Original Scientific Paper https://doi.org/10.24867/GRID-2026-p79

The Influence of Paper Substrate and Coating Formulation on Barrier Properties of Packaging Papers

ABSTRACT

Strict EU PPWR and REACH regulations are pushing the packaging industry towards more sustainable and recyclable packaging materials. Although paper is an excellent sustainable and recyclable substrate, demanding packaging applications (e.g. barrier packaging papers) require functional coatings to provide appropriate barriers against moisture and grease and at the same time meet food contact safety requirements. Based on this demand, the presented study investigates the development of bio-based barrier coatings and their application on two different paper substrates (varying in grammage, surface and structural properties). Base papers were characterized for grammage, thickness, porosity (penetration dynamic analysis) and surface morphology (scanning electron microscopy). Following, four different bio-based coating solutions with different additives ((1) starch, (2) starch + bark-based CNF, (3) starch + bark-based CNF + AKD, (4) starch + bark-based CNF + PVA)) were developed and applied on the selected base papers using the rod coating technique.

The assessment of the coated paper samples regarding water vapour, oil and grease barrier properties was carried out. WVTR (at 50% and 85% RH), Cobb Unger, and KIT tests were evaluated and the results demonstrate how the selected paper base properties and coating formulation (with different additives) influence the barrier properties and provide solutions for over 95% bio-based barrier coating packaging paper.

The results of the study showed that the greatest influence on the barrier properties had the base paper grammage and starch coating, while the additives provided fine-tuning of the final barrier performance.

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1. Introduction

Fibre-based packaging is more and more popular due to its high recyclability rate; however, its porous, hydrophilic, and moisture-sensitive nature does not fulfil the requirements for food packaging. To enhance the functional properties of the substrate, the paper must be coated to impart barrier properties against gases, water vapor, oils, and greases. The final performance of the developed coated paper material depends on both the base paper characteristics and the coating formulation.

The characteristics such as porosity, roughness, and density of the uncoated base paper significantly influence the barrier properties attainable by coatings. A smooth, dense substrate enables flawless, consistent coating, which is crucial for attaining low permeability; in contrast, porous or uneven substrates may induce coating imperfections that serve as pathways for gas

and moisture ingress even after coating (Järnström et al., 2026; Marinelli et al., 2023; Rovera et al., 2020).

Besides base paper properties, coating and its functional additives are important for achieving barrier properties. Due to its natural origin, biodegradability, low cost, and excellent film-forming ability, starch is a promising material for developing sustainable barrier coatings for paper and paperboard (Li et al., 2019). When properly formulated and applied, starch-based coatings substantially improve barrier properties; however, the barrier performance is often limited by its sensitivity to moisture and humidity. Further enhancement can be achieved through the incorporation of functional additives that have to be selected carefully to improve barrier performance without compromising the recyclability of the coated paper (Bakker et al., 2022; Chi et al., 2023). Nanocellulose (CNF) as a reinforcing and tortuosity-building filler can improve barrier properties, particularly gas barrier; however, its hydrophilicity can limit performance when exposed to higher relative humidities (Afra, Mohammadnejad & Saraeyan, 2016; Chi et al., 2023; Lengowski et al., 2023; Wang et al., 2017). It can be obtained from different raw materials, one of them being the use of discarded bark biomass to produce bark-based CNF (Borrega et al., 2026). To overcome its hydrophilicity, alkyl ketene dimer (AKD) can serve as a sizing agent and hydrophobic modifier and polyvinyl alcohol (PVA) as a water-dispersible binder with favourable barrier characteristics (Christophliemk et al., 2017; Huang et al., 2023). AKD is a classic internal sizing agent that reacts with cellulose hydroxyl groups to impart hydrophobicity, thereby reducing water uptake and improving oil resistance. It forms covalent linkages with the cellulose surface, contributing to moisture barrier improvements that are particularly relevant when starch-based coatings are hydrophilic and prone to water uptake (Roberts, 1997). On the other hand, PVA is widely used as a water-soluble, non-toxic binder in starch-based coatings due to its film-forming ability and compatibility with CNF. PVA contribute to barrier performance through its dense film-forming capability and its potential to improve continuity and reduce defects that act as diffusion pathways. In starch-based systems, PVA often acts in synergy with other additives (CNF and AKD) to achieve balanced barrier properties without drastically increasing coat (Huang et al., 2023). When these components are combined in well-designed formulations, the coating can synergistically reduce water vapour transmission rate (WVTR) and enhance grease/oil resistance, while supporting recyclability of the cellulose substrate.

The influence of PVA and AKD additives in coatings on the barrier properties of packaging paper was investigated by Shen et al. (2021). The results showed that PVA coating reduced the WVTR by forming a continuous film that sealed the pores of the surface. The incorporation of AKD improved the water resistance further, resulting in lower Cobb values. They demonstrated that combining a film-forming polymer with a hydrophobic agent can improve the barrier properties of the paper.

