Surface Characteristics and Repulpability Performance of Cellulose-Fiber-Based Packaging Materials Coated with Aqueous Dispersions of Wood-Bark-Derived Suberin

Johanna Lyytikäinen,[a,]* Rajesh Koppolu,[b] Minette Kvikant,[c] Chunlin Xu,[c] and Ville Leminen[a]

Recyclability is an important feature of packaging materials. Although packaging materials made from cellulose fibers such as those found in paper or paperboard can typically be recycled through repulping, the application of coatings, especially polymeric plastic coatings, often impairs their recyclability, leading to increased amounts of rejects and low fiber yields. Herein, the surface properties and repulpability performance of paperboard substrates coated with aqueous dispersion of wood bark-derived suberin, stabilized using synthetic surfactants or bio-based surfactants, were investigated. The results were compared with commercial polyethylene coated material. Surface properties of the materials were investigated through surface imaging, water absorption, and wettability measurements. Repulpability was evaluated based on the amounts of rejects after two screening stages. Fiber analysis was performed for the materials that passed both the screenings. All suberin-coated materials showed hydrophilic surface characteristics and greater water absorbency than the reference material. Repulpability analysis revealed that the suberin coatings resulted in a lower amount of rejects than coated reference material. These results highlight the potential of suberin coatings in developing recyclable and sustainable packaging solutions for cellulose fiber substrates.

DOI: 10.15376/biores.21.1.2315-2329

Keywords: Coating; Paperboard; Recyclability; Repulping

*Contact information: a: Department of Mechanical Engineering, Packaging Technology, Lappeenranta-Lahti University of Technology LUT, Lappeenranta, Finland; b: Sustainable Products and Materials, VTT Technical Research Centre of Finland Ltd, Espoo, Finland; c: Laboratory of Natural Materials Technology, Åbo Akademi University, Turku, Finland; *Corresponding author: johanna.lyytikainen@lut.fi*

INTRODUCTION

Petroleum-based packaging materials have typically been used as packaging materials due their good performance in converting processes and as barrier materials. However, due to their sustainability and recyclability issues, there has been a growing interest in more sustainable packaging materials (Lee *et al*. 2017; Sid *et al*. 2021). In addition, microplastics and migration of chemicals from them may harm not only the environment but also humans (Sid *et al*. 2021). As an alternative, fiber-based packaging materials are renewable, recyclable and biodegradable; however, their porosity and hydrophilicity result in poor barrier properties. Therefore, their barrier properties are commonly improved using conventional, petroleum-based plastics. The use of plastics as coatings reduces the biodegradability and recyclability of fiber-based packaging materials. (Runte *et al*. 2015; Triantafillopoulos and Koukoulas 2020; Kathuria and Zhang 2022).

Paper or paperboard with good barrier properties can be obtained using the dispersion coating method. Dispersion coatings are water-based coatings and are considered more recyclable and repulpable than plastic coatings. However, their repulpability is typically affected by the coat weight and for example type of coating components (Sridach *et al*. 2007; Ovaska *et al*. 2017). Synthetic latex has commonly been used as a coating component in dispersion coatings, although other bio-based components have also been utilized in combination with different additives and fillers (Samyn *et al*. 2010; Ovaska *et al*. 2017). Additives such as surfactants used in coating formulations are typically made from fossil-based resources but could also be derived from more sustainable resources (Dax *et al*. 2013; Kwan *et al*. 2022).

Bio-based and biodegradable coating components and their barrier performance in fiber-based packaging materials have been widely studied. The hydrophilicity of these coatings presents a challenge, as typically the packaging material should retain its barrier performance also at elevated humidities or have water resistance characteristics (Andersson 2008). Hydrophobicity could be increased by modifying of the coating components or introducing hydrophobic components (*e.g.*, waxes) in the coatings (Rastogi and Samyn 2015; Yadav *et al*. 2024). Suberin, a hydrophobic biopolymer, has also been used in aqueous dispersions to improve hydrophobicity as well as water vapor (Korpinen *et al*. 2019; Hu *et al*. 2024) and grease barrier properties of fiber-based materials (Hu *et al*. 2024).

Repulpability and biodegradability are important features of fiber-based packaging materials. The growing utilization of recycled fibers has been driven by environmental concerns and has led to new products with lower costs (Jamnicki *et al*. 2012; Belle *et al*. 2024). However, the fiber quality and amount of recovered fibers may be reduced after several recycling times, affecting the properties of new end-products (Belle *et al*. 2024). In general, when recycling plastic-coated fiber-based materials, coatings typically end up as rejects during repulping. Subsequently, they are further incinerated or utilized as raw materials for other products (Runte *et al*. 2015; Triantafillopoulos and Koukoulas 2020; Kathuria and Zhang 2022). Although there are different solutions to remove plastic coatings during repulping, the disintegrated, small pieces of plastic coatings may still pass fine screening and end up in paper manufacturing processes (Triantafillopoulos and Koukoulas 2020). In addition, fibers tend to adhere to plastic coatings, affecting the fiber yield after repulping.

The utilization of bio-based coatings can have many advantages from an environmental perspective. For example, a nanofibrillated cellulose layer under a polyethylene (PE) coating layer can increase the recyclability of the coated material (Al-Gharrawi *et al*. 2021). However, sustainable coatings, such as those based on polylactic acid or wax, are known to reduce the fiber yield during repulping (Rastogi and Samyn 2015; Zhang *et al*. 2016; Lee *et al*. 2017). Similar to plastics, waxes tend to adhere to fibers (Samyn *et al*. 2010).

Although coated materials may be repulpable, repulped materials may still cause difficulties in papermaking processes (Andersson 2008; Ovaska *et al*. 2017). Small fractions and nanoparticles of coatings can enter the accept stream during repulping and enter the papermaking process, affecting runnability as well as properties of the end product (Popil and Schaepe 2005; Andersson 2008; Zhang *et al*. 2016). In repulping processes, the substrate and coating may increase the amount of dissolved and colloidal substances (DCS). In general, DCS negatively affect the papermaking process. However, some DCS from recycled pulps can also improve the strength of materials (Hubbe *et al*. 2012; Miao *et al*. 2013).

