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Design for Circularity: Lignocellulose-based Thermoplastics

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Abstract booklet
Oral presentations

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The Role of Forests in the Transition to a Circular Society

The bioeconomy had its hype within the political sphere almost two decades ago. Since then, the critics have gained momentum, and we see several initiatives focusing rather on how to restrict the use of biobased materials from forests than on promoting such use.

Examples of this is the EU strategy on forestry, the discussion and agreements on the LULUCF regulation, the updated renewable energy directive, maybe also the packaging and packaging waste regulation.

What went wrong – if wrong it was – and how should supporters of the bioeconomy proceed in the current landscape? Where will biobased materials be used in the future and how will society perceive them?

To the first question, what went wrongt, there are many answers. Some would argue that it's based on a lack of knowledge and understanding from the decision makers, others would say the honeymoon phase just transitioned into a more realistic view on the relationship between society and the forestry sector. Many would blame Frans Timmermans, former Climate Commissioner of the EU Commission, and say it's all because they don't have Nordic volumes of forests in the Netherlands.

One truth is at least that there are challenges in relation to the bioeconomy, which the industry didn't want to talk about in the beginning, but which now can't be avoided. Like the fact that you can't replace all fossil fuels with biomass, since the shear volume of human consumption is unsustainable. Like the yet-not-solved challenge of bio diversity, like the lack of circularity of some biobased products. Like the fact that bioenergy means the carbon is emitted immediately while a new tree takes time to grow.

I am a supporter of the bioeconomy, but I sincerely believe these challenges must be tackled, both in words and in action, if the bioeconomy is to be accepted as a positive solution to climate change, if it is to take the role that it should have in our society.

And I know there is progress, I am sure it will be made totally clear during these days. New better solutions need to be swifttly adopted, new ways of producing long-lasting materials will solve some conflicts, resource efficiency is crucial and not least – circularity is a key to making biobased materials an important part of a sustainable future.

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Degradation of lignocellulose-derived materials

Biobased materials can have environmental benefits with respect to fossil-based materials, however, to make a real difference our materials also need to have managed and sustainable end-of-life. For some applications, biodegradation in compost or other targeted environments is an optimum end-of-life option. Biopolymers, like cellulose, are inherently biodegradable, but sensitive relationship exist between susceptibility to degradation and chemical modification for obtainment of desired material properties and processability [1,2]. In our recent work we developed materials with enhanced (bio)degradability through degradation promoting additives, such as embedded enzymes [3] or carbon dots [4] or by incorporation of dynamic covalent bonds [5]. As an example, we demonstrated the effectiveness of embedded enzymes in triggering the degradation of cellulose acetate under simulated composting conditions and in aqueous environments, while carbon dots were shown to effectively catalyse the deacetylation of cellulose acetate under simulated sunlight leading to regeneration of cellulose with inherent biodegradability. Currently, we are investigating how chemical modification of cellulose fibers or lignin influences the degradation process.

[1] N. B. Erdal, M. Hakkarainen, Biomacromolecules 23 (2022) 2713-2729
[2] N. Yadav, M. Hakkarainen, Chemosphere 265 (2021) 128731
[3] N. K. Kalita, M. Hakkarainen, Biomacromolecules 24 (2023) 3290−3303
[4] N. Yadav, K.H. Adolfsson, M. Hakkarainen, Biomacromolecules 22 (2021) 2211–2223
[5] S. Subramaniyan, et al., ACS Sustainable Chem. Eng. 11 (2023) 3451–3465

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On the development of novel cellulose derivatives for microplastic flocculation

Many studies have been evaluating the potential of cellulose as a suitable substitute for common synthetic polymers prevalent in industrial applications. However, cellulose manipulation is challenging mainly due to its poor solubility in common solvents and a lack of thermoplasticity. Addressing these limitations requires strategic chemical modifications to enhance both cellulose reactivity and boost its applicability. In this work, controlled modification strategies, specifically cationic and hydrophobic modifications, have been employed to achieve derivatives with varying degrees of substitution, molecular weight, and hydrophobicity, aiming at their use as novel bio-based flocculation agents for microplastic removal from wastewaters. The cationization process involved a dual step: initial oxidation with periodate followed by a reaction with a cationic agent (Girard T reagent). Simultaneously, hydrophobic modification of cellulose was pursued, utilizing renewable feedstocks (i.e., plant oils). Fatty acids were extracted from vegetable oils, and cellulose was esterified in methanol. The ecotoxicity of cationic-modified and hydrophobically-modified cellulose derivatives, each with varying substitution degrees (i.e., DS +0.3, +1, +1.8, and DS -1H), was evaluated in four representative species from different freshwater trophic levels. The obtained derivatives were also evaluated regarding their flocculation performance in effluents contaminated with microplastics by laser diffraction spectroscopy (LDS), optical and scanning electron microscopy. The biofloculants developed in this study were found to successfully aggregate and remove model microplastics from aqueous media. Overall, this work demonstrates that "greener" approaches based on bio-based flocculants can be promising solutions for removing microplastics from aqueous media thus contributing to minimize their potential negative effects on aquatic environments.

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Thermoplastic and Thermoset Biocomposites from Several Biopolymer Resins and Cellulose, Hemicellulose, and Lignin Additives

The shift towards sustainable materials has catalyzed significant innovation in the field of composite materials, particularly those integrating renewable biomass fibers. Among these, cellulose stands out as a highly promising biopolymer. Derived from plant fibers, cellulose boasts several properties that make it an excellent candidate for reinforcing materials. Its biodegradability and renewability align with the growing environmental consciousness, while its nanoscale manufacturability opens up diverse applications in various industries.

The summary of the presentation includes several case studies on vegetable oil acrylate nanocellulose composites, polybutylene succinate wood fiber composites and cellulose, hemicellulose, and lignin's plastics. The main conclusions of the ongoing research are presented below.

Chemical modification of cellulose significantly enhances the thermal, tensile, and thermomechanical properties of polybutylene succinate composites. Masterbatch preparation is a more sustainable method for producing PBS/nanocellulose composites than solution or direct melt mixing. The biodegradation of PBS/cellulose composites can be controlled by the type, modification route, and filler content of the cellulose.

In turn, the excellent processing and performance of acrylated oil /lignocellulose resin materials are enhanced by lignocellulose fillers, outperforming other bio-based acrylate resins and hybrids in mechanical, thermal, wetting, and durability properties. Ultra-low nanocellulose loads in resin ensure outstanding mechanical performance and long-term durability for 3D printing applications.

Nanocellulose from hemp and birch biomass can be also recombined with hemicellulose and lignin to create wood-like biopolymer papers, films, and foams. However, the structure of biocomposites can be adjusted by varying lignin and hemicellulose content, green chemical functionalization, and physical cross-linking. Varying the ratios of xylan, lignin, nanocellulose, citric acid crosslinkers, and dicumyl peroxide initiators can fine-tune the performance and biodegradation of the obtained biocomposite films. Bio-foams from xylan, lignin, and nanocellulose can be tailored from soft to rigid for various thermal applications.

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Water transfers in cellulose: bound water comes first!

In various processes, water or vapor flows through or is extracted from cellulosic materials. It appears that during such transfers the interaction of (free) water or vapor with bound water, i.e., absorbed in the amorphous regions between microfibrils, can play a major role on the dynamics. First, bound water alone can diffuse rapidly in the fibers, and finally over the whole fiber network, allowing a material with a porosity filled with oil to dry anyway [1]. For a standard air-filled system the water transfer relies on water vapor transport through the porosity, bound water transport in the fiber network, and exchange (sorption-desorption) between the two phases (see Fig.1a), which as a whole may be described by a simple diffusion process with a diffusion coefficient depending on saturation [2]. Then, for low porosity cellulose fiber stacks, bound water transport may be dominant [2].

This has an important implication on the long-term evolution of an aqueous droplet, possibly containing particles such as pigments and viruses or solute such as ions and polymers, and reaching the surface of a cellulosic sample. It is generally considered that such a droplet somewhat spreads, penetrates the structure, stabilizes and eventually dries. In fact, it may be shown [3] from NMR (nuclear magnetic resonance) relaxometry and MRI (magnetic resonance imaging) that, instead of drying, the water is absorbed in nanoscale pores in the cellulose fibers and diffuses throughout the entire structure (see Fig.1c). Thus, the initial (free) water rapidly disappears from the porosity (see Fig.1b), while the non-absorbed solute or particles remain stuck to the solid surfaces in the initial region of liquid penetration. Bound water also appears to play a major role when some free water penetrates or is extracted from a cellulose fiber stack. It for example ensures the transport of water up to the surface of the sample, even in the ultimate stages for very low free water concentration [4-5].

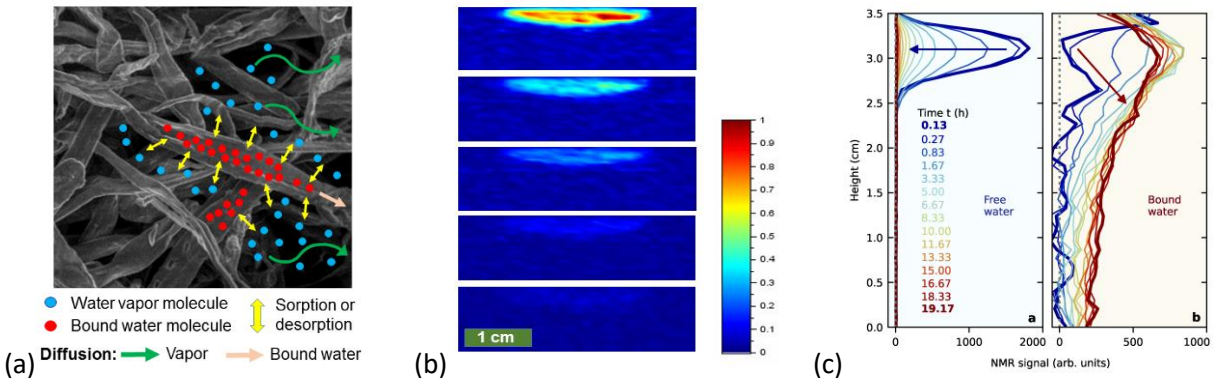

Figure 1: (a) The water transport and exchange processes in a cellulose fiber stack; (b) Evolution (as seen from successive MR images) of the free water from a droplet in cellulose over about 1 hour; (c) Evolution (from 1D MRI) of the free water (left) and bound water (left) distributions over time along sample axis.

