Review

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Designing functional wood materials for novel engineering applications

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Abstract: Wood has great potential to become a key material for future bio-economy, thanks especially to its intrinsic renewability and CO₂-storing capacity. Improved functionalization treatments can make wood materials valid substitutes for less ecofriendly ones, expanding and widening their application range. However, further research is needed. This mini-review highlights some of the most recent developments in the design of functional wood materials, critically discussing their current limitations and obstacles to their implementation.

Keywords: functional wood materials; insulation; sensor materials; sustainability; transparent wood.

1 Introduction

Recent years have witnessed a tremendous scientific interest for the development of functional wood materials (Berglund and Burgert 2018; C. Chen et al. 2020; H. Zhu et al. 2016). The CO₂-storage capacity and renewable nature of wood are both factors of increasing relevance in the context of resource depletion and climate change mitigation efforts. Wooden buildings already can act as long-term carbon sinks, thanks especially to passive timber construction based on cross-laminated timber and Glulam (Churkina et al. 2020). However, there is another fundamental driver towards exploring the full potential of wood functionalization to gain functional materials, which is represented by the hierarchical structure of wood itself. Wood is a robust but porous scaffold, in which

functionalizing agents can be embedded to make functional wood materials tailored for a variety of engineering applications (Berglund and Burgert 2018; Burgert et al. 2015; C. Chen et al. 2020; Kim et al. 2016; Slagman et al. 2018; H. Zhu et al. 2016). The objective to achieve specific properties or functionality by making direct use of the wood micro and nanostructure distinguishes this approach from prominent wood modifications such as acetylation or thermal treatments, which are applied to achieve property improvements, like dimensional stability or resistance against decay (Alfredsen et al. 2013; Militz and Lande 2009). Furthermore, using the wood structure in a top-down approach is substantially different from using disassembled wood cell wall components, like nanocellulose, to assemble a new material bottom-up. The advantage here is that no disassembly and subsequent assembly processes are required, and that the beneficial fiber directionality can be maintained in the entire process (Keplinger et al. 2019). However, a disadvantage compared to nanocellulose is that wood functionalization is more demanding and somewhat still limited. Moreover, while the rapid development in the field of 3D printing of nanocellulose-based inks grants a high flexibility in shaping structures (Markstedt et al. 2017; Siqueira et al. 2017), for wood the degree of design freedom is determined by the structure of the chosen species. Recent developments towards structure-retaining delignification treatments have been especially useful to compensate in part for these disadvantages, without compromising the raised assets of using wood as a scaffold. In delignified wood both the hierarchical structure and fiber directionality is maintained, but the impregnability and porosity of the cell wall is increased (at least in the wet state), facilitating subsequent functionalization treatments (Grönquist et al. 2019; Song et al. 2018). Moreover, a greatly improved deformability in the wet state, together with an enhanced compressibility in densification treatments, allow the production of cellulose scaffolds with complex shapes as well as to make structural changes in a much simpler way compared to native wood (Frey et al. 2018, 2019a,b).

In light of the limitations in design freedom introduced by the wood structure, selecting the most appropriate wood

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species for the desired application, with or without prior delignification, is of utmost importance to enable its proper functionalization. Trees provide a broad range of structural variability, in terms of wood porosity and additional structural specifications. The density of wood can vary from as low as 100 kg m^{-3} to more than 1200 kg m^{-3} , depending on the species, which then reflects in its porosity. Some applications, like those related to functional transparent wood, can profit from the use of rather low-density species (H. Chen et al. 2020), while for liquid transport selecting hardwood species with frequent and even-sized vessels might be favorable (Goldhahn et al. 2020; Zhu et al. 2017b). Regardless of the most proper choice of the wood species, it is of utmost importance to actively exploit its structure to ensure the efficiency of any kind of functionalization treatment. Instead of serving only as a passive, mechanically stable scaffold, the structure of wood must become an integral part of the functionality of the resulting hybrid material. This is a fundamental criterion for developing wood materials with superior functionalities, able to substitute less eco-friendly materials in the near future.

