of the main polymer chain. The final degradation stage (400−460 °C) represents the splitting of the C−C backbone of PVA.

The initial decomposition temperature in the second degradation stage as well as the residual mass is slightly higher in the case of extract-incorporated films. This may be caused by the formation of hydrogen and covalent bonds between PVA and the polyphenols present in the extract, resulting in a delay in the decomposition of main polymer chains.[36]

3.3. Solubility and Water Vapor Permeability. The WS and WVP of the films are important parameters in determining the exchange of moisture between the sample and the environment that affects the quality of a commodity. The WVP of the PP film was lesser than the films incorporated with extract at all concentrations (Table S3). The WVP was the least for PP ($1.99 \pm 0.72 \times 10^{-7}$ g m h^{-1} Pa^{-1}), while that of PE1, PE5, and PE10 were 2.28 ± 0.08, 2.31 ± 0.17, and 2.35 ± 0.33×10^{-7} g m h^{-1} Pa^{-1}, respectively. This can be attributed to an increase in void volume as a result of the destruction of the compact film network brought about by the aggregation of

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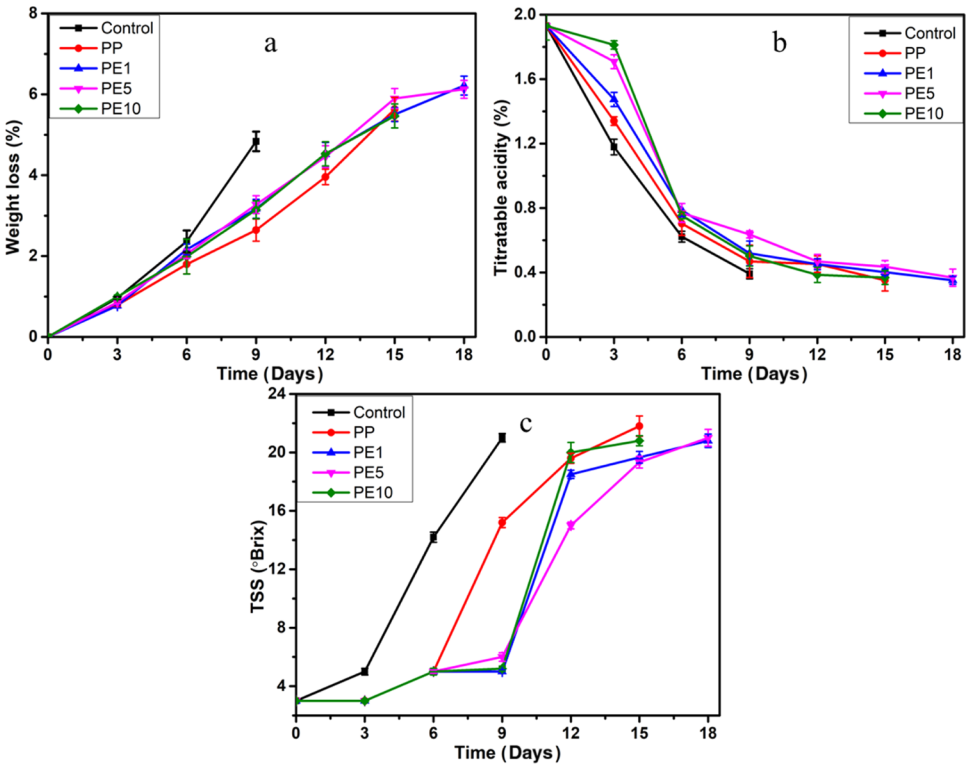

Figure 6. Physicochemical properties of bananas during storage. (a) Weight loss percent, (b) TA, and (c) TSS. The values are mean ± SD obtained from three replications.

the constituents of the extract.[37] This can consequently influence the channel of water vapor through the films.[36] The WS of the films increased from 33.19 ± 1.48% for PP to 37.59 ± 1.88, 40.25 ± 1.33, and 46.49 ± 2.39% for PE1, PE5, and PE10, respectively. The increase in solubility with the increasing concentration of extract may be caused by the interaction between the hydrophilic groups of the phytochemicals present in the films with water molecules. This, in turn, allows the film network to bind more intensely with water and thereby improves the hydrophilicity of the blend.[37] These findings are supported by the microscopic images of the films (Figure 1).

3.4. Physicochemical Analysis of Bananas. The assessment of various physicochemical properties allows us to track the ripening of bananas during the storage period. The FESEM image of an uncoated peel shows a significantly higher roughness than the coated one (Figure 5). On the other hand, the peel coated with a polymer solution retained its structural integrity much more than the control. It can be concluded from this that the treatment improves the stability of the banana peels, which helps in delaying the ripening and consequently enhances the storage life.

