Properties and Hydrophobization of Nonwoven-Woven All-Cellulose Composites

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All-cellulose composites (ACCs) have been fabricated by using a variety of cellulosic sources, versatile technologies, and are sustainable alternatives for traditional composites. In this study, nonwoven-woven ACC laminates were created from wood-based Spinnova short fibers and Lyocell fabrics *via* partial dissolution and an NaOH-urea solvent system. The less-known wood-based Spinnova fiber is created for the textile industry, but it also has great potential for the composite industry. To identify the mechanical properties of ACCs—which greatly influence the range of material application—tensile, impact, and flexural tests were conducted. The mechanical properties indicated only moderate properties, which are influenced by high porosity and weak fiber bonding. Despite this, valuable information on the nonwoven-woven structured ACCs was obtained. To improve the ACC laminate's ability to resist moisture, bio-based coatings (*e.g.*, commercially available birch bark betulin and suberin acid mixture) were applied on the surface of ACCs and it successfully improved the wetting resistance. The results of contact angle analyses demonstrated that the highest contact angle of 128° was measured for betulin-coated laminates and the best stable hydrophobicity calculated a minute after the beginning of the experiment were observed at 109° for the uncommercial pressurized hot ethanol (PHE) extract of birch bark.

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INTRODUCTION

Renewable resources, notably wood-derived materials, present viable alternatives to conventional fossil-based substances and play an essential role in addressing climate change challenges. The Intergovernmental Panel on Climate Change (IPCC) emphasizes the pivotal importance of sustainably produced woody biomass in advancing a green circular bioeconomy while emphasizing the critical need for sustainable forest stewardship (Shukla *et al.* 2019). Wood-based products contribute to achieving the target of carbon neutrality, as they encapsulate atmospheric carbon dioxide, which is sequestered during the transformation of these products into finished goods (Bergman *et al.* 2014; Kazulis *et al.* 2017). Moreover, these bio-based products are biodegradable, thereby having a reduced environmental impact during post-disposal, which is in contrast to the ecological threat

posed by fossil-based plastics (Borrelle *et al.* 2020; Wojnowska-Baryła *et al.* 2020). Nonetheless, evaluating the sustainability of the product is a complex procedure and heavily depends on the technology and processes that are utilized as well as the type of biomass that is converted (Yang *et al.* 2021).

All-cellulose composites (ACCs), prepared through diverse technologies and solvent systems from cellulosic raw materials, are frequently lauded as environmentally benign owing to their renewable origins and their facilitation of recycling on account of their single polymer nature (Matabola *et al.* 2009; Ma *et al.* 2011). In ACCs, the matrix and reinforcement phases exhibit exceptional compatibility, thereby enabling effective stress transfer and adhesion at their interfaces (Yousefi *et al.* 2013). Furthermore, ACCs are noted for their low density and superior mechanical and thermal characteristics, which are attributes related to the robust crystalline cellulose structures and elevated thermal degradation thresholds (Karu and Kaup 2002; Nishino *et al.* 2004; Gildl-Almutter *et al.* 2012; Huber *et al.* 2012; Uusi-Tarkka *et al.* 2023).

However, ACCs have a few drawbacks. As with all bio-based materials, moisture absorption is a primary concern, and this leads to dimensional instability, decreased mechanical properties, and susceptibility to mold and decay fungi (Singh *et al.* 1996; Schirp and Wolcott 2005; Dhakal *et al.* 2007; Gardner *et al.* 2008; Alamri and Low 2012; Cai 2020; Lopez 2020). These limitations have spurred the development of surface treatments designed to enhance the hydrophobicity of cellulosic materials, thereby extending their service life. In the pursuit of waste minimization and reduction of toxic residues, sustainable bio-based coatings are favored. Notable among these are birch-derived compounds such as suberin acids and betulin, which can be used as surface treatments to bolster the protective capabilities of cellulose-based materials. Suberin is a complex structural polymer containing fatty acids, glycerol, and a few aromatics found in plant cell walls (Korpinen *et al.* 2019; Serra and Geldner 2022). This macromolecule is responsible for the development of the cell wall barrier and controls the fluxes of gases, water, and solutes as well as protects plants from biotic and abiotic stress (Pollard *et al.* 2008). Betulin is a triterpenoid found in the bark of birch trees and exhibits a broad spectrum of protective and antibacterial properties (Król *et al.* 2015; Demets *et al.* 2022). Being a rich source of high-value chemicals, birch bark has garnered great interest in forest biorefineries (Normand *et al.* 2014).

There has also been increased interest in utilizing lignocellulosic raw materials—such as pulp, plant fibers, and regenerated fibers—as a resource for the composite industry. Recently, wood-based Spinnova-Lyocell fabric was successfully used as raw material for the fabrication of ACC (Uusi-Tarkka *et al.* 2022, 2023). However, there are no published results regarding ACCs composed of alternating layers of woven Lyocell fabric and non-woven Spinnova fabric. This paper reports the production of an ACC composite structured with needle-punched fabric, which is created from a combination of woven Lyocell fabric and nonwoven Spinnova fabric. It is anticipated that this approach will lead to further tailoring of the properties and performance of the ACC composite. The objective of this study was to characterize the mechanical performance (tensile, impact, and flexural properties) of the obtained ACCs produced using the NaOH-urea solvent system. Furthermore, the use of birch-derived surface coating agents to improve the hydrophobicity of ACC was investigated. The study utilizes both commercially sourced suberin and betulin coatings as well as a novel birch bark-derived pressurized hot ethanol (PHE) extract. The PHE extract is a mixture of birch bark triterpenoids (particularly betulin) and suberin fatty acids (Zhao *et al.* 2020).

