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REVIEW

# Turning Natural Herbaceous Fibers into Advanced Materials for Sustainability

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## Abstract

Considering the growing concerns about natural resource depletion, energy inequality, and climate crises, biomass-derived materials—the most abundant organic matter on the planet—have received a lot of attention as a potential alternative to petroleum-based plastics. Herbaceous biomasses and extracted cellulose have recently been extensively used in the development of high-performance and multifunctional materials. Herbaceous biomass has sparked interest due to its species diversity, abundance, low cost, lightweight, and sustainability. This review discusses the structure versus property relationships of various sources of herbaceous biomasses (e.g., sugarcane, straw, and bamboo) and their extracted biomaterials, as well as the latest emerging applications from macro- and microscales to nanoscales. High-strength structural materials, porous carbon materials, multichannel materials, and flexible materials are examples of these applications, which include sustainable electronics, environmentally friendly energy harvesting, smart materials, and biodegradable structural buildings.

**Keywords** Sustainability · Herbaceous biomass · Biomaterials · Biodegradable · Advanced applications

## Introduction

Over the last decade, sustainability and the effects of industrial practices on the environment have received tremendous attention. Concerns regarding climate change have risen globally, and environmental experts have warned that ecosystems worldwide have suffered irreversible damage. Future generations may face the depletion of natural resources, energy inequality, and widespread climate change if human behavior towards the consumption and disposal of non-renewable or environmentally harmful materials does not change quickly [1, 2]. Material developers and suppliers can make a difference by implementing sustainable and responsible business practices and reducing their dependence on non-renewable resources such as fossil fuels. Natural materials have emerged as viable alternatives to petroleum-based plastics in a variety of applications, including everyday products and advanced materials.

Cellulose is the primary product of plant photosynthesis and is the most abundant organic matter on earth [3]. Cellulose-based materials are attractive in the industry because of their abundance, cost efficiency, carbon neutrality, and excellent biodegradability [4, 5]. As research in the field of natural and cellulose-based materials has advanced, so have the applications for these materials, which include materials for environmental pollution control, optical electronics, renewable energy harvesting, sustainable packaging, and biodegradable structural materials [6]. Currently, most of the research interest in this field is focused on wood-based materials. Nevertheless, the main source of wood, the global forest reserves are dwindling. According to the estimation by the Food and Agriculture Organization of the United Nations in 2015 [7], the countries with the greatest deforestation area during 2010s were Brazil (18.9 million ha), the Democratic Republic of the Congo (11 million ha), Indonesia (8 million ha), Angola (5.6 million ha), and Canada (4.5 million ha). Trees efficiently absorb carbon dioxide and protect the environment [8, 9]; excessive logging

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could cause environmental issues such as climate change, soil erosion, loss of habitat, and environmental degradation [10, 11]. Furthermore, trees typically take many years to grow and mature. For example, pine trees are one of the most commercially important tree species for construction and wood pulp but they typically take ~ 25–30 years to harvest. Although some fast-growing tree species have been genetically engineered to be ready for logging in just a few years, the development of these high-turnover forests can lead to a rapid loss of soil fertility [12]. Therefore, cellulose-based materials from non-wood sources must also be developed to increase human reliance on renewable resources.

Herbaceous biomass is a ubiquitous non-woody cellulose source that can be found in different forms. Most herbaceous biomass species exhibit similar compositions and hierarchical porous structures to wood [13–15] (Fig. 1a), but with a much shorter harvest time, lower prices, and no deforestation concerns (Fig. 1b). Herbaceous biomass has long been used for fuel, animal feed, fertilizers, construction, textiles, food, and chemicals [16–20], but its utilization rate and efficiency are typically low (Fig. 1c). More than $1.55 \times 10^8$ MT of agricultural waste is generated every year, most of which is left on the ground after harvest and used as landfill materials or directly burned, causing air and water pollution [21, 22]. Finding new applications for herbaceous biomass could add significant value to these waste or byproduct streams while avoiding environmentally harmful practices.

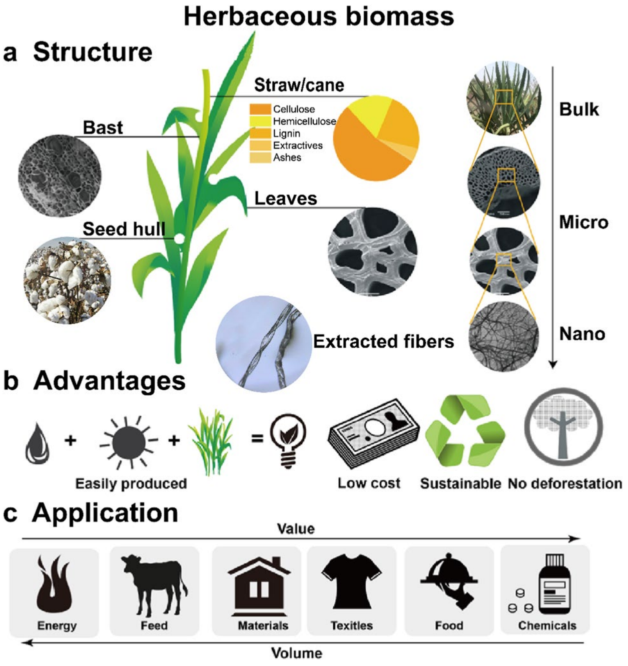

**Fig. 1** Schematic illustration of the main topics of this review. **a** Compositions and morphologies of herbaceous biomass from different sources and plant parts [27–32], **b** its advantages, and **c** its applications

Herbaceous biomass-derived materials can be designed in terms of structure and compositions from macro- to micro- and to nanoscales due to their hierarchical porous structure. The use of herbaceous biomass-derived materials has greatly expanded in recent years, particularly in advanced functional applications. Although both herbaceous biomass and wood are primarily composed of cellulose, hemicellulose, and lignin, their morphologies can differ greatly, resulting in varying densities, strengths, mass (water and nutrients) transport properties, and thermal conductivities. It is crucial to understand these differences to develop modification or compatibilization strategies for herbaceous biomass-derived materials that serve in applications such as high-strength structural materials, carbonized conductive materials, ion-selective membranes, and highly flexible substrates. Moreover, in the preparation of functional materials, herbaceous biomass requires less energy, instrument investment, and chemical treatment than wood [23].

The paper aims to review the structure-property relationships of various sources of herbaceous biomass (cane, straw, leaf, bast, and seed) and their extracted cellulose materials, as well as the latest high-performance applications using material ranging from macro- to micro- and to nanoscales. Several excellent reviews have been published to date on cellulose-based and wood-based advanced functional materials [24–26]. However, the literature lacks a detailed compilation and comparison of recent advances in the applications of herbaceous biomass from different sources. This review will link various morphologies and compositions of herbaceous biomass with applications in emerging fields such as sustainable electronics, environmentally friendly energy harvesting, smart materials, and biodegradable structural materials. Although herbaceous biomass is a promising sustainable material, its widespread adoption faces numerous challenges, and both scientists and governments must collaborate to promote it.

## Compositions and Structure of Herbaceous Biomass

Herbaceous biomasses are complex, hierarchical, and porous 3D composites, mainly having cellulose, hemicellulose, lignin, extractives, and ashes. Because of the species diversity and various plant organs (e.g., cane, straw, leaves), herbaceous biomass from different origins exhibits significantly variable biological structures, morphologies, and chemical compositions. A thorough knowledge of these differences is necessary to tailor effective modification strategies for high-performance multifunctional materials.

## Compositions of Herbaceous Biomass

Cellulose, hemicellulose, and lignin are the primary components of herbaceous biomass and contribute to the formation of cell walls that serve as its supporting skeleton. The weight of these three components accounts for 80–95% of the total mass of most herbaceous biomass species (e.g., the cellulose content of cotton is as high as 90 wt%) [33, 34]. The composition and distribution of herbaceous biomass significantly affect the physical properties such as strength, hydrophilicity, and hydrophobicity, determining whether it is suitable for the extraction of cellulose fibers and nanocellulose.

### Cellulose

Cellulose is a linear hydrophilic polymer with molecular formula $(C_6H_{10}O_5)_n$, consisting of D-glucose linked by β-1,4 glycosidic bonds [35]. The crystallinity, cellulose content, and degree of polymerization (DP) are the most important factors to analyze when determining the suitability of specific species or parts of a herbaceous plant.

The crystallinity of cellulose is a critical microstructural parameter that can be used to predict the mechanical performance of cellulose fibers [36]. Cellulose chains arrange linearly into highly ordered crystallites held together by van der Waals forces and inter- and intra-molecular hydrogen bonding in the crystalline regions of cellulose microfibrils [37, 38]. The amorphous regions behave as disordered structures because of their randomly arranged hydrogen bonds. Therefore, the mechanical strength of crystalline cellulose is significantly higher than amorphous cellulose; the tensile strength is 7.5 GPa, and the elastic modulus is 105–220 GPa [39]. Meanwhile, the elongation, water uptake, and reactivity for chemical modification of cellulose increase with decreasing crystallinity [36, 40, 41]. The crystallinity of natural cellulose mainly depends on its source and physical or chemical treatment (Table 1). Herbaceous biomass provides cellulose with a wide range of crystallinity. For example, the crystallinity of natural cotton (77.0%), flax (70.0%), and sisal (71.7%) is slightly higher than that of other herbaceous biomasses like rice straw (51.9%) and poplar woods (52.7%) listed in Table 1.

