MOISTURE AND OIL RETENTION IN COATED PAPERS FOR FLEXIBLE FOOD PACKAGING: A STUDY ON BARRIER PROPERTIES AND SURFACE CHARACTERISTICS

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Abstract: *Coated paper exhibits promising potential for application in flexible food packaging due to its positive sustainability perception among consumers, recyclability potential, good formability, and printability. One crucial aspect of food packaging is its ability to retain moisture in food items such as fruits/vegetables and prepared meals, as well as to prevent the loss of oil in products with high oil content. Additionally, maintaining a low humidity environment is important for preserving the crispness of dry foods, such as cookies and spices. In its natural state, paper possesses weak barriers against moisture and oil. Therefore, the coating applied to the paper must offer adequate barrier properties against water vapor, water, and oil to meet these requirements and ensure sufficient shelf life for the food items. Within the framework of the CORNET-TETRA project HBC.2021.0288 REPAC², a total of 15 commercially available coated papers were selected based on their production process, composition, and performance characteristics such as heat sealability, as well as permeability to oxygen gas and water vapor. To assess moisture and oil retention, water vapor permeability was measured at 23°C and 85% relative humidity, and Cobb water and oil absorption were evaluated after 1800 seconds. Additionally, water and oil contact angles were measured to assess the polarity of the coated surface. Notably, the water vapor permeability values, which are crucial for preserving dry foods, exhibited high variation among the different coated papers,* *ranging from 0.2 g/m²/d to immeasurably high values. The Cobb water and oil absorption values, important for respectively preserving moist and oily foods, showed a slightly lesser extent of variation, ranging from 0.5 to 95 g/m² for water and 0.8 to 46 g/m² for oil. In conclusion, this study provides valuable insights into the relationship between the barrier properties and physical surface properties of coated papers utilized in flexible food packaging.*

Keywords: Flexible food packaging, coated paper, Cobb absorption, water vapor permeability, surface characterization

1 INTRODUCTION

The European Green Deal sets important policy objectives to further advance the sustainable transition of the packaging value chain. One key objective is to ensure the reusability or recyclability, in an economically viable manner, of all packaging in the EU market (European commission, 2022). Given the long history of mechanical recycling of paper, the consumer preference for paper over plastic, and the renewable nature of paper fibers, a transition from plastic to coated paper is being considered in many primary flexible food applications. An increasing number of brand owners are widely promoting their shift to paper in the media to demonstrate their commitment to a sustainable transition.

However, uncoated paper exhibits low barrier properties, which necessitates the application of a functional coating. The transition to coated paper introduces additional considerations for food companies. Specifically, the barrier properties play a critical role in determining product shelf life (Adibi et al., 2023). Moisture absorption from the surrounding environment can lead to a loss of crispness in dry products, while liquid leakage may result in packaging stains, rendering it unappealing to consumers.

Within the context of the CORNET-TETRA REPAC² project (HBC.2021.0288), commercially available materials for primary food applications were utilized, and their barrier and surface properties were evaluated (UHasselt, 2022). This study aims to investigate potential correlations between the obtained results, aiming to gain comprehensive insights into the relationship between the barrier properties and the physical surface properties. Such insights can contribute to enhancing flexible packaging of food products, thereby ensuring and maintaining their quality and safety standards.

2 MATERIAL AND METHODS

2. 1 Materials

During the REPAC project, 15 food-grade and heat-sealable materials were selected, each differing in various aspects such as barrier properties, production process, and polymer origin in the coating. To maintain confidentiality, which is inherent to this type of research project involving commercial materials, exclusively descriptions, as supplied by the companies, were used. Out of the 15 materials, 12 were coated with a dispersion coating, 2 with an extrusion coating, and 1 with a wax coating. Thirteen materials were supplied as commercially available packaging papers, while two materials ('Dispersion 3' and 'Dispersion 11' in Table 1) were coated in the laboratory.

2. 2 Surface characterization

Contact angles were measured using static droplets of 3 µl water and sunflower oil, using a microsyringe, on the seal coating of each sample. Measurements were done on an OCA 50 contact angle device (Dataphysics Instruments GmbH, Filderstadt, Germany) and fitting of the droplet geometry by a tangent procedure averaging left and right contact angle. Samples are attached to a microscope slide. An average of 10 measurements was taken for each material.