In this mini-review a selection of recent developments in the field of functional wood materials is discussed, both for native and delignified wood substrates, focusing specifically on the connection to smart building applications via embedded adaptability or responsiveness. The interplay of functionalized wood materials and regulation of living comfort, energy efficiency, and even energy harvesting in buildings has great potential. The mini-review provides not only a selection of important scientific activities and progress in the field, but also critically discusses the current limitations and potential for future development, identifying the core challenges that need to be addressed. Research on enhancing the mechanical performance of wood materials by delignification and densification treatments will not be discussed, as it has been covered already in a recent review (Keplinger et al. 2020).

2 Transparent wood

Native wood is not transparent to visible light due to a combination of scattering (from micro-sized channels) and absorption (by lignin and other chromophores). Scattering in wood is caused by its complex hierarchical tissue structure and the inhomogeneous distribution of bio-macromolecules with refractive index (RI) values ranging from 1.53 (cellulose) to 1.61 (lignin). Removing or decolorizing lignin leaves wood with a bright white color, but substantially opaque due to the large RI difference between

cellulose and air. By filling the channels with properly selected materials, the RI mismatch can be reduced and light reflection suppressed, increasing transparency. In 1992, Fink reported a procedure by which wood samples (up to 4 mm-thick) could be “optically dissolved” i.e., made transparent to facilitate its investigation by means of optical microscopy (Fink 1992). The procedure involved two steps: first a chemical bleaching to remove light-absorbing species such as lignin, followed by a RI matching obtained by impregnating wood with low-viscosity liquid formulations having a RI comparable to that of the bleached wood (Figure 1a).

Fink’s work targeting to facilitate optical microscopy of wood is seminal as it contains the fundamental concepts that have been revived more than 20 years later by two research groups (Y. Li et al. 2016; M. Zhu et al. 2016a). Both groups were attracted by the engineering possibilities offered by transparent wood as a bio-based, lightweight, load-bearing, and low-cost material (Figure 1). From everyday uses such as a substitute for conventional glass in windows, to the automotive sector and optoelectronics, a variety of possible applications were envisaged already, and were further empowered by the development of multifunctional transparent wood composites. In the following, the most relevant aspects of transparent wood fabrication are discussed, comparing the original procedure of Fink with those followed in recent years, and then a selection of proposed engineering applications for multifunctional transparent wood.

Lignin is the major responsible in wood for visible light absorption. Hence, one either needs to remove lignin from the wood matrix or to chemically modify its structure eliminating the chromophores responsible for its color. In his original report, Fink achieved structure-retaining delignification by immersing the wood samples in aqueous sodium hypochlorite NaClO , followed by thorough washing with water. Then the samples were dehydrated with increasing concentrations of acetone or ethyl alcohol to prepare them for impregnation. Berglund and collaborators chose to treat wood samples with sodium chlorite a procedure already reported by Yano et al. (2001). A milder approach allows to partially decolorize lignin by dipping wood in an alkaline hydrogen peroxide solution, stabilized with sodium silicate. This procedure only destroys the chromophores that are responsible for the brown color of lignin, reducing optical absorption while preserving the mechanical properties of wood to some extent (Li et al. 2017a).

Otherwise, wood samples can be first boiled in a solution of sodium sulfite and sodium hydroxide, and then transferred into a hydrogen peroxide solution to complete the removal of lignin (M. Zhu et al. 2016a). Most recently a

Figure 1: Examples of transparent wood. (a) Preparation of transparent wood by successive delignification and refractive index (RI) matching of a wood sample (cross-section). Images adapted and reprinted with permission from Wiley-VCH, copyright 2016 (M. Zhu et al. 2016a). (b) A picture of transparent wood and (c) optical transmission spectra of transparent wood samples with different thicknesses (LRW: delignified wood, PMMA: pure bulk poly(methyl methacrylate)). Images reprinted with permission from American Chemical Society, copyright 2016 (Y. Li et al. 2016).

photocatalytic procedure has been reported for bleaching balsa wood by first treating it with an alkaline concentrated hydrogen peroxide solution and then expose it to 20 W-ultraviolet (UV) light for up to 2 h, or to solar light for longer times (Xia et al. 2021a,b).