Previously, research has been conducted on the barrier properties of suberin-based coatings, however, less on their recyclability. Herein, the repulpability performance of paperboard substrates coated with aqueous suberin dispersions was investigated. Different suberin dispersions for coating were stabilized using conventional nonionic surfactants, polyvinyl alcohol (PVOH), and bio-based surfactants —namely modified hemicellulose and amphiphilic cellulose nanofibers (ACNFs). The surface characteristics in addition to tensile and tear indices were determined from all the materials studied. The repulpability experiments were performed with the suberin coated materials and the coated commercial polyethylene (PE) -coated material was used as reference material in repulpability studies. In addition, the repulpability of uncoated paperboard material was studied to evaluate the effects of the novel dispersion coatings on the repulpability performance.

EXPERIMENTAL

Preparation of Aqueous Suberin Dispersions

For coating of paperboard substrate, four different aqueous suberin dispersions were prepared. To obtain suberin for the aqueous dispersions, a modified pilot-scale alkaline ethanol–water fractionation process was employed to extract suberin hydrolysate from birch bark (Korpinen *et al*. 2019). The characteristics of suberin are described in detail in a publication by Hu *et al*. (2024).

To produce the PVOH-stabilized (Kuraray Poval 35-80, Kuraray) aqueous suberin dispersion, dry suberin powder was melted at 95 °C under slow mixing in a 10-L reactor. A 7% PVOH solution was added to molten suberin (at 15% (w/w) relative to suberin). Water was added with continuous mixing until a uniform dispersion was obtained.

The suberin dispersion stabilized by nonionic surfactants, namely sorbitan monooleate 80 (Span 80) and polysorbate 80 (Tween 80) with a ratio of 0.65:0.35) was prepared by mixing the surfactants (2% with respect to dry suberin) in water with mixing at 1000 to 2000 rpm for 2 min using a high-shear mixer equipped with an ultramix head (Silverson, L5M-A, USA). The pH of the solution was adjusted to 12 using sodium hydroxide (NaOH), and suberin was slowly added to the solution with mixing. The suberin–surfactant dispersion was further mixed at 2000 to 3000 rpm for 30 min, after which the pH was adjusted to 7. The dispersion was mixed for an additional 30 min at 2000 to 3000 rpm using a mixing head with a slotted design. The prepared dispersion was filtered using a metallic sieve before the coating process. For preparing suberin dispersions stabilized using ACNFs or hemicellulose (fatty-acid-grafted galactoglucomannan [GGM]), a procedure similar to that of nonionic surfactants was employed, with addition levels being 0.1% and 5% (w/w) with respect to dry suberin, respectively. The synthesis of fatty acid grafted GGM has been reported separately (Kvikant *et al*. 2025).

The ACNFs were prepared using the sequential esterification of kraft pulp with n-octyl succinic anhydride (OSA) in a deep eutectic solvent (DES) composed of imidazole and triethylmethylammonium chloride (TEMACl), followed by mechanical disintegration using a microfluidizer (Qasim *et al*. 2023). The DES was prepared by combining imidazole and TEMACl in a 7:3 molar ratio. This mixture was heated to 80 °C in an oil bath with continuous stirring at 100 rpm for ~15 min to obtain a uniform solution. Small pieces of dry kraft pulp sheets (3.15 g) were dispersed into the DES with stirring for 15 min. Subsequently, OSA (3.9 g; molar ratio = 1:1 relative to the anhydroglucose unit) was added and stirred at 80 °C and 100 rpm. After a reaction time of 2 h, ethanol (100 mL) was added to the mixture, and the modified pulp was washed thoroughly with ethanol (200 mL) and deionized water (200 mL). The ACNFs were stored at −17 °C until nanofibrillation and further analysis. To homogenize the ACNF suspension (0.5 wt%), the Ultra-Turrax mixer (IKA T25, Germany) was used at 10,000 rpm for 10 min before isolating the ACNFs using a microfluidizer (Microfluidics M-110EH-30, USA). The suspension was passed through 400- and 200-µm chambers five times at a pressure of 150 MPa to obtain the ACNFs.

An amphiphilic GGM (AGGM) derivative was produced from spruce wood extract obtained through pressurized hot water extraction. The AGGM was supplied by CH-Bioforce. To ensure proper modification and impurity removal, the spruce extract was subjected to ethanol precipitation. The GGM concentrate was diluted to 20 wt.%, precipitated in 85% ethanol, and filtered to collect the precipitate. This process was repeated twice, with the precipitate being redissolved to a 20 wt.% concentrate each time. After the final precipitation, the GGM extract was washed sequentially with acetone, methyl tert-butyl ether, and 2-propanol, and then dried in a vacuum oven at 40 °C.

The purified GGM product had a weight average molar mass (M_w) of 8.7×10^3 g/mol and a number average molar mass (M_n) of 6.8×10^3 g/mol, as determined *via* high-pressure size exclusion chromatography. Its sugar composition was determined to be ~4:1:1 (mannose:glucose:galactose) *via* acid methanolysis. The degree of acetylation, quantified *via* high-performance liquid chromatography, was 0.20.

An *in situ* esterification reaction was performed to synthesize the AGGM derivative. Stearic acid was dissolved in dimethyl sulfoxide (DMSO) at 65°C and activated with 1 equivalent of 1,1-carbonyldiimidazole under stirring until no more carbon dioxide was produced. Stirring was continued for an additional hour to ensure complete activation. The purified GGM product was dissolved in the DMSO solution and mixed for 15 h at 65 °C. The resulting mixture was precipitated in acetone, filtered, washed with acetone, and dried in a vacuum oven at 40 °C to isolate AGGM.

Paperboard Substrates

A pigment-coated folding boxboard (MetsäBoard Prime FBB Bright, Metsä Board Oyj, Finland) with a grammage of 210 g/m² and a three-layer structure was used as the substrate for the suberin-based dispersion coatings. A paperboard with a one-sided PE coating (MetsäBoard Pro Bright 1PE, Metsä Board Oyj, Finland) was used as reference material in repulpability studies. The grammage of the PE-coated paperboard material was 245 g/m².

