Perspective

Developing fibrillated cellulose as a sustainable technological material

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Cellulose is the most abundant biopolymer on Earth, found in trees, waste from agricultural crops and other biomass. The fibres that comprise cellulose can be broken down into building blocks, known as fibrillated cellulose, of varying, controllable dimensions that extend to the nanoscale. Fibrillated cellulose is harvested from renewable resources, so its sustainability potential combined with its other functional properties (mechanical, optical, thermal and fluidic, for example) gives this nanomaterial unique technological appeal. Here we explore the use of fibrillated cellulose in the fabrication of materials ranging from composites and macrofibres, to thin films, porous membranes and gels. We discuss research directions for the practical exploitation of these structures and the remaining challenges to overcome before fibrillated cellulose materials can reach their full potential. Finally, we highlight some key issues towards successful manufacturing scale-up of this family of materials.

Exiting the fossil fuel era towards a sustainable future will require high-performing renewable materials with low or even net-zero carbon emission. Cellulose is a promising candidate as the most abundant renewable biopolymer on Earth, where it exists as a structural component in the cell walls of plants and some species of algae, as well as biofilms secreted by bacteria (Fig. 1a)[1]. In addition to its advantage as a potentially sustainable material, cellulose enables multiple functions and transformative applications that derive from its unique multidimensional structure. Cellulose fibres can be separated into fibrils of decreasing diameter (ranging from less than 100 μm to around 2–4 nm) that are ultimately composed of ordered linear cellulose molecular chains (Fig. 1b). Owing to this hierarchical structure, fibrillated cellulose features substantial tunability in terms of its morphology and fibril size[2], which enables unique mechanical, optical, thermal, fluidic and ionic properties that far surpass those of the parent cellulose fibres.

In this Perspective, we explore the emerging potential of fibrillated cellulose, particularly as a sustainable and practical alternative to current technological materials. For clarity, we use the term 'fibrillated cellulose' to describe cellulose fibres that have been broken down into smaller fibrils[3] and we note that nanoscale versions are also referred to as nanofibrillated cellulose, cellulose nanofibres and nanocellulose in the literature. Wood has been modified via various top-down approaches to take advantage of these cellulose fibres within the cell walls to produce structures such as super-strong wood, transparent wood and cooling wood for lightweight and energy-efficient building applications[4]. However, such engineered wood does not involve breaking down the cell walls or the cellulose fibres into smaller, free-standing fibrils, making it a separate material category that is beyond the scope of this discussion.

Fibrillated cellulose has attractive, tunable properties and is biocompatible, suggesting the potential for practical implementation and commercialization. Furthermore, fibrillated cellulose is much less expensive than metal and petroleum-based nanomaterials (approximately 2020 US$0.60 per dry kilogram for papermaking-grade fibrillated cellulose and approximately US$20 per dry kilogram for nanoscale fibrillated cellulose)[5] and can be manufactured at industrial scale, providing an additional economic advantage. The accelerated adoption of fibrillated cellulose is expected to facilitate the shift from petroleum- to bio-based products in support of a more sustainable circular economy[6] (Fig. 1c).

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 and biodegradable technologies, as well as far-term applications in optoelectronics, bio-engineering and membrane science (Fig. 1d). In this Perspective, we will discuss the potential, progress and challenges of fibrillated cellulose for various practical uses with growing market potential, including multiscale fibres, bioplastics, nanopaper, porous membranes and soft gels. We believe these growing applications, increasing biorefineries and the commercialization of fibrillated cellulose indicate its importance as a sustainable technological material.

Multiscale fibres

Cellulose has appealing intrinsic mechanical properties, with a theoretical modulus of about 100–200 GPa (about 63–125 GPa g^{-1} cm^3) and tensile strength of about 4.9–7.5 GPa (about 3.0–4.7 GPa g^{-1} cm^3) in its

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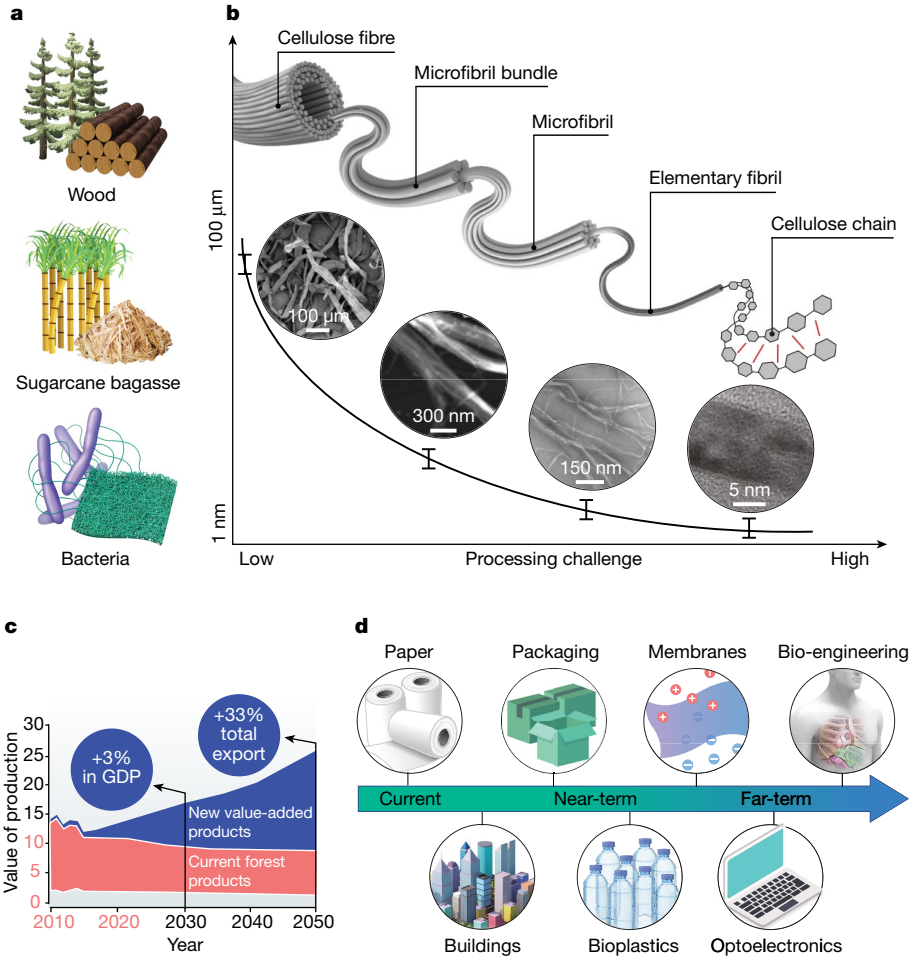

