VTT Technical Research Centre of Finland

Pilot-scale demonstration of novel tandem coating process

Toriseva, Juuso; Lahti, Johanna; Kuusipalo, Jurkka

Published in:
Journal of Applied Polymer Science

DOI:
10.1002/app.53959

Published: 20/06/2023

Document Version
Publisher's final version

Received: 4 January 2023 | Revised: 17 March 2023 | Accepted: 20 March 2023
DOI: 10.1002/app.53959

RESEARCH ARTICLE

Journal of Applied Polymer Science WILEY

Pilot-scale demonstration of novel tandem coating process: Combining dispersion and extrusion coating with enhanced barrier properties

Juuso Toriseva[1,2] | Johanna Lahti[1] | Jurkka Kuusipalo[2]

[1]Biopolymer and Composite Solutions, VTT Technical Research Centre of Finland Ltd, Tampere, Finland
[2]Paper Converting and Packaging Technology, Tampere University, Tampere, Finland

Correspondence
Juuso Toriseva, Biopolymer and Composite Solutions, VTT Technical Research Centre of Finland Ltd, Visiokatu 4, Tampere 33720, Finland.
Email: juuso.toriseva@vtt.fi

Funding information
Tampere University

Abstract

Increasing environmental and legislative demands have accelerated the development of new packaging materials and processes toward more sustainable alternatives. Furthermore, barrier properties of the packaging have become increasingly important, as food waste creates environmental problems. Nowadays, multilayer structures are typically used to create required barrier properties to the packages. The aim of the study was to combine dispersion and extrusion coating into consecutive tandem process to produce two-layer structure with enhanced barrier properties. By combining two processing steps into one, time and resources are saved. The study presents demonstration of the tandem coating in pilot-scale. The process was tested with several different materials and combinations and the processing conditions were optimized to produce optimal layer structure and properties. The properties of samples were analyzed with microscopy imaging, pinhole measurement and barrier measurements including oxygen, water vapor and grease permeation. Single layer of dispersion coatings could not produce continuous film, but the extrusion coatings filled the pinholes and enabled low permeation values. The permeabilities of the tandem coated structures were promising and generally lower than the sum of two individual layers. Moreover, the tandem coated structures produced with sustainable alternatives, performed better than traditional materials in oxygen and grease permeation measurements.

KEYWORDS

barrier properties, dispersion coating, extrusion coating, packaging materials, tandem coating

1 | INTRODUCTION

Global phenomena, such as climate change and resource scarcity, and consumer awareness have generated a strong demand for development of novel packaging materials. Increasing greenhouse gas emissions and pollution of oceans have created concerns and more stringent legislation, which are impacting to the use of plastics in packaging applications. Endeavors toward more sustainable solutions are realized by developing both materials and processes.[1–3]

Extrusion coating was developed to combine a molten thermoplastic polymer to a moving flexible substrate.[4] The main sections of extrusion coating equipment are hopper, extruder, adapter, die, laminator and auxiliary equipment.[5,6] The final product is normally paper, or paperboard coated with layer of plastic film, but other materials such as aluminum foil can be extrusion coated as well.[4–6]

Extrusion coating combines advantages and properties of both the polymer and the substrate. In packaging applications, the paper or paperboard substrate forms a rigid structure that is lightweight, low-cost and sustainable. The polymer coating in turn, enables closing of the package by heat sealing and provides liquid, gas and grease barrier, which prevent permeation and migration in and outward, and protect the product from external factors that can cause deterioration.[7–9]

The coating material can be practically any kind of thermoplastic polymer that can be processed into molten film in the extruder and drawn into thin layer on top of a substrate. Hence, extensive range of structures and products are developed and produced to serve the demands of numerous applications.[4,5] The most common coating materials are still fossil-based polyolefins for example, polyethylene (PE), polypropylene (PP) and their copolymers as well as polyethylene terephthalate (PET) and polyamide (PA).[5,10,11] However, research and development of bio-based and biodegradable polymers is advanced distinctly in the past decade and the applications are constantly increasing due to legislative and environmental reasons.[10–13]

Barrier performance is one of the main properties and targets of the extrusion or dispersion coated materials. Pinhole free coatings, which produce water vapor, oxygen and grease barrier are required for various food packaging products. The barrier performance of polymer varies depending on several factors such as polarity and crystallinity along with functional groups.[7,8] Biopolymers have promising potential to replace current fossil-based coating materials and structures. However, many of bio-based materials are hydrophilic and hence the barrier properties are depended on the ambient humidity.[2,3,10] Oxygen and water vapor permeation properties of commonly used polymers are presented in Table 1.

Dispersion coating refers to a coating process, where aqueous polymer suspension is applied onto the surface of paper or paperboard to form continuous film after drying.[5,15] The goal is to achieve a continuous barrier layer and hence coatings are sometimes referred as barrier dispersions or coatings.[15,16] The use and interest toward polymer dispersion coatings is constantly increasing, which is resulting from environmental awareness and legislation.[17,18] Dispersion coatings are often considered as an environmentally friendly alternative for packaging, since generally the products can be fully recycled.[5,15,18]

TABLE 1 Oxygen and water vapor transmission rates for commonly used polymers.[5,6,14]

Polymer	OTR 23°C/0% RH [$cm^3 \times 25\ \mu m/m^2 \times d$]	WVTR 38°C/90% RH [$g \times 25\ \mu m/m^2 \times d$]
Low density polyethylene, LDPE	4400–8000	12–24
High density polyethylene, HDPE	1600–2800	4–12
Cyclic olefin copolymer, COC	720–1000	0.8–2
Polypropylene, PP	2400–4200	7.2–12
Polyethylene terephthalate, PET	48–92	20–50
Polyamide 6, PA6	8–60	155–324
Polystyrene, PS	5425	109–155
Polyvinylidene chloride, PVDC	0.16–14	0.3–3.4
Ethylene vinyl alcohol, EVOH	0.12–1.2	14.4–84
Polylactic acid, PLA	465–2400	120–620
Poly(3-hydroxybutyrate-co-3-hydroxyvalerate), PHBV	600–1400	60–100
Polycaprolactone, PCL	400–2400	320
Polybutylene adipate terephthalate, PBAT, Ecoflex®	3200	340
Ethylene vinyl acetate/Ethylene methyl acrylate, EVA/EMA	9300–15,500	15.5–139
Ethylene acrylic acid/Ethylene methacrylic acid, EAA/EMAA	3100–7750	15.5–25

Barrier dispersion coatings used in packaging consists of polymer particles and additives in water. Typical polymers used in dispersion coatings include styrene-butadiene, acrylates, metacrylates, vinyl acetates and polyolefins.[5,15] Furthermore, the use of bio-based polymers is growing similarly as in extrusion coating applications.[15,17] Dispersion coatings can be applied with different processes and coaters. The coating methods are usually named after the metering devices such as blade, rod, air knife, gravure and curtain coaters.[5,17] Regardless of the processing method, the outcome should be uniform and continuous polymer film onto the substrate.[15]

The most notable applications of dispersion coated paper and boards are food and liquid cartons.[15,16] Furthermore, dispersion coatings can be used as precoating in multilayer structures with extrusion coating. Both coatings can concentrate on same characteristic such as specific barrier, or provide individual properties, which are combined in the final product.[5,15]

The idea of tandem coating is to improve efficiency and sustainability by developing a process that contains less steps and increase the production. Energy consumption and costs can be decreased, since unnecessary logistics, temporary storages and real estate costs are reduced. This is industrially extremely relevant since the packaging industry is constantly driving toward more economical and sustainable solutions. Moreover, the shift toward future requirements can be advanced further by using more sustainable material choices such as bio-based and biodegradable coating materials.

Multilayer structures of bio-based materials for barrier applications have been researched for example by Poulose et al.,[2] Koppolu et al.,[19] Vähä-Nissi et al.[20] and Vartiainen et al.[21] However, these are produced with multiple and low speed processing steps to achieve the properties. Compared to the studies above, the tandem coating process significantly reduces processing time and resources, which makes it industrially more feasible. Currently, corresponding studies in the similar scale as presented tandem coating process are not available.