EXPERIMENTAL

Production of Cellulose Fabric

Two different fabric structures were produced and labeled "thin" and "thick" ACC. The thin fabric was fabricated with five layers of nonwoven Spinnova web, with a surface weight of 100 g/m^2 and four layers of woven Lyocell fabric with a surface weight of 128 g/m^2 in alternating order. The thick fabric structure was fabricated in a similar manner, with nine layers of nonwoven Spinnova web and eight woven Lyocell fabric placed in alternating order (Table 1). The nonwoven Spinnova web was made from Spinnova fiber (Spinnova Ltd, Jyväskylä, Finland) with a linear density of 3 dtex and 60 mm fiber length in a Mesdan 337A carding machine. The Spinnova fibers are man-made pulp cellulose fibers, which are mechanically refined but not regenerated. The Lyocell fabric was a plain weave fabric. The warp density is 34 yarn/cm, and the weft density is 32 yarn/cm. The yarn was ring-spun with a linear density of 170 dtex and 780 Twist/m, consisting of fiber 1.3 dtex, 38 mm. The Lyocell fabric was obtained from Lenzing AG, Austria.

The targeted nonwoven/woven fabric ratio was aimed to be 50:50; however, the final ratio was approximately 55:45, with the woven Lyocell fabric being in excess. This difference was apparently due to fiber loss from the nonwoven web while processing. The nonwoven web was uneven from the edges, and fibers were lost during the process; therefore, the ratio of 55 wt.% of Lyocell fabric and 45 wt.% of Spinnova fiber can be found in ACCs. The assembled layers were attached together using needle punching (James Hunter Machine Co, Boston, MA, The United States). The fabrics and webs went through the machine once from each side (the fabric was needle punched from the two sides); the thick fabric was needle-punched twice on each side. The machine yields a stitch density of 78 stitches/cm^2 in each pass.

Fabrication of the All-cellulose Composite

Sodium hydroxide (12 wt.%) and urea (7 wt.%), both supplied by Sigma-Aldrich (Darmstadt, Germany), were mixed with water to create the cellulose dissolving solvent. This solution was chilled to -12 °C and continuously stirred before use. The prepared nonwoven-woven thin and thick fabrics were cut into 100 × 100 mm sheets and dipped in tap water to wet the materials. The sheets were then immersed in the NaOH-urea solvent system for two seconds and placed on an even surface for one minute. After letting the solvent penetrate and let material to react with the solvent, the sheets were carefully pressed on an even surface to allow the solvent to spread evenly throughout the material. Subsequently, the sheets were immersed in water buckets for 72 h, during which the water was changed multiple times until the pH level of the material was neutral.

The sheets were first pressed once at room temperature to remove excess water before forming the composite laminate on a hot press adapted from the procedure presented in Uusi-Tarkka (2022, 2023) and a series of pre-studies. The nonwoven-woven fabrics were pressed (10 bar) at 120 °C according to the following schedule: 3 × 5 seconds + 3 × 1 minute + 1 × 5 min + 1 × 10 min, with a 20-second break to release the steam after each interval. After the hot press procedure, the newly formed ACC laminates were placed under weight (1000 g) at room temperature for 24 h in order to retain their flat structure. The newly produced materials were weighed at room temperature (21 °C) (Table 1).

Table 1. Characteristics of the Utilized Fabric Structures and the Produced ACCs

	Number of plies	Weight class (g/m^2)	Thickness (mm)	Density (g/cm^3)
ACC thin	4 + 5 (Lyocell/ Spinnova web	1000	1.5	0.67
ACC thick	8 + 9 (Lyocell/ Spinnova web	2000	2.8	0.71

Production of Birch Bark Pressurized Hot Ethanol Extract

Pressurized hot ethanol (PHE) extraction of birch bark was performed in a custom-built high-pressure batch reactor (Parr Series 4584 reactor; Parr Instrument Company Ltd, Moline IL, USA) with a reactor volume of 5.5 L. The PHE extraction was done using technical grade ethanol (ETAX B 92.4 wt.%, ALTIA Oyj, Rajamäki, Finland) as the solvent. The pH of the extraction solution was adjusted to pH = 12, with a 25% ammonia solution (Suprapur, Merck KGaA, Darmstadt, Germany). A mechanically separated birch bark from Mondi Powerflute Ltd. (Kuopio, Finland) was used for the extraction.

The extraction process was performed in two steps: first, 150 g of dried and ground birch bark was extracted in 1 L ethanol/ammonium hydroxide (EtOH-NH_4OH) solution for 5 h at 120 °C. After overnight cooling to 20 °C, the obtained liquid and solid fractions were separated using suction filtration. In the second step, the solid residue from the first step was extracted and separated again in accordance with the same procedure. The solvents utilized were removed by evaporation before its use. Moreover, only the PHE extract from the second step was utilized in the surface treatment experiments.