Fig. 1 | An overview of fibrillated cellulose. a, Several common source materials of fibrillated cellulose. **b**, Schematic description of the hierarchical structure and manufacturing challenge of fibrillated cellulose. The degree of fibrillation refers to the extent to which the fibres have been longitudinally split into thinner fibrils[119]. The microscopy images were taken from refs. [1,120–122] **c**, Forecast of the total production value of forest-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. The units are in millions of 2015 euros (2020 US$1,187 million) and the data used to draw the curve are an estimate. Data from ref. [6] with adaptations provided by authors at the VTT Technical Research Centre of Finland for use in communications on behalf of the Finnish Bioeconomy Cluster, FinnCERES[123]. **d**, A roadmap of fibrillated cellulose technologies, including current application in paper, near-term applications in speciality packaging, bioplastics, lightweight structural materials, and energy-efficient buildings and transportation, as well as far-term technologies, including porous membranes for energy and water, optoelectronics and bio-engineering.

crystalline form[7–9], both of which are higher than most metals, alloys, synthetic polymers and many ceramics (Fig. 2a). This mechanical strength partially derives from the densely distributed hydroxyl groups (three groups per anhydroglucose unit) on the cellulose molecular chains, which are critical for forming abundant inter- and intramolecular hydrogen bonds (Fig. 2b), especially within the fibrils. Van der Waals interactions are also important owing to their longer interaction range compared with hydrogen bonding. Furthermore, the fibril network provides physical entanglement, which helps to toughen the material[10]. As building blocks, these cellulose fibrils can be processed into various macroscopic structures (for example, composites and macrofibres), which feature enhanced mechanical properties as a result of these molecular interactions. Also, given the low density (about 1.6 g cm^{-3}) of the constituent fibrils, cellulose-derived products are particularly attractive as lightweight structural materials.

Much progress has been made to improve further the mechanical properties of materials made of fibrillated cellulose through advanced structural design and engineering of the fibrils at multiple length scales (Fig. 2c)[11–17]. Reducing the size of the fibril building blocks and porosity of the final products is an effective way of improving the mechanical strength and toughness. For example, the mechanical tensile strength of films made of only nanocellulose fibrils (that is, no other polymers) can reach up to about 300–500 MPa, which is much higher than conventional paper made from loosely packed microscale fibres[16–22]. Aligning cellulose fibrils is another effective design and engineering strategy to reduce structural defects (such as pores), to enhance the interface between cellulose fibrils and fibril aggregates and to strengthen the molecular interactions at multiple length scales[23–25].

The rich hydroxyl groups on fibrillated cellulose also provide opportunities for chemical functionalization and hybridization with other building blocks (for example, graphene oxide[26], graphite[27], clay[28], polymers[29], and so on) to further improve the mechanical properties. As a result of such modification, some recently developed cellulose composites have demonstrated a tensile strength of about 400–1,000 MPa and a high toughness of up to about 30 MJ m^{-3} (refs. [22,27,29]). These values are comparable to those of carbon-based and glass-fibre-based composite materials used in vehicles. Assembling cellulose fibrils into macrofibres with a similar diameter to commercial fibres (for example, carbon fibres and glass fibres) provides another general strategy to incorporate fibrillated cellulose for structural applications. As a demonstration of this concept, bacterial cellulose nanofibrils have been assembled into macrofibres by a wet-twisting and dry-fixing method