Cellulose content is another important parameter in determining whether the raw material is suitable for production of cellulose fibers and nanocellulose. High cellulose content is desirable because of the low energy and chemical consumption associated with a potentially higher yield. Some species of herbaceous biomass naturally contain high cellulose content, such as cotton (90 wt%), flax (72 wt%), and hemp (70 wt%), making them potent raw materials for textile, paper, and cellulose extraction industries.

The final parameter to consider when evaluating cellulose sources is the DP, which corresponds to the chain length of β-D-glucose in a cellulose molecule. In addition to the cellulose crystallinity, the molecular chain length strongly influences the physical properties of the fiber [42]. The DP also indicates the degree of cellulose degradation that occurs during extraction processes involving chemical, enzymatic, or mechanical treatments. In some cases, cellulose degradation may control the properties of the final product (such as dissolving pulp and nanocellulose). In general, cellulose degradation during processing is undesirable because it reduces yield and fiber strength, ultimately increasing production costs. Extraction techniques must be optimized and tailored toward the specific material being processed to extract the highest quality cellulose possible while balancing cost and energy consumption.

### Hemicellulose

Hemicellulose is an amorphous branched polysaccharide comprising hexoses and pentoses such as xylose, glucose, mannose, galactose, and arabinose [43]. According to the lamella model, cellulose microfibrils are embedded in amorphous substances, including hemicellulose, lignin, and pectin [44]. Hemicellulose, a hydrogen bond donor, is tightly bound to cellulose microfibrils via numerous hydrogen bonds, contributing to the mechanical strength of plant cell walls [45]. In addition, hemicellulose plays a key role in physically separating microfibrils from one another. Electrostatic repulsion generated by negative carboxyl groups on the surface of hemicellulose inhibits aggregation of the microfibrils [45]. Furthermore, hemicellulose has a low DP (normally 50–300 for hemicellulose and 1000–6000 for cellulose) and strong water swelling properties [45, 46]. Therefore, retaining hemicellulose facilitates the defibrillation process (e.g., refining and homogenization).

Most herbaceous biomasses typically contain 20–35 wt% or less hemicellulose, excluding cotton fiber, which has little to no hemicellulose. The hemicellulose in herbaceous biomass is mainly arabino-4-O-methylglucuronoxylan, whereas softwood and hardwood hemicelluloses are O-acetyl-galactoglucomannan and O-acetyl-4-O-methylglucuronoxylan, respectively [47, 48]. Different hemicellulose types are conducive to unique biomechanical properties, with glucomannans increasing the elastic modulus in compression and xylans significantly elevating the elongation at break under tension [49].

### Lignin

Lignin is a hydrophobic aromatic polymer containing three main types of phenylpropane units (coniferyl alcohol [G], sinapyl alcohol [S], and paracoumaryl alcohol [H]) linked by ether, ester, and carbon-carbon bonds [50]. Lignin serves as a binder within the middle lamellae and between microfibrils. It also augments the mechanical strength and hydrophobicity of herbaceous biomass [51]. Moreover, lignin is the primary source of color in herbaceous biomass. Therefore, it is usually partially or completely removed to defibrillate cellulose fibers and nanofibers from the complex matrix or reduce the color degree of cellulose-based materials. Most herbaceous biomass species have a lower lignin content (normally 20 wt% or less, Table 1) and DP when compared to wood, making the extraction and purification of cellulose easier. Furthermore, in addition to the G units and S units (mainly G units in softwood and G and S units in hardwood), the lignin in herbaceous biomass also contains H units. A large number of H units are linked to other lignin structures through ester bonds, which are easily cleaved by alkaline-based delignification processes [50].

### Extractives and Ashes

In addition to the aforementioned main components, lignocellulosic biomass typically contains a small number of extractives and ashes. The extractives include resin acids, fatty acids, fats, fatty alcohols, steroids, waxes, terpenes, phenols, rosin, and many other organic compounds [52]. Aside from carbon, hydrogen, and oxygen, numerous other elements exist as ions (the main components of ashes) in the plant, including silicon, calcium, magnesium, iron, potassium, sodium, copper, zinc, and manganese [53]. These materials could adversely affect the cellulose extraction process. For example, the resin acids and fatty acids in the extractives can be softened to an oily substance by heating or saponified when delignified with alkaline treatment, which hinders chemicals penetration, resulting in reduced degree of delignification and increased chemical consumption [54]. The ashes present during delignification decompose oxidative delignification agents (e.g., hydrogen peroxide, oxygen, ozone) into less selective hydroxyl radicals, thereby reducing the DP of cellulose. Herbaceous biomass usually contains a lower content of resin acids and fatty acids than wood but a higher content of ashes [50]. Moreover, the ash content in wheat (~4–9 wt%) and rice (~15–20 wt%) is significantly higher than that of other herbaceous biomasses, and more than 60 wt% of it is in the form of $SiO_2$, which is markedly different from wood [33]. Therefore, suitable extraction methods and conditions must be designed for raw materials from various sources to obtain high-quality extracted cellulose materials.

## Structure of Herbaceous Biomass

Herbaceous biomass, analogous to wood, has a hierarchical porous structure from the nanoscale to the macroscale, but morphology varies greatly across these length scales. At the macroscale, herbaceous biomass exhibits some unique structures and properties. Contrary to the trunk or branches of a tree with a solid center, the herbs' stem (cane and straw) has a hollow and segmented structure with thin walls, making it light-weight, flexible, and a good insulator [70]. Furthermore, the periphery of a herbaceous stem is the strongest part contrary to the trees where it is the centeral part. There is no radial (lateral) conduction cell in the stem except nodes that gives the herbaceous stem a more linear structure than wood [70]. Finally, mechanical stress is distributed more effectively in herbaceous biomass-derived structural materials than in wood-derived materials since knots within trees act as stress concentrators [70].

Other differences in various species of trees and herbaceous biomasses include density, strength, and thermal conductivity, all of which are primarily determined by their microstructure. As illustrated in Fig. 2a1–c5, the microstructures of different herbaceous biomasses are complex and diverse. The morphology of herbaceous biomass cells from different sources can vary in length, diameter, cell wall thickness, and cell wall-lumen ratio. Moreover, these cells are mostly oriented in the direction of growth and are held together by amorphous lignin to form 3D porous aligned microstructures.

Cell morphology is primarily determined by the type of cells and herbaceous biomass. The main cell types found in herbaceous biomass are fiber cells, parenchyma cells, vessels, sieve tubes, companion cells, epidermal cells, and stone cells [71]. Fiber cells (also called *fibers*) are long and thin, and provide structure and support, whereas parenchyma cells have a large cavity and thin walls for nutrition storage. Vessels have large diameters for transporting water and nutrients from roots, whilst sieve tubes and companion cells have numerous pits for transporting photosynthate from leaves to other parts of plant. Epidermal cells contain high ash contents for the protection of internal tissues. Finally, stone cells found in bamboo have thick cell walls, small cell cavities, and a high degree of lignification, all of which contribute to the superior strength [71]. Among these cell types, fiber cells are the predominant in stem, normally accounting for ~ 40–70% of total cells by area. In addition, fiber cells possess high tensile strength, elasticity, flexibility, and cellulose content, making them useful in products such as papers and textiles.

Furthermore, air-filled pores exist in the lumen of mature fiber cells, which provide herbaceous biomass with some desirable characteristics such as lightweight, high thermal resistivity, and insulation. The same type of cells from different sources can also have significantly different morphologies. For example, as summarized in Table 1, the average length of extracted fiber cells from cotton (18 mm) and flax (33 mm) is much higher than that from rice straw (1.4 mm), hardwood (0.4–1.6 mm), and softwood (1.4–6.0 mm). These differences

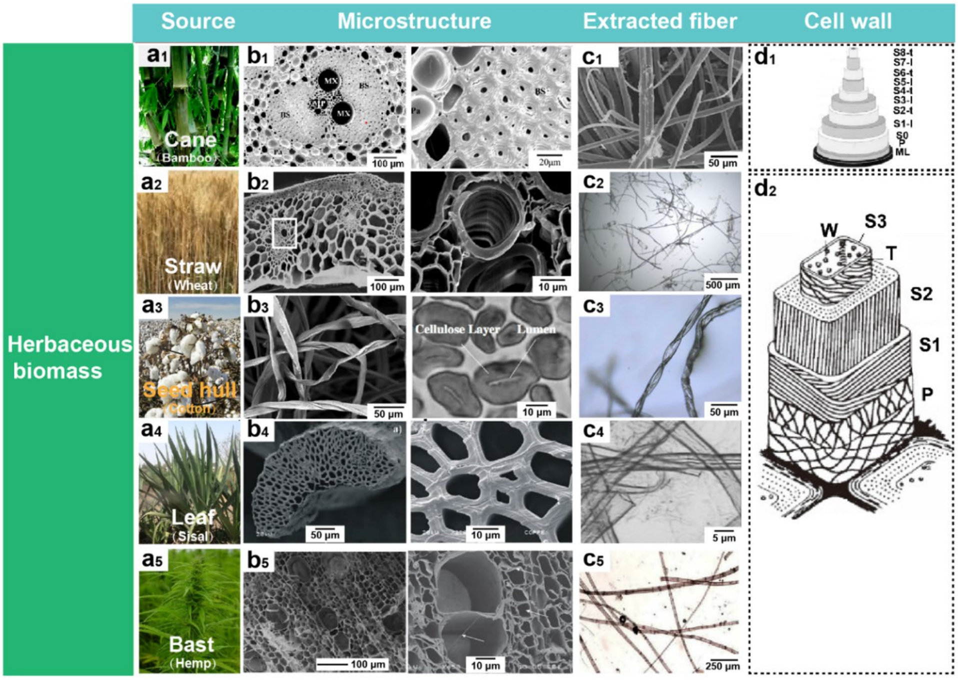

**Fig. 2** Structure of different sources of herbaceous biomass. ($\mathbf{a}_1$–$\mathbf{a}_5$) Images of different sources of herbaceous biomass (cane, straw, seed hull, leaf, and bast) [28, 29, 77–79] and ($\mathbf{b}_1$–$\mathbf{b}_5$) their microstructures [31, 80–84], ($\mathbf{c}_1$–$\mathbf{c}_5$) extracted fibers [32, 85–87], and ($\mathbf{d}_1$, $\mathbf{d}_2$) cell wall structure [88, 89]

in cell morphology can determine the performance and potential applications of a given herbaceous biomass or herbaceous biomass-derived material. For instance, the bamboo cane contains more fiber cells that are longer and have thicker cell walls than the cane or straw of other herbaceous biomass, making bamboo a better choice for structural materials used in high-strength applications.