Surface Treatment of ACCs

ACC laminates were placed in an oven at 105 °C for 20 h prior to the surface treatments to investigate the weight gain of the samples and amount of immersed surface agents. Commercial suberin acid mixture (Suberinno™) and betulin (Betuinno™) from Innomost Ltd. (Kokkola, Finland) and the prepared birch bark PHE extract solution were diluted into ethanol in the ratio 1:200; the pH values were adjusted to pH = 10 with 25% ammonia solution. Thereafter, the thin ACC laminates (20 mm × 10 mm) were dipped in a small bowl with different surface treatments for one second. Five laminates were treated for each surface treatment agent. The specimens were placed in an oven at 105 °C for 20 h. Only the thin ACC laminates were surface-treated.

Table 2. Coating Agents, Dilution Rations, and Concentrations Utilized

	Dilution Ratio	Concentration of the Substance (mg/cm^2)
Betulin	1:200	0.87
Suberin	1:200	0.87
PHE extract	1:200	0.67

Mechanical Testing

Tensile testing was performed using Zwick/Roell 050 universal tester (Ulm-Einsingen, Germany) using laser cut specimens of the size 100 mm x 10 mm. Test speed was set to be 2 mm/min until failure, and a load cell of 2.5 kN was connected to the machine. Each ACC laminate was measured using nine replicate specimens. The Charpy impact testing was performed according to the ISO 179 standard using a Zwick instrument (Ulm-Einsingen, Germany) with a 5 J pendulum arm. The specimens were un-notched, and a minimum of six replicates were tested for each ACC laminate both flatwise and edgewise.

Three-point flexural testing was performed according to the USI 14125 standard using Tinius Olsen H10KT universal machine (Horsham, PA, United States) with a loading rate of 10 mm/min and a 2.5 kN load cell. Eight flat replicate specimens (80 × 15 mm) were utilized for each ACC laminate; the specimens were laser cut.

Statistical evaluation of the data from the mechanical testing was completed using one-way analysis of variance (ANOVA) and Tukey-Kramer post hoc analysis, with a reference level of $p < 0.05$ suggesting a significant difference. Tests were performed with six to nine replicates per sample. All data were plotted with OriginPro 2024 software (OriginLab, USA).

Contact Angle Measurements

Contact angle measurements were performed on a drop-shaped analyzer—DSA25 by Krüss, (Hamburg, Germany). The drop size was set to 2 µL and at least eight replicate measurements were performed for each ACC laminate. The analyzer automatically collected three readings each second, and the mean values were calculated. The evolution of the contact angle was recorded for 60 seconds and contact angles of 1 s, 15 s, 30 s, 45 s, and 60 s were utilized in this study.

Scanning Electron Microscopy (SEM)

Hitachi S-4800 (Hitachi, Tokyo, Japan) was used to capture SEM images. To prevent charging of the specimen surface, the specimens were gold coated (2 nm) using a Cressington sputter coater 208 (Watford, the United Kingdom).

Microcomputed Tomography (Micro-CT)

X-ray micro computed tomography (Micro-CT) imaging of the dry nonwoven-woven fabrics and the ACCs was performed with Nikon XT H 225 (Nikon Metrology NV) micro-CT X-ray tomography device at room temperature. The sample was placed on a holder and imaged with 85 kV (peak) voltage, image pixel size of 10 µm, and 4476–7200 projections. Further, 16-bit .tiff images were converted to 8-bit .bmp images, binarized, and analyzed in 3D with CT Analyzer (Bruker Belgium N.V.).

RESULTS AND DISCUSSION

Tensile Strength, Elongation, and Young's Modulus

The tensile strength result indicates that there was no significant difference between the thin (8.26 MPa) and thick ACCs (7.79 MPa) when comparing the results of nine replicate samples. Recently, woven Lyocell-Spinnova ACC laminates obtained a tensile strength of 28.6 MPa (Uusi-Tarkka *et al.* 2022) and 35 MPa (Uusi-Tarkka *et al.* 2023). The main difference between these ACCs and those fabricated in this study was the type of reinforcement. In the present study, the ACC was partially composed of fabrics manufactured from short fibers, which do not yield the same mechanical properties as woven fabric manufactured by using yarns interlaced into a two-dimensional architecture. The process of ACC dissolution also yields dissimilar material structure that may have significant impact on the observed tensile properties, causing more variance on the tensile strength of thin ACCs. The tensile strength of the nonwoven fabric relies on the orientation and distribution of the short fibers, which are held together only by entanglements created during the needle-punching of the fabric (Russel 2007; Tausif *et al.* 2017). Such material

is also more heterogeneous in structure. One method for increasing the tensile strength in composites made from nonwoven reinforcements is to increase the needling density, which can improve structural integrity (Tausif *et al.* 2017). However, excessive needling can also cause holes and fiber breakage, which may have negative effects on mechanical properties (Hearle and Purdy 1971).