2 | MATERIALS AND METHODS

The trial runs were performed at Tampere University (TAU), Paper Converting and Packaging Technology pilot line. The schematic picture of the pilot line is presented in Figure 1. The coating steps and progress of the substrate are emphasized with yellow and red respectively. The substrate runs through dispersion and extrusion coating steps and the drying units are placed in between.

Bleached virgin-fiber board, Cupforma Natura 195 g/m^2 (Stora Enso, Imatra, Finland) was used as the paperboard substrate. Three barrier dispersion products (Epotal®, BASF, Ludwigshafen am Rhein, Germany) were utilized in dispersion coating. Extrusion coating polymers used were low density polyethylene (LDPE) CA7230 (Borealis, Wien, Austria) and ecovio® PS 1606 (BASF). The coating materials and their main properties are presented in Table 2. CA7230, Epotal® S440eu and Epotal® SP106D can be considered as traditional fossil-based polymer materials. On the other hand, ecovio® PS1606 and Epotal® 8422X offer more sustainable alternatives. PS1606 is bio-based, biodegradable and compostable material according to the standards EN 13432 and ASTM 6400. 8422X has reduced carbon footprint and it includes bio-based material, but it is not biodegradable.

The samples of the study were produced in TAU pilot line, illustrated in Figure 1, which includes both dispersion and extrusion coating stations. Three air dryer units and five infrared (IR) drying units are placed right after the dispersion application station to evaporate water and to form continuous polymer layer. Glossy chill roll was used in the extrusion coating laminator unit. The course of the substrate was varied with assistance rollers compared to the normal one step processes to enable the tandem coating. The process parameters of dispersion and extrusion coating are presented in Tables 3 and 4 respectively. The process parameter values are based on the recommendations of material manufacturers. Furthermore,

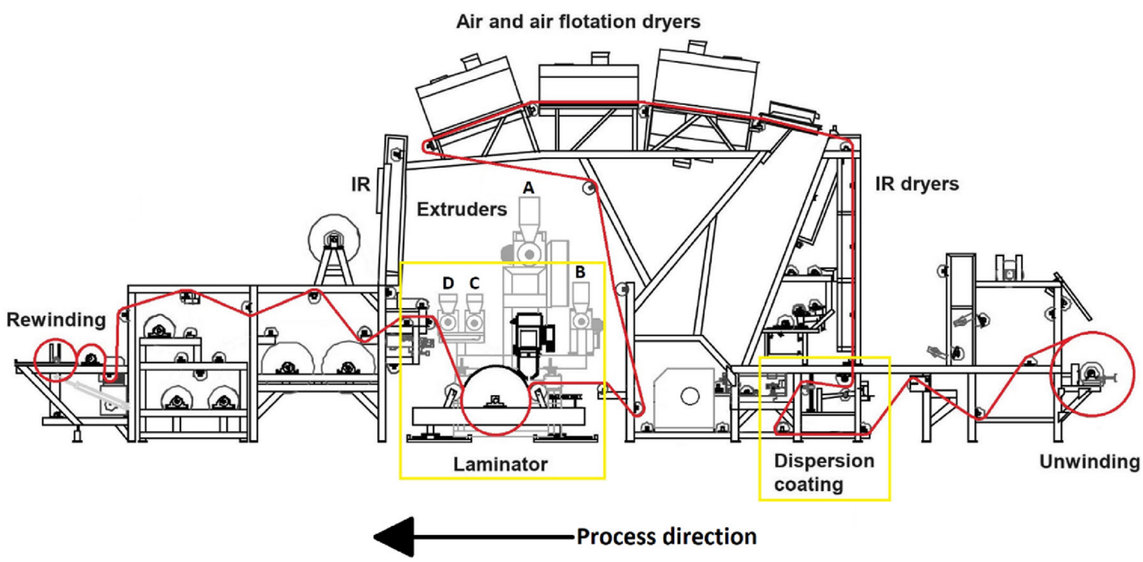

FIGURE 1 Schematic picture of tandem coating at TAU pilot line. [Color figure can be viewed at wileyonlinelibrary.com]

TABLE 2 Coating materials and their main properties.[22–26]

Material	Producer	Polymer	Density [g/cm^3]	Melt flow rate [g/10 min, 190°C/2.16 kg]	Viscosity [mPas]	Solids content [%]
Extrusion coating						
CA7230	Borealis	Low density polyethylene	0.923	4.5		
ecovio® PS1606	BASF	ecoflex® (Polybutylene adipate terephthalate) + Polylactic acid	1.24–1.26	18–24		
Dispersion coating						
Epotal® S440eu	BASF	Styrene-acrylic copolymer	1.04		600–1500	46.5–48.0
Epotal® SP106D	BASF	Styrene-butadiene copolymer	1.04		30–300	49–51
Epotal® 8422X	BASF	Styrene-butadiene copolymer (Reduced carbon footprint)	1.04		225	50

TABLE 3 Process parameters of dispersion coating.

Dispersion coating parameter		Epotal® S440eu	Epotal® SP106D	Epotal® 8422X
Application method		Rod	Rod	Rod
IR dryers [kW]	1	6	6	6
	2	4.2	4.2	4.2
	3	8.4	8.4	8.4
	4	4.2	4.2	4.2
Temperature after IR dryers [°C]		80	84	76
Air dryers [°C]	1	165	165	160
	2	160	160	160
	3	160	160	160
Temperature after air dryers [°C]		100	102	98
Chill roll temperature [°C]		13	13	15
Line speed [m/min]		40	40	40

observations and adjustments were made based on the characteristics of pilot line before producing the samples to ensure the optimal processing conditions and consequently high-quality coatings.

The coating materials, presented in Table 2, were used to produce samples with varying material combinations and thicknesses. Altogether 13 samples were produced and analyzed in this study to compare the properties of each structure. The samples and their layer structures are presented in Table 5. Three samples were produced with dispersion coating and containing a single polymer layer. Furthermore, four test points are single polymer layer structures made with extrusion coating of CA7230 or ecovio® PS1606. The rest of the samples (altogether eight test points) are two-layer structures, wherein tandem coating process is utilized to combine the dispersion and extrusion coating steps and to achieve properties from both layers. Illustration of the samples is shown in Figure 2, which presents the visual appearance of the sample after dispersion coating and subsequent extrusion coating processes.

After the trial runs the coated samples were analyzed with the following methods. Coating weight average and standard deviation of the grammage were measured right after the processing. The weight of a specific surface area (100 cm^2) was measured with analytical scale to determine coating weight. Five parallel samples were measured to reduce instability and calculate the standard deviation. Samples with only dispersion coating layer were dried before weighing to remove residual moisture, which might cause excessive error with low coating weights.

The number of pinholes was measured using three test solutions. Colored turpentine, ethanol/water and water/detergent mixtures were used to study pinhole occurrence. Approximately 0.1 m^2 area of coated sample surface was applied with thin layer of the test solution. The samples were examined from the backside to see if any solution penetrated through the coating and wetted the paperboard.

The barrier properties of the samples were evaluated by measuring the permeability through the structures with oxygen gas (O_2), water vapor (H_2O) and olive oil. Oxygen transmission rate (OTR) measurements were performed with Mocon OxTran 2/21 MH (Ametek Mocon Inc., Minneapolis) according to the standard ASTM D3985. The measurement conditions were 23°C temperature and 50% relative humidity (RH). The oxygen content was 10% and nitrogen (N_2) functioned as a carrier gas. The sample area was 50 cm^2 and the measurements were executed simultaneously with two parallel samples in A and B chambers. The average permeability of A and B chambers was employed as the OTR value of each sample. The measurement time was around 7 h for all the samples and the state of equilibrium was reached between 2 and 3 h. The measurement graph (Sample SP106D + CA7230) is presented in Figure 3 as an example.

TABLE 4 Process parameters of extrusion coating (screw speeds corresponding melt temperature and pressure values are presented in sequence).

