Emerging Applications of Cellulose Nanofibers

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Abstract

Cellulose is the most abundant biopolymer on Earth. In addition, it is renewable, biodegradable, and relatively cheap. Cellulose nanofibers (CNFs) have been produced most commonly from plants, algae, and bacteria. They can be isolated, e.g., from wood-derived fibers that have been microrefined to microlevel and even to nanolevel. In this chapter, we comprehensively review the unique properties and emerging applications of CNFs. We anticipate that CNFs as a new environmentally friendly material will be widely used in many areas such as reinforcement of polymers, energy production and energy storage, environmental protection and improvement, and healthcare. Therefore, there is a necessary to do more research on the potential emerging applications of CNFs.

Keywords

Cellulose nanofibers · Unique properties · Energy production · Energy storage · Environmental protection and improvement · Healthcare and biomedical application

Introduction

Cellulose-based materials have been extensively highlighted because of the increasingly prominent environmental problems. Cellulose has unique properties over conventional polymers including renewability, good biocompatibility, excellent mechanical properties, tailorable surface chemistry, and interesting optical properties. The growing demand for environmental sustainability has encouraged research on various cellulose-based materials to minimize the environmental impact of conventional hazardous materials [1]. To date, nanocellulose holds promise in many different applications including energy production, energy storage, environmental protection and improvement, food, pharmaceuticals, and tissue engineering [2, 3]. However, the poor suspension properties of cellulose in water, as well as its poor dispersion in hydrophobic polymers, limit their wider applications. To overcome this drawback, individualization of cellulose fibers into nanocellulose along with their surface modification and molecular grafting has been intensively investigated.

Traditionally, cellulose is extracted from plant tissue (trees, cotton, etc.) and it also can be produced, using certain bacterial species yielding a very pure cellulose product so-called “bacterial cellulose.” In plants, cellulose is typically present as a mixture together with hemicelluloses, pectin, lignin, oils, and other substances. Therefore, isolation of cellulose nanofibers is conducted by physical, chemical, and biological treatments such as cryocrushing [4], grinding [5], high-pressure homogenization [6], ultrasonication [7], acid hydrolysis [8], TEMPO-mediated oxidation [9], and enzyme hydrolysis [10]. The isolated nanofibers have different structures, length, diameter, functionalities depending on the sources and preparation processes [5–10]. To date, several types of cellulose nanomaterials have been

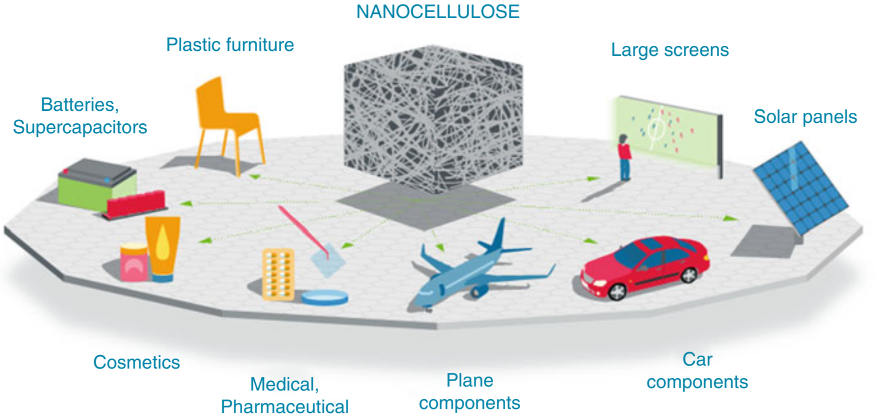

Fig. 1 Potential applications of cellulose nanofibers [12]

developed in recent years and they were classified depending on their geometry: cellulose microfibers (CMFs), cellulose nanofibers (CNFs), cellulose nanowhiskers (CNWs) or nanocrystals (CNCs), and cellulose nanoparticles [11]. As shown in Fig. 1, applications of nanostructured cellulose are almost infinite due to the properties of electrical conductivity, high absorption, and thermo-stability as well as lightweight but with eight-times higher tensile strength than steel. Besides, the material can consist of sheets and other structures, such as laminates or transparent films, and can be integrated into many different high-performance materials for the customer and industrial and biomedical applications [11].

Cellulose is the most appropriate natural polymers for the preparation of various types of nanomaterials. It is the most abundant natural polymer with unique properties such as low density, hardness, and ability to structural and chemical modification. The geometric dimensions of nanocellulose may vary depending on the origin of cellulose microfibrils and the physical and chemical treatments applied to cellulose. Cellulose nanoparticles have a spherical shape with a diameter of 50–150 nm [13]. CNFs and CNWs are different in shape, size, and composition. CNFs consist of cellulose nanofibrils with a high aspect ratio, i.e., 5–20 nm in diameter and several micrometers in length. The geometric dimensions of CNWs may vary depending on the origin of cellulose microfibrils and the physical and chemical treatments applied to cellulose. CNWs produced from wood are typically 3–5 nm in diameter and 100–300 nm in length [14]. CNFs and CNWs have the advantage of creating low environmental impact in their production and disposal and present high barrier properties to oxygen and other gases. CNFs and CNWs can be used to manufacture large screens [15], solar panels [16], batteries [17], supercapacitors [18], and even materials that react to external stimuli such as heat, light, electricity, pH, or pressure [19–22]. However, CNFs have shown improved performance over CNWs due to their higher aspect ratio. For example, both CNFs and CNWs have shown reinforcing effects in polymer nanocomposites. At the same concentration, CNFs led to higher strength and modulus than did CNWs due to CNFs larger aspect ratio and fiber entanglement [23]. There are different reasons motivating the interest of CNFs as a special category of cellulose nanomaterials: (i) the nanoscale dimension (diameter) and high specific surface area, (ii) the very high length up to several micrometers, (iii) the high intrinsic mechanical strength along with good flexibility, and (iv) their inherent tendency to form strong entangled network [24].

