Comparison of the Effects of Two Biodegradable Coatings on the Characteristics of White-top Linerboard Used in Packaging Food Materials in Cold Environments

Jafar Ebrahimpour Kasmani,[a,*] and Ahmad Samariha [b]

Effects of two biodegradable coatings were compared relative to the characteristics of white-top linerboard. To coat the surface of the paperboard, nano-polyurethane was sprayed onto the surface using a nozzle. Subsequently, the samples were placed inside a refrigerator and freezer for a period of 2 and 4 months. In the second stage, nano-polyurethane was again sprayed onto the surface, using a nozzle, to improve the performance of the coating material. To further enhance the coating, the surfaces of the coated white-top linerboard were coated with a nanoclay using a laboratory coater. Subsequently, the samples were placed inside a refrigerator and freezer for a period of 2 and 4 months. The properties of the samples were measured thereafter. The results showed a reduction in water absorption of the samples after coating and freezing. This can be attributed to the penetration of the coating solution into the paper pores, resulting in a decrease in pore diameter and, consequently, a decrease in water permeability through the paper pores. In the coated and frozen samples, an increase in thickness and surface smoothness was observed, but most of the mechanical strength properties decreased. These changes were more pronounced in the dual-layer coatings.

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INTRODUCTION

The use of paperboard in packaging has a wider application compared to plastic materials due to features such as its low density and biodegradability. However, ordinary paperboard has weak barrier properties relative to water, gas, and oil permeability. To improve the barrier properties of paperboard packaging, the use of synthetic polymers such as polyethylene, latex, and polyvinyl alcohol can be problematic in terms of environmental impact. In order to solve these problems, films and coatings based on polysaccharides, proteins, fats, or their combinations have been introduced into the packaging industry (Aloui *et al*. 2011).

White-top linerboard can have high hardness or softness (depending on the type). The top ply layer of the paperboard usually is white, whereas the outer ply on the back may be white or gray. A white ply is widely used as a surface layer in various industries such as industrial, pharmaceutical, food, printing, publishing, and packaging.

However, it is important to note that some types of cardboard are used in humid environments. Such environments can cause the packages to become soft and lose their strength. As a result, even with the slightest movement, the cardboards can tear apart, leading to the destruction of the packaging. In addition, the entry of moisture into the cartons can cause damage to the contents. In such conditions, it is recommended to use cardboards that are resistant to moisture and water. It is important to consider the use of starch, a water-based adhesive, in the bonding of three-layer, five-layer, and seven-layer cartons. However, it should be noted that paper and cardboard, being hygroscopic materials, have a propensity to absorb moisture from the surrounding environment. In high relative humidity conditions, these materials can experience a loss of up to 10% of their initial strength (Aloui *et al.* 2011). Therefore, when utilizing starch as an adhesive, careful attention must be paid to environmental factors, such as humidity levels, to ensure the integrity and strength of the cartons.

As a water-based material, starch adhesive also loses its bonding strength in high humidity conditions, resulting in reduced adhesion for attaching cardboard layers (Reis *et al*. 2011). The decrease in the strength of the cardboard layers on the one hand and the starch glue on the other hand will cause the loss of the cardboard resistance, especially in the full (packaged) state, and will cause a decrease in the quality of the product packaging. To compensate for these problems, covers are used on cardboards (Asadi Khansari and Dehghani Firouzabadi 2013).

Coatings improve the final properties of paper and cardboard. New compounds have been introduced for the coating of paper and cardboard. Many paper products, ranging from cups to food packaging, are coated with petroleum-based polyethylene, which hinders their recyclability and contributes to plastic waste generation. With regard to the increasing human attention to the environment, sanitation and health, it is considered that in the near future, all non-degradable materials based on petroleum derivatives will be removed from packaging industries and bio-composites perishable for all packages. These alternatives provide a sustainable option for replacing petroleum-based coatings and improving the recyclability of paper products. Additionally, research and advancements in the field of sustainable packaging aim to reduce the environmental impact of paper coatings while maintaining their functionality and performance.

The potential benefits of using two coatings instead of a single coating are worth exploring in the context of this study. While a single coating may seem simpler, it often falls short of achieving desired results for several reasons. A single coating can develop small holes or defects that compromise its protective barrier, allowing moisture or harmful agents to penetrate the underlying material and reducing performance and durability. Incorporating a second coating offers an opportunity to address these weaknesses. The second coating can serve as a repair or reinforcement layer, effectively sealing any existing holes or defects in the primary coating. This additional layer enhances the overall performance and durability of the coated material, reducing the risk of moisture absorption and damage. Using two coatings provides a potential advantage over a single coating approach. It may establish a more robust and reliable protective barrier, contributing to improved performance and longevity of the coated materials. This study aims to assess the effectiveness of this dual-coating approach and evaluate its suitability for applications where moisture resistance and durability are crucial factors.

A group of nanomaterials known as bio-nanomaterials have a biological origin, making them biocompatible, biodegradable, and renewable (Asadi Khansari *et al.* 2015). Among them, nanocellulose and nano-polyurethane have gained significant attention due to their intriguing inherent properties, including high surface area, high aspect ratio (length to diameter ratio), abundance of resources, low density, high mechanical strength, renewability, and biodegradability (Hubbe *et al*. 2008).

To enhance the properties of paper for various applications, improving certain characteristics such as surface properties (smoothness and surface porosity), barrier properties (resistance to air permeability and moisture), and mechanical properties (tensile strength and tear resistance) are essential. To achieve this, various processes are employed, including the use of different types of paper pulp, virgin fibers, costly additives, fillers, refining, lamination, coating, and more. Some of these treatments result in increased capital investment, operating costs, and ultimately an overall increase in costs (Julkapli and Bagheri 2016). One of the processes used in this regard is applying surface treatments to paper, which can have lower production costs compared to other process treatments. Among surface treatments, paper coating holds a significant position (Rhim *et al*. 2006).

Cardboard manufacturers try to produce cardboards that have high strength. The goal is to prevent fruits from being crushed, especially in the case of export fruits that have to travel a long distance. Therefore, the aim of this research was to use biodegradable nanomaterials for coating, which not only have biodegradability and renewability but also enhance the water absorption capacity of the white-top linerboard. Appropriate measures are being considered to manufacture high-quality boards suitable for use in cold chain packaging industries, addressing the challenges associated with the use of boards in these industries.

