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Fibrillated Cellulose: Towards a Sustainable Technological Material

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Preface: Cellulose is the most abundant biopolymer on Earth, found ubiquitously in trees, agricultural crops, and other plant biomass. Through different fibrillation processes, the fibers comprising cellulose can be separated into building blocks of different dimensions that extend down to the nanoscale. As a sustainable material, fibrillated cellulose exhibits significant advantages compared to other nanomaterials in addition to its multifunctional mechanical, optical, thermal, and fluidic properties. The last decade has witnessed exciting research progress in fibrillated cellulose material design and unconventional applications that have opened a new field of research with abundant but largely unexplored opportunities. This Perspective discusses the emerging science and technology of fibrillated cellulose and its development as a sustainable technological material, as well as research opportunities for addressing global challenges.

Introduction

The exit from the fossil carbon era into a sustainable future will require high-performing renewable materials with low or even net-zero carbon emission. Cellulose is a clear candidate as the most abundant renewable biopolymer on Earth, where it exists as a structural component in the cell wall of plants, in some species of algae, and as biofilms secreted by bacteria (Figure 1a)¹. In addition to its advantage as a sustainable material, cellulose enables multiple functions and transformative applications that derive from its unique multidimensional structure, which can be separated from fibers into fibrils of decreasing diameter (ranging from $< 100\ \mu\text{m}$ to $\sim 2\text{--}4\ \text{nm}$) that are ultimately composed of ordered linear cellulose molecular chains (Figure 1b). In this Perspective, we use “fibrillated cellulose” as a general term to describe the cellulose fibers that have been broken down into such fibrils².

Due to this hierarchical structure, fibrillated cellulose features significant tunability in terms of morphology and fibril size³, enabling mechanical, optical, thermal, fluidic, and ionic properties that far surpass those of cellulose fibers. In addition, fibrillated celluloses are biocompatible and significantly less expensive than metal nanowires and petroleum-based nanomaterials ($\sim \$0.60/\text{dry kg}$ for papermaking grade and $\$20/\text{dry kg}$ for nanoscale fibrillated cellulose)⁴. Furthermore, the accelerated adoption of fibrillated cellulose is expected to facilitate the shift from petroleum- to bio-based products in support of a circular economy^{2,5} (Figure 1c).

Due to these advantages, there have been exciting advances in the research and development of fibrillated cellulose over the past decade. With improved fundamental understanding and control of this hierarchical structure, we anticipate that fibrillated cellulose could form the foundation of economically viable, sustainable solutions towards a range of near-term applications in high-performance structural materials, energy efficient buildings and biodegradable and sustainable technologies, as well as far-term applications in optoelectronics, bio-engineering, and membrane science (Figure 1d).

Figure 1. An overview of fibrillated cellulose. **a**, Several common source materials of fibrillated cellulose. **b**, Schematic description of the hierarchal structure and manufacturing challenge of fibrillated cellulose. The degree of fibrillation refers to the extent to which the fibers have been longitudinally split into thinner fibrils⁶. **c**, Forecast of the total production value of wood-based products in the Finnish bioeconomy, used here as an example of the possible impact of new advanced materials, including those from fibrillated cellulose, which can drive the exports and gross domestic product (GDP) growth of a nation (adapted from Ref ⁵). **d**, Roadmap of fibrillated cellulose technologies including current application in paper, near-term applications in specialty packaging, bioplastics, lightweight structural materials, and energy efficient buildings and transportation, as well as far-term technologies including novel membranes for energy and water, optoelectronics and bio-engineering.

Structural applications

Cellulose possesses appealing intrinsic mechanical properties with a theoretical specific modulus of $\sim 95000 \text{ N}\cdot\text{m/g}$ and specific tensile strength of $\sim 5000 \text{ N}\cdot\text{m/g}$ ^{7,8}, both of which are higher than most metals, alloys, synthetic polymers and many ceramics (Figure 2a). This mechanical strength partially derives from the densely distributed hydroxyl groups (3 per anhydroglucose unit) on the cellulose molecular chains, which are critical for forming abundant inter- and intramolecular hydrogen bonds (Figure 2b), especially within the fibrils.