The cell wall structure influences the mechanical performance and anisotropy of herbaceous biomass. Unlike numerous microstructural differences between herbaceous biomass species, the cell wall structures of most of them are quite similar with a primary wall and a two- or three-layered secondary wall [72]. In the secondary layers, nanoscale cellulose microfibrils are embedded in a matrix of hemicellulose and lignin, oriented at different angles (Fig. 2d2) [73]. The fiber cells and parenchyma cells in bamboo, as an exception, contain more secondary wall layers than those of other herbaceous biomass. For a 3-year-old bamboo plant, ~ 23% of the fiber cells in the phloem have 7–8 cell wall layers (Fig. 2d1) [72]. Its multilayer structure composed of microfibrils arranged in the longitudinal and transverse directions is conducive to the remarkable mechanical performance of bamboo (tensile strength of ~ 100–300 MPa, compression strength of ~ 40–80 MPa, and flexural strength of ~ 50–150 MPa) [74–76].

## Extraction of Cellulose Materials From Herbaceous Biomass

### Extraction of Cellulose Fibers From Herbaceous Biomass

Cellulose fibers with lengths ranging from a few to tens of millimeters and diameters ranging from a few to tens of microns can be isolated from the complex matrix of cellulose, hemicellulose, and lignin within herbaceous biomass. Commercial cellulose fibers are of three types: lignin- and hemicellulose-retaining pulp (including high-yield pulp and unbleached pulp), hemicellulose-retaining pulp (bleached pulp), and high-cellulose content pulp (dissolving pulp) [90]. Extraction processes typically involve multiple steps and complex chemical and mechanical treatments to separate fibers from one another. Numerous classes of chemicals, such as alkalis, acids, oxidants, organosolvs, ionic liquids, and enzymes, have been used in the extraction processes to increase the dissolution of the binder (lignin) [91]. An ideal fiber extraction process should avoid cellulose degradation and effectively remove lignin while remaining cost-effective, energy-efficient,

**Table 1** Summary of different sources of herbaceous biomass and trees (for comparison) with different compositions, extracted fiber sizes, and crystallinities

| Source | | | Compositions | | | | Fiber size (extracted) | | Crystallinity | | Reference |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Cellulose (%)[a] | Hemicellulose (%)[a] | Lignin (%)[a] | Ashes (%)[a] | Length (mm) | Diameter (μm) | Original (%) | Extracted (%) | |
| Herbaceous biomass | Cane | Bamboo | 26–43 | 15–26 | 21–31 | 1.5–3 | 2.7 | 14 | 53.7 | 64.5 | [33, 34, 55–57] |
| | | Sugarcane bagasse | 32–44 | 27–32 | 19–24 | 1.5–5 | 1.7 | 20 | 42.6 | 76.0 | [33, 34, 58, 59] |
| | Straw | Wheat straw | 29–35 | 26–32 | 16–21 | 4–9 (3–7 $SiO_2$) | 1.4 | 15 | 57.5 | 77.8 | [33, 34, 60] |
| | | Rice straw | 28–36 | 23–28 | 12–16 | 15–20 (9–14 $SiO_2$) | 1.4 | 8 | 51.9 | 63.8 | [33, 34, 57, 61] |
| | Seed hull | Cotton | 90 | 6 | 1 | 0.1–0.2 | 18 | 20 | 77.0 | – | [33, 62] |
| | Leaf | Pineapple leaf | 36 | 23 | 28 | 2.9 | 2.9 | 12 | 49.0 | 64.0 | [63, 64] |
| | | Sisal | 73 | 1 | 13 | 0.6–1.0 | 3.0 | 20 | 71.7 | 72.0 | [33, 34, 65] |
| | Bast | Kenaf | 55 | 13 | 12 | 2–5 | 5 | 21 | 48.2 | 77.3 | [33, 34, 66] |
| | | Hemp | 70 | 15 | 3 | 2.9 | 25 | 25 | 57.4 | 77.1 | [33, 34, 67] |
| | | Flax | 72 | 18 | < 1 | 2–5 | 33 | 19 | 70.0 | – | [33, 34, 68] |
| Tree (for comparison) | Wood | Hardwood | 38–49 | 19–26 | 23–30 | 1 | 0.4–1.6 | 10–40 | 52.7[b] | 69.3[b] | [33, 50, 69] |
| | | Softwood | 40–45 | 7–14 | 26–34 | 1 | 1.4–6.0 | 20–50 | 56.0[b] | 73.2[b] | [33, 50, 57] |

[a] Content of cellulose, hemicellulose, lignin, and ashes expressed in weight percentage of dry matter

[b] The crystallinity of hardwood and softwood is from poplar and fir, respectively

and environmentally friendly. Currently, alkaline pulping (NaOH, $Na_2S$) and sulfite pulping ($Na_2SO_3$, $NaHSO_3$) are the most popular methods for removing lignin in the middle lamella, allowing fiber cells to be easily separated [90]. Although the chemical pulping process can remove most of the lignin in biomass under certain conditions of temperature and pressure, complete removal of lignin through pulping is economically unfeasible because over-pulping can cause severe carbohydrates degradation. Therefore, oxidants ($O_2$, $O_3$, $H_2O_2$) combined with alkali (NaOH) are often used after pulping to remove the residual lignin and cleave the chromophore groups in lignin while maintaining the high yield and low degradation rate of cellulose fibers [90]. Besides that, cold alkaline extraction or prehydrolysis using acid, steam, or hot water is frequently used to remove hemicellulose [90].

The raw material used to extract cellulose fibers is also an important determinant of cost, fiber quality, and pollution load. The low lignin content in herbaceous biomass is beneficial because it requires less energy and chemical treatments during pulping (most of the lignin removal and fiber separation) and bleaching (removal of remaining lignin residues) compared with other feedstocks. For instance, the average production cost for wood pulp is \$441/MT, whereas that for wheat straw and rice straw pulp is \$282/MT (36% cost savings) and \$279/MT (37% cost savings), respectively [92]. As shown in Table 1, extracted fibers from different herbaceous sources also provide a wide range of DP, crystallinity, and size distributions, which are all compatible with the mechanical strength and thermal stability of cellulose fiber-based materials.

## Extraction of Nanocellulose From Herbaceous Biomass

Cellulose fibers are classified as nanocellulose if at least one of their dimensions is less than 100 nm. Nanocellulose can be extracted from lignocellulosic feedstock through chemical, mechanical, or enzymatic methods, or a combination of these. Recently, nanocellulose and its derived materials (filaments, membranes, aerogels, and hydrogels) have gained popularity in many emerging fields, including biomedicine, energy storage and conversion, smart materials, and environmental pollution control [4]. Nanocellulose materials are desirable in a wide range of applications due to their high aspect ratio (10 to 100 for cellulose nanocrystals [CNCs] and tens to hundreds for cellulose nanofibrils [CNFs]), lightweight, high elastic modulus, transparency, low cost, biocompatibility, and low thermal expansion rates [93]. Additionally, the market for nanocellulose is expected to reach 35 MMT/year by 2050 [94]. Many companies around the world have been involved in commercial nanocellulose production, including Innventia (100 kg CNF/day), the US Forest Service (500 kg CNF/day), CelluForce (1 MT CNC/year), and BioVision Technologies (4 MT CNC/year) [95]. However, because of the strong inter-fibrillar hydrogen bonding, the production of nanocellulose typically requires high energy or/and the use of harsh chemicals. Numerous studies have focused on finding inexpensive raw materials, reducing the energy and chemical consumption for extraction, and increasing the chemical recycling rate to make nanocellulose commercially more competitive [96]. Among these methods, finding suitable low-cost raw materials is the simplest way to reduce the production cost of nanocellulose. Different feedstocks can result in various properties of nanocellulose such as morphology, crystallinity, and thermal behavior. Annual plants, crop residues, and industrial bioresidues from herbaceous biomass, such as sugarcane bagasse and wheat straw, are promising non-woody feedstocks for nanocellulose preparation because of their low cost, diversity, and widespread availability. Different feedstocks may be well suited for specific applications depending on the properties of the nanomaterials extracted from them.

