# Multi-layer Barrier Coating Technology Using Nano-fibrillated Cellulose and a Hydrophobic Coating Agent

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A multi-layer barrier coating technology was developed using nano-fibrillated cellulose (NFC) alongside a hydrophobic, paraffin-free biowax for manufacturing an eco-friendly functional packaging paper. Anionic NFC was prepared by isolating hardwood-bleached kraft pulp (Hw-BKP) using a micro-grinder, and cationic NFC was prepared by the quaternization reaction of the anionic NFC. Thereafter, a three-layer barrier-coated paper was manufactured using cationic and anionic NFCs and biowax. The air permeability and water vapor transmission rate (WVTR) of the three-layer barrier-coated paper were measured, and its coverage and coating layer structure were observed by scanning electron microscopy (SEM). The air permeability of the three-layer barrier-coated paper was more than 15,000 s and those WVTR was 67.1 g/m$^2$/day. Its coverage and surface were considerably uniform and smooth. Thick and effective barrier coating layers were formed as indicated by SEM images. Therefore, it was concluded that a multi-layer barrier-coated paper with considerably high barrier properties could be produced using cationic and anionic NFCs with high gas barrier properties and biowax with high moisture barrier properties. Further, the structure could be used as a functional packaging paper with high barrier properties.

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*Keywords: Barrier coating; Nano-fibrillated cellulose (NFC); Quaternization; Biowax; Gas barrier; Water vapor transmission rate; Electrostatic attraction*

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## INTRODUCTION

Recently, many consumers have become interested in packaging technology or materials that keep food freshness. Because high oxygen and moisture permeability increase food spoilage and reduce food shelf life, it is important to block them to satisfy the demands of the consumers (Nechita and Roman 2020; Jin *et al*. 2021; Trinh *et al*. 2022). To block oxygen or moisture, packaging materials made of synthetic polymer materials with high barrier properties are being used (Huang *et al*. 2022). However, there is a concern about serious environmental pollution due to the overflow of plastic packaging materials. And this requires the development of eco-friendly packaging materials that keep food fresh (Norton 2020).

Among the traditional packaging materials, plastic and paper are most important materials (Deshwal *et al*. 2019; Guzman-Puyol *et al*. 2022). Paper is more eco-friendly than plastic, but it needs to be supplemented with functional packaging material due to its high air permeability and hydrophilicity (Nair *et al*. 2014; Zhang *et al*. 2014; Omran *et al*. 2021; Tao *et al*. 2022). Barrier coating technology provides adequate oxygen and moisture to food by providing barrier properties against oxygen and moisture (Yu *et al*. 2019). To obtain high barrier properties, synthetic polymers with excellent barrier properties are often used as barrier coatings for packaging materials. Synthetic polymers used in barrier coatings are not biodegradable, difficult to recycle, and cause environmental pollution by generating hazardous substances when incinerated (Fotie *et al*. 2020; Wu *et al*. 2021). Therefore, there is increasing interest to replace synthetic polymers with eco-friendly materials.

Nano-fibrillated cellulose (NFC) is an eco-friendly functional polymer made from woody cellulose fiber (Julkapli *et al*. 2017; Fotie *et al*. 2020; Xu *et al*. 2021). It has a low density, high aspect ratio, high biodegradability, high tensile strength, and stiffness (Julkapli *et al*. 2017; Wang *et al*. 2020). Additionally, NFC is an effective barrier coating agent because of excellent film formation, transparency, and gas barrier properties, and it can replace synthetic polymers that cause environmental pollution (Afra *et al*. 2016; Fotie *et al*. 2020; Kargupta *et al*. 2022). However, the NFC made from woody cellulose fibers has a negative charge and hydrophilic property because of the presence of carboxylic acid and hydroxyl groups on the cellulose molecule (Sjöström 1989; Nishiyama 2009).

Several studies have been conducted to improve the barrier properties of paper with NFC coatings. However, it was found to be difficult to increase the coating weight due to the low solid content of NFC (Lavoine *et al*. 2014; Mousavi *et al*. 2017; Lee *et al*. 2020). Moreover, the hydrophilic NFC cannot reduce the water vapor transmission rate (WVTR) of the barrier-coated paper (Nair *et al*. 2014). To overcome these problems, a multi-layer barrier coating technology was developed using NFCs and hydrophobic biowax, as shown in Fig. 1. Conventional barrier coatings provide barrier properties with a single-layer. However, gas and moisture barrier properties can be simultaneously imparted to the packaging paper by forming a coating layer with an NFC layer, which has gas barrier properties, and a biowax layer, which exhibits moisture barrier properties and is highly recyclable in a multi-layer barrier coating system.