These hydrogen bonds are essential for the organization of cellulose molecules in crystal structures, as well as the mechanical integrity within the fibril aggregates and interactions within the cellulose fiber wall. Van der Waals interactions are also important due to their longer interaction range compared with hydrogen bonding. Furthermore, the fibril network provides physical entanglement, which plays another important role in toughening the material⁹. The extraordinary mechanical properties that result from these interactions, along with a low density ($\sim 1.5 \text{ g cm}^{-3}$), make cellulose-derived products attractive as lightweight structural materials.

Significant progress has been made to further improve the mechanical properties of materials made of fibrillated cellulose through advanced structural design and engineering (Figure 2c)^{10–15}. For example, conventional laboratory made paper made from cellulose-rich fibers demonstrates a low mechanical tensile strength of ~ 20 – $\sim 120 \text{ N}\cdot\text{m/g}$ due to the random arrangement of the large fibers in handsheet-making and the material's low density. However, this value can be improved to $\sim 160 \text{ N}\cdot\text{m/g}$ by decreasing the size of the cellulose fibril building blocks to the nanoscale and increasing the density of the film¹⁵. Aligning cellulose fibrils is another effective structural design and engineering strategy to enhance hydrogen bonding at the molecular scale^{16–18}. Following this strategy, a record-high specific mechanical tensile strength of approximately $1050 \text{ N}\cdot\text{m/g}$ and specific Young's modulus of approximately $58000 \text{ N}\cdot\text{m/g}$ in the fibril oriented direction were achieved by assembling oriented nanoscale cellulose building blocks into bulk cellulose macrofibers/filaments (density of approximately 1.5 g/cm^3)¹¹.

In terms of a top-down, potentially more scalable approach, a combination of chemical and mechanical treatments of wood can result in a high-performance, lightweight, yet mechanically strong building material composed of fibrillated cellulose with 10-fold enhancement in mechanical strength (reaching $\sim 600 \text{ MPa}$) compared with natural wood^{16,19}. The resulting densified and laminated wood, or the so-called “super-wood”, features intertwined and collapsed cell walls and aligned cellulose nanofibrils with rich intra- and interfibrillar hydrogen bonds within the cell walls, which contribute to the material's substantially enhanced strength. Future efforts to further improve the mechanical strength should focus on further reducing the number of defects in cellulose materials, maximizing the interface between cellulose fibrils and fibril aggregates, and strengthening molecular interactions (e.g., covalent bonding, molecular entanglements, van der Waals interactions, and hydrogen bonding), with a potential goal of achieving a strength of up to 2–3 GPa.

Due to their potential for large-scale manufacturing and light weight cellulose materials are particularly attractive for structural applications for improved energy efficiency. For example, a 10% weight reduction in vehicles is expected to result in a ~ 6 – $\sim 8\%$ savings in fuel consumption²⁰. In the search for a more light-weight and sustainable material for use in cars, the Ministry of the Environment of Japan recently launched the Nano Cellulose Vehicle project²¹, which aims to develop cellulose nanofiber composite resins for a 10–50% weight reduction in automotive

components (e.g., engine parts, the hood, and other structural features) and a total weight reduction of over 10% for the entire vehicle (Figure 2d). Given fibrillated cellulose's superior mechanical properties (Figure 2a) and significantly lower weight than traditional car component materials, these efforts could soon lead to improved mileage and lower greenhouse gas emissions without sacrificing driver safety.

Figure 2. Fibrillated cellulose as a lightweight structural material. **a**, Comparison of the specific tensile strength and modulus of several recently developed cellulose structures (e.g., a graphite-carbon nanofibril (CNF) composite²² and aligned cellulose macrofibers¹¹) compared with conventional structural materials, including polymers, concrete, wood, metals, and alloys. Derived from Cambridge Engineering Selector Edupack Software (Granta Design Limited, Cambridge, UK, 2017). **b**, A schematic description of the molecular structure of cellulose with abundant intrafibrillar and possibly also interfibrillar hydrogen interactions. **c**, Timeline of the improvement of the mechanical tensile strength of manmade cellulose structures^{10–15}. **d**, Lightweight vehicle with cellulose materials composing different auto parts, showing a total weight reduction of more than 10%²¹.