This chapter discusses the unique properties of cellulose nanofibers and their emerging applications in the reinforcement of polymers, energy harvesting, and storage, environmental protection, and improvement, drug delivery, tissue engineering, healthcare, foods, and constructions.

Reinforcing Materials

Cellulose nanofibers are generally used for composite reinforcement because the crystalline part of cellulose has a high stiffness (modulus of ca.150 GPa and tensile strength of ca.10 GPa) [19, 20]. A significant enhancement of thermal and mechanical properties was achieved with the addition of a small amount cellulose nanofibers to the polymer matrix [21]. CNF reinforced polymers with low cost, fire resistance, low density, nontoxicity, biodegradability, and other characteristics have attracted a lot of attention in the composite material field. So far, the effect of CNFs as a reinforcing component to the composite has been studied extensively [25]. For instance, the mechanical properties of the reinforced polylactic acid nanocomposites were shown to change as the content changes. The elastic modulus of PLA was increased from 1.17 to 2.12 GPa by adding 5% wt% CNFs, and the tensile strength was doubled [26]. Saba and his coworkers [27] have proved that the incorporation of CNF filler enhanced the thermal stability and remarkably improved properties for all epoxy nanocomposites compared to neat epoxy. Pelissari [22] indicated that compared with the control film, the polymers reinforced with CNFs present higher tensile strength, Young's modulus, water-resistance, opacity, crystallinity, and thermal stability. All films fabricated from CNFs after enzymatic pretreatment are visually transparent, although their transmittance is only within the range of 2–14%. Regardless of the wood species, all films made from wood CNFs have a transmittance of about 14% [28].

CNFs become very important because incorporation of reinforcement has been related to improvement in overall performance of biopolymers. Saba and his coworkers [27] validate that the addition of CNFs could enhance mechanical, thermal, and barrier properties of biopolymers that have many drawbacks. Figure 2 shows the effect of different amounts (0.5%, 0.75%, and 1% by wt) of CNFs loading on the tensile strength of epoxy resin composites. From Fig. 2a, it is evident that the tensile strength of epoxy enhances by adding CNFs. The developed nanocomposites became relatively stiffer and tougher than pure epoxy because of the hard and stiff nature of CNF particles. Figure 2a shows that the incorporation of 0.5% and 0.75% CNFs into the epoxy resin primarily increased the tensile strength. Such improvements in the mechanical properties also revealed for the scalable CNFs derived from the bleached

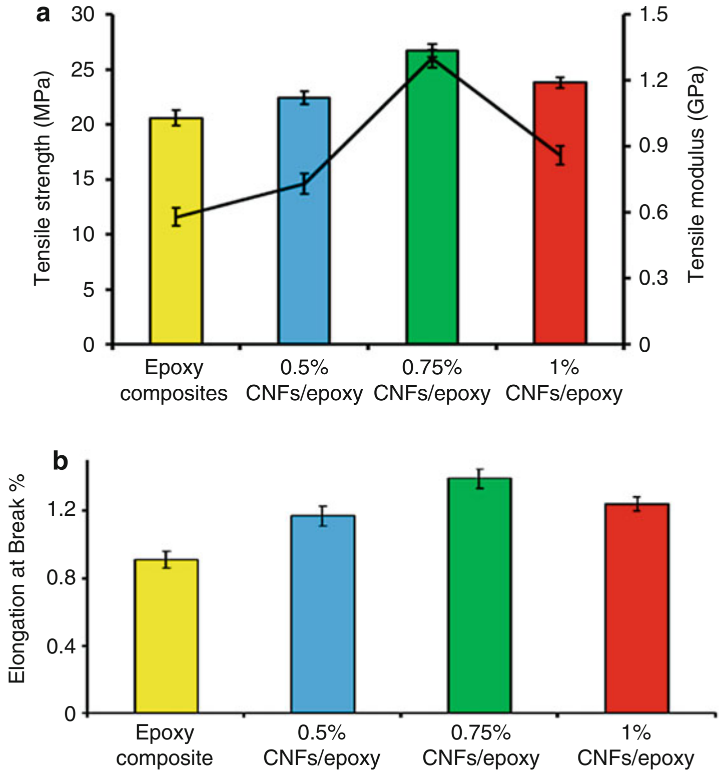

Fig. 2 Effect of CNFs filler loadings on the mechanical properties of epoxy: (**a**) Impact on tensile strength. (**b**) Impact on elongation at break. (Copyright 2017, Elsevier [29])

Eucalyptus Kraft pulp-based nanocomposites. The considerable increase in the tensile strength with the incorporation of CNFs is attributed to the homogeneous dispersion of the nanofibers in the epoxy matrix, which induces a better transfer of the longitudinal stress between the epoxy matrix and CNFs. Figure 2b illustrates the elongation at break values when the longitudinal stress/load was applied to the nanocomposite. The obtained trends for the elongation at break are similar to that of the tensile strength and tensile modulus (Fig. 2a). The results indicate that the elongation at break value increased from 0.5% to 0.75% by the addition of CNFs, while the further addition of CNFs up to 1% decreased the elongation at break value. This can be ascribed to the higher tensile strength value and stiffness offered by the immobilization of polymer chains by the network in the 0.75% CNFs filler as compared against the 0.5% and 1% CNFs loading in the epoxy composites.