EXPERIMENTAL

Materials

A commercial nanoclay product with trade name of Cloisite 30B® was introduced from Southern Clay (Southern Clay Products Inc., Texas, USA). Cloisite-30B® is a natural montmorillonite modified with a quaternary ammonium salt, having a d-spacing of 18.5 ◦A and modifier concentration of 90 meg/ 100 g clay. The nano-polyurethane used was obtained from a specific characteristic startup company, as described in Table 1. Cationic starch was prepared from potato starch produced by Lyckeby Amylex Company (Slovakia). Cationic starch used has the following properties: pH of approximately 6, degree of substitution (DS) of approximately 0.035 mol/mol, protein content of 1.5%, nitrogen content of 0.25%, and moisture content of 10% based on fresh weight. White-top linerboard is one of the types of packaging paper, whose top surface is white and has good printing quality. The grammage of Rasha Caspian white-top linerboard is 131 $g.m^{-2}$, and in the production of this paper, about 30% virgin pulp is used.

Table 1. Specifications of Nano Polyurethane

Appearance	**Light yellow liquid**
Type	Transverse self-joining
Emulsion property	anionic
Percentage of solids	33%
Viscosity	378 mPa.s
Particle size	10-70 nm

Base Paper Coating

First coating

Initially, the white-top linerboard was restrained on a plywood and coated by a spray gun. As much as possible, the goal was to cover the surface evenly. At this stage, nano-polyurethane was used for coating, and due to the impossibility of the exact amount of coating, papers with a coverage of about 15 g/m^2 (the weight of the applied coating is 15 g/m^2) were used for the initial coating.

Second coating

For the second layer application, initially, 4 g of nanoclay was combined with 100 g of distilled water and stirred for 30 min at 50 °C. Subsequently, 2.5 g of styrene butadiene latex and 0.5 g of D200 dispersant from Simab Resin Company were added, and the mixture was stirred again for 20 min. This solution, along with cationic starch at a 5% concentration (based on dry weight), was utilized as a retention aid to enhance coverage and achieve a more uniform distribution of nanoclay particles on the paper surface.

The starch coating solution was prepared by dissolving 5 g of starch in 100 mL of distilled water, resulting in a 5% concentration. The mixture was then cooked at 90 °C for 30 minutes. Afterward, it was maintained in a constant temperature water bath at 50 °C and used for surface coating. A suspension of nanoclay particles was prepared by gradually adding 100 mL of nanoclay colloid solutions to 900 mL of gelatinized starch solution. The mixtures were homogenized and stirred using a magnetic stirrer unit for 30 min.

The coating solutions were applied to paper sheets using an Auto Bar Coater (GBC – A4 GIST Co., Ltd). A volume of 27 mL of the coating solution was poured onto the paper width-wise from one end, and an applicator rod was immediately swept along the length of the paper sheets. The coating speed was set at 25 mm/s. The coated paper sheets were air-dried at room temperature for 24 hours. Subsequently, the samples were placed in a freezer at approximately -15 °C for 2 and 4 months. The specific codes and percentages of the compounds used in the coatings and treatments are summarized in Table 2. Prior to characterization, all samples were conditioned at 27 °C with 65% relative humidity for at least 24 h, following ISO 187 standard.

Table 2. Codes and Conditions of Treatments

No	Code	Description
1	W_0	Control sample
2	W_2	The control sample was frozen for 2 months
3	W_4	The control sample was frozen for 4 months
4	C_{10}	Sample coated once
5	C_{12}	The sample was coated once and frozen for 2 months
6	C_{14}	The sample was coated once and frozen for 4 months
7	WC_{20}	The sample is coated twice
8	WC_{22}	The sample was coated twice and frozen for 2 months
9	WC_{24}	The sample was coated twice and frozen for 4 months

Measurement of Paper Properties

The measurement of paper properties was conducted according to TAPPI standards. The physical properties included thickness (T411-Om89), water absorption (T441 om-96), and surface smoothness (T555om-04). The mechanical properties included tensile strength index in the machine direction and cross direction (T404-Om92), tear strength index in the machine direction and cross direction (T404om-04), stiffness index

(T489 cm-97), and brightness (T403-Om91). The contact angle was measured to determine the wettability of various paper sheets using water as the probe liquid (Kocak 2001).

Microscopic Studies

Microscopic studies were conducted to examine the surface morphology and internal structure of the paper samples. The scanning electron microscopy (SEM) images were obtained using a FESEM instrument (model: 3-XMU Mira) and were analyzed by SEM experts and image processing specialists.

Statistical Calculations

The experimental design was completely randomized, and the obtained measurement results were processed using SPSS software (version 23) for data analysis. One-way analysis of variance (ANOVA) was employed for data analysis and to compare means, the Duncan test was utilized.

RESULTS AND DISCUSSION

To examine the statistical difference between the means of the properties under investigation, an analysis of variance (ANOVA) test was conducted. The obtained F-value from this test and the corresponding significance level are presented in Table 3.

Table 3. Statistical Analysis of Handsheet Samples

Properties	F-value
Thickness (µm)	249.127*
Water absorption (g/m^2)	90.97*
Surface smoothness (S)	63.174*
Contact angle (degree)	28.11*
Burst index (kPam^2/g)	101.972*
Tear index MD (mNm^2/g)	81.789*
Tear index CD (mNm^2/g)	173.452*
Tensile index MD (mkN/m)	62.075*
Tensile index CD (mkN/m)	25.530*
RCT CM (KN/m)	8.126*
RCT CD (KN/m)	22.214*
Brightness (%)	21.849*

Note: * 95% significance level; ^{ns} no significance

Thickness

A one-way analysis of variance (ANOVA) revealed a statistically significant difference among the thickness values of the 9 tested paper types at a significance level of 5%. The thickness values were grouped into six categories across all treatments. Figure 1 illustrates the average thickness variations for the 9 paper types. The highest thickness value was associated with the white-top linerboard that had been coated twice and frozen for 2 months, while the lowest value belonged to the control sample.