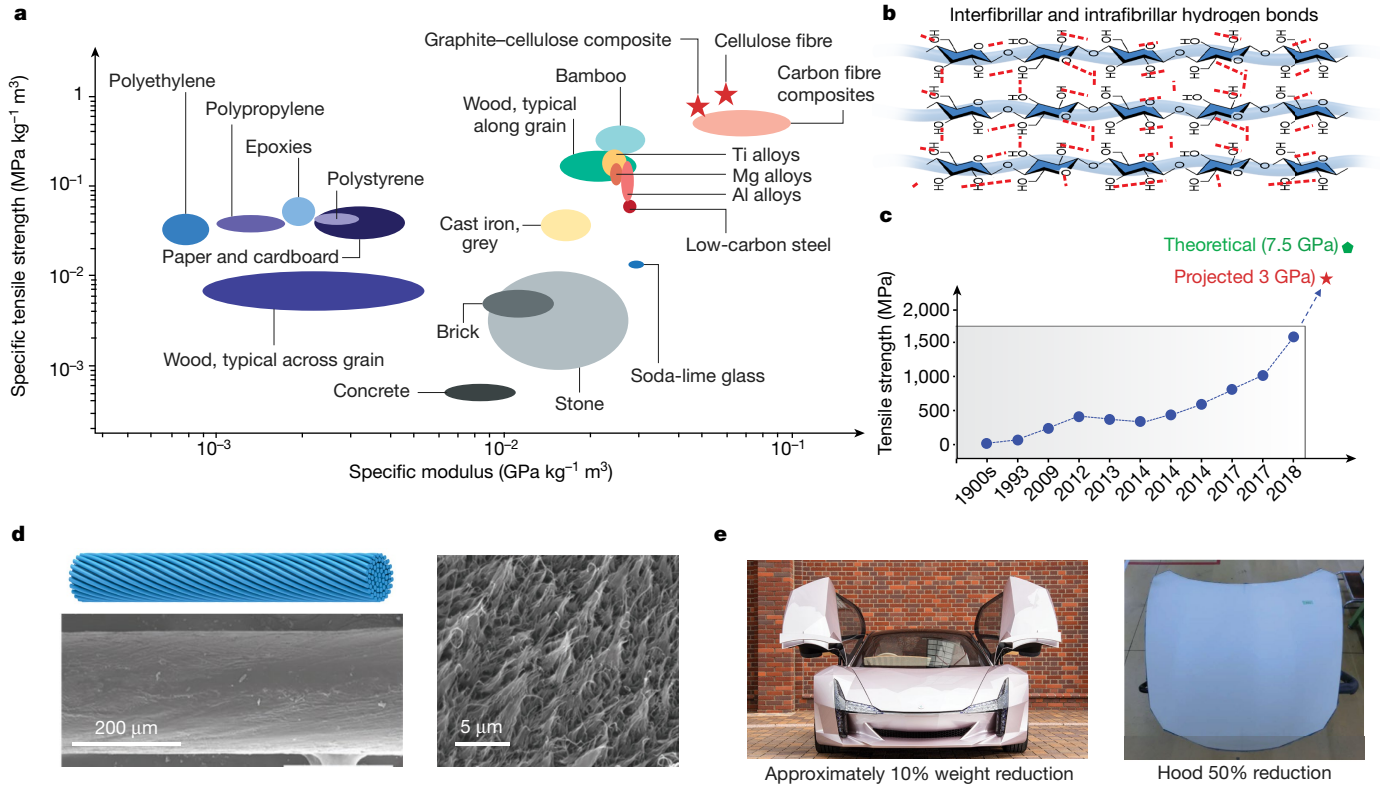

Fig. 2 | Fibrillated cellulose as a lightweight structural material. **a**, Comparison of the specific tensile strength and modulus of several recently developed cellulose structures (for example, a graphite–cellulose composite[27] and aligned cellulose macrofibres[12]) compared with conventional structural materials, including polymers, concrete, wood, metals and alloys. Drawn using Cambridge Engineering Selector Edupack Software (Granta DesignLimited, https://www.ansys.com/products/materials/granta-edupack). **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 human-made cellulose structures[11–16,20,21]. **d**, Strong cellulose macrofibres comprised of numerous aligned fibrils at multiple length scales[30]. **e**, Lightweight vehicle with cellulose materials composing different automobile parts, showing a total weight reduction of more than 10% (ref. [32]) (photos are provided by Ministry of the Environment, Japan).

that resulted in a tensile strength of around 800 MPa and a modulus of around 66 GPa (Fig. 2d)[30]. Similarly, a record-high mechanical tensile strength of around 1.6 GPa and a Young's modulus of around 86 GPa were achieved by assembling oriented nanoscale cellulose fibrils into bulk cellulose macrofibres through microfluidic spinning[12]. However, it remains challenging to manufacture long, continuous macrofibres with such high strength using fibrillated cellulose owing to structural defects such as pores, voids and inhomogeneous aggregation.

Given their potential for large-scale manufacturing and light weight, cellulose materials are particularly attractive for structural applications with improved energy efficiency. For example, a 10% weight reduction in vehicles is expected to result in a saving of about 6%–8% in fuel consumption[31]. In the search for a more lightweight and sustainable material for use in cars, the Ministry of the Environment of Japan recently launched the Nano Cellulose Vehicle project[32], which aims to develop nanocellulose composite resins for a 10%–50% weight reduction in automotive components (engine parts, the hood and other structural features) and a total weight reduction of over 10% for the entire vehicle (Fig. 2e). Given fibrillated cellulose's superior mechanical properties and much lower weight than traditional car components, these efforts could soon lead to improved mileage and lower greenhouse gas emissions without sacrificing driver safety.

Bioplastic

In addition to structural applications, fibrillated cellulose may serve a role in the growing demand for more sustainable alternatives to plastic. 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 the annual primary production due to plastic wastage from prior years[33–35]. As a result, there has been growing interest in bioplastics made from renewable and sustainable materials for packaging, textiles and other consumer products[36,37]. Most petroleum-based plastics take hundreds of years to degrade owing to their crosslinked covalent bonds, particularly the strong carbon–carbon bonds. In contrast, the oxygenated molecular chains of cellulose can be degraded by bacteria, fungi and yeasts that occur naturally in soil[38]. This combination of excellent biodegradability, outstanding mechanical strength, and the high heat and chemical resistance of fibrillated cellulose suggests its potential as an alternative for plastics such as polyethylene and polyethylene terephthalate.

Various 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 fields[39]. In general, the processing of fibrillated cellulose involves water dispersion, which is different from the melt-extrusion process of conventional plastics. However, the latter method can also be used when fibrillated cellulose is hybridized with synthetic polymers to improve extrusion processability[40,41].

The development of films made of either pure or polymer-hybridized fibrillated cellulose is another way to reduce plastic usage[42]. There are already commercial applications of cellulose in grease and oxygen barrier packaging, agricultural mulch films, containers, adhesives,

hygienic disposables, home furnishings and textile sizing agents[43]. The global demand for different types of fibrillated cellulose films is expected to continue to grow owing to the material's decreasing cost, increasing world population and demand, and government legislation towards sustainability. Efforts are well on the way to improve the performance of fibrillated cellulose further in practical applications, where traits such as water resistivity, durability and process scalability are necessary, as we discuss in more detail below.