Despite the high interest in barrier coatings for paper packaging, a clear gap exists in terms of studies concentrating on the evaluation of the impact of each individual component on the obtained barrier performance. Besides, research specifically investigating the application of bark-derived cellulose nanofibrils (CNF) in such coatings remains limited. Thus, the present work aims to fill this research gap by analysing the effects of the coating formulation (including bark-based CNF) and its individual components on the barrier properties of coated paper. The study investigates the effect of two different base papers (with different properties) and four coating formulations with different functional additives (bark-based CNF, AKD, PVA) on the resulting barrier properties of coated paper. This approach offers a thorough understanding of the interactions between the substrate and coating formulation and their contribution to the barrier performance of the developed coated paper materials to water vapour, oils and greases.

2. Matherials and Methods

In the presented research, two different base papers (Paper 1 and Paper 2) produced by Goričane papermill (Slovenia) were used for coating application. The base papers were firstly characterized regarding basic and surface properties. Grammage (ISO 536), thickness (ISO 534), smoothness (ISO 8791-2), porosity (ISO 5636-3) and filler content at 500°C (ISO 1762) were determined. Additionally, the surface morphology and cross-section of the base papers were characterised using a scanning electron microscope (SEM; JEOL JSM-6060 LV, Japan).

Additionally, the samples were characterised with a penetration dynamics analyser Emtec PDA.C 02 (Emtec, Germany). During this method, a testing sample is placed in contact with a testing liquid. The instrument continuously monitors the drop in the ultrasonic signal due to the penetration (capillary absorption) of 16% isopropyl alcohol (IPA) to determine the porosity of the sample surface.

Following this, four different bio-based coating solutions based on starch (S) A4692 (Avebe, The Netherlands) with different additives to enhance the barrier performance were developed. As additives, unbleached bark-based cellulose nanofibers (CNF) VTT20 produced in the scope of the SuperBark project, alkyl ketene dimer (AKD) and polyvinyl alcohol (PVA) Exceval AQ-4104 (Kuraray, Japan) were used. All final developed coating solutions had a bio-based content of at least 95%.

The developed coating solutions (Table 1) were applied in one layer (10 g/m^2) on the base papers with a laboratory rod coater RK Print Coater. After coating,

the samples were dried in a ventilation oven under controlled conditions at 105°C for one minute.

The prepared coated samples were evaluated regarding barrier properties important for food packaging paper. Water vapour transmission rate (WVTR) was determined in accordance with the gravimetric method (ISO 2528 standard) at 50% as well as 85% relative humidity and 23°C. Oil absorption (Cobb Unger 30 s) was evaluated according to SCAN-P 37:77 and grease resistance (KIT test) according to TAPPI T559 standard procedure. All samples were conditioned under standard conditions in accordance with ISO 187 prior to testing.

Table 1
Developed coating solutions and contents of the components used

	Solid content (wt%)	**The content (%) of each component in the coating formulation**			
Sample		Starch	CNF	AKD	PVA
S	25	100	0	0	0
S+CNF	20	97	3	0	0
S+CNF+AKD	20	92	3	5	0
S+CNF+PVA	20	92	3	0	5

3. Results and Discussion

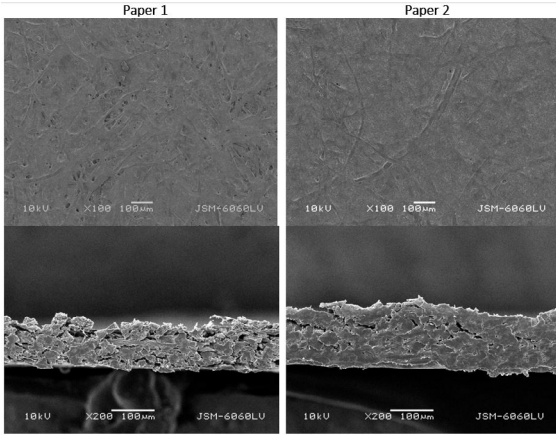
The evaluation of the selected base papers revealed that Paper 1 and Paper 2 differ considerably in their structural and surface properties as presented in Table 2. Paper 1 had a lower grammage (51.2 g/m²), thickness (57 µm), and filler content (7%) compared to Paper 2 (90 g/m², 90 µm, and 13%, respectively). Due to lower filler content and different surface pre-coating, Paper 1 also exhibited higher Bendtsen smoothness and porosity values (90 mL/min for both), whereas Paper 2 showed values of 50 and 1.5 mL/min. Both papers were sized using a rosin-based sizing agent.