Cellulose nanofibers are also promising materials for producing foams with excellent properties, better than traditional polymers and other renewable fibrous material. Wicklein et al. [30]. produced thermally insulating and fire-retardant lightweight anisotropic foams based on CNFs and graphene oxide (GO) using freeze-casting suspensions of GNFs, GO, and sepiolite nanorods. The obtained nanocomposite foam shows better fire-retardant properties than traditional polymer-based insulating materials. The foams are ultralight and exhibit excellent combustion resistance as well as a thermal conductivity of 15 mW m^{-1} K^{-1}, which is about half that of expanded polystyrene.

Ho et al. [31] developed composite materials consisting of cationic nanofibrillated cellulose and layered silicates by high-shear homogenization followed by pressure filtration and vacuum hot-pressing and then tested their mechanical behavior and water vapor barrier performance (Fig. 3). The authors also investigated the mechanical behavior and water vapor barrier performance of a number of hybrid films formed by 13 different clays and micas, although using instead cationic nanofibrillated cellulose (in the range of 0–85 wt%). The water vapor barrier and mechanical properties (tensile strength, E-modulus, strain at break) of the composite films were investigated. In their research, the water vapor permeability of composite films with 50% inorganic contents was 0.025 mg μm m^{-2} s^{-1} kPa^{-1} measured in the

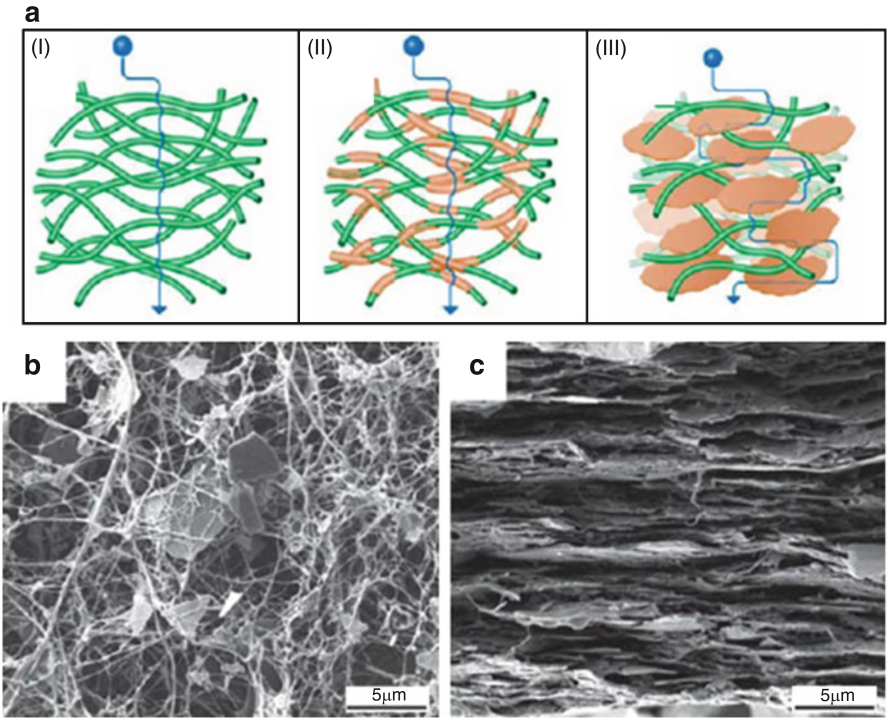

Fig. 3 Composites made of cationic cellulose nanofibers (CNFs) and layered silicates show excellent water vapor barrier and mechanical properties: (**a**) Three model types for water vapor diffusion pathways: (i) neat CNFs, (ii) trimethylammonium-modified CNFs, and (iii) the commonly used “fiber-brick composite.” (**b**) SEM images of a dried suspension of CNFs–micacomposite, and (**c**) a fracture surface of the CNFs–mica film cross-section [31]. (Copyright 2012, American Chemical Society)

condition of 85% relative humidity. The composite films with the best barrier properties showed a tensile modulus of 9.5 GPa, a strength of 104 MPa and a strain at break of 1.9% in the condition of 43% relative humidity.

The lignin aerogels that are characterized by high porosity and compressibility would have promising implications in bioengineering field, sound-adsorption, and damping materials. However, it is a challenge of creating this aerogel from adhesive lignin. CNFs working as green adhesion agent with strong mechanical performance help to solve this problem [32]. As shown in Fig. 4, CNFs and lignin particles dispersed in the ionic liquids which are composed of the anions and cations. Anions are represented by spherical particle and cations are represented by ellipsoids with a polar headgroup. Figure 4 presents the dissolved state of the starting cellulose, lignin/1-butyl-3-methylimidazolium chloride (BMIMCl) solution. The gel formation process is shown in Fig. 4b. The formation of wet gels lies in the interactions between ions and water molecules. Figure 4c shows the schematic illustration of the nanostructure of samples after drying, in which lignin particles are entrapped by cellulose nanofibers and formed hydrogen bonds with them [32].