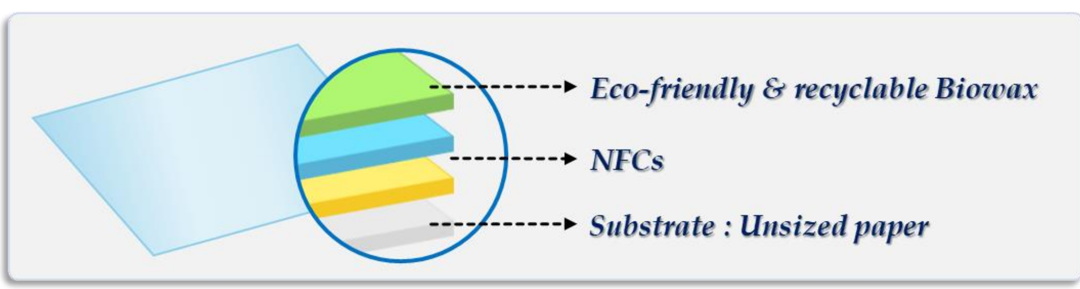


**Fig. 1.** Concept of a multi-layer barrier coating using NFCs and biowax

In this study, a multi-layer barrier-coated paper was produced using NFCs and hydrophobic biowax. The first barrier coating was conducted with cationic NFC, and the second and third layers were coated on the same side as the first coating layer using anionic NFC and biowax. Thereafter, the Gurley air permeability and WVTR of the three-layer-coated paper were measured to evaluate the gas and moisture barrier properties. Additionally, the coverage and coating layer structure were observed by scanning electron microscopy (SEM).

## EXPERIMENTAL

### Materials

Hardwood-bleached kraft pulp (Hw-BKP), supplied by Moorim Paper (Jinju, Republic of Korea), was used to make the NFCs. Glycidiyltrimethylammonium chloride (GMA, Sigma-Aldrich, St. Louis, USA) and N,N-dimethylacetamide (DMAc, Sigma-Aldrich, St. Louis, USA) were used to prepare cationic NFC, and 1 M potassium hydroxide flake(KOH, Daejung, Siheung, Republic of Korea) was used as a catalyst. Ethyl alcohol ($C_2H_5OH$, Daejung, Siheung, Republic of Korea) and n-hexane ($C_6H_{14}$, Thermo Fisher Scientific, Massachusetts, USA) were used for the solvent exchange of NFC pads to measure the fiber width.

A biowax emulsion (Topscreen Bw200, Solenis, Gimcheon, Republic of Korea) was used as a coating agent for imparting hydrophobicity to the filter papers. Biowax can be applied to coating for food packaging due to consist of paraffin-free plant oil. Unsized paper with a grammage of 95 g/m$^2$ was used as the base paper for the multi-layer barrier coating.

### Methods

*Preparation of NFCs*

Anionic NFC was made by refining and micro-grinding. The 1.57% Hw-BKP was soaked in tap water and refined to 450 ± 5 mL Canadian Standard Freeness (CSF) using a laboratory Hollander beater. The refined pulp slurry was diluted to 1.0% consistency for fibrillation. Thereafter, the pulp slurry at 1.0% consistency was fibrillated using a Super Mass Colloider (MKZA6-2, Masuko Sangyo Co., Ltd., Kawaguchi, Japan) at 1,500 rpm. The pulp slurry was then fed into the grinder, and the fibrillation was performed nine times.

Cationic NFC was made from anionic NFC *via* the quaternization reaction described by Chaker and Boufi (2015) and Lee *et al*. (2019). The water was removed from an anionic NFC slurry using a centrifuge (LaboGene 1248, Gyrozen Co., Ltd., Gimpo, Republic of Korea), and the solids were solvent-exchanged twice using DMAC. After the air-drying, the NFC was conditioned at room temperature for 12 h, followed by addition of 0.8 g of GMA per 1 g of oven-dried NFC and approximately 15 mL of 1 M KOH as a catalyst. Thereafter, the reaction was performed in a constant temperature water bath at 70 °C for 6 h. After the reaction was completed, the cationic NFC was washed five times with distilled water, and a centrifuge was used to remove the remaining unreacted chemicals.