Table 2
Basic properties of the base papers used

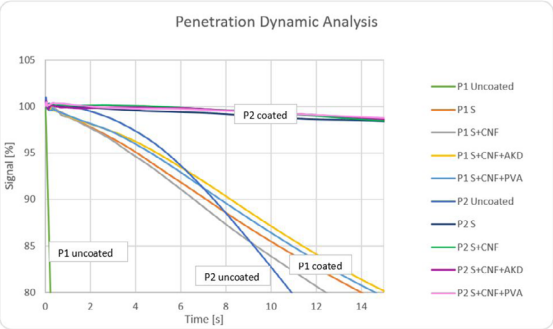
	Paper 1	**Paper 2**
Grammage (g/m²)	51.2	90
Filler at 500°C (%)	7	13
Thickness (µm)	57	90
Smoothness Bendtsen (mL/min)	90	50
Porosity Bendtsen (mL/min)	90	1.5
Type of sizing agent	Rosin sizing agent	Rosin sizing agent

The differences in porosity, thickness, and surface structure between the two paper substrates were also clearly observed in the SEM images (Figure 1). Paper 1 exhibited a more open and heterogeneous surface structure, whereas Paper 2 showed a denser and more compact structure with a smoother surface.

To gain a deeper insight into paper porosity, penetration dynamics analysis of all coated and uncoated samples was performed. For the evaluation of porosity, 16% isopropanol (IPA) was used as it is a very wetting fluid, so penetration is governed mainly by capillary absorption and the natural pore structure of the substrate. The decrease in ultrasonic signal intensity in the first 15 seconds after the sample was in contact with the testing liquid is presented in Figure 2. The penetration dynamics of the two uncoated base paper substrates, Paper 1 (P1) and Paper 2 (P2), show a significant difference. The uncoated Paper 1 shows an extremely rapid drop in the signal, reaching the 95% threshold almost immediately, which indicates a very porous network with fast capillary absorption. In contrast, uncoated Paper 2 shows a considerably lower penetration rate, indicating a significantly denser pore structure of the base. All coated samples on Paper 1 decrease the porosity of the paper (the signal drops to 95% between 4 and 6 seconds), which is good evidence that the coatings are successfully blocking the open surface pores and limiting capillary transport. However, the porosities of coated samples on Paper 1 are very similar to uncoated Paper 2. The coatings on the denser Paper 2 achieve almost complete barrier performance against 16% IPA. The signal intensity of all coated P2 samples is almost the same over the entire 15-second measurement time and is above 98%, with excellent resistance to wetting and penetration. PDA results show that the application of coatings seals the porous network of more porous base papers (Paper 1) efficiently, but the combination of dense base paper (Paper 2) with barrier coatings forms a near-impermeable structure to polar wetting liquids.



» **Figure 1:** *SEM images of the base papers. Surface (top) and cross-section (bottom)*

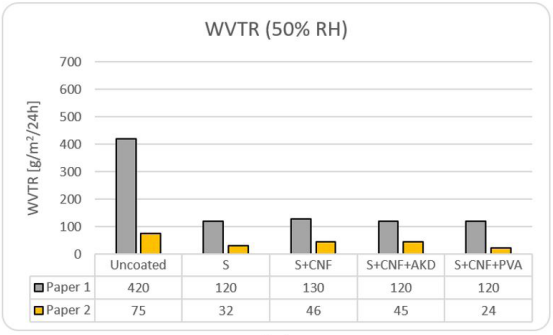


» **Figure 2:** *Penetration dynamic chart with 16% IPA as a testing liquid. P1 presents samples on Paper 1 and P2 presents samples on Paper 2*

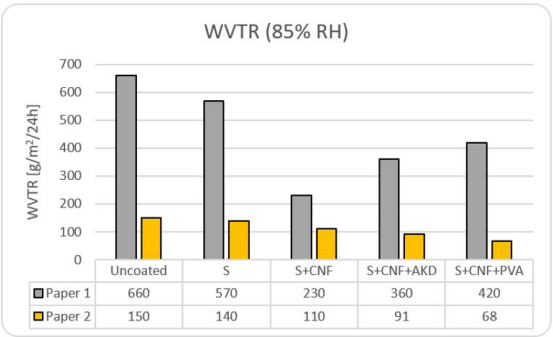
Water vapour transmission rate (WVTR) measurements at 50% and 85% relative humidity (RH) revealed significant differences in barrier performance between the two substrates (Figure 3). Paper 2 outperformed Paper 1 by more than five times when uncoated; however, WVTR at 50% RH is almost halved in comparison to WVTR at 85% RH. Large differences were also evident when both papers were coated with the developed coating formulations, which could be attributed to the thickness and porosity of the base papers. At 50% RH, uncoated Paper 1 exhibited high WVTR (420 g/m^2/24h), which decreased sharply upon applying coating S (120 g/m^2/24h); however, additives in the coating formulations did not further decrease WVTR. WVTR of samples on Paper 2 systematically reduced from 75 g/m^2/24h (uncoated) down to a minimum of 24 g/m^2/24h with the S+CNF+PVA formulation. Higher humidity (85% RH) increased WVTR overall. On Paper 1, the S+CNF formulation provided the strongest barrier enhancement, reducing WVTR to (230 g/m^2/24h), whereas additional AKD or PVA proved counterproductive. Conversely, samples on Paper 2 displayed an improvement with each added component from Uncoated (150 g/m^2/24h) to the sample coated with S+CNF+PVA coating solution (68 g/m^2/24h). Overall, S+CNF is optimal for upgrading the porous Paper 1 at high RH, while S+CNF+PVA yields the best performance among coating solutions on the denser Paper 2 substrate at both relative humidities.