Table 3. Tensile Strength, Strain, and Young's Modulus Values for Thin (1.5 mm) and Thick ACCs (2.8 mm)

	Tensile strength (MPa)	Standard error	Strain %	Standard error	Young's modulus (GPa)	Standard error
Thin ACC	8.26	0.78	1.04	0.15	0.48	0.045
Thick ACC	7.79	0.29	2.18	0.16	0.69	0.023

Elongation values were measured to be 2.18% and 1.04% for the thick and thin ACCs, respectively ($N = 9$, $p < 0.05$). The thick ACC was able to stretch and prevent breakage better, resulting from a higher number of fiber-fiber interactions and surface area that contributed to higher strain at break, thereby resulting in greater elongation. This result is in keeping with previous studies in which ACCs have been produced using the same dissolution method (Piltonen *et al.* 2016; Uusi-Tarkka *et al.* 2022). Much higher elongation values, 11% to 14%, have also been reported with woven ACC laminates (Chen *et al.* 2020; Uusi-Tarkka *et al.* 2023). The smaller elongation values with nonwoven-woven ACCs are likely the result of a lower degree of bonding among the short fibers. The observed elongation results resembled, or even were lower than those of a paperboard (Yokoyama *et al.* 2007; Tervahartiala *et al.* 2018) and some grades of paper (Zeng *et al.* 2013) rather than the values conventionally seen in nonwoven biocomposites (Anuar *et al.* 2018).

Young's modulus of nonwoven-woven ACCs followed a similar trend as elongation and revealed a significant difference ($p < 0.05$) between thin and thick ACCs—0.48 GPa and 0.69 GPa, respectively. Adak and Mukhopahyay (2016) created Lyocell fabric-based ACCs using different compression pressures. In their study, Young's modulus of 1.78 GPa was obtained when the same compression pressure (10 bar) was used as that in the present study. At a five-bar pressure, they received a Young's modulus of approximately 0.6 GPa, which was on the same level as that obtained for the thick ACC in our study. It was also observed that the Young's modulus was highly dependent on void content, and the higher pressure increased the ACC's compactness and improved the fiber-matrix adhesion (Adak and Mukhopahyay 2016). The nonwoven-woven ACCs fabricated in this study most likely had high porosity, which contributed in part to a low Young's modulus.

It is also rather likely that the cellulose dissolution method utilized in this study was not able to compete with the effectiveness of ionic liquids. It has been reported earlier that the NaOH-urea solvent has limited capacity to dissolve cellulose with a high degree of polymerization (Jin *et al.* 2007), which can result in an insufficient matrix fraction. A low proportion of regenerated matrix phase can be a cause for low stiffness and diminished mechanical properties of the ACC laminates (Huber *et al.* 2012; Dormanns 2016).

Impact Strength

Charpy impact strength exhibits the energy absorbed by the specimen during a fracture. In this study, the impact strength values were lower for the flatwise specimens compared to the edgewise specimens (Fig. 1). In addition, the thick ACCs had higher impact strength compared to the thin ACCs, which is most likely due to higher mass and density in thick ACCs. Prior studies have revealed that higher density can positively influence impact strength and resistance to puncture (Komorek *et al.* 2022). Lee and Kang (2000) demonstrated that maximum impact resistance properties were achieved with a modest needle-punching density, which was 30 penetrations/cm^2 in their study. Excessive punching density deteriorates sample impact resistance due to the damage to fibers, decreased thickness, and easier crack initiation, which may have also influenced the ACC specimen in the current study.

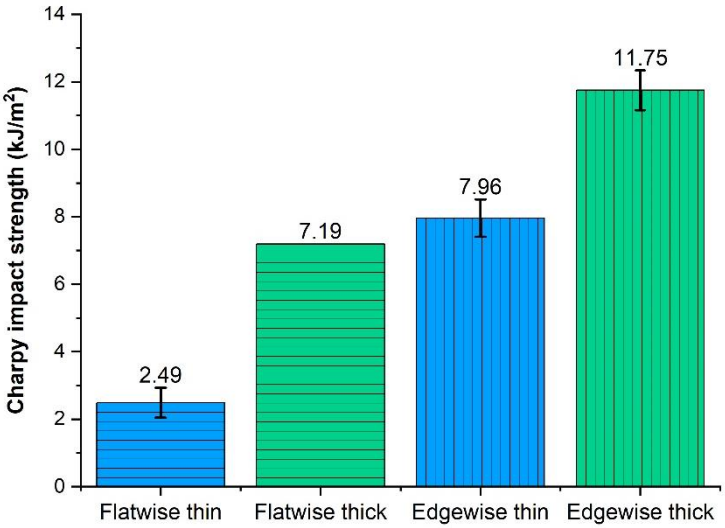

Fig. 1. Charpy impact strength for thin and thick ACCs measured as flat and edgewise. Error bars signify the standard deviation within the group (N = 7).

Flexural Properties

Flexural stress (MPa) and strain (%) were 4.82 MPa and 1.88%, respectively, for the thin ACC, and 6.70 MPa and 4.21%, respectively for the thick ACC (Figs. 2 a and b). These results are much lower compared to previously reported viscose textile ACC laminates with flexural strain of as high as 135.2 MPa, which also outperforms numerous commonly used biocomposites (Huber *et al.* 2013). Baghaei *et al.* (2020) reported flexural strength values of 29 to 34 MPa for ACC laminates, which were produced by using end-of-life textile as reinforcement. Long fibers and a woven structure offer higher strength and flexibility than short fibers. Moreover, regenerated fibers are characterized by a combination of strength and high strain, whereas plant-based fibers tend to be strong and stiff with low or moderate strain at failure (Mohanty *et al.* 2000; Adusumali *et al.* 2006). The Spinnova short fibers used in this study are man-made pulp cellulose fibers, which are mechanically refined but not regenerated, an aspect that also contributes to the test results. Bending resistance and flexural strain, as well as tensile properties, could be improved by adding a larger proportion of woven material in the nonwoven-woven structured composite (Skrifvars *et al.* 2019). The failure mode of materials was not investigated in detail as a part of this study.