Materials for building energy efficiency

In the U.S., buildings account for ~30% of all energy-related greenhouse gas emissions and 40% of energy consumption. Furthermore, 70% of this energy is consumed by heating, cooling, and lighting (Figure 3a)²³. Common building materials, including concrete, steel, glass, and brick, face challenges in energy efficiency, CO₂ emissions, and environmental pollution²⁴. As a result, new and more sustainable material solutions for energy efficiency have been explored using fibrillated cellulose via various phonon and photon engineering strategies.

In order to reduce cooling and heating usage, phonon engineering of fibrillated cellulose can be applied towards the development of thermally-insulating building materials. Walls in particular are a major focus of these efforts, with the goal of achieving a thermal insulation R-value of 50^{25,26}. By tuning the spacing between cellulose fibrils comparable to the mean free path of air molecules (~70 nm at room temperature and 1 atm), the thermal motion of air molecules can be suppressed and heat transport can be impeded. Either the bottom-up approach from fibrillated cellulose or the

top-down approach through wood delignification can achieve a high porosity material (> 90%) composed of cellulose fibrils that decreases the thermal conductivity to a value comparable to or even lower than that of air at $0.028 \text{ W/m}\cdot\text{K}^{27-29}$.

Photon engineering of cellulose in a wide spectrum range has also been explored towards designing more energy efficient building materials. Cellulose exhibits an intrinsically low emissivity in the solar spectrum (< 5%, 200–1000 nm) and high emissivity in the infrared range (> 85%, 1–25 μm). In scenarios where high solar reflectivity and high infrared emission are desired, such as in passive cooling applications³⁰, fibers/pores with dimensions from tens of nanometres to tens of micrometres in delignified wood provide strong broadband back-scattering of incoming light, which renders the material extremely white and non-absorptive of solar radiation³¹. Such “cooling wood” can strongly emit infrared radiation via the atmospheric transparency window (8–13 μm) without energy consumption (inset of Figure 3b). Therefore, when applied as roofing and siding, this fibrillated-cellulose-based material can help reduce building cooling needs, decreasing the building temperature by 2–12 °C with a 24 h cooling energy of $53 \text{ W/m}^{2,31}$.

While the building envelope deals with heating and cooling for thermal comfort, windows are another source of heating and cooling energy loss due to the poor thermal insulation of glass. As an alternative, fibrillated cellulose demonstrates potential as a highly transparent yet thermally insulating window material. Uniform interconnecting pores ~100 nm in size can be obtained within high aspect ratio fibrillated cellulose and polysiloxane fibers to reach a thermal conductivity as low as $8 \text{ mW}/(\text{mK})$ with a visible range transparency of over 90%²⁸ (inset of Figure 3b), which allows the material to be applied as an energy efficient glass window replacement. Besides decreasing cellulose fiber sizes, high visible range transparency (~90%) can also be achieved by infiltrating index matching polymers into the microscale air pockets of wood frameworks^{32,33,34}. Such transparent wood has been reported in the U.S., Europe, and Asia since 2016, with continuing efforts on commercialization.

Overall, compared with commonly used building materials, cellulose-based compounds exhibit a desirable combination of thermal and optical management capabilities, energy efficiency, strength, lightweight, sustainability, and relatively low embodied energy (i.e., the total energy required for manufacturing) (Figure 3c). The combination of these advantages strongly suggests the potential of fibrillated cellulose in sustainable building applications.

Figure 3. Phonon and photon engineering of fibrillated cellulose towards energy efficient buildings. **a**, The amount of energy consumed by direct use, transportation, residential and commercial buildings, and industry in the U.S. (adapted from Ref²³). **b**, Advanced cellulose technologies for building energy efficiency. Cellulose materials are used for various functions, such as radiative cooling wood for roofing material³¹, optically transparent and thermally insulating cellulose films for windows²⁸, strong cellulose-graphite composites for mechanical support²², and thermally insulating cellulose for the building envelope^{29,27}. λ_{mfp} is the mean free path of air molecules, which is $\sim 70\ \text{nm}$ at room temperature under 1 atm. **c**, A comparison between commonly used construction materials and advanced cellulose materials for buildings in terms of energy efficiency, thermal insulation, strength, density (weight by volume), sustainability, and embodied energy efficiency.