References: [1] Y. Zou et al., *Cellulose*, 30, 7463–7478 (2023); [2] Y. Zou et al., *PNAS Nexus*, 3, pgad450 (2024) ; [3] L. Yan et al., submitted (2024) ; [4] M. Cocusse et al., *Science Advances*, 8, eabm7830 (2022) ; [5] N. Ben Abdelouahab et al., *Cellulose*, 28, 5321-5334 (2021)

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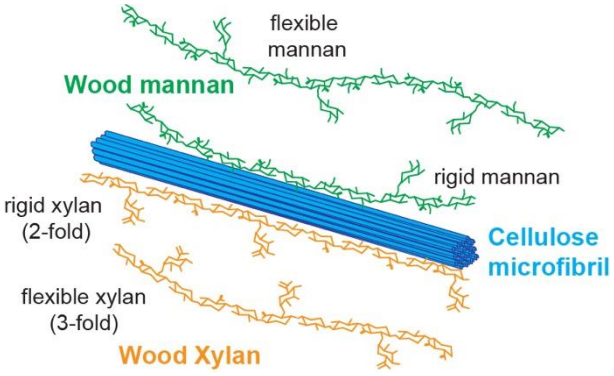
Hemicelluloses as the Janus "two-faced" components in wood modulating interactions with cellulose and lignin

Hemicelluloses are key components in lignocellulosic biomass, interlocking cellulose microfibrils and lignin. In hardwoods, acetylated glucuronoxylan is the main hemicellulose component with minor presence of acetylated glucomannan. In softwoods, contrarily, we find both acetylated galactoglucomannan and arabinoglucuronoxylan in significant amounts. In the last decade, fundamental progress has been made on elucidating the presence of regular repeating molecular motifs in wood xylans and mannans using glycomic mass spectrometric approaches. The occurrence of major intramolecular domains in the structure of hardwood and softwood xylans with even pattern of substitution is required for their conformational adaptation to cellulose surfaces [1,2]. Moreover, the occurrence of minor domains with clustered decorations might pay a role in modulating interaction with lignin through lignin-carbohydrate complexes (LCCs) [3,4]. This dual behaviour enables hemicelluloses to both assemble into rigid and flexible domains on the surface of cellulose fibres [5], modulating the mechanical properties and bridging simultaneously with lignin. However, there are still many unanswered questions on the role of hemicelluloses in the molecular architecture of woody tissues. In this presentation the latest developments on elucidating the structure of wood hemicelluloses will be presented, in relation to molecular models of the architecture of lignocellulosic fibres and their application for the development of lignocellulosic materials.



[1] N. J. Grantham, et. al. Nature Plants, 3(11), 859-865.
[2] A Martínez-Abad, et al. Plant Physiology, 175(4), 1579-1592.
[3] A Martínez-Abad, et al. Green Chemistry 20 (11), 2534-2546
[4] P. Sivan, et al. Carbohydrate Polymers, 122434.
[5] J Berglund, et al. Nature Communications 11 (1), 1-16

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Exploring cellulose carbonization pathways using reactive force field methods

Carbon materials are important for our everyday life, as they are essential components in a range of products from electrical devices to fiber reinforced plastics. Currently used carbon materials are almost solely produced from non-renewable precursors, and there is a growing need for sustainable alternatives. One of the candidates is cellulose, which is both renewable and abundantly available. Unfortunately, cellulose-based carbon materials remain notably inferior to fossil-based ones, largely because carbon structure formation in cellulose pyrolysis is not adequately understood.

In our ongoing work, we study the chemical and structural pathways of cellulose carbonization using atomistic simulations based on the ReaxFF reactive force field. We both evaluate the capability of the method to reproduce known reactions and products of cellulose pyrolysis, and predict reactions that lead to amorphous condensed phase structures at early stages of the carbonization process. Our modelling work is linked to an experimental campaign to understand the transformation of cellulose into an intermediate thermostable condensed phase and ultimately carbon [1, 2].

Our first modelling studies focus on the mechanism and kinetics of chain scission at high temperatures and heating rates, which are accessible using (unbiased) molecular dynamics (MD) simulations [3, 4]. Many of the predictions are compatible with mechanisms proposed for cellulose fast pyrolysis, but the absence of anhydrosugar forming reactions suggests that the high temperatures might hinder correspondence with practical conditions. To confirm that this is not a shortcoming of the force field, we are performing systematic force field evaluation for cellulose. Moreover, we study crosslinking reactions between cellulose chains at significantly lower temperatures using biased MD simulations. Simulations with cellulose only show no crosslinking, whereas cellulose and maltosan produce stable crosslinks, as does maltosan only. This suggests that levoglucosan end groups could play a role in the formation of an amorphous cross-linked network.

[1] Fliri, et al., J. Appl. Anal. Pyrol. 175 (2023) 106153
[2] Fliri, et al., J. Anal. Appl. Pyrol. 181 (2024) 106591
[3] A. Paajanen, et al., Cellulose, 24(7) (2017) 2713–2725
[4] A. Paajanen, et al., Cellulose 28(14) (2021) 8987–9005

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The role of lignin structure for swelling properties of wood: hierarchical multiscale modeling

Softwood branches develop compression wood (CW) in the lower and opposite wood (OW) in the upper part. These wood types differ in structure at several length scales. Distinctive differences are found at cell level (different cell geometry), cell wall level (CW lacks S3 layer and has a larger microfibril angle than OW) and at the level of chemical composition of the lignin matrix. While OW mostly contains guaiacyl (G) units, CW is known to contain a substantial fraction of 4-hydroxyphenyl (H) lignin.

In this study, the impact the lignin difference has on wood hygro-expansion is studied by the means of hierarchical modelling as support to macroscopic investigation of CW and OW hygro-expansion [1]. By using atomistic models and Molecular Dynamics computer simulations of lignin systems at different levels of hydration, swelling coefficients for the different lignin matrices are obtained and implemented in a Finite Element (FE) model of the cell wall, as illustrated in Figure 1.

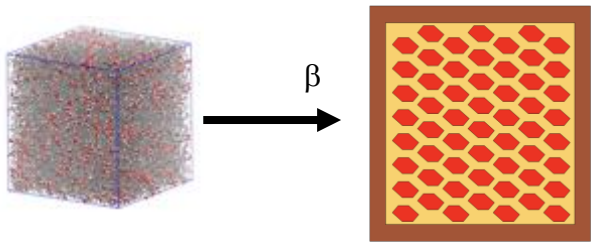

Figure 1: Schematic illustration of hierarchical modelling approach by obtaining swelling coefficient β from atomistic simulations (left) to use in FE model (right).

It was found that, despite the minor difference in chemical composition, there are differences in lignin swelling, structure and water dynamics. CW lignin is found to have a higher uniaxial swelling coefficient, since the phase separation between lignin and water is more pronounced. This behaviour is linked to structural differences, where intermolecular $\pi - \pi$ stacking is more common in CW lignin and hydrogen bonding to water more pronounced in OW lignin. The effect of these differences was further investigated at next hierarchical level as matrix swelling coefficients of lignin in a FE model.

[1] M. Hartwig-Nair, et al., *Wood Sci. Technol.,*(2024). doi.org/10.1007/s00226-024-01548-z

Wood Nanoscience and Nanotechnologies

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I will give an overview of our published work on nanotechnologies using cellulose nanomaterials, with a focus on assembly and functionalization strategies of **wood nanocellulose** aimed at specific properties, with an eye toward high impact applications including energy, electronics, building materials and water treatment, including nanomanufacturing and light management in transparent nanopaper for optoelectronics (as a replacement of plastics); mechanical properties of densely packed nanocellulose for lightweight structural materials (**super wood,** replacement of steel, Nature 2018); artificial tree for high-performance water desalination and solar steam generations; mesoporous, nano-ionic **wood thermoelectrics** (Nature Materials, 2019); radiation **cooling wood** (Science, 2019); **transparent wood**; **solid state batteries** (Nature 2021), and 3D moldable wood (Science 2021, Cover).

Bio: Liangbing Hu received his B.S. in physics from the University of Science and Technology of China (USTC) in 2002, where he worked on colossal magnetoresistance (CMR) materials for three years. He did his Ph.D. in at UCLA, focusing on carbon nanotube based nanoelectronics (2002-2007). In 2006, he joined Unidym Inc as a founding scientist. He has published over 450 research papers. Currently he is a professor at Yale University. He is also the Founder of Inventwood LLC. (www.inventwood.com).

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Future applications for lignin and LCCs

Lignin is an underutilized resource that is gaining increased interests. While lignosulphonates have been commercially utilized for long, the viable application of kraft lignin has been more challenging due to e.g. its poor solubility, low reactivity and heterogeneity. One emerging way of overcoming these challenges is the formation of lignin nanoparticles (LNPs). In this presentation we will present some recent results on applications of LNPs, lignin containing nanocellulose and lignin carbohydrate complexes (Figure 1). While unmodified LNPs are hydrophilic, they can form multifunctional hydrophobic and breathable coating layers on wood or textile surfaces either via esterification of the lignin prior to particle formation [1] or via epoxidation [2]. Interestingly, thin layers of esterified LNPs were sufficient to increase the water contact angle of pure cotton from 43 to 130°. However, if slightly thicker and continuous layers are used, low oxygen and water vapor transmission rates are achieved showing potential for application as barrier coatings. Water soluble lignin carbohydrate complexes (LCCs) also shows potential as barrier coatings [3].

Acetylation of lignin prior to particle formation enabled production of tiny LNPs. One monolayer of these particles formed a transparent, colourless and superhydrophilic layer applicable as antifogging or anti icing coatings. Layer-by-layer deposition together with cationic poly-L-lycin enabled to form fotonic coatings with bright colours ranging from blue, red and violet depending on the layer thickness of the coating [4]. As a final example we demonstrate the applicability of the particles for emulsion and foam stabilization enabling formation of solid, strong but still biodegradable films or solid foams together with cellulose nanofibrils [5].

Figure 1. AFM image of LNPs, examples of strong, water-resistant nanocomposite, multifunctional textile coating, transparent or photonic colours and foams using LNPs.