To promote the use of fibrillated cellulose in competition with plastics will require: (1) life-cycle assessment 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 in use and biodegradability when disposed (see the 'Challenges' section for detailed discussion); and (4) a recycling process for fibrillated-cellulose-containing composites. We note that while fibrillated cellulosic materials can be extracted from plants, the processing could be chemically and energy intensive, which from the life-cycle assessment point of view may not necessarily be sustainable. For example, extensive chemical processing is usually used to achieve the high degrees of fibrillation necessary to produce cellulose nanomaterials. These chemistries include concentrated sulfuric acid hydrolysis[44] and 2,2,6,6-tetramethylpiperidine-oxyl (TEMPO)-mediated oxidation. To avoid the use of such chemically and energy-intensive treatments, researchers are exploring recyclable chemicals, such as solid di-carboxylic acids[45] and enzymes[46–48], particularly novel enzymes such as lytic polysaccharide monooxygenases[49,50]. Lytic polysaccharide monooxygenases promote oxidation at the C1 or C4 position of the cellulose macromolecule, which induces chain breaks, facilitating the fibrillation of cellulose fibres under milder conditions[49,50]. With the aim of improving the sustainability of highly fibrillated cellulose, we can look to the knowledge and expertise accumulated by the pulp and paper industries to develop greener fabrication methods for fibrillated cellulose-based bioplastics.

Thin films and coatings

Like the microscale cellulose fibres that compose traditional paper, nanofibrillated cellulose or nanocellulose can be assembled into free-standing thin films and coatings on substrates with a thickness typically smaller than about 100 μm, which are often referred to as 'nanopaper'. The structure of nanopaper provides a range of attractive properties that suggest this material for various applications. For example, the tunable porosity and pore size of nanopaper enables its optical properties (for example, transmittance and haze) to be modulated depending on the desired application. With this aim, an early work reported a nanopaper based on nanofibrillated cellulose featuring an optical transmittance of up to about 70% in the visible range and a mechanical strength of up to 223 MPa (ref. [51]). Subsequently, highly transparent nanopaper (up to 92%) has been reported by different research groups, with a wide range of transmittance haze for different applications (for example, high haze for solar cells and low haze for displays; Fig. 3a)[52–57]. Nanopapers featuring carefully tuned microstructures have also demonstrated high forward transmittance, high reflection (with a brightness of up to 80%)[58] and even structural colour[59].

In addition to optical properties, the high packing density of the nanoscale fibrils produces a smooth surface with high gas barrier resistance (for example, against oxygen)[16], while the porous structure yields a low thermal conductivity. As a result, nanopaper has been incorporated into several emerging applications. For example, the excellent gas barrier properties (oxygen permeability of less than 40 cm^3 μm m^{-2} day atm) of nanopaper have resulted in the material's commercial use for packaging applications[60]. Meanwhile, window coatings have been developed using self-assembled cellulose nanofibres with a high transmittance (around 90%), a low optical haze (around 6%) and a low thermal conductivity (around 8 mW mK^{-1}) for improved energy efficiency in buildings[61]. Cellulose is also considered a possible coating material for radiative cooling applications owing to its high emissivity in the infrared range[62,63].

Flexible and transparent nanopaper is also particularly attractive for optoelectronics, as a replacement for plastic—the default material of choice for such applications owing to the need for outstanding mechanical flexibility. 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 (for example, solar cells) or outward (for example, light-emitting diodes) transmission/coupling of light[52–57]. 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 the 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 (see Fig. 3a). 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 solar cells and organic light-emitting diodes[52,64]. Transistors on nanopaper have also been demonstrated[65], which can be used to turn on and off the pixels in active matrix organic light-emitting diode displays. However, the optoelectronic application of fibrillated cellulose faces the same challenges as plastics, particularly in terms of lifetime, such as its ultraviolet stability in solar cells for over 20 years, and its thermal stability in devices such as organic light-emitting diodes that often function at a high current density (about 10 mA cm^{-2}).

Porous membranes

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 people do not have access to electricity[66]. To address such challenges, fibrillated cellulose is a natural choice as an ionic and fluidic material because one of its native functions involves transporting water in plants. Fibrillated cellulose can be readily processed into highly tunable, mesoporous structures, enabling the fabrication of multifunctional membranes (Fig. 3b). Furthermore, the surface and bulk properties of such two- and three-dimensional cellulose membranes can be modified for specific applications, such as by chemical modification of the surface functional groups, crystallinity control (crystalline versus amorphous), crystal structure engineering (cellulose I versus cellulose II), as well as tuning the diameter and orientation of the cellulose building blocks to control the resulting pore sizes and distribution.

Using the pore size of a cellulose membrane, the mass transport behaviour can be categorized into three length scales: (1) bulk behaviour when the pore size is much larger than the Debye length of ions (for example, the capillary effect); (2) nanoscale behaviour when the channel size ranges from 1 nm to 100 nm (for example, nanofluidic ion transport due to the electrical double layer effect; and Knudsen diffusion when the characteristic length of the pore/channel is comparable or smaller than the molecular mean free path); and (3) sub-nanometre behaviour, where continuous transport models fail. In particular, fibrillated cellulose can demonstrate sub-nanometre behaviour such as regulated ion transport upon the intercalation of sodium ions between the cellulose molecular chains, which opens numerous channels approximately 0.6 nm in diameter in the elementary fibrils, in which new transport phenomena of ions and fluids can occur[67].