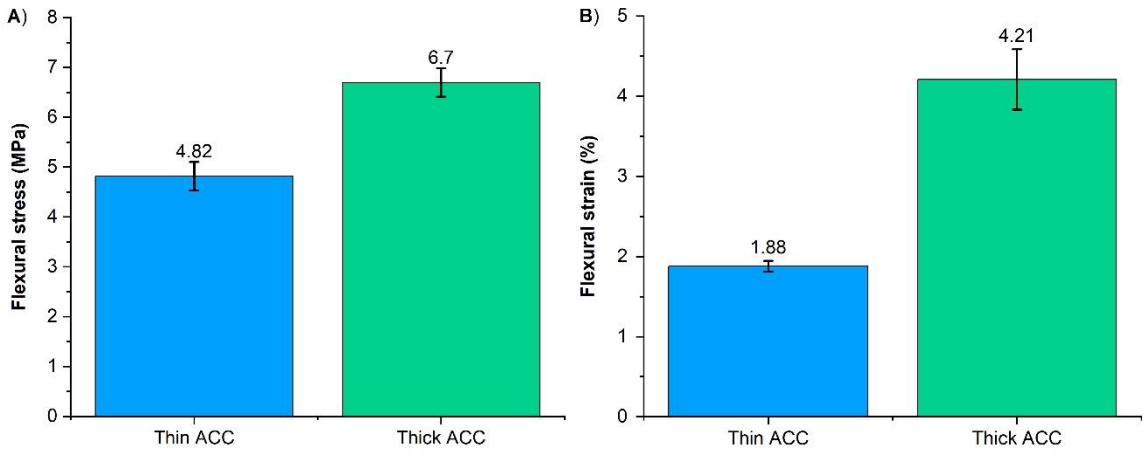

Fig. 2. a) Flexural stress (MPa), b) flexural strain (%) for the thin (1.5 mm) and thick (2.8 mm) non-woven ACC laminates. Error bars signify the standard deviation within the group (N = 6, $p < 0.05$).

Micro-CT Visualization and Porosity Assessment

Micro-computed tomography (micro-CT) is used to internally analyze materials non-destructively and can identify porosity and the type of voids. The results of this study revealed that the dry fabrics (thin and thick nonwoven-woven fabric) had higher porosity (%) and higher pore surface area (mm^2/mm^3) compared to the obtained ACCs (Table 4). It was also evident that porosity was higher for thin materials, which corresponds to higher density for the thick materials. However, the average pore size (mm^3) was higher for ACCs compared to nonwoven-woven fabrics. The main reason for this result is that there were abundant small sized pores (pore count) found in nonwoven samples, which affected the average value of the pore volume (Fig. 3).

Micro-CT results also concluded that there was no considerable difference between the average pore sphericity (shape factor) between the samples. Fig. 4 a) reveals the irregular morphology and void content in the thin ACC sample. Plies of layered woven and nonwoven fabric are evident in Fig. 4 b). During the tensile testing, there was a delamination of the assembled fabric layers, since the needle-punched nonwoven layers could not hold the plies together when stress was applied. This does not come as a surprise, as the weak bonding between the layers was revealed in the micro-CT image (Fig. 4 b).

Table 4. Characteristics of Porosity for Thin and Thick Lyocell-Spinnova Nonwoven-Woven Fabrics Prior Dissolution Treatment, and for Thin and Thick ACC Laminates After Partial Dissolution and Hot-Pressing

Pore characteristics	Thin nonwoven-woven	Thick nonwoven-woven	Thin ACC	Thick ACC
Porosity (%)	16.6	14.5	16.1	13.7
Pore surface area (mm^2/mm^3)	19.26	17.22	16.83	14.27
Average pore volume (mm^3)	$6.2{\cdot}10^{-4} \pm 1.9{\cdot}10^{-6}$	$5.5{\cdot}10^{-4} \pm 1.6{\cdot}10^{-6}$	$8.8{\cdot}10^{-4} \pm 6.0{\cdot}10^{-6}$	$8.0{\cdot}10^{-4} \pm 4.9{\cdot}10^{-6}$
Average pore sphericity (-)	0.37 ± 0.00	0.38 ± 0.00	0.38 ± 0.00	0.38 ± 0.00

