FINAL REPORT

SUSBINCO
Sustainable binders and coatings

SUSBINCO

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Content

Executive summary

Humanity is facing numerous major challenges, including the energy crisis, climate change and other environmental problems. We must seek solutions to ensure a sustainable future. SUSBINCO addresses to those challenges by substituting fossil-based raw materials with bio-based solutions in producing bio-based binders and coatings for packaging, paints, adhesives, sealants and abrasives. These solutions are urgently needed to produce safe alternatives and reduce greenhouse gas emissions.

SUSBINCO brings together a consortium of several organisations and 11 industrial partners. SUSBINCO is a co-innovation project funded by Business Finland from 1 September 2021 to 31 July 2024 with a total budget of EUR 10.1 million.

The research organisations started the project tasks by sharing the know-how and infrastructure to complement each other, having strongly supported the joint development of research and innovation. As a result, many tasks were conducted under collaboration, which was not in the initial plan. SUSBINCO has a strong emphasis on the use of bio-based waste and side streams, such as suberin from bark, nanocellulose from sawdust, hemicellulose and lignin as side streams in forest product processes. Regarding the technology renewal, pilot-scale experiments such as production of suberin and coating were successfully conducted. A sustainability assessment was also performed from the design of the experiment on a laboratory scale. This will certainly benefit upscaling and improve the feasibility of the future processes. The environmental impact of the raw materials and their products was also evaluated to ensure that the direction of development could be more sustainable.

Six industries are conducting their own parallel projects, and the other five are joining in a steering role. Industry involvement is evident not only through their active steering in consortium and work package meetings, but also in the form of open-minded discussions, facilitating knowledge exchange in both directions. Moreover, the industrial partners have provided strong support with their expertise covering the entire value chain.

SUSBINCO's mission is to increase and enhance the global business opportunities for the companies and the industries in the consortium and network by jointly developing new sustainable binders and coatings that meet market needs. The innovations developed are aimed at overcoming acute difficulties and grasping untapped market opportunities. The total value of the global packaging market was USD 850 billion in 2016; the global paints and coatings market was USD 146 billion (2019), and the global adhesives and sealants market was USD 52 billion (2017). All of the markets are expecting strong growth and are trending towards bio-based solutions. SUSBINCO ties in significant impact along with the consortium partners' parallel projects. Companies with parallel projects alone are expecting to have over EUR 532 million worth of exports in 2030 through the bio-based coatings and binders business.

Professor Chunlin Xu
Åbo Akademi University
Scientific Director of the project

Christiane Laine
UPM
Chairperson of the steering group

SUSBINCO in brief

Author/coordinator Taina Kujanpää

SUSBINCO in numbers

Duration	1 Sept 2021–31 July 2024
Budget	EUR 10.1 million
Company budget	EUR 5.2 million
Research institution budget	EUR 4.9 million
Number of persons involved	approx. 80
Number of publications	10

The consortium

During the SUSBINCO project, the primary objective was to pioneer innovative solutions by substituting conventional fossil-based binders and coatings with eco-friendly bio-based alternatives, suitable for diverse applications in packaging, paints, adhesives, sealants, and abrasives.

The overarching goal was to expand and enrich global business opportunities for companies and industries within the consortium and network. A key milestone was aiming for increased adoption of bio-based coating materials, which was validated through the successful testing of 100% pilot coating during the project duration.

The project partners collaboratively developed sustainable binders and coatings that effectively addressed market demands, ensuring alignment with environmental objectives while also meeting industry needs.

SUSBINCO brought together a wide consortium of operators in the field of sustainable materials. The consortium consisted of six industry core partners with their own parallel project, and seven research institutes responsible for high-level scientific research. In addition, five collaborative partners contributed to the project implementation.

Industry core partners

- CH-Polymers
- Metsä Board
- Mirka
- Montinutra
- Teknos
- UPM-Kymmene

Research institutes

- Lappeenranta-Lahti University of Technology (LUT University)
- Natural Resources Institute of Finland (Luke)
- Tampere University (TAU)
- University of Eastern Finland (UEF)
- University of Oulu (UO)
- VTT Technical Research Center of Finland (VTT)
- Åbo Akademi University (ÅAU)

Collaborative partners

- Brightplus
- CH-Bioforce
- Kiilto
- MetGen
- Valmet Technologies

Project management

The project management of SUSBINCO was based on a steering group of the overall project, a group of work package leaders (WP), individual WP groups and a project coordinator. The steering group consisted of the project partners' representatives. Its main function was to oversee the overall management of the project.

The research activities were organised around five substance work packages which were supported by WP0 for management and dissemination: WP1 Production of raw materials for coating experiment, WP2. Controlled preparation of aqueous dispersions, WP3. Introduction and progress on lignin-based coatings, WP4. Application and testing, and WP5. Environmental impacts, end-of-life, and safety matters.

WP1 focused on preparing raw materials and developing bio-based surfactants for the purpose of dispersant, binders, and resins for coatings.

In WP2, the focus was on formulating and analysing the aqueous bio-based coating dispersions by bringing together the expertise of research organisations and companies.

WP3, introduction and progress on lignin-based coatings – focused on developing innovative crosslinking methods without any toxic chemicals for the lignin-based resins used primarily in coating materials.

WP4 provided valuable information on testing and optimising the functionality of the novel bio-based dispersions in the coating and converting processes.

WP5 focused on testing and evaluating environmental impacts, end-of-life and safety aspects of new materials developed in the project for sustainable binders and coatings as well as final products.

The scientific director of the project was Professor Chunlin Xu from Åbo Akademi University.

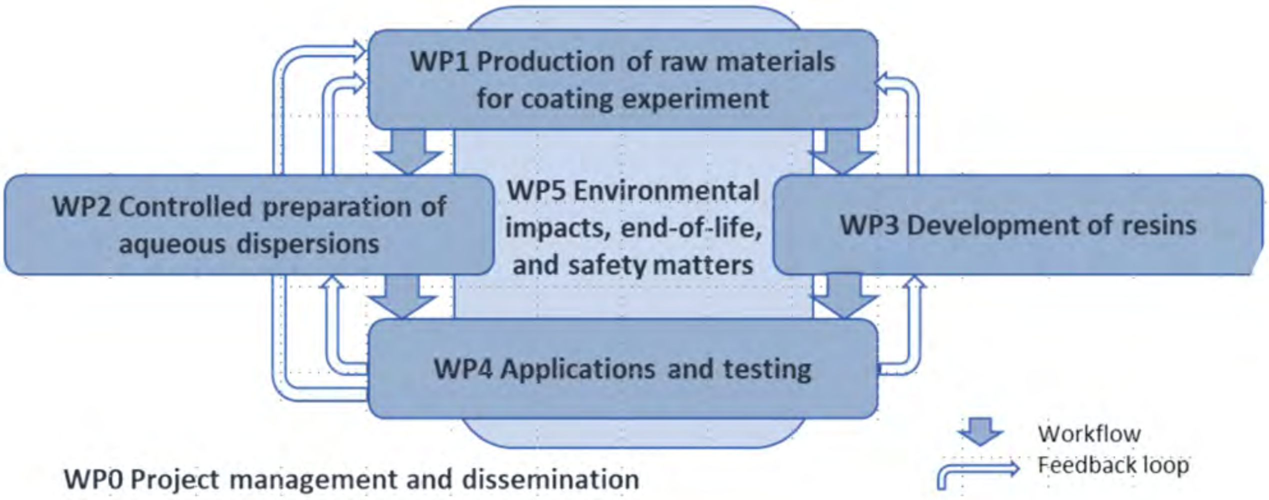
The chairperson of the steering group was Christiane Laine, UPM.

The five work packages were managed by WP leaders from research institutes. The companies took an active part in the activities of the WP groups and in guiding the project. The collaboration between the companies and researchers was very close and successful.

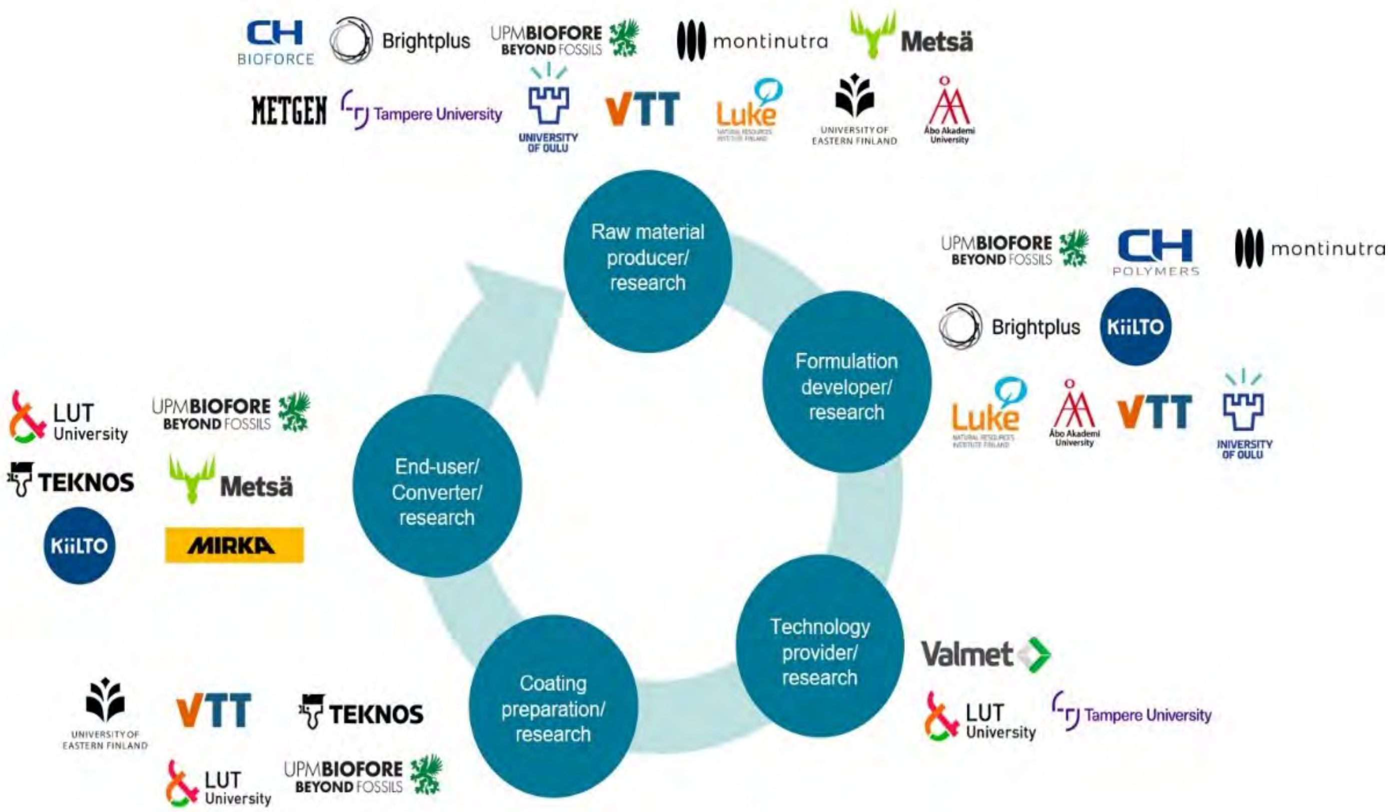
The project coordination was contracted from CLIC Innovation Oy, and the coordination activities were led by Aila Maijanen, Head of Bioeconomy, CLIC Innovation.

SUSBINCO steering group	
Jarkko Leivo	Brightplus
Lari Vähäsalo	CH-Bioforce
Misla Lagus	CH-Polymers
Soilikki Kotanen	Kiilto
Matti Heikkilä	MetGen
Terhi Saari	Metsä Group
Petter Andersson	Mirka
Jaakko Pajunen	Montinutra
Marjaana Mussalo	Teknos
Christiane Laine	UPM
Olli Tuovinen	Valmet Technologies
Pekka Saranpää	National Resources Institute Finland
Ville Leminen	LUT University
Henrikki Liimatainen	University of Oulu
Tomas Björkqvist	Tampere University
Reijo Lappalainen	University of Eastern Finland
Katariina Torvinen	VTT Technical Research Centre of Finland
Chunlin Xu	Åbo Akademi University

Work contents



Value network



International collaboration

Åbo Akademi

International collaboration was initiated with the Max Planck Institute of Colloids and Interfaces. Dr. Daniel Dax, a group leader in the Department of Sustainable and Bio-inspired Materials, made two visits to Åbo Akademi, in spring and autumn 2022. Dax provided crucial expertise in optimising the esterification process required for synthesis. The collaboration continued through Minette's research mobility, consisting of two visits to characterise the bio-based surfactants, one in spring 2023 and the other in the autumn of 2023. There, Dr. Svitlana Filonenko, an expert in colloidal chemistry and a group leader at the Max Planck Institute, functioned as a host, and has been a vital collaborative partner throughout the past year.

University of Oulu

The University of Oulu started international collaboration with the Cellulose & Wood Materials research group in Empa, Switzerland. The group is headed by Dr. Gustav Nyström and focuses on cellulose and wood as a basis for new smart materials with embedded functionalities. The hybrid biomaterials created from suberin and cellulose nanofibres were analysed in collaboration with Nyström's group, and doctoral researcher Umair Qasim joined the group for three months during spring 2024.

Tampere University

International collaboration was started to address the challenge of achieving an industrially utilisable assessment of microfibrillated cellulose quality, initially as off-line laboratory analyses but also assisting in reaching the final goal of online analyses in production lines. The international collaborator, in addition to SUSBINCO partners ÅA and UO, is the High Yield Pulping Technology group of Professor Per Engstrand at FSCN Research Centre in Mid Sweden University, Sundsvall, Sweden. One area of expertise of the group is mechanical attrition of biomaterials, including microfibrillation and fibrillation-level analysis. In particular, the use of optical measurement branded PulpEye/CrillEye for MFC quality analysis gives possibilities that are lacking nationally.

LUT University

LUT has collaborated with the Norwegian food research institute in Nofima, Norway in the field of LCA of the bio-based coatings in packaging applications. Regarding the converting and effect of converting on the properties of dispersion coatings, researchers from Politecnico Di Milano Polimi, Italy joined the research group for three and a half months during the project period. The research visit was hosted by Dr. Leminen's group.

University of Eastern Finland (UEF)

UEF has collaborated with Karlstad University in Sweden on comprehensive valorisation of birch, focusing in particular on the use of thermochemical techniques to produce biochar/hydrochar and liquid distillates. In SUSBINCO, the focus was on purifying and using phenols for paper coatings. In the Academy of Finland WoodPro joint project, on the other hand, other sustainable utilisation schemes were studied with Professors Ali Mohammadi, Karin Granström and Maria Sandberg.

VTT Technical Research Centre of Finland (VTT)

International collaboration was established with Tecnalia, a research and technology development centre in Spain, which hosted VTT. The research visit was initiated with Dr. Aitor Barrio, a materials researcher at Tecnalia. Dr. Barrio and his colleagues have expertise in polyurethane development from bio-based materials. Research scientist Dr. Miriam Kellock from VTT visited Tecnalia for two weeks in Spring 2023 to collaborate on the development of lignin-based polyurethanes. She then continued the development work at VTT.

Work Package 1

Production of raw materials for coating experiment

Summary

The consortium aims to enable the use of up to 100% bio-based content in developed binders and coatings. That would require innovations on all coating components, among them barrier components and dispersants. Moreover, sustainability is crucial in the development. The use of waste was therefore the starting point for developing all bio-based raw materials. This includes the use of bark to extract suberin and to produce phenolic fractions from its reside, the use of sawdust to produce hemicelluloses using hot-water fractionation and nanocellulose derivatives using sustainable approaches, and the use of the resulting waste stream, i.e. lignin. More importantly, understanding of the basics behind phenomena is essential, and thus we had a strong focus on sharing know-how and infrastructure from the beginning.

The aim of WP1 was to prepare raw materials and develop bio-based surfactants for dispersant, binders, and resins for coatings. WP1 targeted to promote the transition of nanocellulose, hemicellulose, lignin, and extractives to bio-based components of binders and dispersants in aqueous dispersion coating applications. The more specific WP1 objectives were:

1) to produce suberin from bark on a pilot scale for formulation and coating trials in WP2 and WP4;
2) to investigate the proof-of-concept for phenolic fractions from slow pyrolysis and hydrothermal liquefaction (HTL) distillates and test them for dispersion coatings;
3) to develop nanocellulose and hemicellulose surfactants with tailored hydrophilic-lipophilic balance (HLB) as dispersants for aqueous dispersion formulation in WP2 and coating in WP4;
4) to thoroughly understand abrasive nanocellulose fibrillation in ultra-thin plate gaps in order to enable the use of nanocelluloses as part of the binder formulations;
5) to investigate the proof-of-concept for the synthesis of bio-based latex using hemicellulose-based surfactants as a template in emulsion polymerisation;
6) to prepare lignin samples and characterise them for the product development in WP3; and
7) to develop characterisation methods and share the analytical tools within the consortium, supporting the need of understanding of each coating component and evaluating their application performance.

As the outcome of WP1 in close interaction with WP2, several highlights can be mentioned here. Suberin production was successfully upscaled from the lab scale to the pilot scale. There were two trials, and over 10 kg suberin was produced and tested by all the relevant partners. Amphiphilic nanocellulose derivatives were produced with sustainable approaches, and they were well characterised before sending them to WP2 for formulation testing and to WP4 for coating trials. Fatty acid-grafted hemicellulose surfactants were synthesised and well characterised with interactional collaboration with the Max Planck Institute. Such hemicellulose-based surfactants were tested in formulation in WP2 and further in coating trials in WP4, and were also sent to WP5 to support LCA, toxicity and composability testing. Novel phenolic fractions were produced and primarily tested with their surface property. Abrasive fibrillation trials were conducted to allow the development of fibrillation techniques. Lignin samples were received from several industrial partners and well characterised before sending to WP3. The synergy for forming a strong consortium was boosted, and all partners have contributed by sharing analytical tools and infrastructure.

Contact person

Chunlin Xu
Åbo Akademi
chunlin.xu@abo.fi

1.1 Raw material and bio-based building blocks for dispersion coating

Description

Suberin: Simultaneous extraction of suberin fatty acids, i.e. suberin hydrolysate and betulin was optimised on a laboratory scale at Luke. For small-scale extraction with accelerated solvent extraction (ASE) unit and a two-litre mixing reactor, the outer bark was separated manually from the inner bark. The yield was thus higher than in piloting. Since no pure bark was available in the market, birch bark was crushed and sieved to obtain an outer bark-rich fraction for the larger-scale extractions at VTT Technical Research Centre of Finland. Suberin hydrolysate was obtained with alcoholysis, including multiple steps, among them evaporation, filtration and acidification.

Phenolic fractions, University of Eastern Finland: For the planned coating experiments, a set of distillates were produced by slow pyrolysis and hydrothermal liquefaction (HTL) and screened for their overall composition and their phenolic content with NMR spectroscopy. Industrial slow pyrolysis liquids were also examined.

Centrifugal partition chromatography (CPC) was chosen for the separation of phenols from the selected distillates due to its suitability for challenging samples such as pyrolysis and HTL liquids. Arizona (AZ) solvent system proved to be suitable for the phenol purification, typically with a purity of >70% and gram quantities in a single run. The techniques are scalable to industrial scale.

Åbo Akademi University's contribution involves two main tasks: the development of bio-based surfactants in the form of amphiphilic hemicellulose derivatives, and the investigation of high bio-content latexes using hemicellulose-based surfactants as a template in emulsion polymerisation for barrier-coating applications. The central focus of these efforts was on ensuring sustainability and environmental safety in material development. Hemicelluloses, sourced as a side stream from the pulp industry, were specifically used, with a particular emphasis on galactoglucomannans found in softwoods. The overarching goal was to ensure a production process that is suitable for pilot-scale applications, and thus close collaboration with sustainability evaluation at WP5 was important.

The University of Oulu group focused on developing green dispersing agents based on functionalised nanocelluloses. The main aim was to produce new amphiphilic nanocelluloses to enable stable and processable aqueous coating formulations in combination with suberin. Furthermore, nanocellulose can have a dual role in the formulations, i.e. they can also improve the features of the final coating (mechanical strength, barrier properties, recyclability, biodegradability, etc.), and their adhesion and compatibility with other bio-constituents was addressed.