[1] Babaeipour et al Mater. Adv.5 (2024) 6114.
[2] Henn et al Appl Mat Interface 13 (2021) 34793.
[3] Asikanius et al Frontiers in Energy Res 9 (2021) 784.
[4] Henn et al Chem Eng J. 475 (2023) 145965.
[5] Zou et al Mater Adv. (2024)

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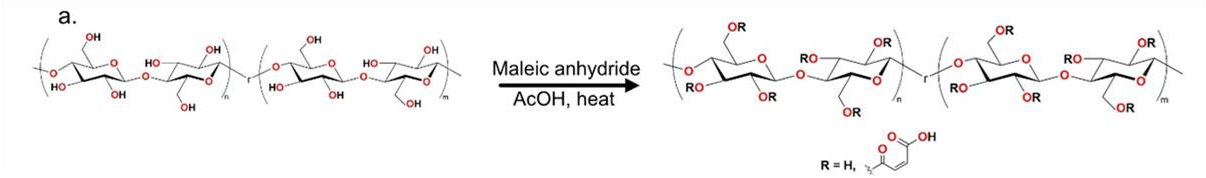
New routes for functionalization of cellulose-rich fibres

The chemical modification of delignified cellulose-rich fibres has a very long tradition and has received an enormous renewed interest following the development of nanocellulose materials. These bio-based, renewable and biodegradable nano-components have opened a new route for a bottom-up engineering of biobased materials with vast application areas. However, in order to fully liberate the fibrils from the fibres and to induce a colloid stability of the prepared fibrillar dispersions [1] it is necessary to oxidize the fibres to introduce charges to the cellulose. During the last decades numerous methods have been suggested to accomplish this but the drawback with many of these methods is that they are costly and the regeneration of the used chemicals is not simple and usually not available at large scale. This has indeed hampered the development of the application of nanocelluloses at large scale.

In a recent development [2,3] it has been found that the treatment of cellulose-rich fibres with maleic anhydride in acetic acid according to scheme 1, opens for a new way of efficiently modifying cellulose-rich fibres and for a green and cost-efficient preparation of cellulose nanofibrils (CNFs).



Scheme 1. Maleilation of cellulose-rich fibres by using maleic anhydride in acetic acid.

Apart from introducing carboxyl groups to the fibres, the modified fibres will also contain an alkene group allowing for a further modification of the fibres or the fibrils from the modified fibres. The degree of modification of the fibres can simply be tuned by changing time and temperature during the reaction and charges between 300 and 1000 μeq./g were easily achieved [3]. The charges will naturally cause a swelling of the fibre wall which enables for an efficient defibrillation of the fibres wall and for the preparation of colloidally stable CNF dispersions.

By using the modified *fibres*, without fibrillation, we have also been able to create strong hydrogels by polymerizing PEGs inside and outside the fibre wall creating an interpenetrating polymer network with the modified fibrils within the fibre wall and between the fibres [3]. Furthermore, the modified fibres have also been used as a substrate for polymerizing acrylate oligomers/polymers from the fibres allowing for the preparation of cellulose fibres with a total charge of around 7000 μeq./g which show excellent ability to remove contaminants from aqueous media. Finally, due to the chemical structure of the maleilated fibres the attached groups can be simply removed by an increase in pH of the modified fibres.

[1] Benselfelt, T. et al, ChemSusChem 2023, 16, e202201955
[2] Olsén, P. et al. Swedish Patent Application 2430053-5, 2024
[3] Cortes Ruiz, et al, Submitted for publication 2024 and PhD thesis, KTH 2024
[4] Atoufi, Z., PhD Thesis, KTH, 2024

By- or - Co-Products: Manure, hop straw, black liquor as source for fibres and polymers

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Agriculture and manufacturing processes often yield co- and by-products. We show that the utilisation of by- or co-products as raw materials could generate valuable products. Utilizing herbivore manure as source for cellulose outsources biomass collection to animals and disintegration of plant biomass into fibres occurs in their digestive system, which can reduce the energy input for fibre production. We will show that high biogas yields can be achieved using manure as substrate and that the remaining fermentation residue allows for effortless isolation of cellulose nanofibres, leading to a significant reduction of the environmental impact compared with traditional systems based on wood. The anaerobic fermentation of manure or hop straw residue enables the up-concentration of lignin on cellulose fibres, which after disintegration results in nanolignocellulose fibres with high lignin contents. Nanolignocellulose fibres are useful particulate emulsifiers. In contrast if wood is pulped to produce cellulose fibres lignin is separated and often used as fuel. We use black liquor as source for production of lignin-based resins which upon curing results in strong, stiff thermosetting resins for a range of applications.

Nanostructured lignocellulosic composites with multiple functions

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A stated goal for lignocellulosic materials is to replace synthetic plastics. For obvious reasons, all synthetic plastics cannot be replaced in all applications. It may instead be fruitful to ask: What are the unique characteristics of lignocellulosics which give them potential to replace fossil-based plastics and composites, and what are the shortcomings? The mechanical properties are often an advantage. The cellulose crystal has an axial modulus higher than 100 GPa at low density, due to high molecular orientation and dense packing. The composite nature and nanostructure of load-bearing secondary plant cell walls is another unique advantage [1]. In the secondary wood cell wall, the dominating organization of elementary cellulose fibrils is as parallel individual entities with SAXS and SANS correlation distances in the range 4-10 nm. This structural perfection is not practically possible to achieve in man-made nanocomposites. Instead, nanoparticle/nanofiber agglomeration, lack of preferred orientation and low reinforcement content severely limits their mechanical properties in large structures.

The nanostructure of lignocellulosic materials makes it possible to also add other functions than mechanical performance through nanoparticles, dyes, functional polymers etc [1]. Examples will be provided together with challenges associated with different starting substrates such as cellulose nanofibrils, pulp fibers and wood. Composites with high optical transmittance are one example where nanostructural perfection is important, and new wood material functions can be obtained. The use of a polymer matrix may impede eco-friendly characteristics, but a polymer matrix can certainly address the problem of moisture sensitivity in lignocellulosics.

Recently, there is also an interest in the role of lignin as an intrinsic polymer binder in lignocellulosic materials. There are strong arguments in favor of increased use of lignin-containing materials, and some research questions related to the molecular structure of processed lignin will be suggested.

[1] LA Berglund, I Burgert, Bioinspired Wood Nanotechnology for Functional Materials, *Adv. Mater.* (2018), 30, 1704285. https://doi.org/10.1002/adma.201704285

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Moisture induced activity of lignified cells and tissues

Fibrous plant cells are known for their intrinsic multifunctionality and activity which changes with time and environmental conditions. Despite developmental changes in living cells, dead cells can perform tasks autonomously, to actively respond to environmental triggers [1,2]. The most prominent trigger is water, determining swelling, mechanics, degradability or conductivity of the cell walls and consequently the tissues and materials at larger scales. This inherent activity is often seen as a disadvantage, especially for applications that require high reliability, such as in the building sector. Other drawbacks for building applications are the inherent high biological variability. While variable and changing properties are clear limitations for certain applications, they also offer the opportunity to use certain plant materials for new applications or to provide inspiration for the development of new materials. The challenge of exploiting material variability requires a dual approach: a deep understanding of the natural material and its role in the plant organism and for applications, and creative experimentation to break new ground. Here, new insights into the relationship between water, cell wall structure, swelling and mechanical properties are presented to illustrate the property space of secondary cell walls [3,4]. Functionality for the organism will be briefly addressed. Examples of design experiments to exploit the property space will be presented [5].

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[1] M. Eder, et al., Science. 543-547 (2018) 362

[2] M. Eder, et al., Advanced Materials. 2001412 (2021) 33

[3] N. Horbelt, et al., Wood Science and Technology. 1305-1318 (2021) 55

[4] T. Zhan et al., Wood Science and Technology. 1359–1377 (2021) 55

[5] C. Wenig et al., Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences 20200345 (2021) 379

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Synchrotron imaging with x-ray scattering or spectroscopy contrast for the characterization of thermoplastics

Scanning imaging with a focused X-ray beam allows to record a 2D scattering pattern or a energy spectrum spatially resolved in extended samples. Small- and Wide angle X-ray scattering (SAXS/WAXS) probes structures from several hundreds of nanometer down to the Ångström scale and provide valuable information on polymeric materials, such as local crystallinity and nanostructure sizes and orientation as illustrated in Figure 1.[1] Raster-scanning a macroscopic sample with a micrometer-sized beam, one can extract information from each scattering pattern to construct images with different contrasts. This allows inside into structures formed after thermoplastic molding, both for conventional [2,3] and cellulose-based polymers [4]. If instead an energy spectrum is probed, chemical information on elements (X-ray fluorescence, XRF) or more detailed information about the types of chemical bonding an element is involved (X-ray absorotion near-edge spectroscopy, XANES) can be obtained to a very high spatial resolution of a few tens of nanometers. In the case of ligno-cellulose fibers, sample preparation and the right embedding material is a challenge, but if solved lignin can be located on single cellulose fibers, and it opens up to study local modifications. In this talk an overview of these synchrotron scanning imaging methods will be given, together with examples focusing on thermoplastic materials.

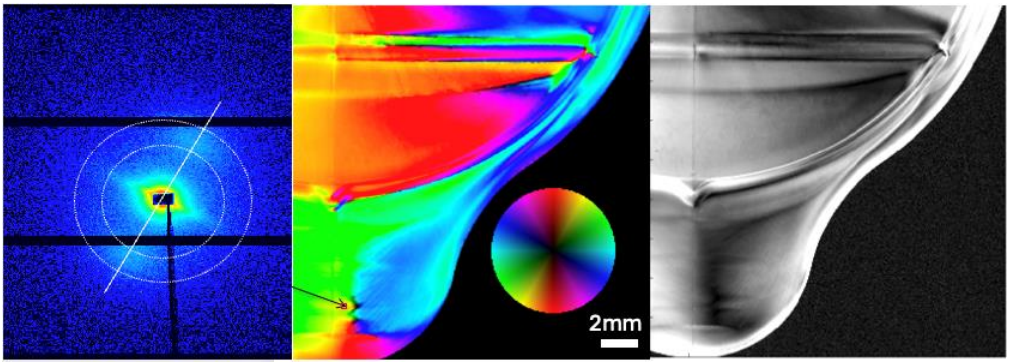

Figure 1: SAXS pattern (left) reveal structure and orientation of polyethylene in a injection molded packaging material. In scanning imaging a scattering pattern is recorded in each point, and information such as nanostructure orientation (middle) or degree of orientation (right) can be extracted in each pixel used to create images with different contrast.

