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Enhanced Preservation of Climacteric Fruit with a Cellulose Nanofiber-Based Film Coating

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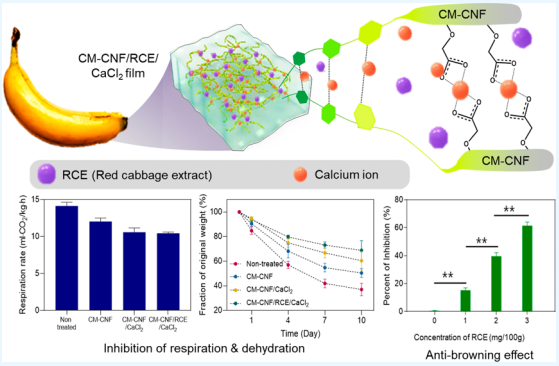
ABSTRACT: Bananas are a typical climacteric fruit with high respiration and ethylene production rates after harvest, and they show rapid ripening−senescence phenotypes. Here, we demonstrate that carboxymethylcellulose nanofibers (CM-CNFs) and red cabbage extracts (RCE) can be used as a unique film coating formulation for enhancement of the shelf-life of fruit. A CM-CNF suspension solution is created through a process involving chemical modification, followed by mechanical grinding. It has a high aspect ratio that allows for the creation of a thin and transparent film on the surface of bananas. The cross-linked CM-CNF hydrogel forms a dense film layer on the banana surface during dehydration and prevents respiration and weight loss. RCE contains polyphenols acting as antioxidants, which prevent the appearance of black dots on the banana peels. It serves to mitigate the browning of banana skins and also hinders the respiration process, consequently slowing the aging of bananas.



1. INTRODUCTION

Bananas are a representative climacteric fruit that is widely cultivated in tropical, subtropical climates and is one of the most consumed fruits for their sweet tastes and flavors in the world.[1,2] Since bananas have respiration and ethylene production rates much higher than those of other fruits after harvest, they can show rapid ripening−senescence phenotypes. Due to the release of ethylene gas and the weak physical properties, they are easily softened and damaged by physical pressure and chilling injuries.[3] Recently, the need for biodegradable packaging materials has been growing in the storage of fruits to increase shelf-life. Especially, the combination of biodegradable polymers and bioactive natural molecules provides the fruit coating with multifunctionality such as gas regulating, antibacterial, and ultraviolet (UV) barrier properties.[4−7] Despite the excellent barrier coating of fruits, biomass-based coating materials are still needed in terms of sustainable resources.

Cellulose nanofibers (CNFs) are based on the sustainable natural polysaccharide that is abundant in nature.[8] CNFs are considered for film coating of fruits due to their excellent mechanical properties, transparency, biodegradability, and chemical modifications.[9−11] Especially, carboxymethyl CNFs (CM-CNFs) are prospective in fruit coating due to the easy gel formation by electrostatic interaction with divalent cations.[12] The advantages of CM-CNF-based film coating are the natural resources and minimal chemical modification needed to produce the coating material. CNFs are based on the biomass in nature, and their nanofibrillation is mostly available by mechanical grinding. The hydrophilicity of CNFs enables the use of aqueous conditions in the process, and the protection performance can be enhanced by combining with additive molecules because CNFs retain the physical structure of fibers.

Red cabbage extracts (RCE) contain polyphenols, including anthocyanin, kaempferol, and quercetin, that function as antioxidants, increasing the shelf life of fruits.[13−16] Moreover, they can protect the plant cells, reducing the harm caused by intense sunlight.[17,18] Recently, a natural deep eutectic solvent formulation containing polyphenols has been used for food coating due to its remarkable antifrosting, antimicrobial and antioxidant properties.[19,20] A CM-CNF suspension with RCE can be used as a unique film coating formulation for antibrowning properties by protecting fruits from UV lights. The development of black spots observed during the storage of bananas is triggered by melanin production involving polyphenolic compounds and enzymes.[21−23] Essentially, melanin formation is initiated by the breakdown of cell walls, resulting from the generation of radicals and oxidation processes.[24,25] The UV protection offered by RCEs prevents the generation of radicals and oxidation within plant cells, thereby safeguarding the integrity of the cell walls.

We introduce an alternative to the film coating of fruits such as bananas with CM-CNFs containing RCE for maintaining the

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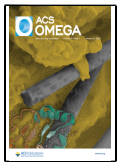

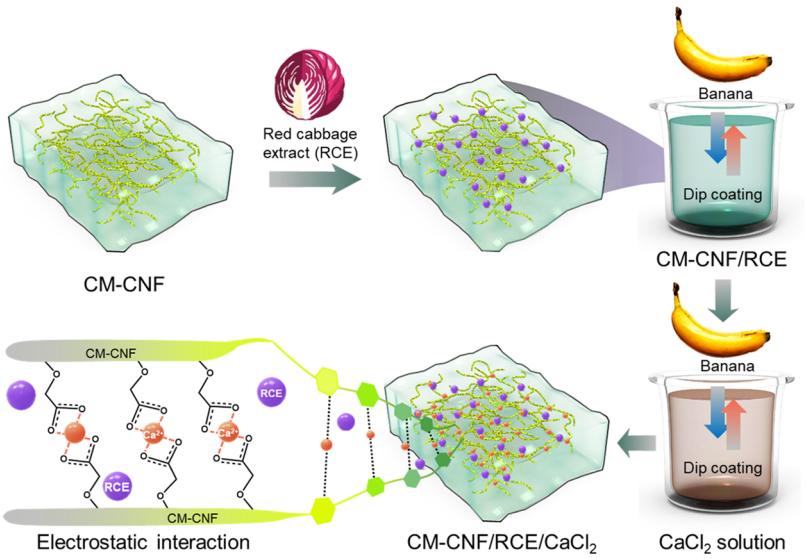

Figure 1. Procedure of CM-CNF/RCE/$CaCl_2$ film processing on the surface of bananas.

freshness. Due to the negativity of CM-CNFs, they can be cross-linked to form a hydrogel by treating with multivalent cations such as calcium ions (Figure 1). The cross-linked hydrogel forms a dense film layer on the fruit surface during dehydration and prevents respiration and weight loss. Besides, the antioxidant RCE in the film inhibits oxidation of the cell structure by suppressing radical formation; thus, it prevents the browning effect of bananas. The freshness of fruits is crucial in the food industry. These issues continue to be present in the traditional packaging approach for fruit preservation, and CM-CNF/RCE is one potential material for the enhancement of the shelf-life of fruits.