The Tampere University group addressed the challenge of producing sustainable microfibrillated cellulose (MFC) as a component for binders and coatings in a purely mechanical process that can be scaled to industrial production. In addition to the scale-up challenge, the problematic factors of MFC production are conventionally either high energy consumption or high chemical load and costs with pre-treatments. The work included fibrillation pre-trials where tool surfaces were developed with industry partners for a refiner-like process to improve energy efficiency. These trials assisted fibrillation trials of never-dried pulp and the same pulp stored as pulp sheets to investigate the benefit of avoiding hornification during pulp drying.

Results

The chemical composition of the outer bark differs considerably from the inner bark. Birch outer bark contains only small amounts of carbohydrates and lignin (Fig. 1). The suberin content is 51.4% (DW) and the amount of extractives, which included mainly triterpenoids such as betulin, is also large, at approximately 31.8% (DW) (Fig. 2). Birch inner bark does not contain suberin, and hence the carbohydrate content is much higher than that of outer bark (Fig. 1).

Since the industrial birch bark contains both outer bark and inner bark, and also a small amount of wood (xylem), the extractive and suberin content is much lower than with pure outer bark, which was separated manually for the lab-scale extraction. Additionally, the carbohydrate and lignin contents are considerably higher. It is therefore important to separate outer and inner bark in order to improve the suberin yield. Outer and inner bark are also structurally different, and so separation is possible with simple mechanical methods (air, water, crushing).

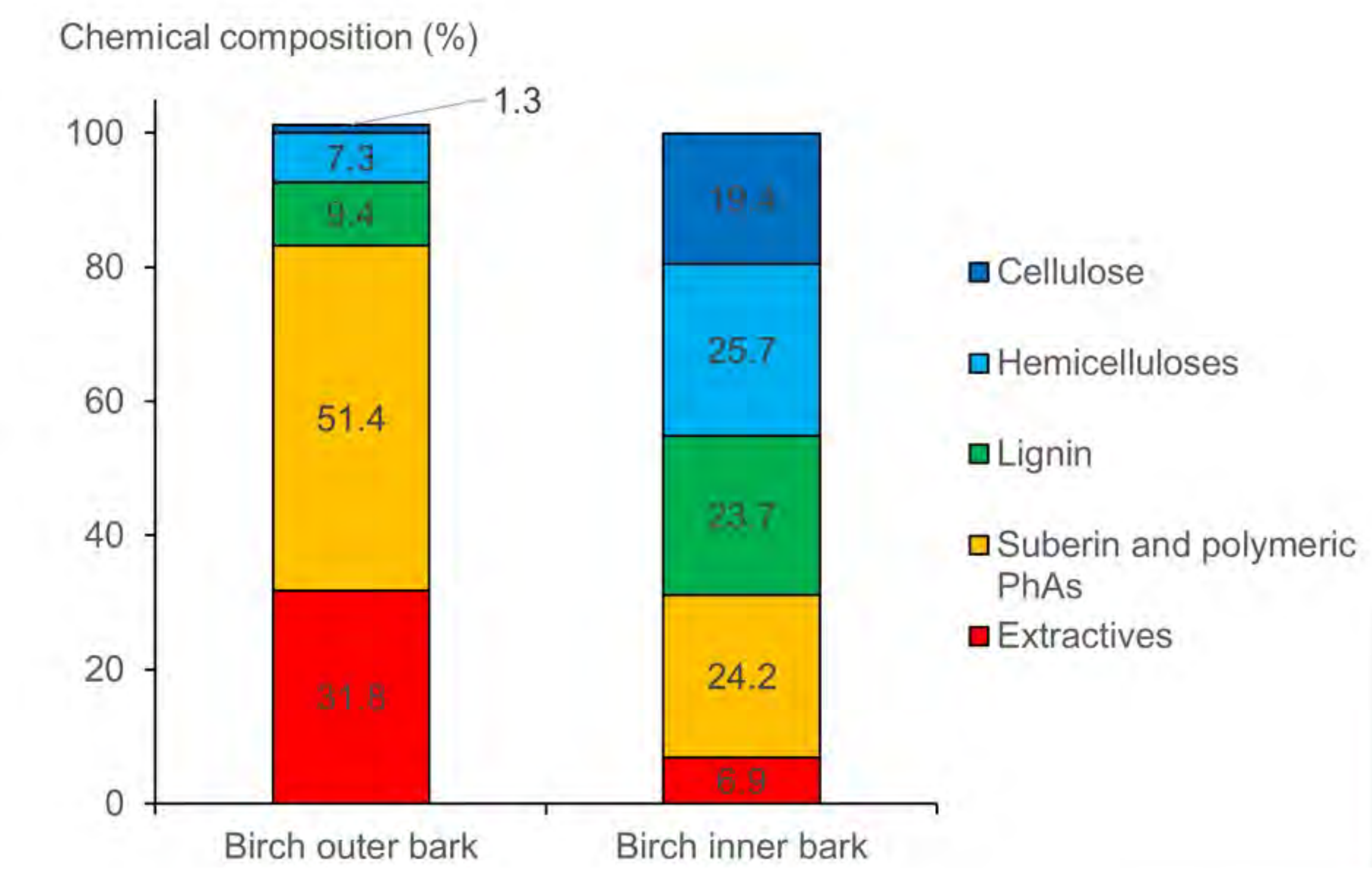


Figure 1 shows the difference in chemical composition between inner and outer bark.

Suberin fractions were isolated from birch outer bark using a lab-scale and pilot-scale ethanol/water extraction and subsequent alkaline ethanol/water hydrolysis. The ethanol/water extraction enabled the removal of betulinol, and the resulting extracts allowed the hydrolysis to isolate the suberin fractions (Fig. 3). After the isolation process, the obtained suberin fractions formed a brown solid cake. The 1800 L pilot batch yielded over 10 kg of suberin product (Fig. 4).

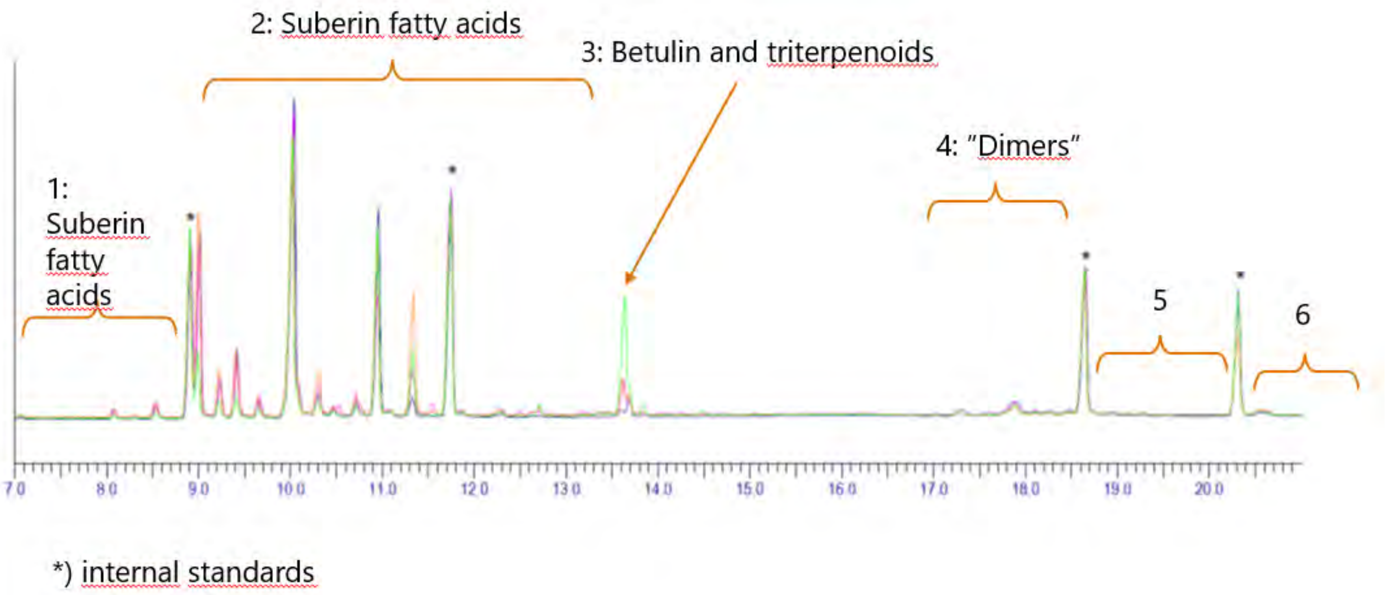


Figure 2. Gas chromatogram showing the major components of the suberin hydrolysate obtained from an outer bark fraction. The internal standards used for quantification are marked with an asterisk (*). Suberin fatty acids eluted first (Section 1 and 2), then betulin and trirepenoids. "Dimers" (section 4) include glycrol backbone, if any, and the trace amounts of steryl esters (section 5) and triacylglycerols (section 6) may originate from the xylem fraction.

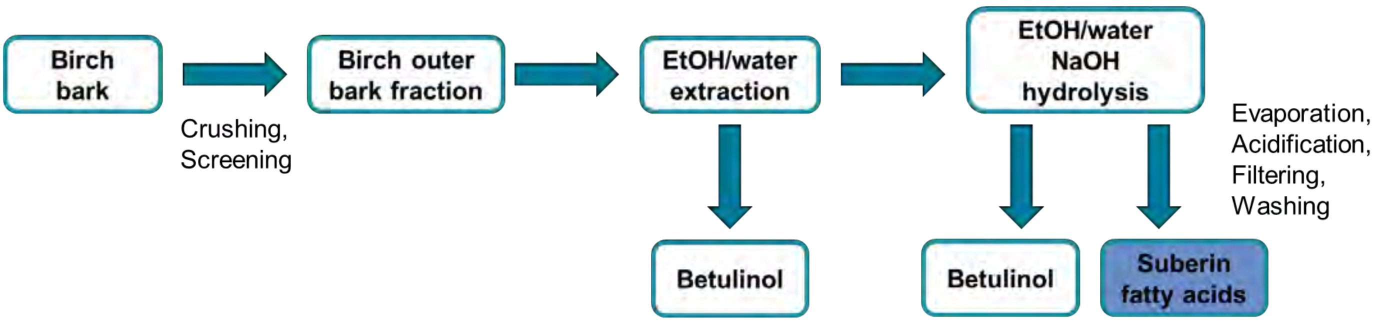


Figure 3. The simplified extraction process. The alkaline hydrolysis of the extracted bark and the subsequent betulin separation and suberin fatty acid precipitation was carried out by a slightly modified procedure (Korpinen et al. 2019).

Figure 4. Laboratory-scale extraction with ASE and the scaling up of suberin production at VTT.

Compound mg/g	Combined laboratory batch Luke	Laboratory batch Luke 2 L	Semipilot batch VTT 130 L	Pilot batch crude VTT 1800 L	Pilot batch washed VTT 1800 L	Pilot batch VTT 1800 L
18-hydroxy-18:1 acid	31.6	85.7	103.7	121.3	126.6	72.8
9,10-epoxy-18-hydroxy-18:0 acid	186.4	179.4	149.0	267.2	254.1	107.4
9,10,18-trihydroxy-18:0 acid	100.1	63.5	91.6	167.5	155.0	145.2
Betulinol	64.8	8.0	18.2	79.1	85.2	110.3

Table 1. The major fatty acid composition of suberin hydrolysate in different-scale extractions.

Major fatty acids of the hydrolysate are shown in Table 1. The 9,10-epoxy-18-hydroxy-18:0 acid dominates in almost all of the extracts except the second large-scale extraction (1800 l). The variation in fatty acid composition is natural, and due to the complex structure of suberin it is difficult to assure a certain composition or quality. However, the fatty acid composition does not have a major impact on the final product quality.

Publications

Hu, L., Koppolu, R., Hämäläinen, R., Kanerva, H., Nick, T., Toivakka, M., Korpinen, R., Saranpää, P., Qasim, U., Liimatainen, H., Xu, C., Anghelescu-Hakala, A. Suberin-based aqueous dispersions for barrier packaging applications. ACS Sustainable Chemistry & Engineering. https://doi.org/10.1021/acssuschemeng.4c02244.

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Korpinen R.I., Kilpeläinen P., Sarjala T., Nurmi M., Saloranta P., Holmbom T., Koivula H., Mikkonen K.S., Willför, S., Saranpää P.T. The Hydrophobicity of Lignocellulosic Fiber Network Can Be Enhanced with Suberin Fatty Acids. Molecules 2019, 24, 4391. https:// doi.org/10.3390/molecules24234391.

Contact person

Risto Korpinen
Natural Resources Institute Finland (Luke)
risto.korpinen@luke.fi

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BENEFITS FOR PARTICIPATING COMPANIES

Teknos Oy

Teknos finds new expert network and partnerships valuable for future product development
Teknos Oy has been participating in many WPs in SUSBINCO project, which has provided insights for material research, material synthesis, application related processes and testing of coatings. Benefits for the company R&D have been learning new testing methods, seeing how the pilot coating process works, understanding different aspects of polymer synthesis raw materials and process and exploring different bio-based raw material sources and material characteristics. This will facilitate development work in the future, as experience from materials and their processability can be benchmarked to materials studied in SUSBINCO.

The research network of institutes and companies has improved Teknos' understanding of relevant and skilful operators in the bio-based research field. These contacts will benefit in future research programmes, but also in finding the right players to complete different value chains.
Public communication related to the SUSBINCO project has resulted in new contacts with Teknos on a global level. The topic of sustainable binders and coatings is highly relevant, and attracts plentiful interest within Teknos' value chain. Ways to source new materials and utilise developed solutions in different applications have created new contacts for Teknos.
Results achieved during the project have given insight into both the potential of new raw materials and the challenges related to availability, processing and achievable properties for the new materials. Project results are helping Teknos to plan and deliver future coating solutions with a realistic timeframe as the results help to evaluate materials that are at different readiness levels. Teknos has been able to create formulation platforms containing increased bio-based contents, which gives an opportunity to develop and commercialise a range of new coating products for different applications.

Pasi Virtanen, SUSBINCO project manager

Two birch-based slow pyrolysis liquids were selected based on their composition and availability. The process is displayed in Figure x. Distribution tests were carried out to find the optimal solvent systems (AZ H, L and N) for the CPC runs. The phenolic yield per run was approx. 30 wt%, with an overall purity of approx. 70%. The sample miscibility appeared to be a limiting factor. Based on the NMR analysis, the fractions were typically mixtures of 2 to 4 methoxyphenols, but single-compound fractions (purity >90%) were also obtained. This is a good achievement, with only a single purification step. The phenols were produced in at a scale of approx. 10 grams.

Plans for the industrial scale-up were made, but the runs would have required excessive financial resources. However, based on the performed experiments the scale-up could be straightforward, and can be resourced in future projects. A PhD thesis article based on this study is being written.

Contact person

Noora Jokinen
University of Eastern Finland (UEF)
noora.jokinen@uef.fi

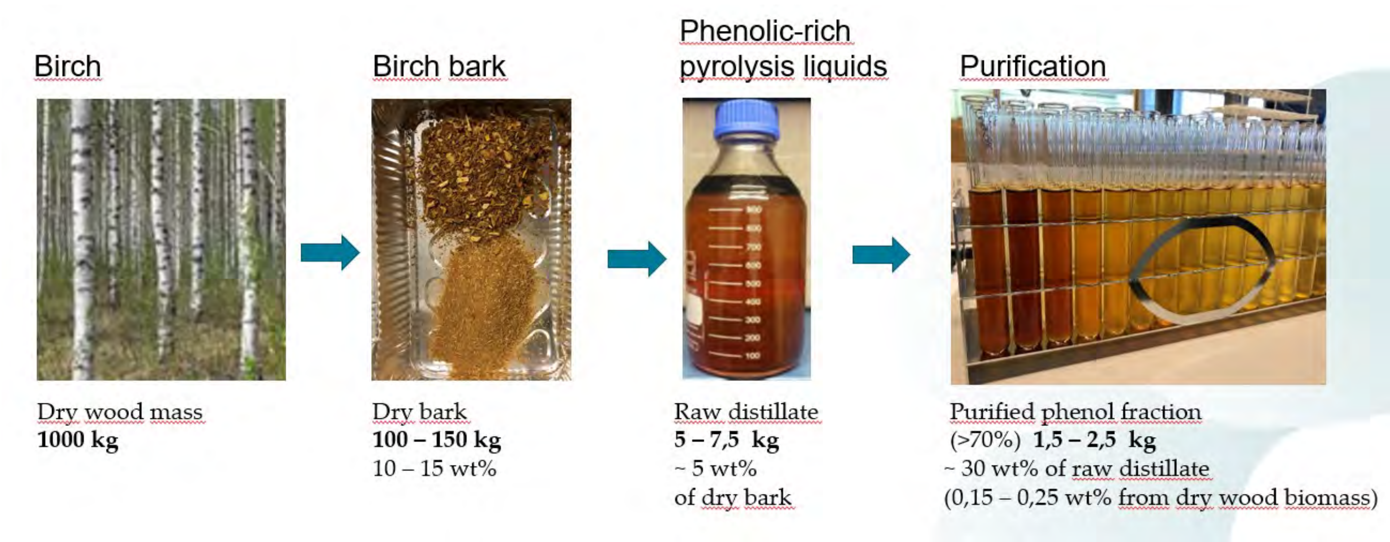

Figure 5. A schematic description of the process from feedstock to phenol extraction from woody biomass.

The bio-based surfactant is produced by functionalising the GGM. This is achieved through esterification by grafting on naturally occurring fatty acids on the GGM chain, creating an amphiphilic GGM derivative. Low and high substitution degrees, as well as three different fatty acid chain lengths, were employed to modify the physicochemical properties of the surfactant. The surfactants were tested as a stabiliser for oil-in-water emulsions, keeping the emulsions stable for up to 6 weeks. Furthermore, the surfactants have also successfully worked as dispersants in suberin coatings. A manuscript based on this result is almost ready for submission.

Bio-latex incorporating 50% bio-based materials was developed, substituting the conventional hazardous surfactant sodium dodecyl sulphate (SDS) with galactoglucomannan-galactoglucomannans-based biosurfactant. The latex particles are less than 100 nm in size and are well stabilised by steric and ionic mechanisms, resulting in free-standing films with good barrier properties. The paper-based board coating trials are in the implementation stage and are expected to form the basis of the second manuscript.

Publications

Kvikant, M., Filonenko, S., Dax, D., Toivakka, M., Xu, C. (2024). Amphiphilic Hemicellulose Derivatives as Stabilizers in Oil-in- Water Emulsions. Manuscript to be submitted.

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Dax, D., Eklund, P.,Hemming, J., Sarfraz, J., Backman, P., Xu, C., Willför, S. Amphiphilic Spruce Galactoglucomannan Derivatives Based on Naturally-Occurring Fatty Acids. Bioresources 2013, 8(3), 3771–3790.

Yong, Q., Xu, J., Wang, L., Tirri, T., Gao, H., Liao, Y., Toivakka, M., & Xu, C. (2022). Synthesis of galactoglucomannan-based latex via emulsion polymerization. Carbohydrate Polymers 291, Article 119565. https://doi.org/10.1016/j.carbpol.2022.119565.

Contact person

Chunlin Xu
Åbo Akademi University
chunlin.xu@abo.fi

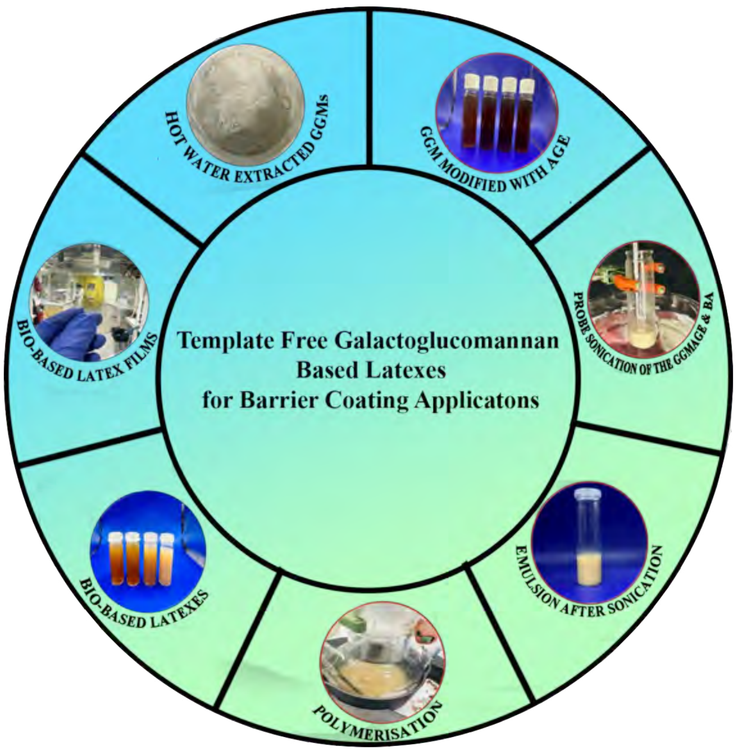

Figure 6. A schematic description of the process from feedstock to phenol extraction from woody biomass.