According to Fink, polymerizable mixtures suitable for impregnation should: “(1) yield transparent, crystal-clear preparations, (2) offer a choice of variable refractive indices, (3) show only little shrinkage during polymerization, and (4) allow ease of handling” (Fink 1992). Most often a mixture of monomer(s) (e.g., vinyl and/or methacrylic) and oligomers is used instead of pure monomer(s). Impregnation with pre-formed polymers is also possible, but it depends on factors such as the average molecular weight, the concentration, and the nature of solvent(s). Monomer(s), prepolymer(s), initiator(s) (when necessary), additional solvent(s), and excipients are mixed together and then used to impregnate the sample. This can happen by exchanging the acetone or ethanol solvent in wood with the polymerizing mixture, replenishing it periodically and allowing it to diffuse over relatively long times (≥ 24 h), or by forcing the mixture to enter the pores of wood by applying vacuum. Polymerization can then happen by thermal or photo-initiation, depending on the chosen system. In case of conventional free radical polymerization, the radical-scavenging effect of atmospheric oxygen needs to be taken into account. Usually, the impregnated wood samples are sandwiched between glass (or quartz, in case of UV-mediated photopolymerization) slides to reduce contact with atmosphere.

The cellulose volume fraction is of importance for the transmittance and the transparency of the resulting wood

composite is higher the better the polymer fills the matrix and adheres to the cell wall surfaces (without leaving empty, or air-filled, spaces that would lead to scattering), and its RI matches that of the cellulose matrix. A variety of polymers have been used so far to make transparent wood composites, including: epoxy resins, thiol-ene resins, poly(dimethylsiloxane) (PDMS), poly(methyl methacrylate) (PMMA), poly(styrene) (PS), poly(vinyl pyrrolidone) (PVP), and poly(vinyl alcohol) (PVA). Transparent wood displays both high transmittance (over 85% for 1 mm-thick samples) (Figure 1b,c) and high haze in a broad wavelength range, between 400 nm and 1100 nm (Y. Li et al. 2016). The high haze is due to the presence of light scattering centers in wood fibers and channels (microcurvatures with bumps, microstripes, and microcavities), and the efficient forward scattering thanks to microfibers and microchannels that guide the incident light along the axial direction. Transmittance and haze can be further optimized through molecular and nanoscale materials design. For example, with the help of acetylation, the compatibility between the template and the infiltrated polymer is improved. Eliminating the interface debonding gap results in higher transmittance and lower haze. With 1.5 mm thick transparent balsa, the transmittance was increased from 83% to 92% and haze decreased from 70% to 50%. In this way, cm-thick transparent wood samples could be obtained (Li et al. 2018).

Scattering in bleached wood can be reduced also by decreasing the amount of free space (that is, air) inside the scaffold. Mechanical compression of delignified wood can thus result in reduced scattering, since densification reduces porosity (Zhu et al. 2017a). However, the degree of

transparency depends upon the sample thickness: while thin (*ca.* 50 μm) compressed delignified wood appears transparent thanks to ballistic light transport, it becomes opaque when thickness is up to a few centimeters. The anisotropy of the resulting material can be controlled by using wood samples cut in different directions. For example, compressed delignified wood obtained from radially-cut wood is highly anisotropic, while that obtained from cross-section is isotropic (Zhu et al. 2018). It is worth mentioning that wood infiltration can be conducted also without preliminary bleaching. Wu et al. (2020) contributed a study on the optical properties of native wood impregnated with PMMA, in which six wood species with highly different lignin contents were taken into consideration.

2.1 Functional transparent wood

Already in the 1992 report, Fink proposed to disperse pigment particles or fluorescing powders into the infiltrating monomer solution, to increase the optical contrast between different wood tissue details. In 2017, the Berglund group reported the preparation of luminescent transparent wood by impregnating delignified wood substrates with dispersions of silicon Si or cadmium selenide CdSe quantum dots (QDs) in methyl methacrylate (MMA) monomer/oligomer mixtures, followed by polymerization (Li et al. 2017b) (Figure 2). Photoluminescence mapping of the resulting composite revealed a homogeneous distribution of emission centers. Using a similar strategy, Gan et al. (2017) obtained luminescent magnetic wood by

incorporating core-shell iron oxide@europium-doped yttrium vanadate $\gamma\text{-Fe}_2\text{O}_3\text{@YVO}_4\text{:Eu}^{3+}$ nanoparticles. Bi et al. (2018) reported the use of luminescent transparent wood as a light-emitting layer for white-LED applications. Delignified wood was impregnated with poly(acrylic acid) (PAA) containing a mixture of fluorophores obtained from the hydrothermal treatment of citric acid and urea. The resulting composite was then glued onto a conventional LED chip.