Coating Process

The coating was done using a roll-to-roll minipilot coater (Surface Treatment Concept, SUTCO) at VTT, Espoo, Finland. A reverse gravure coating method was employed, utilizing a gravure roller with a surface volume of 125 cc/m². The paperboard (MetsäBoard Prime FBB Bright, Metsä Board Oyj, Finland) was used as the substrate for the coatings. The coating speed was 7 m/min, and a combination of infrared and air dryers was used to dry the wet suspensions. The solid contents of the suberin-based dispersions containing the nonionic surfactants, PVOH, hemicellulose, and ACNFs were 20%, 22%, 17% and 15%, respectively.

Material Characterization

The grammage and thickness of uncoated and coated materials were measured according to ISO 536 (2012) and ISO 534 (2005) standards, respectively. The tearing resistance of materials was measured according to the ISO 1974 (2013) standard. All materials were conditioned at 23 °C and 50% relative humidity before testing.

The thickness of the suberin-coated materials was calculated based on the thickness of the base and coated substrates. The coating thickness of the base substrate in the coated materials and reference PE-coated material have been reported in a study by Lev *et al.* (2024).

The repulpability of the suberin-coated and reference material was analyzed based on the quantities of coarse rejects, fine rejects, and DCSs, following the Cepi Recyclability Laboratory Test Method (Confederation of European Paper Industries 2020). The contents of DCS (dissolved and colloidal substances) in disintegrated samples were determined through evaporation. The coarse screening for the samples was performed using a screening plate with 5-mm-diameter holes, and fine screening was performed using a screening plate with 0.15-mm gaps. The amounts of DCSs and rejects are reported as percentages. The composition of the materials that passed through the fine screening plate was assessed visually to characterize the remaining fractions.

A scanning electron microscopy (SEM) device (Hitachi SU3500, Tokyo, Japan) with a secondary electron detector was utilized to capture surface images of the coated and uncoated materials. The samples were sputter-coated with an Au target before imaging. The working distance was ~7 mm, and the acceleration voltage was 5 kV.

Contact angles of water were measured using the Theta optical tensiometer (Biolin Scientific AB, Gothenburg, Sweden). Water with a drop volume of 3 µL was used as the test liquid, and five measurements were performed for each material. The materials were conditioned at 23°C and 50% relative humidity before measurements.

The water absorptiveness (Cobb60) of the materials was measured following the ISO 535 (2014) standard using a test area of 25 cm^2. Five measurements were performed for each material, and the resulting values are reported in grams per square meter.

A Fiber Tester (Lorentzen and Wettre, Kista, Sweden) was used to determine the fines content, shape factor, and kinks per millimeter of the fibers in the accept fraction after the fine screening phase. The objects were defined as fines if the length of the object was <200 µm. The amount of fines was calculated from the amount of fibers and reported as percentage values.

RESULTS AND DISCUSSION

Material Properties

Results for the coat weight, which is expressed as a percentage of the total grammage of the material, coating thickness, and tear index of all materials, are presented in Table 1. The reference material had lower coating thickness than the suberin-based coated materials. Among the suberin-based coatings, formulations incorporating ACNFs exhibited lower coat weight and coating thickness than those containing other stabilizers, probably due to the lower solid content of the coating solution used in the coating process.

Table 1. Coat Weight and Coating Thickness of the Materials

Coating composition	Coat weight (%)	Coating thickness (μm)
PE	6.5	10*
Base for suberin coating	-	-
Suberin–Nonionic surfactants	10.0	35
Suberin–PVOH	11.2	38
Suberin–AGGM	9.2	31
Suberin–ACNF	8.6	28

* Lev *et al*. (2024)

Note: The coat weights were calculated from the total grammage of the materials, where applicable, and were reported as percentages.

The tensile and tear indices for the material are presented in Fig. 1. The wetting and penetration of a coating solution into a material during the coating and drying of the coated material can reduce its tensile properties (Rhim and Kim 2009). Reduced tensile properties were observed for the suberin-coated materials compared with the base substrate. Among all the materials, the tear index was the highest for the PE-coated material and was similar in the machine direction (MD) and cross direction (CD). In general, plastic coatings can show good mechanical strength, which may affect their behavior during recycling (Al-Gharrawi *et al*. 2021; Belle *et al*. 2024). The suberin-coated materials showed similar tear indices in both directions. In the CD, the tear resistance of the suberin-coated materials increased compared with that of the uncoated substrate, except for the material in which the coating contained hemicellulose. Although the coat weight seemed to affect the tear resistance, the effect varied among the suberin-coated materials probably because of the mechanical strength and uniformity of the coating layer. Wetting of materials affects the strength properties, but the results may partly indicate the behavior of the coating under disintegration during repulping (Zhang *et al*. 2016).

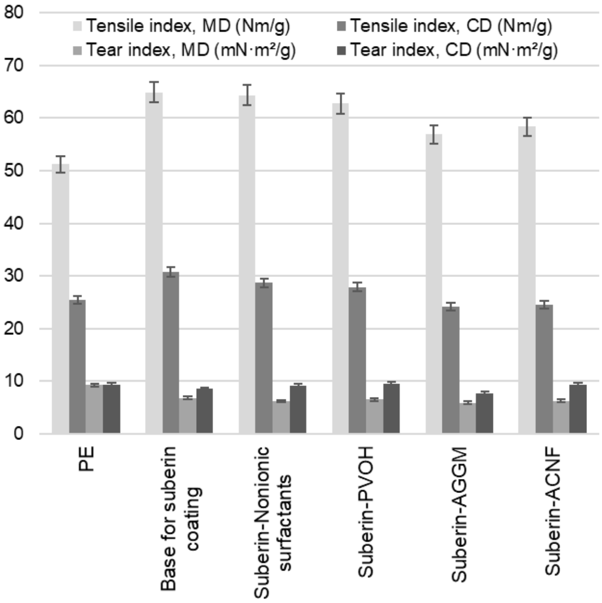

Fig. 1. Tensile and tear indices of the materials. MD and CD refer to the machine and cross directions, respectively

SEM Analysis

SEM images of all material surfaces were obtained to evaluate coating coverage and uniformity, which may further affect the wetting and water absorption behavior of the materials in addition to the coating composition. As expected, the PE-coated material showed a uniform surface free from defects (Fig. 2A). Some defects were observed on the substrate used as the base material for the coatings (Fig. 2B).