The roughness of the sealing sides, which come into contact with the food product, an average of 10 measurements with the air leak method was taken for each sample, following ISO 8791:2, using the L&W Bendtsen tester and through a visual assessment of the arithmetical mean height (Sa), taken an average of two measurements for each sample, following ISO 25178, using Keyence 3D topographical imaging with magnification lens 20x. These results are expressed in mL/min and µm, respectively.

The main component of the seal-side is identified through attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR) by fitting the spectra with a dedicated database. This analytical approach serves to elucidate the components as described by the companies and may yield a more precise nomenclature for said components. Furthermore, this test has the potential to unveil novel components if the previously described components were absent within the seal-side of the paper.

2. 3 Barrier characterization

There are two types of barriers tested in this study:

i.) Barrier against liquids, specifically sunflower oil and water, determined by weighing the absorbed mass after an exposure of 1800 s, taken an average of five measurements for each sample, following ISO 535, using the Rycolab Semiautomatic Cobb water and oil absorption tester;

ii.) Barrier against water vapor, determined using a Modulated Infrared Sensor with single measurements, following ASTM F1249, using the Permatran-W model 3/33 MG, where the outer side is exposed to 23°C and a relative humidity of 85% and the carrier gas is at the seal side.

3 RESULTS AND DISCUSSION

The table presented below provides grammages the ATR-FTIR main identified components of the seal-sides, and outcomes of all coated papers. Among the 15 samples, 4 samples showed an extremely high Water Vapor Transmission Rate (WVTR), which could not be accurately measured. As a result, a value of 1000 $g/m^2/d$ was assigned to these samples for the purpose of facilitating statistical analysis. It should be noted that this assigned value underestimates the true WVTR value. Based on the unsatisfactory barrier performance and relatively large standard deviations, it can be inferred that the coated papers produced under laboratory conditions, 'dispersion 3' and 'dispersion 11', are likely of inferior quality compared to most of the coated papers manufactured under industrial conditions. Additionally, 'dispersion 2' and the wax-coated paper also exhibited poor barrier properties, suggesting a similar deficiency in the coating process used for these materials. This deficiency in coating quality is substantiated by the FTIR results. In three out of the four materials exhibiting the lowest performance in terms of WVTR, cellulose is identified as the primary component. Cellulose is a material which is not heat sealable in its natural state (Bamps et al., 2023), which suggests that the infrared radiation directly impinges upon the paper surface or a cellulose layer, where seal coating should ideally be present if the paper were uniformly coated. The extremely low WVTR-values of 'dispersion 1' and 'extrusion 1' are respectively better as or comparable to a 25 µm film of polyethylene or polypropylene (1 - 4 $g/m^2/d$), which are polymer types known for its excellent water vapor barrier properties. In both coated papers, polyethylene was identified within the sealing layer, which is well-known for its excellent moisture barrier properties. With such low WVTR, it is possible, disregarding other nutritional requirements such as oxygen content, to consider the coated paper for extremely moisture-sensitive products that are at risk of losing crispness, such as cookies, crackers, and potato chips (Robertson, 2013).

The results of the Cobb tests conducted with water and sunflower oil under extended exposure of 1800 seconds, provide indications of the ability of the coated papers to retain water and oil within the packaging, simulating a significant shelf life limiting factor for food products with high moist and oil content. As evident from the table, the previously mentioned 'dispersion 1' and 'extrusion 1', but also 'extrusion 2' can be considered for moist food applications. For oil-rich food products, there are multiple coated papers that may be suitable. 'Dispersion 4' stands out positively with a relatively low oil absorption rate of 0.8 g/m². Due to the unclear spectra, potentially resulting from a blend within the sealing layer, the identification of a primary component that could account for the low oil absorption value was not achievable. The relatively favorable values mentioned above demonstrate the potential of these coated papers to retain liquids. However, due to the lack of scientific literature providing $Cobb_{1800s}$ results in relation to shelf lifes of various food categories, it is not possible to discuss a one-on-one relation with shelf life.