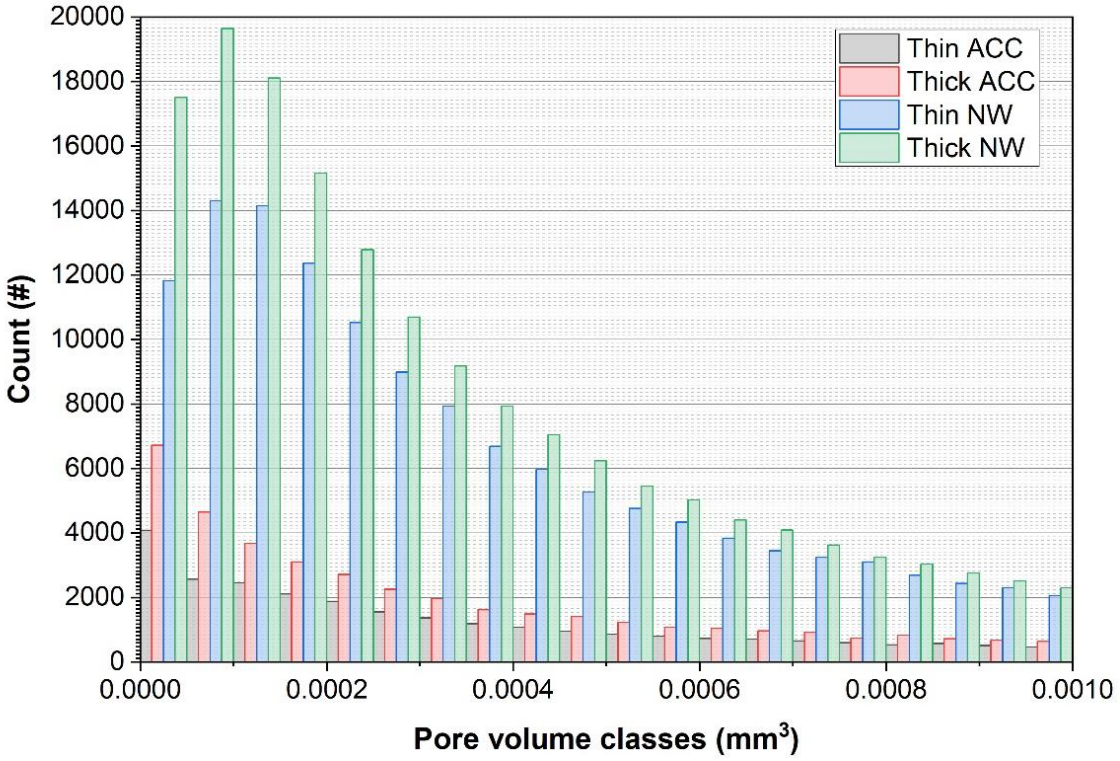

Fig. 3. Quantity of micro-CT analyzed pores for thin and thick Lyocell-Spinnova nonwoven-woven fabrics and for thin and thick ACC laminates

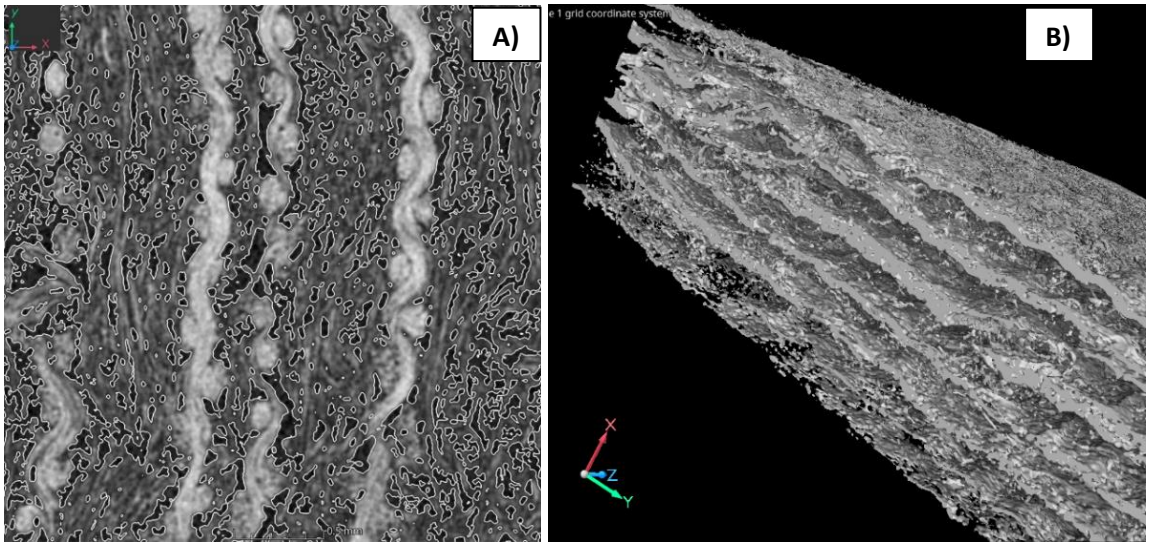

Fig. 4. Micro-CT visualization of the layered structures of a) thin ACC (1.5 mm) and b) thick ACC (2.8 mm)

Contact Angle Measurements on Surface-Modified ACCs

Surface modifications were applied by using suberin fatty acid mixture, betulin, and the PHE extract of birch bark. The results of the contact angle measurements are presented for the following times: 1 s, 15 s, 30 s, 45 s, and 60 s. All surface treatments significantly increased the contact angle of ACCs, since untreated ACCs immediately absorbed water and yielded a contact angle value of 0°.

In the beginning of the contact angle measurement (1s), betulin obtained the highest value, 128°; however, it also decreased rapidly during the experiment period and had the second broadest standard deviation range at the end (60 s) of measurement (Fig. 5). The highest contact angles were determined to be 104° and 110° for the suberin fatty acid mixture and PHE extract, respectively. The measured contact angles revealed that the PHE extract had the most consistent performance, ranging from 110° to 109°. Therefore, when the level and duration of hydrophobic performance was combined, the PHE extract appeared to have the best properties of the three.