[1] Bunk et al. New. J. Phys. 123016 (2009), 11
[2] Björn L. et al. ACS Appl. Polym. Mater. 4852-4864 (2024) 6
[3] Björn L. et al. ACS Appl. Polym. Mater. 6429-6440 (2023) 5
[4] Lo Re. G. et al. Chem. Eng. J. 141272, (2023), 458.

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Deciphering wood cell wall nanostructure and moisture behavior via x-ray scattering and modelling

X-ray and neutron scattering provide unique tools for characterizing the nanoscale structure of lignocellulosic fibers under various conditions. For instance, they allow observing changes of the nanostructure inside unmodified cell walls or *in situ* during different processes. Especially, they can be used to follow the interactions of the main structural components of the fibers, cellulose microfibrils, with moisture. These extraordinary capabilities of scattering methods come with the price of rather demanding data analysis in reciprocal space, which in the case of wood is further complicated by the heterogeneous hierarchical structure and the presence of multiple structural components.

Our recent collaborative works have used molecular modelling to support the analysis of X-ray scattering data from wood-based samples. With this combination, we have studied the molecular-level phenomena related to drying of wood, linking previously reported shifts in diffraction peak positions to aggregation-induced deformations of cellulose crystals [1]. We have further investigated the role of lignin in the moisture-induced swelling of microfibril bundles by conducting similar studies using delignified wood, and revealed same type of behavior as in untreated wood [2]. Moreover, the atomistic models have allowed us to improve the accuracy of the interpretation of X-ray scattering data measured from cellulosic samples [3].

To better address the heterogeneous structure of materials like wood, we have also been exploring the possibilities of machine learning to obtain nanostructural information specific to different tissue types in plant samples. This includes for instance utilizing clustering and regression algorithms to determine the average properties of each tissue type and to predict their characteristic scattering patterns.

[1] A. Paajanen et al., Nano Lett. 5143-5150 (2022) 22
[2] A. Zitting et al., Small Struct. (2024)
[3] A. Zitting et al., Cellulose 8107-8126 (2023) 30

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Synthetic Polymer-Free Fibreboards Derived from Oil Palm Waste

Empty fruit bunches (EFB) of oil palm are the abundant lignocellulosic residues from the palm oil mill process, which are commonly discarded as mulch on land or incinerated. In this work, we show a simple method mimicking paper-making process to upcycle EFB fibres into high-value boards without the need for formaldehyde binders. This was achieved by utilising cellulosic fibres from wood pulp as binders. The concept utilises the cellulose network from pulp fibres to hold the otherwise loose EFB fibres together to produce robust and rigid EFB boards. Mechanical refinement was performed using a re-circulating colloid mill to improve the binding performance of the pulp fibres. The results indicate that performing longer refining times to the pulp fibres leads to the production of denser EFB boards with enhanced mechanical properties, air resistance, and particle retention efficiency. This is because the refining stage applied to the pulp fibres induced fibrillation and the addition of fines, enhancing the contact area between the pulp fibres used as binders and the EFB fibres. Furthermore, EFB boards containing 30 wt% of pulp fibres refined for 30 minutes possessed better flexural properties (EF = ~2.9 GPa; σ F = ~22 MPa), compared to commercial particle board (PB) and medium density fibreboard (MDF) for furniture application. This work also shows that EFB boards can be moulded in special shapes either in never-dried state or after being formed and dried into panels, with a steaming step introduced to re-wet the boards. This capability presents good potential for furniture and packaging applications that require products with complex shapes. A lifecycle analysis model (LCA) using ReCiPe 2016 method was also developed to measure the environmental impact of upcycling EFB using this technique. The results showed that the EFB boards possessed lower environmental impact on global warming potential and all end-point categories (human health, ecosystem and resources) compared to the commercial MDF. This can be attributed to the absence of synthetic adhesives used as the binder and the avoidance of *business-as-usual* scenarios from EFB.

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Wettability of fatty acid esterified cellulose surfaces from molecular simulations

Surface functionalization treatments can be used to reduce the moisture sensitivity of cellulosic materials, which is particularly relevant for textile and packaging applications. One such approach is to use enzymatic methods to link plant-based lipids to cellulose surfaces. Structure-function relationship studies have shown evidence of long-chain fatty acids (C10–C18) introducing hydrophobic character with water contact angles > 90º [1,2]. Here, we investigate the hydrophobicity introduced to cellulose with short to long-chain fatty acid esterification using molecular dynamics simulations [3]. We develop software tools for building models of functionalized cellulose microfibrils and surfaces, and use the models to study how fatty acid grafting affects the surface structure, the organization of interfacial water and water-cellulose work of adhesion.

Our simulations show distinct structural effects due to the fatty acid carbon tails aggregating near the substitution sites on the hydrophilic cellulose surfaces. The degree of substitution and carbon tail length both influence the structure of the interfacial water, and medium to long chain fatty acids tend to form a patchy surface with variable water accessibility. Analysis of hydrogen bond density shows the formation of a depletion layer near the grafted surfaces. The water contact angle, as determined through the cellulose-water work of adhesion, shows a linear response to the graft chain length for C4–C12 side chains. After this, the contact area and work of adhesion between water and cellulose remain unchanged, reaching contact angle saturation for long-chain fatty acids. Such computational approach has potential use in the development and optimization of surface modification treatments for lignocellulosic materials.

[1] J. Peydecastaing, et al. Cellulose 18 (2011) 1023–1031
[2] P. Willberg-Keyriläinen, et al. Heliyon 5 (2019) 11
[3] N.K. Karna, et al. J. Colloid Interface Sci. 589 (2021) 347–355.

Air-dried rigid bio-foam prepared by the Pickering emulsion approach

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Despite increasing environmental consciousness, producing foam from renewable lignocellulosic materials remains challenging due to issues such as low strength, difficulties in controlling porosity, uneconomical freeze-drying techniques, and poor water resistance. The Pickering emulsion principle is a unique method of making biobased foams that allows the tuning of foam's porosity and mechanical characteristics by controlling the interfacial interaction between the foam components. Although many reports have demonstrated the preparation of dry foam by harnessing cellulose through a Pickering emulsion template, these approaches rely on freeze-drying [1, 2] or oven-drying processes[3, 4] to obtain bio-based dry foams. This study aims to reduce energy consumption during the drying process of foam by designing a pickering emulsion template that allows the fabrication of dry foams at room temperature. The result showed that a stable emulsion of cellulose, lignin, polyvinyl alcohol, and alkene solvent with a smaller droplet diameter (3 – 8 µm) compared to the previous study (~12.5 µm)[5] was obtained. Dry foams produced after overnight drying at room temperature had higher strength at 50% strain (272.8 kPa) compared to other studies by using freeze[2] (82.41 kPa) or oven-dried foam[3] (< 50 kPa). Our proposed methodology shows the potential to reduce the use of foam derived from fossil fuels without sacrificing the drying process, desired morphology, and strength properties.

References

(1) Agustin, M. B.; Nematollahi, N.; Bhattarai, M.; Oliaei, E.; Lehtonen, M.; Rojas, O. J.; Mikkonen, K. S. Lignin nanoparticles as co-stabilizers and modifiers of nanocellulose-based Pickering emulsions and foams. *Cellulose (Lond)* **2023**, *30* (14), 8955-8971. DOI: 10.1007/s10570-023-05399-y From NLM PubMed-not-MEDLINE.
(2) Wang, H.; Dinesh; Kim, J. Development of lightweight, high-strength, and highly porous ligno-nanocellulosic foam with excellent antioxidant and insulation properties. *Carbohydr Polym* **2024**, *326*, 121616. DOI: 10.1016/j.carbpol.2023.121616 From NLM PubMed-not-MEDLINE.
(3) Liu, S.; Gao, Y.; Zhang, R.; Wu, R.; Lu, P. Hydrophobic bio-based foam with improved mechanical strength and antibacterial activity via rosin coating. *Industrial Crops and Products* **2024**, *216*. DOI: 10.1016/j.indcrop.2024.118750.
(4) Parın, F. N.; Dinç, H.; Parın, U.; Kıldalı, E.; Taner, G. Preparation and characterization of Pickering foams by mechanical frothing and emulsion templating. *Express Polymer Letters* **2024**, *18* (3), 282-295. DOI: 10.3144/expresspolymlett.2024.20.
(5) Kimiaei, E.; Farooq, M.; Grande, R.; Meinander, K.; Österberg, M. Lignin Nanoparticles as an Interfacial Modulator in Tough and Multi-Resistant Cellulose–Polycaprolactone Nanocomposites Based on a Pickering Emulsions Strategy. *Advanced Materials Interfaces* **2022**, *9* (27). DOI: 10.1002/admi.202200988.

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Fabrication of Hydrophobic Cellulosic Foam Using Sustainable Technology

Biobased lightweight materials have received high attention from both academia and industry as a replacement for fossil-based materials.[1] Cellulose has risen as a very interesting resource to create a range of products that allow us to go from a fossil-based society to a bioeconomy.[2,3] Well-designed cellulosic foams can be applied in variety of applications including mattresses, sealing and insulation products, water purification and many other applications. Cellulosic fibre is highly water-absorbent; they need certain level of hydrophobization to be resistant to wetting by polar liquids such as water. Aqueous foam can be used as a transform medium to form a biobased foam from cellulosic fibres together with chemicals to design the structure according to the intended application. Here, we disclose how to use the biodegradable chemicals during the foam fabrication process to design a hydrophobic biobased foam with high mechanical properties. Chemi-thermomechanical (CTMP) and bleached chemi-thermomechanical (BCTMP) pulps are used for fabrication of hydrophobic foams in this study. We prepare an organocatalytic aqueous formulation (OAF), which contain a hydrophobic suspension derived from alkoxysilanes and an organocatalyst.[4] The combination of OAF, a sustainable polyelectrolyte and pulp, resulted in a hydrophobic foam with high strength property (E-modulus improved from 2.75 to 22 kPa).[5] Elemental mapping images show an even distribution of the OAF and the chitosan at the lignocellulose fibers of the foam. The fabricated biobased hydrophobic foam has a great potential for decontamination applications such as removal of oil from water. All ingredients are approved as non-toxic chemicals and can be readily provided in large quantities, which allows for scalability in a cost-effective manner.