2. EXPERIMENTAL SECTION

2.1. Materials. Never-dried bleached eucalyptus kraft pulp (79.4 ± 0.6% cellulose, 18.8 ± 0.2% hemicellulose, and a small amount of lignin and byproducts) was supplied by Moorim P&P (Ulsan, Korea). Bananas and red cabbage (*Brassica oleracea L. var. capitata f. rubra*) were purchased from a local market. Methanol (99.5%, Samchun, Seoul, Korea), ethanol (anhydrous 99.9%, Samchun), isopropyl alcohol (99.5%, Samchun), sodium hydroxide (98%, Samchun), monochloroacetic acid (MCA ≥ 99.0%, Sigma-Aldrich, St. Louis, MO), sodium chloride (≥99.0%, Sigma-Aldrich), magnesium chloride (anhydrous ≥ 98.0%, Junsei, Tokyo, Japan), calcium chloride (anhydrous ≥ 97.0%, Sigma-Aldrich), 1,1-diphenyl-2-picrylhydrazyl (free radical ≥ 97.0%, TCI, Tokyo, Japan), and L-tyrosine (≥98.0%, Sigma-Aldrich) were also purchased.

2.2. Preparation of the CNF Coating Solution. First, the pulp was ripped by hand, which was beaten using a laboratory valley beater for about 30 min.[26,27] For carboxymethylation, the following process was carried out using beaten pulp. The wet pulp (dry weight, about 70 g) was solvent-exchanged to ethanol and immersed in isopropanol/methanol (4:1, 4000 mL) solution with sodium hydroxide (11.2 g) for mercerization. 14 g of MCA was added to the pulp slurry, and the solution was stirred for 90 min at 700 rpm. Subsequently, the pulp slurry solution was exchanged with deionized water at pH 7.0 and passed through a grinder (Super Masscolloider, Masuko Sangyo Co., Ltd., Japan) to produce the CM-CNF suspension. The operation speed was about 1500 rpm, and the gap distance of the grinder stones was 100 μm. The final concentration of the pulp suspension was about 1.5%.

The content of the carboxyl group was measured using the ionic conductivity method. 1 g of CM pulp in sodium form was converted to the proton form until all acid groups received hydrogen ions as counterions and was then titrated with 0.01 M NaOH. The total amount of carboxyl group and degree of substitution were calculated according to eqs 1 and 2[26,28]

$$\text{carboxyl group content (mmol/g)} = \frac{C_{NaOH} \times V_{NaOH}}{w} \quad (1)$$

$$\text{degree of substitution (\%)} = \frac{162 \times C_{NaOH} \times V_{NaOH}}{w - 80 \times C_{NaOH} \times V_{NaOH}} \times 100 \quad (2)$$

where C_{NaOH}, V_{NaOH}, and w are the concentration of the NaOH solution, the volume of the NaOH solution consumed over a flat area, and the oven-dried weight of the sample, respectively.

Fourier transform infrared spectroscopy (FTIR) (Nicolet iS5, Thermo Fisher Scientific, Waltham, MA) was measured to analyze the chemical structure of the CM-CNF films. Scanning was carried out in the range from 500 to 4000 cm^{-1} with 32 scans at a resolution of 4 cm^{-1}.

Nanoscale images of CM-CNFs were obtained by using a transmission electron microscope (TEM, JEM1010, JEOL, Tokyo, Japan). The CM-CNF suspension was dropped on a glow-discharged carbon copper grid, and the fibers were negatively stained with a 10 μL uranyl acetate solution (1% w/v). The dimension of CM-CNFs was analyzed using ImageJ software (1.52a NIH, Bethesda, Maryland).

2.3. Preparation of the CNF/RCE/$CaCl_2$ Film. Ten gram of red cabbage was added to a falcon tube, and then 30 mL of acidified ethanol (85 mL of ethanol and 15 mL of 1.5 mol/L HCl) was added and subjected to constant agitation for 1 h. The supernatant was separated by a filter manually. Finally, it was dried in a vacuum desiccator overnight.[29,30]

The 1% (w/w) CM-CNF solution containing 3% RCE was used as the coating material. Before being dipped into the CM-CNF solution, the bananas were rinsed with deionized water twice. Bananas were dipped into the CM-CNF solution and,

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ACS Omega 2024, 9, 1069−1076

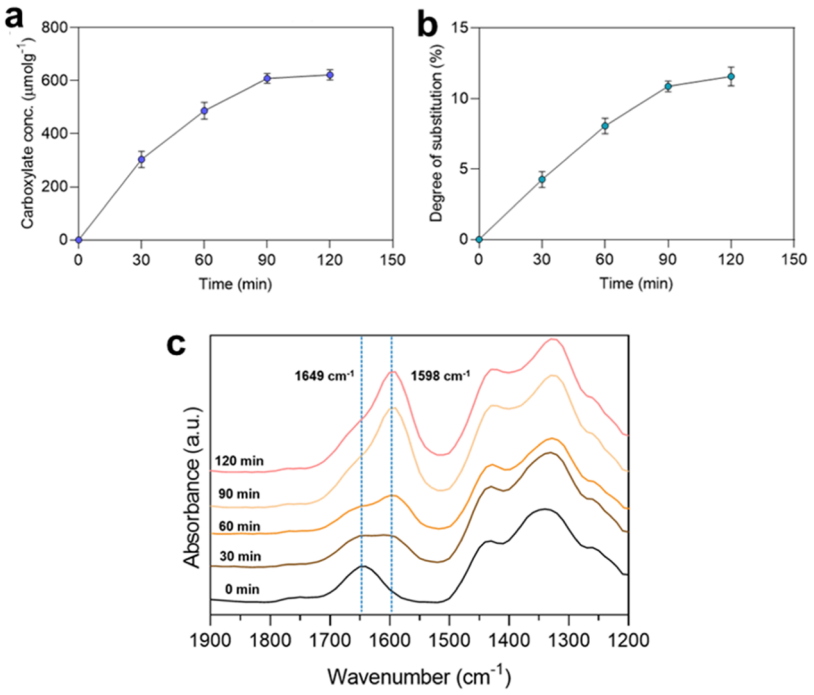

Figure 2. (a) Carboxylate concentration of pulp cellulose by carboxylation reaction with monochloroacetic acid. (b) Degree of substitution with reacting pretreatment. (c) FTIR spectrum of CM-CNF film over carboxylation time from 0 to 120 min.

subsequently, into 2% $CaCl_2$ solution for 3 min. The samples were dried at 25 °C overnight and stored at 25 ± 2 °C for 10 days.