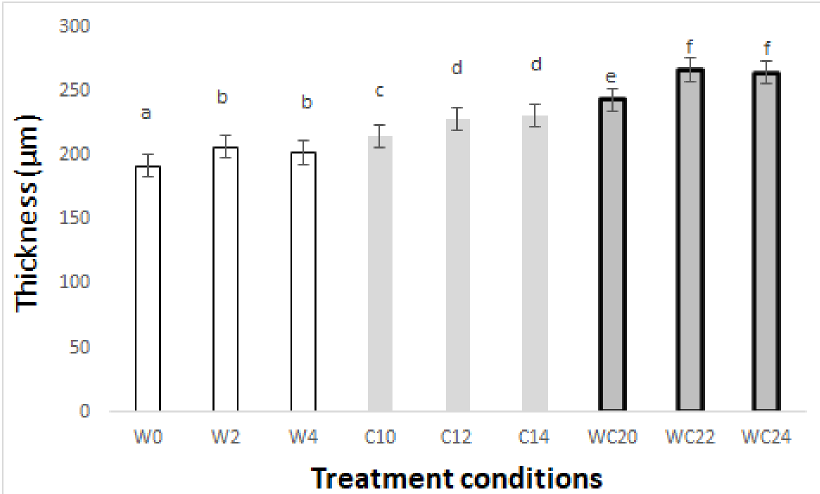

Fig. 1. Comparison of the average thickness of different papers (small letters indicate the Duncan ranking of the averages at a 95% confidence interval), (W_0: Control sample, W_2: The control sample was frozen for 2 months, W_4: The control sample was frozen for 4 months, C_{10}: Sample coated once, C_{12}: The sample was coated once and frozen for 2 months, C_{14}: The sample was coated once and frozen for 4 months, WC_{20}: The sample is coated twice, WC_{22}: The sample was coated twice and frozen for 2 months, WC_{24}: The sample was coated twice and frozen for 4 months)

The increase in thickness in dual-layer coatings was significantly higher than that of single-layer coatings. The deposition of the coating on the substrate led to an increase in thickness. Such deposition results in the formation of a coating layer, and the thickness of this layer is influenced by the properties of the polymer and the amount of solid materials in the coating solution (Qie *et al*. 2023). When the coating is applied in the form of a single layer or dual layers, there is increased molecular contact between the coating components, which may weaken the compressive forces of the polymer chains and consequently open up the coating matrix further. As a result, the thickness increases, indicating the loss of homogeneity and uniformity in the coating layer (Seo *et al*. 2020).

Water Absorption

ANOVA revealed a statistically significant difference among the water absorption values of the 9 tested paper types at a significance level of 5%. The water absorption values were grouped into nine categories across all treatments. Figure 2 shows the average water absorption variations for the 9 paper types. Figure 2 demonstrates that the lowest water absorption value was associated with the white-top linerboard that had been coated twice, while the highest value belongs to the control samples that were frozen for 4 months.

Water absorption is considered one of the barrier properties of coating papers. Generally, the water absorption of paper depends on two factors: the porous structure of the sheet and the interaction between fibers and water. Coating cardboard with nano-polyurethane and nano-clay provides water absorption properties to the cardboard. The water absorption in samples coated and freeze-dried with nano-polyurethane and samples coated and freeze-dried with nano-polyurethane and nano-clay was lower than that of the untreated and non-freeze-dried control samples. This reduction in water absorption was more pronounced in dual-layer coatings, which can be attributed to the small size of the nano-particles and, consequently, their larger specific surface area. These particles can easily fill the voids and gaps between the fibers, resulting in a decrease in water absorption.

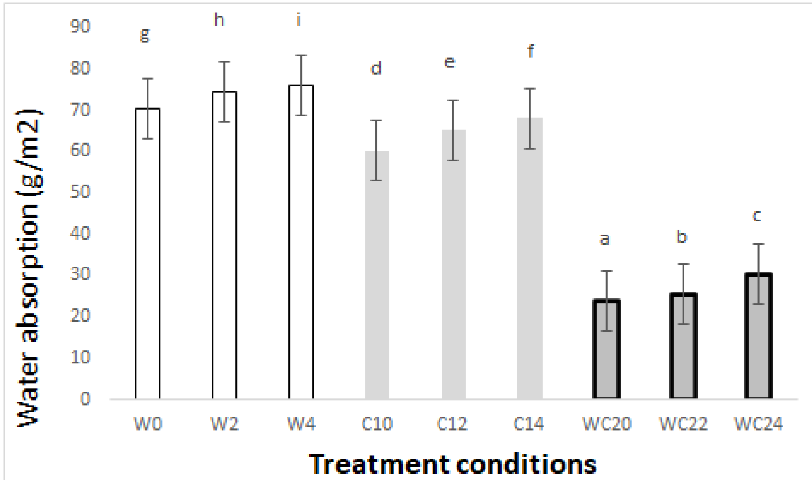

Fig. 2. Comparison of average water absorption of different papers

During the coating process, the access of water to the fibers and the formation of hydrogen bonds with the functional groups present in the fibers are reduced, leading to a decrease in water absorption. Regarding the water resistance of the samples, it should be noted that the low hydrophilicity of nano-polyurethane and nano-clay causes these materials to absorb fewer water molecules. Due to covering the surface of the paper and closing the pores and empty space between the fibers with nano-polyurethane, less water reaches the fiber network, the empty space between the fibers, and the fibers themselves.

Additionally, this reduction may be due to the fact that by coating the surface of the paper, we have created a hydrophobic surface, resulting in a decrease in surface wettability and water absorption.

Vähä-Nissi *et al*. (2017) concluded that due to the extremely small size of the nanoparticles, they can easily fill the voids and gaps in the defective matrix of the coating. As a result, the easy diffusion of water or moisture becomes difficult, leading to a decrease in the water wettability of starch films. Similar findings were reported by Tambe *et al*. (2016).

The film-forming properties of coating materials play a crucial role in determining their water absorption characteristics. Coatings with good film-forming properties tend to form a continuous and uniform layer on the substrate, which helps to create a barrier against water penetration. Incorporating flat mineral pigments such as nanoclay into coatings can enhance their water barrier properties. Nanoclay particles have a plate-like structure, and when dispersed within the coating matrix, they create a tortuous pathway for water molecules. This means that water absorption is impeded as it needs to navigate through a complex path around the nanoclay particles. This tortuous pathway increases the diffusion length for water and slows down its penetration into the coating. Furthermore, nanoclay particles can also contribute to the reinforcement of the coating matrix by improving its mechanical properties, such as stiffness and strength. This reinforcement can further enhance the resistance of the coating to water absorption. This can be beneficial in various applications where water resistance is desired, such as in packaging materials or protective coatings.