Papermaking "Flexible Transparent Nanopapers"

Nanocellulose has shown some potential applications in the field of papermaking and coating. The addition of nanocellulose (e.g., CNWs and CNFs) to the base paper is typically carried out by one of the following strategies: (i) the direct addition into

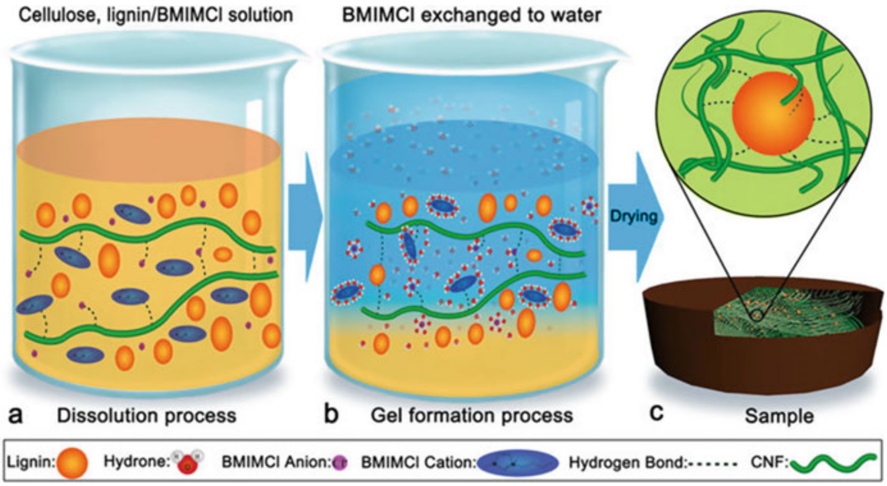

Fig. 4 Synthesis of lignin aerogel as an adhesion agent with strong mechanical performance, sound-absorption and thermal insulation: (**a**) dissolution of CNFs in lignin/1-Butyl-3-methylimidazolium chloride (BMIMCl) solution; (**b**) Wet gel formation process through the interactions between CNFs, ions and water molecules; and (**c**) Nanostructure of the samples after drying, in which lignin particles are entrapped by CNFs [32]. (Copyright 2016, Nature)

the pulp, (ii) the incorporation in multiply paperboard, (iii) the integration in the wet-end chemistry as a retention agent, and (iv) the development of new hybrid papermaking materials [33]. The use of CNFs in the paper stock formulation, by 10 wt% to an unrefined pulp, can increase the tensile strength from 20 to 34 N with a significant grammage reduction [34]. In contrast to traditional paper, the dense packing of CNFs with a pore size less than 100 nm causes only the ink carrier to be absorbed through the pores while the inkjet inks remain on the paper surface [35].

Nanopapers made of cellulose nanofibers and clay nanosheets provide good oxygen gas barrier resistance, where the oxygen molecules penetrated much more slowly due to nanofiber entanglements [36]. Transparent cellulose nanofiber(CNF)/ clay nanopaper is considered as a sustainable and efficient fire-retardant coating for its unique masonry structure. The treatment of clay nanopaper mainly depends on the physical hybrid assembly of nanoparticles and is environmentally friendly and very simple due to its water-based process. The clay nanopaper is manufactured in colloidal water, which is the mixture of CNFs in the form of montmorillonite(MTM) nanosheets and clay colloid aqueous solution. The colloidal mixture is treated by filtration and evaporation, as it is shown as in Fig. 5a. The MTM platelets in resulting nanopaper show strong in-plane orientation and are embedded in a CNFs network which is tough and ductile (Fig. 5a) [37].

The ordered nanocomposite structure in Fig. 5 has interesting properties. The strength is in the range of mineralized tissue in nature and can be superior to other synthetic polymer/clay bulk nanocomposites (Fig. 5b). The gas barrier property is an interesting property due to preferential orientation of clay. For instance, the oxygen permeability of the clay nanopaper is two times higher than that of the multilayer polymer laminates and metal/polymer composites currently used, and in the same range as other promising technologies such as SiOx coatings which are very brittle, and even easy to crack (Fig. 5c).

More recently, much effort has been spent on preparing flexible electronics or printable electronics on cellulose nanopaper with the promise of low cost, excellent flexibility, light weight, transparency, inertness, recyclability, and high mechanical strength when compared to glass (silicone)-based or plastic-based electronics [38]. Cellulose nanopapers made of CNFs show some unique characteristics over conventional papers, including high transparency, enhanced barrier properties, high surface smoothness, low thermal expansion, and further possibilities for surface functionalization [39]. Cellulose nanofibers show improved performance and unique transparency and flexibility over other conventional materials like glass and plastics. Glass has well-suited low thermal expansion for use in electronic devices, but it is fragile. Plastic with excellent flexibility is one of the promising materials in flexible electronic devices; however, the extremely large thermal expansion coefficients which exceed 200 ppm K^{-1} limit its usage in flexible electronic devices successfully. In addition to the advantages mentioned above, CNFs can suppress light scattering due to the small interstices between the fibers; hence, the CNF films become transparent [40]. Recently, Choi et al. [40] fabricated foldable organic memory on cellulose nanofibril paper by employing initiated chemical vapor deposition (iCVD) for the polymerization of the resistive switching layer and inkjet printing (silver-