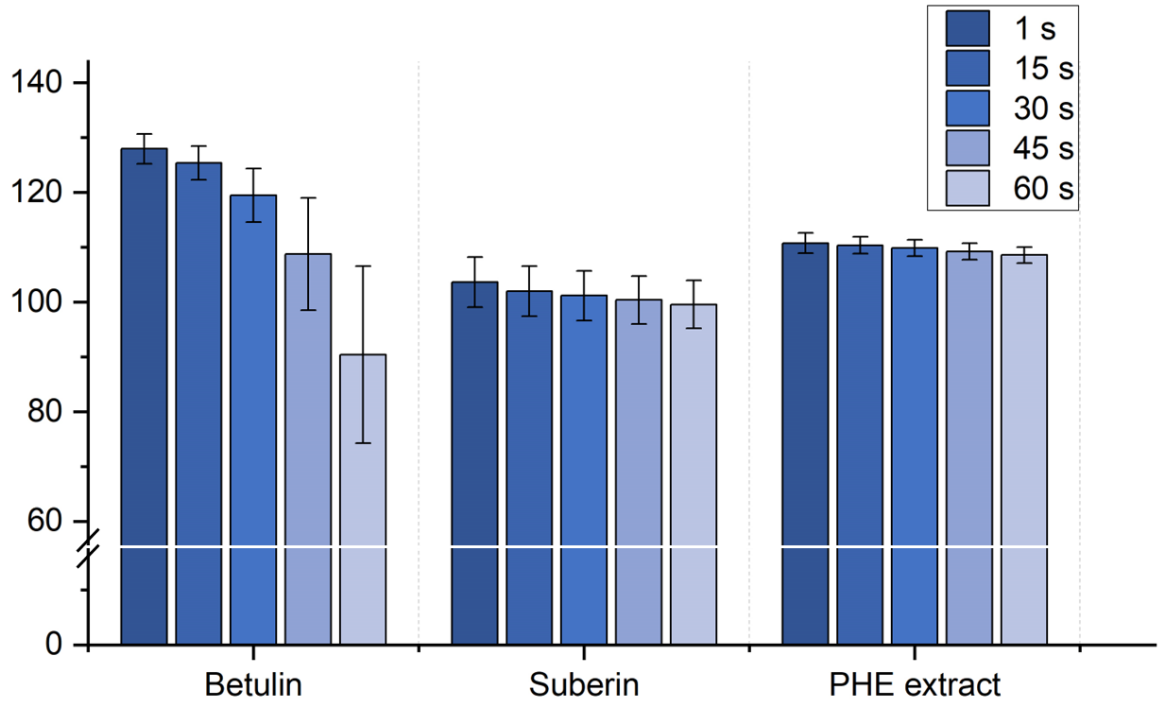

Fig. 5. Contact angle values for the time periods ranging from 1 second–60 seconds (N = 30). Time series for suberin and PHE extract revealed no statistically significant deterioration of performance from 1 second–60 seconds; moreover, the time series for betulin-coated ACCs revealed a significant difference ($p < 0.05$) only between the sampling at 1 second and 60 seconds and at the droplet evolution at 15 seconds and 60 seconds.

Scanning Electron Microscopy (SEM)

The surface of the modified ACC laminates and untreated ACC were examined using SEM (Figs. 6 and 7). The disoriented Spinnova fiber network was particularly evident in untreated ACC. Applied treatments covered the surfaces, and a strong consolidation of the fiber structure appeared to occur on the surface of suberin and, to a lesser extent, with ACCs treated with PHE extract. Small particle-like structures were evident on the surface of betulin-treated ACC. Solid crystal structures were also evident in Fig. 8b. This crystallization most likely occurred during the drying process in the presence of unevaporated ethanol. Heterogenous coating pattern on betulin likely explains why hydrophobicity decreased rapidly during contact angle measurements. The ACC treated with PHE extract included portions of betulin and suberin, and the surface morphology of both was smoother than in uncoated reference ACC specimen (see Figs. 6 and 7).