A peculiar application of luminescent wood is to make laser cavities. Vasileva et al. (2017) prepared luminescent wood by impregnating delignified balsa wood with rhodamine 6G (Rh6G, $1 \times 10^{-3} \text{ mol L}^{-1}$), a well-known organic laser dye, followed by MMA monomer/oligomer impregnation and polymerization. The resulting Rh6G-doped transparent wood (TW-Rh6G) was used as an active gain medium to achieve lasing (Figure 2a). The slope efficiency of the TW-Rh6G laser was 11%, with a threshold pump energy of 0.7 mJ. Notably, the emission spectrum of TW-Rh6G had a narrower ($\approx 5\text{--}6 \text{ nm}$) peak width, measured as the full width at half maximum (FWHM), compared to that of free Rh6G photoluminescence spectrum. This resulted from the superposition of lasing modes produced by individual resonators, in that case cellulose fibers of different lengths and diameters. By comparing the results obtained for TW-Rh6G and for a reference medium (PMMA-Rh6G), the lasing action from TW-Rh6G could be attributed to the collective effect of cellulose fibers working as an assembly of small, partially-ordered uncoupled Fabry–Perot type resonators. Since in TW-Rh6G optical feedback is not based on a classical external resonator, but instead is obtained through scattering on the wood fiber boundaries, the fine structure of the lasing modes is more similar to that of dye molecules in other quasi-random

Figure 2: Examples of functional transparent wood. (a) Effect of wood scaffold anisotropy on the photoluminescent emission of transparent wood loaded with quantum dots (QDs). The intensity of the light emission collected at the sample edge is a function of the relative wood fiber direction. An isotropic sample reference (QDs embedded in glass) is also shown for comparison. Image reproduced with permission from Wiley-VCH, copyright 2017 (Li et al. 2017b). (b) Transparent wood as a light-management coating to improve the performance of a conventional GaAs photovoltaic cell. Image adapted and reproduced with permission from Elsevier, copyright 2016 (M. Zhu et al. 2016b).

media, e.g., glass beads. As such, this type of source could be more appropriately defined as a quasi-random laser. It is also worth mentioning that the diameter of separate fibers was not found to be a critical issue for the lasing performance: the shape of the emission spectra would be the same as long as the samples' width would be larger than the penetration depth of the pump light (200–300 μm).

In 2019, the Zhou group showed the assembly of electroluminescent devices by sandwiching a conventional inorganic phosphor (zinc sulfide doped with copper, ZnS:Cu), embedded in epoxy resin, between two layers of transparent wood (Zhang et al. 2019b). Wood was first delignified, impregnated with epoxy resin for RI matching, then made conductive by spray-coating silver nanowires (AgNWs) on the surface. The lower hygroscopicity and thermal conductivity of epoxy resin-impregnated wood reduced both humidity uptake and thermal expansion, respectively, factors which in conventional devices can lead to failures due to mismatch between substrate and active layer. As such, superior performances were claimed, especially in terms of resistance against humidity and temperature changes, using transparent wood substrates instead of conventional paper- and plastic-based ones (Zhang et al. 2019a). Recently, delignified wood was impregnated with a polymerizable deep eutectic solvent (PDE) based on acrylic acid and choline chloride, followed by *in situ* photopolymerization to obtain a transparent, electrically conductive (0.16 S m^{-1}) composite (Wang et al. 2019). The incorporation of fluorescent moieties in such a composite could lead to very interesting results.

Lang et al. (2018) made polymer-based electro-chromic devices (ECD) using transparent wood as substrate. They deposited a thin (*ca.* 140 nm) layer of poly(3,4-ethylene-dioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) on transparent birch wood by blade-coating. Then, they enhanced the electrical conductivity of the PEDOT:PSS layer by around three orders of magnitude treating with 1 M *p*-toluenesulfonic acid in ethylene glycol (EG) at 40 °C. The resulting devices displayed a vibrant magenta-to-colorless color change ($\Delta E^* 43$, $\Delta\%T 38\%$) with a high coloration efficiency ($590 \text{ cm}^2 \text{ C}^{-1}$) while requiring low energy and power inputs (3 mW h m^{-2} , 2 W m^{-2}) and driving voltage of only 0.8 V, making it attractive for applications as e.g., optical memories and smart windows.