Extrusion coating parameter	CA7230	ecovio® PS1606
Extruder diameter [mm]	40	40
Extruder L/D ratio	24	24
Screw speed [rpm]	100/140	120/140
Melt temperature [°C]	314/310	235/235
Melt pressure [bar]	77/100	102/114
Chill roll temperature [°C]	13	15
Nip pressure [bar]	5.2	5.5
Air gap [mm]	280	160
Die gap [mm]	0.63	0.63
Line speed [m/min]	40	40

TABLE 5 Samples and their layer structures.

Structure	Substrate	Dispersion coating polymer	Extrusion coating polymer	Sample name
Substrate/Dispersion	Cupforma Natura	Epotal® S440eu	—	Epotal® S440eu
Substrate/Dispersion	Cupforma Natura	Epotal® SP106D	—	Epotal® SP106D
Substrate/Extrusion	Cupforma Natura	—	CA7230	CA7230 (100 rpm)
Substrate/Extrusion	Cupforma Natura	—	CA7230	CA7230 (140 rpm)
Substrate/Dispersion/Extrusion	Cupforma Natura	Epotal® S440eu	CA7230	S440eu + CA7230 (100 rpm)
Substrate/Dispersion/Extrusion	Cupforma Natura	Epotal® S440eu	CA7230	S440eu + CA7230 (140 rpm)
Substrate/Dispersion/Extrusion	Cupforma Natura	Epotal® SP106D	CA7230	SP106D + CA7230 (100 rpm)
Substrate/Dispersion/Extrusion	Cupforma Natura	Epotal® SP106D	CA7230	SP106D + CA7230 (140 rpm)
Substrate/Dispersion	Cupforma Natura	Epotal® 8422X	—	Epotal® 8422X
Substrate/Extrusion	Cupforma Natura	—	ecovio® PS1606	ecovio® PS1606 (120 rpm)
Substrate/Extrusion	Cupforma Natura	—	ecovio® PS1606	ecovio® PS1606 (140 rpm)
Substrate/Dispersion/Extrusion	Cupforma Natura	Epotal® 8422X	ecovio® PS1606	8422X + PS1606 (120 rpm)
Substrate/Dispersion/Extrusion	Cupforma Natura	Epotal® 8422X	ecovio® PS1606	8422X + PS1606 (140 rpm)

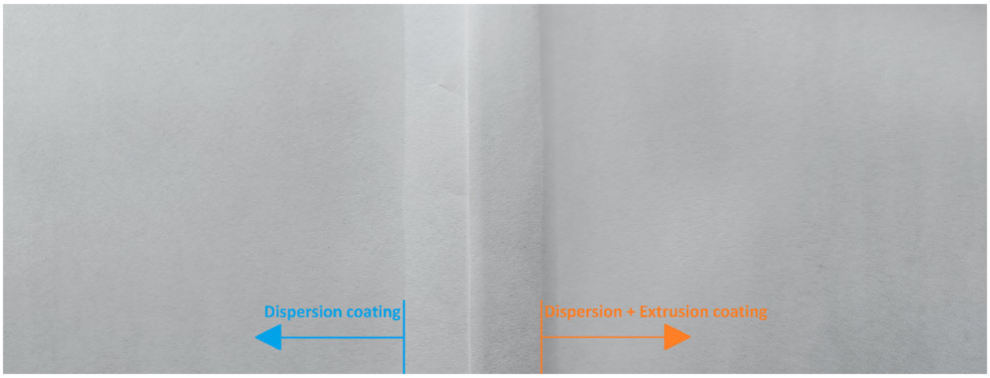

FIGURE 2 Samples after dispersion and extrusion coating process steps, uncoated paperboard is seen between the coatings. [Color figure can be viewed at wileyonlinelibrary.com]

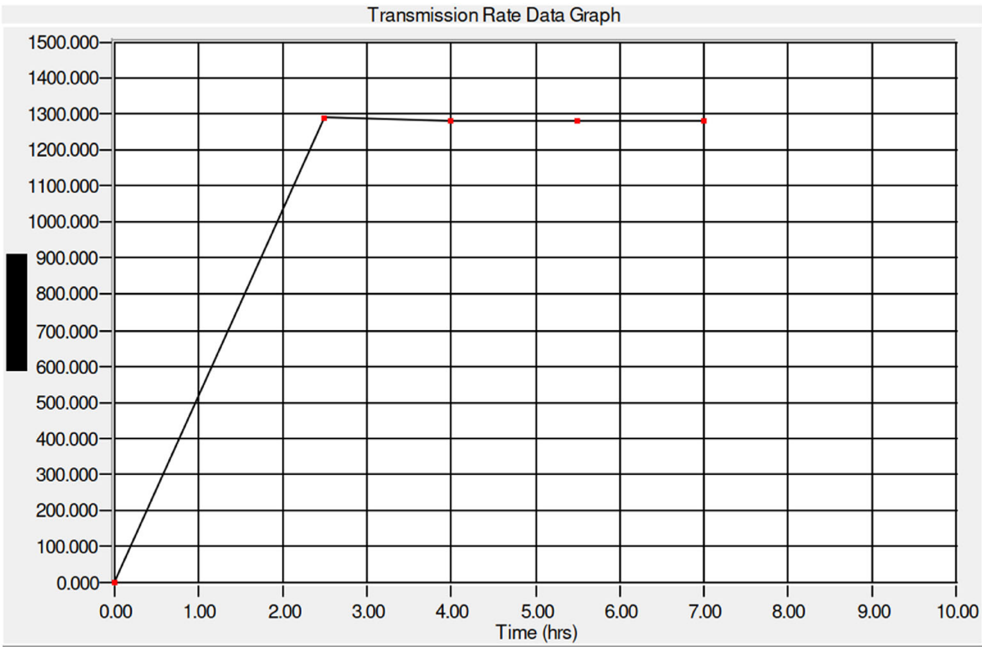

FIGURE 3 Oxygen transmission rate graph. [Color figure can be viewed at wileyonlinelibrary.com]

Water vapor transmission rate (WVTR) was studied with water and desiccant method according to the standard ASTM E96. The samples were sealed with wax on top of a cup including anhydrous calcium chloride granules (2–5 mm) as a desiccant. The sample cups were conditioned under 23°C and 50% RH environment in Espec PR-2J climate chamber. The sample cups were weighed at regular intervals to define the increase of the weight as a function of time, and hence the permeability of water vapor. Five parallel measurements were done at five points in time to determine the average WVTR value and standard deviation for each sample. The measurement time was altogether 2 days, and the change of mass was weighed approximately at 12 h intervals to ensure that the permeability has reached the equilibrium.

Rate of grease penetration was examined according to standard ASTM F119-82. The samples were set onto fluorescent thin layer chromatography (TLC) sheets. Six separate drops of olive oil were applied on each sample and the drops were pressed with weights. The samples were heated at 40°C in Firlabo AC120 (Froilabo, Lyon, France) oven and the grease penetration was observed as a function of time. The penetration was determined by examining color change of TLC sheets under the samples with ultraviolet (UV) light of at regular intervals. The grease penetration was determined at the points of the first and last (6th) drop penetrated through the sample structure.