2.4. Evaluation of CNF/RCE/$CaCl_2$ Film-Coated Bananas. 1% (w/w) CM-CNF solutions were poured into a mold of circular disk shape (diameter: 0.8 cm; thickness: 0.1 cm). And then, 2% of various metal ions in the aqueous solution, including NaCl, $MgCl_2$, and $CaCl_2$, were dropped into the prepared mold. After 10 min, the hydrogel specimens were immersed in PBS for 24 h. The water droplets on the surface of the samples were clearly absorbed by dried tissue. The rheological behavior of hydrogels was analyzed by using a digital rheometer (MARS III, Thermo Scientific, Newington, NH). Shear strain sweep oscillatory rheometry was performed using a parallel plate geometry (8 mm) with a gap size of 800 μm under a 0.001 to 0.1% strain range.

Bananas were weighed at the beginning of the experiment after air drying and thereafter each day during storage. Weight loss was expressed as the percentage loss of the initial total weight. The weight change was calculated using eq 3.[11]

$$\text{weight change (\%)} = \frac{(M_i - M_f)}{M_i} \times 100 \quad (3)$$

where M_i is the initial mass and M_f is the final mass of the bananas. It was expressed as a fraction of the original weight (%).

The firmness of the bananas was measured by using a penetrometer. A CT-3 texture analyzer (Brookfield Co., Middleborough, MA) was used for firmness measurements. Fruit firmness (N) was measured at the equatorial plane of the fruit using a flat probe with a 100 mm diameter at a speed of 2 mm/s and a strain of 5 mm.[31]

The respiration rate was determined by using static methods. A banana was stored in 1 L Nalgene polypropylene jars with a septum in the lid for sampling gas. The jars were stored at room temperature. Gas sampling was carried out after 2 h with 1 mL syringes by means of a needle connected to a gas chromatograph (YL 6400, Younglin Instrument, Seoul, Korea). Three replicates were performed for each sample. The respiration rate was calculated using eq 4[32]

$$\text{respiration rate } (\text{mL}\cdot\text{CO}_2\cdot\text{kg}^{-1}\cdot\text{h}^{-1}) = \frac{(\Delta\text{CO}_2)}{100} \times V_{\text{headspace}} \times \frac{1000}{m} \times \frac{60}{t} \quad (4)$$

where ΔCO_2 is the difference between the initial and final concentration of CO_2, $V_{headspace}$ is the empty volume of the jar (mL), m is the mass of the strawberries, and t is the sampling time (min).

After firmness analysis, the bananas were cut into small pieces, wrapped with gauze, and squeezed using a plastic squeezer. To determine the total soluble solids content (TSS), 0.3 mL of squeezed juice was added to a refractometer prism (PR-101, Atago, Japan). After TSS analysis, the banana juice was added to an acidometer (GMK-708, G-won Hitech Co., Korea) to measure the percentage of acidity (%).

2.5. Analysis of Antibrowning Effect of the RCE. The antioxidant activity of the RCE was determined using a DPPH assay.[33] This was achieved by mixing 3.8 mL of a standard DPPH methanol solution (0.004%) with 0.2 mL of each sample in different concentrations. The radical quenching potency of the RCE solutions and ascorbic acid as a standard material was also measured at different concentrations (10, 20, 40, 80, 160, and 200 μL/mL). A DPPH standard solution was used as the control sample. The absorbance (Ab) values were analyzed at 517 nm, and DPPH radical quenching was calculated according to eq 5

$$\text{DPPH assay} = \frac{\text{Ab}_{\text{DPPH}} - \text{Ab}_{\text{sample}}}{\text{Ab}_{\text{DPPH}}} \times 100 \quad (5)$$

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ACS Omega 2024, 9, 1069−1076