## Extraction of CNFs From Herbaceous Biomass

CNFs typically exhibit an entangled web-like morphology, with fibril diameters ranging from 10 to100 nm and lengths of several micrometers (Fig. 3b). CNFs were first prepared by homogenizing wood fiber in 1983 [97]. Since the 1990s, mechanical treatments such as homogenization, microfluidization, grinding, disk refining, and sonication have been developed for CNF preparation. Mechanical methods mainly use shear forces to break the hydrogen bonds between the microfibrils and separate them from the cell wall. Mechanical processes for CNF production typically give high yields (90–95 wt%) of CNFs with high aspect ratios, greater degrees of fibrillation, and good thermal stability (degradation temperature ~ 250–350 °C) [98].

A single homogenization process can consume 20,000–30,000 kWh/MT of energy [99]. Several pretreatment strategies have been developed to reduce the energy consumption of these fibrillation processes, such as adding charged species to the cellulose surface, lowering the DP of the cellulose, and swelling the cellulose with a suitable solvent. Figure 3a depicts typical pretreatment methods used in CNFs production. Each of the pretreatment methods in Fig. 3a, such as TEMPO oxidation for imparting surface charges, the use of organic solvents for cellulose swelling, and enzymatic and mechanical pretreatments (e.g., refining, cryo-crushing), can significantly decrease the energy consumption to less than 1,000 kWh/MT [100].

Cellulose fibers in some herbaceous biomasses naturally have a low DP, eliminating the need for mechanical, chemical, or enzymatic treatments to lower the DP. For example, wheat straw has a DP of 1025, and wheat straw pulp

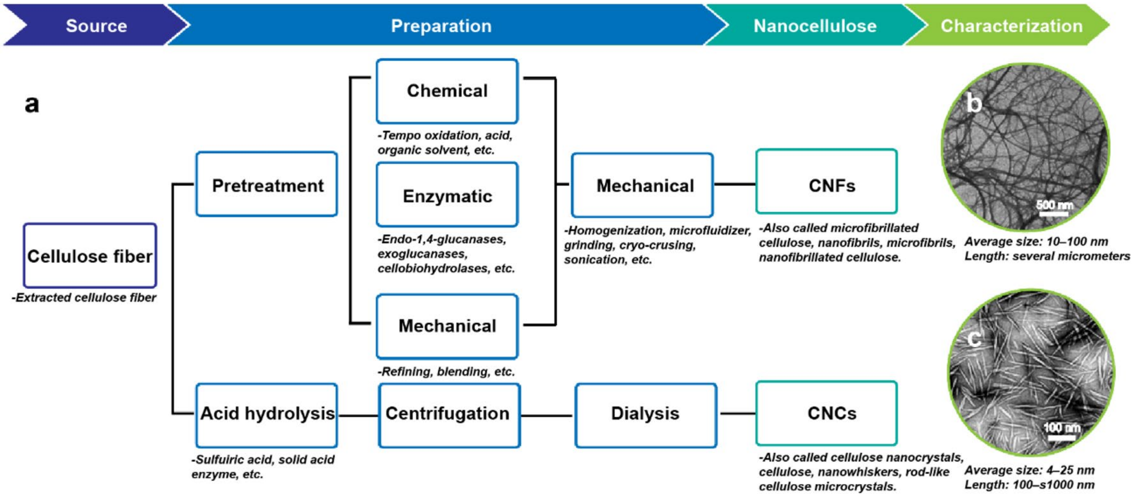

**Fig. 3** Typical nanocellulose preparation methods. **a** Schematic illustration of typical nanocellulose preparation methods. TEM images of **b** entangled web-like CNFs [27] and **c** rod-like CNCs [116]

requires fewer treatment cycles and can be processed at a higher range of concentrations to obtain CNFs than softwood sulfite pulp, which has a DP of 2249 [101]. The use of herbaceous biomass with a low DP as the raw material for nanocellulose preparation is a promising way to save energy and reduce chemical consumption. As previously stated, the high hemicellulose content in some species of herbaceous biomass (e.g., wheat straw, rice straw, sugarcane bagasse) helps to reduce the energy requirement for CNF production. Herbaceous biomass such as wheat straw, rice straw, sugarcane bagasse, bamboo, kenaf, pineapple leaves, and cotton have been reported for preparation of nanocellulose [96, 102, 103]. However, to the best of the authors' knowledge, no systematic research has been conducted on the relationship between energy consumption and raw materials from different sources [45].

The feedstock used for CNF extraction also has a significant impact on the resulting crystallinity and aspect ratios, which affect the mechanical performance and thermal stability of CNFs. CNFs with a higher crystallinity typically exhibit higher strength and better thermal stability but less elasticity and water retention. The crystallinity of CNFs extracted from various feedstocks using the same fibrillation process can differ significantly. Chen et al. compared the crystallinity of cellulose fibers and CNFs after fibrillation from bamboo, rice straw, and flax [104]. The crystallinity of fibers from each feedstock before and after fibrillation changed by less than 1% and maintained the high crystallinity of CNFs (64% for bamboo and rice straw and 82% for flax).

CNFs with high aspect ratios are preferable for high-strength applications, particularly reinforcing composites. CNFs with higher aspect ratios tend to form stiffer interconnected networks or web-like structures through nanofiber entanglement, which is one reason for the higher tensile strength for CNF membranes than for CNC membranes with relatively low aspect ratios [105]. However, because of the entangled web-like structural morphology of CNFs, determining average length of CNFs from microscopy (transmission electron, scanning electron, or atomic force) results alone is tricky. Few studies have reported average lengths of prepared nanocellulose, and no standard measurement technique exists for calculating average dimensions of CNFs accurately. Several indirect methods have been developed to determine the lengths of CNFs. Tanaka et al. evaluated the average lengths of CNF by a shear viscosity method [106]. Shinoda et al. reported a linear relationship between the DP and average length of TEMPO-oxidized CNFs, indicating that the DP could be used as an estimate of CNF length [107]. Kleinebudde et al. studied the effects of homogenization on cellulose fibers with different DPs [108]; they found that cellulose with a higher DP resulted in CNFs with greater particle size (higher lengths and widths). In general, cellulose fibers from different herbaceous biomasses can form CNFs of varying dimensions because of the wide range of DPs. Among them, some herbaceous biomasses with very high cellulose DP (e.g., cotton, flax, hemp [109, 110]) are promising raw materials for the preparation of high–aspect ratio CNFs.

## Extraction of CNCs From Herbaceous Biomass

Unlike CNFs, CNCs are highly crystalline rod-like particles typically 4–25 nm in diameter and 100–1000 nm in length (Fig. 3c). CNCs were first prepared by Randy et al. using sulfuric acid hydrolysis of the amorphous region of cellulose in 1949 [111]. High concentrations of sulfuric acid can lead to equipment corrosion, large amounts of chemical waste, and excessive water consumption. Various hydrolysis strategies have been developed to address these problems, including the use of solid acids and enzymes (Fig. 3a) [112]. Currently, sulfuric acid is still the most commonly used acid in CNCs preparation because of the good dispersibility of the resulting CNCs. Sulfuric acid can convert the hydrogen bonds on the cellulose surface into sulfate groups by esterification, which increases its electrostatic repulsion and inhibits agglomeration and flocculation.

The morphology, crystallinity, and yield of CNCs are influenced by cellulose sources, chemical compositions, and hydrolysis conditions (temperature, time, acid/fiber weight ratio, and acid concentration). CNCs with high crystallinity can be obtained by increasing the degree of acid hydrolysis, but they often have reduced dimensions and yield. CNCs with a high yield, crystallinity, and aspect ratio can be produced without using strong hydrolysis conditions if the optimal feedstock is employed. Hsieh compared the morphology of CNCs from cotton and rice straw and found that the prepared cotton CNCs (200–400 nm length, < 10 nm width, 20–40 aspect ratio) exhibited much higher lengths and aspect ratios than those of rice straw (117 nm length, 11.2 nm width, 10.5 aspect ratio), even though the hydrolysis time of cotton was longer (60 min for cotton and 45 min for rice straw) [113]. Additionally, the chemical composition of the raw materials also affects the hydrolysis process and performance of the resulting CNCs. Feng et al. compared rice straw-derived CNCs and holoCNCs (CNCs containing hemicellulose, silica, and residual sulfonates) under the same hydrolysis conditions (64 wt% $H_2SO_4$, 45 °C, 45 min) [114, 115]. The width of holoCNCs (4.1 ± 1.6 nm thick, 6.4 ± 1.8 nm wide, and 113 ± 70 nm long) obtained was similar to but shorter than that of CNCs (4.7 ± 1.3 nm thick, 6.4 ± 1.2 nm wide, and 143 ± 31 nm long). Moreover, the holoCNCs had a lower charge (0.128 mmol/g) and crystallinity (84.4%) than CNCs (0.237 mmol/g, 90.7%), but they were more thermally stable (maximum mass loss temperature of 284 °C) than CNCs (235 °C).

## Application of Biomaterials From Herbaceous Biomass

As discussed, herbaceous biomasses comprising carbon-based lignocellulosic fibers have various macro-, micro-, and nano-morphologies, as well as different chemical compositions, endowing them with unique mechanical, thermal, and transport properties. These unique structures and properties could make them useful in a variety of applications such as ultra-strong structural materials, high-performance carbonized conductive materials, ion-selective transportation and adsorption, and highly flexible devices.