Biodegradable and sustainable technologies

The growing demand and use of non-biodegradable materials, such as plastic, has caused severe global plastic pollution (also known as "white pollution"). In 2010, the global primary production of plastic was 270 million tons, yet the global plastic waste for the same year was 275 million tons, exceeding annual primary production due to plastic wastage from prior years^{35–37}. As a result, there has been a growing interest in bioplastics made from renewable and sustainable materials (e.g., cellulose) for packaging, textiles, and other consumer products^{38,39}.

Most petroleum-based plastics take hundreds of years to degrade due to their crosslinked covalent bonds, particularly the strong carbon-carbon bonds, which are difficult for bacteria to digest. However, the oxygenated molecular chains of cellulose, can be fragmented by certain fungi and bacteria using a battery of hydrolytic and oxidative enzymes^{40,41}. The unique combination of excellent biodegradability, outstanding mechanical strength and high heat and chemical resistance of fibrillated cellulose suggests its potential as an alternative for plastics such as polyethylene and polyethylene terephthalate.

In pursuit of cellulose-based products, industrial processes, such as electrospinning, roll-to-roll processing, and additive 3D printing, can be adopted to fabricate a large variety of fibrillated cellulosic bioplastic products, which have been successfully demonstrated and commercialized in a wide range of application fields⁴². In particular, the hybridization of polymers with fibrillated cellulose can improve the material's extrusion processability. This could enable the fabrication of products like fibrillated-cellulose-based drinking straws, particularly given consumer pressures that have led companies such as Starbucks to phase out plastic drinking straws by 2020⁴³.

The development of films made of either pure or polymer-hybridized fibrillated cellulose is also growing rapidly due to their potential as a replacement for plastics⁴⁴. Already there are commercial applications of cellulose in grease and oxygen barrier packaging, agricultural mulch films, containers, adhesives, hygienic disposables, home furnishings, and textile sizing agents⁴⁵. The global demand of different types of fibrillated cellulose films is expected to continue to grow due to the material's decreasing cost, increasing world population and demand, and government legislation towards sustainability. Efforts are well on the way to further improve the performance of fibrillated cellulose in practical applications, where traits such as water resistivity, durability, and process scalability are necessary, as we discuss in more detail in later sections.

To promote the usage of fibrillated cellulose in competition with plastics will require: (1) life cycle assessment (LCA) and degradability studies to confirm the environmental impact; (2) continuous optimization of the fabrication process of fibrillated cellulose to reduce the cost; (3) a careful balance between stability and biodegradability in the resulting composites (see Challenges section for detailed discussion); (4) a recycling process for fibrillated-cellulose-containing composites; and (5) the development of less water-sensitive cellulose-based materials without losing recyclability. Note that while fibrillated cellulosic materials can be extracted from plants, the processes themselves can be chemical and energy intensive, which from the LCA point-of-view may not necessarily be sustainable. For example, at relatively high degrees of fibrillation for producing cellulose nanomaterials, extensive chemical processing is commonly used, such as concentrated sulfuric acid hydrolysis to produce cellulose nanocrystals⁴⁶, as well as 2,2,6,6-tetramethylpiperidine-oxyl (TEMPO)-mediated oxidation. Using recyclable chemicals, such as solid di-carboxylic acids⁴⁷ or enzymes^{48,49}, can provide potential solutions, especially novel enzymes like lytic polysaccharide monooxygenases (LPMOs)^{50,51}. LPMOs promote oxidation at the C1 or C4 position of the cellulose macromolecule, which induces chain breaks, facilitating the fibrillation of cellulose fibers under milder conditions^{50,51}. One benefit to consider is the amount of knowledge and expertise accumulated in the pulp and paper industries that can be adopted for fibrillated cellulose extraction with the hope of achieving a more sustainable and green process for the fabrication of fibrillated cellulose-based biodegradable products.

Far-term technologies

While fibrillated cellulose is currently poised as a sustainable material for structural and biodegradable applications, substantial efforts have also been devoted to find new applications based on the unique properties of cellulose nanofibrils. Here we highlight the far-term outlook of fibrillated cellulose in optoelectronics, ionic and fluidic transport, and bio-engineering, with intriguing potential that warrants further study.