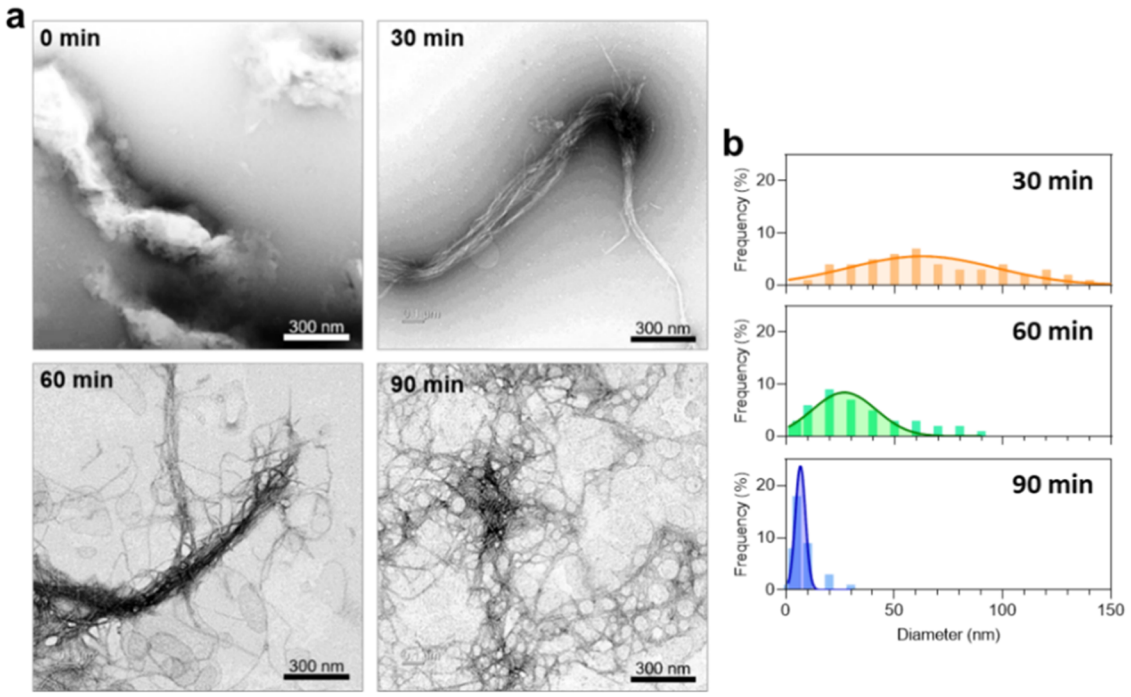

Figure 3. (a) TEM images of CM-CNF over carboxylation time from 0 to 120 min. (b) Gaussian curve of CM-CNF with reaction times of 30, 60, and 90 min. The scale bar is 300 nm.

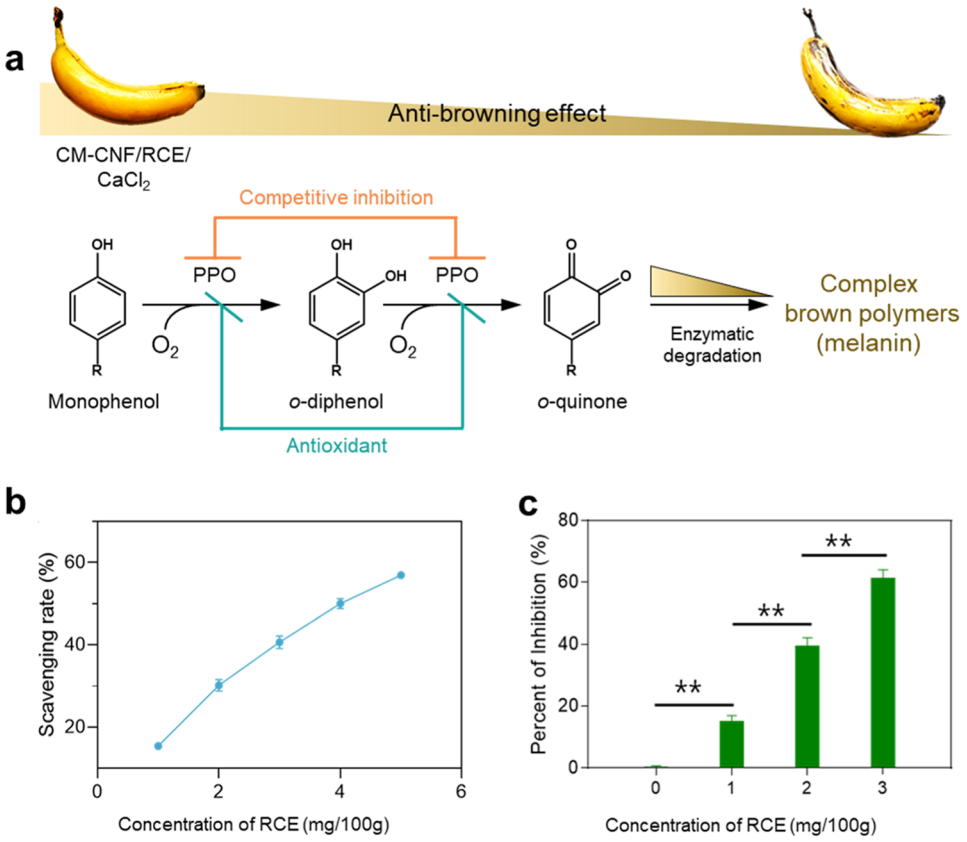

Figure 4. (a) Mechanism of antibrowning effect by RCE. (b) DPPH radical scavenging assay of RCE ($n = 3$). (c) Tyrosinase inhibition assay of RCE. ** indicates a p-value <0.01, and statistical analysis was performed using the one-way ANOVA test.

Tyrosinase inhibition assays were performed with minor modifications.[34,35] Tyrosinase (1000 U/mL) from mushroom solution was prepared at a concentration of 100 U/mL in a phosphate buffer solution at pH 6.5. Tyrosinase mushroom solution (150 μL) and phosphate buffer solution at pH 6.5 (300 μL) were mixed with 0−3% concentrations of RCE (0.8 mg). The mixture was then incubated at 25 °C for 5 min before 300 μL of 2.5 mM tyrosine solution was added, and the reaction was

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ACS Omega 2024, 9, 1069−1076

measured at 475 nm. The percentage of inhibition of tyrosinase activity was calculated using eq 6

$$\text{inhibition } (\%) = \frac{A - B}{A} \times 100 \quad (6)$$

where A represents the difference in absorbance of the control sample and B represents the difference in absorbance of the test sample.

2.6. Statistical Analysis. All experiments were carried out in triplicate, and the average results were shown as the mean ± standard deviation. Statistical analysis of the results was performed using one-way analysis of variance (one-way ANOVA) with Tukey mean analysis. This analysis was performed using IBM SPSS statistics version 25 (SPSS Inc., Chicago, IL).

3. RESULTS AND DISCUSSION

To optimize the condition of carboxylmethylation on the CNF, carboxylate contents and the degree of substitution (DS) were evaluated by reaction time. The carboxylate content of CM-CNF was controlled by varying the reaction time up to 120 min (Figure 2a). Specifically, CM-CNF without pretreatment showed 9.12 μmol/g, while CM-CNFs pretreated for 30, 60, 90, and 120 min showed 289.47, 462.77, 602.35, and 622.33 μmol/g, respectively. The DS was 4.56% for 30 min, 8.23% for 60 min, 10.87% for 90 min, and 11.37% for 120 min (Figure 2b). Both carboxylate content and degree of substitution increased with pretreatment time, and the optimal pretreatment time was determined to be 90 min, considering the steady value of the curves.