Owing to these different transport mechanisms and the material's inherent advantages in terms of tunability, sustainability, abundance and scalability, fibrillated cellulose membranes have been considered attractive candidates for a wide range of applications in the

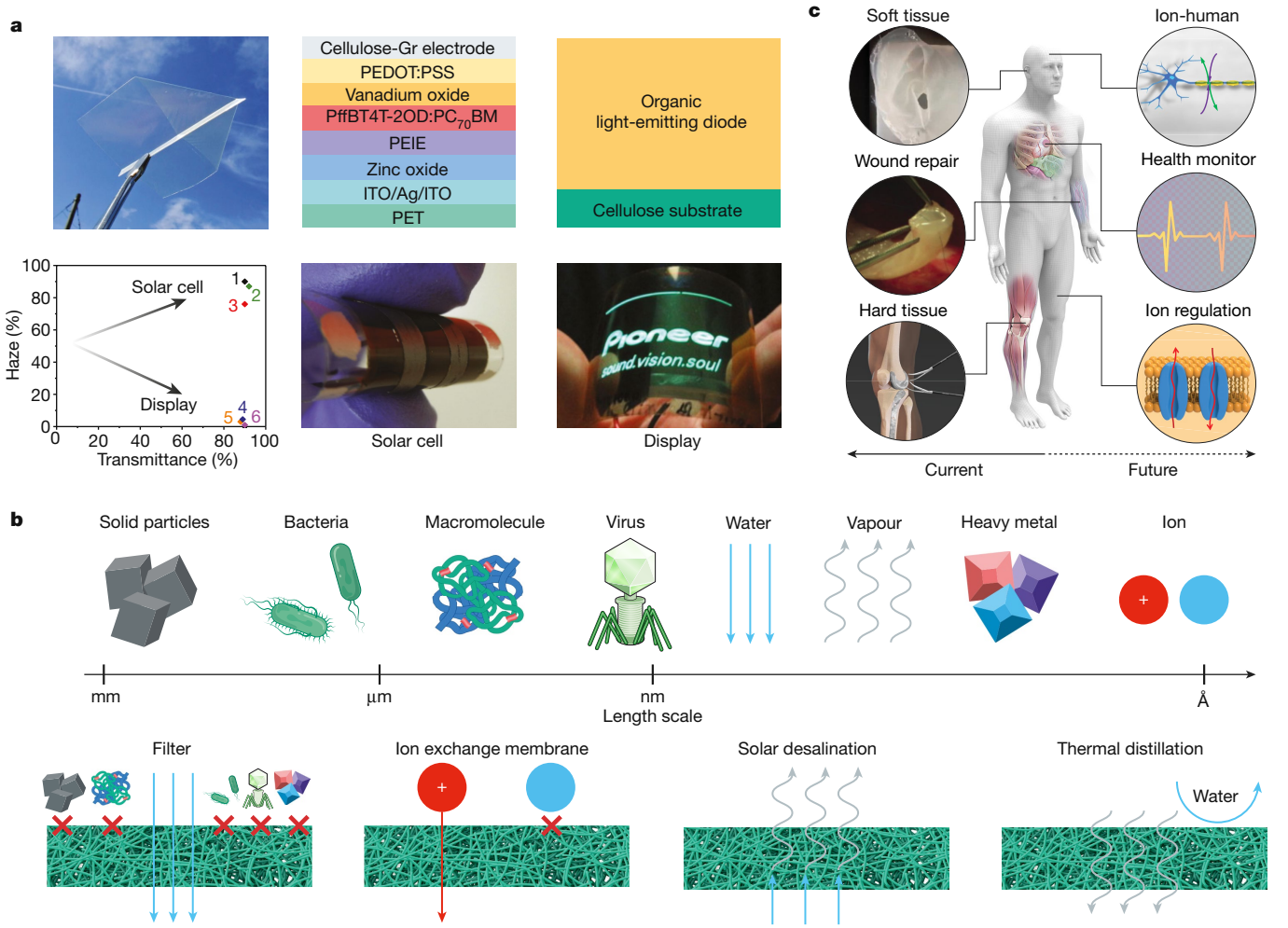

Fig. 3 | Fibrillated cellulose for far-term technologies. a, Nanopaper for optoelectronics. Left, a photograph of transparent nanopaper (top) and the optical properties (bottom) of several selective nanopapers showing a high optical transmittance and tunable transmittance haze[52–57]. Middle, a layer diagram (top) and photograph (bottom) of a nanopaper-based solar cell[124]. Right, a layer diagram (top) and photograph (bottom) of an organic light-emitting diode display[64]. Gr, graphene; PEDOT:PSS, poly(3,4 -ethylenedioxythiophene) polystyrene sulfonate; PEIE, polyethylenimine ethoxylated; PET, polyethylene terephthalate. **b**, Selective transport of multiscale mass (from solids to ions) across different length scales in various fibrillated cellulose membranes for filtration, ion selectivity, solar desalination, and thermally efficient distillation. **c**, Fibrillated cellulose soft-gels for bio-applications, including wound repair, soft and hard tissue engineering[87], ion regulation[67,72], the human (ion)–machine (electron) interface, and health monitoring.

water-energy nexus (Fig. 3b), including removal of heavy metals or viruses (mainly by size exclusion)[68,69], batteries/supercapacitors/ionic devices (ion-selective membranes)[70–72], solar desalination[73], water/vapour filtration (selective vapour transport and free water blockage[74]), and thermal energy harvesting (thermally driven ion separation)[67]. For example, the enhanced ion selectivity within the confined channels in fibrillated cellulose leads to a greatly increased electrical signal under a thermal gradient, resulting in an ionic Seebeck coefficient of 24 mV K^{-1}, which could be used for low-grade heat harvesting. These novel fluidic transport mechanisms in fibrillated cellulose membranes suggest great technological potential in water and energy applications.