## Application of Herbaceous Biomass at the Macroscale

### High-Performance Structural Materials

The cane in herbaceous biomasses is the strongest component and their primary support structure. The cane of bamboo has a tensile strength (~ 100–300 MPa) comparable to structural steel (~ 360–550 MPa) and compressive strength (~ 40–80 MPa) superior to concrete (~ 8–20 MPa), despite being light-weight [117–120]. Therefore, bamboo has become a popular structural material in furniture, houses, and bridges (Fig. 4a) [70]. The high mechanical strength and toughness of bamboo canes come from their unique rod-like structure and high aspect ratio. A mature bamboo plant can grow to be 5–12 m tall. The bottom of these tall plants withstands large bending forces caused by the weight of the plant and external factors such as wind and climbing animals. Trees resist these bending forces by thickening their trunks, whereas bamboo possesses a denser macrostructure and ultrastructure than most trees, allowing it to provide enough bending resistance [70]. In addition to its mechanical strength, bamboo has unique properties that make it amenable for use as a structural material. The nodes of a bamboo cane stiffen it and enable it to avoid buckling. Bamboo's unique root system (50% of its mass) makes it one of the fastest-growing plants in the world, and some types of bamboo can grow more than 0.9 m a day. Furthermore, the average harvest time for bamboo is 3–5 years, whereas the harvest time of most trees used for construction ranges from 20 to 60 years [121]. Besides, bamboo is widely distributed in tropical and subtropical regions and is commonly found in Africa, Asia, and America [121]; more than 490 species of bamboo are available in the United States and Canada [122]. Finally, bamboo is more carbon-neutral than commercial steel, and the carbon dioxide emitted during processing is about one-fifteenth of commercial steel. Therefore, bamboo is a promising, sustainable, light-weight, and robust alternative to more traditional structural materials.

Further enhancing the strength and toughness of bamboo could expand its applications. Currently, the tensile strength of engineered bamboo is one- or two-tenths of the theoretical strength of elementary fibril (a universal structural unit of cellulose microfibril, ~ 7.5 GPa) [39]. Several factors in natural bamboo restrict the increase in its mechanical strength, such as the brittle lignin networks, weak interactions between parenchyma cells and sclerenchyma cells (fiber cells and sclereids), and defects. Several methods have been developed to overcome these issues by softening the fiber cell wall and increasing the density of bamboo. Recently, Li et al. densified bamboo via flattening

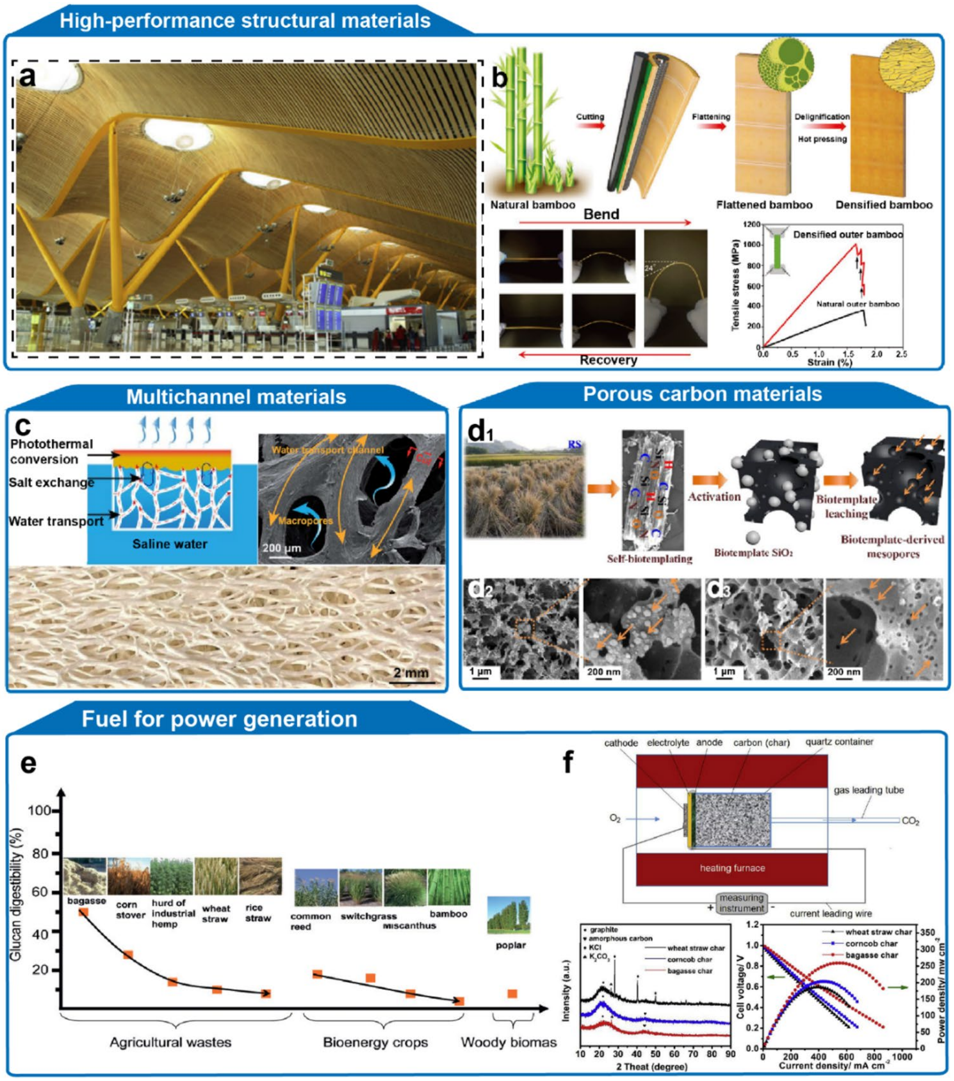

**Fig. 4** Macroscale application of using herbaceous biomass for high-performance structural materials, fuel, porous carbon materials, and multichannel materials. **a** Photograph of bamboo ceiling [131]. **b** Schematic of preparation of bamboo-based structural material with high tensile strength and flexural strength [121]. **c** Schematic of loofah-based multichannel materials for solar desalination [124]. **$d_1$** Schematic of preparation of biotemplate-derived porous carbon materials from rice straw, and scanning electron microscope images of **$d_2$** silicon-containing and **$d_3$** silicon-removed porous carbon [13]. **e** The glucan digestibility (important parameter for bioethanol production) of various non-pretreated herbaceous biomasses and wood [129]. **f** Schematic of sugarcane bagasse-, corn stalk-, and biochar-fueled direct carbon solid oxide fuel cells with high power output [130]

(70% reduction in thickness), delignification, and hot pressing, which reduced defects, increased hydrogen bonding, and significantly augmented mechanical properties [121]. The densified bamboo exhibited a very high tensile strength (~ 1 GPa), toughness (9.74 MJ $m^{-3}$), and specific strength (777 MPa $cm^3$ $g^{-1}$), each of which was significantly higher than most structural materials (Fig. 4b). In another study, Chen et al. used partial delignification and microwave heating to achieve fast and simple densification of bamboo, resulting in high tensile strength (560 MPa), toughness (6.8 MJ $m^{-3}$), and specific strength (560 MPa $cm^3$ $g^{-1}$) [123].

## Multichannel Materials

Multichannel structures are advantageous in many applications, including solar desalination, ion-selective transportation, and salinity gradient power generation. Because of biodiversity, herbaceous biomass provides more diverse multichannel structures on macro-, micro-, and nanoscales than wood, as well as different chemical compositions. Several multichannel structures aligned together provide continuous transport paths for ions and water, and high cellulose contents can increase the hydrophilic properties of herbaceous biomass, increasing water and ion transport efficiency.

Lignocellulosic biomass-based solar desalination can efficiently convert solar energy to thermal energy for pure water generation. A bilayer structure design containing a top layer as the light absorber and a bottom layer as the water pumper (Fig. 4c) has been widely adopted by researchers. Multiple channels play an important role in continuously pumping water from a bulk liquid to the heating surface. An ideal desalination system should meet the following standards: (1) efficient solar energy conversion enabled by broad-spectrum light adsorption, (2) low heat dissipation to the bulk liquid, (3) little to no salt accumulation, (4) efficient pumping of water from the bulk liquid, and (5) low cost [124]. For lignocellulosic biomass-based evaporators, salt accumulation and pumping efficiency are key factors that impede the evaporation rate and efficiency. Wood has densely packed channels and a relatively low hydrophilic surface because of the high content of hydrophobic lignin. Therefore, wood-based bilayer evaporators exhibit a relatively low evaporation rate (~ 1 kg $m^{-2}$ $h^{-1}$) and evaporation efficiency (~ 70–80%) and can significantly accumulate salt [125]. Recently, Liu et al. reported an efficient loofah-based evaporator with multichannel structures and highly hydrophilic surfaces [124]. Loofah, a natural sponge, has a loose structure comprising macropores and interconnected fibers, as well as numerous aligned microchannels (Fig. 4c). Loofah has a high cellulose content (60–65 wt%), making it highly hydrophilic. Combined with its multichannel structure, loofah can efficiently transport seawater from the bottom layer to the top layer during desalination. In addition, the fluffy structure of loofah (containing numerous macropores) and multiscale channels can effectively avoid salt accumulation. The evaporation rate and evaporation efficiency were as high as 1.42 kg $m^{-2}$ $h^{-1}$ and 89.9%, respectively, under 1 Sun of illumination [124].