Table 1. Average values and standard deviations of surface and barrier characteristics (water contact angle (WCA), oil contact angle (OCA), Bendtsen rougness, surface roughness (Sa), Cobb water and oil absorptions, water vapor transmission rate (WVTR)).

Name - grammage (g/m²) – description (ATR-FTIR identified main component)	***WCA (°)***	***OCA (°)***	***Bendtsen roughness (mL/min)***	***Sa (µm)***	***$Cobb_{1800s}$ water (g/m²)***	***$Cobb_{1800s}$ oil (g/m²)***	***WVTR (g/m²/d)***
Dispersion 1 - 67 – ethylene, metacrylic acid, acrylate copolymer	*97.0 ± 3.7*	*20.2 ± 1.2*	*31.3 ± 0.1*	*0.60 ± 0.07*	*1.8 ± 0.2*	*3.0 ± 0.4*	*0.2*
Dispersion 2 – 45 – acrylic, polyethylene vinyl acetate copolymer	*15.0**	*34.4 ± 2.5*	*430.9 ± 1.9*	*4.13 ± 0.08*	*59.6 ± 1.2*	*25.0 ± 1.5*	*1000*
Dispersion 3 – 103 – cellulose nanocrystals	*53.1 ± 4.3*	*44.8 ± 6.3*	*900.4 ± 0.5*	*4.33 ± 0.04*	*95.1 ± 4.7*	*46.0 ± 1.1*	*1000*
Dispersion 4 – 71 – proprietary vegetable wax	*119.2 ± 3.1*	*56.6 ± 4.8*	*399.4 ± 0.1*	*3.29 ± 0.23*	*39.8 ± 20.6*	*0.8 ± 0.4*	*794.1*
Dispersion 5 – 45 – polyvinylalcohol	*104.0 ± 1.0*	*51.8 ± 1.5*	*26.4 ± 0.0*	*1.41 ± 0.17*	*22.9 ± 0.2*	*2.7 ± 0.1*	*85.1*
Dispersion 6 – 80 – proprietary polymeric component	*42.2 ± 2.5*	*54.8 ± 6.1*	*104.7 ± 0.0*	*1.52 ± 0.08*	*24.4 ± 0.2*	*2.9 ± 0.6*	*141.1*
Dispersion 7 – 69 – acrylic copolymer	*103.6 ± 1.0*	*50.1*	*129.6 ± 0.0*	*2.72*	*16.9 ± 1.0*	*6.0 ± 1.0*	*105.7*

		± 0.8		± 0.42			
Dispersion 8 – 71 – vacuum metalized	*92.1 ± 2.0*	*47.4 ± 2.3*	*686.8 ± 0.0*	*4.03 ± 0.05*	*61.5 ± 3.3*	*3.0 ± 0.6*	*167.6*
Dispersion 9 – 55 – vacuum metallized	*97.9 ± 3.5*	*52.9 ± 1.2*	*160.2 ± 0.0*	*2.04 ± 0.17*	*40.0 ± 0.4*	*10.2 ± 0.4*	*64.0*
Dispersion 10 – 65 – polyolefin	*97.5 ± 2.5*	*54.3 ± 1.9*	*45.2 ± 0.0*	*1.10 ± 0.28*	*42.7 ± 0.4*	*2.7 ± 0.6*	*124.8*
Dispersion 11 – 41 – polyvinylalcohol	*46.5 ± 4.1*	*30.3 ± 6.1*	*466.1 ± 0.6*	*3.06 ± 0.89*	*32.9 ± 1.2*	*15.9 ± 0.9*	*1000*
Dispersion 12 – 102 – acrylic acid copolymer	*91.1 ± 2.4*	*19.1 ± 4.2*	*33.1 ± 0.0*	*1.17 ± 0.04*	*20.0 ± 4.1*	*3.1 ± 0.6*	*146.4*
Extrusion 1 – 98 – polyethylene	*97.7 ± 2.2*	*37.2 ± 3.8*	*76.7 ± 0.0*	*3.40 ± 0.76*	*0.6 ± 0.2*	*3.4 ± 0.6*	*4.2*
Extrusion 2 – 100 – polyolefin	*93.2 ± 3.2*	*35.6 ± 8.2*	*441.8 ± 0.0*	*4.27 ± 0.21*	*3.2 ± 0.3*	*3.6 ± 0.5*	*23.8*
Wax – 44 - – ethylene copolymer and wax	*101.4 ± 4.5*	*65.2 ± 2.5*	*485.2 ± 0.7*	*2.62 ± 0.09*	*29.3 ± 1.5*	*9.2 ± 1.5*	*1000*