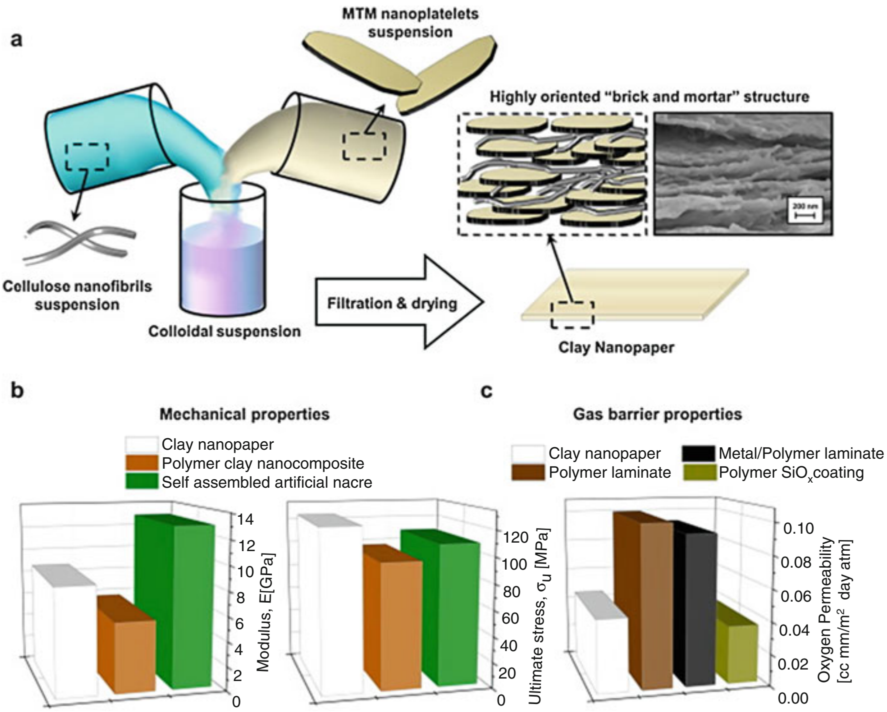

Fig. 5 Transparent CNFs/clay nanopaper as a sustainable and efficient fire-retardant coating (**a**) manufacturing process and oriented structure of CNFs/clay nanopaper; (**b**) comparison graphic of clay nanopaper with 50% MTM and other clay nanocomposites on mechanical properties; and (**c**) comparison graphic of measurement under humid conditions (23 °C 50% R.H.) and existing measurement on oxygen barrier properties. (Copyright 2015, Elsevier Ltd. [37])

based ink) of the electrode (Fig. 6). The device exhibits a low operation voltage of 1.5 V enabling battery operation compared to previous reports and wide memory window. The inkjet-printed pattern formed on cellulose nanofibril paper shows stable conductivity under a folded state and reliable nonvolatile memory performance even after folding the device into origami.

Energy Storage and Energy Harvesting

With the increasingly serious environmental pollution and people's awareness of environmental protection, the battery based on toxic metals such as Pd and Cd will be restricted, which prompted people to look for new environmental battery materials. The Li-ion batteries were featured by their high operating voltage, fast charge and discharge property, and long cycle life and have been therefore extensively concerned. The Li-ion batteries have been applied in some areas like mobile phone,

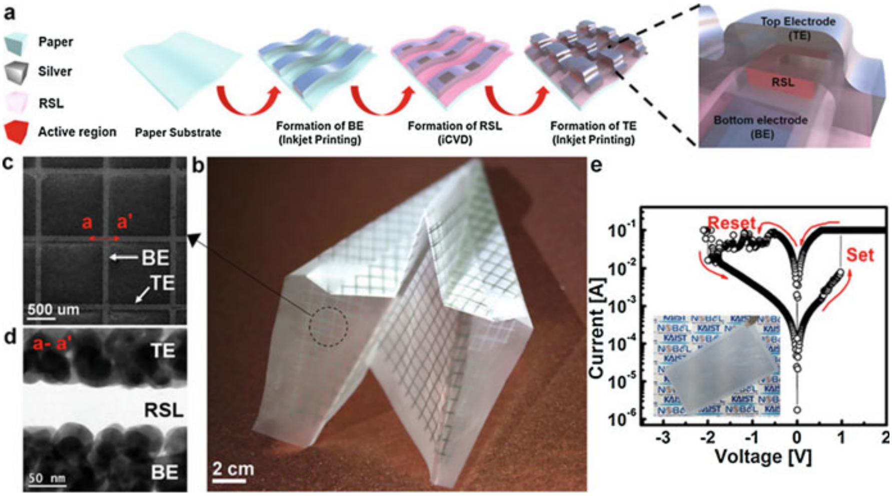

Fig. 6 Schematics of the fabrication process of cellulose nanopapers and their integration in substrate-based memory and printed electronics devices: (**a**) Inkjet printing and room temperature iCVD are suitable for fabricating the device on a paper substrate. (**b**) Fabricated nanopaper-based memory in the form of an airplane demonstrating the foldable memory feature. (**c**) Top view SEM image of the fabricated device. (**d**) Cross-sectional TEM image of the direction along a-a′ of Fig. 6c. (**e**) I-V characteristic of the fabricated device showing memory operation, (inset) the nanopaper based memory before origami [41]. (Copyright 2016, Nature)

laptops, digital camera, electric vehicles, and aerospace. But Li-ion battery can no longer meet the requirement of future application due to its limited energy density.

Cellulose is of many advantages such as film-forming property, good compatibility, structural stability, and mechanical properties compared to metals, glass, and plastics. A series of research on applying CNFs to constitute diaphragm material, electrode material, and electrolyte materials have been carried out. CNFs applied in diaphragm exhibit the advantages of higher thermal stability, higher ion conductivity, better cycle performance, and better safety performance compared to traditional diaphragm material such as PP [42]. Bacterial cellulose, a typical biomass material, attracted researcher's attention and exhibited promising application for electrochemical battery, because its properties of a large degree of polymerization and high crystallinity can extend the capability for graphitization during carbonization resulting in high conductivity and superior structure, and it is often used as electrolyte materials [43].