References

[1] Mort, R.; Vorst, K.; Curtzwiler, G.; Jiang, S. RSC Advances. 4375–4394 (2021) 22.
[2] Ottenhall, A.; Seppänen, T.; Ek, M. Cellulose. 2599–2613 (2018) *25* (4).
[3] Ferreira, E. S.; Cranston, E. D.; Rezende, C. A. ACS Sustain Chem Eng**.** 8267–8278 (2020) *8* (22).
[4] Cordova, A.; Alimohammadzadeh, R. Composition Intended for Use as a Coating. WO2022136398A2
[5] Cordova, A.; Alimohammadzadeh, R. A sustainable process for fabricating lightweight products from cellulose-based pulps. US Patent application.

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Investigation of the effect of the interaction between main components of bio-based barrier dispersion coatings on its film formation

Nowadays, food packaging is a major source of global plastic waste, which is a significant environmental issue. One possible solution is applying barrier coatings, which enable the use of more environment-friendly plant-based materials like paper and paperboard for food packaging by significantly enhancing the barrier properties of these porous and moisture-absorbing materials. Water-based barrier dispersion coatings possess many advantages, such as facilitated repulping, recycling, and, in some cases, composting of the coated plant-based packaging. Their advantages also include low emissions of volatile organic compounds, some of which may be a health hazard. Additionally, barrier dispersion coatings are promising safe alternatives to fluorochemicals in food packaging. However, the film formation mechanism remains at the core of the design of dispersion coatings with good barrier properties, and the development of cost-efficient barrier dispersion coatings still requires substantial research efforts. Currently, interest in bio-based barrier dispersion coatings has increased due to the possibility of an even greater reduction in the use of fossil-based materials. It should be noted that bio-based dispersions demonstrate even more complex film formation mechanisms than synthetic dispersions due to the recalcitrance of many natural materials.

Our study delves into the area of the interaction between the components of bio-based barrier dispersion coatings, shedding light on these coatings' complex film formation mechanisms. We studied the interaction between the bio-based dispersion's core polymer cellulose acetate butyrate with the stabilizer poly(vinyl alcohol). Variations in chemical structure and molecular weight, the effect of the ratio of the components in dispersion, and the size of the particles on the film formation properties were studied. Fourier-transform infrared spectroscopy and quartz crystal microbalance with dissipation monitoring were employed for qualitative and quantitative evaluation of the interactions between the dispersion components. We employed surface-sensitive atomic force microscopy, a mainstay for investigating the film formation process at the nanoscale, enabling the evaluation of various parameters' impact. Moreover, some dispersions were coated onto paperboard, the obtained coatings’ morphology was evaluated with scanning electron microscopy, and the coatings’ barrier properties were tested. As a result, the effect of different coating dispersion parameters on film formation and coatings’ morphology and barrier properties were analyzed, and main trends in the structure-properties relationship were revealed.

This work represents a step towards the development of bio-based dispersion coatings, a promising way for sustainability. By studying these coatings' complex film formation mechanisms, our research contributes to the ongoing efforts to find sustainable alternatives to plastic food packaging.

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Dispersion of bacterial cellulose in organic solvents and its assembly as an ultra-low grammage coating on woven fabric

Current permeable system of chemical and biological (CB) protective suit is mostly multi – layered, consisting of a permeable outer shell fabric for evaporative cooling and an integrated layer of active carbon lining as an adsorbent layer against hazardous aerosol and gas. Aspiring for an enhanced particulate filtration efficiency in the shell fabric layer and hence an extended lifespan of the inner carbon adsorbent, a versatile method was pitched to explore the feasibility of incorporating bacterial cellulose (BC) in protective garment. In this study, BC suspensions prepared in different concentrations and with various organic solvents were first vacuum filtered onto woven cotton fabrics and nonwoven films of BC with grammage < 1 g m^{-2} were achieved. Air permeabilities of these fabricated fabrics were characterised and particulate filtration efficiencies were also simulated with a sodium chloride aerosol challenge in a small – scale wind tunnel. For instance, a prototype coated with 1 g m^{-2} of BC – in – water suspension had a filtration efficiency of 83% while sustaining an air permeance of 20 mm/s. Despite the hydrophilic nature of BC, it was discovered that similar particulate filtration performance could be achieved with 0.5 g m^{-2} of BC when an ethanol – water mixture was employed during filtration. Filtration efficiency of woven fabric coated with 1 g m^{-2} of BC – in water/ethanol suspension also increased from 83% to 93%, hinting a better dispersion of BC in a ‘less polar’ medium. The solubility parameter between different organic solvents and BC was therefore investigated in this work and the potential of realising different porous structure in BC nonwoven films by controlling solvent polarity was demonstrated.

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Combined full-field tomography and SWAXS imaging at ForMAX

With the rising demand for sustainable products, forest-derived materials are crucial for addressing societal and economic needs worldwide. Renewable natural composites, such as plant and wood fibers, exemplify these materials due to their exceptional mechanical properties [1], stemming from the hierarchical arrangement of cellular structures and the ultrastructural characteristics of cellulose microfibrils. Understanding the structural properties of these materials on micro-, nano-, and atomistic scales is essential when considering the development and manufacturing of biodegradable products with desired traits.

Synchrotron-generated X-ray full-field tomography and scattering techniques are invaluable for generating multiscale structural information on wood and other hierarchical materials. At the ForMAX beamline at MAX IV Laboratory [2], the combination of full-field microtomography imaging (µCT) and small- and wide-angle X-ray scattering (SWAXS) allows for in-situ, time-resolved characterization of these materials.

In this project, we characterized the nanoscale properties of cellulose microfibrils in selected microscale structures of young birch branches using a combination of µCT and SWAXS imaging techniques. The microfibrils' orientation and crystalline properties were determined and matched with the cellular structures visible in tomography data. The results emphasize the outstanding suitability of the combined X-ray imaging techniques for the multimodal structural characterization of hierarchical materials and the unique capabilities of the ForMAX beamline.

[1] Jakob, M, et al., Prog. Mater. Sci. 100916, (2022), 125
[2] K. Nygård et al., J. Synchrotron Rad. 31, (2024), 2

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Enhanced Material Analysis through the Integration of Rheological and Scattering Methods

We present an experimental methodology that integrates rheology with temperature and moisture control in conjunction with Small-Angle X-ray Scattering (SAXS) and Wide-Angle X-ray Scattering (WAXS). This approach allows us to investigate material properties at various length scales simultaneously.

The core of our experimental setup is the Anton Paar MCR702 rheometer, outfitted with a custom-modified SWAXS cell that enables temperature and moisture control. This versatile cell can accommodate different sample holders, making it suitable for a wide range of materials. For solid samples, we use holders for bar-shaped specimens, while for softer materials, extensional fixtures can be used. Liquid samples can be measured using parallel plate, cone and plate or Couette cell configurations. The rheometer is mounted on a movable table at the MAX IV synchrotron in Lund, Sweden, facilitating the investigation of material properties in both time and space during a single experiment. Our system operates within a temperature range of approximately 25 °C to 200 °C. When combined with moisture control, the temperature range is naturally limited but can manage humidity levels from a few per cent up to around Rh = 90%.

The presentation will highlight the versatility of our setup by showing examples of different materials exposed to various humidity and temperature conditions. We will illustrate how this integrated approach provides new insights into the interaction between mechanical properties and structural changes at different length scales, offering a deeper understanding of material behavior that was previously out of reach.

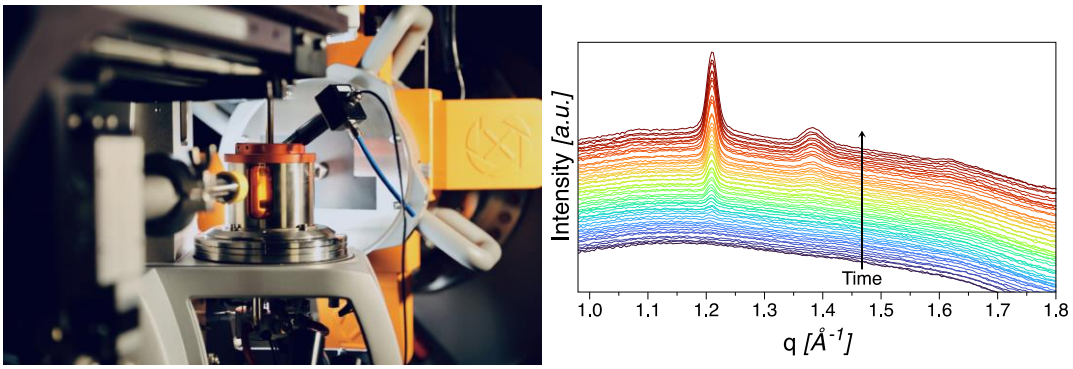

Figure 1 Rheometer with SWAXS cell at MAX IV (left). Example of a time evolution of WAXS signal (right).

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X-ray Computed Tomography aided Finite Element Modelling to Analyse the Hygromechanical Behaviour of Wood

Microscopic X-ray computed tomography aided finite element modelling (XCT FE modelling) is a popular method within material science to predict material properties and behaviour of heterogenous materials, such as wood. The method uses information from synchrotron or lab-based tomograms to build, calibrate and validate finite element models. This approach allows for situation specific simulations through incorporation of material variability and complexity of geometry. XCT FE modelling can be categorised in five subsequent steps: *source*, *experiment*, *image processing*, *model development*, and *output* (Florisson et. al. 2023). This process is far from automated, which can result in human-induced errors.