Surface Smoothness

A one-way analysis of variance indicated a statistically significant difference among the surface smoothness values of the 9 tested paper types at a significance level of 5%. The surface smoothness values were grouped into three categories across all treatments. Figure 3 illustrates the average surface smoothness variations for the 9 paper types. Figure 3 demonstrates that the highest surface smoothness value is associated with the second-coated white-top linerboard, while the lowest value belongs to the first-coated white-top linerboard samples.

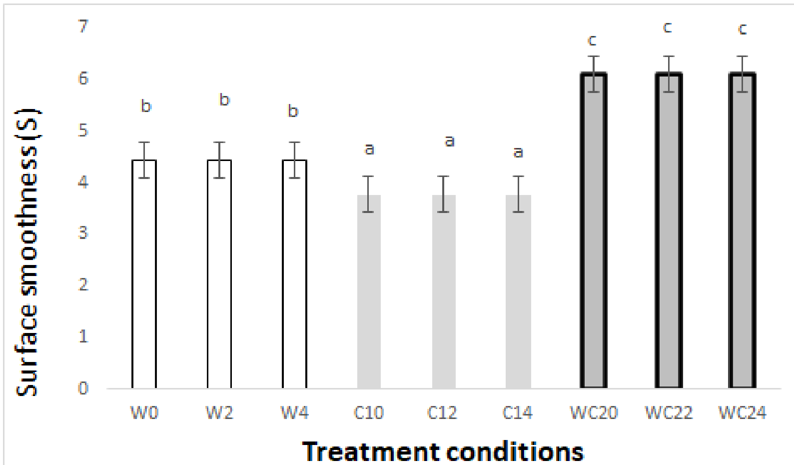

Fig. 3. Comparison of the average surface smoothness of different papers

The surface smoothness of papers, which is one of the indicators of printability, varies significantly. The use of double coating resulted in increased smoothness. In this research, the surface of the paper coated twice with nano-polyurethane and nano-clay exhibited relatively good uniformity and smoothness due to the higher specific surface area of nano-clay and its favorable particle shape. The coating process fills the cavities and empty spaces between fibers, leading to an improvement in surface smoothness. However, with the implementation of supercalendering operations, this feature will undoubtedly show further improvement compared to the control sample (Asadi Khansari and Dehghani Firouzabadi 2013).

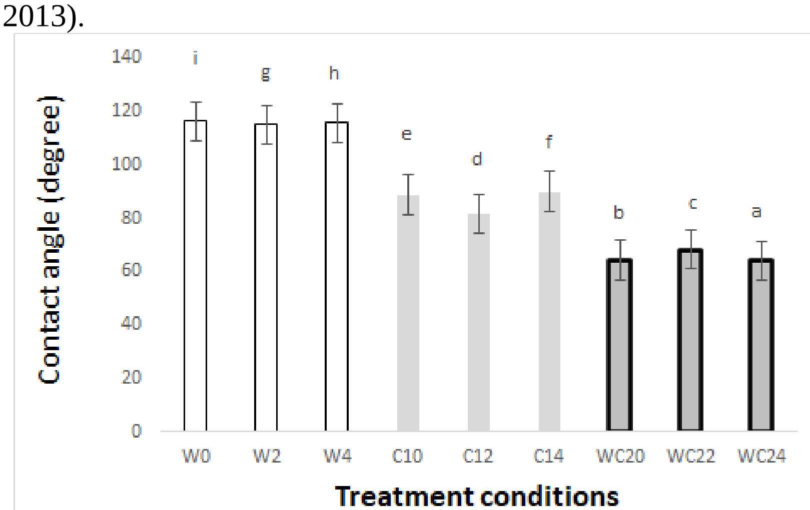

Fig. 4. Comparison of the average contact angle of different papers

Contact Angle

ANOVA revealed a statistically significant difference among the contact angle values of the 9 tested paper types at a significance level of 5%. The contact angle values were grouped into six categories across all treatments. Figure 4 illustrates the average contact angle variations for the 9 paper types. Figure 4 demonstrates that the highest contact angle value is associated with the control sample, while the lowest value belongs to the samples that have been coated twice and frozen for 4 months.

Tensile Index in the Direction of the Machine

A one-way analysis of variance demonstrated a statistically significant difference among the tensile index in the direction of the machine values of the 9 tested paper types at a significance level of 5%. The tensile index in the direction of the machine values were grouped into five categories across all treatments. Figure 5 illustrates the average variations in tensile index in the direction of the machine for the 9 paper types. Figure 5 demonstrates that the highest value of tensile index in the direction was associated with the control sample, while the lowest value belonged to the white-top linerboard samples that had been coated twice and frozen for 4 months.

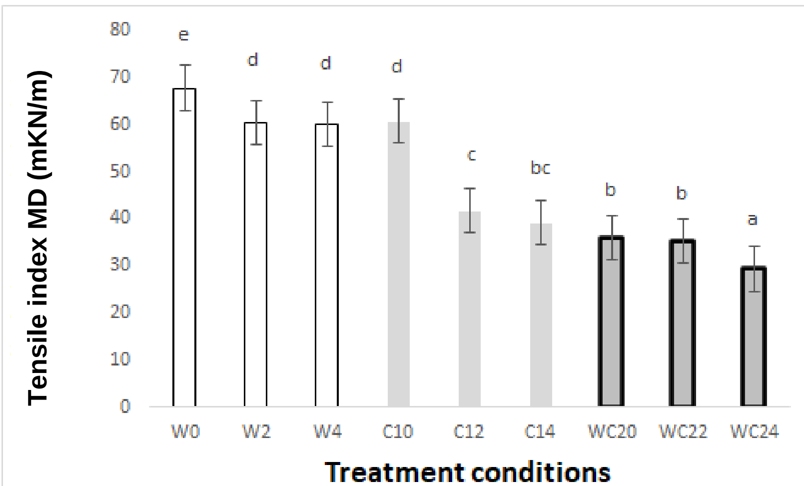

Fig. 5. Comparison of the average tensile index in the direction of the machine of different papers