In recent years, a need for mechanically flexible and strong batteries has arisen to power technical solutions such as active radio-frequency identification (RFID) tags and bendable reading devices. Leijonmarck [44] proposed that the battery can be integrated into a single flexible paper structure for manufacturing flexible and strong batteries. Nano-fibrillated cellulose (NFC) is used as a binder of electrode material and diaphragm material. The battery paper is manufactured by continuous filtration of water dispersions containing battery components, which is a kind of papermaking process. The thickness of the battery paper is 250 μm and the strength of the battery paper is high. When the electrolyte is immersed, the strength is up to 5.6 MPa. The battery paper also shows good cycling performances with reversible capacities of 101 $\mathrm{mAhg^{-1}}$ $LiFePO_4$ at cycling (1C) and 146 $\mathrm{mAhg^{-1}}$ $LiFePO_4$ at cycling (C/10). It is equivalent to an energy density of 188 $\mathrm{mWhg^{-1}}$ in full paper battery at cycling (C/10).

Figure 7 shows carbonaceous bacterial cellulose (BC) coated with amorphous Fe_2O_3 nanoshells is introduced as a flexible framework for Li-ion batteries (LIBs) [45]. The carbonized BC nanofibers exhibit good electrical conductivity and mechanical stability. The amorphous nature of the Fe2O3 shell provides an enhanced capacitive-like lithium storage and flexible structure of the electrode compared with its crystalline counterpart [45]. In order to address the critical issues, such as short recycle life and low coulomb efficiency, Huang et al. [17] designed advanced Li-S batteries based on high carbonized bacterial cellulose (CBC) and high sulfur. The configuration of the Li-S battery consists in four layers i.e., relatively thick CBC aerogel, relatively thin CBC aerogel, separator, and Li anode from bottom to top. The resulting product showed good electrical conductivity and mechanical stability.

Supercapacitors with the properties of fast charge speed, long recycle life, and high energy conversion rate, and so on are promising energy storage devices. However, the traditional binder used in supercapacitor has the disadvantages of poor mechanical property, usage of the toxic solution in the manufacture, high price, and difficult to recycle. Cellulose nanofiber has become an alternative to traditional adhesives due to the pore structure and the character of absorbing electrolyte easily. In order to obtain higher cell voltages and energy densities, Tammela et al. [18] designed a capacity whose negative electrode employed carbon

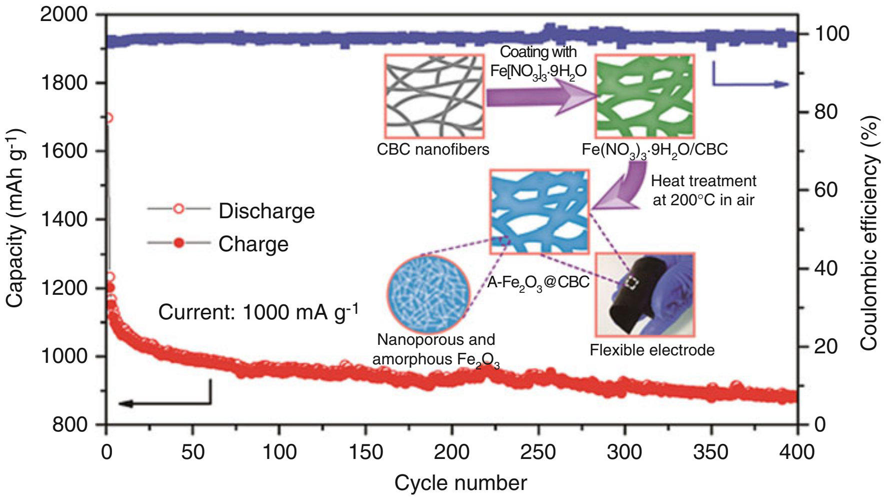

Fig. 7 Carbonized bacterial cellulose nanofibers coated with amorphous Fe_2O_3 nanoshells as a flexible anode for high-performance lithium-ion batteries. (Copyright 2016, Elsevier [45])

nanofiber(C-nf) obtained by straightforward heat treatment of PPy-cellulose, while the positive electrode employed PPy-cellulose. Interestingly, the negative electrode exhibited a very similar porous "spaghetti-like" nanofiber structure, and the positive electrode nanofibers with a diameter less than 100 nm are intertwined and form a three-dimensional network. They attained that the specific gravimetric capacitances of PPy-cellulose electrodes were 146 F/g, and the energy density was 33 J/g.

With the development of micro-electro-mechanical systems, the potentially implantable glucose fuel cell based on the electrochemical reaction between glucose and oxygen at two separated electrodes has extensively attracted the researchers' attention. Since oxygen and glucose are present in body fluid at the same time, it is necessary to separate the anode and the cathode in order to avoid a drastic reduction in the cell voltage because of mixed electrode potentials. Noble metal catalysts are preferable for implantable glucose fuel cell applications than reaction-selective enzymatic catalysts or microbial catalysts, which are employed in implantable glucose fuel cell now, for their long-term stability and sterilizability. Platinum nanoparticles (Pt NPs), as the most known catalysts, could not be used in glucose fuel cell directly because they are catalytic for glucose oxidation and oxygen reduction, which could induce an electrochemical short circuit.

In order to solve the problem of mixed potentials, Liu et al. [46] designed a conductive hydrogel electrode membrane based on bacterial cellulose (BC), multi-walled carbon nanotubes (MWCNTs), and Pt NPs. Their research showed that even with a very limited catalytic activity in the case of a platinum plate cathode, the performance of the fuel cell is close to that of an implanted RANEY® type platinum fuel cell.