Cellulose nanofibres with amphiphilic characteristics were developed as green dispersing agents. A dual-functioning deep eutectic solvent system based on triethylmethylammonium chloride and imidazole was harnessed as a swelling agent and a reaction media for the esterification of cellulose with n-octyl succinic anhydride (OSA) (Fig. 7). The modified amphiphilic cellulose was further converted into nanofibres (ACNFs) with improved surface activity due to the balance of hydrophobic and hydrophilic characteristics. The amphiphilic nature of ACNFs promoted their performance as stabilisers in oil-in-water Pickering emulsions, and self-standing films of ACNFs showed high contact angles for all the tested ACNF variants.

Publications

Qasim U., Suopajärvi T., Sirviö J.A., Backman O., Xu, C. & Liimatainen, H. (2023). Pickering Emulsions and Hydrophobized Films of Amphiphilic Cellulose Nanofibers Synthesized Using Ring-Opening Esterification in Deep Eutectic Solvent. Biomacromolecules 2023, 24, 9, 4113–4122. https://pubs.acs.org/doi/10.1021/acs.biomac.3c00472.

Contact person

Henrikki Liimatainen
University of Oulu
henrikki.liimatainen@oulu.fi

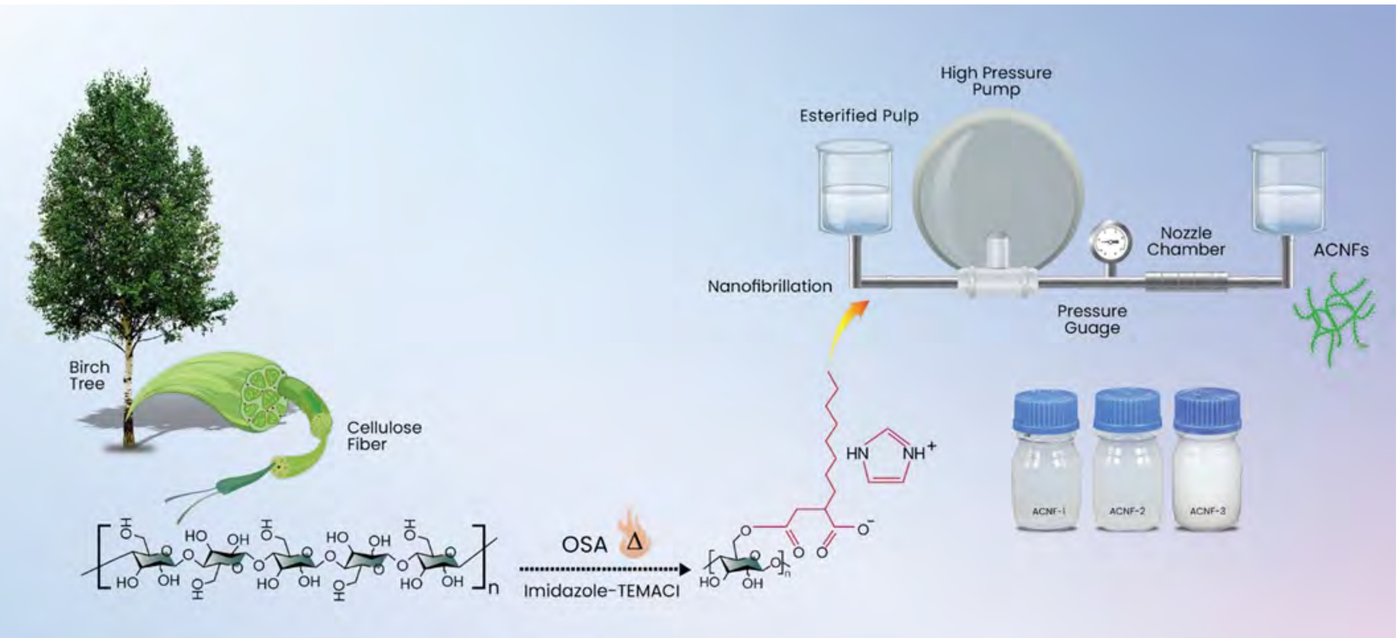

Figure 7. Amphiphilic nanofibers synthesised in deep eutectic solvent. (Ref: Qasim et al., Biomacromolecules)

The pre-trials investigated the fibrillation potential of fibrillation tool surfaces with various surface roughness when the six step cyclic fibrillation trials were otherwise constant. Figure 8 shows the influence of surface roughness (MC2, MC2RB and MC3) on fibrillation degree, here shown as viscosity development as a function of cumulative specific energy consumption (SEC). The steepest curve (NC151222/MC2RB) shows significant energy savings at relatively high fibrillation level compared to the least-developed fibrillation curve (NC131022/MC2RB).

Fibrillation trials of never-dried pulp and the same pulp stored as pulp sheets shows significantly different reaction to initial fibrillation steps, but finally converge. Viscosity as a function of SEC is presented in Figure 9. The benefit of avoiding hornification of pulp during drying is therefore substantial at relatively high fibrillation level, but negligible after the final fibrillation step. Strong bonds due to hornification is likely the reason to diverge initial fibrillation reactions during slightly differing fibrillation intensity at initial fibrillation steps.

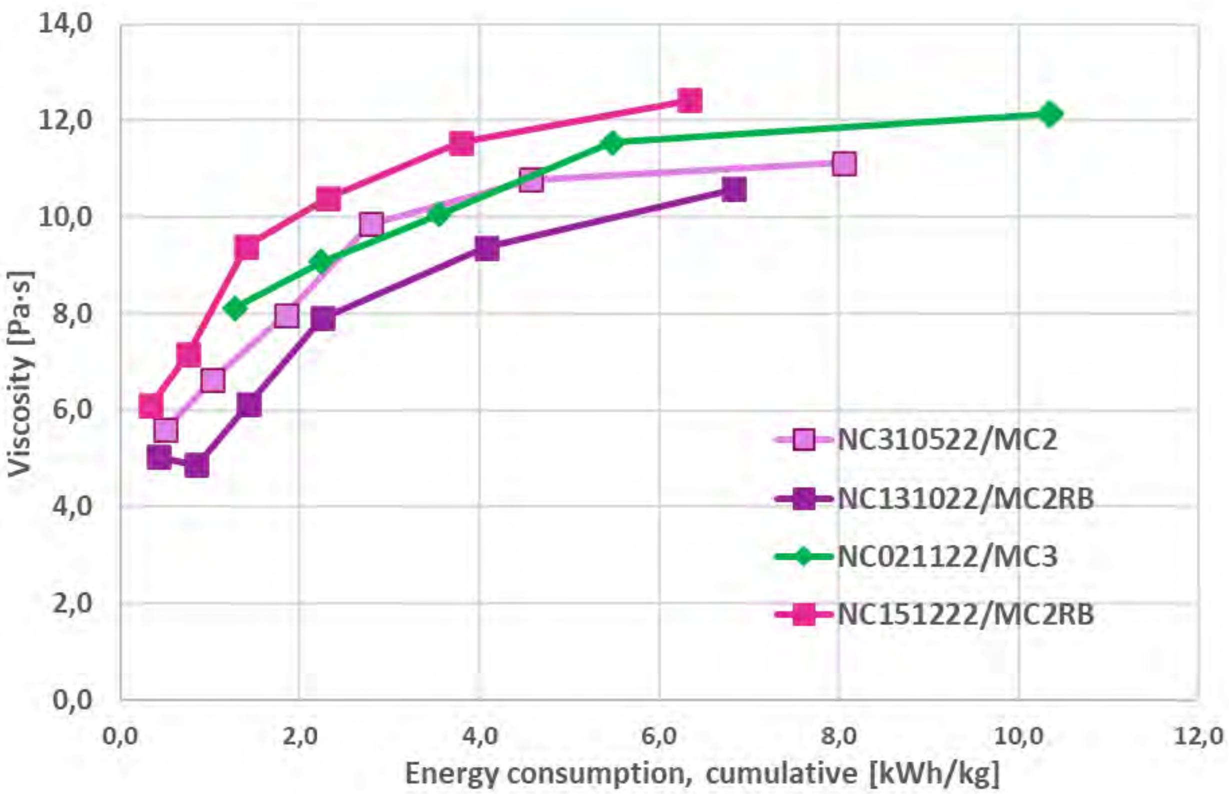

Figure 8. Viscosity of MFC as function of cumulative specific energy consumption for the pretests.

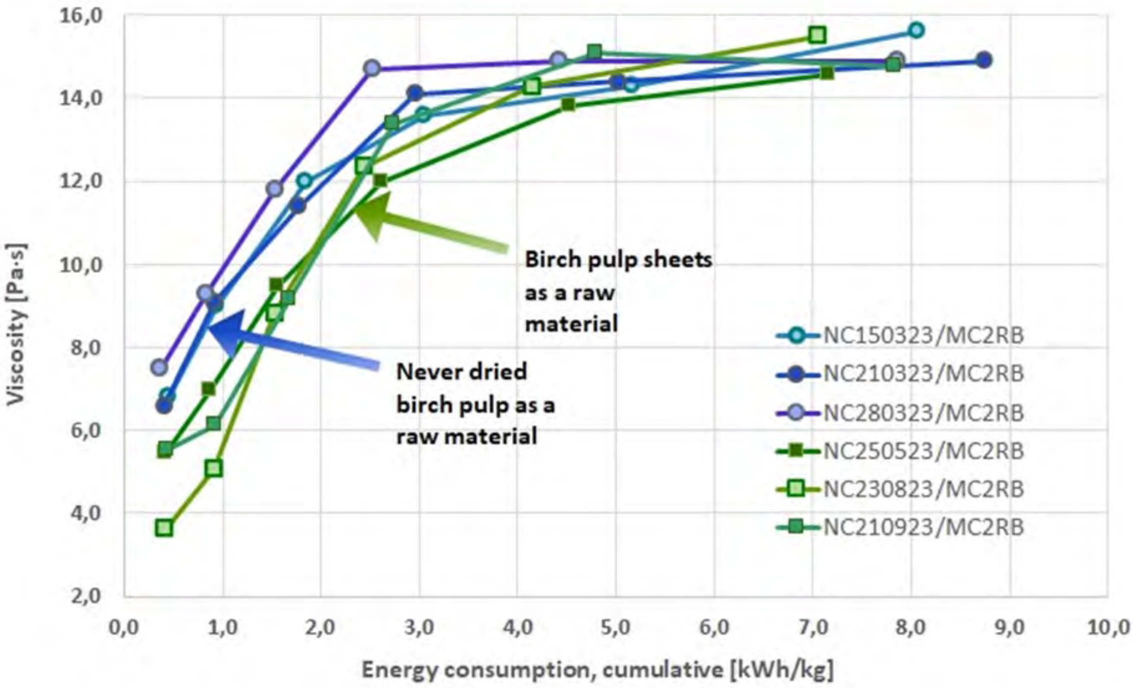

Figure 9. Viscosity as function of SEC for MFC fibrillated from newer dried pulp and the same pulp stored as pulp sheets.

Contact person

Tomas Björkqvist
Tampere University
tomas.bjorkqvist@tuni.fi

1.2. Characterisation of lignins

Description

Lignins used in WP3 coating work were characterised in terms of composition, functional group content and molar mass distribution, properties that affect the reactivity of lignins towards crosslinkers and the viscosity and solid content of coating formulations (WP3). For example, a high molar mass may result in a high viscosity that makes a coating formulation difficult to apply, while the desired (high) solid content of the formulation cannot be attained, because water needs to be added to reduce the viscosity.
To give examples of how functional groups affect reactivity, aliphatic hydroxyl groups are important for crosslinkers such as isocyanates in polyurethane coatings, while phenolic hydroxyls are required for other types of lignin crosslinkers such as the polyamine polyethyleneimine (PEI). A high lignin content is generally preferred, but aliphatic hydroxyls located on carbohydrate impurities can contribute positively to crosslinking and coating properties in polyurethane coatings. Other types of impurities (proteins, extractives and ash) should be minimal for all types of coatings.

Results

The lignins showed considerable differences with regard to their lignin contents that ranged from 78% to 97% (Fig. 10). The main non-lignin components (impurities) were the carbohydrates and proteins in Boreal Bioproducts' lignin, the ash in Metnin Ultra, and the proteins in wheat straw soda lignin. Part of these impurities (insoluble ash and proteins) may also show up as "lignin" in the gravimetric Klason lignin determination.

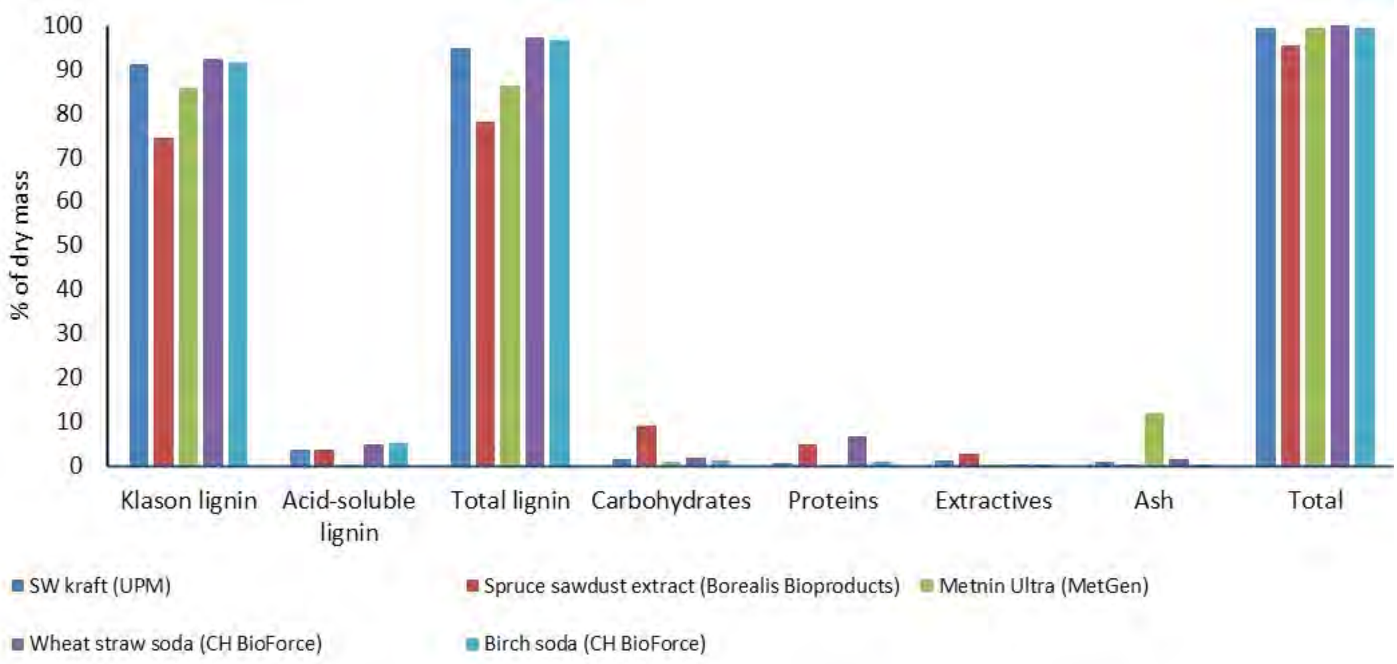

Figure 10. Chemical composition of lignins

In terms of functional groups (aliphatic and phenolic hydroxyls and carboxyls) determined by 31P NMR, there were clear differences between the lignins (Fig. 11). Boreal Bioproducts' lignin was notably different from all the others by virtue of its high aliphatic hydroxyl content, due to its high carbohydrate content. In general, the differences reflect the pulping or extraction processes used to obtain the lignins. The botanical origin of the lignins (softwood, hardwood or grass) explain the ratios of the different types of phenolic hydroxyls, namely guaiacyl, syringyl and p-hydroxyphenyl units.

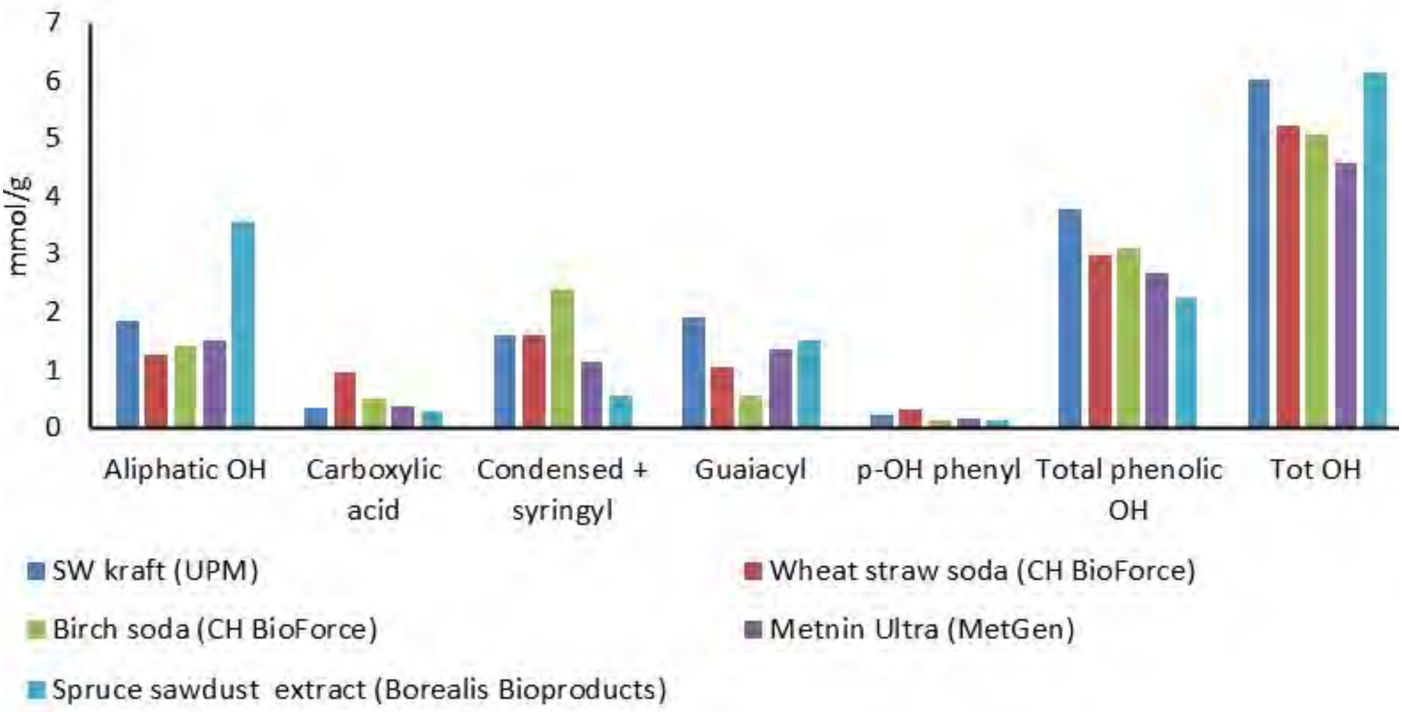

Figure 11. Functional group distribution of lignins by 31P NMR

The lignins were also characterised by 2D HSQC NMR to ascertain the types of linkages between the lignin phenylpropane units. The most abundant lignin interunit linkages were b-aryl ether (b-O-4), resinol (b-b), and phenyl coumaran (b-5) structures. The main linkage type of all samples (Boreal Bioproducts lignin was not analysed) was b-O-4. b-5 structures were observed only for SW kraft and Metnin lignins. For birch soda lignin, almost 45% of the linkages were of the b-b type. Methoxyl contents were similar (11-13%) except for the only HW lignin, birch soda (20.5%).

As for the molar mass distribution of the lignins determined by SEC (Fig. 12), the highest values were those of the hardwood lignins (birch soda and Metnin) with Mw values of 4,500-5,500 Da. By far the lowest value (1,300 Da) was seen for the Boreal Bioproducts sawdust extract. The explanation for this may be that rather than true polymeric lignin, which is not extractable with water only, it mainly comprises other types of low molar mass phenolic substances, such as stilbenes and lignans.