3.4.1. Weight Loss, TA, and TSS. The change in physicochemical properties of bananas in terms of weight loss, TSS, and TA during storage is presented in Figure 6. The quality and value of fresh produce during postharvest storage can be greatly influenced by the loss of moisture resulting in weight reduction. The surface coating of these produces with an edible film can minimize water loss, thereby enhancing the shelf life.[38] The trend of the reduction in the weight of banana fruits during the storage period was measured every day (Figure 6a). The percentage of weight loss was identical for all of the sample groups up to the first 3 days. This may be influenced by water evaporation from the film surface, resulting in a decrease in weight. The visible change was observed starting from the fourth day when the weight loss was visibly higher in the uncoated group than in any coated fruits. The weight loss continued during the study period, where the control fruit experienced a weight loss of 4.84 ± 0.24% on day 9, while the weight loss of PP, PE1 PE5, and PE10 was 2.64 ± 0.27, 3.17 ± 0.23, 3.27 ± 0.22, and 3.15 ± 0.2%, respectively. The reduced weight loss in coated samples indicates that the semipermeable PVA-based coating could effectively reduce the moisture loss and mass transfer from the sample surface.[39] Among the coated samples, the weight loss was higher in the samples with extract-incorporated coatings, as compared to PP. This may be could be attributed to the increased WVP as explained in Section 2.3.4. The control reached maximum stage marketability owing to the full ripeness on the ninth day; hence, it was not analyzed further. The coated fruits, however, showed a slower ripening and were analyzed till day 18. The slower ripening and weight loss from the coated fruits indicate the effectiveness of the semipermeable film in mass transfer reduction.[8] The weight loss in the fruit coated with the PP solution was lesser than the ones containing extract at all concentrations. This may be explained by the disturbance of film integrity caused by the aggregation of the extract constituents, allowing moisture to pass through more freely as explained in the preceding section.

The TA of all the sample groups showed a declining trend throughout the storage period (Figure 6b). The reduction in TA of bananas during storage is greatly influenced by the metabolic activities occurring continuously. Organic acids are utilized during metabolic processes like respiration and result in a decreased acidity of the fruit with time.[40] This reduction can thus be correlated with the various changes in a banana

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Table 1. Coated and Uncoated Bananas during Storage at 25 ± 1 °C[a]

Group/Day	Control	PP	PE1	PE5	PE10
0					
3					
6					
9					
12	X				
15	X				
18	X	X			X

[a]X indicates that the sample group has attained full maturity and is not analyzed further.

during ripening and storage. In addition, organic acids are converted into sugars during ripening, resulting in the declination of TA.[41] The slower declination in the TA of coated samples can be ascribed to the inhibition of respiration rate caused by the reduced O_2 transfer through the barrier film.[42]

The TSS of all of the samples increased throughout the storage period. The change is more pronounced in the control samples where the TSS increased from 3 °Bx on day 0 to 5 ± 0.23, 14.2 ± 0.35, and 21 ± 0.29 °Bx on days 3, 6, and 9, respectively (Figure 6c). The control fruit attained maturity on day 9, while the TSS remained 15.2 ± 0.35, 5 ± 0.12, 6 ± 0.29, and 5.2 ± 0.17% for PP, PE1, PE5, and PE10, respectively. PP reached a maximum TSS of 21.8 ± 0.69 °Bx on day 15, while PE10 attained a maximum value of 21 ± 0.35 °Bx on the same day. Similarly, both PE1 and PE5 attained maximum acceptable ripening on day 18 with TSS of 20.8 ± 0.46 and 21 ± 0.58 °Bx, respectively. This increase in TSS in all sample groups can be attributed to the hydrolysis of starch and pectic substances present in the pulp into soluble sugars.[43]

The rate of change of various physicochemical properties is lower in the coated samples as compared to the uncoated ones, where the coatings with extract showed better retention of quality for a longer period. The bioactive compounds present in the extract are secondary metabolites synthesized by plants as a defense mechanism against biotic and abiotic stresses.[9] These compounds are efficient antioxidant and antimicrobial agents that have been associated with delaying and decreasing fruit decay.[44] In addition, certain phenolic compounds are reported to inhibit or delay the biosynthesis of ethylene, the modulator of fruit ripening.[45] In addition, some phenolic compounds are reported to reduce the presence of enzymes such as catalase, which play an important role in fruit senescence and ripening.[46,47] Previous studies have hypothesized that antioxidant enzymes such as superoxide dismutase, catalase, and peroxidase play an important role in both senescence and fruit ripening. This has been attributed to the association between ripening-related gene expression and oxidative stress response.[10] This could eventually result in a delay in ripening the extract-incorporated coatings.