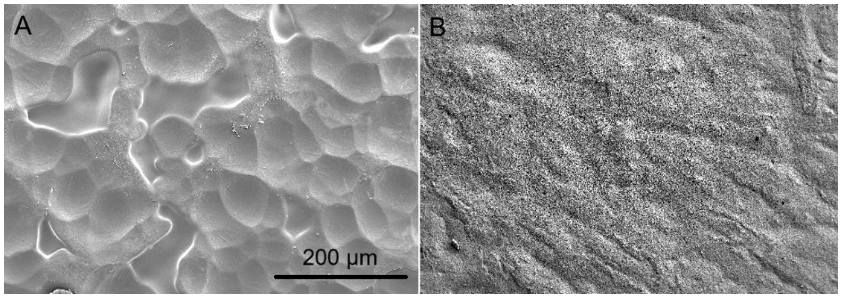

Fig. 2. SEM surface images of reference (a) PE coating, and (b) the uncoated substrate. Scale bar = 200 µm.

The different stabilizers led to differences between the coated surfaces (Fig. 3). Compared to the coating with synthetic nonionic surfactants (Fig. 3A), the PVOH-containing coating (Fig. 3B) had more defects, which may be due to air in the coating paste attributed to the foam-forming properties of PVOH (Gottberg *et al*. 2019). The large proportion of PVOH in comparison to that of the other suberin coatings likely led to the higher coverage of the base substrate. The use of ACNFs apparently led to a rougher surface, as the nanofibers seemingly remained on the top of the coating but were covered by suberin (Fig. 3D). However, few defects were observed in the coating. In the case of the suberin–AGGM coating (Fig. 3C), air in the coating paste probably caused blistering and resulted in clear ruptures of the coating layer, which may have affected the mechanical strength of the material, as shown in Fig. 1.

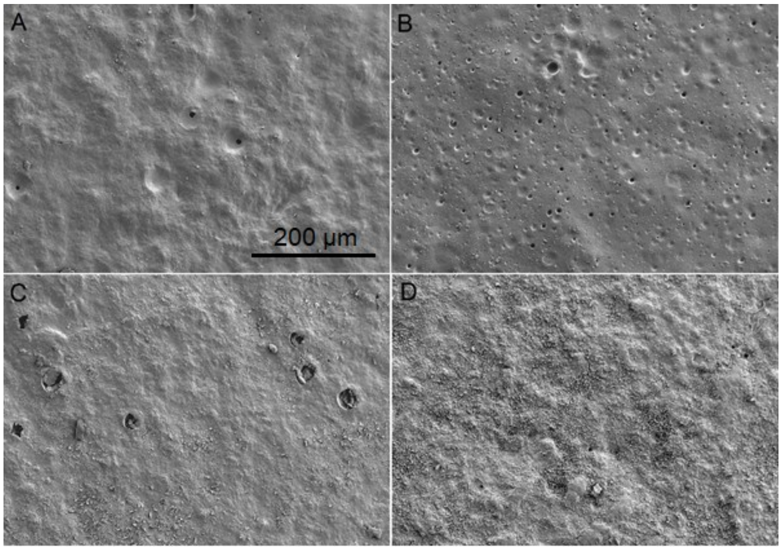

Fig. 3. SEM surface images of the suberin coatings containing (a) the nonionic surfactants, (b) PVOH, (c) AGGM, and (d) ACNFs. Scale bar = 200 µm.

Surface Wettability and Water Absorption

The contact angles of water were measured to assess the wettability of the coated and uncoated surfaces (Fig. 4A). In general, different coatings and surface modifications can affect the hydrophobicity or hydrophilicity of the surface. As the water contact angle was >90° for the PE-coated material, the surface was considered hydrophobic (Cirisano and Ferrari 2021). Meanwhile, other surfaces were considered hydrophilic, including the base substrate with a water contact angle of 80°. The surface wettability of the AGGM- and ACNF-containing coatings was high. In particular, the AGGM-containing coating absorbed water rapidly onto the surface probably because of its defects and the hydrophilicity of hemicellulose (Hubbe *et al*. 2012; Lozhechnikova *et al*. 2014). The high hydrophilicity of the suberin–AGGM coating indicated that the hydrophilic parts of AGGM may have affected the water absorption of the coating (Lozhechnikova *et al*. 2014). Coatings prepared from ACNFs have been reported to generate hydrophobic surfaces (Qasim *et al*. 2023). In this study, the combination of ACNFs with suberin led to increased wettability of the resulting coating.

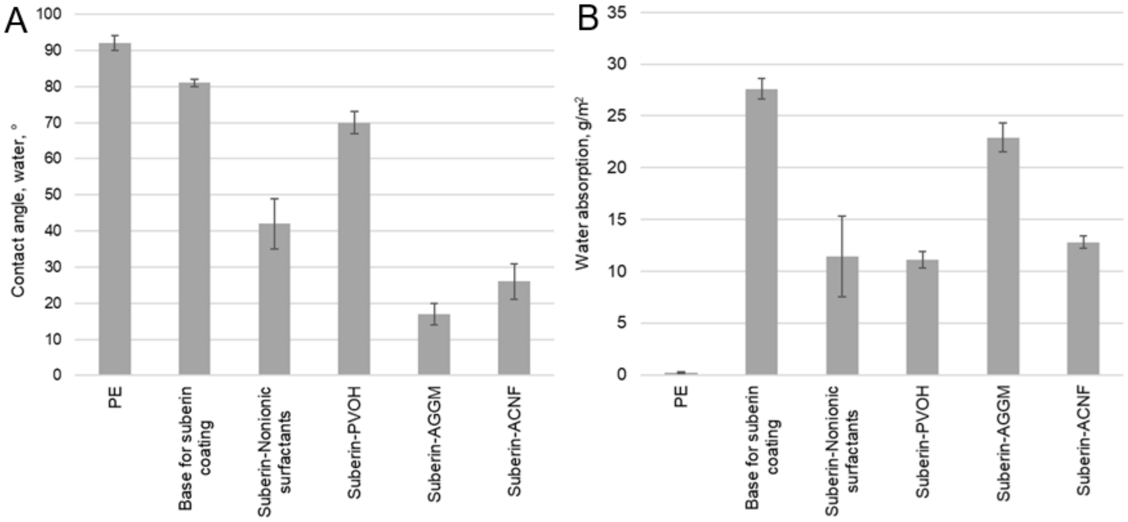

Fig. 4. (a) Contact angles of water and (b) water absorption on the coated surfaces and the base substrate.