Soft-gel

Fibrillated cellulose is considered a biocompatible material with applicability to a range of advanced bio-engineering fields, such as wound dressing[75], tissue engineering[76], drug delivery[77], medical diagnostics[78], smart sensors[79] and electronic skin[80]. Soft-gels (for example, hydrogels and ionic gels) made of fibrillated cellulose with the potential for integration with living tissue are particularly attractive for such bio-related applications (Fig. 3c)[76,81–83]. Various fibrillated cellulose based soft-gels with three-dimensional macromolecular networks and excellent water binding and retaining capability have been synthesized through different processes such as gelation, ionic crosslinking, spinning and 3D printing[84]. The interactions among building blocks, water molecules and/or ions during gel formation determine the structure and properties of the resultant soft-gels, and thus influence their use.

Given their biocompatibility, water-retaining capability, tunable mechanical properties and ability to regulate gas, liquid and ions inside the porous network structure, fibrillated cellulose soft-gels have advantages for bio-engineering. For example, in wound dressing, the 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 conformability[81]. The porous structure also ensures good permeability for gas and liquid exchange, which is beneficial for wound recovery. Wound-dressing products made of bacterial cellulose or nanocellulose, such as FibDex, are available in clinical applications, some of which perform better than traditional ones[82,85].

In addition, fibrillated cellulose soft gels hold promise in tissue engineering, providing a porous, robust and biocompatible matrix for the construction of artificial organs and prostheses (for example, ears) via 3D printing and self-assembly[86–88]. Fibrillated cellulose has also been used for cell immobilization and drug delivery[77,89], in which the large surface charge is beneficial for binding with drugs while the biodegradability and biocompatibility helps to minimize side effects. Products made of nanofibrillated cellulose hydrogel can be biocompatible with human cells and tissues but free from any animal- or human-derived material, making it suitable for advanced three-dimensional cell culturing and other biomedical applications[85]. Combining this capability of tissue engineering and ion manipulation, we foresee other potential uses of cellulose-based soft-gels in biomedical devices at the human–machine interface using ions instead of electrons (Fig. 3c).

Challenges

Although proof-of-concept materials and devices have been demonstrated, there are still obstacles in the transition of fibrillated cellulose from the laboratory to market. The major challenges include sustainability, the balance between biodegradability and product durability or 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 (for example, papermaking and wood and textile manufacturing) without compromising the sustainability and performance of fibrillated cellulose materials.

Sustainability

A resource such as fibrillated cellulose is only truly sustainable when its processing is also sustainable. Evaluation of the sustainability of fibrillated cellulose requires consideration of both technical economic and life-cycle assessment analyses based on pilot-scale data. Unfortunately, such information is generally proprietary and therefore we discuss this point based on the information available and subject to a number of assumptions.

The sustainability and energy requirements for the production of fibrillated cellulose are tied not only to the biomass source but also to the processing methods employed. Chemical pulp fibres are the typical source in the production of fibrillated cellulose, which are obtained from industrial processes that are environmentally friendly and cost-effective[90,91]. The pulp fibres are then chemically pre-treated, involving a catalytic reaction in aqueous phase at atmospheric pressure and low temperature[3,45]. Such pretreatments are important for substantially reducing the energy consumed in the mechanical defibrillation that follows. As an alternative source, bacteria are also widely used to produce fibrillated cellulose, generally through static and agitated cultures as well as the recently developed bioreactor-based production. Static and agitated culture approaches have been limited by low yields and long culture periods, making them inefficient for large-scale production[92,93]. However, recent advances in bioreactor-based production has greatly lessened the culture time required, improved the production yield and reduced the production cost[92,93], representing a promising path towards more sustainable and scalable fabrication of bacterial cellulose for commercial use in food, biomedicine and other fields[81,94,95].

Water utilization is another factor affecting sustainability metrics. At present, mechanical fibrillation processes are mainly conducted at low solid contents (for example, 2 wt%). However, the water in the finished fibrillated cellulose products is often used by end users during applications. In cases where a dewatering or drying step is applied after the fibrillation process, most of the water can be recycled. Furthermore, advanced technologies, such as twin-screw extrusion, can be used to fibrillate cellulose fibres at considerably higher solid contents (15–30 wt%), which substantially reduces the water footprint[96]. Clearly, fibrillated cellulose has strong potential as a sustainable material, but only if it is processed sustainably, which will require further study, particularly when scaled up.

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, the dimensional stability of cellulose-based materials, derived from low hygroscopicity and high water resistance, is also necessary to enable a desirable product durability and lifetime. Generally, cellulose hybrid materials (for example, nanocellulose polymer composites) show enhanced durability, but it comes at the sacrifice of biodegradability to some degree. This tradeoff is an issue that must be mitigated if cellulose is to serve as both a sustainable and practical alternative to traditional petroleum-based plastics.

In this regard, we can use knowledge from the paper and wood industries about scalable and facile approaches that are known to stabilize cellulose products. The general strategy is to improve the hydrophobicity and reduce the hygroscopicity. Many strategies have been proposed, such as surface coating, acylation, esterification and hybridization with other components[97]. Some of these are particularly appealing as green technologies, such as protonation treatment[97] and hybridization with natural polymers (for example, lignin)[98]. The paper industry also routinely uses surface sizing (through impregnation or coating of waterproofing components on the surface of paper) and internal sizing[99] (in which the waterproofing component is added into the wood pulp) to reduce paper hygroscopicity; Fig. 4a). Indeed, all chemical treatments that have been developed for modifying materials such as cotton and wood-based fibres could be applicable to fibrillated cellulose. However, we note that these chemicals often have a cationic charge that allows them to adsorb on the anionic fibres, which may cause flocculation and the loss of the small dimensions of the fibrillated cellulose, from which many of the material's advantages derive. An alternative approach is to first form a fibrillated cellulose structure followed by a post-treatment with the desired chemistry to ensure the targeted end-use properties are still achieved.