## Porous Carbon Materials

The performance of electrical energy storage (EES) devices (e.g., capacitors, fuel cells, and batteries) strongly depends on the physicochemical properties of the electrode materials. Porous carbon electrodes are currently one of the most widely used electrode materials. Ideal porous carbon electrodes have a high specific surface area (SSA), well-controlled pore systems, a large active surface, good electrical conductivity, and stability. Among these qualities, the well-controlled pore systems can effectively increase the SSA, improve the specific capacitance, and maintain a good rate capability. Currently, the preparation of most porous carbon electrodes relies on fossil fuel-derived precursors and complex synthesis systems, which consume large amounts of energy and are environmentally harmful. Therefore, inexpensive and environmentally friendly carbon sources and preparation methods for porous carbon electrodes are highly desirable.

Herbaceous biomass has naturally well-aligned hierarchical pore structures that are used for the transmission and transportation of water, ions, and nutrients. Different herbaceous biomass types and plant parts possess distinct pore structures, demonstrating a wide variety of pore hierarchies. These unique pore structures could be inherited after a simple carbonization process, endowing their derived carbon materials with a high SSA and well-controlled pore systems. Carbonized herbaceous biomass also maintains good mechanical properties, ensuring its mechanical stability in energy storage and conversion devices. Furthermore, for the derived carbon materials, rich hydroxyl groups on the surfaces of herbaceous biomass can improve its wettability and be functionalized to regulate its active sites. Herbaceous biomass-derived porous carbon is therefore a promising alternative to fossil fuel-derived materials for EES devices.

To prepare herbaceous biomass-derived porous carbon materials with ideal physicochemical properties, various methods, including heteroatoms doping, hydrothermal treatment, and carbonization and activation, have been developed [126]. Among them, carbonization combined with activation is the most adopted method because of its simplicity and the high SSA of prepared carbon materials. The carbonization and activation processes can retain the original interconnected pore structures in herbaceous biomass, thus providing a continuous and short electronic transmission channel. Several parameters significantly influence the properties of the derived carbon, including carbonization temperature (normally between ~ 400-1000 °C), activation methods, and chemical compositions of herbaceous biomass. A suitable carbonization process can endow the derived carbon materials with a high SSA, guaranteed high yield, abundant active sites, and high degree of graphitization, resulting in high conductivity. The activation process can further promote the pores and channels formation via chemical etching, which is beneficial to the transportation and storage of electrolyte ions.

The composition of biomass plays a crucial role in the polymorphism of derived carbon. Feng et al. examined the potential correlation between compositions (cellulose, hemicellulose, and lignin) and carbonization temperature in the final carbon product [127]. They concluded that the highly ordered crystalline cellulose structure leads to an increased tendency to form ordered and semi-graphitized carbons compared with lignin and hemicellulose. In addition, the metal salts existing in herbaceous biomass could promote the formation of graphitic carbon because of the catalytic effects [128]. Other inorganic compounds, such as $SiO_2$ existing in herbaceous biomass, can also serve as the biotemplates for increasing the SSA. Recently, Chen et al. reported high-performance porous carbon from rice straw by chemical activation and a self-biotemplating strategy (Fig. 4d1) [13]. Figures 4d2 and d3 show the hierarchical porous carbon after activation and the removal of biotemplated $SiO_2$. The mesopore ratio increased from 11.5% to 23.7% after $SiO_2$ removal and further increased to 28.0% after chemical activation. The resulting porous carbon exhibited good specific capacitance (357 F $g^{-1}$ at 0.5A $g^{-1}$, 1 M $H_2SO_4$ for a three-electrode system) and high energy density (29.3 W h $kg^{-1}$ at 900 W $kg^{-1}$).

## Fuel

Herbaceous biomass has long been used as a source of energy. Household herbaceous biomass fuels have been gradually replaced by fossil fuels since the 1910s, when coal became affordable and available [126]. Because of its abundance, renewability, low cost, and carbon neutrality, herbaceous biomass has recently attracted significant attention for energy production as concerns about climate change and the depletion of fossil fuels have grown. With rapid developments in the field of energy production, herbaceous biomass-derived fuels, including biogas, bioethanol, bio-oil, synthesis gas, and biochar, could be used for furnaces, turbines, engines, and fuel cells.

Biofuel (e.g., bioethanol) derived from herbaceous biomass and agricultural waste has been produced commercially, but its cost needs to be reduced further before it can be widely adopted. As a raw material, herbaceous biomass is a potential candidate in biofuel and energy production. Different compositions of herbaceous biomass, which include cellulose, hemicellulose, lignin, ashes, moisture, and extractives, determine its fixed carbon, calorific value, volatile component, inorganic element composition, and content of enzyme inhibitors generated [126]. Therefore, different sources will significantly affect the microbial fermentation and thermal transformation processes. Sathitsuksanoh et al. compared the glucan digestibility (an important parameter for determining the potential of raw materials for bioethanol production) of various non-pretreated herbaceous biomasses and wood (Fig. 4e) [129]. The digestibility of glucan was reported in the order of sugarcane bagasse (47%), corn stover (23%), common reed (19%), switch grass (17%), industrial hemp (14%), wheat straw (11%), rice straw (10%), poplar (9%), miscanthus (8%), and bamboo (3%). Qiu et al. compared the biochar derived from wheat straw, corncob, and sugarcane bagasse and used it as fuel for direct carbon solid oxide fuel cells (Fig. 4f) [130]. Under the same pyrolysis conditions, biochar derived from sugarcane bagasse and corn stalk exhibited lower crystallinity and a larger SSA than wheat straw biochar because of its higher cellulose content and lignin and lower content of ashes. Therefore, the obtained power output of direct carbon solid oxide fuel cells powered by sugarcane bagasse biochar (240 mW cm$^{-2}$) and corn stalk biochar (204 mW cm$^{-2}$) was higher than that of wheat straw biochar (187 mW cm$^{-2}$). Overall, herbaceous biomass provides a variety of raw materials for bioconversion to biofuels, some of which have great potential to become future energy suppliers.

## Application of Herbaceous Biomass at the Microscale

Flexible electronics have significantly developed in recent years because of their flexibility, portability, and high applicability to dynamic applications. Unlike commonly used non-biodegradable polymers, cellulose is inherently sustainable, biodegradable, biocompatible, and has rich surface chemistry. These properties enable it to be used in making paper and textile-based electronics that are environmentally and biologically friendly, as well as in flexible electronics [132]. In addition, the hydrogen bonding and van der Waals forces in and between the cellulose molecules combined with the physical entanglement of fibers offer excellent mechanical properties for paper and textiles [4]. Moreover, the randomly distributed or aligned fibrous network formed during paper forming or weaving makes the resultant paper and textiles porous, light-weight, and flexible [133]. Paper and textiles made from herbaceous biomass are inexpensive and have been industrialized for centuries. The output of herbaceous biomass-derived textiles in 2019 is estimated to be 26 million tons, accounting for 40% of the total output [134, 135]. The output of herbaceous biomass-derived paper in 2020 is estimated to be 40 million tons, accounting for 10% of the total [136]. Paper and textiles made from herbaceous biomass are considered as eco-friendly, energy-efficient, cost-effective, and scalable alternatives for building flexible electronics like circuits, biosensing devices, EES devices, and generators.

### Electronic Circuits

Paper made from herbaceous biomass is a promising substrate for flexible electronic circuits due to its low cost, eco-friendliness, lightweight, excellent flexibility, and mechanical robustness. In addition, the low stretchability of paper can be improved by kirigami and origami designs, making it suitable for wearable electronics or building 3D electronics. However, the inherent hygroscopicity of cellulose paper inevitably affects its stability and robustness in wet environments, causing unfavorable mechanical degradation and irreversible deformation. This problem can be solved by surface chemical modification (e.g., silanization). Moreover, copy paper is usually hydrophobically treated and filled with mineral fillers to reduce ink diffusion, penetration and surface roughness, as well as enhance printability and writability [132].