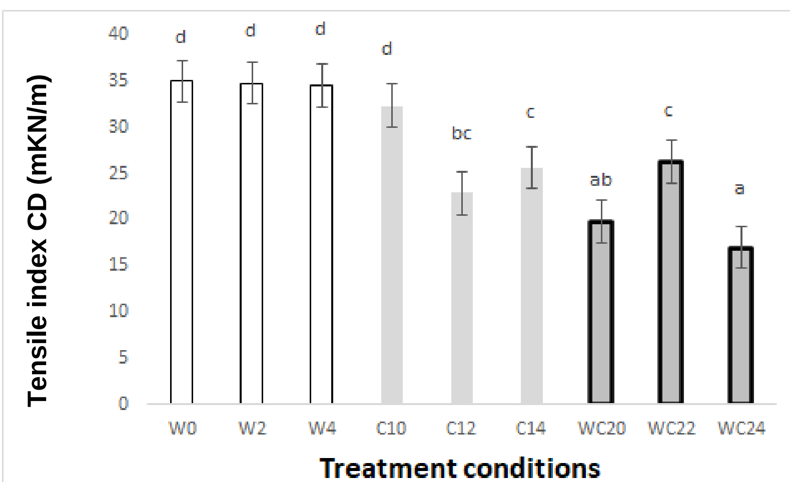

Fig. 6. Comparison of the average tensile index in cross-machine of different papers

Tensile Index in the Cross-Machine Direction

A one-way analysis of variance revealed a statistically significant difference among the tensile index in the cross-machine direction values of the 9 tested paper types at a significance level of 5%. The tensile index in the cross-machine direction values were grouped into four categories across all treatments. Figure 6 illustrates the average variations in tensile index in the cross-machine direction for the 9 paper types. Figure 6 demonstrates that the highest value of tensile index in the cross-machine direction was associated with the control sample, while the lowest value belonged to the white-top linerboard samples that have been coated twice and frozen for 4 months.

Burst Strength Index

ANOVA indicated a statistically significant difference among the burst strength index values of the 9 tested paper types at a significance level of 5%. The burst strength values were grouped into five categories across all treatments. Figure 7 illustrates the average variations in burst strength index for the 9 paper types. Figure 7 demonstrates that the highest value of burst strength index was associated with the one-coated liner paper, while the lowest value belonged to the samples of white-top linerboard that have been coated twice.

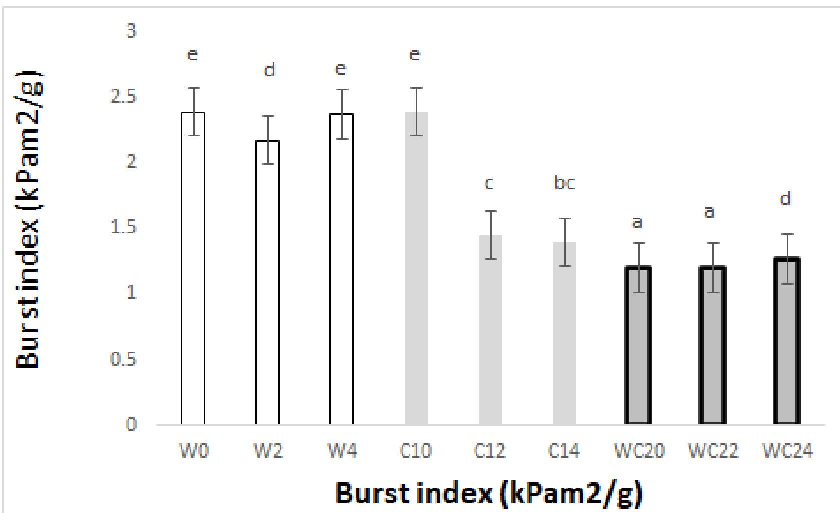

Fig. 7. Comparison of the average burst index strength of different papers

Tear Index in the Direction of the Machine

A one-way analysis of variance (ANOVA) revealed a statistically significant difference among the tear index in the direction of the machine values of the 9 tested paper types at a significance level of 5%. The tear index in the direction of the machine values were grouped into five categories across all treatments. Figure 8 illustrates the average variations in tear index in the direction of the machine for the 9 paper types. Figure 8 demonstrates that the highest value of tear index in the direction of the machine was associated with the control sample that had been frozen for 4 months, while the lowest value belonged to the white-top linerboard samples that had been coated twice and frozen for 2 months.

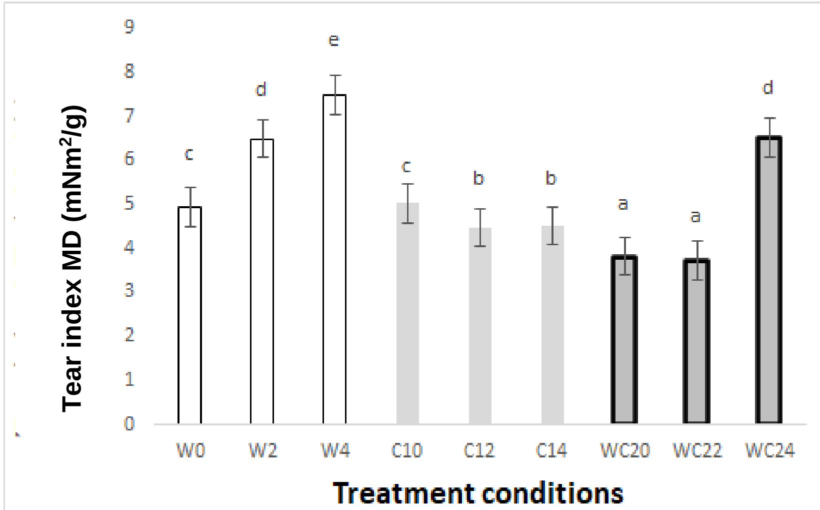

Fig. 8. Comparison of the average tear index in the direction of the machine of different papers

Tear Index in the Cross-Machine Direction

A one-way analysis of variance (ANOVA) showed that there was a statistically significant difference among the tear index in the cross-machine direction for the 9 tested paper types at a significance level of 5%. The tear index in the cross-machine direction were grouped into six categories across all treatments. Figure 9 illustrates the average variations in tear index in the cross-machine direction for the 6 paper types. Figure 9 demonstrates that the highest value of tear index in the cross-machine direction was associated with the control sample that has been frozen for 4 months, while the lowest value belonged to the white-top linerboard samples that had been coated twice and frozen for 2 months.