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Table 2. Change in Ion Leakage and Chlorophyll Content of Peels during Storage at 25 ± 1 °C[a]

	days						
treatment	0	3	6	9	12	15	18
				Ion Leakage (%)			
control	15.15 ± 1.18^{a}	28.58 ± 3.40^{acd}	53.57 ± 2.82^{a}	85.61 ± 2.33^{a}	NA	NA	NA
PP	15.15 ± 1.18^{a}	14.70 ± 2.32^{b}	21.43 ± 1.42^{b}	56.36 ± 2.95^{b}	61.88 ± 3.03^{a}	85.09 ± 2.15^{a}	NA
PE1	15.15 ± 1.18^{a}	19.34 ± 1.71^{bc}	27.92 ± 4.08^{b}	20.81 ± 2.61^{cd}	49.14 ± 3.44^{b}	82.78 ± 3.00^{a}	85.14 ± 2.36^{a}
PE5	15.15 ± 1.18^{a}	21.13 ± 1.76^{bc}	32.48 ± 1.43^{b}	23.16 ± 2.37^{d}	52.95 ± 1.68^{ab}	82.80 ± 2.76^{a}	86.09 ± 2.28^{a}
PE10	15.15 ± 1.18^{a}	24.46 ± 1.80^{bd}	25.19 ± 2.84^{b}	26.50 ± 2.57^{d}	57.22 ± 2.15^{ab}	91.23 ± 1.78^{a}	NA
				Total Chlorophyll (mg g^{-1})			
control	0.20 ± 0.01^{a}	0.17 ± 0.01^{c}	0.16 ± 0.01^{b}	0.05 ± 0.01^{c}	NA	NA	NA
PP	0.20 ± 0.01^{a}	0.24 ± 0.00^{b}	0.23 ± 0.01^{c}	0.08 ± 0.01^{b}	0.06 ± 0.00^{b}	0.04 ± 0.01^{ac}	NA
PE1	0.20 ± 0.01^{a}	0.20 ± 0.01^{d}	0.18 ± 0.00^{b}	0.17 ± 0.01^{a}	0.10 ± 0.01^{a}	0.07 ± 0.01^{a}	0.05 ± 0.01^{a}
PE5	0.20 ± 0.01^{a}	0.26 ± 0.01^{ab}	0.21 ± 0.01^{ac}	0.19 ± 0.01^{a}	0.08 ± 0.01^{a}	0.05 ± 0.01^{ad}	0.04 ± 0.01^{a}
PE10	0.20 ± 0.01^{a}	0.27 ± 0.00^{a}	0.20 ± 0.01^{a}	0.20 ± 0.01^{a}	0.08 ± 0.01^{a}	0.03 ± 0.01^{bcd}	NA

[a]The values are means ± standard deviation for three replicates. Means sharing the same letters within columns are not significantly different. NA, not analyzed.

The progression of ripening of the study groups of bananas can be visualized from the digital images as shown in Table 1. There was an apparent difference in the shelf life of the treated and untreated groups. These images prove that the coating of banana peels' surface improves the shelf life of bananas, as explained in the preceding and subsequent sections.

3.4.2. Ion Leakage and Chlorophyll Content. The change in ion leakage from fruit peels allows us to assess the viability of fruit peels as it indicates the integrity and stability of membranes.[38] Banana peels from all study groups lost integrity with ripening, resulting in 85.61 ± 2.33% ion leakage on day 9 for the uncoated group, while that of PP was 56.36 ± 2.95% and that of PE1, PE5, and PE10 remained below 30% (Table 2). The higher membrane stability in the coated fruits and more so in the groups containing the extract can be credited to the preservation of the peels' cell wall. This may be accredited to the high antioxidant activity of compounds like gallic acid present in the extract.[48] The findings indicated that the increase in ion leakage was considerably slowed down in the coated banana peels that allow the fruits to be stored for longer periods.[8,49]

The measurement of the total chlorophyll (Table 2) showed a higher content in the treated bananas, whereas the untreated ones retained a lesser concentration of the pigment. The better preservation of the green pigment in the coated bananas may be ascribed to the modification of the fruit atmosphere by the coating, which slowed down and delayed the ripening. Additionally, the antioxidative action of phenolic compounds like gallic acid may delay the oxidation and consequent degradation of chlorophylls. The findings of the current study are in agreement with previous works[42] where coating significantly affected the photosynthesis of pigments in bananas.

4. CONCLUSIONS

The application of PVA-based coating was successfully studied to delay the ripening and increase the shelf stability of green bananas. The incorporation of gallic acid-rich extract further improved the functionality and efficiency of PVA coatings. The addition of extract to PVA increased the water solubility and water vapor permeability of the films. The coatings remained sufficiently transparent after extract addition, allowing proper visualization of the coated sample. The fruits without any treatment attained complete maturity and thus commercial acceptability on the ninth day. The fruits coated with PP as well as PE10 remained acceptable till day 15, while the ones with PE1 and PE5 reached full ripeness on day 18. The data from physicochemical analyses of the pulp as well as the peels showed that there was continuous variation in the properties during storage; however, coated samples exhibited slower change. The investigation and the results obtained will help design simple and eco-friendly systems for improving the shelf life of commodities like bananas that are perishable in nature. This paper will be helpful to the researchers focusing on the utilization of natural-based coatings and packaging materials for food and allied applications.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.2c05271.

Composition of the coating groups (Table S1); atomic percentages of carbon and oxygen from XPS spectra (Table S2); water vapor permeability and water solubility of coating films (Table S3); survey spectra of (a) PP, (b) PE1, (c) PE5, and (d) PE10 (Figure S1) (PDF)

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Notes

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