Scalable screen printing, inkjet printing, printing of liquid metal, and direct writing have been developed for building electronic circuits with paper [132]. Siegel et al. fabricated flexible paper-based circuits by depositing metallic wires via evaporation, sputtering, or airbrushing on paper substrates [137]. Unlike circuits using glass fiber, ceramic, or polyimide as substrates, paper-based circuits can be folded repeatedly, built into 3D structures, trimmed with scissors, and disposed of by incineration (Fig. 5a). Li et al. prepared paper-based circuits via mechanical sintering and recycled the obtained circuits (Fig. 5b) [138]. The resulting circuits exhibited high electrical stability after 10,000 folding cycles, with only ≤ 4% changes in resistance. Upon recycling they exhibited almost the same electrical stability during the

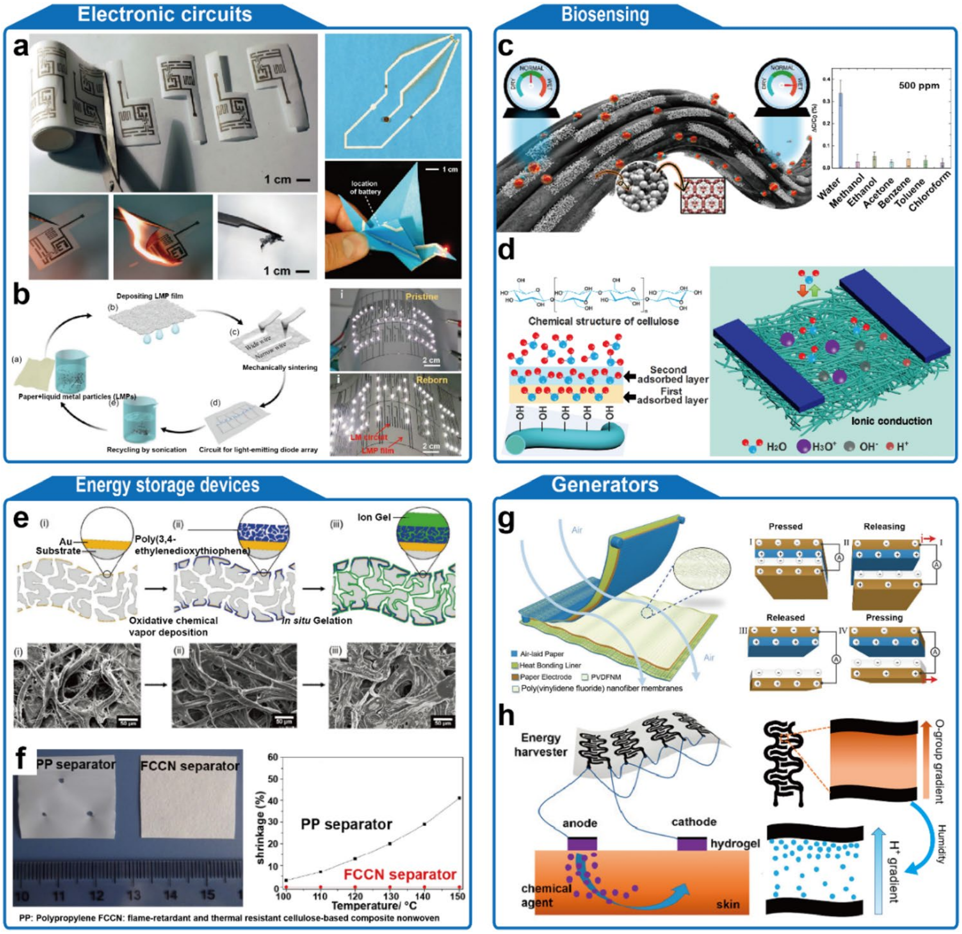

**Fig. 5** Schematic illustration of cellulose fiber-based paper and textiles as a promising flexible material for (**a**, **b**) electronic circuits [137, 138], (**c**, **d**) biosensing [140, 141], (**e**, **f**) EES devices [142, 143], and (**g**, **h**) generators [139, 144]

folding process, as well as almost the same thermal characteristics as the pristine circuits (Fig. 5b). Although paper presents many merits in flexible circuits, its inherent porous structure causes discontinuity, which limits its performance and uses in complicated applications [132]. In general, low-cost paper-based flexible electronic circuits have shown great potential in applications such as smart packaging, medical diagnosis, and disposable electronic devices, but their development is still at its infancy, and more efforts are needed to improve the performance and broaden the application scope.

## Biosensors

Recently, paper- and textile-based flexible sensors have been continuously used to detect various signals of living organisms, including temperature, humidity, pH, biopotential, respiration, pressure, strain, light, and biochemical compositions [132]. In these sensors, paper and textiles take on the role of substrates or active materials.

Paper and textiles made from herbaceous biomass are promising flexible substrates because of their high mechanical strength, flexibility, and biocompatibility. In addition, compared with nonbreathable substrates, the porous structure of paper and textiles provides them with excellent breathability (> 6000 g m$^{-2}$ day$^{-1}$) [139]. Recently, Rauf et al. fabricated flexible linen textile-based humidity sensors by direct-coating an active layer of a metal–organic framework (Fig. 5c) [140]. The resulting sensor exhibited a highly selectivity of 0.6 fF per %RH (relative humidity) with a detection limit of ~ 0.71% RH. Moreover, this sensor also showed a high degree of selectivity toward water vapor in the presence of several volatile organic compounds (Fig. 5c).

Unlike hydrophobic substrates, the abundant hydrophilic hydroxyl groups on the surface of paper and textiles allow them to be both active materials and substrates for humidity detectors. The abundant hydroxyl groups can cause physisorption of water molecules and can produce conductive ions ($H_3O^+$ and $OH^-$), which could slightly increase conductivity (Fig. 5d) [141]. However, this kind of sensor has the shortcomings of low response/recovery speed and low humidity sensing response at low humidity. Overall, paper and textiles made from herbaceous biomass could be promising flexible substrates or active materials for flexible biosensing devices, and some of them exhibit good sensitivity and signal quality.

### EES Devices

EES devices, including electrochemical batteries, supercapacitors, and biofuel cells are closely related to people's daily lives. Traditional EES devices are typically stiff and rigid, and cannot easily meet the requirements of next-generation electronics for flexibility, lightweight, and wearability. Paper- and textile-based EES devices have the potential to meet these requirements. Moreover, the hierarchically porous structure and hydrophilic nature of these devices could increase the loading of conductive materials and minimize inactive materials, which facilitates the formation of a good electrode-electrolyte interface and electrons/ions transfer, thereby producing high-performance EES devices. As shown in Fig. 5e, Liu et al. reported a flexible paper-based supercapacitor via vapor printing. The high porosity and SSA of paper resulted in good electrical performance because of the excessive loading of active materials [142]. The resulting paper-based supercapacitors exhibited a high specific capacitance (3.92 F $cm^{-3}$), energy density (~ 1 mWh $cm^{-3}$), and power density (0.42 W $cm^{-3}$).

Furthermore, a low thermal expansion is also desired for EES devices. Compared with most polymer-based materials, cellulose-based paper and textiles have low thermal expansion rates at a temperature range of 100–200 °C. In addition, the thermal performance of paper and textiles can be further improved through various modification strategies. Zhang et al. fabricated a heat-resistant and flame-retardant paper by incorporating a nitrogen-phosphorous complex as a flame-retarding agent (Fig. 5f) [143]. The paper demonstrated good thermal stability and flame resistance. When used as the separator, the resulting lithium-ion battery exhibited thermal stability and stable cycling performance even at 120 °C.

### Generators

Flexible generators that can collect energy from the surrounding environment (such as motion, sweat, and body temperature) and convert it into electricity are ideal for self-powered wearable electronics. Considering the low price, environmental friendliness, breathability, and biocompatibility, paper- and textile-based generators have attracted widespread attention, especially in some newly developed energy-harvesting technology, such as triboelectric nanogenerators (TENGs), hydroelectric nanogenerators (HENGs), and piezoelectric nanogenerators.

Cellulose paper and textiles can easily lose electrons when rubbed against materials that are likely to gain electrons (i.e., the contact electrification effect). Coupling with the electrostatic induction effect is the main mechanism of paper- and textile-based TENGs. Recently, Yang et al. fabricated a flexible TENG using a paper substrate and multi-walled carbon nanotube-coated paper electrode (Fig. 5g) [144]. The resulting TENG exhibited excellent mechanical stability under mechanical deformation (folded, curled, and tailored) and could be used to charge commercial capacitors or as a self-powered sensor to capture human motion signals. Moreover, the TENG also showed remarkable washing durability, proving the possibility of using paper-based TENGs for durable wearable electronics.

The abundant hydroxyl groups on the surface of cellulose paper and textiles make them usable in moisture-induced energy generation. The hydroxyl groups can absorb water and produce a large amount of hydrogen ions via a hydrolysis effect (Fig. 5h). Subsequently, hydrogen ions can diffuse along the concentration gradient and thus generate electricity. Based on this premise, Xu et al. designed a pencil-paper-based HENG, which can generate a voltage of 480 mV for more than 2 h from ambient humidity through a single-unit device (0.87 $cm^2$) [139].

Cellulose paper and textiles are also piezoelectric materials because of the orderly arrangement of the polar hydroxyl in the cellulose crystal, enabling them to generate electricity under mechanical stress [145]. However, commercial piezoelectric polymer has a longitudinal piezoelectric constant of 21 pC $N^{-1}$, whereas that of natural cellulose is only 0.4 pC $N^{-1}$ [146]. Therefore, high-piezoelectric activity materials need to be added into cellulose fibers to improve their piezoelectric performance. Besides being inexpensive, flexible, biocompatible, and breathable substrates, paper and textiles made from herbaceous biomass can also be active materials for various flexible generators because of the triboelectric, hydroelectric, piezoelectric, and electrostatic effects of cellulose.

## Application of Herbaceous Biomass at the Nanoscale

Nanocellulose (CNFs and CNCs) has been used in numerous applications because of its high aspect ratio, high crystallinity, hydrophilicity, abundant hydrogen bonds, high SSA, and chirality. The four primary types of herbaceous biomass-derived nanocellulose materials or composites are filaments, membranes, hydrogels, and aerogels. These materials or composites have been used in a variety of applications, including high-transparency and smooth substrates, self-healing materials, shape memory materials, sensors, and ion-conductors.