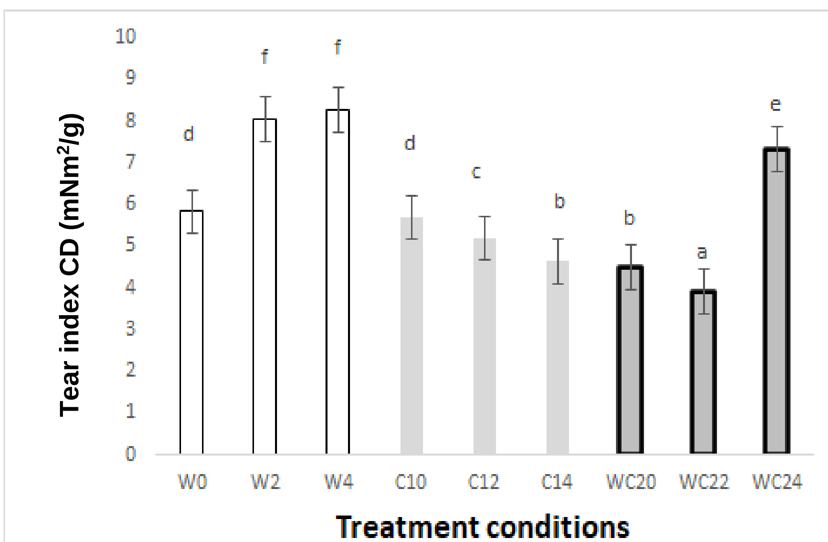

Fig. 9. Comparison of the average tear index in cross-machine of different papers

Ring Crush Test in Machine Direction

A one-way analysis of variance (ANOVA) indicated that there was a statistically significant difference among the ring crush test in machine direction values in the machine direction for the 9 tested paper types at a significance level of 5%. The ring crush test in machine direction values were divided into two groups across all treatments. Figure 10 illustrates the average variations in ring crush test in machine direction for the 9 paper types. Figure 10 demonstrates that the highest value of ring crush test in machine direction was associated with the control sample that had been frozen for 4 months, while the lowest value belonged to the white-top linerboard samples that have been coated twice and frozen for 4 months.

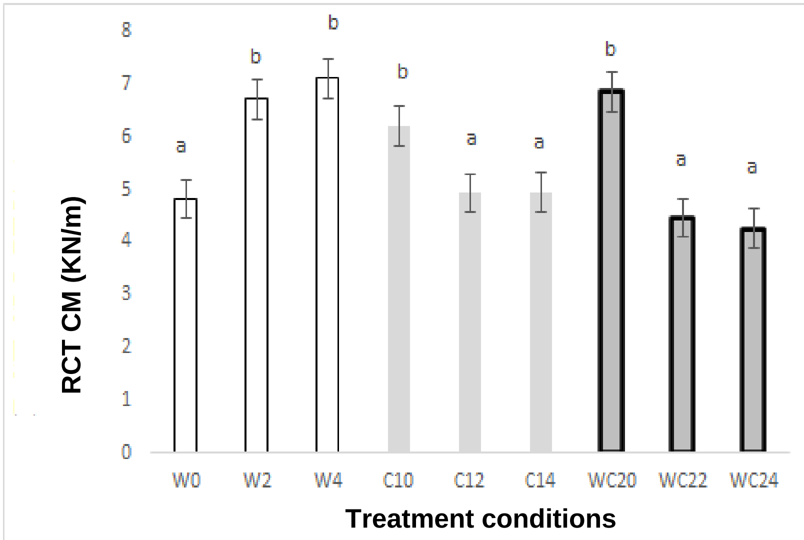

Fig. 10. Comparison of the average ring crush test in the direction of the machine of different papers

Ring Crush Test in Cross-Machine Direction

A one-way analysis of variance revealed that there was a statistically significant difference among the ring crush test in cross-machine direction for the 9 tested paper types at a significance level of 5%. The ring crush test in cross-machine direction were grouped into five categories across all treatments. Figure 11 illustrates the average variations in ring crush test for the 9 paper types in the cross-machine direction. Figure 11 demonstrates that the highest value of ring crush test in cross-machine direction was associated with the control sample that had been frozen for 4 months, while the lowest value belonged to the white-top linerboard samples that had been coated once and frozen for 2 months.

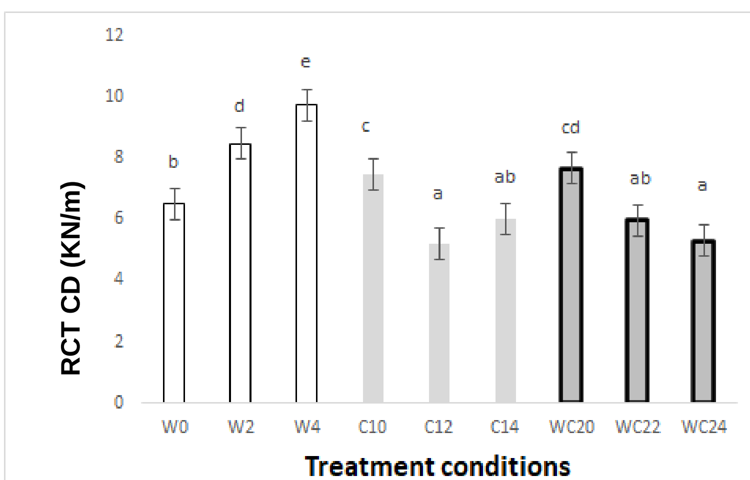

Fig. 11. Comparison of the average ring crush test in cross-machine direction of different papers

Brightness

A one-way analysis of variance (ANOVA) indicated that there was a statistically significant difference among the brightness values for the 9 tested paper types at a significance level of 5%. The brightness values were categorized into three groups across all treatments. Figure 12 illustrates the average variations in brightness for the 9 paper types. Figure 12 demonstrates that the highest value of brightness was associated with the control sample that had been frozen for 4 months, while the lowest value belonged to the samples that have been coated once.