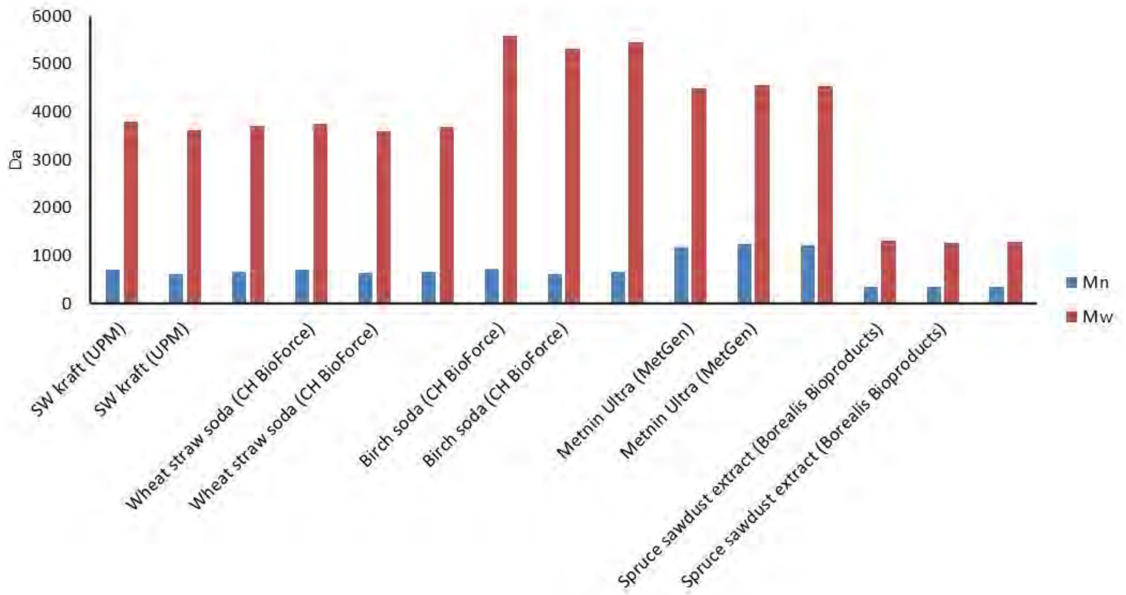

Figure 12. Molar mass distribution of lignins determined by SEC. Mw = weight-average molar mass; Mn=number-average molar mass

Contact person

Petri Widsten
VTT Technical Research Centre of Finland
petri.widsten@vtt.fi

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BENEFITS FOR PARTICIPATING COMPANIES

Boreal Bioproducts – Montinutra

Parallel project enabling pilot operations with public synergetic research on hemicellulose modifications

Boreal Bioproducts (Montinutra Ltd) had a parallel company project over the first two years of the SUSBINCO consortium. In the parallel company project, research pilot scale production operations were developed and commissioned, creating the cornerstone for our current operations and development. The research pilot technology is based on pressurised hot water extraction and solely utilises side stream wood and bark from saw milling. With the help of the research pilot operations, novel hemicellulose and lignin product samples were prepared for the SUSBINCO public research partners in kind.

SUSBINCO research contributed to crucial understanding of the characteristics and modifications of our pressurised hot water extracted hemicelluloses and lignins. Based on the project, the results seem to differ from those from other products that were sampled and tested. The project yielded promising results in the fields of modifying and using hemicelluloses for surfactants and as dispersants, for example, which have high industrial volume potential, and also increase the value of the utilised side streams. The research on co-polymerisation approaches to making bio-based emulsion polymers shows potential for further research and development, for example for industrial applications to substitute fossil based latexes.

Jaakko Pajunen, Managing director, Boreal Bioproducts

1.3 Characterisation of raw materials and method development

Description

The consortium partners started the project by sharing information on all available infrastructure and knowledge. We set up a document for saving and categorising the tools available for WP1 and WP2 by all the partners.

Some examples include:
- Composition analysis at both LUKE and ÅAU
- Fibre analysis tools at TAU, OU and ÅAU
- Coating infrastructure at ÅAU and VTT, serving other partners
- Analytical platform for physicochemical properties at ÅAU, available for all partners

Results

One highlight of the result was the determination of hydrophilic-lipophilic balance (HLB) of suberin conducted at ÅAU. Experimental methods were developed to determine the HLB of suberin, formulating emulsions using hemicellulose derivatives as surfactants and evaluating the stability of the emulsions. Emulsions stabilised with the model compounds Spans and Tweens exhibited strong colloidal stability, while emulsions stabilised with galactoglucomannan-grafted-fatty acids (GGM-FA), a natural surfactant, displayed its potential by producing stable emulsions. The study showed promising results on the stability of suberin emulsions and the potential application of hemicellulose derivatives as surfactants, providing valuable insights for the packaging and coating industries.

Several collaborative tasks arose during the project, demonstrating the synergy benefits of collaboration. The surface property of phenolic extractions from UEF was studied by contact angle at ÅAU, and showed certain level of hydrophobicity. Comparison of fibres via different processing approaches is under investigation in a joint effort by TAU and ÅAU.

Publications

Öhman, Thomas. (2023). Characterisation of the stability for suberin-in-water emulsions. Master's thesis, Åbo Akademi University, Turku, Finland.

Contact person

Chunlin Xu
Åbo Akademi University
chunlin.xu@abo.fi

Controlled preparation of aqueous dispersions

Summary

The aqueous dispersion formulations are an attractive option for developing new sustainable coatings, e.g. in paper and board products, and for replacing current laminated layers, which are based on non-renewable raw materials. Therefore, formulation and knowledge of the behaviour of aqueous dispersions is crucial for the adoption of new sustainable coatings by the industry. WP2 produced water dispersions for barrier coating applications using bio-based binder compounds (suberin and distillate extracts) and surfactants (commercial polyvinyl alcohol (PVOH), functionalised nanocelluloses and hemicellulose derivatives).

The primary aim was to develop technically feasible mixtures with a high content of biomaterials on a laboratory scale using the materials developed in WP1, and then scale up the production of the most feasible formulations. The specific objectives were: i) to develop aqueous dispersion formulations based on bio-based binders i.e. extraction compounds (WP1: suberin and pyrolysis fractions) and their mixtures with bio-dispersants (WP1: functionalised nanocelluloses and hemicelluloses derivatives); ii) to address the feasible preparation protocols for stable aqueous dispersions, iii) to analyse the properties of aqueous dispersion formulations, and iv) to scale up the most promising aqueous dispersions for the coating purposes (to be further harnessed in WP4 and WP5).

Various approaches based on thermomechanical or mechanical methods were used to form water dispersions from suberin. All the dispersing agents, i.e. commercial PVOH, functionalised nanocellulose and hemicellulose resulted in stable suberin dispersion, but the feasible dispersing method and the required dispersant dosage varied significantly. All the suberin dispersions had an average particle size below 20 µm and rheological properties feasible for coating purposes. Moreover, the behaviour of dispersions and their performance in coatings was notably affected by the conditions of the aqueous medium. Finally, the preparation of suberin dispersions stabilised with biodispersants and PVOH were successfully scaled up, and the formulations were used for pilot-scale barrier coating and converting experiments (WP4).

Contact person

Henrikki Liimatainen
University of Oulu
henrikki.liimatainen@oulu.fi

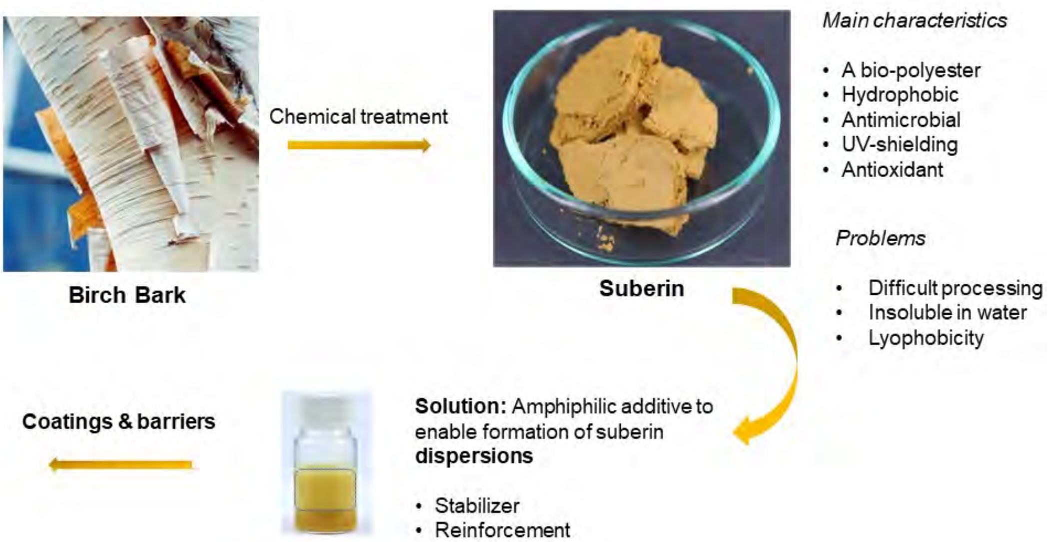

Figure 13. Preparation of aqueous suberin dispersions stabilised with commercial PVOH, functionalised galactoglucomannan (GGM) and amphiphilic nanocelluloses.

2.1. Controlled preparation of aqueous dispersions

Description

The Åbo Akademi University group's contributions are to formulate and evaluate water dispersions for coating applications using bio-based binder compounds, e.g., suberin, and synthesised surfactants obtained from WP1 in a lab scale. The preparation protocols of controlled water dispersions based on suberin only, suberin with commercial dispersant, and suberin with functionalised galactoglucomannan (GGM) was demonstrated through analysis and testing of coating formulations. The optimal coating formulation was applied to a coating test on both the laboratory scale and pilot scale process.

The University of Oulu group focused on developing preparation protocols for bio-based aqueous barrier coating dispersions based on suberin, producing stable dispersions using functionalised nanocellulose surfactants and analysing and testing the behaviour and properties of suberin dispersions stabilised with nanocelluloses.

The research at VTT focused on protocol development at laboratory scale for the preparation of aqueous dispersions produced from suberin using a thermomechanical method. The dispersions were stabilised by commercial stabilisers, including polyvinyl alcohol, and characterised to evaluate their suitability for barrier coating applications. The parameters for controlling the particle sizes of dispersions which affect the dispersions quality, and their stability were investigated. The optimised protocol was selected to scale up the process to the pilot scale.

Nanocellulose qualities consist of a broad range of fibrillated material, and industrially exploitable quality assessment analyses for this material group are limited. To analyse the properties of microfibrillated cellulose (MFC) as a component for aqueous dispersion, gel point analysis was exploited as a quality assessment method to compare with optical microscope and SEM imaging analyses and rheological measurements.

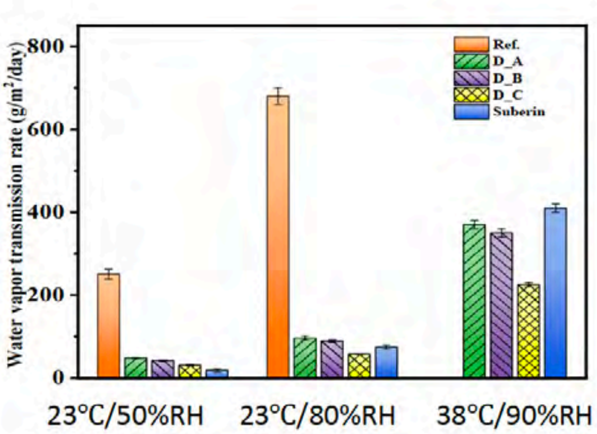

Figure 14. (left side) Overview of suberin isolation from birch bark and formation of aqueous suberin dispersions for barrier coatings for paper and paperboard substrates. (Right side) Water vapour transmission rate (WVTR) of coated paperboard samples in different coating conditions, 23 °C/50% RH, 23 °C/80% RH and 38 °C/90% RH. (D.A-D.C: Commercial barrier dispersion A, B and C).

Results

Water-based suberin dispersions were prepared in the presence of poly (vinyl alcohol) (PVOH) addition as the dispersing agent using high-shear post treatment (Figure 14, left side). The prepared suberin dispersion exhibited very high stability, homogenous particle size distribution and very high barrier behaviour (Figure 14, right side). In addition, water-based suberin dispersions were made by using modified hemicelluloses (galactoglucomannan, GGM) as the stabiliser. The most suitable modifications were determined by using static multiple light scattering technology to analyse the stability of the dispersions. The impact of pH adjustment during preparation was shown to be a crucial factor determining the extent of water sensitivity of the final suberin coatings.

Publications

Hu, L., Koppolu, R., Hämäläinen, R., Kanerva, H., Nick, T., Toivakka, M., Korpinen, R., Saranpää, P., Qasim, U., Liimatainen, H., Xu, C., Anghelescu-Hakala, A. Suberin-based aqueous dispersions for barrier packaging applications
ACS Sustainable Chemistry & Engineering.
https://doi.org/10.1021/acssuschemeng.4c02244

Contact person

Chunlin Xu
Åbo Akademi University
chunlin.xu@abo.fi

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BENEFITS FOR PARTICIPATING COMPANIES

Valmet Technologies Oy

Valmet is a leading global developer and supplier of process technologies, automation and services for the pulp, paper, and energy industries.

Valmet joined the SUSBINCO consortium because of the focus on the megatrends that are the most important for Valmet's business: a resource-efficient and clean world. The need for more efficient use of water, natural resources, and chemicals is increasing in all areas of industry, including pulp mills, which are key customers for Valmet. This requires new technologies and operational solutions, which can only be developed through collaboration between research institutes and companies.

The key topics for Valmet were:

Use of sustainable natural raw materials in suspensions and coating

- Suberin and hemicelluloses as a potential natural and sustainable paper/board coating and dispersing material
- New knowledge on recycling and eco-toxicity of suberin-containing fibre products.
- Initial information of scale-up of the suberin extraction process
- The role and quality demands of MFC in sustainable natural raw materials in suspensions and coating
- The capability and modelling of a new, energy-efficient method for MFC production by mechanical attrition
- New methods for MFC quality assessment
- Lignin as a potential raw material in corrosion coatings of metal surfaces

In addition to increasing the knowledge of these topics within Valmet, the SUSBINCO project has brought more skills and knowledge to research and business partners, which can initiate even more fruitful collaboration in the future.

Olli Tuovinen, Development Manager, Valmet Technologies Oy
Heli Kangas, Development Manager, Valmet Technologies Oy

The aqueous dispersions of suberin were stabilised with amphiphilic cellulose nanofibres (ACNF) for producing sustainable barrier coating formulations. The dual-functioning ACNF, synthesised in a deep eutectic solvent, functioned as an efficient suberin dispersant and reinforcing agent in the barrier coatings. The nanofibrillar percolation network of CNF provided steric hindrance against the coalescence of the suberin particles. The low CNF dosage of 0.5 wt.% resulted in a suberin dispersion with optimal viscosity (208.70 Pa.s), enhanced stability (instability index of <0.001), and reduced particle size (9.37 μm). The dispersion of suberin and CNF was further converted into self-standing films with superior UV-blocking capability, good thermal stability, improved hydrophobicity (increase in water contact angle from 61° ± 0.15 to 83° ± 5.11), and antioxidative and antimicrobial properties against gram-negative bacteria.

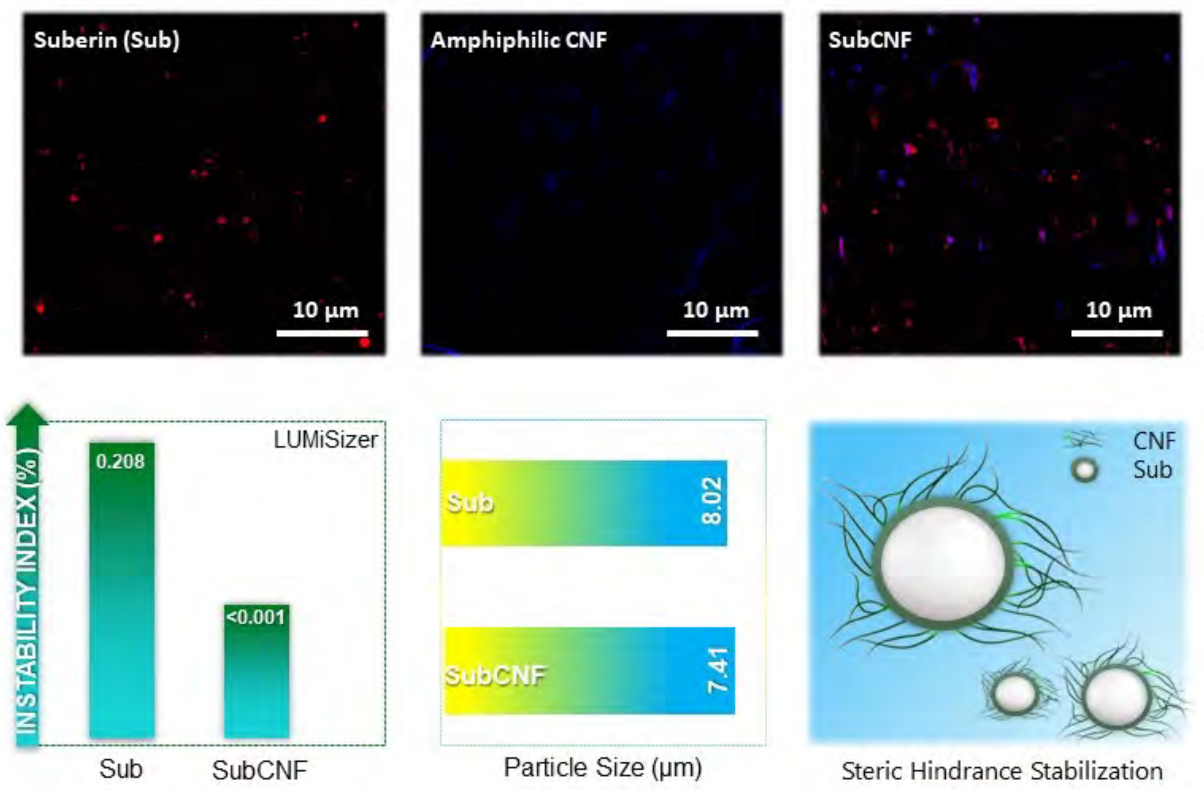


Figure 15. Aqueous suberin (Sub) dispersions stabilised with amphiphilic cellulose nanofibres (CNF). Ref: Qasim et al.

Publications

Qasim, U., Sirviö, J.A., Suopajärvi, T., Hu, L., Pratiwi, F.W., Hong Lin, M.K.T., Anghelescu-Hakala, A., Ronkainen, V-V., Xu, C. & Liimatainen, H. (2024) Multifunctional biogenic films and coatings from synergistic aqueous dispersion of wood-derived suberin and cellulose nanofibers. Carbohydrate Polymers. https://doi.org/10.1016/j.carbpol.2024.122218.

Contact person

Henrikki Liimatainen
University of Oulu
henrikki.liimatainen@oulu.fi

The protocol for preparation of stable aqueous suberin dispersion using the thermomechanical method was successfully established. The controlled preparation process included optimising parameters such as suberin melting temperature, dispersion preparation temperature, dispersion stabiliser dosage and its addition, and post-treatment alternatives. The optimised suberin aqueous dispersions were homogeneous, with particle size between 0.3 and 20 microns, and could be stored for months without any separation of the phases.

Publications

Hu, L., Koppolu, R., Hämäläinen, R., Kanerva, H., Nick, T., Toivakka, M., Korpinen, R., Saranpää, P., Qasim, U., Liimatainen, H., Xu, C., Anghelescu-Hakala, A. Suberin-based aqueous dispersions for barrier packaging applications. 10.1021/acssuschemeng.4c02244.