Optoelectronics. In addition to conducting and semiconducting elements, optoelectronic devices also require electron insulating materials for substrates, encapsulation, and dielectric layers. Furthermore, these materials often need to be optically transparent to accommodate the inward (e.g., solar cells) or outward (e.g., light emitting diodes) transmission/coupling of light. Plastic substrates are by default the material of choice for such applications due to their outstanding mechanical flexibility and optical transparency. However, as an alternative substrate, paper made of nanofibrillated cellulose (so called “nanopaper”) is optically transparent due to largely reduced scattering⁵². Compared with plastics, nanopaper exhibits distinct advantages, including 1) a mesoporous structure enabling fluid (ink, for example) absorption for improved processability, and 2) tunable fibril and pore size for light coupling to enhance optoelectronic performance.

The multifunctionality and printability of nanopaper suggests its potential for enabling large-scale optoelectronics, and several proof-of-concept devices have been reported in the literature (Figure 4a)^{53,54}. Unlike other transparent substrates, the high scattering haze (> 90%) of nanopaper promotes light coupling into or outward from the active layer, which has been used to increase the energy efficiency for GaAs solar cells and organic light-emitting diodes (OLEDs)^{53,54}. Transistors on nanopaper have also been demonstrated⁵⁵, which can be used to turn on and off the pixels in active matrix OLED displays. However, the optoelectronic application of fibrillated cellulose faces the same challenges as plastics, particularly in terms of lifetime, such as its UV stability in solar cells for >20 years, and its thermal stability in devices such as OLEDs that often function at a very high current density (~10 mA/cm²).

Cellulose as an ionic and fluidic material platform. Water and energy scarcities are among the most challenging crises in modern society. Currently, over 2.1 billion people lack access to safe, readily available water, and around one billion do not have access to electricity⁵⁶. To address such challenges, fibrillated cellulose is a natural choice as an ionic and fluidic material considering one of its inherent functions, namely transporting water in plants. Additionally, it can be readily processed into various mesoporous structures, including multifunctional membranes (Figure 4b). The surface and bulk properties of such 2D/3D cellulose membranes can be modified for specific applications, such as by chemical modification of the surface functional groups, crystallinity control (crystalline vs. amorphous), crystal structure engineering (cellulose I or cellulose II), as well as tuning the diameter and orientation of the cellulose building blocks to control the resulting pore sizes/distribution.

Based on the pore size of a cellulose membrane, the transport behavior of ions and fluid can be categorized into three length-scales: (1) bulk behavior (e.g., the capillary effect), where the pore size is much larger than the Debye length of ions; (2) nanoscale behavior (e.g., ion selectivity due to the electrical double layer effect), where the channel size ranges from 1 nm to 100 nm; and (3) sub-nanometer behavior, where continuous transport models fail. As an excellent sub-nanometer material, sodium ion intercalation into cellulose molecular chains can open numerous ~0.6 nm

channels in the elementary fibrils, in which discrete transport behavior of ions and fluids will likely occur⁵⁷.

Due to these different transport mechanisms, fibrillated cellulose membranes have been considered attractive candidates for a wide range applications in the water-energy nexus (Figure 4b), including heavy metal/virus removal (mainly by size exclusion)^{58–60}, batteries/supercapacitors (ion selective membranes)^{61,62}, solar desalination^{63,64}, water/vapor filtration (selective vapor transport and free water blockage⁶⁵), and thermal energy harvesting (thermally-driven ion separation)^{57,66}. For example, the enhanced ion selectivity within the confined nanoscale channels in fibrillated cellulose leads to a significantly increased electrical signal under a thermal gradient, resulting in a record-high ionic Seebeck coefficient of 24 mV/K, which could be used for low-grade heat harvesting⁵⁷. Due to such novel fluidic transport mechanisms in fibrillated cellulose membranes, we can foresee significant technological potential in water and energy applications.