The water absorptiveness of the materials is shown in Fig. 4B. The reference PE-coated material exhibited the lowest water absorption. In general, the structure of the coating layer, the characteristics of the coating components and larger amount of water may affect the water absorptiveness of the material (Mesic *et al*. 2010). The uncoated substrate showed high water absorption capacity even though its water contact angle was ~80°. Higher water absorption leads to higher wetting of the material and may indicate good disintegration and repulping performance. However, other characteristics, such as mechanical durability of coatings under wetting and the raw materials of the coating and substrate, should also be considered in terms of repulpability (Al-Gharrawi *et al*. 2021; Belle *et al*. 2024).

Compared with the uncoated material, the suberin-coated materials showed reduced water absorption. The hydrophobic characteristics of suberin may have reduced water absorption by the materials even though defects were present on the coatings. The reduction in water absorption was more evident for the coatings containing the nonionic surfactants and PVOH, whereas the coating with hemicellulose showed increased water absorption. Defects in the PVOH-containing coating were apparently more on the surface and not within the coating. Meanwhile, more severe defects were observed in the hemicellulose-containing coating.

Repulpability

Typically, the weight and composition of a coating affect the repulpability of materials (Ovaska *et al*. 2017). Herein, the PE-coated material led to a high amount of coarse rejects (Fig. 5A) as the PE coating withstood disintegration and was retained as large pieces on the screening plate. Meanwhile, the pigment coating on the paperboard substrate was fragmented during disintegration, passed through the coarse screening plate, and was collected in the accept fraction. In the case of the suberin-based coatings, the amount of coarse rejects was low, which indicated that the coatings disintegrated into pieces small enough to pass through the coarse screening plate. However, foaming occurred during the coarse screening phase for the suberin-based coatings (Fig. 5B and 5C), which may prevent the material passing the screening plate and thus the amount of reject retained on the plate. In addition, when hemicellulose or ACNFs were used in the coatings, the formed foam was more stable compared with other materials. Nevertheless, foaming did not seem to affect the screening process as the amount of rejects was similar for all the suberin-based coatings.

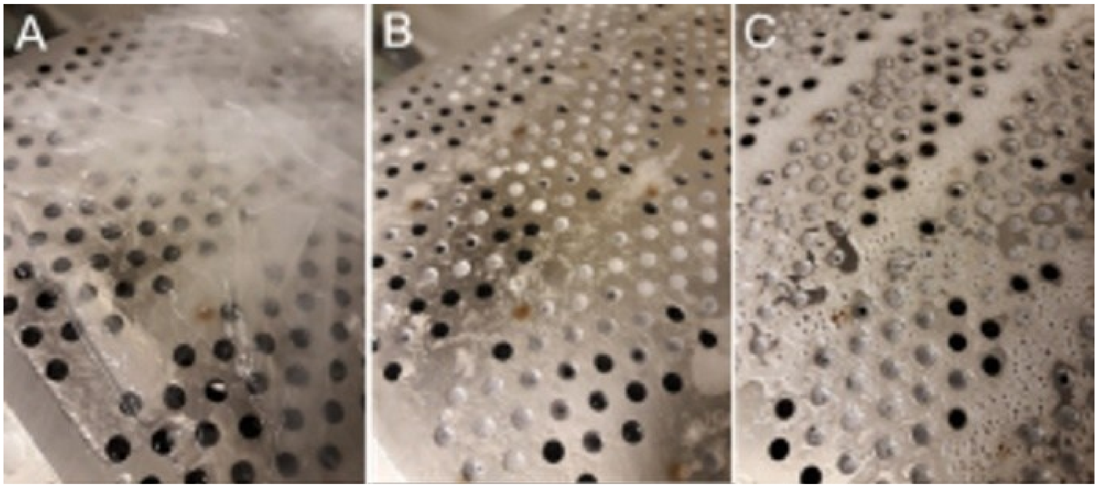

Fig. 5. Coarse rejects from (a) PE, (b) suberin–nonionic surfactants, and (c) suberin–ACNF coated materials.

During fine screening, most fibers and the coating material in the case of the suberin-containing coatings passed through the screening plate with narrow gaps. However, the longest fibers and a few fiber bundles were left on the screening plate, except in the case of the PVOH-containing coating, for which small fragments of the suberin-based coating were left on the fine screening plate. This was probably because of the higher coat weight and lower wettability and water absorption capacity of the PVOH-containing coating compared with those of the other suberin-containing coatings. Nevertheless, the suberin-based coatings did not notably affect the repulpability of the substrates compared to the base substrate in terms of the reject proportions. In general, fibers may attach to coatings because of the penetration of the coating solution into the substrate, which can increase the reject rate and reduce fiber yield (Zhang *et al*. 2016; Al-Gharrawi *et al*. 2021).

The calculated amounts of rejects and colloidal and dissolved substances are presented in Table 2. As shown in Fig. 5, the amount of rejects was the largest for the PE-coated material. In addition, as shown in Table 1, the amount of coating was 6.5% of the total material content and the total reject was 6.8%. This could indicate that the fibers remained on the screening plate along the plastic coating and that some fibers could have attached to the coating (Al-Gharrawi *et al*. 2021). The amount of coating for the suberin coatings was between 8.6% and 11%, expressed as a percentage of the total grammage of the material, whereas the total amount of reject was small for the suberin-containing coatings. This may indicate that the coatings disintegrated and probably dissolved during the repulping process, resulting in low reject rates. With their low amounts of total rejects, the suberin-coated materials could also be considered repulpable.

DCS were also determined from all the samples according to the test method (Table 2). Typically, DCS negatively affects papermaking, but in the case of recycling and depending on the DCS components, they can also strengthen materials in some cases (Miao *et al*. 2013). The coated reference material had lower amounts of DCS than the other tested materials. Compared with the uncoated substrate, the amount of DCS increased when the suberin coating was applied. Meanwhile, the larger amounts of DCS in the PVOH-containing coating could be explained by its higher coat weight and thus larger share of the coating in the sample compared with the other samples in repulping. Similarly, DCS correlated with the coat weight of the AGGM- and ACNF-containing coatings.