** Complete absorption of droplet upon contact*

An overview of the relation between contact angles of oil and water for the different coating grades is shown in Figure 1. It indicates that the present selection of industrial coatings includes grades that either can provide sufficient oleophobicity, hydrophobicity and/or a combination of both depending on the coating type.

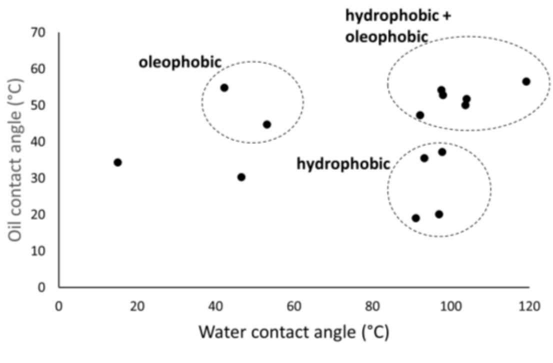

Figure 1. Relation between water contact angle and oil contact angle for different coating grades, prodiving a selection tool for papers with oleophobic and/or hydrophobic barrier properties.

Due to the uncertainty regarding the existence of linear relationships between contact angles, roughness and barrier properties, an investigation into monotonic correlations is conducted by calculating Spearman coefficients and p-values of the variables. The findings are presented in Table 2. The strongest significant relationship, within a 95% confidence interval, is observed between Bendtsen roughness and surface roughness. Additionally, there are significant correlations, highlighted in Table 2, between oil and water contact angles, WVTR and Bendtsen roughness, Cobb water absorption and Bendtsen roughness, Cobb water absorption and WVTR, Cobb oil absorption and Bendtsen roughness, and Cobb oil absorption and surface roughness. There are no significant correlations with grammage (not shown in table).

Although relationships between contact angles and barrier properties have been discussed in the literature (Rastogi et al., 2015) (Tambe et al., 2016), no correlation was found between the different sets of results in this study. Rastogi et al. mentions that hydrophobicity is a primary requirement for the creation of a barrier layer on papers. Tambe et al. discusses that it is not surprising that Cobb water absorptions and water contact angles are inversely correlated. Possibly, the absence of correlation between contact angles and barrier properties can be explained by a lack of quality of some of the coatings.

However, several correlations were found between roughness of coated surface and barrier properties of coated paper, a relationship that has been previously mentioned in the literature (Thitsartarn et al. 2020) (Lee et al. 2021). Thitsartarn et al. refers to authors who report that increasing surface roughness generally enhances hydrophobicity. Other authors are cited who state that for hydrophilic surfaces, greater surface roughness leads to increased wettability. Hydrophobicity and particle size are utilized in the discussion to relate low Cobb absorptions, albeit conducted at much lower exposure times, to the effective filling of the porous structure of paper-based materials to prevent water absorption. Lee et al. discussed the WVTR values by positively relating them to surface roughness.

Table 2. Spearman coefficients and p-values of variables.

Variable	***By Variable***	***Spearman ρ***	***p-value***
Surface roughness Sa	*Bendtsen roughness*	*0.779*	***0.001***
Water contact angle	*Bendtsen roughness*	*-0.286*	*0.302*
Water contact angle	*Surface roughness Sa*	*-0.246*	*0.376*
Oil contact angle	*Bendtsen roughness*	*0.118*	*0.676*
Oil contact angle	*Surface roughness Sa*	*-0.096*	*0.733*
Oil contact angle	*Water contact angle*	*0.525*	***0.045***
WVTR	*Bendtsen roughness*	*0.652*	***0.008***
WVTR	*Surface roughness Sa*	*0.368*	*0.178*