Cross sectional images of the samples were taken with optical microscope, Carl Zeiss Light Microscopy model Axioskop 40 (Carl Zeiss AG, Oberkochen, Germany). The samples were prepared in cross direction (CD), before examination with the microscope. Axioskop 40 was also used to evaluate the size of pinholes. The dispersion coated samples were applied with colored turpentine solution and imaged from above. Scanning electron microscopy (SEM) was used to study and analyze the sample structures further. Analytical high resolution scanning electron microscope, Zeiss UltraPlus FE-SEM (Carl Zeiss, AG,

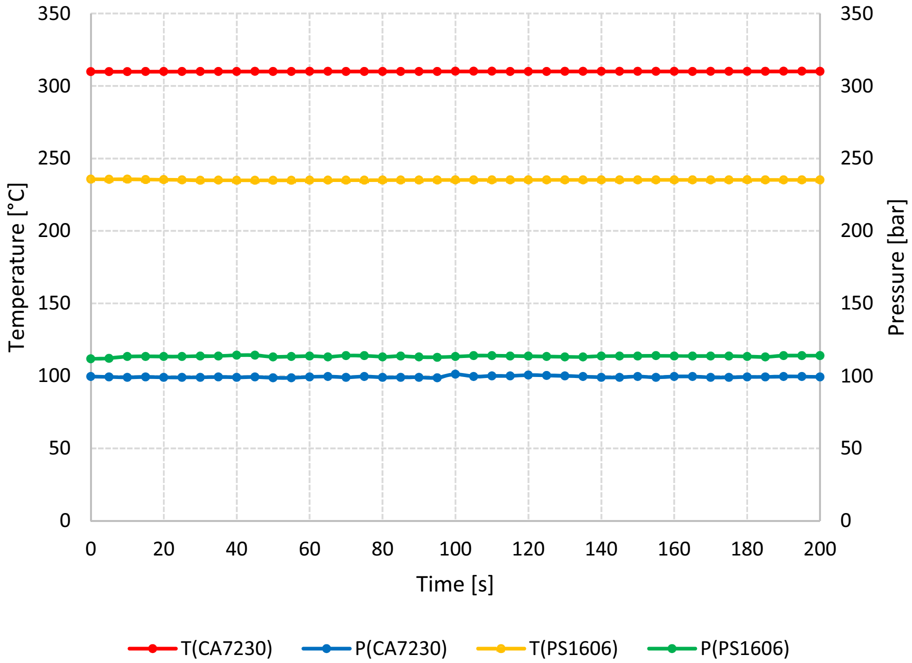

FIGURE 4 Mass temperature and pressure as a function of time during the trial runs. [Color figure can be viewed at wileyonlinelibrary.com]

Oberkochen, Germany), was used to take SEM pictures. The samples were prepared with cross section polisher (CSP) and sputtered with platinum/palladium (Pt/Pd) before scanning.

3 | RESULTS AND DISCUSSION

The coated paperboard samples were characterized for coating weight and its standard deviation, pinholes and barrier properties including WVTR, OTR and grease penetration. The melt temperature and pressure data of extrusion coating step were data logged during the trial runs to examine any fluctuations, which could lead to defects or discontinuities in the coating. Figure 4 presents the melt temperature and pressure for both extrusion coating materials with constant 140 rpm screw speed setting. Figure 4 shows that no significant variation in the temperature or pressure is registered. Hence, the extrusion coating step can be considered steady and properly adjusted. All the extrusion and tandem coated samples were produced within the 200 s interval, from which the extrusion temperature and pressure data is logged.

3.1 | Coating weight and pinholes

Coating weight and standard deviation were measured during the trial run to verify that the targeted layer structures were achieved. Furthermore, standard deviation of coating weight can be related to the runnability of extrusion coating layer. Lower standard deviation of coating weight signifies more uniform coating process and hence thickness profile. All dispersion coating layers are roughly on the same coating weight scale. Extrusion coating layers, both LDPE (CA7230) and ecovio® (PS1606), were produced with two different screw speeds to get more versatile samples. Moreover, coating weights of the polymers are intentionally different, LDPE being lower than ecovio®, which is based on the processability, properties and previous experience with the polymers. Coating weight and standard deviation are presented in Table 6.

Pinhole measurements were performed for all sample structures. The results are presented also in Table 6. The results show that pinholes appear neither in extrusion coating nor in tandem coating samples. Furthermore, water-detergent solution does not penetrate the structures, which can be explicated by too low detergent content and thus excessive contact angle and poor wetting. Dispersion coating samples (Epotal® S440eu, SP106D and 8422X) include various amounts of pinholes, when using turpentine or ethanol-water solutions. In S106D and 8422X samples, pinholes are divided rather evenly across the sample width, whereas S440eu samples show pinholes mainly at the edges of sample area. However, this does not have significant effect on the barrier measurements, since none of the samples are entirely pinhole free, which ruins the barrier performance regardless. The issue is solved with subsequent extrusion coating step, which blocks the discontinuities and enables the barrier performance of dispersion coating layer.

The main reason for high number of pinholes in dispersion coated samples is the single polymer layer.

TABLE 6 Coating weight and pinhole results of the samples.

Sample	Coating weight [g/m^2] Average	STDEV	Pinholes Turpentine	Ethanol + Water	Water + Detergent
Epotal® S440eu	6.9	0.6	Yes (<50)	Yes (<50)	No
Epotal® SP106D	9.0	0.4	Yes (>50)	Yes (>50)	No
CA7230 (100 rpm)	12.3	1.5	No	No	No
CA7230 (140 rpm)	16.6	0.4	No	No	No
S440eu + CA7230 (100 rpm)	22.3	1.6	No	No	No
S440eu + CA7230 (140 rpm)	30.6	1.3	No	No	No
SP106D + CA7230 (100 rpm)	24.9	1.9	No	No	No
SP106D + CA7230 (140 rpm)	28.1	1.3	No	No	No
Epotal® 8422X	7.2	0.9	Yes (<50)	Yes (<50)	No
ecovio® PS1606 (120 rpm)	25.2	2.2	No	No	No
ecovio® PS1606 (140 rpm)	29.7	2.0	No	No	No
8422X + PS1606 (120 rpm)	36.1	1.6	No	No	No
8422X + PS1606 (140 rpm)	39.7	2.6	No	No	No

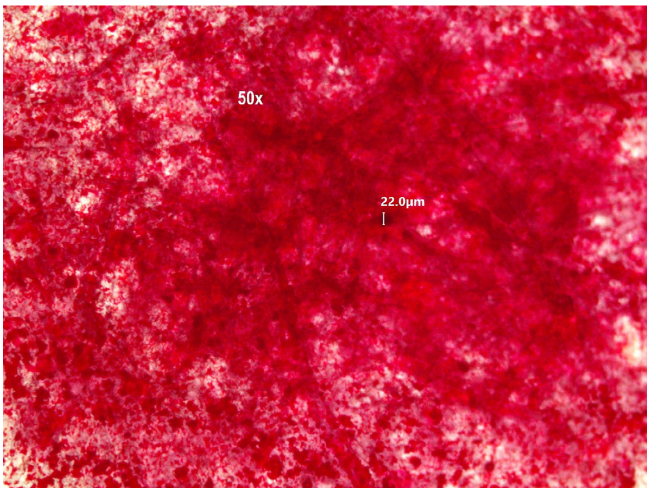

FIGURE 5 Optical microscopy image illustrating the pinhole size. [Color figure can be viewed at wileyonlinelibrary.com]

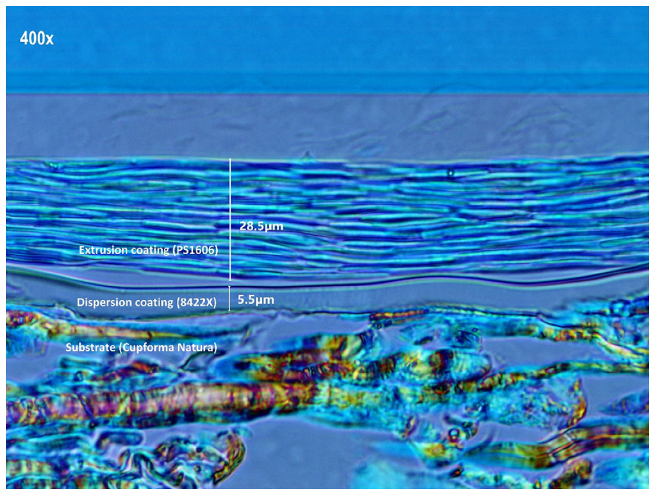

FIGURE 6 Cross sectional image of the tandem coated sample. [Color figure can be viewed at wileyonlinelibrary.com]

Hence, the coating weight is low and substrate roughness can penetrate the polymer layer and hinder the spreading, resulting to uneven and discontinuous coating. Furthermore, differences between viscosities and hence dissimilar application and spreading induce variation between the dispersion coated samples. 8422X and especially S440eu dispersions present slightly lower number of pinholes than SP106D, which indicates that higher viscosity values facilitate the application process at TAU pilot line. S440eu samples contain pinholes mostly at the edges, whereas SP106D and 8422X are covered more evenly.