*Characterization of anionic and cationic NFCs*

The fiber width of the prepared anionic and cationic NFCs was analyzed using a field emission scanning electron microscope (FE-SEM, JSM-7610F, JEOL, Tokyo, Japan) to ensure that the NFCs had been ground to the nano-scale after micro-grinding. Wet NFC pads were prepared as test specimens to measure the fiber width using the vacuum filtration system. The wet NFC pads were dried *via* the solvent exchange method using ethyl alcohol, acetone, and n-hexane to prepare the test specimens.

Afterward, FE-SEM images of the pads were captured, and the fiber width was measured with image analysis using a three-dimensional (3D) image software (MP-45030TDI, JEOL, Tokyo, Japan). The particle size for 0.2% NFC slurry was measured using a particle analyzer (1090 LD, CILAS, Orléans, France). The laser scattering-based particle size measurement is not a perfect method for detecting the fiber dimensions because NFCs have a high aspect ratio (Gantenbein *et al*. 2011). However, particle size data were used to indirectly indicate the size differences between NFCs (Park *et al*. 2018). The zeta-potential for 0.01% NFC slurry was measured using a zeta-potential analyzer (Nano ZS, Malvern Panalytical, Malvern, UK).

*Evaluation of the barrier properties of a barrier-coated paper depending on NFC type*

In previous studies, the most difficult point in the barrier coating process using NFCs was that the desired coating weight could not be sufficiently achieved to show high gas barrier property (Mousavi *et al*. 2018; Lee *et al*. 2020; Yook *et al*. 2020). Thus, the concept of multi-layer barrier coating that forms two or more barrier coating layers on one side of the paper was developed. To select the NFC to be used for the multi-layer barrier coating, a two-layer barrier coating was conducted using anionic and anionic NFCs, and cationic and anionic NFCs.

The NFC slurries at 2% concentration were prepared for the barrier coating. The barrier coating was performed using a laboratory rod coater (Auto bar coater, HanTech Co., Ltd., Daejeon, Republic of Korea). The top side of a base paper with a grammage of 95 g/m$^2$ was coated using a rod coater, and the paper was dried in an air dryer at 105 °C for 150 s and then passed through a cylinder dryer at 120 °C to prevent the barrier-coated paper from curling. After the top side of the paper was coated with anionic or cationic NFC, the process was repeated on the same side using anionic NFC. During the two-layer barrier coating, the coating speed was adjusted at 70 mm/s, and the rod number used was 22.

The two-layer barrier coated papers were conditioned at 23 ℃ and 50% relative humidity (RH) before measuring their coating weight and air permeability. Gurley air permeability (TAPPI T460 om-02 (2006)) was measured using an air permeability tester (4110N, Gurley Precision Instruments, New York, USA). The coverage of the barrier-coated papers was evaluated using the SEM images of their surface captured using an FE-SEM (JSM-7601F, JEOL, Tokyo, Japan) analysis.

*Evaluation of the barrier properties of a multi-layer barrier coated paper*

To obtain the low air permeability and high moisture resistance of the barrier-coated paper, a three-layer barrier coating was performed with cationic and anionic NFCs, and a hydrophobic biowax. The cationic NFC was used for the first layer, and anionic NFC was used for the second layer. Thereafter, a biowax layer was formed on the last layer of the barrier-coated paper to afford high hydrophobicity. As a control, a one-layer barrier-coated paper was prepared with only biowax. Figure 2 shows the detailed three-layer barrier coating conditions for each layer.

To evaluate the gas barrier property of the three-layer barrier-coated paper, the air permeability was measured according to the TAPPI T460 om-02 (2006) protocol using the Gurley air permeability tester (4110N, Gurley Precision Instruments, New York, USA). The WVTR was measured at 23 °C and 50% RH using a WVTR tester (w3/031, Labthink, Jinan, China). The surface and cross-section of the three-layer barrier-coated papers were analyzed using an SEM (JSM-6380LV, JEOL Ltd., Tokyo, Japan).