WVTR	*Water contact angle*	*-0.360*	*0.187*
WVTR	*Oil contact angle*	*0.142*	*0.613*
Cobb water absorption	*Bendtsen roughness*	*0.579*	***0.024***
Cobb water absorption	*Surface roughness Sa*	*0.321*	*0.243*
Cobb water absorption	*Water contact angle*	*-0.282*	*0.308*
Cobb water absorption	*Oil contact angle*	*0.279*	*0.315*
Cobb water absorption	*WVTR*	*0.697*	***0.004***
Cobb oil absorption	*Bendtsen roughness*	*0.600*	***0.018***
Cobb oil absorption	*Surface roughness Sa*	*0.539*	***0.038***
Cobb oil absorption	*Water contact angle*	*-0.414*	*0.125*
Cobb oil absorption	*Oil contact angle*	*-0.339*	*0.216*
Cobb oil absorption	*WVTR*	*0.384*	*0.158*
Cobb oil absorption	*Cobb water absorption*	*0.243*	*0.383*

To visualize the correlations between the roughness (Sa, Bendtsen) and barrier (Cobb water + oil, WVTR) variables, Figure 2 displays three XY-scatterplots. In all three graphs, the relationship appears to be asymptotic, with high absorption or permeability levels at high roughness values. It is also interesting to focus on the outliers, characterized by high roughness and low absorption and permeability. Two such outliers that appear in all three graphs are the two extrusion coated papers, exhibiting high surface roughness values of 3.4 and 4.27 µm, as well as low liquid absorptions and WVTR. Presumably, the melt-processing during extrusion helps and limits defects through which liquids and/or gases can penetrate. In addition to surface roughness, material properties of the coating are also important. The FTIR results reveal that these two outliers are coated with polyethylene, a polymer widely recognized for its excellent moisture barrier properties.

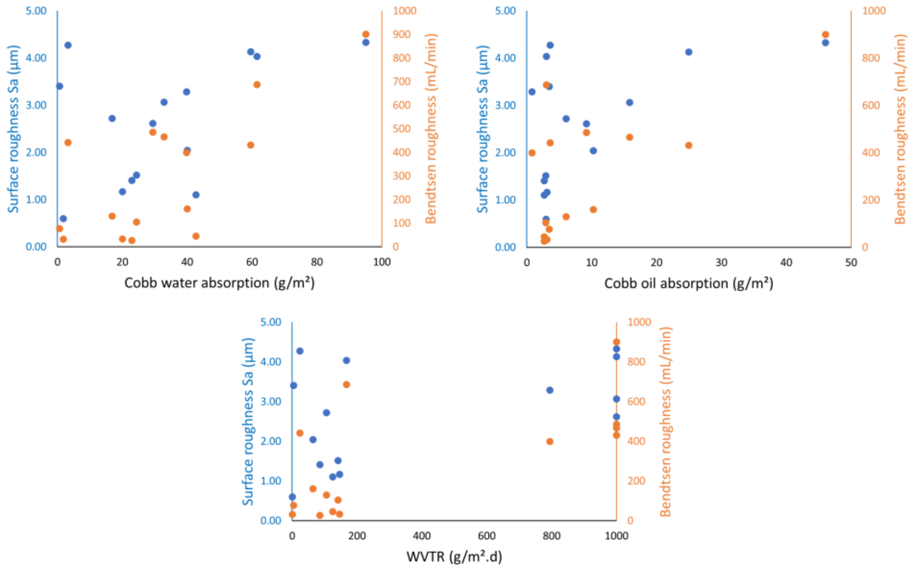

Figure 2. XY-scatterplots of roughness (Sa, Bendtsen) and barrier (Cobb water + oil absorptions, WVTR) variables.

4 CONCLUSION

This study emphasizes the importance of a smooth surface coating to achieve good barrier performance. However, this is only one of the factors influencing barrier properties, as evidenced by the deviating results of the extrusion-coated papers in the study, which achieved good barrier properties despite having high roughness, and by material properties, such as the excellent moisture barrier of polyethylene. Further research would be valuable to explore other aspects in detail, such as more details on the chemical composition of the coating as well as the nature and porosity of the underlying paper. In conclusion, this study provides an overview of the variations in surface characteristics and barrier properties among coated papers available in the market for primary food applications.

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