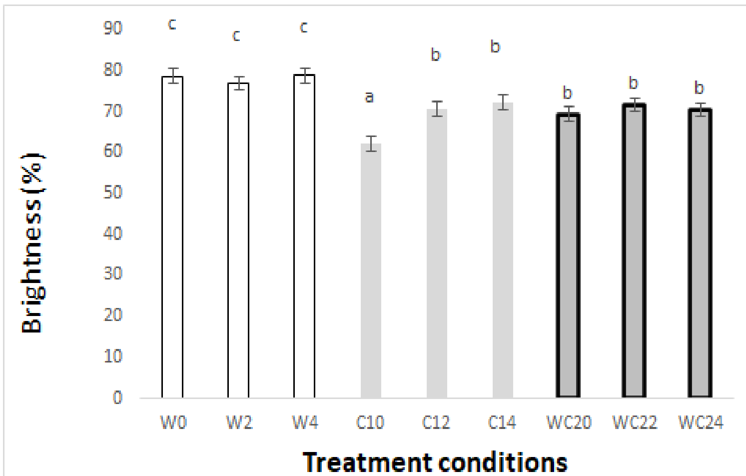

Fig. 12. Comparison of average brightness of different papers

The applied coatings, whether single-layer or double-layer, led to a reduction in the mechanical properties. These results indicate that the flexibility of the coated paperboards was higher compared to uncoated ones. The decrease in tensile strength and tear resistance may be due to the penetration of the coating solution into the fiber network, resulting in cellulose fiber swelling and disruption of fiber-to-fiber connections. It appears that the increase in the solvent (water) content leads to an increase in the penetration of the coating materials into the fiber network and a decrease in tensile strength. This reduction in tensile strength and tear resistance is consistent with the findings of Reim *et al*. (2006) and Park *et al*. (2000a,b). Factors influencing the tear index include fiber length, inter-fiber bond strength, and individual fiber strength. Due to the presence of nanoparticles between the fibers and the weakening of the fiber-to-fiber bonds, the inter-fiber bond strength usually decreases, while the individual fiber strength remains relatively unchanged. Coatings, especially in the case of double-layer coatings, have contributed to a decrease in tear resistance. This can be attributed to the low consumption of nanoparticles and their nanoscale dimensions. The small size of these particles and their limited ability to create physical distance between the microfibrils of the fibers have had a minor impact on the relative reduction of bond area and, consequently, the paper's strength.

Microscopic Studies of the Structure of Paper (FE-SEM)

Figure 13 displays scanning electron microscopy (FE-SEM) micrographs for the surfaces of the following white-top linerboard samples: (a) control white-top linerboard (4-month freeze), (b) control white-top linerboard (no freeze), (c) white-top linerboard coated once, (d) white-top linerboard coated once (4-month freeze), (e) white-top linerboard coated twice, and (f) white-top linerboard coated twice (4-month freeze).

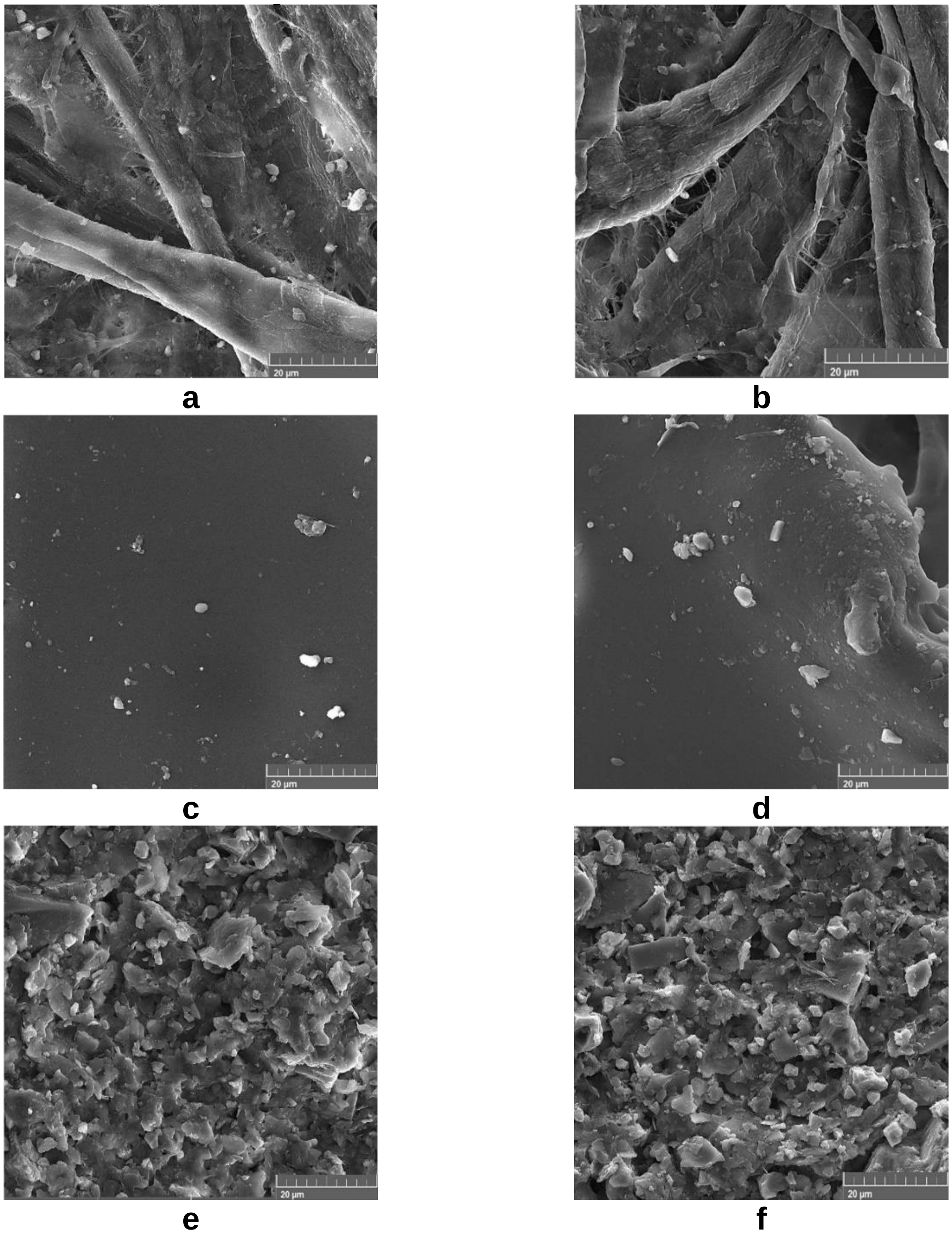

Fig. 13. Surfaces of the following white-top linerboard samples (a) control white-top linerboard, (b) control white top liner (4-month freeze), (c) white-top linerboard coated once, (d) white-top linerboard coated once (4-month freeze), (e) white-top linerboard coated twice, and (f) white-top linerboard coated twice (4-month freeze)