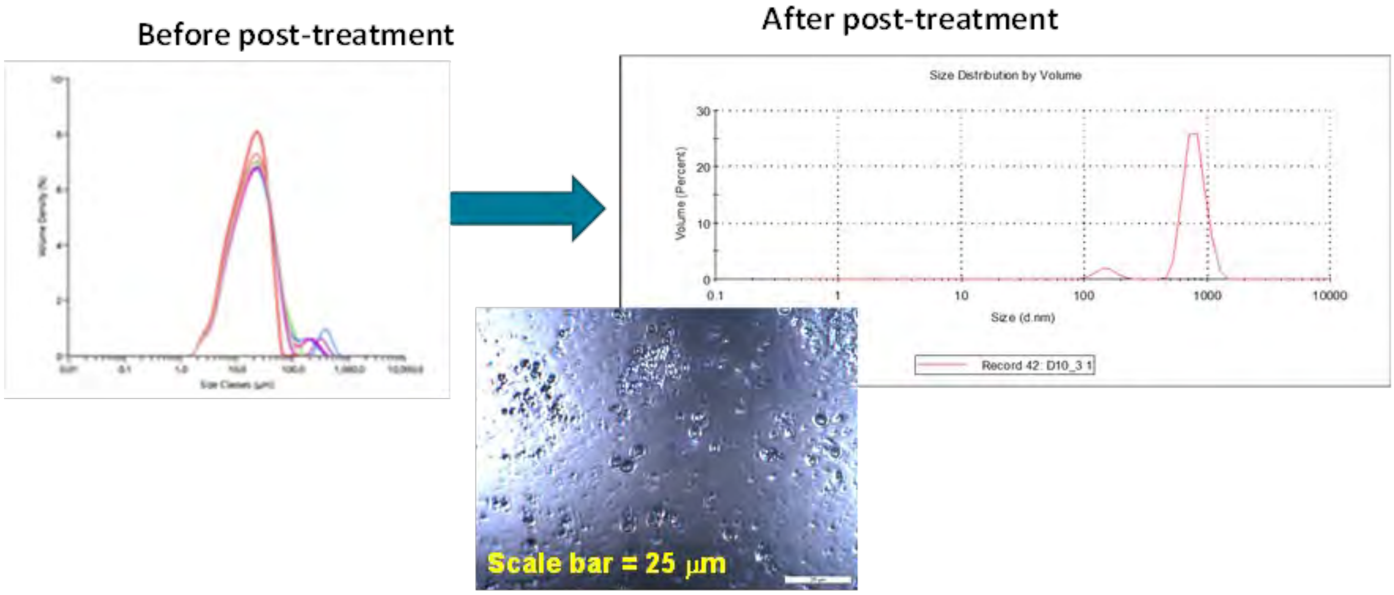


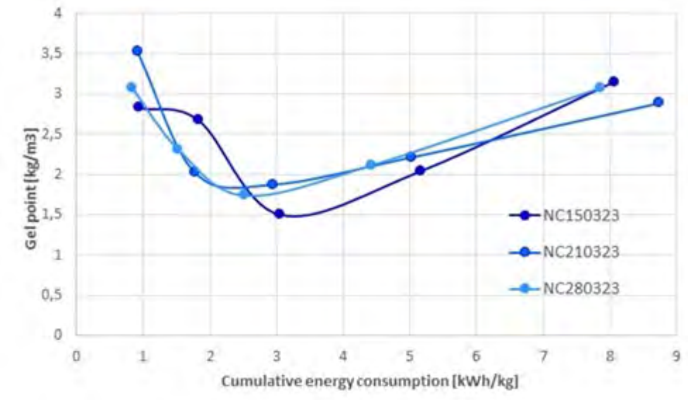
Figure 16. Effect of post-treatment for reduction of particle sizes of suberin dispersions.

Contact persons

Adina Anghelescu-Hakala
VTT Technical Research Centre of Finland
adina.anghelescu-hakala@vtt.fi

Roosa Hämäläinen
VTT Technical Research Centre of Finland
roosa.hamalainen@vtt.fi

Gel point is a crucial property when MFC is used as a surfactant or stabiliser for aqueous dispersions with bio-based binder compounds. Gel point measurement, based on sedimentation tests, is a sensitive analysis showing contrast between different MFC fibrillation level samples when e.g. viscosity measurement results appear unchanged. Figure 17 shows the results of the gel point analysis of three parallel fibrillations on both the never-dried pulp andthe same pulp stored as pulp sheets. Figure 18 shows examples of representative polarised optical microscope pictures of corresponding fibrillation levels shown as points in Figure 17. The lowest gel point value is achieved at modest fibrillation level before most of the fibre material is transferred to nano-scale material.



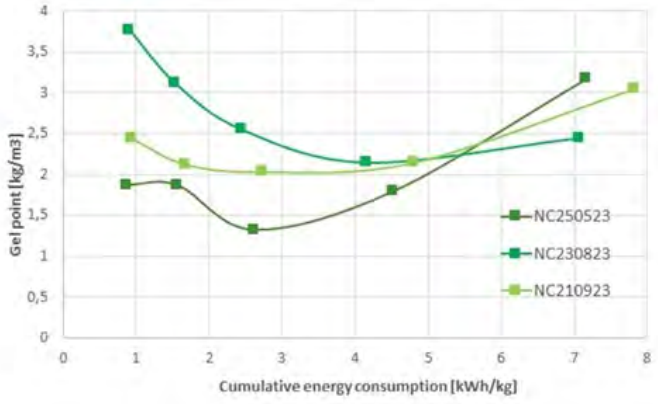


Figure 17. Gel point values for MFC fibrillated from never-dried pulp (left side) and from the same pulp stored as pulp sheets (right side) as a function of cumulative energy consumption.

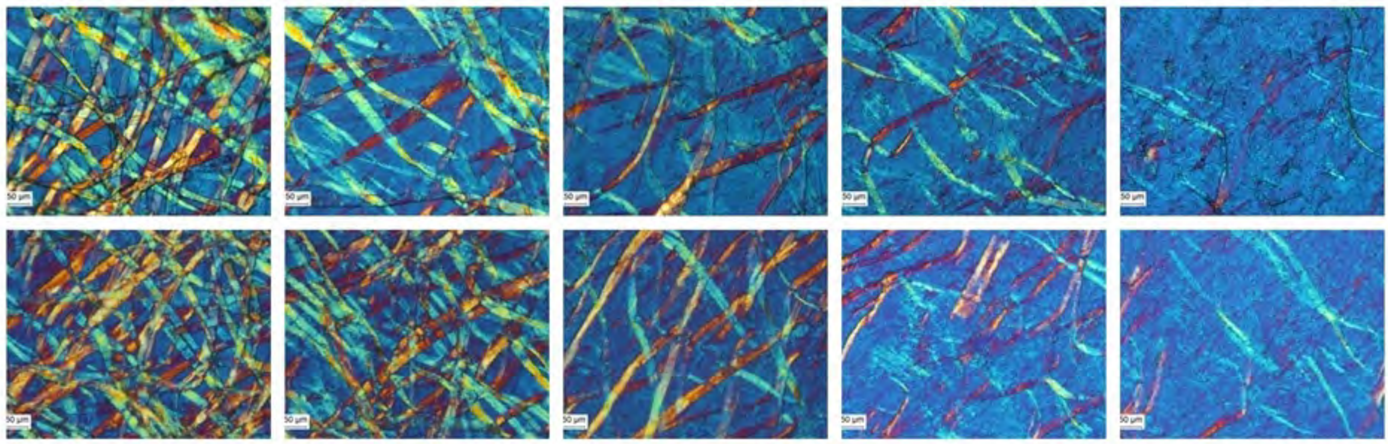


Figure 18. Representative polarised optical microscope pictures of corresponding fibrillation levels shown as points in Figure 17. First row: Samples of never-dried pulp from fibrillation cycles 2 to 6, and second row: samples of the same pulp stored as pulp sheets from fibrillation cycles 2 to 6.

Contact person

Tomas Björkqvist
Tampere University
tomas.bjorkqvist@tuni.fi

2.2. Scale-up of suberin dispersion formulations

Description

The scaling up of selected protocols developed at laboratory scale for preparation of suberin-based dispersions was performed at VTT. The aim was to identify an appropriate production process for scaling up of suberin dispersion formulations, and to provide sufficient formulation amounts for coating applications and evaluation of their coating performances. In this task, larger amounts of three type of suberin dispersion formulations were produced at 10 litre scale: i) suberin dispersion stabilised by polyvinyl alcohol (PVOH) prepared by thermomechanical method based on a VTT protocol; ii) suberin dispersion stabilised by amphiphilic cellulose nanofibres (CNF) developed at University of Oulu (UO) using a mechanical method, and iii) suberin dispersion stabilised by functionalised galactoglucomannan (GGM) developed at Åbo Akademi (ÅAU) using a mechanical method. The suberin dispersions prepared by thermomechanical method were first successfully scaled up to 500 g (up to 6 times) in the laboratory. Further scaling up to semi-pilot scale was performed in the VTT pilot using a Lödige 10L reactor. All the dispersions were visually examined right after preparation. The particle sizes and shapes were analysed with a laser diffraction particle size analyser and an optical microscope. Other investigated properties were solid content, conductivity and pH. The microfluidisation post-treatment was applied for the scaled-up dispersions for further reduction of particle sizes, as it was demonstrated that this post-treatment improves the homogeneity and stability of suberin dispersions and therefore their quality and suitability for application as barrier coatings. The scaled-up dispersions were used for the coating trials in a bigger scale using the SUTCO pilot line at VTT.

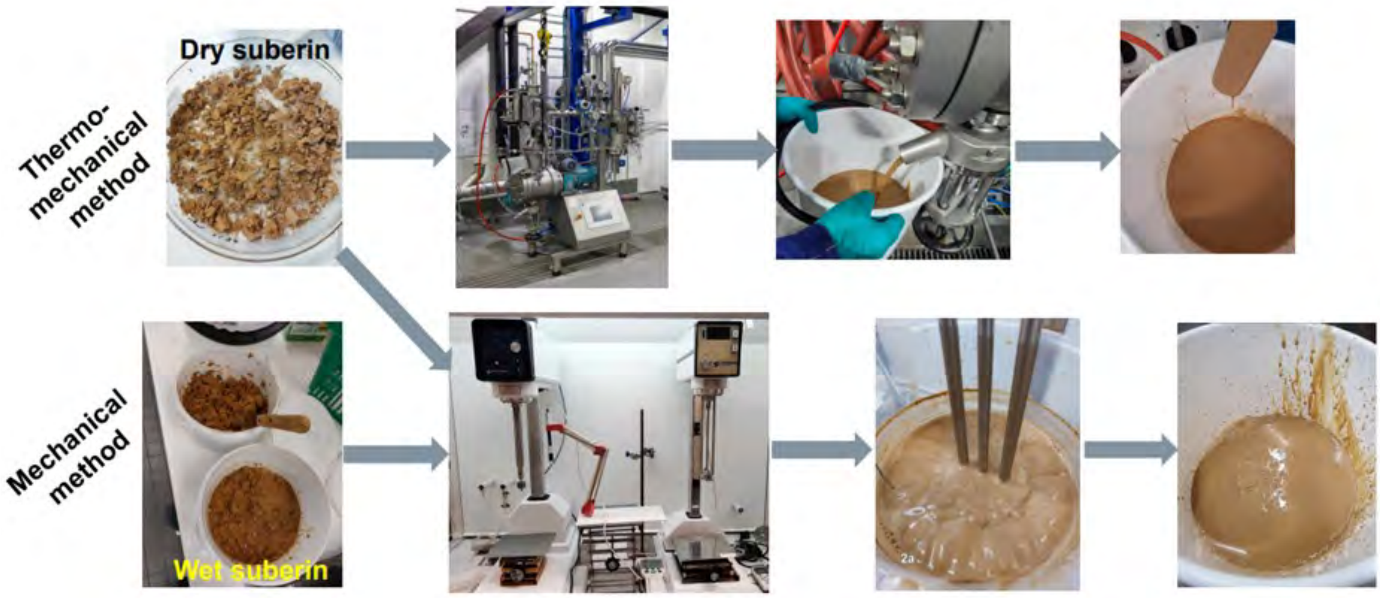


Figure 19. Production of suberin dispersions at semi-pilot scale using thermomechanical and mechanical methods.

Results

For the first campaign of pilot coating trials with SUTCO, a suberin dispersion stabilised by PVOH and prepared by thermomechanical method was used. For production of this formulation based on the first suberin batch extracted at VTT, the laboratory protocol was scaled up by a factor of 6 to produce a 0.5 kg dispersion. The effect of Ultra Turrax post-treatment was investigated for reduction of particle sizes providing increased population of particle sizes below 1 µm. The produced suberin dispersion was successfully used in the first pilot coating trials at VTT.

For the second pilot campaign of coating trials with SUTCO, the suberin dispersion prepared by a thermomechanical method was further scaled up to 8.8 kg using a Lödige 10 L reactor. The post-treatment was performed by microfluidisation for increasing the population of low particle sizes.

In addition, two other suberin dispersions were produced using mechanical method for the same pilot trials. In the case of these latest dispersions, bio-based surfactants were used for stabilisation of dispersions: amphiphilic cellulose nanofibres (CNF) and functionalised galactoglucomannan (GGM). The laboratory protocols were successfully optimised for production of these dispersions in 10 litre scale. Due to excessive foaming, microfluidisation post-treatment was not applied in the case of the dispersions stabilised by bio-based surfactants.

Dispersion experiment	pH	Brookfield viscosity (mPas)				
		Spindle	50 rpm	Torque (%)	100 rpm	Torque (%)
Suberin 2_D.11	4,44	RV05	1 760	22	1 588	39,5
Suberin 2_D.12	4,5	RV06	1 560	7,8	1 390	14
		RV06	1 000	5,1	940	9,1

Table 2. Basic characterisations of suberin dispersion prepared by thermomechanical method and scaled up in the laboratory.

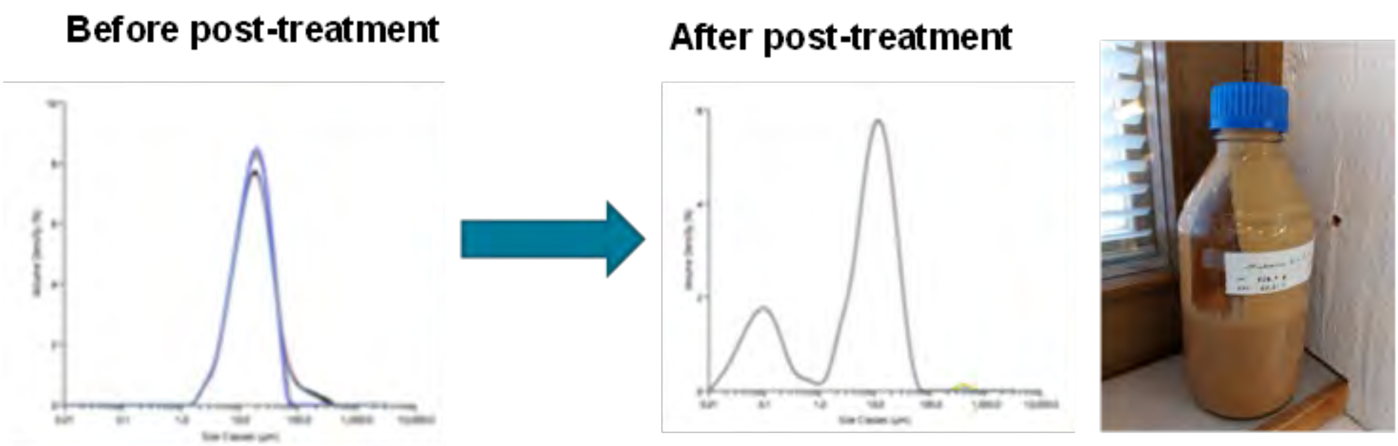


Figure 20. Reduction of particle sizes of suberin dispersion by Ultra Turrax post-treatment.

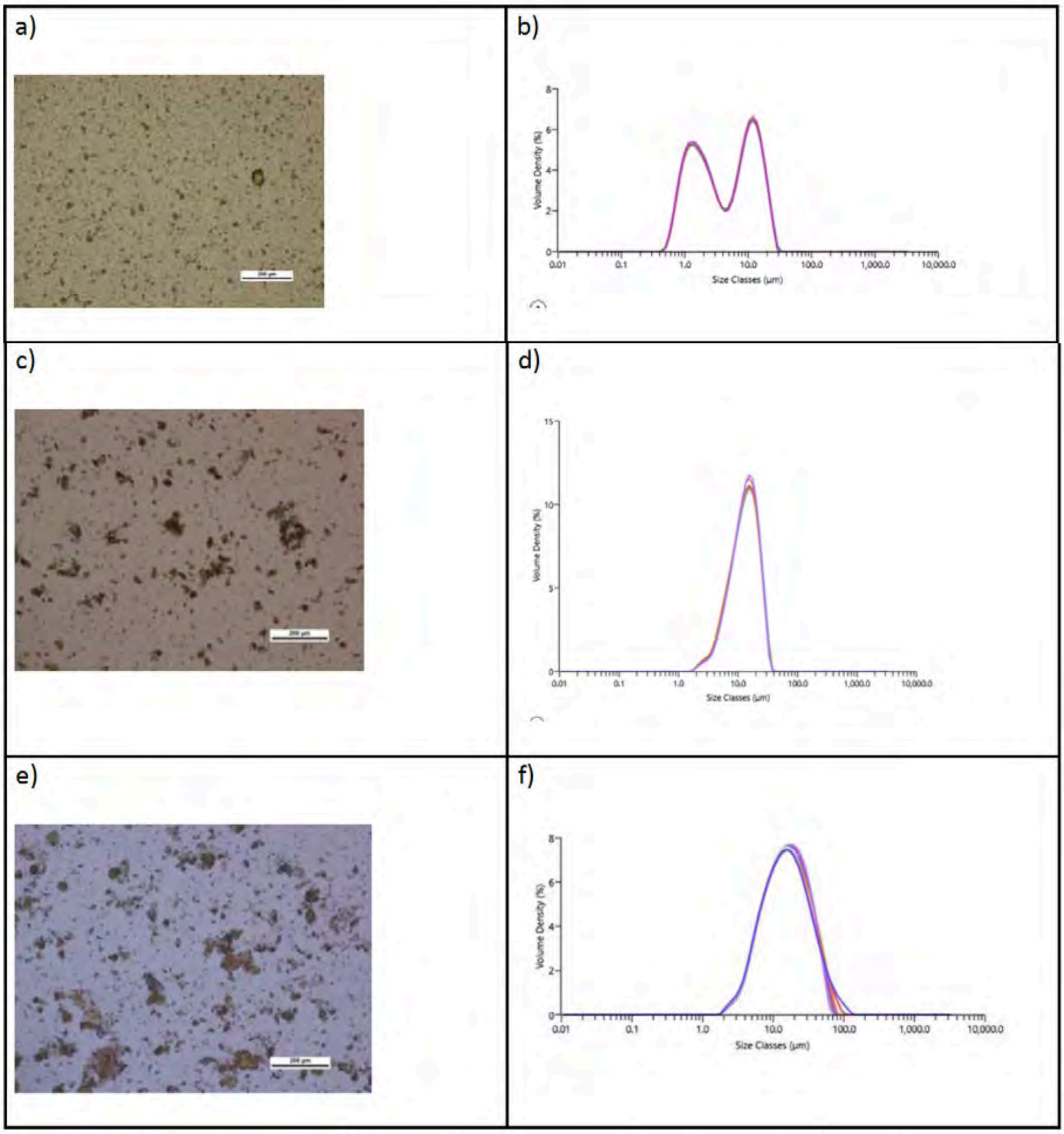


Figure 21. Characterisation of suberin dispersions produced at semi-pilot scale. Microscope images and particle size distributions. a) Suberin dispersion stabilised by PVOH. Microscope image; b) Suberin dispersion stabilised by PVOH. Particle size distribution; c) Suberin dispersion stabilised by CNF. Microscope image; d) Suberin dispersion stabilised by CNF. Particle size distribution; e) Suberin dispersion stabilised by modified GGM. Microscope image; f) Suberin dispersion stabilised by modified GGM. Particle size distribution.

Contact person

Adina Anghelescu-Hakala
VTT Technical Research Centre of Finland
adina.anghelescu-hakala@vtt.fi

Development of resins

Summary

Lignin-based coatings have the potential to replace petroleum-based coatings such as phenol-formaldehyde for different types of products such as solid wood, metals and abrasives. In WP3, technical lignins (kraft, soda and others) provided by company partners were combined with two types of crosslinkers: polyethyleneimine (PEI) and isocyanates to prepare the bio-based coatings that were then tested for relevant properties such as water resistance, hardness, elasticity and adhesion to substrate. The work was divided between partners based on the type of coated substrate: wood (VTT), metal (Teknos) and abrasives (Mirka).

VTT's lignin-PEI coatings on wood were visually pleasing but had insufficient water resistance. Lignin-PU coatings (lignin + isocyanate) also produced visually appealing coatings that also showed a good level of hardness. Water absorption of the wood was reduced by all of the coatings, but the largest improvement was obtained with a low molecular weight lignin with a high number of isocyanate-reactive aliphatic hydroxyl groups. The hardness levels and the best water resistance performance were comparable to those obtained with industrial wood coatings.

Teknos developed coatings for wood based on SW kraft lignin and PEI, a lignin-derived bioester and melamine, and a lignin nanopolyol and polyisocyanate. The kraft lignin-PEI coatings had good dry and wet adhesion, but became brittle at higher coating weights. The bioester-melamine coatings also had good adhesion, but the films were not homogeneous. The nanopolyol-isocyanate coating formulations became very viscous, which resulted in inhomogeneous films. Strategies were suggested to overcome these issues and produce industry-level coatings.