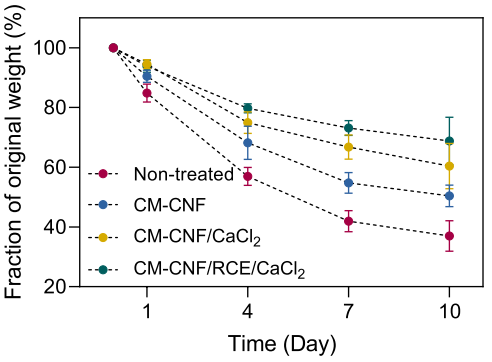

Figure 5. Fraction of the original weight of bananas coated with CM-CNF, CM-CNF/$CaCl_2$, and CM-CNF/RCE/$CaCl_2$ films for 10 days (n = 3).

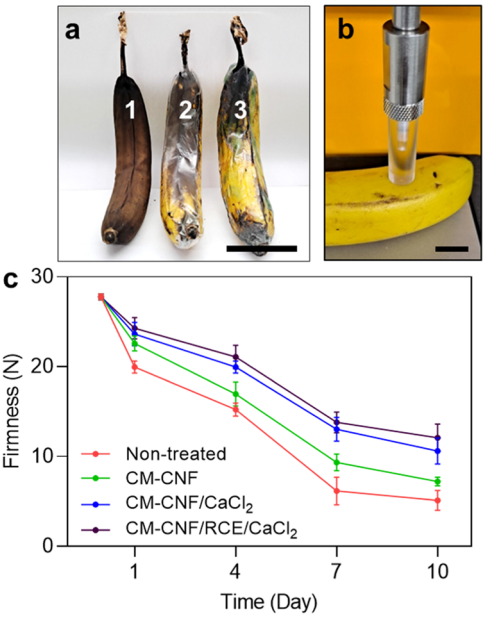

Figure 7. (a) Image of bananas that remained ripe for 10 days at room temperature. 1: nontreated, 2: CM-CNF/$CaCl_2$-coated banana, 3: CM-CNF/RCE/$CaCl_2$-coated banana. The scale bar is 5 cm. (b) Firmness was measured by a texture analyzer at the three random points. The scale bar is 1 cm. (c) Firmness of the banana coated with CM-CNF film for 10 days (n = 3).

While the untreated CNF showed a strong peak at 1649 cm^{-1}, the carboxymethylation of CNFs additionally increased to an absorbance peak at 1598 cm^{-1} as the pretreatment time increased (Figure 2c). This was assumed to be due to the symmetric stretching vibrations of the carboxylate group.[36] Similar to the carboxylate content, CM-CNFs at 90 and 120 min did not show significant differences in peak intensity, indicating the optimal carboxymethylation conditions.

The thickness of CM-CNFs decreased, and nanofibrillation was enhanced as the carboxylation time increased (Figure 3). Untreated cellulose pulp fibers were not separated properly and showed thick bundles of fibers, while treated CM-CNFs showed higher levels of nanofibrillation (Figure 3a). The measured thickness of the CM-CNFs was 51.23 nm for 30 min, 27.45 nm for 60 min, and 8.31 nm for 90 min (Figure 3b). Nanofibrillation

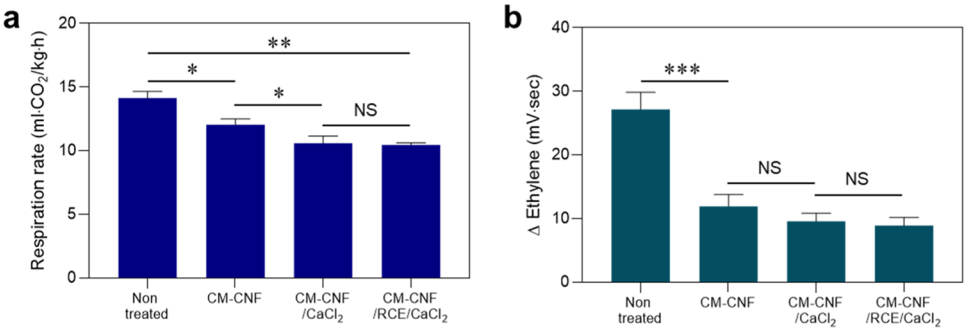

Figure 6. (a) Respiration rates of bananas coated with different CNF films at 10 days. (b) Integrated area of ethylene emitted in bananas coated with CNF films over 4 days (n = 3). NS indicates no significance, * indicates a p-value of <0.05, ** indicates a p-value of <0.01, *** indicates a p-value of <0.005, and statistical analysis was performed using a one-way ANOVA test.

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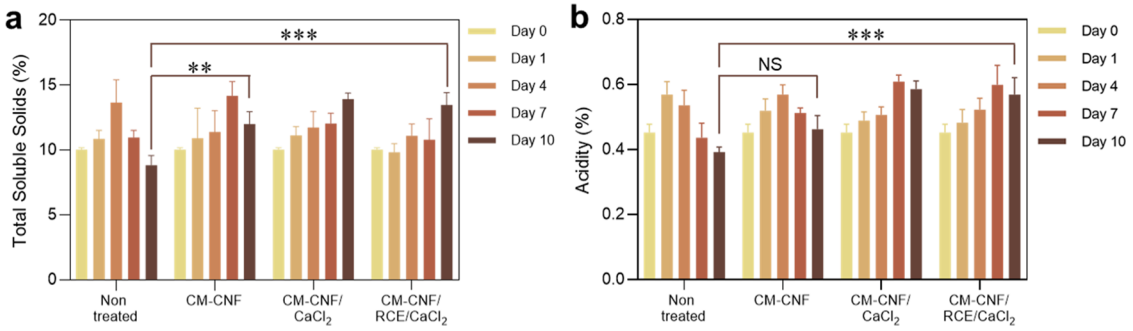

Figure 8. (a) Total soluble solids and (b) acidity of bananas coated with CM-CNF film within 10 days (n = 3). NS indicates no significance, ** indicates a p-value of <0.01, *** indicates a p-value of <0.005, and statistical analysis was performed using a one-way ANOVA test.

resulted from the electrostatic repulsion between CM-CNFs, followed by mechanical grinding.