Foldable electric devices that can be put in a pocket and opened out when used like newspapers and magazines have attracted many researchers' attention. The transparent conductive substrates that are most used in portable electronic devices such as laptops and E-books are made of doped metallic oxide glass which is heavy and brittle. In order to address the issues above, transparent conductive plastic substrates using carbon nanotubes, silver nanowires, or graphene instead of doped metallic oxides have been developed. The process of manufacturing transparent conductive plastic substrates is complex, and the foldability of transparent conductive plastic substrates cannot satisfy the demand for the future foldable electronic device.

Masaya et al. [47] designed a process to fabricate optically transparent, electrically conductive nanofiber paper using 15-nm-wide cellulose nanofibers and 50-nm-wide silver nanowires. As shown in Fig. 8a, traditional papers (left) were fabricated with cellulose fibers with a diameter ranging from 15 to 50 μm; the nanofiber paper (middle) was produced by using cellulose fibers with a diameter of 15 nm, and high electrical conductive paper (left) was fabricated by depositing silver nanowires onto nanofiber papers. The nanofiber paper showed a high total transmittance of 91.4% at a wavelength of600 nm (Fig. 8b). Three posttreatments, i.e., heating method (upper), pressing method (middle), and dropping method (lower), have been used to deal the silver-nanowire-coated nanofiber paper (Fig. 8d), and the total transmittance of the resulting sample is presented in Fig. 8c.

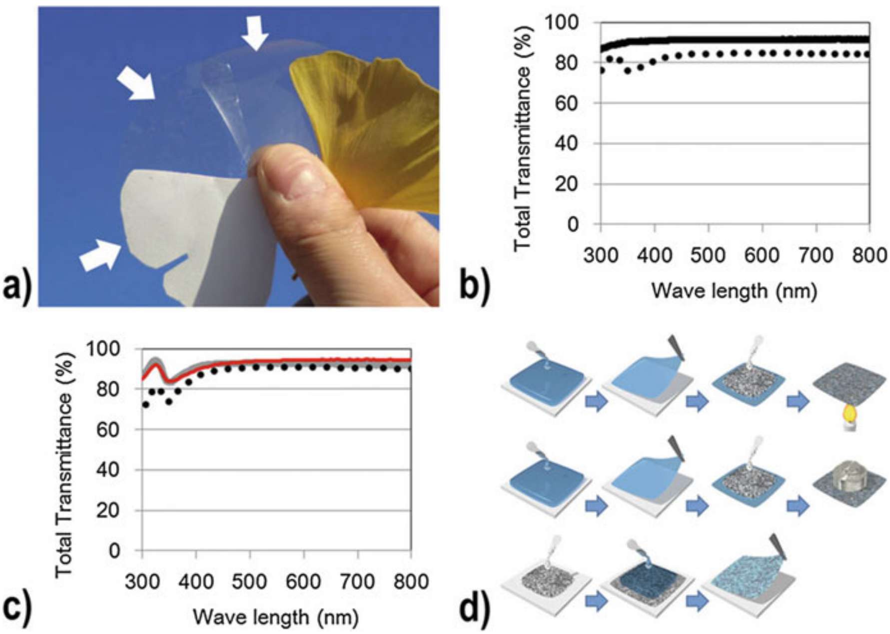

Fig. 8 Transparent conductive nanofiber paper for foldable solar cells. (**a**) Transparent conductive nanofiber paper (right), transparent nanofiber paper (center), and traditional white paper (left). (**b**) Optical transmittance of two kinds of transparent nanofiber papers in which dotted line represented the transparent conductive nanofiber paper and solid line representing the normal transparent nanofiber paper. (**c**) Optical transmittance of Ag-nanowire layers fabricated on the transparent nanofiber paper by three different measures, i.e., heating method, pressing, and dropping which are shown in dotted line, gray line, and redline, respectively. (**d**) Three Fabrication processes of transparent conductive nanofiber paper where the CNF dispersion is shown as blue and the Ag nanowire suspension is shown as black. The heating method, pressing method, and dropping method are shown from top to bottom. (Copyright 2015, Nature [47])

Sensing Devices

Sensors are extensively employed in automobiles, aerospace, safety, indoor air quality, environmental control, food, industrial production, and medical sectors. The demand for sensors has exponentially increased in recent years. Despite the sensor research has been established, there is still a demand for new and improved sensors across the different industries. CNFs-based sensors are more sensitive than traditional sensors because of the high surface-to-volume ratio and increase in the detection limit. Besides, CNFs are particularly suitable for biosensor because of their biocompatibility with enzymes, microorganisms, and antigens. Therefore, more and more researchers become interested in CNFs-based sensors. Xu and his colleagues [48] successfully assembled an alcohol-sensing device comprising a thin-film sensor made of graphene nanosheets (Gr) and bacterial cellulose nanofibers (BC). Gr/BC sensors demonstrate fast response/recovery times and a wide range of alcohol detection (10–100%). This is a facile, green, low-cost route for the assembly of ethanol-sensing devices with potential for vast application. Figure 9 shows the assembly process from raw material to the sensor device. Gunji [49] also reported that the SiO_2/SnO_2 nanofibers synthesized by TEMPO-oxidized cellulose nanofibers have a high gas-sensitivity to 1000 ppm ethanol.

He et al. [50] reported that the electrical conductivity and mechanical properties of cellulose nanofiber/polyaniline (CNFs/PANI) composite films isolated from bamboo indicated that they could be used potentially in biological sensors. Pang et al. [51] reported a kind of room temperature ammonia gas sensor which was sensitive based on cellulose/TiO_2/PANI composite nanofibers. Qin et al. [52] reported a cellulose nanofiber/cationic conjugated polymer hybrid aerogel sensor for nitroaromatic vapors detection. CNFs can effectively prevent aggregation of the conjugated polymer cationicwater-solublepoly[9,9-bis[3-(N,N-dimethyl)-N-ethylammonium)propyl)-2,7-fluorene-alt-1,4-phenylene]dibromide (CPFD) backbones.