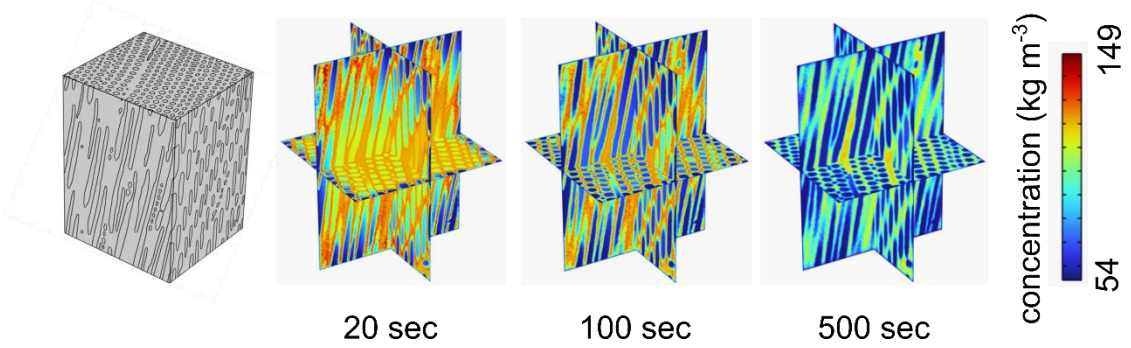

Figure 1: XCT FE moisture flow simulation of a Norway spruce branch compression wood microstructure (image courtesy to Emmy Lano and Isac Norberg)

At Uppsala University, an XCT FE modelling methodology for wood was developed and the bottlenecks within this process were identified. Two case studies were used to test the methodology and some of its bottlenecks. The first study focused on segmentation and meshing strategies to geometrise the microstructure of wood for finite element modelling. The second study focussed on the proper identification of hygroexpansion properties of opposite wood (OW) and compression wood (CW) found in Norway spruce branches. The properties were obtained with an ex-situ post-mortem scanning approach. By incorporating the variability of material and geometry into a numerical model through XCT, a more naturalistic material behaviour could be simulated and a better prediction of material properties could be obtained.

[1] Florisson, S. & Gamstedt, E. (2023). An overview of lab-based micro computed tomography aided finite element modelling of wood and its current bottlenecks. Holzforschung, 77(11-12), 793-815. https://doi.org/10.1515/hf-2023-0061

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Synchrotron techniques as a new way to look into the fundamentals of the pulping process and wood fibre properties

Considering modern environmental demands to replace fossil-based materials with renewable and biodegradable analogues, the scientific community started to put more attention to understanding the pulping process and its impact on wood fibre structure and properties. Wood is one of the most abundant natural materials, the market value of which is rapidly dropping due to the emergence of electronic and plastic substitutes. In order to be able to use wood components in the production of smart materials and be competitive economically with synthetic analogues, it is necessary to have a good understanding of structure-property relationships. We want to assess this by means of different synchrotron techniques, including Wide Angle X-Ray Scattering (WAXS), X-Ray 3D Nanotomography, and X-Ray Multiprojection Imaging (XMPI).
Kraft cooking is one of the well-established methods for pulp production, and numerous studies were performed previously to characterize the impact of each step on the wood structure and properties [1]. Usually, for characterization such studies require sampling before and after treatments. One needs to stop the experiment at different steps to perform sampling and characterization during the experiment. This may induce false visualization of chemical and mechanical changes. In this work, we attempted to study pulping in situ with WAXS using a specially customized reactor without the need for termination of the experiment. First tests in water were already performed at DESY, P03 beam line, and have proven the potential of the reactor.

[1] B.T.T. Dang, H. Brelid, H. Theliander, Holzforschung, 1-7 (2015)

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Understanding the impact of lignin types on the yield and morphology of cellulose-lignin based fibers: advanced characterization of lignin leaching with magnetic resonance methods

The shift towards forest-based products aligns with the growing emphasis on sustainability and the need to reduce dependency on fossil resources. Cellulose–lignin-based carbon fibers (CFs) have emerged as a promising alternative to expensive fossil-based polymers used for precursor fibers. However, the performance of the bio-based carbon fibers needs further improvement. One of the persisting challenges is the lignin leaching during fiber spinning which limits the carbon fiber yield and the efficiency of the process.
This work focuses on comparing lignin leaching by varying the molecular weight and sources of lignin: soft wood kraft lignin (SKL), low and high molecular weight fractions of SKL and wheat straw lignin are used.

Magnetic resonance imaging (MRI) is used for an in-situ characterization of the lignin leaching on a model set-up for filaments coagulation[1]. The MRI visualizations and evaluation of the mass transport during the coagulation are complemented with UV absorbance measurements on coagulation baths during a spinning process to determine the amount of leached lignin. The spun fibers are analyzed using Klason analysis and gel permeation chromatography (GPC) to assess the lignin content. Additionally, liquid-state NMR is used on the different lignins and on dissolved spun fibers[2] to examinate structural changes in lignin and cellulose, as well as any potential linkages or interaction points between the two molecules.

The combined methodologies suggested that the lignin leaching is very dependent on the molecular weight and the chemical structure of the lignin. A substantial lignin content decrease is noticed when lignin with higher content of carboxylic acid groups is used – i.e. wheat straw lignin - followed by the low molecular weight fraction of SKL.

Another part of this study demonstrated that lignin leaching likely results in defects, defibrillation, and pores in the spun fiber. Conversely, a lower lignin content in the spun fiber led to an apparent increase in tensile strength, likely due to the relatively higher cellulose content.

Maximizing the potential of bio-based carbon fibers remains challenging. Emphasis should be placed on minimizing lignin leaching and optimizing the operating conditions for fiber spinning to improve their quality and performance.

[1] Hedlund A, Macromolecules 2017, 50 (21), 8707–8719 .
[2] Fliri, L. et al. Nat. Protoc. 2023, 18 (7), 2084–2123.

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Effect of fiber aspect ratio on the extrusion instabilities in wood polymer biocomposites

Wood polymer composites (WPCs) are key industrial ingredients in the context of overarching societal efforts to promote and increase the use of renewable materials. Specifically, the use of wood fiber-based biocomposites as renewable materials can lead to a significant reduction in the consumption of plastic materials. The WPCs employed in this study are custom compositions based on the DuraSense® by StoraEnso, comprising polypropylene reinforced with different wood fiber concentrations (up to 40 wt%) and different aspect ratios. Usually, the extrusion flow of unfilled polymers exhibits a smooth surface (no instability) at low shear rates, followed by the appearance of instabilities such as sharkskin, stick-slip, and gross melt fracture at higher shear rates. However, in stark contrast to the dynamics of neat polymers, the extrusion flow of WPCs can be dominated by the presence of so-called surface tearing instabilities, even at very low shear rates. Melt flow instabilities (MFIs) of WPCs were mapped in single-screw extrusion via inline optical spectral analysis for the first time in this study. The MFIs were detected and quantified by applying an inline image analysis with the help of an optical visualization setup placed at the die exit. Further, space-time diagrams were constructed, subsequently, the temporal and spatial periodicities of the MFIs were determined as a function of shear rate, and Weissenberg number using 2D-Fourier transform analysis. Fourier transform spectrograms represent different characteristic frequency peaks for various instabilities. Surface tearing appeared from the lowest shear rates investigated, followed by a gradual decay in spectral intensity with increasing shear rates/slip velocities. Increasing shear rates, drying, wood fiber content, and increasing fiber size showed mitigating effects on surface tearing. However, surface tearing in undried samples was still present even at the highest shear rates and high wall slip velocities. A regime where the extrudate surface is dominated by bubbles at high shear rates and low wood fiber contents in undried WPCs was identified [1].

[1]. Pashazadeh, Sajjad, et al. "Mapping surface defects in highly-filled wood fiber polymer composite extrusion from inline spectral analysis." Composites Science and Technology 242 (2023): 110133.

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Transforming Poplar Wood to high performance bio composites

Abstract:

Wood is a renewable, CO_2-storing natural material with widespread use in the construction sector, thanks especially to its favorable mechanical properties. Its combined high specific strength and stiffness make it useful as a core material for advanced structural sandwich panels, and its unique structure allows its use for developing advanced functional materials such as membranes and nanogenerators [1]. However, compared to other engineering materials and composites, wood has important disadvantages such as moisture-driven swelling and shrinking, and low mechanical properties in the wet state [2, 3].

We report a novel, fully bio-based high-performance composite obtained by reconstructing wood. This composite was developed by the delignification of poplar wood followed by its relignification i.e. impregnation with Kraft lignin. The physical and chemical properties of the reconstructed wood, and especially its microstructure, were investigated using a combination of scanning electron microscopy (SEM), fluorescence imaging, X-ray diffraction (XRD), and small-angle X-ray scattering (SAXS), and tensile experiments. The microstructure of reconstructed wood displayed strongly deformed cell walls, which helped forming cohesive interfaces. Compared to native wood and densified wood, the reconstructed wood exhibited significant superior mechanical properties, reaching a tensile strength of about 250 MPa and yield stress of 130 MPa. In addition, this material displayed a very high-water resistance. Reconstructed wood, born from the essence of wood, holds great promises as a more sustainable solution for construction and manufacturing of high-performance materials.

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[1] Richard Wool and X Susan Sun. *Bio-based polymers and composites*. Elsevier, 2011.

[2] Roger M Rowell and W Bart Banks. Water repellency and dimensional stability of wood. *Gen. Tech. Rep. FPL-50. Madison, WI: US Department of Agriculture, Forest Service, Forest Products Laboratory; 1985: 24 pages*, 50, 1985.

[3] Sophie Marie Koch, Manuel Pillon, Tobias Keplinger, Christopher Hubert Dreimol, Stephan Weinkötz, and Ingo Burgert. Intercellular matrix infiltration improves the wet strength of delignified wood composites. *ACS Applied Materials & Interfaces*, 14(27):31216–31224, 2022.

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Biodegradable and Flexible Wood-Gelatin Composites for Soft Actuators

Compliant materials are essential for many emerging soft robotics applications. Concerns about sustainability and end-of-life options for these materials are increasing, as they are mainly petroleum-based and non-recyclable. Although alternative bio-derived soft materials like gelatin have been explored, they often fail to meet the mechanical performance requirements for soft actuating systems. To address this, we reinforced a compliant and translucent gelatin-glycerol matrix with structure-retained delignified wood, creating a flexible and fully bio-based composite called DW-flex. This DW-flex composite exhibits highly anisotropic mechanical behavior, with greater strength and stiffness in the fiber direction and high deformability perpendicular to it. Introducing such anisotropy into otherwise isotropic soft materials enables more complex movement patterns. To demonstrate DW-flex's capability and potential, we developed and modeled a fin ray-inspired gripper finger that deforms based on a twist-bending-coupled motion, adjustable by changing the fiber direction. Additionally, we designed a proof-of-concept demonstrator suitable for gripping a soft object with a complex shape, such as a strawberry. We show that this composite is entirely biodegradable in soil, offering more sustainable solutions for soft actuators in robotics applications.