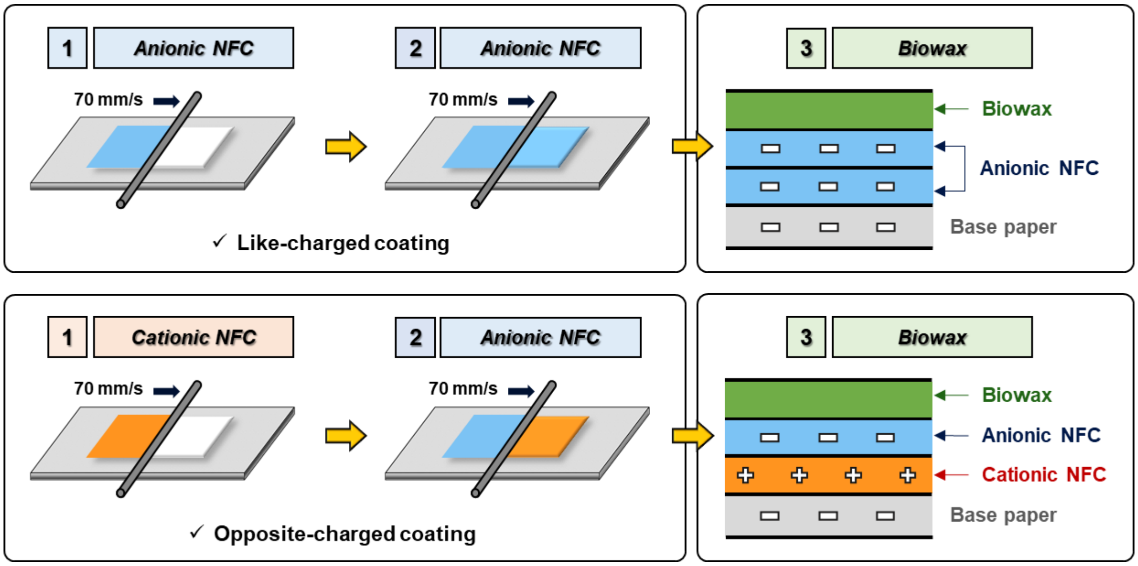


**Fig. 2.** Flow diagram of multi-layer barrier coating using NFCs and biowax

# RESULTS AND DISCUSSION

## Characteristics of the Two NFCs

Table 1 shows the fiber widths and average particle sizes of the anionic and cationic NFCs. Both NFCs showed a lower fiber width than 100 nm, which is a standard between micro- and nano-fibers (Chinga-Carrasco 2011; Isogai *et al.* 2011), and there was no difference in the level. The particle sizes of the anionic and cationic NFCs were approximately 11 μm and showed similar levels. The cationic NFC was made from anionic NFC; thus, the quaternization reaction did not affect the change in the physical size of the anionic NFC.

**Table 1.** Fiber Widths and Average Particle Sizes of NFCs

| | Fiber Width from FE SEM (nm) | Average Particle Size from Light Scattering (μm) |
|---|---|---|
| Anionic NFC | 38.69 | 11.89 |
| Cationic NFC | 37.50 | 11.35 |

Table 2 shows the average, maximum, and minimum zeta-potentials of the anionic and cationic NFCs. The average zeta-potential of the anionic NFC was −17.4 mV, and the maximum zeta-potential was −6.5 mV, indicating that all nanofibers were anionic. However, the cationic NFC prepared by the quaternization reaction of anionic NFC showed an average zeta-potential of +31.7 mV and the lowest zeta-potential of +19.5 mV, indicating that the charge of the anionic NFC was completely reversed to be cationic through the quaternization reaction.

Therefore, it was confirmed that the cationic NFC, which was prepared from the mechanically treated anionic NFC through the quaternization reaction, had a similar physical size as the original anionic NFC; however, its zeta-potential was completely reversed to cationic.

**Table 2.** Zeta-potential of NFCs

| | Average Zeta-potential (mV) | Minimum Zeta-potential (mV) | Maximum Zeta-potential (mV) |
|---|---|---|---|
| Anionic NFC | -17.4 | -26.4 | -6.5 |
| Cationic NFC | +31.7 | +19.5 | +49.4 |

### Barrier Properties of the Two-layer Barrier-coated Paper Depending on NFC Type

To determine the best combination of NFCs for improving the coating weight and gas barrier properties, two-layer barrier coating was performed using anionic and anionic NFCs, and cationic and anionic NFCs. Figure 3 shows the coating weights of the two-layer barrier coated papers. As the number of barrier coating layers increased, the coating weight of the barrier coated paper also increased. The coating weight of the two-layer barrier-coated paper using cationic NFC and anionic NFC was 8.3% higher than that of the two-layer barrier-coated paper using only anionic NFCs. This was attributed to the attractive forces between the cationic NFC and anionic base paper and the cationic NFC and anionic NFC, which further increased the coating weight in the two-layer barrier coating process (Hubbe 2021).