The size of pinholes is examined with microscopy image, which is presented in Figure 5. The actual pinhole is detected as darker spot within the image area and the turpentine solution has spread radically within the fibrous structure underneath the coating. Average size pinhole was imaged from Epotal® 8422X coated sample. The pinhole diameter is 22 μm, which is shown in Figure 5. The imaging was focused to a single pinhole and thus larger and smaller sizes also appear. However, the hole size is more than enough to reduce oxygen and water vapor barrier performance, which are analyzed later.

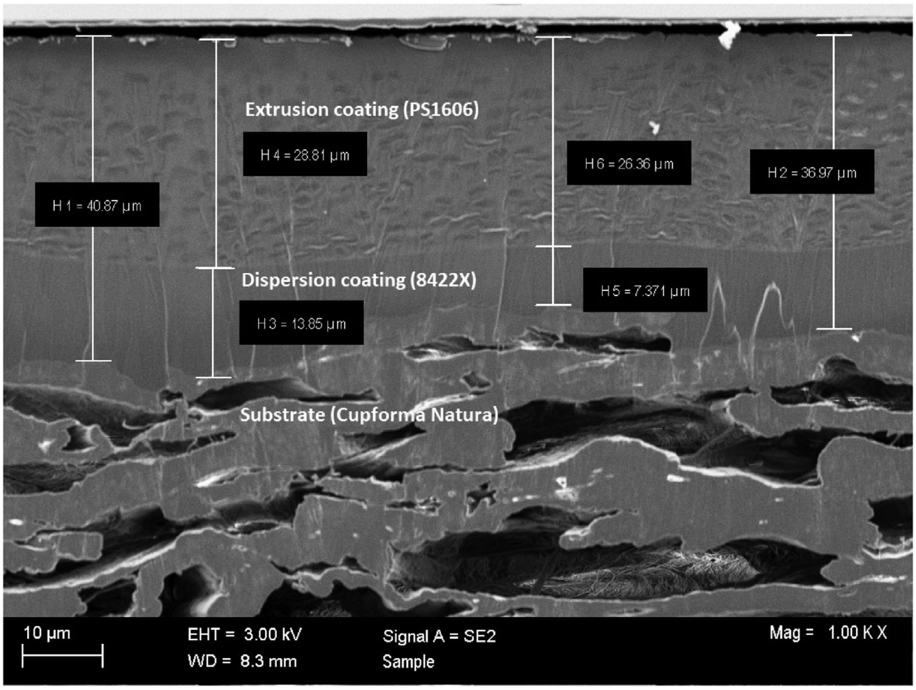

FIGURE 7 Scanning electron microscopy image of the tandem coated sample.

3.2 | Optical and scanning electron microscopy images

Cross sectional and SEM images of the tandem coating structures are presented in Figures 6 and 7 respectively. Both images were taken from the same sample, where Epotal® 8422X is the dispersion coating layer and ecovio® (140 rpm) is the extrusion coating layer. Hence, the images and consequently layer structure and thicknesses can be compared. Figures 6 and 7 show that the coating layers of both dispersion and extrusion polymer are continuous and uniform in the illustrated sample area. This indicates that both coating processes have been successful and stable. However, the examined areas are only a fraction of the whole coated surface area and thus hasty conclusions should not be made. This limitation can be confirmed by looking at the pinhole results shown in Table 6, which clearly express that all dispersion coating layers include plenty of pinholes and therefore the dispersion coating layers are not consistent but contain points of discontinuity.

The layer thicknesses of dispersion and extrusion coating layer correspond rather well to each other and furthermore to the coating weight results presented in Table 6. In the dispersion coating layer, most of the thickness values are nearly the same as coating weight, which correlates well with the density value of 1.04 g/cm^3 (Table 2). Theoretically, the extrusion coating layer should be slightly thinner as the coating weight is around 30 g/m^2 and the density is around 1.25 g/cm^3 (Table 2). The most significant result is the variation in dispersion coating layer thickness in Figure 7, which emphasizes the roughness of the substrate and uneven application of the polymer. This induces thicker sections as in Figure 7, but also thinner sections, which cause the formation of pinholes presented in Table 6.

3.3 | Oxygen transmission rate

Oxygen barrier performance was defined by measuring oxygen transmission rate (OTR) from all samples prepared. The permeation through the sample is dependent on the polymer layer or layers and the substrate material. However, the coating layers can be compared, since the substrate is constant, and its barrier effect is negligible. The barrier effect of the substrate is irrelevant, because OTR values are already over the measurement limit with dispersion coating layer, and hence the actual OTR of substrate value cannot be measured. OTR results are presented in Table 7 and illustrated in Figure 8.

The results can be analyzed from two different standpoints. One viewpoint is to compare the materials and especially traditional materials to the sustainable alternatives. The other perspective is to evaluate different processing techniques and particularly the benefit of the tandem coating process compared to the separate dispersion and extrusion coating steps.

The first and the most obvious result and observation is that only single layer of dispersion coating polymer is not enough to achieve measurable barrier and OTR value. Furthermore, by looking at the pinhole results in

TABLE 7 Oxygen transmission rate (OTR) results of the samples (23°C and 50% RH).

	Coating weight [g/m^2]		O_2TR [cm^3/(m^2 × d)]		23°C, 50% RH
Sample	**Average**	**STDEV**	***A***	***B***	**Average**
Epotal® S440eu	6.9	0.6	Over	Over	Over
Epotal® SP106D	9.0	0.4	Over	Over	Over
CA7230 (100 rpm)	12.3	1.5	9781	14,414	12,098
CA7230 (140 rpm)	16.6	0.4	4715	3439	4077
S440eu + CA7230 (100 rpm)	22.3	1.6	6803	6984	6893
S440eu + CA7230 (140 rpm)	30.6	1.3	4724	5254	4989
SP106D + CA7230 (100 rpm)	24.9	1.9	1228	1282	1255
SP106D + CA7230 (140 rpm)	28.1	1.3	1146	1150	1148
Epotal® 8422X	7.2	0.9	Over	Over	Over
ecovio® PS1606 (120 rpm)	25.2	2.2	973	1069	1021
ecovio® PS1606 (140 rpm)	29.7	2.0	741	701	721
8422X + PS1606 (120 rpm)	36.1	1.6	67	74	70
8422X + PS1606 (140 rpm)	39.7	2.6	53	55	54

Table 6, the conclusion appears indisputable. The outcome is expected, because of single and thin coating layer. Hence, the actual oxygen barrier performance of dispersion coatings cannot be confirmed, since the coating layer is not continuous and contains pinholes, which oxygen molecules can easily penetrate. This is earlier researched for example by Johansson et al.[19,20] However, the goal of this research is to prepare and compare results of single run processes and therefore only one layer of dispersion coating is applied.

Comparison of extrusion coated samples reveals that OTR of LDPE is much higher than OTR of ecovio®. However, ecovio® is not an explicit oxygen barrier material either, compared to ethylene vinyl alcohol (EVOH) presented in Table 1. Furthermore, it is notable that the coating weight of ecovio® is significantly higher than that of LDPE. OTR values of extrusion coated samples also show the distinct effect of screw speed and consequently coating weight. Both traditional and sustainable materials produce lower OTR value when the layer thickness is increased, which is evident considering the mechanism of permeation.

The final and most important aspect in OTR results is to compare the tandem coating to single step processes and to evaluate the possible benefits. Examination of the results in Table 7 and Figure 8 shows distinct and profitable effect of the two-step tandem coating. The majority of OTR values are substantially lower within the tandem coated samples. Furthermore, the same potential is seen in both traditional and sustainable materials. The only significant aberration is with tandem coatings of S440eu and CA7230, where the OTR is higher than the result of single layer of CA7230 at 140 rpm. It can be considered as a combination of poor oxygen barrier of LDPE and pinholes of S440eu. Furthermore, the level has reduced significantly compared to LDPE at 100 rpm, which indicates that the variation is rather high, when OTR values are elevated.