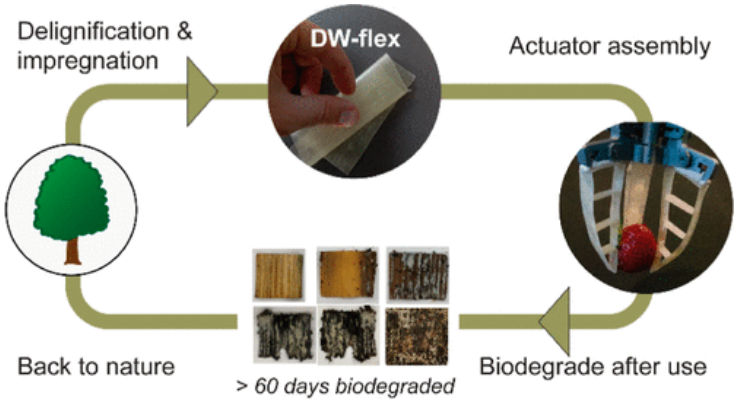

Figure 1. Graphical abstract of this work. Wood is delignified and infiltrated by a gelatin-glycerol matrix. Then, a gripper is assembled for gripping soft objects with a complex geometry. The composite can finally be biodegraded and returned to nature.

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Development of active polylactic acid (PLA)-based packaging film containing artichoke (*Cynara scolymus*) leaf extract and enzyme

The global population growth has led to an increased demand for minimally processed food, resulting in a limited shelf life of food products and high consumption of synthetic plastic packaging [1]. Consequently, the food packaging industry is seeking bioactive packaging materials that can prolong the shelf life of food while providing a sustainable alternative to non-degradable plastics [2]. Meanwhile, plant residues, such as artichoke leaves, are often discarded, creating environmental concerns. Global artichoke production is estimated at 1978 Kton annually, with 70-80% discarded in the form of leaves, stems, and roots [3]. However, artichoke leaves are rich in bioactive compounds with potential use in active packaging. Therefore, this study aims to formulate a compostable and active PLA-based packaging film by incorporating a PLA-degrading enzyme and bioactive artichoke leaf extract (ALE). For this purpose, ALE was recovered using subcritical water extraction based on previously optimized conditions in a two-cycle process (solid-to-liquid ratio of 1:10, temperature of 180 °C for 15 minutes). The bioactive PLA-based blend was formulated with 0, 2.5, and 5% wt (of total solids) of both ALE collected from the second cycle and PLA-degrading enzyme, and a homogeneous mixture was prepared using a co-rotating twin-screw extruder. Stretch films with a thickness of around 240-270 µm were produced by compression molding and characterized for their mechanical properties and bioactivity. The antioxidant results for the PLA-based films showed the highest activity for the film containing 5% of both ALE and enzyme, with 81.07, 61.14, and 74.94% DPPH scavenging activity after three oxidative cycles for 15-minute intervals. Mechanical analysis indicated that the addition of ALE and enzyme decreased the tensile strength from 80.36 MPa in the control film to 54.98 MPa in the film containing 5% of both enzyme and ALE. Similarly, Young's modulus values decreased from 2138.93 MPa in the control film to 1797.84 MPa in the film containing 2.5% of both enzyme and ALE. The softening effect of the ALE and enzyme was beneficial for stretch films, intended for wrapping food products of various shapes and sizes. Additionally, the bioactivity results are promising for enhancing the shelf life of food products and reducing food waste.

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Keywords: Active packaging, Food loss, circular economy, Plant waste

References

[1] Khan, M. R. et al., Food Packag. Shelf Life. 101313 (2024) 44
[2] Momtaz, M. et al., Sci. Rep. 519 (2024) 14
[3] Bittencourt, G. et al., J. Supercrit. Fluids. 106028 (2023) 201

The mysterious glassy state and its transitions – a brief overview

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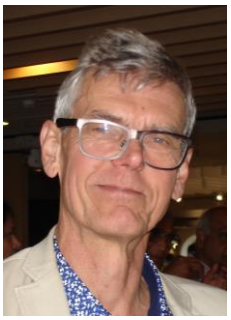

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Amorphous polymeric materials and phases undergo several thermal transitions, which involve changes in the molecular mobility without marked change in the molecular order. At very low temperatures in the glassy state, the molecular mobility is restricted to vibrations. At some higher temperature, the polymer chains begin to change conformation and these changes are subtle involving only a few polymer segments. These processes which occur in the glassy structure are referred to as sub-glass processes. At even higher temperatures when the amorphous polymer becomes markedly softer – a fully amorphous uncrosslinked polymer shows a three order of magnitude decrease in the Young's modulus – the glass has been converted to a polymeric liquid. This is the glass transition. A taste of a number of related topics are presented in the lecture: (i) the kinetic and the thermodynamic glass transitions, traffic jam and the Kauzmann paradox; (ii) methods for the assessment of transition temperatures and the fictive temperature; (iii) physical ageing, engineering aspects, memory effect and models; (iv) confined amorphous phases in semicrystalline polymers; (v) facts about cellulose and other wood-related polymers. Recommended reviews concerned with fundamental aspects are refs. [1] and [2]. Refs. [2] and [3] provides information about methods to study glassy polymers and their transitions. Ref. [4] presents structure-property relationships for glassy polymers.

[1] U. W. Gedde and M. S. Hedenqvist, Fundamental Polymer Science, 2nd Edition, Chapters 5 and 6, Springer Nature Switzerland AG, Cham, Switzerland (2019).
[2] R. H. Boyd and G. D. Smith, Polymer Dynamics and Relaxation, Cambridge University Press, Cambridge, UK (2007).
[3] U. W. Gedde, M. S. Hedenqvist, M. Hakkarainen, F. Nilsson and O. Das, Applied Polymer Science, Chapters 1, 3 and 5, Springer Nature Switzerland AG, Cham, Switzerland (2021).
[4] U. W. Gedde, M. S. Hedenqvist, M. Hakkarainen, F. Nilsson and O. Das, Applied Polymer Science, Chapters 6 and 7, Springer Nature Switzerland AG, Cham, Switzerland (2021).

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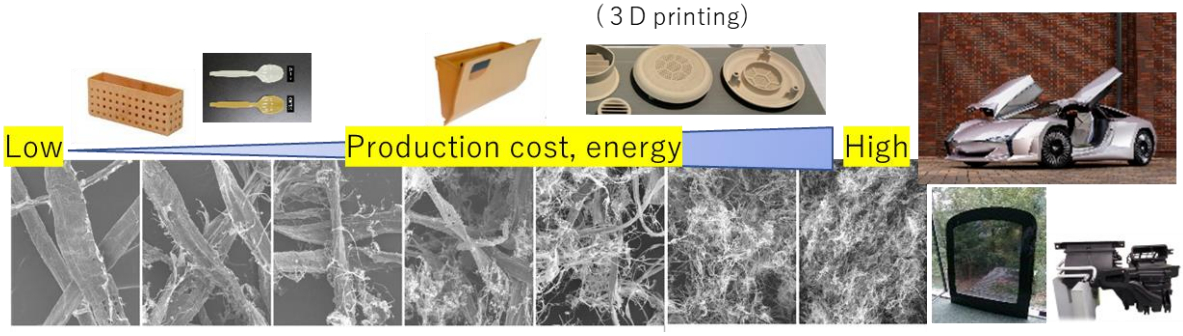
Bio and Material Recycling for a Circular Economy Society

High elasticity, high strength, and low linear thermal expansion resulting from semi-crystalline extended polymer chain structure are fundamental to structural application of cellulose. In addition, cellulose, which is made from atmospheric carbon dioxide, is renewable and completely different from metals, ceramics, and petroleum-derived plastics, which emit carbon dioxide in the manufacturing process. Therefore, cellulose is a material that supports carbon-neutral society. We have developed lightweight, high-strength materials in 2001 and optically transparent materials with low linear thermal expansion in 2003 using cellulose nanofibers. We have also been working on reinforcing plastics with chemically modified cellulose nanofibers from 2005. In 2019, we combined these materials to create an NCV (Nanocellulose Vehicle), and driving tests have shown that a 16% weight reduction achieved with NCV leads to a 11% improvement in fuel efficiency (NCV movie, https://www.youtube.com/watch?v=06H8wP9axjU). Subsequently, based on these results, we have been developing cellulose nanofiber reinforced bioplastics (HDPE, LDPE, PBS, PHBH) and cellulose nanofiber reinforced recycled plastics as materials for a circular economy society. Since these materials are easily material recyclable, LCA has shown that their use contributes significantly to GHG reduction. In the future, it will be important to develop and implement in society cellulose fiber-reinforced plastics on the basis of their performance due to the semi-crystalline extended polymer chain structure and biodegradability, with a range of element sizes from nano to micron, low energy consumption during manufacturing, and cost advantages.



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Hemicellulose and cellulose interactions as a bioinspired approach for tuning water interactions and viscoelastic properties of cellulose networks

Physical modification via polymer adsorption constitutes one of the simplest means for modifying cellulose nanomaterials.[1] Moreover, utilizing the naturally occurring interactions of carbohydrates, such as hemicelluloses and chitin, with cellulose has proved a versatile tool for nanocellulose modification.[2] We demonstrate the potential of biobased carbohydrate polymers, i.e., mixed-linkage glucans (MLGs), for tailoring water interactions and viscoelastic properties of nanocellulose networks.[3] These grass cell wall hemicelluloses have recently gained attention for their interesting interactions with cellulosic materials, including their ability to efficiently gel and hydrate nanocellulose networks.[4,5] The presentation discusses the adsorption behavior of MLGs on nanocellulose surfaces, which somewhat deviates from the well-known polyelectrolyte adsorption theories. Moreover, the influence of co-solute ions, especially chaotropic species like nitrates, on the MLG layers' structural properties and water interactions is highlighted (Figure 1).