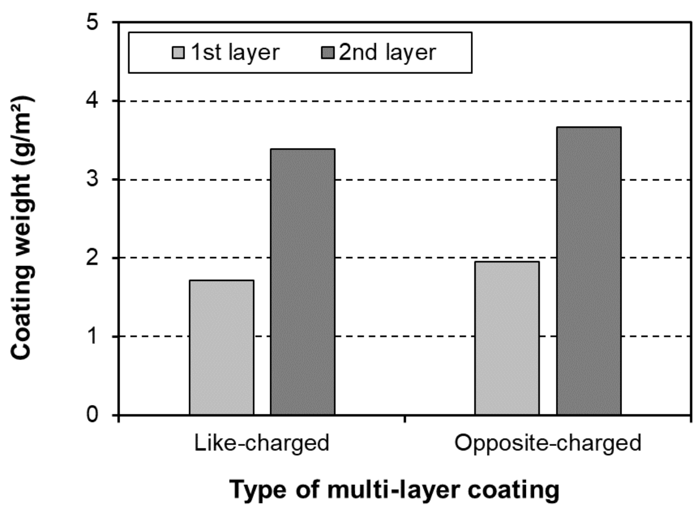


**Fig. 3.** Coating weight of the two-layer barrier coated paper using NFCs

Figures 4 and 5 show the air permeability and coverage of the two-layer barrier-coated paper. The Gurley air permeability of the two-layer barrier-coated paper using cationic and anionic NFCs was approximately 38,000 s, and the air permeability of the two-layer barrier-coated paper using only anionic NFCs was approximately 10,000 s, indicating that the cationic NFC was more effective for improving the gas barrier property. When observing the coverage of the two-layer barrier-coated paper depending on the NFC type, the two-layer barrier-coated paper using cationic and anionic NFCs showed better coverage than that using only anionic NFCs. According to the ionic bonding mechanism described by Hubbe (2021), it has been reported that bonds formed with opposite-charged ionic groups can improve barrier properties of film in comparison to a default system. This prior study supports the finding that the opposite-charged coating exhibited higher coating weight and Gurley air permeability value in comparison to like-charged coating due to electrostatic attraction by different ionic properties. Therefore, it was confirmed that the combination of the cationic and anionic NFCs for a multi-layer barrier coating was effective to improve the gas barrier properties.