Another direction for transparent wood applications is as a structural material for photovoltaic devices such as solar cells. Thanks to the high haze, transparent wood can be designed as a light diffusing layer. High haze means large scattering angles, which increase the length of the light path inside solar cells, so that efficiency is improved. The application of transparent wood to solar cells has been first

described by the Hu group in 2016. Transparent wood was used as a broadband light management layer to enhance the trapping of photons inside the active layer in a standard gallium arsenide GaAs solar cell (M. Zhu et al. 2016b) (Figure 2b). More recently, the Berglund group described the use of transparent wood as substrate for a low temperature (<150 °C)-processed perovskite solar cell (Y. Li et al. 2019). Zou et al. (2020) proposed a two-layer transparent wood, with channels oriented perpendicular (0/90°) to each other, obtained from rotary-cut veneer to increase the performance of a conventional silicon solar cell. Finally, the application of luminescent transparent wood for luminescent solar concentrators (LSCs) has been suggested, but not yet demonstrated in practice (Li et al. 2017b).

Thanks to its anisotropic light scattering properties, the use of transparent wood for smart windows has been proposed, to help the modulation of transmitted sunlight, and even improve thermal management (T. Li et al. 2016). Yu et al. (2017) demonstrated a heat-shielding transparent wood by introducing near infrared (NIR)-absorbing cesium tungstate Cs_xWO_3 nanoparticles in a transparent wood matrix. Tested as windows in small-scale model buildings, a strong absorption of light was observed at wavelengths from 780 to 2500 nm, resulting in lower temperature increase under continuous solar radiation compared to a glass window reference. Mi et al. (2020b) reported the fabrication of a relatively large-scale (tens of cm wide and long) “esthetic” transparent wood. Douglas fir was selected thanks to its pronounced structural contrast between low-density earlywood (EW) and high-density latewood (LW). Selective removal of lignin from EW with sodium chlorite treatment, followed by impregnation with epoxy resin, allowed to make transparent wood while preserving the natural growth ring patterns. In addition to the aesthetic value, the resulting transparent wood was reported to have better UV-blocking properties, which are ascribed to the presence of residual lignin (Mi et al. 2020b). Fu et al. (2020) recently described a way to obtain luminescent wood without the need to impregnate the scaffold with RI-matching polymers. In this approach, a delignified wood scaffold was first infiltrated with hydrophobic QDs and then densified to a thin film, which showed uniform luminescence thanks to isotropic light scattering.

3 Wood as sensor and energy-harvesting material

Applying mechanical forces to certain materials can result in the production of an electrical charge, a phenomenon

called “electromechanical coupling”. The reverse effect is also possible, that is, the development of a mechanical strain upon application of electrical bias. Piezoelectricity is possibly the best-known example of electromechanical coupling phenomena, discovered in 1880 by the Curie brothers in a variety of crystals (inorganic as well as organic).

On the other hand, research on the piezoelectric behavior of wood and wood materials is much more recent, the first reports dating back to the 1940s (Shubnikov 1940). Fukada (1968) and Bazhenov (1961) were able to measure the generation of electric charges, attributed to piezoelectricity, upon the application of mechanical stress on wood. The same fundamental relations developed for piezoelectricity in monocrystalline materials are believed applicable to wood. The presence of crystalline regions of cellulose, with hydroxyl groups being arranged in a non-centrosymmetric order, is thought to be responsible for the observed phenomenon of piezoelectricity in wood. In the first reported work on wood piezoelectricity, Shubnikov (1940) proposed the concept of “piezoelectric texture” to represent a system consisting of many crystalline particles oriented unidirectionally, as for cellulose crystals in wood. While Fukada (1968) reported that only the shear piezoelectric effect could be measured for cellulose in woody tissues,

later studies revealed the existence of non-zero transverse piezoelectric effects (Hirai et al. 2011).