Bio-engineering. The high biocompatibility, high water-retaining capability, tunable mechanical properties, and good gas permeability of cellulose suggests its potential for integration with living tissue for different uses (Figure 4c)⁶⁷. For example, in wound dressing, the high water-retaining capability of fibrillated cellulose helps it to maintain a moist environment while the tunability of the mechanical properties and shape contributes to the material's excellent conformability⁶⁸. The porous structure also ensures good permeability for gas and liquid exchange, which is beneficial for wound recovery. In addition, fibrillated cellulose holds great promise in tissue engineering, providing a highly porous, robust, and biocompatible matrix to mimic the lamellar structure of natural bone and soft tissues for the construction of artificial organs and prostheses^{69, 70–72}. Fibrillated cellulose has also been used for cell immobilization and drug delivery^{73, 74}, in which the large surface charge is beneficial for binding with drugs while the excellent biodegradability and biocompatibility helps minimize side effects. Combining this capability of tissue engineering and ion manipulation, we foresee other potential uses of fibrillated cellulose in biomedical devices at the human-machine interface using ions instead of electrons (Figure 4c).

a Flexible printed optoelectronic devices

c Fibrillated cellulose in bio-engineering

b Selective transport of multiscale mass in water-energy devices

Figure 4. Fibrillated cellulose for far-term technologies, including optoelectronics, membrane technologies and bio-engineering. **a**, Layer diagrams and photographs of various fibrillated cellulose-based flexible printed optoelectronic devices, including transistors⁵⁵, solar cells⁵⁴, and OLED displays⁵³. Gr: graphene. **b**, Selective transport of multiscale mass (from solids to ions) across different length scales in various water and energy devices based on filtration, ion selectivity, solar desalination, and thermally efficient distillation. **c**, The bio-applications of fibrillated cellulose in wound repair, soft and hard tissue engineering, ion regulation, the human (ion)-machine (electron) interface, and health monitoring.

Challenges

Although proof-of-concept materials and devices have been demonstrated, there are still obstacles in the transition of fibrillated cellulose from the lab to market. The major challenges include the balance between biodegradability and product durability/dimensional stability, as well as fire safety and public health concerns. These challenges can be addressed through innovative material design and structural engineering, as well as adapting mature knowledge from related industrial fields (e.g., papermaking, wood and textiles products) without compromising the sustainability and performance of fibrillated cellulose materials.

Balance between product durability and biodegradability. Strong water absorption is an inherent property of cellulose that can facilitate its biodegradability, as most organisms and enzymes need moisture to be effective in the biodegradation process. However, resistance to biodegradation, including dimensional stability (low hygroscopicity) and water resistance, are also necessary for the product durability and lifetime of cellulose-based materials. This conflict between biodegradability and product durability is a concern that must be mitigated if cellulose is to serve as a viable alternative to traditional petroleum-based plastics.

The solution is to improve the hydrophobicity of cellulosic products when they are in service. Many strategies have been proposed to improve the hydrophobicity and reduce the hygroscopicity of cellulose materials, such as by surface coating, acylation, esterification, and hybridization with other components⁷⁵. Some of these are particularly appealing as green technologies, such as protonation treatment⁷⁵ and hybridization with natural polymers (e.g., lignin)⁷⁶. In this regard, we can utilize knowledge from the paper and wood industries on scalable and facile approaches that are known to stabilize cellulose products. For example, the paper industry routinely uses surface sizing (through impregnation or coating of waterproofing components on the surface of paper) and internal sizing⁷⁷ (in which the waterproofing component is added into the wood pulp) to reduce paper hygroscopicity. Meanwhile, the wood industry has developed various waterproofing treatments using tung oil, polyurethane, and impregnation of polymers into the wood structure (Figure 5a). We anticipate that similar techniques could be applied or adapted to stabilize fibrillated cellulose products.

Other established chemistry could also be used in a green and sustainable way (Figure 5a). For example, the cross-linking or even intercalation of ions into the molecular chain of cellulose could reduce water absorption by forming strong ionic bonds⁷⁸. Meanwhile, reversible chemistry has been demonstrated effective in improving the stability of polymers yet without compromising their biodegradability⁷⁹. For example, Diels-Alder coupling is a rapid reaction that can enable the coupling of linear polymers and nanocrystals, such as that of functionalized cellulose. Linkages made in this manner can undergo thermally induced retro Diels-Alder reactions to realize thermal recycling strategies⁸⁰. Finally, surface super-hydrophobization is another promising technique that could improve the water resistance of fibrillated cellulose products without harming their biodegradability, sustainability, or performance (Figure 5a).