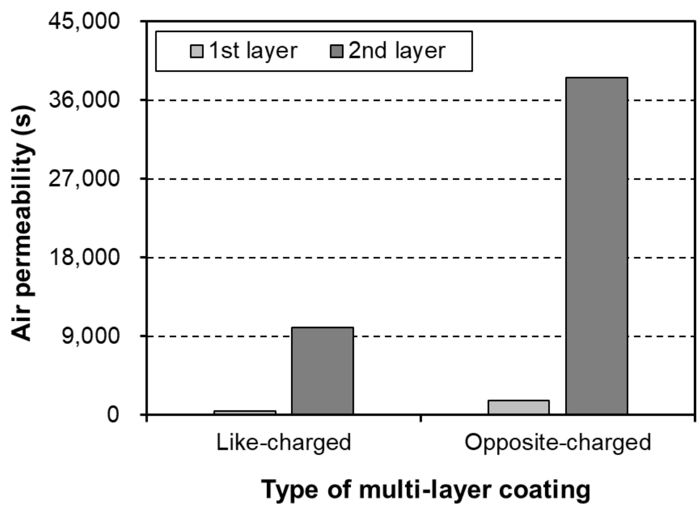


**Fig. 4.** Air permeability of the two-layer barrier-coated paper using NFCs

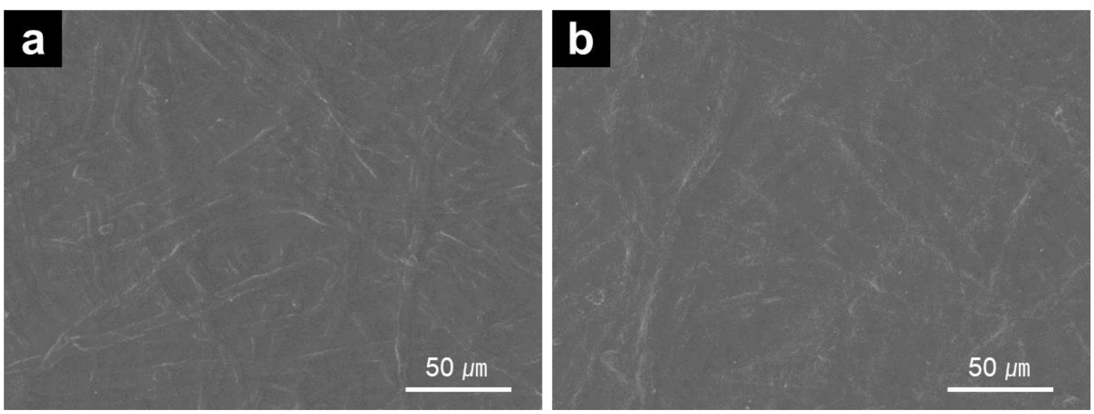


**Fig. 5.** SEM images of the surface of the two-layer barrier-coated paper using NFCs (a: anionic + anionic, b: cationic + anionic)

## Evaluation of the Barrier Properties of the Multi-layer Barrier-coated Paper Using NFCs and Biowax

Table 3 shows the air permeability and WVTR of the three-layer barrier-coated paper using the cationic and anionic NFCs and biowax. The air permeability of the base paper used for the barrier coating was 50 s. After applying the barrier coating to the base paper in the order of cationic NFC and anionic NFC, the air permeability of the two-layer barrier-coated paper rapidly increased to 38,600 s. However, the base paper and the two-layer barrier-coated paper were considerably hydrophilic and porous. Thus, it was impossible to measure the WVTR. After the last barrier coating using biowax was conducted, the air permeability of the three-layer barrier-coated paper was more than 1,500,000 s, and the WVTR was 67.1 g/m$^2$/day, indicating that biowax was considerably effective for improving the barrier properties. However, the single-layer barrier-coated paper using only biowax showed an air permeability of 700 s, and WVTR immeasurable.

**Table 3.** Air Permeability and WVTR of the Barrier-coated Paper Using Cationic and Anionic NFCs and Biowax

| Barrier Coating Layer | Air Permeability (s) | WVTR (g/m$^2$/day) |
|---|---|---|
| No coating (base paper) | 50 | - |
| Cationic NFC + anionic NFC | 38,581 | - |
| Cationic NFC + anionic NFC + biowax | > 1,500,000 | 67.1 |
| Only biowax | 700 | - |

Figure 6 shows the coverage of the barrier-coated paper using cationic and anionic NFCs with biowax in comparison to using only biowax. The three-layer barrier-coated paper showed a uniform and smooth coverage; however, there were many cracks on the surface of the single-layer barrier-coated paper using only biowax. This was because the biowax coating layer was affected by the surface uniformity of the coated paper. After the base paper was coated using NFCs, improved coverage and smoothness were observed. After the NFC coatings, the additional barrier coating using biowax showed better coverage and higher smoothness than the two-layer barrier-coated paper using NFCs. Therefore, it was confirmed that effective barrier coating could not be performed with biowax alone, and the combination of NFCs and biowax showed a considerably high synergistic effect on the improvement of the barrier properties of the three-layer barrier-coated paper.