Other established chemistry could also be used in a green and sustainable way to improve the stability of fibrillated cellulose without compromising its biodegradability or performance (Fig. 4a). For example, the cross-linking or even intercalation of ions into the molecular chains of cellulose could reduce water absorption by forming strong ionic bonds[100]. Meanwhile, reversible chemistry has been demonstrated to be effective in improving the stability of polymers, but without compromising their biodegradability[101]. 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 strategies[102]. Finally, in fibrillated cellulose products (for example, nanopaper), surface super-hydrophobization is another promising technique that could improve the water resistance of fibrillated cellulose products while maintaining overall bulk biodegradability (Fig. 4a).

Fire safety

In terms of practical use, fibrillated-cellulose-based composites and structural designs must also consider ways to improve the material's fire safety. Researchers have demonstrated the ability to modify cellulose with phosphate groups to improve fire-retardancy[103,104]. Another common practice in industry for creating fire-retardant composites includes the combination of cellulose with inorganic particles such as asbestos (aluminium silicate fibre), talc, calcium silicate, calcium carbonate and clay[28,105]. Some recent works have also demonstrated the development of greener fire protection technologies for cellulose-based materials, such as through the formation of strong

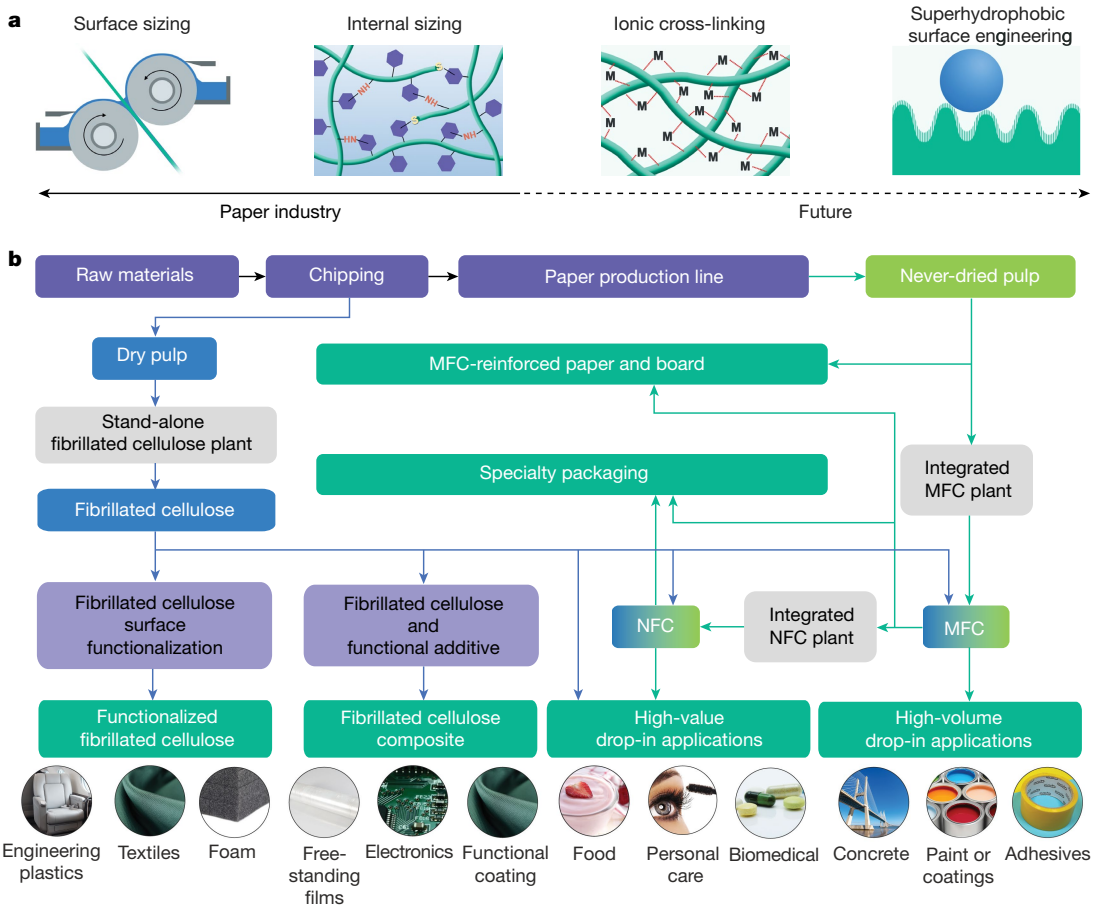

Fig. 4 | Research and industrialization opportunities. a, Strategies towards the balance of increased biodegradability and improved product durability/dimensional stability. Mature technologies from the paper industry (for example, surface sizing and internal sizing) can be readily adopted. Additionally, emerging techniques, such as ionic cross-linking and superhydrophobic structures are under development. **b**, Commercialization routes for fibrillated cellulose. MFC, microfibrillated cellulose; NFC, nanofibrillated cellulose.

ionic bonds[100], the hybridization of flame retardants[106,107], and through structural engineering at multiple length scales to prevent cross-plane heat conduction and gas penetration[108]. In addition, maintaining the mechanical strength of cellulose-based structures under fire exposure is critical for gaining more rescue time. To further improve the fire safety of cellulose-based structures in construction applications could be a future research direction.