Table 2. Repulpability of the Coated Substrates

Coating composition	Coarse reject (%)	Fine reject (%)	Dissolved and colloidal substances (%)
PE	6.6 ± 0.1	0.2 ± 0.0	1.5 ± 0.2
Base for suberin coating	0.1 ± 0.0	0.0 ± 0.0	3.8 ± 0.1
Suberin-Nonionic surfactants	0.3 ± 0.0	0.2 ± 0.0	4.4 ± 0.0
Suberin-PVOH	0.3 ± 0.0	0.1 ± 0.0	5.8 ± 0.2
Suberin-AGGM	0.3 ± 0.0	0.1 ± 0.0	4.9 ± 0.0
Suberin-ACNF	0.3 ± 0.1	0.1 ± 0.0	4.3 ± 0.1

Note: The amounts of coarse and fine rejects and colloidal and dissolved substances are expressed as percentages.

Figure 6 shows the differences between the compositions and examples of coating particles that passed the fine screening plate. Pigment coating particles were present in all compositions. Some particles from the suberin coating were found in the accept fraction after fine screening when the surfactant (Fig. 6A) or PVOH (Fig. 6C) was used in the coating composition. PVOH used in this study also improved adhesion, which could influence the coating fragments observed in the accept fraction after fine screening. Fewer particles were found when ACNF was used in the coating (Fig. 6E). These results seem to correlate with the water absorption results, where the suberin–hemicellulose coating had the largest water absorbency, indicating that the suberin was dissolved during the repulping process with this composition (Fig. 6D). Examples of suberin-based and pigment coating particles are presented in Fig. 6B and 6F.

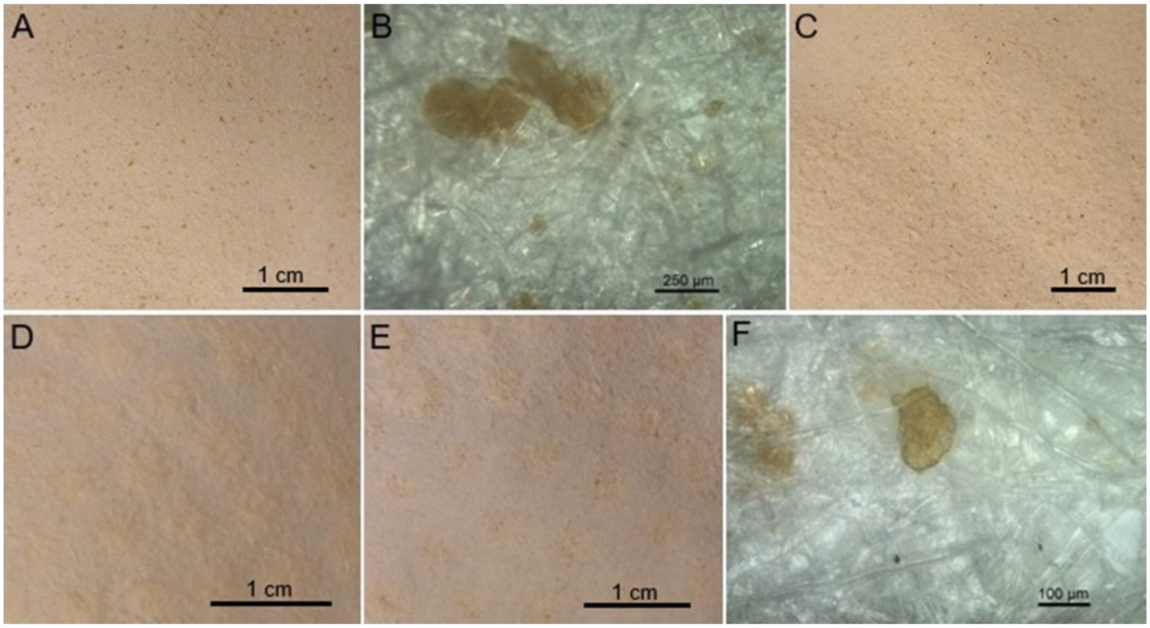

Fig. 6. Accept fractions of suberin-coated materials on filter paper containing (a, b) nonionic surfactants, (c) PVOH, (d) AGGM, and (e, f) ACNF

Fiber Analysis

Changes in the fiber dimensions and proportion of fines after disintegration and fine screening were evaluated (Table 3). No significant changes were observed for the uncoated and suberin-coated paperboard samples. This result agreed with a suggestion that repulping does not notably affect fibers or decrease the fiber length (Hubbe *et al*. 2007). Nevertheless, recycling generally affects fibers, typically resulting in reduced strength after several recycling events (Hubbe *et al*. 2007).

Table 3. Fines Content, Number of Kinks, and Shape Factor of the Samples after Disintegration and Screening Stages

Coating composition	Fines (%)	Kinks/mm	Shape (%)
	After disintegration		
PE	9.3 ± 0.4	0.29 ± 0.01	91.3 ± 0.1
Base for suberin coating	8.2 ± 0.1	0.42 ± 0.01	91.4 ± 0.0
Suberin-Nonionic surfactants	8.0 ± 0.3	0.41 ± 0.01	91.0 ± 0.2
Suberin-PVOH	8.7 ± 0.3	0.38 ± 0.04	91.2 ± 0.1
Suberin-AGGM	8.0 ± 0.3	0.41 ± 0.01	91.1 ± 0.2
Suberin-ACNF	7.9 ± 0.1	0.43 ± 0.00	90.9 ± 0.1
	After fine screening		
PE	9.5 ± 0.2	0.26 ± 0.00	91.9 ± 0.1
Base for suberin coating	8.3 ± 0.3	0.38 ± 0.01	91.6 ± 0.1
Suberin-Nonionic surfactants	8.2 ± 0.1	0.39 ± 0.00	91.4 ± 0.1
Suberin-PVOH	8.1 ± 0.2	0.40 ± 0.01	91.4 ± 0.2
Suberin-AGGM	8.2 ± 0.3	0.39 ± 0.00	91.4 ± 0.2
Suberin-ACNF	8.1 ± 0.2	0.39 ± 0.01	91.6 ± 0.1

In this study, the number of kinks per millimeter of fibers decreased after the screening stages. The decrease in the number of kinks and an increase in the shape factor positively affect a material's mechanical properties, but an increase in fiber curls may increase the three-dimensional elongation of the material (Joutsimo *et al*. 2005; Laukala *et al*. 2023).