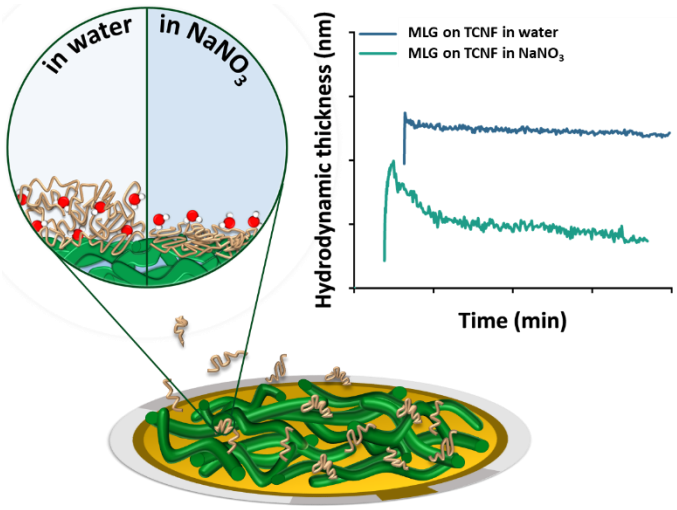

Figure 1. Strong binding of mixed-linkage glucans (MLGs) on TEMPO-oxidized cellulose nanofibers (TCNF) is evidenced via an interfacial investigation using quartz crystal microbalance with dissipation monitoring (QCM-D). MLG adsorption on TCNF is studied in water and controlled salt solutions to elucidate the role of co-solute ions on the interactions. Furthermore, the Voigt model[6] is fitted to the adsorption and energy dissipation data to model the viscoelastic properties and swelling of the MLG adlayers.

[1] F. Rol et al., Prog. Polym. Sci. 241-264 (2019) 88.
[2] E. Kontturi et al., Adv. Mater. 1703779 (2018) 30.
[3] T. Virkkala et al., J Mater. Chem. B. 8788-8803 (2023) 11.
[4] S. Arola et al., Soft Matter 9393-9401 (2018) 14.
[5] J. C. Muñoz-García et al., Biomacromolecules 4180−4190 (2019) 20.
[6] M. Voinova et al., Phys. Scr. 391-396 (1999) 59.

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DNP-NMR as a tool to determine degree of modification in dialcohol cellulose

Dialcohol cellulose is synthesized via periodate-mediated oxidation of cellulose followed by borohydride reduction. This two-step reaction results in a ring-opened polymer structure with primary hydroxyl groups at the C2 and C3 positions of the glucose residues, providing desired processability and the potential for replacing petroleum-based plastics in various applications. The degree of modification, a key parameter dictating the material's mechanical properties, including ductility, brittleness, and moisture sensitivity, must be precisely quantified to facilitate its development. In this study, we demonstrate the application of solid-state NMR spectroscopy enhanced by dynamic nuclear polarization (DNP) to accurately quantify the degree of modification in dialcohol cellulose. By employing multiple-contact cross polarization experiments under DNP conditions, we achieved sensitivity enhancements exceeding a factor of fifty and this high sensitivity enables the rapid quantification within 20-30 minutes, offering a significant improvement over traditional titration and UV-based methods. Additionally, we explore the use of 13C-13C dipolar recoupling experiments for detailed structural characterization, including chemical shift correlations in dialcohol cellulose. Our results highlight the utility of DNP-enhanced solid-state NMR techniques for precise chemical analysis of modified biomass material.

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Recent developments of all-cellulose composites from cellulose fibers – concepts, properties, and applications

Single polymer composites have been under development for some time, and these composites are made of a single polymer type, which is processed so that a composite with a distinct reinforcement is embedded in a matrix, made from the same polymer. Compared to conventional composites, which are composed of two different materials (for example glass fibre reinforcement and a thermoset resin), single polymer composites can be recycled after end-of life in an easier way, as no physical separation of the composite constituents are needed.

All-cellulose composites (ACCs) have shown their potential for this purpose. By first selectively dissolving a cellulose material (preferably as a fabric) and then regenerating the cellulose it is possible to form a single polymer composite. The ACC is composed of a cellulose reinforcement, which is embedded in a cellulose matrix. By doing the processing according to a controlled procedure, it is possible to obtain a composite with tailored structure and aligned cellulose fibres.

The presentation will review our previous and recent research aiming at demonstrating the potential for all-cellulose composites as a structural composite material. The process has involved both ionic liquids and cold NaOH-urea, and laminates of various thickness and dimensions have been made and te achieved properties have been analyzed. In our research we have used both recycled end-of life textiles (cotton denim), as well as non-woven virgin cellulose fabrics.

[1] Baghaei, B., Skrifvars, M. All-Cellulose Composites: A Review of Recent Studies on Structure, Properties and Applications, *Molecules* **2020**, *25*, 2836
[2] Baghaei, B., Johansson, B., Skrifvars, M., Kadi, N. All-Cellulose Composites Properties from Pre-and Post-Consumer Denim Wastes: Comparative Study, *J. Compos. Sci.* **2022**, *6*, 130
[3] Uusi-Tarkka, E.-K., Levanič, J., Heräjärvi, H., Kadi, N., Skrifvars, M., Haapala, A. All-Cellulose Composite Laminates Made from Wood-Based Textiles: Effects of Process Conditions and the Addition of TEMPO-Oxidized Nanocellulose *Polymers* **2022,** *14*, 3959

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The unseen magic of water molecules in wood

We unravel the magic of water molecule behavior in cellulose, hemicellulose and lignin, its mixtures and composites at atomistic level using molecular dynamics (MD).
We show that water molecules are adsorbed at nanoscale first filling the existing pore space between polymeric chains, followed by adsorption induced swelling. This coupling between adsorption and swelling is at the origin of sorption hysteresis observed in wood [1].
The mechanical properties show first stiffening when water molecules fill the initial porosity, followed by a weakening at higher moisture content attributed to a breakage of hydrogen bonds [2]. This hydrogen bond breakage with increasing moisture content is also responsible for the decrease in bond strength between crystalline cellulose fiber and matrix, explaining the shape memory effect of wood [3].
The diffusion in wood polymers enhances with increasing moisture content and is shown to depend on adsorption energy and stiffness of the material [4].
Treatment of wood cell wall with polyethylene glycol (PEG) is a widely used to consolidate waterlogged archaeological wooden artifacts such as the Swedish warship Vasa and Henry VIII's warship the Mary Rose. PEG treatment is shown to effectively stabilize the wood structure and prevent extreme shrinkage and structural collapse that can occur during uncontrolled drying processes [5].

[1] M. Chen, B. Coasne, R. Guyer, D. Derome, J. Carmeliet, Role of hydrogen bonding in hysteresis observed in sorption-induced swelling of soft nanoporous polymers, Nat Com., 2018 29;9(1):3507. doi: 10.1038/s41467-018-05897-9
[2] Zhang C., Chen M., Keten S., Coasne B., Derome D. and Carmeliet J., Hygromechanical mechanisms of wood cell wall revealed by molecular modeling and mixture rule analysis, Science Advances, vol. 7: no. 37, Washington: Amer Assoc Advancement Science, 2021
[3] Chi Zhang, Mingyang Chen, Sinan Keten, Dominique Derome and Jan Carmeliet, Towards unraveling the moisture-induced shape memory effect of wood: the role of interface mechanics revealed by upscaling atomistic to composite modeling, NPG Asia Materials, vol. 13: no. 1, pp. 74, London: Macmillan Publishers, 2021.
[4] Chi Zhang, Ali Shomali, Benoit Coasne, Dominique Derome and Jan Carmeliet, Sorption-Deformation-Percolation Model for Diffusion in Nanoporous Media, ACS Nano, vol. 17: no. 5, pp. 4507-4514, Washington, DC: American Chemical Society, 2023.
[5] Wenqiang Liu, Ali Shomali, Chi Zhang, Benoit Coasne, Jan Carmeliet and Dominique Derome, Nanostructure and interfacial mechanical properties of PEG/cellulose nanocomposites studied with molecular dynamics, Carbohydrate Polymers, vol. 343, pp. 122429, Elsevier, 2024.

Challenges to process nanocellulose for new fiber based materials

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Among lignocellulosic materials, the nanocellulose family emergence over the last decade has been highly promising. Such cellulosic materials have numerous advantages which favor their uses in several various applications like in composites, electronics, medical, food , building or cosmetic industry. Since the beginning, the nanocellulose industry focuses its efforts to up-scale their production and develop the commercialization of these nanoscaled cellulose.

This key step has been more complex than expected despite their promising properties, mainly because of process issues regarding sustainability and energy consumption during their production. One part of the presentation will discuss about how sustainable can be considered the production of nanocellulose.

At the same time, it was not so obvious to find tangibles applications, even in fiber based materials packaging, one of the most commonly described and expected applications.

During this presentation , we will discuss some of the main challenges of their use in the fibre based packaging industry and share how nanocellulose can help for promising solutions by providing some examples from the Cellulose Valley, an industrial excellence chair launched on the topic in Grenoble for 2022-2026. Some example of value-added functionalization of such materials and the application of such nanocellulose on 2D and 3D packaging will also be proposed to conclude and propose perspectives.

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Highly thermal conductive bio nano composite with anisotropic properties

Wood polymer composites (WPCs) are essential in the move towards renewable materials, as they significantly reduce reliance on plastics. However, WPCs typically lack high thermal conductivity, limiting their application. This study presents the pioneering manufacture of 2D Hexagonal Boron Nitride (HBN) and wood fiber (provided by Stora Enso) reinforced Poly Lactic Acid (PLA) composites using extrusion. Initially, the wood fibers were coated with HBN, then compounded with PLA, and subsequently the used as input material in capillary extrusion. Prior to extrusion, the rheological properties of the composites were examined using a rotational rheometer, and extrusion parameters were adjusted based on the composite viscosity. The composites exhibit excellent anisotropic properties due to the orientation of wood fibers and HBN in the extrusion direction. Various compositions, including 15 wt% wood fiber (WF), 45 wt% HBN, 30 wt% WF-30 wt% HBN, and 45 wt% WF-15 wt% HBN, were analyzed. Thermal conductivity tests revealed that the 30 wt% HBN-30 wt% WF composite achieved a 1000% improvement in thermal conductivity compared to pure PLA in the flow direction. Notably, the perpendicular direction to the flow showed a 100% improvement, underscoring the remarkable anisotropic characteristics of the extruded composites.