The best results are achieved by tandem coatings of Epotal® 8422X and ecovio® PS1606, which induce OTR values of 70 and 54 cm^3/(m^2 × d) with coating weights of 36.1 and 39.7 g/m^2 respectively. The improvement in oxygen barrier performance can be considered in consequence of extrusion coating polymer blocking the discontinuities of dispersion coating polymers.[27,28] Regardless, the tandem coating can be considered as more than the sum of dispersion and extrusion coating with respect to oxygen barrier performance.

3.4 | Water vapor transmission rate

Water vapor barrier performance was determined by measuring WVTR from all samples. Similarly as OTR, the coating layers are compared independently, since the substrate is constant and its barrier performance is considered insignificant. WVTR value of substrate was confirmed by a separate measurement according to the same standard (ASTM E96) at the same conditions (23°C and 50% RH). The average result is 636 g/(m^2 × d), which is substantially higher than permeabilities of coated samples and hence the effect of substrate is minor. WVTR results are presented in Table 8 and illustrated in Figure 9. The results can be analyzed again from two

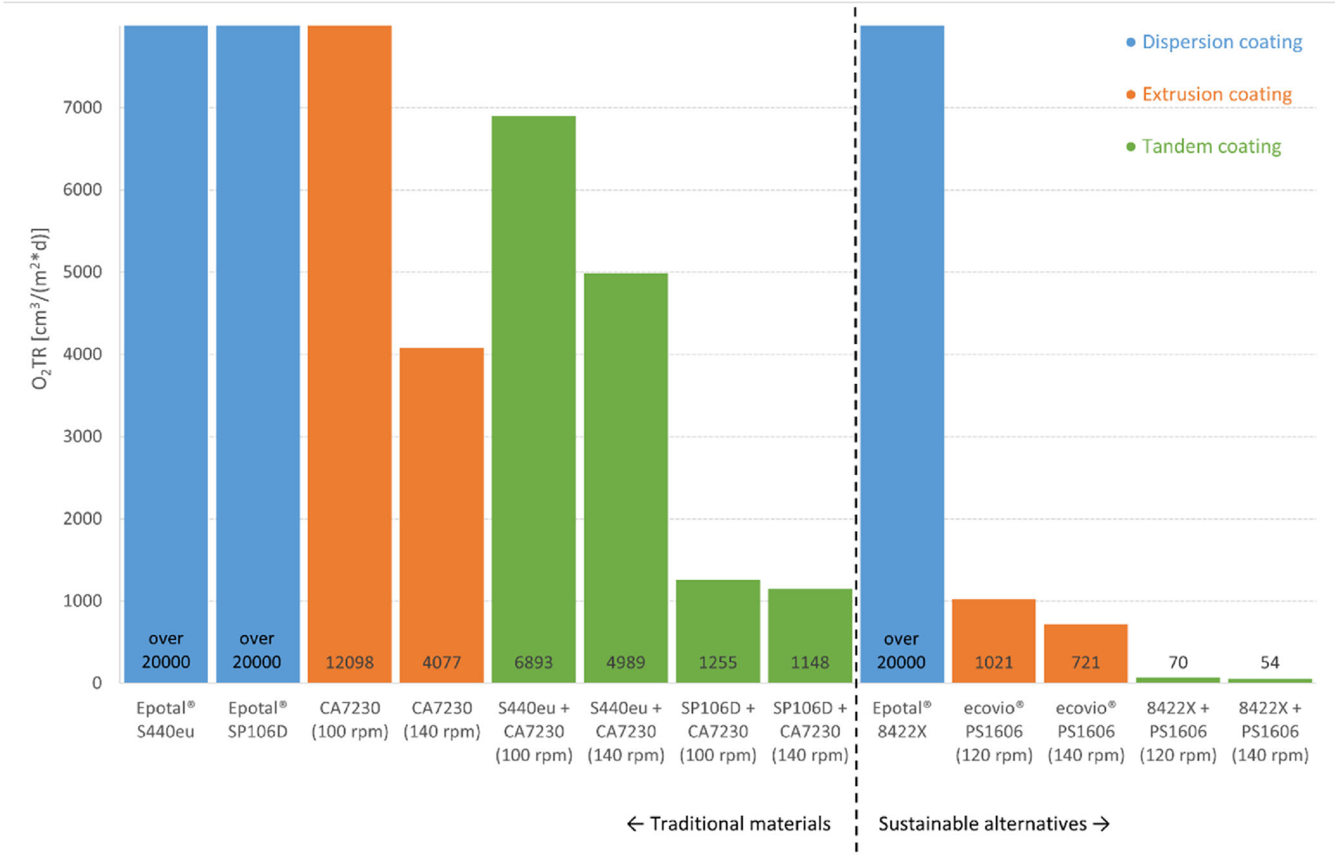

FIGURE 8 Oxygen transmission rate (OTR) results of the samples (23°C and 50% RH). [Color figure can be viewed at wileyonlinelibrary.com]

perspectives. One is to compare the materials and particularly traditional and sustainable alternatives, the other is to evaluate the processing steps and the advantage of tandem coating.

All studied materials provide sufficient water vapor barrier to achieve measurable WVTR value. Pinholes in the dispersion coating samples do not have as significant effect on WVTR as in the case of OTR measurement, where the values were too high to measure. The actual water vapor barrier performance of dispersion coatings should be better when continuous layer is achieved. Comparison of dispersion coated samples show that S440eu presents significantly worse WVTR value than two other samples. Furthermore, WVTR values of SP106D and 8422X are comparable and even better than some of pinhole-free extrusion coated samples.

WVTR values of extrusion coated samples present a clear difference between traditional and sustainable materials. Bio-based extrusion coating polymer ecovio® has poor water vapor barrier performance compared to the dispersion coatings. As expected, LDPE provides an excellent WVTR value, compared to the other studied coating materials as stated in Table 2.

Water vapor barrier performance of LDPE is much better compared to the other studied materials, which makes the evaluation of the effect of dispersion coating challenging in the tandem process. Small decrease of WVTR values is noticed, when comparing the extrusion and tandem coated samples of traditional materials. Comparison of sustainable materials and combinations expose more significant advantage of the tandem coating process. Combination of 8422X and ecovio® improve the water vapor barrier performance substantially, when comparing to a single step extrusion coating, and slightly compared to dispersion coating.

The best results are accomplished by tandem coatings of Epotal® SP106D and CA7230, which produce WVTR values of 2.6 and 3.3 g/($m^2 \times d$) with coating weights of 24.9 and 28.1 g/m^2 respectively. Compared to OTR results, the ability of extrusion coating to block pinholes does not have the same effect and importance. Hence, the best water vapor barriers are ensued mostly by the extrusion coating layer of LDPE, although dispersion coating layer (SP106D) induce rather good barrier performance as well. Regardless, it is reasonable to conclude that tandem coatings will improve the water vapor barrier or at least ensure and secure the existent level.

TABLE 8 Water vapor transmission rate (WVTR) results of the samples (23°C and 50% RH).