Although the reported piezoelectric coefficients were very small (Fukada reported the magnitude of the piezoelectric modulus of wood as approximately 1/20 of that of a quartz crystal), those studies have spurred further investigations thanks to possible applications. A number of different wood species have been shown to exhibit a piezoelectric effect, and non-destructive wood testing by means of direct piezoelectric effect was actually the subject of several investigations (Knuffel and Pizzi 1986; Niemz et al. 1994). Nowadays, research on electromechanical coupling effects in wood and other biomaterials (such as silk and wool) has seen a remarkable resurgence, especially due to promising applications of energy-harvesting devices, wearable sensors, and flexible actuators (Maiti et al. 2019; Wang et al. 2021). If wood could be engineered to generate macroscopic piezoelectric responses, it could have distinct advantages over piezoelectric ceramics and synthetic polymers because of its intrinsic sustainability, excellent mechanical properties, relatively low density, and great relevance as building material.

Recently Sun et al. (2020, 2021) reported about a simple and effective way to strongly enhance the electrical output from electromechanical coupling in wood by means of

Figure 3: Wood sponge piezoelectric nanogenerator. Pictures and microstructures of (a) native wood and (b) delignified wood sponge. (c) Enhanced compressibility of wood sponge. Piezoelectric output of a single (d) native wood cube and (e) delignified wood sponge cube. Images adapted and reprinted with permission from American Chemical Society, copyright 2020 (Sun et al. 2020).

delignification (Figure 3). Balsa wood was found to be especially suitable, most probably thanks to very low cell wall thicknesses. Removal of lignin could be achieved both by chemical (treatment with concentrated acetic acid and hydrogen peroxide mixtures at 80 °C) (Sun et al. 2020) and biological treatments (white rot fungi) (Sun et al. 2021). The resulting “wood sponge” displayed a spring-like micro-structure due to cell wall breakage, leading to enhanced compressibility. In turn, the electrical output produced upon the application of compression forces as small as 13 kPa increased by an order of magnitude. Connecting more wood sponges in series or parallel, respectively the voltage or current outputs could be increased to power small devices such as LEDs or liquid crystal displays (LCDs). A demonstrator wooden floor element, made with 30 wood sponges (each measuring $15 \times 15 \times 14 \text{ mm}^3$) connected in parallel could produce a maximum output current of $\approx 205 \text{ nA}$.

Despite such promising results, and their intrinsic sustainability, piezoelectric wood generators do not appear competitive enough compared to their inorganic counterparts for energy harvesting and the use as bio-based sensor material appears more suitable. For energy-harvesting wood materials, triboelectricity could be a more convenient approach. However, the very low ability of native wood to attract or lose electrons is a major hinder for the development of wood triboelectric generators, but could be solved by proper surface engineering strategies.

4 Wood for energy-efficient buildings

Functional wood materials produced by delignification can also be used as insulation materials or as strong light irradiation reflectors, to guarantee a comfortable interior room climate while reducing energy consumption. Since the removal of lignin by delignification treatments increases the porosity and decreases the density of wood, drying steps that retain the porous structure result in materials with comparably good insulation and mechanical properties. Their thermal conductivity is especially low perpendicular to the fiber direction due to a pronounced anisotropy (He et al. 2020), but they cannot compete with advanced insulation materials such as nanocellulose-based aerogels or silica aerogels (Koebel et al. 2012; Lavoine and Bergström 2017). Transparent wood is also of interest for more energy-efficient windows with thermal conductivities significantly lower than glass, in particular perpendicular to the fiber direction (Mi et al. 2020a).

Figure 4: Transparent wood for energy-efficiency (a) Transparent wood can be used to harvest and guide sunlight. As such it can be used (b) as a rooftop to achieve comfortable and uniform lighting while keeping a constant indoor temperature. Images reprinted with permission from Wiley-VCH, copyright 2016 (T. Li et al. 2016).

A very promising approach is based on the optimization of light reflection by delignified wood, which could lead to a radiative cooling material (Figure 4). A complete lignin removal results in a bright white wood, which has been utilized by T. Li et al. (2019) to develop wood materials that backscatter solar radiation and show pronounced energy emitting properties in the infrared wavelength range resulting in a cooling effect. A comprehensive modeling of energy saving potentials in different US cities based on the “cooling wood” properties revealed that on average $\sim 35\%$ cooling energy could be saved in midrise apartment buildings erected prior to 1980 and $\sim 20\%$ cooling energy could be saved in buildings erected after 2004, if bright-white delignified wood is used as a building material. The modeling showed that cooling energy savings could be especially pronounced under hot and dry conditions.