Fines typically negatively affect the dewatering process in paper machines (Hubbe *et al*. 2007). Notable differences were not observed between the proportions of fines in the suberin-coated samples. Nevertheless, the PE-coated material contained the highest proportion of fines. A minor change in fines content after fines screening was observed for the PVOH-containing coating.

CONCLUSIONS

1. All the substrates coated with suberin dispersions led to lower amounts of rejects than commercial plastic-coated reference material. This was probably because of increased wettability, increased water absorption, and reduced resistance to mechanical stresses during disintegration.
2. Scanning electron microscope (SEM) images revealed defects on the suberin-based coatings, which influenced water absorption by the coatings along with the coating thickness and mechanical properties.
3. When poly(vinyl alcohol) (PVOH) was used as a stabilizer, small coating fragments containing suberin were retained on the fine screening plate. This was possibly due to the higher proportion and adhesion characteristics of PVOH in the coating as well as a higher coat weight compared with other suberin coated materials.
4. Optimizing the coating process can help to obtain required properties of packaging materials while maintaining their recyclability. The behavior and performance of repulped fibers in papermaking should be clarified in the future to better understand the recyclability of novel dispersion-coated materials.

ACKNOWLEDGMENTS

This research was conducted as part of the Sustainable Binders and Coatings (SUSBINCO) project (No. 2822/31/2021), funded partially by the Bio&Circular Finland Program of Business Finland. Metsä Board Oyj, University of Oulu and Prof. Henrikki Liimatainen are acknowledged gratefully for providing materials for the study. Nea Laitinen is thanked for assistance with laboratory work.

Conflict of Interest

The authors declare that they have no conflicts of interest.

REFERENCES CITED

Al-Gharrawi, M. Z., Wang, J., and Bousfield, D. W. (2021). "Improving recycling of polyethylene-coated paperboard with a nanofibrillated cellulose layer," *BioResources* 16(2), 3285-3297. https://doi.org/10.15376/biores.16.2.3285-3297

Andersson, C. (2008). "New ways to enhance the functionality of paperboard by surface treatment – A review," *Packag. Technol. Sci*. 21(6), 339-373. https://doi.org/10.1002/pts.823

Belle, J., Hirtz, D., and Sängerlaub, S. (2024). "Expert survey on the impact of cardboard and paper recycling processes, fiber-based composites/laminates and regulations, and their significance for the circular economy and the sustainability of the German paper industry," *Sustainability* 16(15), article 6610. https://doi.org/10.3390/su16156610

Cirisano, F., and Ferrari, M. (2021). "Sustainable materials for liquid repellent coatings," *Coatings* 11(12), article 1508. https://doi.org/10.3390/coatings11121508

Confederation of European Paper Industries (2020). "Harmonised European laboratory test method to produce parameters enabling the assessment of the recyclability of paper and board products in standard paper and board recycling mills," Brussels, Belgium.

Dax, D., Eklund, P., Hemming, J., Sarfraz, J., Backman, P., Xu, C., and Willför, S. (2013). "Amphiphilic spruce galactoglucomannan derivatives based on naturally-occurring fatty acids," *BioResources* 8(3), 3771-3790. https://doi.org/10.15376/biores.8.3.3771-3790

Gottberg, J., Lappalainen, T., and Salminen, K. (2019). "Polyvinyl alcohol as foaming agent in foam formed paper," *Tappi J*. 18(8), 475. https://doi.org/10.32964/tj18.8.475

Hu, L., Koppolu, R., Hämäläinen, R., Kanerva, H., Nick, T., Toivakka, M., Korpinen, R., Saranpää, P., Qasim, U., Liimatainen, H., Xu, C., and Anghelescu-Hakala, A. (2024). "Suberin-based aqueous dispersions for barrier packaging applications," *ACS Sustain. Chem. Eng*. 12(23), 8902-8912. https://doi.org/10.1021/acssuschemeng.4c02244

Hubbe, M.A., Venditti, R.A., and Rojas, O.J. (2007). "What happens to cellulosic fibers during papermaking and recycling? A review," *BioResources* 2(4), 739-788. https://doi.org/10.15376/biores.2.4.739-788

Hubbe, M. A., Sundberg, A., Mocchiutti, P., Ni, Y., and Pelton, R. (2012). "Dissolved and colloidal substances (DCS) and the charge demand of papermaking process waters and suspensions: A review," *BioResources* 7(4), 6109-6193. https://doi.org/10.15376/biores.7.4.6109-6193

Hubbe, M. A., Gardner, D. J., and Shen, W. (2015). "Contact angles and wettability of cellulosic surfaces: A review of proposed mechanisms and test strategies," *BioResources* 10(4), 8657-8749. https://doi.org/10.15376/biores.10.4.Hubbe_Gardner_Shen

ISO 534 (2005). "Determination of thickness, density and specific volume," International Organization for Standardization, Geneva, Switzerland.

ISO 535 (2014). "Determination of water absorptiveness — Cobb method," International Organization for Standardization, Geneva, Switzerland.

ISO 536 (2012). "Determination of grammage," International Organization for Standardization, Geneva, Switzerland.

ISO 1924-3 (2005). "Determination of tensile properties — Part 3: Constant rate of elongation method (100 mm/min)," International Organization for Standardization, Geneva, Switzerland.

ISO 1974 (2013). "Determination of tearing resistance — Elmendorf method," International Organization for Standardization, Geneva, Switzerland.