Sample	Coating weight [g/m²] Average	STDEV	WVTR [g/(m² × d)] Average	23°C, 50% RH STDEV
Epotal® S440eu	6.9	0.6	125	3.29
Epotal® SP106D	9.0	0.4	20.2	0.40
CA7230 (100 rpm)	12.3	1.5	3.8	0.36
CA7230 (140 rpm)	16.6	0.4	3.4	0.15
S440eu + CA7230 (100 rpm)	22.3	1.6	4.0	0.10
S440eu + CA7230 (140 rpm)	30.6	1.3	2.9	0.21
SP106D + CA7230 (100 rpm)	24.9	1.9	2.6	0.18
SP106D + CA7230 (140 rpm)	28.1	1.3	3.3	0.07
Epotal® 8422X	7.2	0.9	26	0.77
ecovio® PS1606 (120 rpm)	25.2	2.2	72	0.70
ecovio® PS1606 (140 rpm)	29.7	2.0	64	0.68
8422X + PS1606 (120 rpm)	36.1	1.6	18	0.63
8422X + PS1606 (140 rpm)	39.7	2.6	18	0.70

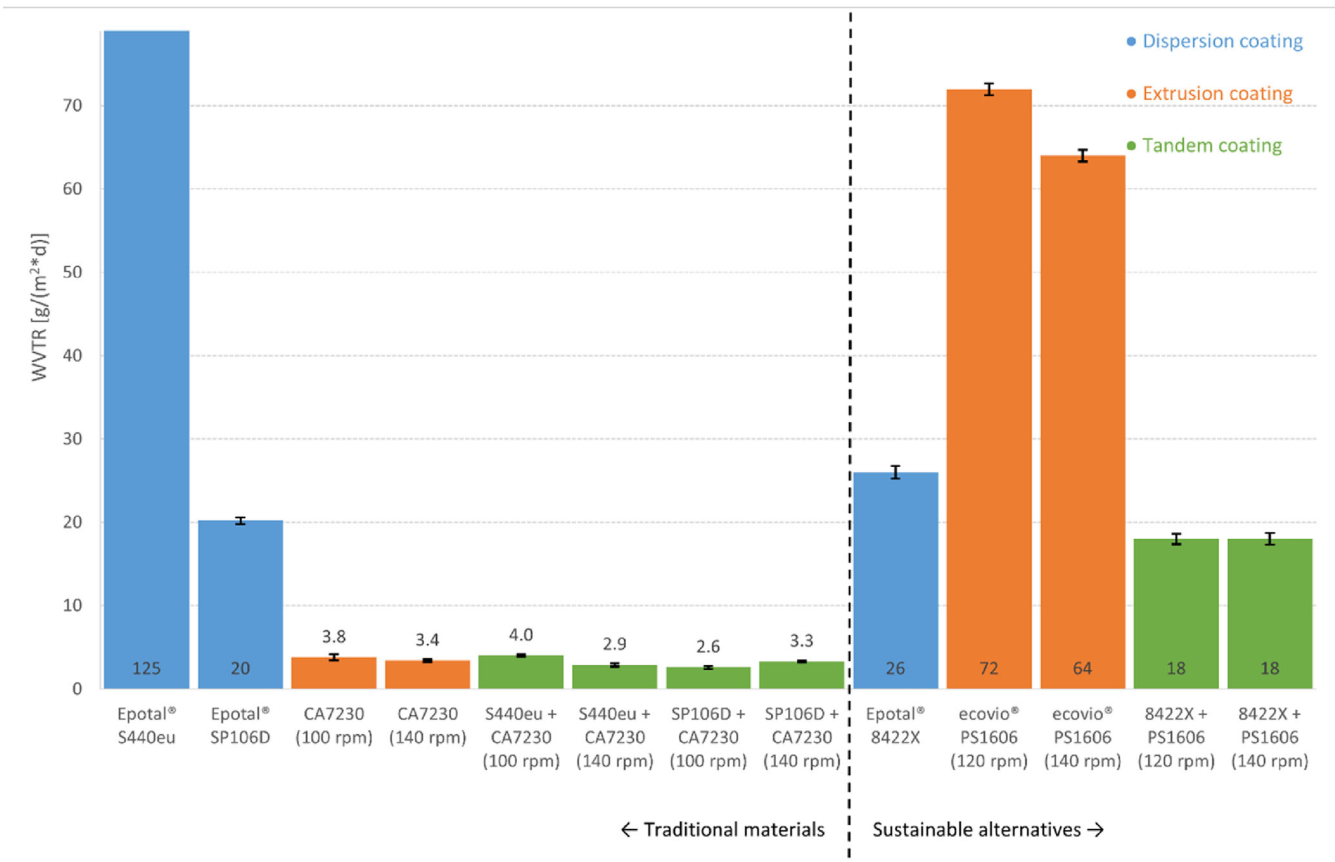

FIGURE 9 Water vapor transmission rate (WVTR) results of the samples (23°C and 50% RH). [Color figure can be viewed at wileyonlinelibrary.com]

3.5 | Rate of grease permeation

Grease barrier performance was examined by measuring rate of grease penetration using olive oil at 40°C temperature. Six parallel measurements were done for each sample and the time for the worst (1st) and best (6th) penetration time was defined. Hence, variation within the test points is observed. Rate of grease penetration is presented in Table 9.

TABLE 9 Rate of grease penetration of the samples (olive oil, 40°C).

Sample	Coating weight [g/m^2]		Rate of grease penetration [olive oil, 40°C]	
	Average	STDEV	Time for 1st penetration	Time for 6th penetration
Epotal® S440eu	6.9	0.6	24 h	30 h
Epotal® SP106D	9.0	0.4	0.5 h	1 h
CA7230 (100 rpm)	12.3	1.5	2 d	2 d
CA7230 (140 rpm)	16.6	0.4	3 d	3 d
S440eu + CA7230 (100 rpm)	22.3	1.6	2 d	2 d
S440eu + CA7230 (140 rpm)	30.6	1.3	2 d	2 d
SP106D + CA7230 (100 rpm)	24.9	1.9	3 d	4 d
SP106D + CA7230 (140 rpm)	28.1	1.3	3 d	4 d
Epotal® 8422X	7.2	0.9	1 h	3 d
ecovio® PS1606 (120 rpm)	25.2	2.2	13 d	16 d
ecovio® PS1606 (140 rpm)	29.7	2.0	13 d	16 d
8422X + PS1606 (120 rpm)	36.1	1.6	21 d	21 d
8422X + PS1606 (140 rpm)	39.7	2.6	21 d	27 d

Grease barrier performance of dispersion coated samples is rather poor overall. Epotal® 8422X is an aberration, because it induces very high variation within the first and sixth parallel measurement. However, this can be justified by the single layer coating and consequently pinholes within the sample area. Therefore, the actual barrier performance of all dispersion coated samples can be higher, if the coating layer is continuous and uniform.

Rate of grease penetration results of extrusion coated samples disclose the distinct difference between the traditional and sustainable material choices. Ecovio® induces noticeably better grease barrier performance than LDPE. Time for penetration is roughly five times longer with ecovio® than LDPE. However, the coating weight and thus thickness of the polymer layers are different, which has an impact on the results and increase the difference.

Evaluation of tandem coating shows differences between the traditional and sustainable material alternatives. Since grease barrier performances of LDPE and both dispersion coatings are low, the combination does not provide significant benefit against the grease penetration. However, the combination of sustainable alternatives presents significant improvement in grease barrier performance, nearly doubling the penetration time compared to extrusion coating and multiplying the penetration time compared to dispersion coating.

The best grease barrier results are realized by tandem coatings of Epotal 8422X and ecovio® PS1606, which produce penetration times of 21 days (1st) and 27 days (6th) with coating weight of 39.7 g/m^2. The advantage of tandem coating is not evident, when comparing the traditional materials, but the sustainable alternatives indicate otherwise. Tandem coating can be considered beneficial, when the rate of grease penetration is slower, and materials provide sufficient barrier results. Furthermore, the grease barrier performance of tandem coated sustainable materials is much better than the results of individual coatings indicate. The extrusion coating polymer is blocking the pinholes and hence assisting and improving the barrier performance of dispersion coating layer.

In summary, the barrier performance of coating structure improves as dispersion and extrusion coating steps are combined to a single tandem process. The outcome is naturally dependent on material choices but in most cases, the permeabilities of the tandem coated structures are lower than the combination of single layer measurement results. This is due to the layer structure, wherein extrusion coating material fills in the pinholes and thus enhances the barrier performance of dispersion coating layer. The result is conclusively seen in oxygen transmission rate values, where the barrier performance improves substantially in the tandem coated structures. The similar effect can also be seen in WVTR, although the impact is not equally significant within the traditional material choices. However, this is an outcome of the very low water vapor permeation characteristics polyethylene, which diminishes the effect of dispersion coating layer. The results of grease penetration measurements validate the conclusion even further especially considering the sustainable materials, where the barrier performance is initially better and less vulnerable to deviation.