To determine the proper cross-linking agent of CM-CNFs, the storage modulus of hydrogels prepared with 2% NaCl, $MgCl_2$, and $CaCl_2$ was characterized (Figure S1). The storage moduli of CM-CNF, CNF/NaCl, CNF/$MgCl_2$, and CNF/$CaCl_2$ were 1252.5 ± 95.2, 1193.2 ± 103.1, 1775.5 ± 76.8, and 2190.1 ± 51.1 Pa, respectively. The difference in the storage modulus was not significant between CM-CNF and CM-CNF/NaCl hydrogels because Na^+ is a monovalent ion. In contrast, CM-CNF hydrogels cross-linked with $MgCl_2$ and $CaCl_2$ showed a significant change in storage modulus. Especially, the CNF/$CaCl_2$ hydrogel showed a higher value than CNF/$MgCl_2$ hydrogel due to the strong binding and higher cross-linking density.

A banana, which is a representative climacteric fruit, was chosen for improvement in the preservation period. It appears with a green color before harvesting, but its color rapidly and naturally changes to yellow during ripening.[37] When it arrives at senescence, black dots and peel discoloration appear on the surface of the banana due to enzymatic degradation, known as the browning effect (Figure 4a).[21,38] The process of browning occurs via polyphenol oxidase (PPO). In the presence of O_2, PPO is transformed into substances known as phenolic compounds via the oxidation process. This process, however, generally does not happen within fresh, unripen fruits because the PPO and the phenolic compounds are separated in the cells.[39] The senescence of banana induces cell degradation, and auto-oxidation of PPO changes quinone into melanin, thus producing black dots on the surface.

The antibrowning effect is activated by the antioxidant effect of RCE, and competitive inhibition converts PPO to RCE. RCE contains natural antioxidant products that scavenge radicals via resonance rings. The scavenging rate was obtained with a DPPH assay using a sample concentration of 1 to 5 mg/100 g. The assay showed that the higher the concentration of RCE, the higher the scavenging rate. In particular, the 5 mg/100 g sample showed a 56.94% scavenging rate (Figure 4b). To confirm the antibrowning effect by competitive inhibition, a tyrosinase inhibition assay was performed (Figure 4c). In general, enzymatic browning occurs in bananas due to the destruction of cell walls and tissues during the ripening stage. Tyrosinase released from the cells comes into direct contact with oxygen, and the substrate (L-tyrosine) is oxidized to melanin. As the concentration of RCE increased from 0 to 3 mg/100 g, the activity of tyrosinase was inhibited up to 61.47%. Inhibited tyrosinase does not proceed with the oxidation reaction, and eventually, melanin production is prevented by competitive inhibition.

Dehydration of bananas is caused by water evaporation and increased respiration rates during ripening. Since bananas are vulnerable to dehydration compared to other climacteric fruits, it is necessary to protect them from dehydration. As shown in Figure 5, a noncoated banana was dehydrated to 36.96% of its original weight after 10 days of storage. In contrast, the banana coated with CM-CNF/RCE/$CaCl_2$ film maintained 68.77% of its original weight after the same storage time.

The respiration rate was a factor used to determine the preservation efficiency of the CNF film coatings. It was evaluated by measuring the CO_2 released from the fruit using gas chromatography (Figure 6a). The uncoated banana showed the highest value, and the bananas coated with CM-CNF/$CaCl_2$ film and CM-CNF/RCE/$CaCl_2$ film showed the lowest respiration rate. Ethylene gas is known as an excellent maturing hormone for fruits and vegetables. It promotes the conversion of amylose into sucrose by accelerating the activation of amylase.[40] Besides, since pectinase is rapidly activated by ethylene, the degradation of pectin, which is one of the structural components of fruit skin, is accelerated.[41] As shown in Figure 6(b), the reduced respiration rate was closely related to the production of ethylene gas. The reduced ethylene gas retarded the ripening of bananas, and the preservation time was prolonged.[42]

The amount of ethylene gas released from the bananas was significantly different between the uncoated and coated bananas during storage (Figure 6b). The barrier property against ethylene gas was excellent for both the CM-CNF/$CaCl_2$ film and the CM-CNF/RCE/$CaCl_2$ film.

During senescence, the color of bananas changed due to the browning effect (Figure 7a). The senescence process is caused by decomposition of the pectin layer through the acceleration of pectinase and results in a decreased fruit texture. The firmness of the bananas was measured to evaluate the decomposition of the pectin layer (Figure 7b,c). The firmness of the bananas decreased as the storage time increased due to natural senescence. However, the banana coated with CM-CNF/RCE/$CaCl_2$ film showed a higher firmness than the noncoated or CM-CNF-coated bananas. This resulted from retardation of the senescence process via a decreased respiration rate and inhibited production of ethylene.

Starch−sucrose transformation during banana ripening involves several enzymatic processes, and starch disappears while soluble sugars accumulate. The senescence of the fruit can be informed by the changes in TSS and acidity because metabolic activity is inhibited by the destruction of cells. The uncoated banana reached the maximum value after 4 days and

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ACS Omega 2024, 9, 1069−1076

showed decreases of TSS and acidity after that (Figure 8a,b). In contrast, CM-CNF/$CaCl_2$ and CM-CNF/RCE/$CaCl_2$ films showed increases of TSS until 10 days and acidity until 7 days, indicating prolonged preservation of fruit freshness (Figure 8).

4. CONCLUSIONS

Bananas could be coated with the CM-CNF hydrogel by a simple dipping process. A dense film was formed by subsequent ionic cross-linking with divalent cationic solution. The CM-CNF hydrogel film applied to the banana surface decreased the weight loss and restricted the respiration rate of bananas, thus reducing CO_2 and ethylene production. This resulted in retaining the firmness during storage and helped with retardation of the senescence process, which was confirmed by the time to reach the maximum TSS and acidity. The antibrowning effect of RCE was also activated by antioxidant effects and enzymatic inhibition. The CM-CNF film coating of bananas was effective in preserving freshness.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.3c07273.

Storage modulus of CM-CNF hydrogels (Figure S1) (PDF)

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Notes

The authors declare no competing financial interest.

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■ REFERENCES

(1) Li, S.; Li, Y. Mechanical and Antibacterial Properties of Modified Nano-ZnO/high-density Polyethylene Composite Films with a Low Doped Content of Nano-ZnO. *J. Appl. Polym. Sci.* **2010**, *116*, 2965−2969.