Mirka applied lignin-PEI coatings on nonwoven and paper abrasives to replace phenol-formaldehyde resins used as adhesives for the metal fragments. Although differences in performance between lignins were seen in sanding tests, the coatings on nonwovens in general performed poorly. However, performance of up to 87% of the reference was obtained with softwood lignin-PEI adhesive for paper abrasives. The main challenge for the paper-based abrasives was the dry matter to viscosity ratio, which was below the usual level.

Contact person

Petri Widsten
VTT Technical Research Centre of Finland
petri.widsten@vtt.fi

3.1. Coatings on wood

Description

The main goal of this task was to use renewable lignins to replace harmful fossil-based chemicals such as phenol and formaldehyde in the production of water-resistant coatings for wood. Polyethyleneimine (PEI) and isocyanate were used to crosslink lignin and produce coatings from lignins provided by industry partners (UPM, CH Bioforce, and Boreal Bioproducts).

Results

The first approach involved optimising the lignin to PEI ratio and reaction conditions for lignin-polyethylene imine (PEI) coatings. Lignin-PEI coatings with a lignin content of about 50 wt-% and total bio-based content of about 80 wt-% were produced at VTT. Only selected lignins produced uniform coatings that were resistant to cracking after water exposure. The coating thicknesses (Fig. A) and penetrations of lignin-PEI coatings were determined by UEF and observed to be in line with the amount of coating material applied onto the surface.

UEF also tested the thickness of the lignin-PEI coatings and the hydrophobicity of transparent lignin coatings applied without a crosslinker. The latter were deposited by spraying an industrial lignin from MetGen (Metnin™ Shield Lite) solution on the wood surface. The hydrophobicity of the surface (contact angle of water) increased with increasing lignin content of the solution (Fig. B). Ecotoxicity testing with Daphnia magna (water flea) revealed some toxicity concerns, but these decreased with the increasing degree of dilution. This was the expected result with the raw coating dispersion.

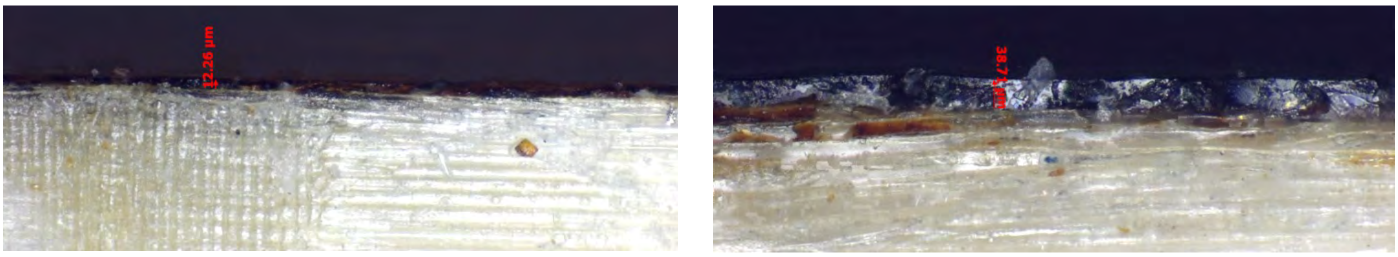


Figure 22. Thickness of lignin-PEI coatings. Left: Thin (30 g/m2) coating (12.26 µm) Right: thick (75 g/m2) coating (38.71 µm). Lignin: UPM SW kraft

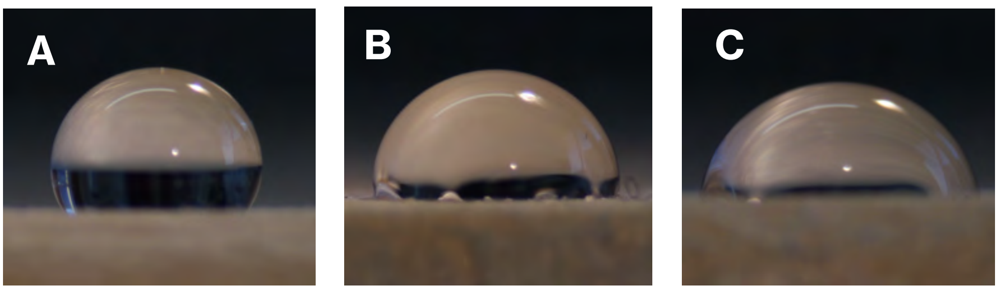


Figure 23. Water contact angles of the industrial lignin (Metnin™ Shield Lite) coatings. The undiluted solution (A) had a contact angle of 1220 (hydrophobic), the 1:1 diluted solution had that of 970 (hydrophobic), and the uncoated reference (C) had 740 (hydrophilic). These values are the average of two samples, with 15 µL water droplets.

Despite certain improvements in water resistance, the lignin-PEI coatings did not perform well enough in this regard, and this line of research for water resistant coatings were discontinued. Development of lignin-PEI coatings was, however, continued at Teknos and Mirka for other applications.

The second approach to produce water resistant lignin-based coatings was to use lignin as the main component in lignin-polyurethane (PU) coatings in which lignin and isocyanate react to form polyurethane bonds. Following a research visit to Tecnalia, Spain to learn about the production of polyurethane coatings, VTT was able to optimise the coating formulation for each lignin and coatings were produced with lignin contents of 54-57 w%. The lignin's botanical origin and processing history influenced the coating properties and the colour of the coating (Fig. C). Coating hardness was in the range of industrially acceptable coatings and could be adjusted by modifying the resin mixture (Fig. D). All the lignin-PU coatings improved the water resistance of wood, but the most significant improvement was achieved with an industrial lignin (Boreal Bioproducts spruce sawdust water extract) having the lowest molecular weight and highest content of the isocyanate-reactive aliphatic hydroxyl groups (Fig. D). To increase the bio-based content in lignin-polyurethane coatings, a partially bio-based diisosyanate could be used in combination with the bio-based lignin components. However, research is still needed to reach fully bio-based polyurethane coatings.

Figure 24. Appearance of lignin-based polyurethane coatings on glass (left) and wood (right). Lignins from Boreal Bioproducts (A), UPM (B) and CH Bio-force (C, D)

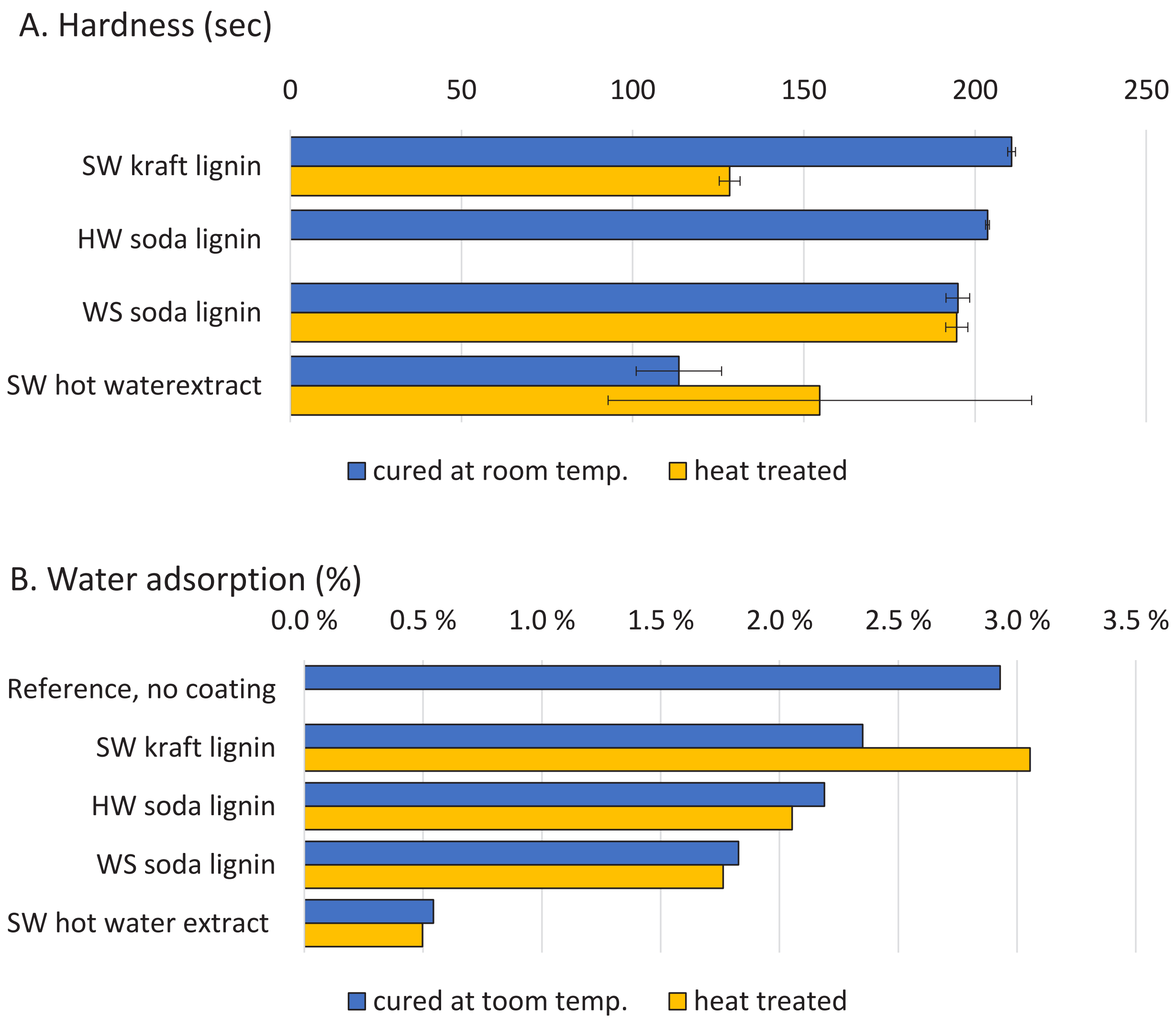


Figure 25. Top: Hardness of lignin-based polyurethane coatings on glass measured with a König pendulum. Bottom: water absorption of lignin-based polyurethane coatings on wood after a 24-hour water exposure (COBB test). Uncoated wood was used as a reference.

Contact persons

Miriam Kellock
VTT Technical Research Centre of Finland
miriam.kellock@vtt.fi

Petri Widsten
VTT Technical Research Centre of Finland
petri.widsten@vtt.fi

Viivi Berg
University of Eastern Finland (UEF)
viivi.berg@uef.fi

3.2. Coatings on metal

Description

Lignin-based coating formulations with different crosslinkers or thermal curing were prepared and applied to metal. The coatings were evaluated for dry and wet adhesion and brittleness.

Results

Coatings were prepared with SW kraft lignin (UPM) (Fig. 26), a lignin-based bioester (MetGen) (Fig. 27) and a lignin nanopolyol – lignin etherified to convert phenolic hydroxyls of lignin into more isocyanate-reactive aliphatic hydroxyls (Fig. 28). The results are compiled in Table 3. Several formulations showed promising properties particularly regarding adhesion, but further research is needed to attain all the target properties and improve processability.

Lignin	Wet coat	Crosslinking	Results, dry film on metal substrates	Possible future work
UPM lignin	Alkalis typical for coating applications were used to dissolve	Polyethyleneimine, temperature curing	Great dry and wet adhesion at lower film thicknesses Brittleness at higher film thicknesses	Optimizing curing temperature, testing plasticizers to reach applicable film thickness in coating industry
Metnin bioester	Highly viscous and very sticky, same alkalis were used to dissolve	Low formaldehyde melamine, temperature curing	Great dry and wet adhesion Non-homogenous film due to the surface tension incompatibility	Testing surfactants
	Highly viscous and very sticky	Temperature curing	Very high film thickness due to high viscosity of bioester Brittle film with poor dry and wet adhesion	Reducing viscosity and film thickness, adding plasticizers, optimizing curing temperature
Metnin Nanopolyol	Highly viscous, was easier to utilize than Metnin bioester	Polyisocyanates, temperature curing	The mixture's viscosity dramatically increased immediately after mixing, leading to a non-homogenous mixture with a rapid reaction.	Testing a non-reactive, low viscous plasticizer and/or diluent, and catalyst to favor the reaction

Table 3. Formulations and properties of lignin-based coatings for metal

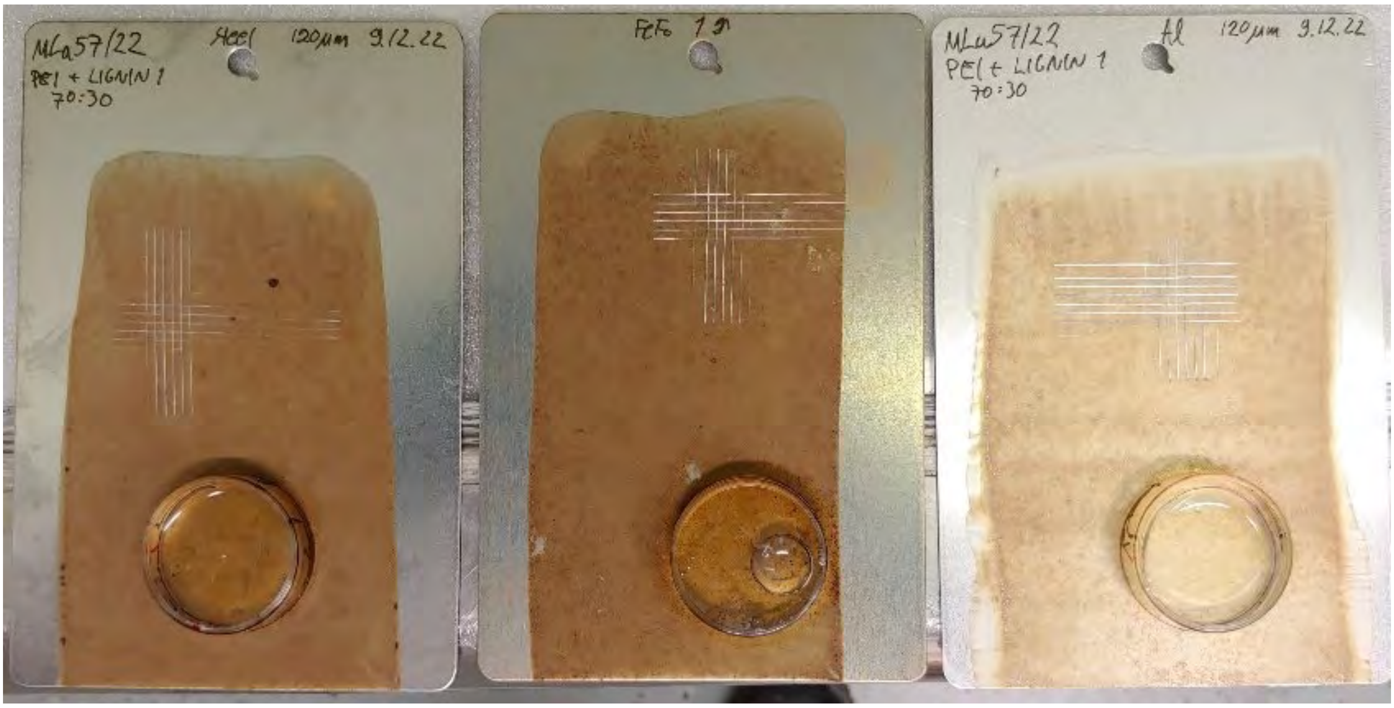


Figure 26. Lignin-PEI coatings on metal, showing the set-up for adhesion testing.

Figure 27. Metnin bioester – demonstration of its high viscosity.

Figure 28. Mixture of metnin nanopolyol and isocyanate

Contact persons

Lena Ammosova
Teknos
lena.ammosova@teknos.com

Pasi Virtanen
Teknos
pasi.virtanen@teknos.com

3.3. Coatings on nonwoven and paper abrasives

The lignin-PEI resins were tested as coatings for nonwoven and paper abrasives (Figs. 29 and 30) to replace the phenol-formaldehyde resins in coating formulations. The resins were first evaluated for their processability, defined by their dry matter to viscosity ratio. Based on this, these coatings were estimated to be better suited for spray coating than for roll coating.

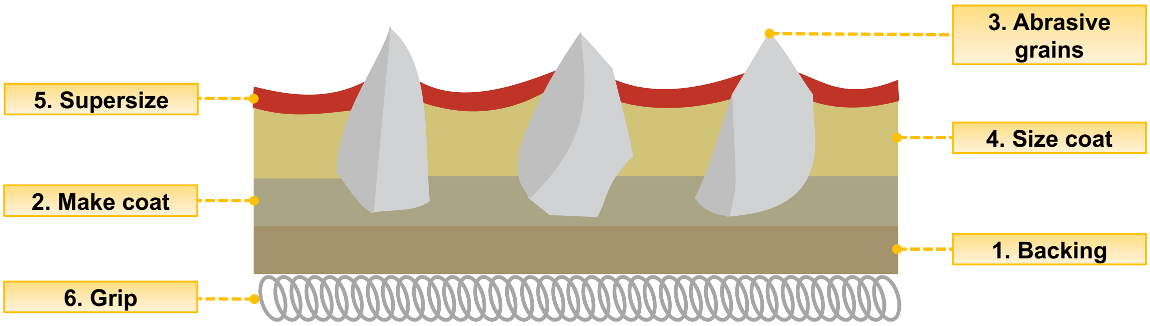


Figure 29. Abrasives typically consist of different layers: 1 the backing material, 2 the make coat, 3 abrasive grains, 4 the size coat and 5 the supersize

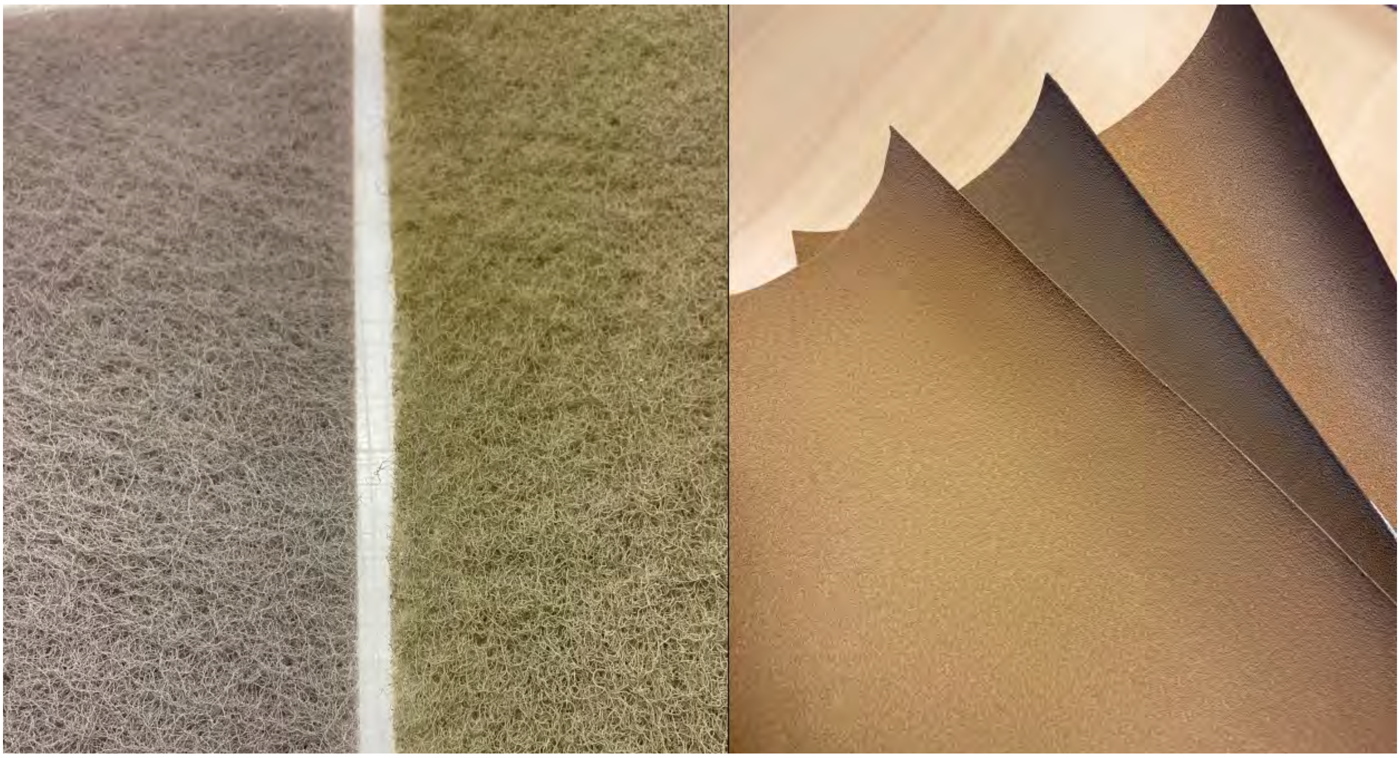


Figure 30. Spray- and roll-coated nonwoven and paper abrasives with lignin coatings

Coatings on nonwoven: It was found that these coatings are not water-resistant enough to perform in spray-coated abrasives. Their performance could be slightly improved in wet and dry sanding by selecting another coating to the outermost coating layer (Fig. 31). Their performance was poor.