### Filament

Filament composed of nanocellulose or nanocellulose composites can be prepared via dry or wet spinning, during which the fiber morphology and orientation is adjusted by changing the processing conditions. However, the cost of nanocellulose-derived filament is still much higher than natural microfiber, and extensive research is aimed at lowering these production costs by developing inexpensive feedstocks and optimizing processing methods. Oksman et al. prepared continuous-aligned CNF-based filaments from herbaceous biomass waste (banana rachis) via dry spinning without using organic solvents, which are typically required for wet spinning processes [145]. Figure 6a illustrates this dry spinning method, in which concentrated and randomly oriented CNF suspensions (~ 6.5–12 wt%) were extruded at different speeds and subsequently dried. In the study, the highest tensile modulus and strength of the resulting CNF-based filaments reached 12.6 GPa and 222 MPa, respectively, by increasing the CNF alignment.

## Nanopaper

In addition to their good mechanical performance, low cost, and low carbon footprint, nanocellulose-based nanopaper derived from herbaceous biomass also has good transparency and low surface roughness. When used as a conductive substrate, the conductivity is proportional to the surface roughness of the substrate. Huang et al. reported a high-performance nanocellulose-based solar cell using nanocellulose derived from cotton linters via rod coating methods to replace traditional polymer substrates (polyethylene terephthalate and polyethylene naphthalate) (Fig. 6b) [146]. The surface of the nanopaper was coated with acrylic resin to further decrease surface roughness (as low as 2.151 nm) and avoid deformation in humidity owing to its high hydrophilicity. The resulting solar cells demonstrated a power conversion efficiency of 4.3%, a high power per weight of 0.6 W g$^{-1}$, and more than 80% efficiency after 50 bends.

CNC-based nanopaper can also be used as intelligent responsive membranes for external stimuli, including temperature, humidity, and pressure changes. Because of the chirality of CNCs, CNC-based membranes can self-assemble via slow evaporation and display a variety of colors [147]. The pitch of such chiral nematic structures can be changed by external stimuli, which results in a color change. Liu et al. prepared a CNC/glycerol nanopaper for intelligent, responsive iridescent membranes [148]. Figure 6c delineates the reversible color responsiveness of these membranes in response to changes in humidity. Glycerol was added as a hygroscopic agent to regulate the helical pitch by controlling the evaporation rate and increasing its responsiveness to moisture.

## Hydrogels with CNFs as Building Blocks

CNF-based hydrogels derived from herbaceous biomass comprise hydrophilic cellulose networks with good water

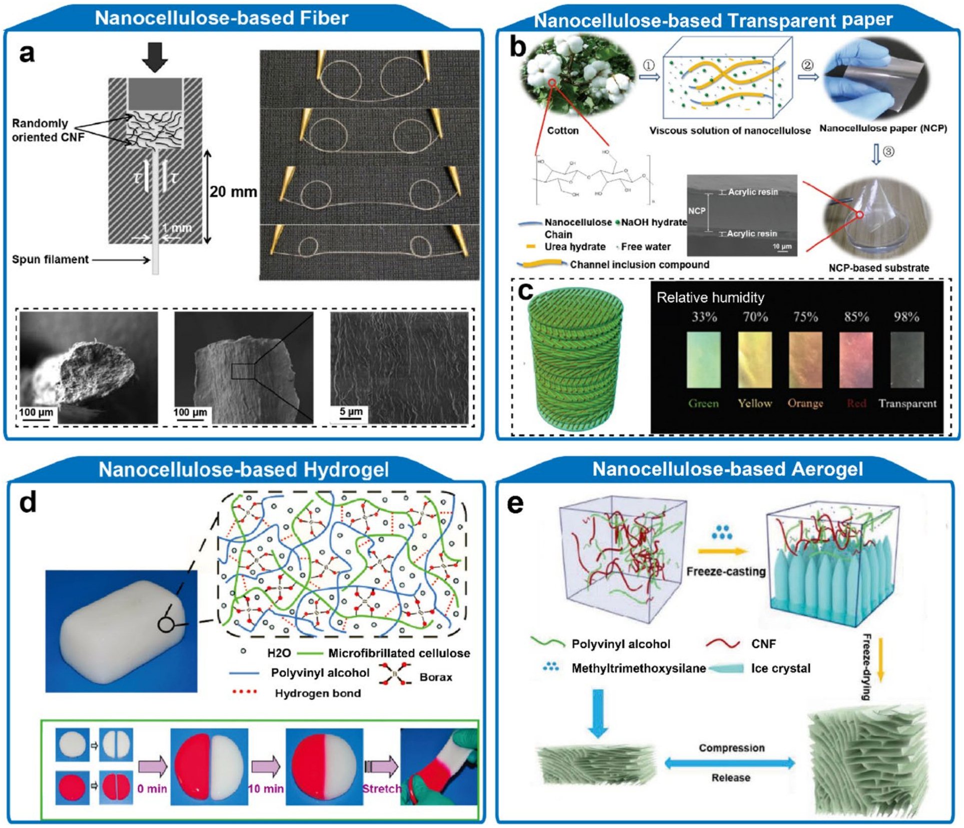

**Fig. 6** Nanocellulose-based functional materials. **a** Schematic illustration of nanocellulose-based fiber preparation, photographs and scanning electron microscope images of the fiber [145]. **b** Schematic illustration of nanocellulose-based transparent membrane [146]. **c** Schematic illustration of CNC-based intelligent responsive membranes and photographs of its color change under different moisture conditions [148]. **d** Schematic illustration of prepared self-healing hydrogels and photographs of its self-healing performance [149]. **e** Schematic illustration of nanocellulose-based aerogel with hierarchical structures [152]

uptake capacity. These hydrogels have been extensively used in many emerging applications, including biomedical devices, environmental remediation, and stimuli-responsive materials [149, 150]. Huang et al. prepared a self-healing and pH-responsive hydrogel composed of bamboo-derived CNFs. After sufficient mixing with polyvinyl alcohol and borax by ball milling, bamboo-derived CNFs form a well-dispersed network in water and generate reversible didiol-borax linkages with borax (Fig. 6d) [149]. The resulting hydrogels possessed good stiffness because of the inherent strength of bamboo-derived CNFs, as well as self-healing behavior due to the reversible didiol-borax linkages.

### Aerogels with CNFs as Building Blocks

Nanocellulose-based aerogels have ultralow densities, thermal conductivities, and high SSAs. These materials have been used in various applications, including insulation, absorbent materials, tissue engineering, and energy storage. However, low mechanical strength (compression modulus of ~ 0.1–2 MPa) and inherent hydrophilicity of nanocellulose aerogels limit their wide range of applications [151]. Strategies to increase reinforcement such as cross-linking with organic polymers and surface hydrophobicity treatments (e.g., esterification, silylation, and amidation) have been extensively used to fabricate mechanically robust and hydrophobic aerogels. Nanocellulose composite aerogels with enhanced mechanical performances have been generated by direct mixing or in situ polymerization with various polymers, such as polyvinyl alcohol, polydopamine, poly(aminophenyl) silsesquioxanes, and polypyrrole [151]. Recently, herbaceous biomass-derived nanocellulose composite aerogels with controllable aligned hierarchical structures have attracted great interest because of their high strength, stiffness, and shape recovery. Zhang reported silylated bamboo-derived CNF aerogels with aligned porous structures by directionally freeze-casting a mixture of CNF suspension and methyltrimethoxysilane (Fig. 6e) [152]. The resulting aerogels exhibited promising strength in the axial direction (specific compressive modulus of 5.50 MPa $cm^3/g$) and excellent shape recovery in the radial direction (92% recovery after 100 cycles at 80% compression).

## Summary and Outlook

As a low-cost, abundant, and sustainable raw material, herbaceous biomass has long been used in many applications, including fuel, animal feed, fertilizer, paper, textiles, food, and chemicals. The development of modification strategies enables the use of advanced herbaceous biomass-derived materials in many emerging fields. For instance, modified herbaceous biomass exhibits good tensile and flexural strength for high-performance structural materials, hydraulic and ionic conductivity for solar desalination and ion-selective transportation, and hierarchical porous structures for energy storage and conversion.

Some typical herbaceous biomasses, such as bamboo and sugarcane bagasse, are also suitable raw materials for extraction of cellulose fibers and nanocellulose with high yield and low chemical and energy consumption. Different species of herbaceous biomass provide a wide range of dimensions, crystallinity, and DP of cellulose, each of which can significantly affect the fiber and nanocellulose properties. The use of fibers and nanocellulose from herbaceous biomass provides an inexpensive and sustainable method to fabricate high-performance multifunctional materials exhibiting flexibility (e.g., filaments, paper, nanopaper, textiles), low thermal conductivity/thermal insulation properties (e.g., aerogels), response to stimuli (e.g., nanopaper, hydrogels), transparency (e.g., nanopaper, hydrogels), and absorbency (e.g., aerogels).

Conclusively, herbaceous biomass shows great potential for scalable fabrication of multifunctional materials. Some challenges require additional research, including development of inexpensive and environmentally friendly modification strategies, increasing the performance and durability of the resulting materials, and creating standardized supply, production, storage, and transportation systems.

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## Declarations

**Conflict of Interest** The authors have no competing interests to declare in the submission of this manuscript, and the manuscript is approved by all authors for publication.

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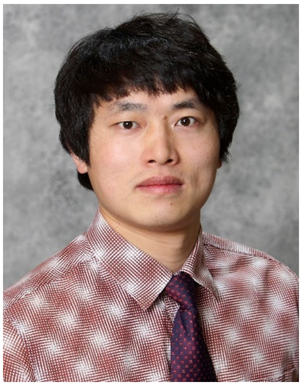

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