The electron microscopy structure revealed that in the coated samples, there were pores and cavities between the coated fibers, and filling these cavities resulted in uniformity and improved print quality. The coating material fills these voids and creates a smooth surface, enhancing the adhesion between the ink and the coated fibers. Additionally, the high-resolution images showed that the coated paperboard had a smoother surface and fewer irregularities, leading to improved ink coverage and reduced ink absorption into the fiber network. This results in sharper and more vibrant prints, as well as reduced ink bleeding and improved color reproduction. The increased surface smoothness and reduced fiber-to-fiber gaps also contributed to better print quality by preventing the spread of ink beyond the intended areas and improving the overall print resolution.

CONCLUSIONS

1. The findings of this study have provided valuable insights into the topic, filling several gaps in knowledge. Firstly, it has been demonstrated that covering and freezing the samples resulted in significant reductions in water absorption, indicating that this combination can effectively enhance the water resistance of the materials. The water absorption of the samples decreased by 197% after being covered and frozen.
2. The coating and freezing process led to improvements in surface smoothness and increased contact angle, suggesting that it is possible to achieve enhanced surface properties and increased hydrophobicity through this approach. The surface smoothness of the samples increased by 38% after coating and freezing, also the contact angle of the samples increased by 80.9% after coating and freezing.
3. The study also revealed substantial decreases in various strength properties, such as tensile index, burst strength index, tear index in the machine direction, and ring crush test in the cross-machine direction, after covering and freezing. This highlights the trade-off between improved water resistance and decreased mechanical strength, which should be carefully considered when developing environmentally friendly coatings for food containers. Tensile and tear index in the direction of the machine, burst strength index, and ring crush test in cross-machine direction were decreased by 88.3, 29, 99.2, and 67.1% after being covered and frozen respectively.
4. Brightness decreased by 13.4% after coating and freezing.
5. In summary, this study has contributed to the understanding of the topic by providing insights into the effects of covering and freezing on the properties of the materials. It has shed light on the potential for environmentally friendly coatings in food containers while raising important considerations about trade-offs and coating strategies for achieving optimal performance.

REFERENCES CITED

Aloui, H., Khwaldia, K., Slama, M. B., and Hamdi, M. (2011). "Effect of glycerol and coating weight on functional properties of biopolymer-coated paper," *Carbohydrate Polymers* 86(2), 1063-1072. DOI: 10.1016/j.carbpol.2011.06.026

Asadi Khansari, R., and Dehghani Firouzabadi, M., (2013). "Introduction of new paper and cardboard in food packaging," *Journal of Packaging Science and Technology* 16(4), 46-57.

Asadi Khansari, R., Dehghani Firouzabadi, M., and Resalati, H. (2015). "The effect of biodegradable coatings on the barrier properties of papers," *Iranian Journal of Wood and Paper Industries* 7(1), 91-101.

Hubbe, M. A., Rojas, O. J., Lucia, L. A., and Sain, M. (2008). "Cellulosic nanocomposites: A review," *BioResources* 3(3), 929-980. DOI: 10.15376/biores.3.3.929-980

Julkapli, N. M., and Bagheri, S. (2016). “Developments in nano-additives for paper industry,” *Journal of Wood Science* 62, 117-130. DOI: 10.1007/s10086-015-1532-5
Park H. J., Si, H. H., Seung, T. L., Dong, H. S., Sang ,Y. C. H., and Keum, T. H. (2000a). “Grease resistance and mechanical properties of isolated soy protein-coated paper,” *Journal of the American Oil Chemists' Society* 77(3), 269-273.
Park, H. J., Kim, S. H., Lim, S. T., Shin, D. H., Choi, S. Y., and Hwang, K. T. (2000b). “Grease resistance and mechanical properties of isolated soy protein-coated paper,” *Journal of the American Oil Chemists' Society* 77(3), 269-273. DOI: 10.1007/s11746-000-0044-2
Qie, R., Zajforoushan Moghaddam, S., and Thormann, E. (2023). “Dopamine-assisted layer-by-layer deposition providing coatings with controlled thickness, roughness, and functional properties,” *ACS Omega* 8(3), 2965-2972. DOI: 10.1021/acsomega.2c05620
Reis, A. B., Yoshida, C. M., Reis, A. P. C., and Franco, T. T. (2011). “Application of chitosan emulsion as a coating on kraft paper,” *Polymer International* 60(6), 963-969. DOI: 10.1002/pi.3023
Rhim, J. W., Lee, J. H., and Hong, S. I. (2006). “Water resistance and mechanical properties of biopolymer (alginate and soy protein) coated paperboards,” *LWT-Food Science and Technology* 39(7), 806-813. DOI: 10.1016/j.lwt.2005.05.008
Seo, K. S., Bajracharya, R., Lee, S. H., and Han, H. K. (2020). “Pharmaceutical application of tablet film coating,” *Pharmaceutics* 12(9), 853. DOI: 10.3390/pharmaceutics12090853
Tambe, C., Graiver, D., and Narayan, R. (2016). “Moisture resistance coating of packaging paper from biobased silylated soybean oil,” *Progress in Organic Coatings* 101, 270-278. DOI: 10.1016/j.porgcoat.2016.08.016
Vähä-Nissi, M., Koivula, H. M., Räisänen, H. M., Vartiainen, J., Ragni, P., Menttä, E., Kaljunen, T., Malm, T., Minkkinen, H., and Harlin, A. (2017). “Cellulose nanofibrils in biobased multilayer films for food packaging,” *Journal of Applied Polymer Science* 134(19), article 44830. DOI: 10.1002/app.44830

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