Coatings on paper: Comparison of different lignin sources was studied on roll-coated abrasives. Samples with lignin-PEI resins as binders in size coatings were tested in practical sanding tests. The sanding performance versus time was recorded. The coatings had limited durability, as the dry matter to viscosity ratio prevented normal level of binder usage in the coating mixture. However, the performance differences of the coatings based on softwood lignin and hardwood lignin are evident (Fig. 32). Softwood lignin gave 87% of the reference performance, but hardwood only 32%.

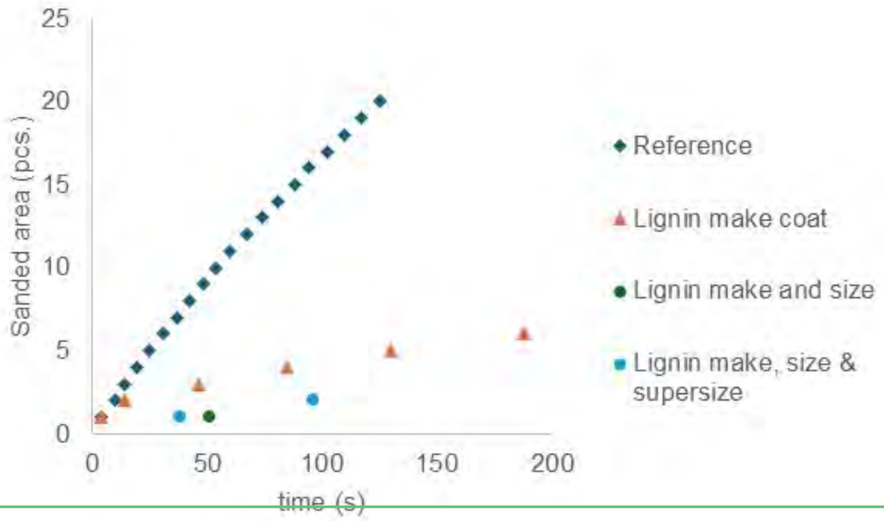


Figure 31. Wet sanding performance of spray coated abrasives with lignin coatings in different coating layers. See Figure 29 for an illustration of the layers in abrasives

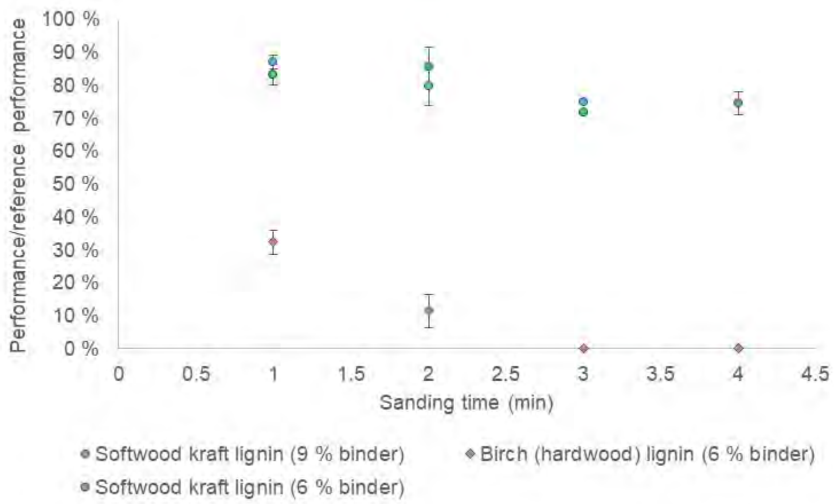


Figure 32. Lignin source effect on sanding performance

Contact persons

Anna Kirveslahti
Mirka
anna.kirveslahti@mirka.com

Petter Andersson
Mirka
petter.andersson@mirka.com

SUSBINCO

Work Package 4

Applications and testing

Summary

In WP4, the objective was to test and optimise the functionality of the novel bio-based dispersions in the coating and converting processes. It is vital that the novel dispersions be investigated and optimised for both coating processes and the subsequent post-processing. WP4 provided valuable feedback for the other work packages, which is crucial for gaining a better understanding of the functionality of the developed solutions and for maximising their potential through process and tooling optimisation. Because material convertibility cannot be evaluated strictly based on laboratory measurements, it is crucial to assess the functionality in pilot-scale and production-scale equipment. The main partners in WP4 were LUT (WP leader), VTT and UEF.

Contact persons

Ville Leminen
LUT University
ville.leminen@lut.fi

Riku Talja
Metsä Board
riku.talja@metsagroup.com

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BENEFITS FOR PARTICIPATING COMPANIES

UPM-Kymmene Oyj

Development of know-how and infrastructure widely serves the Finnish bioeconomic cluster
The SUSBINCO project established a valuable expert network from industrial and research partners that responded to the demand for sustainable wood-based binders. The partners involved have well developed infrastructure and know-how that support industry needs in novel products and strengthen the Finnish bioeconomy cluster. CLIC Innovation's project booster, combined with Business Finland support, enabled substantial efforts towards meeting this central challenge.

Skilful researchers developed processes and methods throughout the value chains, delivering new insights on many levels. The valuable results include among other methods for the visualisation of suberin dispersions, upscaling of aqueous dispersion production, pilot coating and packing trials. UPM R&D competence in the paper packaging materials area was greatly developed with the project partners. Valuable insights were gained from testing methods for end-of life options and in carrying out life cycle assessments for novel suberin-based products. Publications reach the scientific community far beyond the SUSBINCO consortium.

The threshold for process and product development is lowered through the project, because critical steps in terms of technology and sustainability have been identified.

Christiane Laine,
Chairperson of SUSBINCO's steering group and project manager of UPM's company project

4.1. Pilot coating of superin-based dispersions

Description

VTT conducted research on suberin-based dispersions for potential application as barrier coatings. The initial phase involved coating trials at a laboratory scale using a rod coater on two base substrates provided by the project partners. Three commercially available dispersions (from the project partners) were used as a reference point. Their barrier properties were measured and analysed to serve as a benchmark for the subsequent suberin coatings. VTT then formulated their own suberin dispersions stabilised with polyvinyl alcohol (PVOH). These in-house dispersions were rod-coated on a laboratory scale to determine the optimal coating thickness and weight. The same rod coating method was then employed on a pilot scale using the SUTCO coater to coat both the commercial dispersions and the PVOH-stabilised suberin dispersions in a continuous roll-to-roll process. After coating, the barrier properties of these pilot-scale samples were evaluated, and the samples were forwarded to LUT for convertability testing. The project progressed to using larger volumes of suberin dispersions (10 litre scale) stabilised with three materials: PVOH from VTT, fatty acid grafted hemicellulose from ÅAU, and modified cellulose nanofibrils (CNF) from UO. These dispersions were coated on both the paper and paperboard substrates using a reverse gravure coating method on the SUTCO pilot line.

Results

Two coat weights were produced for each substrate-dispersion combination, resulting in a total production of approximately 70-170 metres of coated material for each trial point. Visual representations of the SUTCO coating trials are provided in Figure 1. Following the pilot-scale coating, the barrier properties of the coated samples were characterised and compared to those produced at the laboratory scale. Tables 1 and 2 summarise the coat weights and thicknesses achieved for the suberin-coated paper and paperboard. The barrier properties of the coated papers are further illustrated in Figures 2 and 3. Finally, these pilot-scale coated papers were sent to LUT for further tests to assess their convertibility. The samples were also shared with the partners in WP5 to evaluate the environmental impacts (task 5.1) and end-of-life issues (task 5.2).

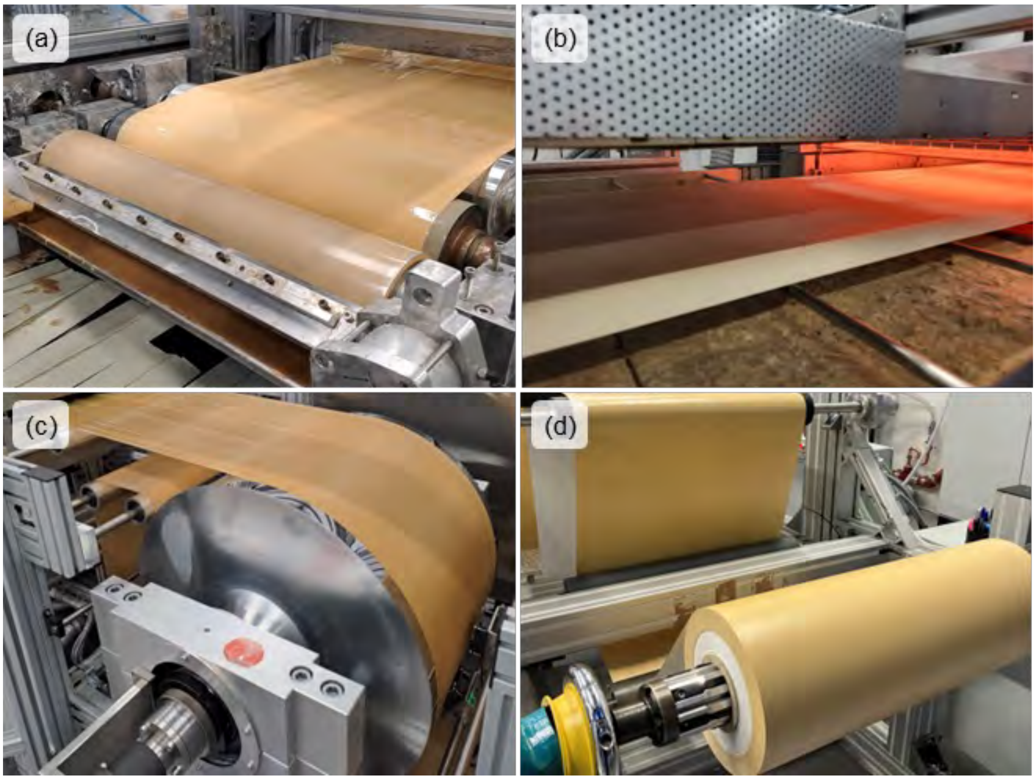

Figure 33. Pilot-scale SUTCO coating of suberin-based dispersions, (a) reverse gravure coating, (b) IR dryers, (c) cooling roller, (d) rewinding

Substrate: Paper (53 g/m2, 60 µm, pre-coated base)			
Dispersion	**Coated length (m)**	**Coat weight (g/m2)**	**Coating thickness (µm)**
S-PVOH	77	15.2 ± 0.8	18.3 ± 1.7
S-HC	81	11.6 ± 0.6	13.7 ± 0.3
S-CNF	175	11.0 ± 0.3	13.3 ± 1.5
Ref-1*	172	20.6 ± 0.3	15.3 ± 0.6
Ref-2*	184	14.5 ± 0.3	13.0 ± 1.0

Table 4: Coat weight and coating thicknesses for suberin- and commercial dispersion-coated paper. (Ref-1 and Ref-2 – commercial reference dispersions, PVOH – Polyvinyl alcohol, HC-hemicellulose, CNF-Cellulose nanofibres).

Publications

Koppolu, R., Nissinen, E., Hämäläinen, R., Anghelescu-Hakala, A., Leminen, V. 2024. Suberin fatty acid derived bio-based dispersion coatings for barrier packaging. 24th IAPRI World Packaging Conference, Valencia, Spain, June 17–21. Submitted.

Liqiu Hu, Rajesh Koppolu, Roosa Hämäläinen, Heimo Kanerva, Tapani Nick, Martti Toivakka, Risto Korpinen, Pekka Saranpaa, Umair Qasim, Henrikki Liimatainen, Chunlin Xu, Adina Anghelescu-Hakala. 2023. Suberin based aqueous dispersions for barrier coatings. ACS Sustainable Chemistry & Engineering. https://doi.org/10.1021/acssuschemeng.4c02244.

Invention disclosures

Preparation of aqueous suberin dispersions for barrier coatings. Rajesh Koppolu and Ilmari Oravala.

Suberin pigment hybrid dispersions for barrier coatings. Rajesh Koppolu and Ilmari Oravala.

Contact person

Rajesh Koppolu
VTT Technical Research Centre of Finland
rajesh.koppolu@vtt.fi

4.2. Converting analysis of coated materials

Description

LUT and the project partners analysed the barrier properties and convertibility of the developed coatings. The performance was compared to commercial benchmarks, and research and development work was done also to understand the packaging convertibility of the coated materials, and how the converting processes and tooling need to be developed to maximise the functionality of the coated papers and paperboards in package converting. The main converting processes that were investigated included creasing for the coated paperboards and heat sealing and vertical form-fill-seal (VFFS) package manufacturing for the coated papers. The mechanical and surface properties of the materials were also analysed. As part of the studies, a literature review was conducted on how converting in general affects the barrier properties of coated packaging materials.

Results

The results from the creasing studies showed that the developed coatings provided promising barrier properties. However, converting can have a significant impact on the barrier properties of the materials. This highlights the need for both development of material properties to consider convertibility, both also the development of converting tooling and processing. Figure 34 shows the changes in the oil and grease resistances (OGR) of the coated materials before and after converting.

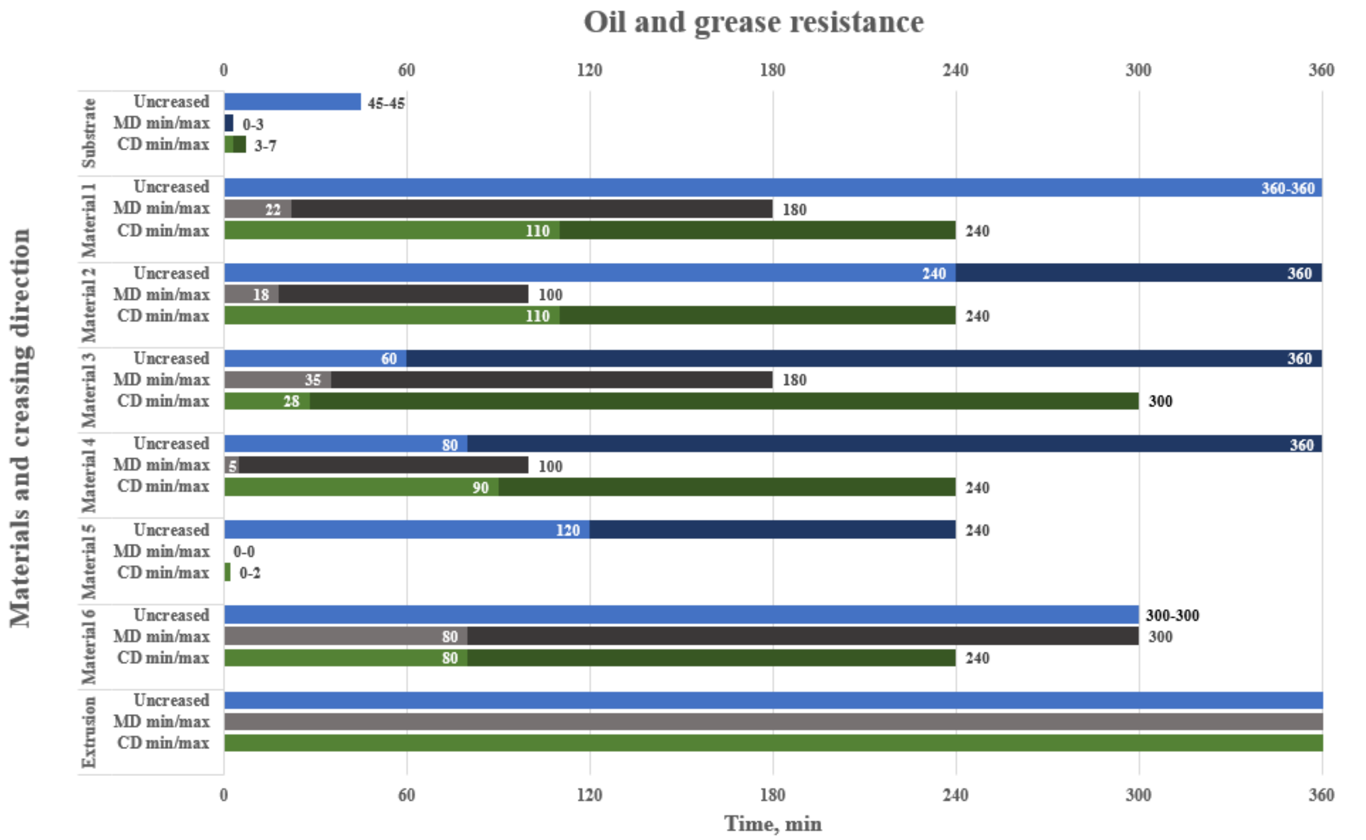

Figure 34. Oil and grease resistances of the samples

Several interesting observations were made related to VFFS converting of coated papers. The material friction can hinder the convertibility, and it was also noted that the converting reduces the barrier properties of the materials. Part of this reduction can be explained by wrinkling in the forming shoulder area (Fig. 35).

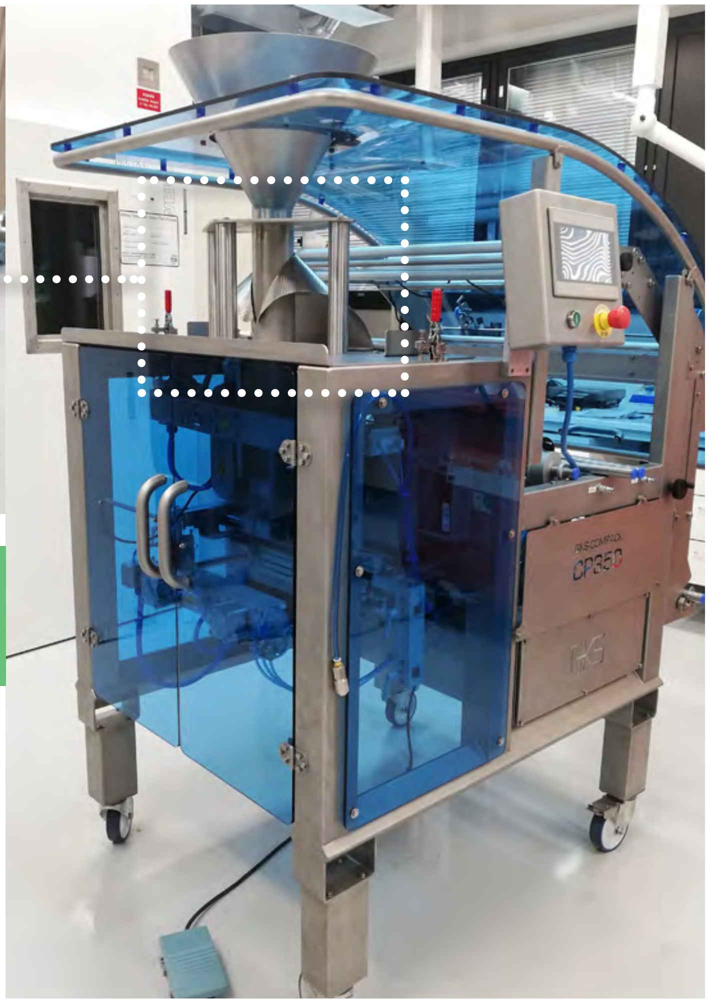

Figure 35. VFFS equipment used in the study, including a close-up of a forming shoulder.

The frictional behaviour of materials varied, and the surfaces of the equipment (especially in the forming shoulder and sealing tools) clearly affect the material runnability in the VFFS process. A Review. Based on the results, several observations related to converting tooling could be made. For example, flat tools in the heat-sealing process should be avoided, and serrated tools are recommended. In the forming shoulder design, material surface pattern and geometry should be considered the minimise the wrinkling in converting, and to ensure smooth runnability of materials.

Publications

Lev, R., Tanninen, P., Lyytikäinen, J., Leminen, V. Converting and its Effects on Barrier Properties of Coated Packaging Materials: Bioresources 18(4), 2023.