Jamnicki, S., Lozo, B., Rutar, V., and Barušić, L. (2012). "A study on the food contact suitablity of recycled paper and board," *Papíripar* 54(4), 14-20

Joutsimo, O., Wathén, R., and Tamminen, T. (2005). "Effects of fiber deformations on pulp sheet properties and fiber strength," *Paperi ja Puu* 87(6), 392

Kathuria, A., and Zhang, S. (2022). "Sustainable and repulpable barrier coatings for fiber-based materials for food packaging: A review," *Front. Mater*. 9, article 929501. https://doi.org/10.3389/fmats.2022.929501

Korpinen, R.I., Kilpeläinen, P., Sarjala, T., Nurmi, M., Saloranta, P., Holmbom, T., Koivula, H., Mikkonen, K.S., Willför, S., and Saranpää, P.T. (2019). "The hydrophobicity of lignocellulosic fiber network can be enhanced with suberin fatty acids," *Molecules* 24(23), article 4391. https://doi.org/10.3390/molecules24234391

Krainer, S., and Hirn, U., (2021). "Contact angle measurement on porous substrates: Effect of liquid absorption and drop size," *Colloids Surf. A.: Physicochem. Eng. Asp*. 619, article 126503. https://doi.org/10.1016/j.colsurfa.2021.126503

Kvikant, M., Dax, D., Toivakka, M., Filonenko, S., and Xu, C. (2025). "Amphiphilic hemicellulose derivatives as stabilizers in oil-in-water emulsions," *Int. J. Biol. Macromol*. 163(4), article 143094. https://doi.org/10.1016/j.ijbiomac.2025.143094

Kwan, I., Huang, T., Ek, M., Seppänen, R., and Skagerlind, P. (2022). "Bark from Nordic tree species – A sustainable source for amphiphilic polymers and surfactants," *Nord. Pulp Paper Res. J*. 37(4), 566-575. https://doi.org/10.1515/npprj-2022-0003

Laukala, T., Tanninen, P., Aghamohammadi, G., and Leminen, V. (2023). "Increasing 3D elongation potential in fixed blank forming *via* fiber curlation," *Cellulose* 30(3), 1449-1466. https://doi.org/10.1007/s10570-022-04984-x

Lee, T. J., Yoon, C., and Ryu, J. Y. (2017). "A new potential paper resource; recyclability of paper cups coated with water-soluble polyacrylate-based polymer," *Nord. Pulp Paper Res. J*. 32(1), 155-161. https://doi.org/10.3183/NPPRJ-2017-32-01-p155-161

Lev, R., Lyytikäinen, J., Matthews, S., Koppolu, R., Tanninen, P., and Leminen, V. (2024). "Investigating the convertibility and barrier performance of a novel suberin dispersion coating during the paperboard creasing process," *Packag. Technol. Sci*. 37(11), 1037-1048. https://doi.org/10.1002/pts.2845

Lozhechnikova, A., Dax, D., Vartiainen, J., Willför, S., Xu, C., and Österberg, M. (2014). "Modification of nanofibrillated cellulose using amphiphilic block-structured galacto-glucomannans," *Carbohydr. Polym*. 110, 163-172. https://doi.org/10.1016/j.carbpol.2014.03.087

Mesic, B., Kugge, C., and Järnström, L. (2010). "Superhydrophobic paper coating containing nonconventional clay," *Tappi J*. 9(11), 33-40. https://doi.org/10.32964/TJ911.33

Miao, Q., Huang, L., and Chen, L. (2013). "Advances in the control of dissolved and colloidal substances present in papermaking processes: A brief review," *BioResources* 8(1), 1431-1455. https://doi.org/10.15376/biores.8.1.1431-1455

Ovaska, S. S., Mielonen, K., Lyytikäinen, J., Hirn, U., and Backfolk, K. (2017). "Determination of repulpability of talc-filled biopolymer dispersion coatings and optimization of repulped reject for improved material efficiency by tailoring coatings," in: *2017 TAPPI PaperCon*, Minneapolis, MN, USA, pp. 36-48.

Popil, R. E., and Schaepe, M. (2005). "Comparative evaluation of the potential for wax-alternative packaging coatings," *Tappi J*. 4(8), 25-32

Qasim, U., Suopajärvi, T., Sirviö, J.A., Backman, O., Xu, C., and Liimatainen, H. (2023). "Pickering emulsions and hydrophobized films of amphiphilic cellulose nanofibers synthesized using ring-opening esterification in deep eutectic solvent," *Biomacromolecules* 9(9), 4113-4122. https://doi.org/10.1021/acs.biomac.3c00472

Rastogi, V. K., and Samyn, P. (2015). "Bio-based coatings for paper applications," *Coatings*. 5(4), 887-930. https://doi.org/10.3390/coatings5040887

Rhim, J. W., and Kim, J.-H. (2009). "Properties of poly(lactide)-coated paperboard for the use of 1-way paper cup," *J. Food Sci*. 74(2), E105-E111. https://doi.org/10.1111/j.1750-3841.2009.01073.x

Runte, S., Putz, H. J., Bussini, D., Limongi, L., and Elegir, G. (2015). "Recyclability criteria for paper based packaging products," *Cellul. Chem. Technol*. 49(7-8), 667-676.

Samyn, P., Deconinck, M., Schoukens, G., Stanssens, D., Vonck, L., and Van den Abbeele, H. (2010). "Modifications of paper and paperboard surfaces with a nanostructured polymer coating," *Prog. Org. Coat*. 69, 442-454. https://doi.org/10.1016/j.porgcoat.2010.08.008

Sid, S., Mor R. S., and Kishore, S. (2021). "Bio-sourced polymers as alternatives to conventional food packaging materials: A review," *Trends Food. Sci. Technol*. 115, 87-104. https://doi.org/10.1016/j.tifs.2021.06.026

Sridach, W., Hodgson, K. T., and Nazhad, M. (2007). "Biodegradation and recycling potential of barrier coated paperboards," *BioResources* 2(2), 179-192. https://doi.org/10.15376/biores.2.2.179-192

Triantafillopoulos, N., and Koukoulas, A. A. (2020). "The future of single-use paper coffee cups: Current progress and outlook," *BioResources* 15(3), 7260-7287. https://doi.org/10.15376/biores.15.3.Triantafillopoulos

Yadav, S., Khan, A., Hamdani, S.S., and Rabnawaz, M. (2024). "Degradable polymeric waxes for paper coating applications," *ACS Appl. Polym. Mater*. 6(6), 3263-3272. https://doi.org/10.1021/acsapm.3c03072

Zhang, H., Bussini, D., Hortal, M., Elegir, G., Mendes, J., and Beneyto, M. J. (2016). "PLA coated paper containing active inorganic nanoparticles: Material characterization and fate of nanoparticles in the paper recycling process," *Waste Manage*. 52, 339-345. https://doi.org/10.1016/j.wasman.2016.03.045

Article submitted: September 26, 2025; Peer review completed: November 22, 2025; Revised version received: December 1, 2025; Accepted: January 9, 2026; Published: January 26, 2026.
DOI: 10.15376/biores.21.1.2315-2329