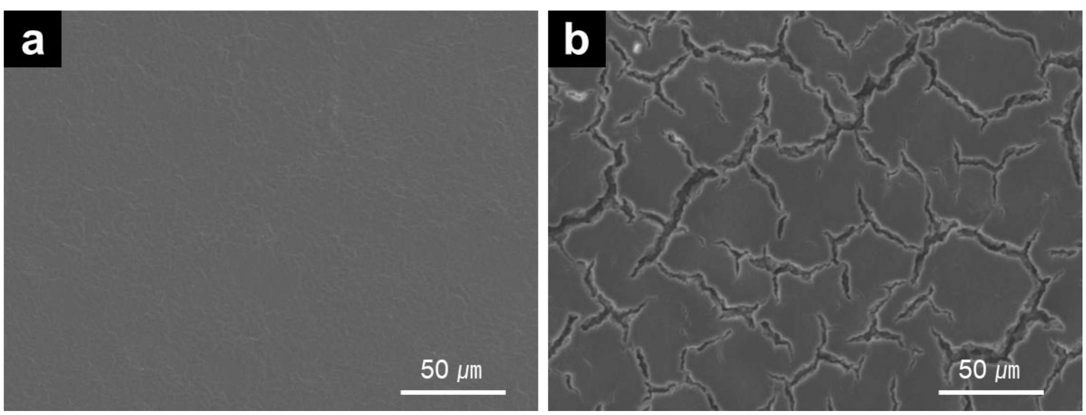


**Fig. 6.** SEM images of the surfaces of (a) the three-layer barrier-coated paper using NFCs and biowax and (b) the single-layer barrier-coated paper using only biowax

Figure 7 shows the cross-section of the coating layer of the three-layer barrier-coated paper. Comparing the cross-section image of the base paper with that of the three-layer barrier-coated paper confirmed that thick and effective barrier coating layers were formed after the barrier coating using NFCs and biowax, and the external surface of the three-layer barrier-coated paper was considerably smooth.

These results were similar to those of Venditti's study (Spence *et al.* 2011), which improved the barrier properties by using beeswax on MFC film. Beeswax and biowax could be completely applied to the MFC or NFC surface to form a thick coating layer and effectively reduce the WVTR and Gurley air permeability. Therefore, it was confirmed that the barrier-coated paper with considerably high barrier properties could be manufactured if the cationic and anionic NFCs with high gas barrier properties and biowax with high moisture barrier properties were used as multi-layer barrier coating agents. Finally, it was concluded that the three-layer barrier-coated paper with NFCs and biowax could be used for food packaging according to literature, which reported that the water permeability range for food packaging was 0.1 g/m$^2$/day to 100 g/m$^2$/day (Palmström and Reese 2022).

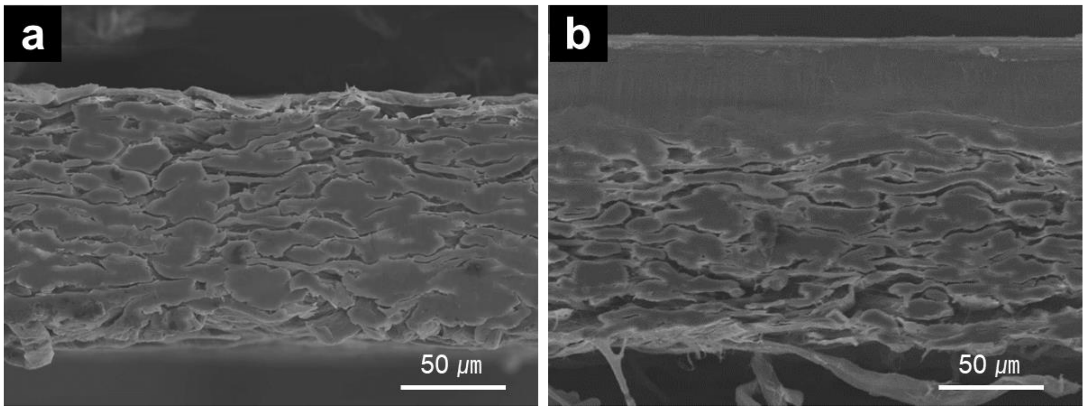


**Fig. 7.** SEM images of the cross-sections of (a) the base paper and (b) the three-layer barrier-coated paper using NFCs and biowax

## CONCLUSIONS

1. Cationic nanofibrillated cellulose (NFC) that was prepared from anionic NFC through the quaternization reaction, had a similar physical size as anionic NFC. Meanwhile, the charge of anionic NFC was completely reversed to a cationic charge.
2. The combination of the cationic and anionic NFCs for a two-layer barrier coating was effective to acquire the desired coating weight for improving the gas barrier property of the barrier-coated paper.
3. A multi-layer barrier coating technology was developed using NFCs and a paraffin-free biowax for manufacturing an eco-friendly functional packaging paper. Three-layer barrier-coated paper with considerably high barrier properties could be manufactured using cationic and anionic NFCs with high gas barrier properties and biowax with high moisture barrier properties.
4. It was concluded that the multi-layer barrier-coated paper could be used as a functional packaging paper with high barrier properties.

## ACKNOWLEDGMENTS

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