Lev, R., Lyytikäinen, J., Matthews, S., Koppolu, R., Tanninen, P., Leminen, V. Investigating the convertibility and barrier performance of a novel suberin dispersion coating during the paperboard creasing process. Packaging Technology and Science. Submitted.

Merabtene, M., Tanninen, P., Pesonen, A., Lyytikäinen, J., Varis, J., Leminen, V. Evaluation of Frictional Influence and Thermal Analysis of Flexible Paper-Based Materials Used in the Vertical Form– Fill–Seal Process: An Experimental Study. Packaging Technology and Science 2024.

Contact person

Ville Leminen
LUT University
ville.leminen@lut.fi

4.3. Testing of resins for protective coatings

Description

At UEF, we have prepared coatings from industrial UV-curable resins and a thermally curable wood distillate. The coatings have been modified with two phenolic mixtures. They are characterised using complementary techniques, and antimicrobial testing is commenced, which also includes the coating dispersions in addition to coatings.

Contact person

Viivi Berg
University of Eastern Finland
viivi.berg@uef.fi

Results

Detailed results will be available as the studies are completed by June 2024.

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BENEFITS FOR PARTICIPATING COMPANIES

Mirka

Most abrasives are single-use products that are thrown away after a short period of use. Because of this, it is crucial that Mirka, as an abrasives company, rethinks the way we make our products and what raw materials we can use. We need to find more sustainable solutions to shape a more environmentally friendly future.
The work with finding environmentally better solutions for abrasive products has already started within Mirka's own Veturi project SHAPE, and the collaboration regarding new bio-based binders and coatings in SUSBINCO has been an interesting and much-needed addition in our aspiration to make more sustainable products.
The key goal for Mirka in SUSBINCO has been to understand the fibrillation process of nanocellulose, and to build a concept for more efficient fibrillation. We believe that nanocellulose is a key raw material needed for a more sustainable future, by replacing fossil-based raw materials in coating and packaging. To be part of this research is of major importance to us, including from a business perspective. We also want to pinpoint where and how we can use raw materials for backings and coatings, as well as packaging that are better for the environment. The work in SUSBINCO has helped us to do so.

Another important aspect is the network of knowledge that has been formed during the project between research and industrial partners. The collaboration with both industrial and research project partners has been of great help for Mirka during the project. We are proud to be part of creating a strong nanocellulose and bio-based coatings hub in Finland, and to move bio-based products closer to the present than to the future.

Petter Andersson
Steering group member
Mirka

Work Package 5

SUSBINCO

Environmental impacts, end-of-life, and safety matters

Summary

The objective of WP5 was to test and evaluate environmental impacts, end of life, and safety aspects of new materials developed in the SUSBINCO project for sustainable binders and coatings and final products. This would ensure sustainable and safe development throughout the WPs and tasks all the way from raw materials to new end-product prototypes. WP5 utilised the analytical and testing results from other WPs for various purposes, including pinpointing potential harmful contaminants, solvent residues and VOC compounds. In addition to standard reference materials and compounds, existing approved products from industrial partners were used as relevant references or base lines for analysis and testing.

WP5 results provided valuable feedback for the other work packages, which is crucial to better minimise potential harmful components and optimise the processing and procedures with respect to environmental safety, composting, recycling (repulping), (eco)toxicity, and mutagenicity. The outcome of the project provides a good basis for further development and approval testing of products and processes related to e.g. food contact applications as required by ISO 22000 food safety certification.

Contact person

Reijo Lappalainen
University of Eastern Finland
reijo.lappalainen@uef.fi

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BENEFITS FOR PARTICIPATING COMPANIES

CH-Polymers Oy

Expanding knowledge and partnership networks through the SUSBINCO project

CH-Polymers joined the SUSBINCO project to develop bio-based and sustainable alternatives to fossil-based materials, in response to the growing demands from brand owners, consumers, legislators and other decision-makers. The project consortium, which includes the entire value chain, enabled the establishment of new partnership networks to develop future bio-based technologies with a significant potential impact. As a participant, CH-Polymers collaborated with an extensive network of industry leaders and research organisations to develop bio-based binders and coatings for various product applications. The SUSBINCO project expanded and strengthened CH-Polymers' partner network and furthered its internationalisation efforts. Valuable insights were provided into suberin extraction, hemicellulose modification and the evaluation of the sustainability and end-of-life of new bio-based solutions.

The developed technologies in the SUSBINCO project stand as a great achievement brought about by close collaboration between expert researchers, which serves as a comprehensive background for further innovations in bio-based solutions for binders and barrier coatings.

Misla Lagus
R&D Manager, Paper & Packaging and Project Manager of CH-Polymers' company project

5.1. Environmental impacts

Description

Task 5.1 was supporting the material and product design by assessing the environmental impacts of the developed coatings already in the early phase of the design process. A life cycle assessment (LCA) method was used. The goal of the study was to find the hotspots and identify the factors which should be minimised or optimised to reduce the environmental impacts of the dispersion coatings production. A formulation consisting of water, suberin and modified hemicellulose was selected for assessment. The entire life cycle, i.e. from material acquisition to the product's end of life, was studied, except for the packaging use phase, as illustrated in Figure 36.

The functional unit of the study was 1 m2 of coated paper and the reference unit was 35 g of dispersion. The impact assessment methods used were IPCC AR6 GWP 100 (version Aug. 2021) and EF 3.0. All impact categories have been included, except toxicity categories due to lack of consistent accurate data for them. Inventory data was iteratively collected from project partners and from the Ecoinvent database. Pilot data on suberin extraction and laboratory data on hemicellulose purification and modification were obtained. LCA for experts (aka GaBi) software was used for life cycle modelling. The allocation of impacts between co-products for suberin and hemicellulose extraction was carried out on a mass basis.

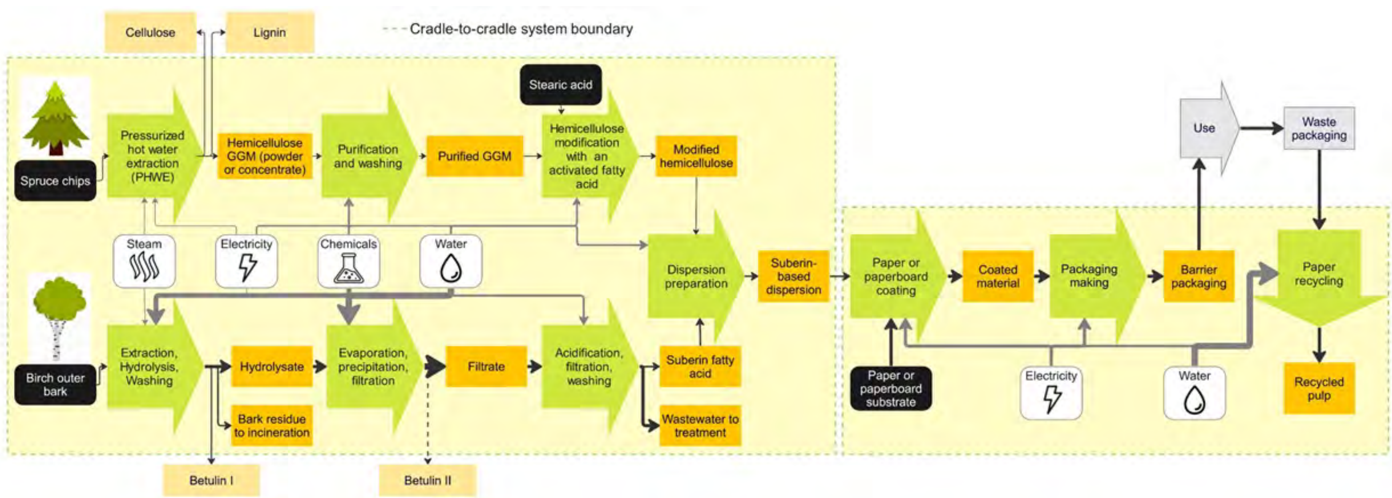

Figure 36. System boundary of the LCA study on suberin-based dispersion coating.

Results

The production of modified hemicellulose and suberin makes the largest contribution to the considered environmental impact categories, as shown in Table X. First, in the baseline scenario, most of the emissions are from laboratory purification and modification due to the intensive use of solvents that have not been recycled. In the scenario where solvents are recovered and circulated in the system, hemicellulose impacts are significantly lowered, resulting in a reduction in total impacts of up to 50% in most categories. Second, hemicellulose modification requires a long stirring process, resulting in high energy consumption. The second energy-intensive stage was the paper coating process, as this involves drying and evaporation of water from the dispersion coating. The most serious contribution to suberin production was extraction, hydrolysis, and washing due to the large amount of ethanol used and steam consumption. Even considering solvent recycling, these steps are intensive. Another optimisation could be improvements in drainage and filtration processes to reduce the amount of circulating water, which subsequently needs to be evaporated. In addition, more efficient extraction of betulin could potentially reduce the impact of suberin production. The packaging recycling process, particularly recycled pulp quality, wastewater content and rejects, must be studied in more detail to draw conclusions about the impact on LCA results.

Impact category	Units	Total	Suberin production	Modified hemicellulose production	Dispersion preparation	Dispersion paper coating	Paper production	Packaging making	Repulping of packaging
IPCC GWP 100, excl. biogenic CO_2	kg CO_2 eq.	0.75	0.07	0.51	0.01	0.08	0.07	1.31E-04	0.01
IPCC GWP 100, incl. biogenic CO_2	kg CO_2 eq.	1.05	0.27	0.59	0.01	0.09	0.04	1.49E-04	0.05
EF 3.0 Acidification	Mole of H+ eq.	3.18E-03	4.48E-04	1.76E-03	4.63E-05	3.38E-04	4.71E-04	5.82E-07	1.15E-04
EF 3.0 Land use	Pt	32.23	5.59	19.41	0.11	0.78	6.32	1.34E-03	0.02
EF 3.0 ozone depletion	kg CFC-11 eq.	4.74E-08	1.92E-08	1.37E-08	5.97E-10	4.35E-09	7.49E-09	7.51E-12	2.01E-09
EF 3.0 particulate matter	Disease incidences	2.59E-08	3.43E-09	1.45E-08	2.40E-10	1.75E-09	4.94E-09	3.02E-12	1.03E-09
EF 3.0 resource use, fossils	MJ	12.40	1.28	6.58	0.34	2.50	1.56	4.31E-03	0.14
EF 3.0 resource use, mineral and metals	kg Sb eq.	4.32E-06	5.34E-07	1.99E-06	1.33E-07	9.70E-07	4.06E-07	1.67E-09	2.80E-07
EF 3.0 Water use	m^3 world equiv.	0.53	0.06	0.23	0.01	0.09	0.11	1.53E-04	0.03

Table 5. Life cycle impact analysis results with highlighted hotspots for baseline scenario

As might be expected, laboratory and pilot level processes have a large environmental impact. Since this is an early design stage assessment, those processes should not be directly compared to the industrial production of coating components. Further improvement and development of the upscaling model together with other WPs will enable a comparative LCA study.

Contact person

Natalia Vinitskaia
LUT University
natalia.vinitskaia@lut.fi

5.2. End-of-life issues

Description

Recycling and biodegradability represent preferable end-of-life options for bio-based packaging materials (Fig. 37). In SUSBINCO, LUT focused on evaluating the repulpability, while VTT assessed the biodegradability of the developed materials. When materials are biodegradable and disintegrate within a specific timeframe and under certain conditions, they become suitable for composting. During composting process, humic compounds are formed as an end product, which is valuable for soil improvement. It is crucial that packaging materials do not negatively impact the treatment process or the quality of humus. To ensure this, ecotoxicological studies (conducted by UEF) were closely linked to the evaluation of biodegradability.

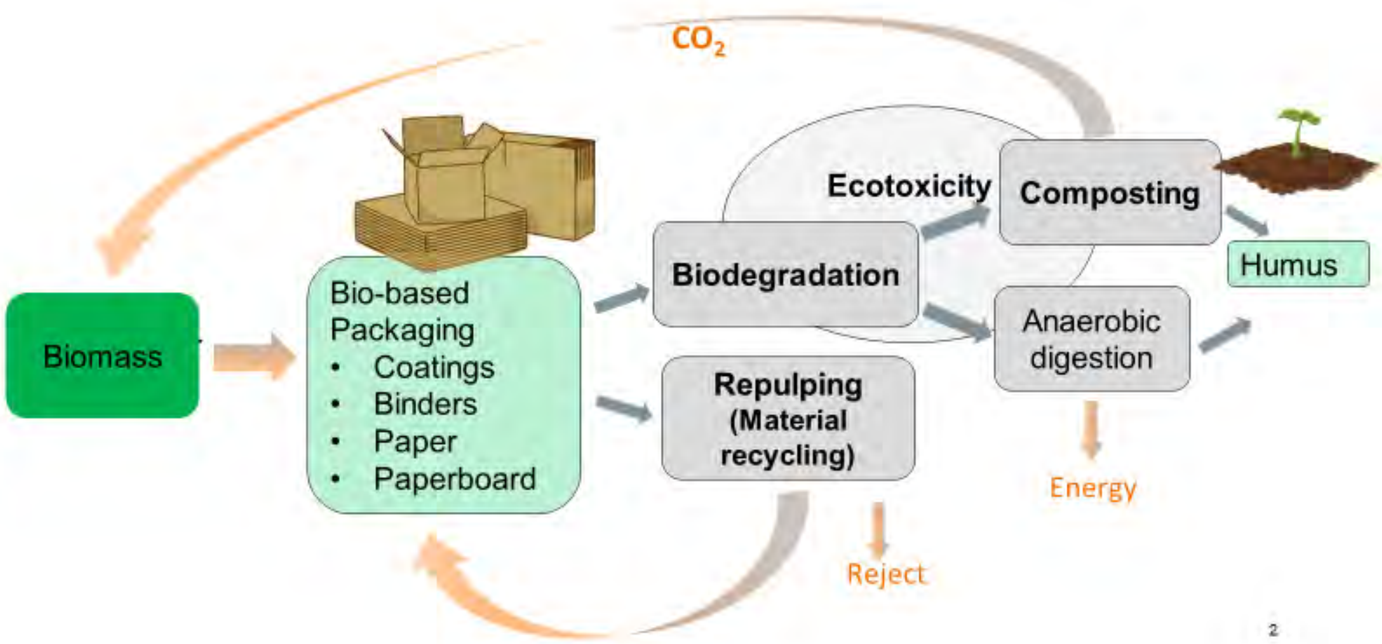

Figure 37. End-of-life options for bio-based packaging in SUSBINCO. Anaerobic digestion is an alternative treatment process for compostability.

Results

The biodegradability of suberin-coated paper/ paperboards and their raw materials was assessed using standardised ISO methods under both soil and aquatic conditions. To evaluate ecotoxicity, samples were taken from the aquatic biodegradability test and subjected to *Daphnia magna* test in UEF. Furthermore, the biodegradability of the final products was examined in industrial composting conditions.

Suberin-coated paper and paperboard exhibit high biodegradability in soil and aquatic conditions, despite of a small reduction due to the suberin coating. Suberin is a complex polyester containing long chain fatty acids and aromatic structures, which might hinder its mineralisation to CO_2 during biodegradation. Standardised biodegradability tests are based on the concept of mineralisation, and this has been shown to yield poor biodegradability values for complex biopolymers like lignin (Vikman et al., 2024). The influence of lignin was clearly evident in the limited biodegradability of birch bark, which serves as raw material for the manufacture of suberin. Furthermore, the results obtained in SUSBINCO also underscore the importance of considering the biodegradability of surfactants and other additives. Notably, the commercial PVA-grade used to prepare the suberin dispersion exhibited poor biodegradability under tested conditions.

Repulpability was investigated from the uncoated and suberin-coated paper and paperboard materials. Repulpability experiments were performed based on the 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 (CEPI recyclability laboratory test method) (Cepi 2020). The main process steps in the method include disintegration of the material, coarse screening and fine screening. Reject is determined from both screening phases. The evaluation of repulpability was performed based on fibre-based packaging recyclability evaluation protocol, beta release (4evergreen), which considers a standard mill case.

The total amounts of reject from the suberin-coated papers and paperboards were less than 1.2% in all cases. The amount of reject was low for the suberin-coated materials and minor also for the uncoated materials. Suberin coatings adhered lightly to the equipment during the disintegration of the material and foaming occurred in some samples during coarse screening. Nevertheless, repulpability issues were not expected for the experimental coatings developed in the project as the total amount of reject was minor for all the materials.

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Contact person

Minna Vikman
VTT Technical Research Centre of Finland (VTT)
minna.vikman@vtt.fi

BENEFITS FOR PARTICIPATING COMPANIES

Metsä Board

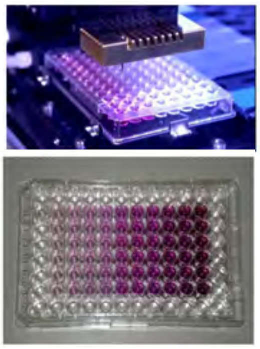
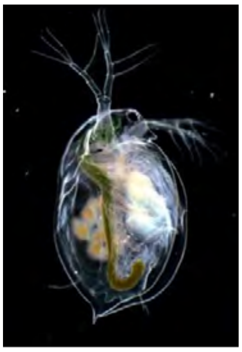
Metsä Group has firmly committed to strategic sustainability 2030 objectives for creating a path to a climate-neutral society and a more sustainable future. This is seen in Metsä Board by reducing and getting rid of use fossil fuels and fossil-based energy. Alongside this, Metsä Board has started to develop solutions to replace fossil raw materials in paperboard products with sustainable solutions. The SUSBINCO project has been supporting Metsä Board's targets by gaining know-how of bio-based and wood-based materials and their usage on the road towards a sustainable future. Collaboration between the research partners deepened through the project, which can be seen as high-quality knowledge.

Riku Talja, Manager, Barrier Development, Metsä Board

5.3. Safety issues

Description

Direct and indirect contact packaging materials, substances, processing aids, coatings, adhesives, and adjunct substances are controlled and conditionally approved for various applications. In order to minimise physical, chemical and biological risks for human health and environment related to the materials and products, proper procedures and testing are required already in the early stages of RDI. The safety issues were studied in this project at three stages of novel bio-based materials: dispersions, raw materials and their ingredients; coated paperboards and samples degraded in liquid biodegradation test. µAmes testing was used to evaluate toxicity and mutagenicity, Daphnia magna (water flea) test was used for ecotoxicity and µ-CTE™ to determine VOC emissions from coated/uncoated paperboards.

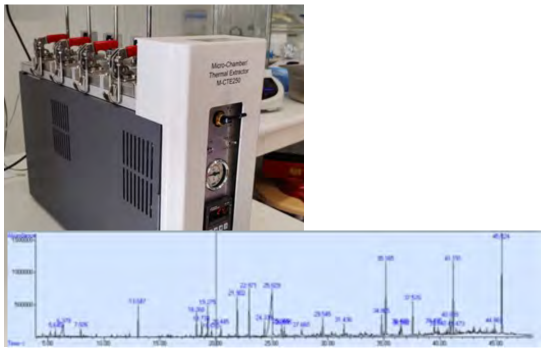


Figure 38.
µAmes *Daphnia magna* *VOC emissions*

Results

Both toxicity and ecotoxicity depends on the concentration of the potentially harmful compounds. In these tests, raw liquid samples turned out to be potentially toxic, as expected, but at diluted concentrations they became less harmful. In most of the tested cases, toxicity was caused by the solvents or other dispersion components, not the main components such as suberin, hemicelluloses or nanocelluloses. Therefore, the possibility of using proper bio-based non-toxic components such as solvents or dispersion agents should be carefully considered.

As indicated in the biodegradation tests (task 5.2.) both the paperboards and their coatings and coating materials degrade in a liquid and soil environment over time. To evaluate the potential ecotoxicity of degradation products, we collected samples from biodegradability test in a liquid environment. Based on the ecotoxicity test, tested 12 materials were not ecotoxic in a Daphnia magna test.

In VOC emission testing, significant differences were observed in the quantity of emissions from different industrial paperboards as well as from the same paperboards coated with different receipts developed in the SUSBINCO project. Detailed identification of compounds, their quantities, and dependence on the temperature is ongoing and will be included in a scientific article.

Contact person

Isa Lyijynen
University of Eastern Finland
isa.lyijynen@uef.fi

Publications

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Invention disclosures
Preparation of aqueous suberin dispersions for barrier coatings. Rajesh Koppolu and Ilmari Oravala.

Suberin pigment hybrid dispersions for barrier coatings. Rajesh Koppolu and Ilmari Oravala.

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