Health and public safety

Although fibrillated cellulose clearly has strong potential in a broad range of fields, and is already being used as a result, we must also consider whether the material is safe for public use. Fortunately, cellulose is considered a relatively benign material. Microcrystalline cellulose is used as a pharmaceutical excipient[109] and is considered safe for human consumption. Although little work has been reported about the impact of fibrillated cellulose on human digestion, recent results from in vitro systems suggest several beneficial effects[110]. Fibrillated cellulose has also been used as a thickener in foods to contribute to a high-fibre diet and even made into synthetic meats (protein–cellulose mixes)[94,111], all of which are strong indicators of the excellent safety of fibrillated cellulose. The paper industry has also used highly mechanically treated cellulose-rich fibres and no resultant risks have been detected. 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 food[112]. 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.

Industrialization opportunities

To benefit from the properties, potential sustainability and 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 to lower costs, but we must consider the morphology, water content and functionality of the fibrillated cellulose, depending on the application. With these factors in mind, we offer our perspective on the industrialization opportunities of this material.

Feedstocks

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. The type of biomass (wood or non-wood, softwood or hardwood) and the pulping technology also have a direct impact on the structure of the cellulose, which ultimately affects the functionality. 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. For example, the fast growth rate of bamboo makes it widely available for fibrillated cellulose and its longer fibrils enable superior mechanical strength compared to shorter fibril feedstocks, making it more competitive for lightweight structural material applications[113,114]. Sugarcane residue (bagasse) is another vast, inexpensive feedstock that could be used for fibrillated cellulose production with process integration opportunities in sugar-based biorefineries[115].

However, in choosing feedstocks for fibrillated cellulose, we must also consider the process–structure–property relationships between the plant source and the application need. In particular, this may require researchers to consider the chemical composition of the feedstock, such as the residual hemicellulose and lignin[116], as well as the structure of the fibre walls and any interactions among the hierarchical cellulose building blocks, and how composition and structure react to different processing conditions[117].

Alternatively, the production of bacterial cellulose depends heavily on the fermentation medium that provides carbon, nitrogen and other macro- and micro-nutrients for bacterial growth. The most efficient growth of bacteria generally requires the supplement of an abundant carbon source (glucose, sucrose, organic acids, and so on) and a minimal nitrogen source.

Different morphologies

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

Wet versus dry products

The water content of fibrillated cellulose will play a critical part 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%, as a wet cake with a solid content of 20–25 wt%, and as a dry powder. For high-volume applications (for example, paper and board products, food, concrete, paints, coatings, inks and adhesives), where fibrillated cellulose is incorporated relatively easily, the wet gel or wet cake forms are preferred as 'drop-in' solutions (Fig. 4b). 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, owing to these systems' incompatibility with water.

Cellulose versus cellulosic materials

Purity, as an influential factor on the manufacturing cost and properties of fibrillated cellulose, must also be considered in its commercialization. Notably, high-purity fibrillated cellulose does not necessarily perform better than low-purity materials, depending on the application. For example, hemicelluloses and lignin are ubiquitous in cellulosic materials. Residual amounts of these components after fibrillation, while lowering the material's purity, can also potentially improve the properties of resulting composites and even add new functions[23,116]. Cellulosic materials are also much more cost competitive than pure cellulose owing to the reduced or less intense processing involved.

Synergy with the paper/wood industries

Pulp and paper producers have sought to develop 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 to 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, whereas standalone manufacturing plants should focus on high-end applications, such as cosmetics, homecare, biomedical care, electronics and so on (Fig. 4b). Already we observe such trends, as several international companies, such as Sappi, Nippon Paper and Kruger, have successfully demonstrated the production of fibrillated cellulose products through either stand-alone or integrated plants. Additionally, scaling of emerging technology can be achieved by incorporation into existing products. For example, cellulose nanofibres are used as a surface treatment in some of these companies' 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 nanofibres in Japan. These trends suggest the future of the paper industry lies with sustainable materials with low environmental impact.

Conclusion

Whereas the bottleneck for technology deployment of many other nanoscale materials is scalable manufacturing, cellulose is produced daily by approximately 3,000,000,000,000 trees[118] and other plants, such as fast-growing bamboo and sugarcane. Therefore, fibrillated cellulose provides a nearly unlimited resource for functional composite materials, and as such its commercialization in a wide range of products has accelerated over recent years as producers in the European Union, Japan, Korea, China and North and South America bring products to market. Successful commercialized examples include skin-care products using fibrillated cellulose as a rheology modifier, sports products such as the badminton racket, using fibrillated cellulose for reinforcement, and ballpoint pens using fibrillated cellulose as a thickening agent. This acceleration is driven in part by consumers' increasing awareness of sustainability and renewability, creating a strong incentive for brand owners to reduce carbon footprints, as well as affecting global regulatory policies. However, the most important driving force in the commercialization of fibrillated cellulose derives from paper and pulp producers, who are facing the challenge of the declining printed market, forcing them to pivot their traditional business models towards biorefineries and the production of biomaterials.

With continuing reduction of cost and increase in performance, we anticipate ongoing 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 substantial progress in manufacturing the material beyond pilot scale. However, further efforts are required to increase the material durability and reduce the cost, particularly for products based on nanoscale fibrillated cellulose. We foresee the use of fibrillated cellulose in printed optoelectronics, affordable clean water and biotechnologies, with commercialization potential pending further study. However, we note that true sustainability, from raw materials to manufacturing and final products, remains a challenge that requires further study from the academic community and industry in order to reduce energy and water consumption and to balance sustainability and performance better (for example, the competition between biodegradability and material stability). In addition, nearly all the development in the past decade of fibrillated cellulose research has been limited to sizes no smaller than the elementary fibril level. There is much more room for exploration of fundamental science and technologies at the molecular level in the sub-nanometre region of this material.

We anticipate 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 industrial processing and fundamental understanding, we anticipate fertile opportunities for fibrillated cellulose as one of the leading low-dimensional materials for advanced solutions towards addressing global water, energy and environmental challenges in a sustainable way.

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Additional information
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