Fire safety. Fibrillated-cellulose-based composites and structural designs must also consider ways to improve the material's fire safety without sacrificing its biodegradability, sustainability, or performance. Towards this aim, researchers have demonstrated the ability to modify cellulose with phosphate groups to improve the fire-retardancy of the material^{81,82}. Another common practice in industry for creating fire-retardant composites include the combination of cellulose with inorganic particles such as asbestos (aluminum silicate fiber), talc, calcium silicate, calcium carbonate, and clay^{83,84}. Some recent works have also demonstrated the development of greener fire protection technologies for cellulose-based materials, such as through the formation of strong ionic bonds⁷⁸, the impregnation of flame retardants into the wood cells^{85,86}, and through structural engineering at multiple length scales to prevent cross-plane heat conduction and gas penetration⁸⁷. In addition, maintaining the mechanical strength of cellulose-based structures under fire exposure is critical for gaining more rescue time. This could be a future research direction to further improve the fire safety of cellulose-based structures in construction applications.

Health and public safety. Cellulose is considered a relatively safe material. Microcrystalline cellulose is used as a pharmaceutical excipient⁸⁸ and is considered safe for human consumption.

While little work has been reported about the effect of fibrillated cellulose in human digestion, recent results from in-vitro systems suggest several beneficial effects⁸⁹. The paper industry has also used highly mechanically-treated cellulose-rich fibers and no risks have been detected with these materials. Regardless, there is a need to manage consumer perceptions of nanomaterials to promote acceptance as we begin to manufacture these compounds at a larger scale. The concerns regarding consumer safety particularly focus on the inhalation risk of dry nanomaterials and the migration of nanomaterials to food⁹⁰. Fortunately, fibrillated cellulose is often supplied in gel forms, and once water is removed during application, there should be no concerns of inhalation given the aggregation of the material.

Global outlook of industrialization opportunities

To benefit from the novel properties and potential applications of fibrillated cellulose, we also have to consider its cost-competitiveness with traditional technologies in potential high-volume markets. Industrial-scale manufacturing will help lower costs, but it is also necessary to consider the morphology, water content, and functionality of the fibrillated cellulose depending on the application. With these in mind, we offer our perspective on a few key factors related to the industrialization opportunities of this material.

(1) Selection and availability of feedstock for global use. The type of cellulose feedstock has an impact on the performance and manufacturing cost of fibrillated cellulose. The pulp price varies depending on the species and pulping process, and different pulp grades require different refining strategies, resulting in varying process efficiencies and energy consumption. Wood, as the main feedstock for the pulp and paper industry, is expected to maintain this position as a source material for fibrillated cellulose. However, pulp and paper feedstocks also include non-wood species, such as bamboo and sugarcane, which could have unique advantages in fibrillated cellulose production.

(2) The use of different morphologies of fibrillated cellulose. The properties and manufacturing costs of fibrillated cellulose also vary depending on its morphology (e.g., size distribution and fibrillation degree). For example, products with smaller cellulose fibril dimensions and a higher fibrillation degree are generally higher cost compared to microfibrils and their bundles. The tradeoff between performance and cost must therefore be carefully considered in the selection of fibrillated cellulose with different morphologies.

(3) Wet vs. dry products. The water content of fibrillated cellulose will play a critical role in its storage, transport, and product use. Fibrillated cellulose is typically available in three different forms: as a wet gel with a solid content of 2–10 wt%, a wet cake with a solid content of 20–25 wt%, and a dry powder. For high volume applications (e.g., paper and board products, food, concrete, paints and coatings, inks, and adhesives), where fibrillated cellulose is incorporated relatively easily, the wet gel or wet cake forms are preferred as “drop-in” solutions (Figure 5b). For cosmetics and biomedical applications that require fibrillated cellulose to be hydrated in an aqueous system, the wet gel or wet cake are also preferred. For applications such as engineering plastics, textiles, foams, films, and solvent-based functional coatings, a dry powder is necessary due to these systems’ incompatibility with water.