(2) Yang, Z.; Zhai, X.; Zhang, C.; Shi, J.; Huang, X.; Li, Z.; Zou, X.; Gong, Y.; Holmes, M.; Povey, M.; Xiao, J. Agar/TiO_2/radish Anthocyanin/neem Essential oil Bionanocomposite Bilayer Films with Improved Bioactive Capability and Eelectrochemical Writing Property for Banana Preservation. *Food Hydrocolloids* **2022**, *123*, No. 107187, DOI: 10.1016/j.foodhyd.2021.107187.

(3) Huang, H.; Wang, L.; Qiu, D.; Zhang, N.; Bi, F. Changes of Morphology, Chemical Compositions, and the Biosynthesis Regulations of Cuticle in Response to Chilling Injury of Banana Fruit During Storage. *Front. Plant Sci.* **2021**, *12*, No. 792384.

(4) Yue, X.; Miao, M.; Feng, X. Incorporating of Oxidized Cellulose Nanofibers@d-Limonene Pickering Emulsion into Chitosan for Fully Biobased Coatings toward Fruits Protection. *ACS Sustainable Chem. Eng.* **2023**, *11*, 15102−15113.

(5) Ding, J.; Hao, Y.; Liu, B.; Chen, Y.; Li, L. Development and Application of Poly (Lactic Acid)/Poly (Butylene Adipate-Co-Terephthalate)/Thermoplastic Starch Film Containing Salicylic Acid for Banana Preservation. *Foods* **2023**, *12*, No. 3397, DOI: 10.3390/foods12183397.

(6) Hao, Y.; Zhang, M.; Liu, B.; Ma, W.; Dong, Q.; Fan, M.; Wang, Y.; Li, L. Nonmigrating Active Antibacterial Packaging: Antimicrobial Mechanism against Staphylococcus Aureus and Its Application in Large Yellow Croaker. *ACS Sustainable Chem. Eng.* **2023**, *11*, 6220−6229.

(7) Roy, S.; Ghosh, T.; Zhang, W.; Rhim, J. W. Recent Progress in PBAT-based Films and Food Packaging Applications: A Mini-review. *Food Chem.* **2024**, *437*, No. 137822.

(8) Kwak, H.; Kim, H.; Park, S.; Lee, M.; Jang, M.; Park, S. B.; Hwang, S. Y.; Kim, H. J.; Jeon, H.; Koo, J. M.; Park, J.; Oh, D. X. Biodegradable, Water-Resistant, Anti-Fizzing, Polyester Nanocellulose Composite Paper Straws. *Adv. Sci.* **2022**, *10*, No. e2205554, DOI: 10.1002/advs.202205554.

(9) Cazón, P.; Velazquez, G.; Ramírez, J. A.; Vázquez, M. Polysaccharide-Based Films and Coatings for Food Packaging: A Review. *Food Hydrocolloids* **2017**, *68*, 136−148.

(10) Pirozzi, A.; Ferrari, G.; Donsì, F. The Use of Nanocellulose in Edible Coatings for the Preservation of Perishable Fruits and Vegetables. *Coatings* **2021**, *11*, No. 990, DOI: 10.3390/coatings11080990.

(11) Kwak, H.; Shin, S.; Kim, J.; Kim, J.; Lee, D.; Lee, H.; Lee, E. J.; Hyun, J. Protective Coating of Strawberries with Cellulose Nanofibers. *Carbohydr. Polym.* **2021**, *258*, No. 117688.

(12) Yu, H.; Hong, H. J.; Kim, S. M.; Ko, H. C.; Jeong, H. S. Mechanically Enhanced Graphene Oxide/carboxymethyl Cellulose Nanofibril Composite Fiber as a Scalable Adsorbent for Heavy Metal Removal. *Carbohydr. Polym.* **2020**, *240*, No. 116348.

(13) Tabart, J.; Pincemail, J.; Kevers, C.; Defraigne, J.-O.; Dommes, J. Processing Effects on Antioxidant, Glucosinolate, and Sulforaphane Contents in Broccoli and Red Cabbage. *Eur. Food Res. Technol.* **2018**, *244*, 2085−2094.

(14) Wiczkowski, W.; Szawara-Nowak, D.; Topolska, J. Red Cabbage Anthocyanins: Profile, Isolation, Identification, and Antioxidant Activity. *Food Res. Int.* **2013**, *51*, 303−309.

(15) Musso, Y. S.; Salgado, P. R.; Mauri, A. N. Smart Gelatin Films Prepared Using Red Cabbage (*Brassica Oleracea* L.) Extracts as Solvent. *Food Hydrocolloids* **2019**, *89*, 674−681.

(16) Mizgier, P.; Kucharska, A. Z.; Sokół-Łętowska, A.; Kolniak-Ostek, J.; Kidoń, M.; Fecka, I. Characterization of Phenolic Compounds and Antioxidant and Anti-Inflammatory Properties of Red Cabbage and Purple Carrot Extracts. *J. Funct. Foods* **2016**, *21*, 133−146.

(17) Ghareaghajlou, N.; Hallaj-Nezhadi, S.; Ghasempour, Z. Red Cabbage Anthocyanins: Stability, Extraction, Biological Activities and Applications in Food Systems. *Food Chem.* **2021**, *365*, No. 130482.