Besides WVTR barrier properties, the prepared coated samples were evaluated also for oil and grease barrier properties.

Oil absorbency was evaluated with the Cobb Unger testing method after 30 seconds of sample/oil contact. A lower Cobb Unger value represents lower oil absorption. Grease resistance was evaluated with the KIT test, where the highest value (12) presents superior grease resistance and where the solution applied on the surface of the sample does not stain the surface. As can be seen from Figure 4 a huge difference is evident between uncoated papers, where uncoated Paper 1 showed extremely poor oil barrier with a Cobb Unger value of 17.1 g/m^2 and a KIT value of 0, while uncoated Paper 2 exhibited much lower oil absorbency (1.4 g/m^2) and a KIT value of 1, further

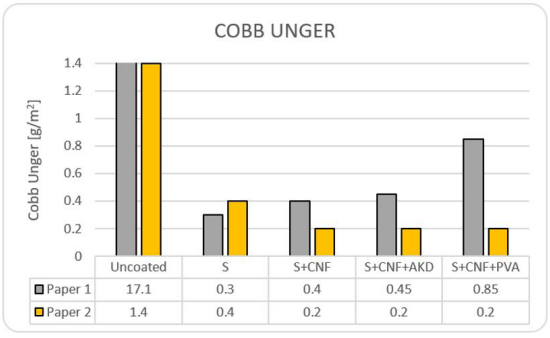


(a)

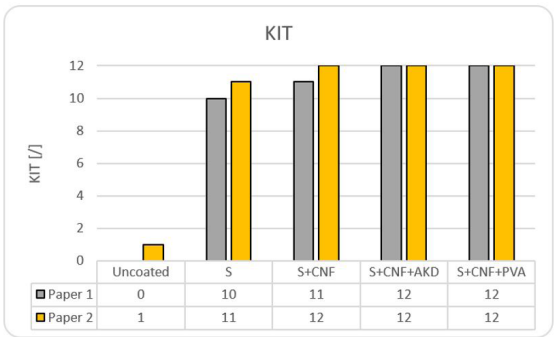


(b)

» **Figure 3:** *WVTR of tested samples at 50% RH (a), and 85% RH (b)*



(a)



(b)

» **Figure 4:** *Cobb Unger (a) and KIT values (b) for tested samples*

confirming the importance of a denser structure and higher grammage of the base paper. After coating application, both barriers increased significantly. With starch coating (S), oil absorbency dropped to 0.3 and 0.4 g/m^2, while KIT values increased to 10 or 11. The addition of CNF and other additives further improved the Cobb Unger on Paper 2, while on Paper 1 the barrier decreased slightly. However, KIT values increased to the highest value (12) and remained the same for all coating formulations with additives (CNF, AKD or PVA) on Paper 2.

4. Conclusion

In conclusion, the evaluation of water vapour and oil and grease barrier properties emphasises the importance of the choice of the base paper substrate. The denser and less porous structure of Paper 2 consistently outperformed Paper 1 under all testing conditions and thus had the most critical impact on overall barrier efficiency. The primary starch coating (S) is critical in establishing the basic barrier, while the addition of functional additives is mostly a fine-tuning process to adjust specific barrier properties. In particular, the CNF was found to be critical for enhancing WVTR performance on the more porous Paper 1 substrate under high relative humidity (85% RH) conditions. However, the fully formulated S+CNF+PVA coating mixture showed the best overall performance, with the lowest water vapour transmission rates and excellent oil and grease resistance. In particular, no significant differences in the grease and oil barrier properties were observed for Paper 2 with different functional additives (CNF, AKD or PVA) added to the starch base, as all formulation combinations provided excellent grease repellency and low oil absorption.

5. Acknowledgment

The SuperBark project (grant agreement No. 101112447) is supported by the Circular Bio-based Europe Joint Undertaking and its members.

Funded by the European Union. Views and opinions expressed are however, those of the author(s) only and do not necessarily reflect those of the European Union or CBE JU. Neither the European Union nor the CBE JU can be held responsible for them.

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