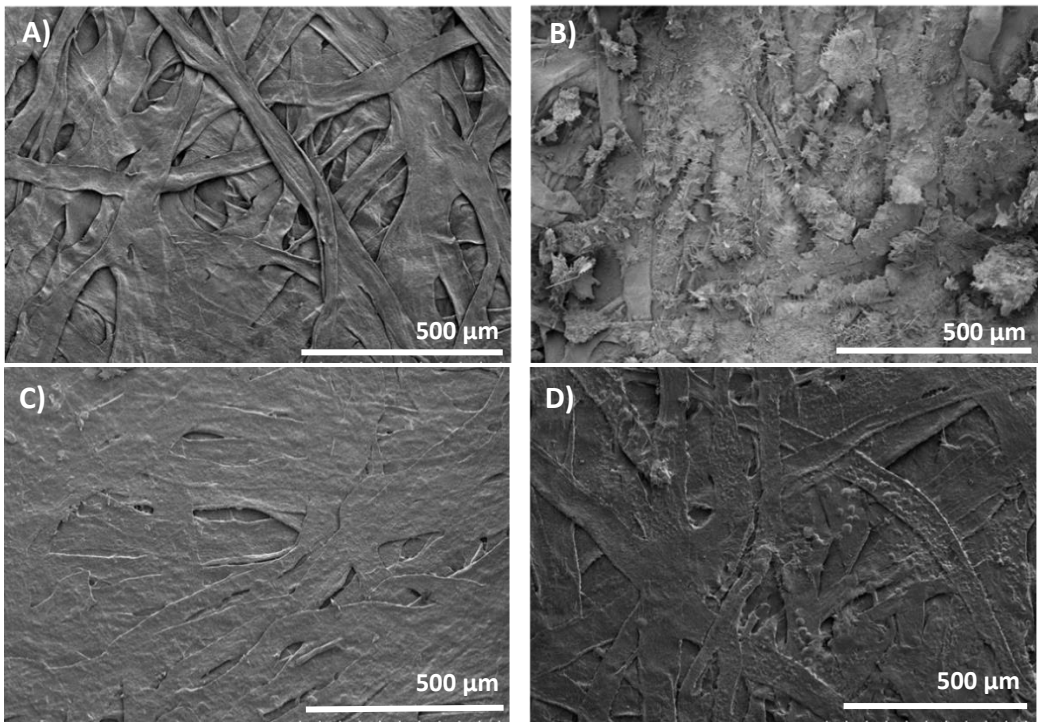

Fig. 6. SEM pictures of the surfaces of ACCs at 100× magnification; a) uncoated ACC, b) betulin-coated ACC, c) suberin-coated ACC, d) PHE-coated ACC

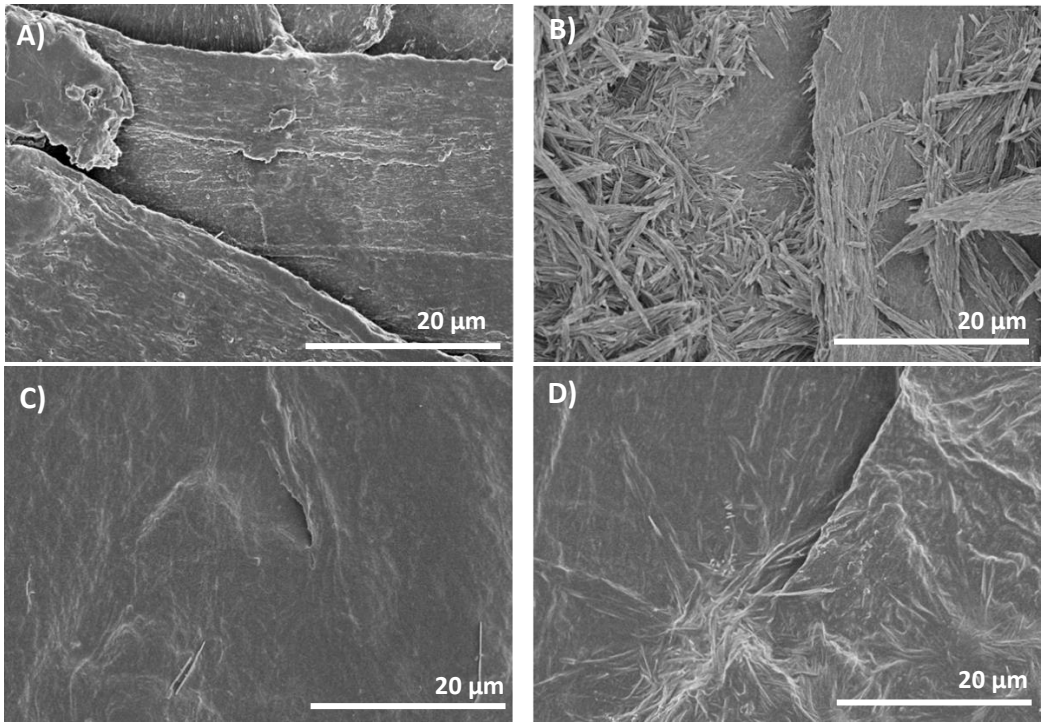

Fig 7. SEM pictures of the surfaces of ACCs at 2500× magnification; a) uncoated ACC, b) betulin-coated ACC, c) suberin-coated ACC, d) PHE-coated ACC

CONCLUSIONS

1. In this study, a Spinnova-Lyocell fabric structured all-cellulose composite (ACC) was developed. The study provides a good foundation for further research and development of nonwoven-woven ACCs.
2. The mechanical testing (tensile, impact, and flexural) revealed that the nonwoven-woven ACCs produced in this study had only modest mechanical properties. The thick ACC performed better than the thin ACC, which was attributed to the higher density and lower pore volume (mm^3) of the thick ACC. A reduction in porosity can increase the compactness of structure and improve the fiber–matrix adhesion of ACCs.
3. The water resistance properties of highly hydrophilic ACCs were dramatically upgraded with birch-based coatings—for example, a commercially available birch bark betulin, suberin acid mixture, and pressurized hot ethanol (PHE) extract of birch bark. All tested surface agents had a positive effect on creating a moisture barrier on the surface of the composites. The highest contact angle of 128° was measured with betulin-coated ACCs, and the best stable hydrophobicity calculated a minute after the beginning of the experiment were observed with the PHE extract.

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