Promising conclusions can also be made regarding the material choices used in the study. The starting point was to utilize tandem coating to produce more sustainable structures compared to traditional packaging materials, since they are environmentally and legislatively preferred in modern-day packaging field. The results clearly show that required barrier performance can be achieved with bio-based materials and the permeability can be even reduced compared to the traditional materials. Especially oxygen transmission rate results show that the lowest permeability values are achieved, when combining Epotal® 8422X dispersion coating and ecovio® PS1606 extrusion coating layers by the tandem coating process. Furthermore, grease barrier results support the conclusion and even water vapor barrier is at moderate level, although it can be problematic to achieve with bio-based materials.

4 | CONCLUSIONS

In this study, different coating processes were researched and compared in the pilot-scale coating line. Furthermore, the performance of different coating materials was evaluated in order to compare traditional fossil-based materials to more sustainable and bio-based structures. The main goal was to investigate the performance and advantages of a tandem coating process, where dispersion and extrusion coating steps are combined into a continuous process. The coating structures were mainly evaluated using permeability values including oxygen, water vapor and grease transmission rates.

This study provides successful pilot-scale demonstration showing that the tandem coating process can be utilized to save valuable processing time and resources as well as to achieve enhanced barrier properties. The results clearly indicate that the barrier performance can be significantly improved with two-layer structure combining two different polymers and coating methods. Furthermore, the study confirms that the results are possible to achieve with single run process. The observations and conclusions made in this study are very promising considering the transition toward more sustainable material alternatives, which are becoming preferred choice in the future. The permeability of bio-based materials can be significantly reduced with tandem coating process and the results are comparable with fossil-based structures.

For subsequent studies, the further upscaling and optimization of the process would be very interesting and could provide important knowledge about the industrial possibilities, because the production rate was constrained by the dispersion coating speed. Furthermore, other important end-product properties such as adhesion and heat sealability could be researched in order to complement the potential of the tandem coating process.

AUTHOR CONTRIBUTIONS

Juuso Toriseva: Conceptualization (equal); data curation (lead); formal analysis (lead); investigation (lead); methodology (lead); resources (lead); validation (lead); visualization (lead); writing – original draft (lead); writing – review and editing (lead). **Johanna Lahti:** Funding acquisition (equal); project administration (equal); supervision (supporting); writing – review and editing (supporting). **Jurkka Kuusipalo:** Conceptualization (equal); funding acquisition (equal); project administration (equal); supervision (lead); writing – review and editing (supporting).

ACKNOWLEDGMENTS

The author wishes to show gratitude to partners from BASF SE for the material support. Furthermore, the author wishes to thank the research group of Paper Converting and Packaging Technology and the staff working at the pilot line and laboratory.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

ORCID

Juuso Toriseva https://orcid.org/0000-0002-9992-286X

REFERENCES

[1] J. Kuusipalo, *J. Polym. Environ.* **2000**, *8*, 39.
[2] S. Poulose, J. Toriseva, J. Lahti, I. Jönkkäri, M. Hedenqvist, J. Kuusipalo, *ASC Appl. Polym. Mater.* **2022**, *4*, 4179.
[3] E. Luoma, T. Rokkonen, A. Tribot, K. Nättinen, J. Lahtinen, *Polym. Renew. Resour.* **2022**, *13*, 1.
[4] B. H. Gregory, *Extrusion Coating: A Process Manual*, Trafford Publishing, USA **2005**, p. 224.
[5] K. Jurkka Ed., *Papermaking Science and Technology, Book 12, Paper and Paperboard Converting*, Second ed., Finnish Paper Engineers' Association, Finland **2008**, p. 346.
[6] B. A. Morris, *Science and Technology of Flexible Packaging: Multilayer Films from Resin and Process to End Use*, Elsevier Inc., USA **2017**, p. 728.
[7] J. Kuusipalo, *J. Polym. Environ.* **2000**, *8*, 49.
[8] J. Kuusipalo, *J. Polym. Environ.* **2001**, *9*, 125.
[9] E. Suokas, J. Kuusipalo, J. Toriseva, K. Nevalainen, V. Ribu. Polymer coated paper and paperboard, SE1851188A1. **2018**.
[10] V. K. Rastogi, P. Samyn, *Coatings* **2015**, *5*, 887.
[11] K. Helanto, R. Talja, O. Rojas, *J. Appl. Polym. Sci.* **2021**, *138*, 51225.
[12] K. Helanto, R. Talja, O. Rojas, *Nord. Pulp Pap. Res. J.* **2021**, *37*, 108.
[13] K. Helanto, R. Talja, O. Rojas, *Nord. Pulp Pap. Res. J.* **2022**, *37*, 212.
[14] M. Fereydoon, S. Ebnesajjad, *Chapter 5 Development of High-Barrier Film for Food Packaging, Plastic Films in Food Packaging*, Elsevier Inc., Oxford **2013**, p. 71.

[15] T. Kimpimäki, A. Savolainen, *Surface Application of Paper Chemicals*, Blackie Academic and Professional, Chapman & Hall, UK **1997**, p. 208.
[16] T. Kimpimäki, *Effect of Drying Temperature on the Coalescence of Polymer Particles and on the Properties of Dispersion Coated Paperboard, Doctoral Thesis*, Vol. 221, Tampere University of Technology Publications, Finland **1997**, p. 100.
[17] S.-S. Ovaska, *Oil and Grease Barrier Properties of Converted Dispersion-Coated Paperboards, Doctoral Thesis*, Vol. 719, Acta Universitatis Lappeenrantaensis, Finland **2016**, p. 92.
[18] R. Bollström, R. Nyqvist, J. Preston, P. Salminen, M. Toivakka, *Tappi J.* **2013**, *12*, 45.
[19] R. Koppolu, J. Lahti, T. Abitbol, A. Swerin, J. Kuusipalo, M. Toivakka, *ASC Appl. Mater. Interfaces* **2019**, *11*, 11920.
[20] M. Vähä-Nissi, H. Koivula, H. Räisänen, J. Vartiainen, P. Ragni, E. Kenttä, T. Kaljunen, T. Malm, H. Minkkinen, A. Harlin, *J. Appl. Polym. Sci.* **2017**, *134*, 8.
[21] J. Vartiainen, Y. Shen, T. Kaljunen, T. Malm, M. Vähä-Nissi, M. Putkonen, A. Harlin, *J. Appl. Polym. Sci.* **2016**, *133*, 6.
[22] Polyethylene CA7230. Product Data Sheet, Borealis AG, Vienna, Austria, 2014, 1st Ed., p. 3.
[23] ecovio® PS1606. Product Information, BASF SE, Global Marketing Biopolymers, Ludwigshafen, Germany, 2014, Version 1.0, p. 5.
[24] Epotal® S 440 eu. Polymer Dispersions for Paper, Technical Information, BASF SE, Dispersions for Adhesives, Construction & Paper Europe, Ludwigshafen, Germany, 2015.
[25] Epotal® SP-106D. Polymer Dispersions for Paper, Technical Data Sheet, BASF SE, Dispersions and Resins Europe, Ludwigshafen, Germany, 2020, p. 2.
[26] Epotal 8422X. Polymer Dispersions for Paper, Technical Information, BASF SE, Dispersions and Resins Europe, Ludwigshafen, Germany, 2017.
[27] P. Johansson, J. Lahti, J. Kuusipalo, *J. Appl. Packag. Res.* **2021**, *13*, 1.
[28] P. Johansson, J. Lahti, J. Vihinen, J. Kuusipalo, *J. Appl. Packag. Res.* **2019**, *11*, 49.

How to cite this article: J. Toriseva, J. Lahti, J. Kuusipalo, *J. Appl. Polym. Sci.* **2023**, *140*(24), e53959. https://doi.org/10.1002/app.53959