(18) Wu, J.; Liu, W.; Yuan, L.; Guan, W.-Q.; Brennan, C. S.; Zhang, Y.-Y.; Zhang, J.; Wang, Z.-D. The Influence of Postharvest UV-C Treatment on Anthocyanin Biosynthesis in Fresh-Cut Red Cabbage. *Sci. Rep.* **2017**, *7*, No. 5232.
(19) Mercadal, P. A.; Picchio, M. L.; González, A. Food-Protecting Films Based on Soy Protein Isolate and Natural Deep Eutectic Solvents: Antimicrobial and Antioxidant Properties. *Food Hydrocolloids* **2024**, *147*, No. 109414.
(20) Fang, X.; Li, Y.; Kua, Y. L.; Chew, Z. L.; Gan, S.; Tan, K. W.; Lee, T. Z. E.; Cheng, W. K.; Lau, H. L. N. Insights on the Potential of Natural Deep Eutectic Solvents (NADES) to Fine-tune Durian Seed Gum for Use as Edible Food Coating. *Food Hydrocolloids* **2022**, *132*, No. 107861.
(21) Palmer, J. K. Banana Polyphenoloxidase. Preparation and Properties. *Plant Physiol.* **1963**, *38*, 508−513.
(22) Griffiths, L. A. Detection and Identification of the Polyphenoloxidase Substrate of the Banana. *Nature* **1959**, *184*, 58−59.
(23) Lee, M.-K. Inhibitory Effect of Banana Polyphenol Oxidase during Ripening of Banana by Onion Extract and Maillard Reaction Products. *Food Chem.* **2007**, *102*, 146−149.
(24) Molina, H. E.; Buta, J. G.; Newman, I. M. Prevention of browning of banana slices using natural products and their derivatives. *J. Food Qual.* **1999**, *22*, 499−511, DOI: 10.1111/j.1745-4557.1999.tb00181.x.
(25) Nguyen, T. Relationship between Browning and the Activities of Polyphenoloxidase and Phenylalanine Ammonia Lyase in Banana Peel during Low Temperature Storage. *Postharvest Biol. Technol.* **2003**, *30*, 187−193.
(26) Im, W.; Lee, S.; Abhari, A. R.; Youn, H. J.; Lee, H. L. Optimization of Carboxymethylation Reaction as a Pretreatment for Production of Cellulose Nanofibrils. *Cellulose* **2018**, *25*, 3873−3883, DOI: 10.1007/s10570-018-1853-9.
(27) Shin, S.; Kwak, H.; Hyun, J. Transparent Cellulose Nanofiber Based Open Cell Culture Platform Using Matrix-Assisted 3D Printing. *Carbohydr. Polym.* **2019**, *225*, No. 115235, DOI: 10.1016/j.carbpol.2019.115235.
(28) Huang, P.; Zhao, Y.; Kuga, S.; Wu, M.; Huang, Y. A Versatile Method for Producing Functionalized Cellulose Nanofibers and Their Application. *Nanoscale* **2016**, *8*, 3753−3759.
(29) Markakis, P. *Anthocyanins as Food Colors*; Elsevier, 1982; pp 163−180.
(30) Prietto, L.; Mirapalhete, T. C.; Pinto, V. Z.; Hoffmann, J. F.; Vanier, N. L.; Lim, L.-T.; Dias, A. R. G.; da Rosa Zavareze, E. pH-Sensitive Films Containing Anthocyanins Extracted from Black Bean Seed Coat and Red Cabbage. *LWT* **2017**, *80*, 492−500, DOI: 10.1016/j.lwt.2017.03.006.
(31) Bang, J.; Lim, S.; Yi, G.; Lee, J. G.; Lee, E. J. Integrated Transcriptomic-Metabolomic Analysis Reveals Cellular Responses of Harvested Strawberry Fruit Subjected to Short-Term Exposure to High Levels of Carbon Dioxide. *Postharvest Biol. Technol.* **2019**, *148*, 120−131.
(32) Marelli, B.; Brenckle, M. A.; Kaplan, D. L.; Omenetto, F. G. Silk Fibroin as Edible Coating for Perishable Food Preservation. *Sci. Rep.* **2016**, *6*, No. 25263.
(33) Goupy, P.; Bautista-Ortin, A.-B.; Fulcrand, H.; Dangles, O. Antioxidant Activity of Wine Pigments Derived from Anthocyanins: Hydrogen Transfer Reactions to the DPPH Radical and Inhibition of the Heme-Induced Peroxidation of Linoleic Acid. *J. Agric. Food Chem.* **2009**, *57*, 5762−5770.
(34) Aramwit, P.; Bang, N.; Srichana, T. The Properties and Stability of Anthocyanins in Mulberry Fruits. *Food Res. Int.* **2010**, *43*, 1093−1097.
(35) Kwak, H.; Shin, S.; Hyun, J. Colorimetric Assay of Tyrosinase Inhibition Using Melanocyte Laden Hydrogel Fabricated by Digital Light Processing Printing. *J. Ind. Eng. Chem.* **2020**, *84*, 252−259.
(36) Onyianta, A. J.; Dorris, M.; Williams, R. L. Aqueous Morpholine Pre-Treatment in Cellulose Nanofibril (CNF) Production: Comparison with Carboxymethylation and TEMPO Oxidisation Pre-Treatment Methods. *Cellulose* **2018**, *25*, 1047−1064.
(37) Baite, T. N.; Mandal, B.; Purkait, M. K. Antioxidant-Incorporated Poly(Vinyl Alcohol) Coating: Preparation, Characterization, and Influence on Ripening of Green Bananas. *ACS Omega* **2022**, *7*, 42320−42330.
(38) Murmu, S. B.; Mishra, H. N. Post-Harvest Shelf-Life of Banana and Guava: Mechanisms of Common Degradation Problems and Emerging Counteracting Strategies. *Innovative Food Sci. Emerging Technol.* **2018**, *49*, 20−30.
(39) Zhu, L.-s.; Shan, W.; Wu, C.; Wei, W.; Xu, H.; Lu, W.; Chen, J.; Su, X.; Kuang, J. Ethylene-Induced Banana Starch Degradation Mediated by an Ethylene Signaling Component MaEIL2. *Postharvest Biol. Technol.* **2021**, *181*, No. 111648.
(40) Fabi, J. P.; do Prado, S. B. R. Fast and Furious: Ethylene-Triggered Changes in the Metabolism of Papaya Fruit during Ripening. *Front. Plant Sci.* **2019**, *10*, No. 535, DOI: 10.3389/fpls.2019.00535.
(41) Mulas, M.; Fadda, A.; Angioni, A. Effect of Maturation and Cold Storage on the Organic Acid Composition of Myrtle Fruits. *J. Sci. Food Agric.* **2013**, *93*, 37−44.
(42) Thakur, R.; Pristijono, P.; Bowyer, M.; Singh, S. P.; Scarlett, C. J.; Stathopoulos, C. E.; Vuong, Q. V. A Starch Edible Surface Coating Delays Banana Fruit Ripening. *LWT* **2019**, *100*, 341−347.

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