5 Wood as a natural filter

Water purification processes rely most often on filtration to remove macromolecules, particles, and microorganisms that could be harmful for human health and for the environment in general. To this end, membranes made from artificial polymers or ceramics are widespread used, with associated problems of fouling and disposal. Developing sustainable filtration systems of simple use and that could be easily and safely disposed is of special relevance for developing countries, which are most affected by diseases caused by contaminated water. Wood is a widespread available biomaterial, which multiscale hierarchical structure and aligned microchannels make highly interesting for filtration and separation purposes. While

chemical modification of wood scaffolds offer many opportunities for e.g., the design of in-flow catalytic reactors and for oil/water separation (Fang et al. 2021; Goldhahn et al. 2020; Xie et al. 2020), here the discussion is focused on the physical filtration properties of native wood tissue.

The xylem offers minimal resistance to the ascent of sap while maintaining small pores (pits) for cell wall passage. Depending on the plant species, the size distribution of these small pores can range from a few nanometers to around 500 nm, making wood xylem (when never dried) an excellent candidate for transporting water while rejecting pathogenic particles (Figure 5). Despite the high scientific and social relevance, relatively little is known about the removal efficiency of native wood for particulate and colloidal material, especially in the size range of interest for water-borne pathogens like viruses (≤ 100 nm), bacteria (0.5–3.8 μm), and protozoa (4.9–12.2 μm).

Boutillier et al. (2014) showed that filtration through the sapwood of pine trees could significantly reduce turbidity and remove bacteria (*Escherichia coli*, rejection exceeding 99.9%) from wastewater by pressure-driven filtration. The filters were prepared simply by removing the bark of pine tree branches (≈ 2.5 cm-long sections of ≈ 1 cm-diameter) and inserting the xylem tissue into a tube. Filtration experiments with hydrophobic Nile Red pigment particles revealed a size cut-off of about 100 nm, with most of the filtration occurring within the first 2–3 mm of the filter. Bordered pits were identified as the functional unit/small pores where actual filtration of the bacteria occurred (Figure 5). Flow rates of about 4 L/day were obtained using 1 cm^2 filter areas at applied pressures of about 5 psi, and 3 cm^3 of sapwood could filter water at the rate of several liters per day. However, such filters were not as effective against viruses, as they could not filter out nanoparticles about 20 nm in size. Most importantly, the filtration

properties of the pine sapwood were found to be drastically reduced after drying. The blockage of xylem filters upon drying is related to the bordered pit aspiration blocking the fluid transport pathways between softwood tracheids. A 100 $\times$ drop in flow rate was observed for these 2.5 cm-thick filters.

Che et al. (2019) modified the internal surfaces of pine wood with silver nanoparticles (AgNPs) to inactivate, in addition of trapping, bacteria from contaminated water. The activity of such 10 mm-thick wood membranes, native or treated to contain 1.25 wt% of AgNPs, was then tested against *E. coli*. The total density of *E. coli* bacteria remaining in the filtrate from the AgNPs/wood membrane was similar to that found for the filtrate obtained using native wood, indicating that the introduction of AgNPs in wood caused no apparent change of its physical filtration properties. However, almost all of the bacterial cells that were not rejected by the AgNPs/wood membrane were dead, unlike the case of native wood.

Vitas et al. (2019) reported a study of particle rejection using beech wood, a choice motivated by larger cell lumen diameter due to vessels. Beech wood cross-sections (thin discs of 18 mm $\times$ 1 mm) were characterized to determine their porosity parameters and ζ -potential. These data were in turn used to predict the theoretical removal efficiency, based only on structural considerations. Using a dedicated setup, parameters such as flux, permeability, specific permeability, and their time-dependency were evaluated, and eventually the wood filters were challenged with micro-particles (5 and 20 μm in diameter) as surrogates for microorganisms. Alteration of the flow and filtration characteristics due to swelling were observed after prolonged testing times (≈ 8 h) and the results showed removal efficiency only for the bigger particles (20 μm diameter).