(4) Cellulose vs. cellulosic materials. In addition to pure cellulose, cellulosic materials that contain some residual hemicelluloses and/or lignin after fibrillation are another attractive group of functional materials for various applications^{16,91}. These residual compounds can potentially

improve the properties of composite materials and even add new functions. Cellulosic materials are also much more cost competitive compared with pure cellulose due to the reduced or less intense processing involved.

(5) Synergy between the paper/wood industries and emerging fibrillated cellulose technologies.

Pulp and paper producers have sought to develop new wood-derived biomaterials and advanced composite materials to offset the decline in the printing segments of the industry. Integrating the manufacturing of fibrillated cellulose products with the existing forest and paper industries would be a synergistic approach for decreasing the production cost at large scales. Integrated fibrillated cellulose manufacturing plants should be able to support large volume applications, such as paper, packaging, and coatings, while standalone manufacturing plants should focus on high-end applications, such as cosmetics, homecare, biomedical care, electronics, etc. (Figure 5b). Several international companies, such as Sappi, Nippon, and Kruger, have successfully demonstrated the production of fibrillated cellulose products through either standalone or integrated plants. Additionally, scaling of emerging technology can be achieved by incorporation into existing products. For example, StoraEnso AB is using cellulose nanofibers as a surface treatment in some of their board products to create smooth surfaces with added strength. The widely used roll-to-roll manufacturing facilities from the paper industry have also been adopted for the mass production of cellulose nanofibers by Japanese companies like Nippon. These trends suggest the future of the paper industry in sustainable materials with low environmental impact.

a Strategies to improve stability without compromising biodegradability

b Commercialization routes

Figure 5. Future academic research and industrialization opportunities. a, Strategies towards the balance of biodegradability and product durability/dimensional stability. Mature technologies from the paper (e.g., surface sizing and internal sizing) and wood (e.g., surface coating and impregnation) industries can be readily adopted. Additionally, emerging techniques, such as ionic cross-linking, superhydrophobic structures, and reversible chemistry design are under development. **b,** Commercialization routes for fibrillated cellulose. FC: fibrillated cellulose; MFC: microfibrillated cellulose; NFC: nanofibrillated cellulose.

Conclusion

Fibrillated cellulose derives from incredibly familiar and commonplace materials, but with the benefit of more than twenty years development by nanoscience. While the bottleneck for technology deployment of many other nanoscale materials is the scalable manufacturing, cellulose is produced daily by $\sim 3,000,000,000,000$ trees⁹² and other plants, such as fast-growing bamboo and sugarcane. As a result, fibrillated cellulose provides a nearly unlimited resource for functional composite materials. With continuing reduction of cost and increase in performance, we anticipate exponential growth in the production and usage of fibrillated cellulose as a sustainable technological material for addressing global challenges. In the near term, fibrillated cellulose has enjoyed promising success as a material for lightweight structures, energy-efficient green buildings, and biodegradable and sustainable technologies, with significant progress in manufacturing the material beyond pilot scale. However, further efforts are required to increase the material durability and reduce the cost, particularly for nanoscale fibrillated-cellulose-based products. In the far term, we foresee the potential use of fibrillated cellulose towards printed optoelectronics, affordable clean water, and biotechnologies, with paradigm shifting applications pending further study. Note that nearly all the development in the past decade of fibrillated cellulose research has been limited to no smaller than the elementary fibril level. There is much room for exploration of fundamental science and technologies at the molecular level in the sub-nanometer region, which could open unprecedented opportunities. We are at the beginning of a new era in the adoption of abundant and sustainable fibrillated cellulose for an exciting array of solutions to address societal needs for low-cost, high-performance materials with minimal environmental impacts. Combining its economic and environmental benefits with the facile physical and chemical tunability of its fibrous structure, fibrillated cellulose is highly competitive among low dimensional materials. When aided by advances in novel industrial processing and fundamental understanding, we anticipate fertile opportunities for fibrillated cellulose as one of the leading low-dimensional materials for advanced solutions towards sustainability and beyond.

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Author contributions

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Competing interests

The authors declare no competing interests.