Most recently, a comprehensive study by Ramchander et al. (2021) addressed not only the problem of virus

Figure 5: Wood as a filter. (a) Photograph of a pine branch (~ 1 cm diameter) showing xylem (wood) and bark, and schematic representation of water flow (blue arrows) through conifer xylem (the red circles represent the bordered pits where filtration occurs). (b, c) SEM images showing *Escherichia coli* cells accumulated on the margo after filtration of contaminated water (scale bars: 10 and 2 μm , respectively). Images adapted and reprinted with permission from (Boutillier et al. 2014), copyright 2014, the authors. Published by Public Library of Science.

filtration with wood filters, but also several other characteristics that are critical for practical water filtration applications, such as their structural stability over the course of its shelf- and operational life, and susceptibility to common fouling agents. Insights on xylem behavior of three different wood species were coupled with those on how thickness and area of the wood filter can affect its performance. Xylem filters were found to have a highly non-linear dependence of flow resistance on thickness upon drying, and a tendency for self-blocking. To retain some permeance in dried filters, the filter thickness was reduced, and an optimized procedure involving first the swelling and then the dehydration with ethanol of the wood filter was described. The relevance of this work is multifold, as it provides guidelines for the design and fabrication of gravity-operated xylem filters with shelf life >2 years, that can provide >3 log removal of *E. coli*, MS-2 phage, and rotavirus (from synthetic test waters) and coliform bacteria (from contaminated environmental waters). Finally, a prototype filtration device was proposed, working with daily- to weekly-replaceable xylem filters.

6 Future perspectives

The application-driven wood functionalization treatments discussed here are only a selected extract of the recent and ongoing research activities in the field of native and delignified wood materials, without claims of completeness. These examples should be representative of a rapidly developing field, powered by the increasing awareness of environmental challenges and associated need to promote more efficient use of natural resources, coupled with new (nano)technological developments facilitating more sophisticated functionalization of wood materials and cellulose scaffolds. Having said this, it is also important to emphasize that many open challenges remain to be tackled to ensure the transitional period from basic research to practical application being as short as possible.

Upscaling of materials' production, from laboratory to application-relevant sizes, is one of these key challenges, even though functionalizing a bulk material by a top-down approach greatly facilitates upscaling compared to bottom-up assembly processes (like those applied for nanocellulose). The efficient treatment of larger samples can be affected by diffusion-based processes, a limitation which can be circumvented by assembling/stacking functionalized veneers to larger structural elements (Frey et al. 2019a). In case a high porosity needs to be retained by the final material, common laboratory-scale approaches like freeze-drying may not be affordable at the industrial level.

Strict adherence to green chemistry guidelines is also required to help establish a leading role for functionalized wood materials in future engineering applications. It makes no sense to acclaim the renewable nature and CO₂-storing capacity of wood and cellulose scaffolds, if their functionalization requires procedures and chemicals with negative sustainability, recyclability, health and environmental safety aspects. This is a general challenge in the wood sector and beyond, but in view of the foreseen regulations enforcing circular economy, the earlier the emerging field of functional wood materials will be able to cope with these requirements, the faster its wide adoption could be ensured.

Another important challenge is associated to the performances, which can be achieved by functionalizing wood. Even though new developments and improved processes will result in enhanced wood materials, the application-specific performances of many less eco-friendly materials may remain out of reach, such as those requiring high electrical conductivity. Here, the solution might rather come from a more global change of perspective, not only of the scientific community, but especially of industry and society. Advantages in sustainability, recyclability, health and environmental safety must weigh more heavily, and the individual unit related performance should not be the main decision criteria. For instance, the degree of efficiency of photosynthesis of plants is comparably low (Zhu et al. 2008), but this is irrelevant in view of the quantity of plant leaves. Likewise, a functional wood material in a building application may have a lower efficiency than a more competitive, less ecofriendly material,

Figure 6: Selected applications of functional wood materials in smart buildings: indoor thermal and light management, sensing of and energy harvesting from mechanical movement, improved barrier effects.

but the disadvantage could be overcompensated by using large amounts of this wood material, for instance by using it as decorative wall, ceiling or floor element with embedded functionality (Figure 6).

If these challenges could be overcome, smart buildings would enter a new era, by integrating functional bio-based materials in which wood, thanks to its wide availability and excellent properties, may play a key role. But this requires rapidity of action, since a supply of wood for a bio-based circular economy requires implicitly a sustainable forestry, and the accelerating climate change has also strong impacts on forests and trees in many countries.

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