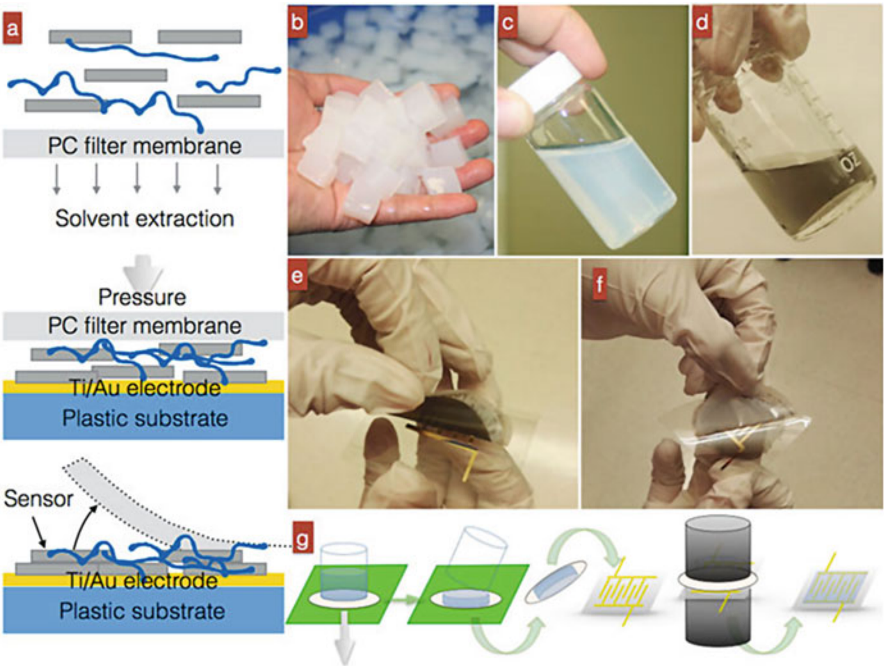

Fig. 9 Fabrication of alcohol-sensing device based on graphene nanosheets (Gr) and bacterial cellulose nanofibers (BC): (**a**) Gr/BC sensors are fabricated by vacuum filtration. (**b**) The BC hydrogels at 0.5 wt%. (**c**) The extraction process of 1 wt% aqueous suspension in BC after TEMPO treatment. (**d**) Gr/BC colloidal mixture with 40 wt%Gr volume. (**e**) At 40 wt% Gr wet base load Gr/BC device delamination sputtered Ti/Au electrode interdigital on a plastic substrate. (**f**) The process stripped the substrate from dry Gr/BC vacuum compression/drying at 60 °C. (**g**) The whole process of equipment assembly [48]. (Copyright 2017, Nature)

CNFs/CPFD aerogels have a large number of cavities due to their porous structure, which is enough to allow nitroaromatic compound (NAC) vapor to diffuse into the gel. Therefore, CNFs/CPFD gel sensors have high sensitivity for NAC steam.

Electronic Devices

The main application of nanocellulose in electronic devices is in making transparent flexible substrates, which is one important component of transparent flexible electronic devices affect the flexibility, transparency, weight, and even life of the resulting product. The most commonly used substrates are based on plastic, which has low degradability and can easily cause environmental pollution. Traditional cellulose papers as conductive films are also widely used in manufacture of electronic devices. However, the conductivity of conductive material deposited on traditional paper is lower than that of conductive material deposited on plastic for traditional paper's high surface roughness and high porosity. The cellulose nanopapers are a desirable choice for a new generation of environment-friendly electronic device substrate material because of its smooth surface, high transparency, low thermal expansion coefficient, high strength, biodegradability, and flexibility.

Gao et al. [53] prepared CNFs-based flexible transparent conductive paper successfully. The transparent conductive paper has good optoelectronic properties, excellent flexibility, and mechanical properties. It was reported that CNFs can also be used as a template for the effective decontamination of radioactive cesium [54–56]. CNF composites display good mechanical properties with low thermal expansion [57–59] and can serve as substrates for flexible electronics. The CNF composite substrates for flexible electronics are good alternatives to overcome the electronic waste since CNF pays the way for biodegradable substances [60, 61] and Eichhorn et al. [19] developed organic display system from CNFs. Such products can bring about changes in the communications display system and even spread to the entire industry of electronic telecommunication [62, 63].

Ou and his colleagues demonstrated a fabrication process of original paper's hierarchical structure into transparent nanopaper by ILs-polishing, as shown in Fig. 10. Because the refractive index of cellulose and air in voids is different, the original paper based on micron cellulose fibers and voids shows opacity. The voids between cellulose fibers ranged from nanoscale to microscale (Fig. 10b, c). The paper gets hyaline owing to the uniformity of refractive index on paper when the air in the void is completely removed or replaced by cellulose (Fig. 10d). Part of the surface dissolution of cellulose fibers is caused by the forced pressure resulting from the collapse of cellulose cell walls during hot pressing (Fig. 10e). Therefore, the high density and the uniform refractive index along the thickness of the paper are due to the fact that most of the voids in the air are replaced by cellulose. The perfect optical and mechanical properties are obtained by ILs-polishing the paper gotten by hot pressing of (Fig. 10f) [64]

Jung et al. (2015) reported a kind of high-performance flexible microwave and digital electronics that consume the smallest amount of potentially toxic materials on

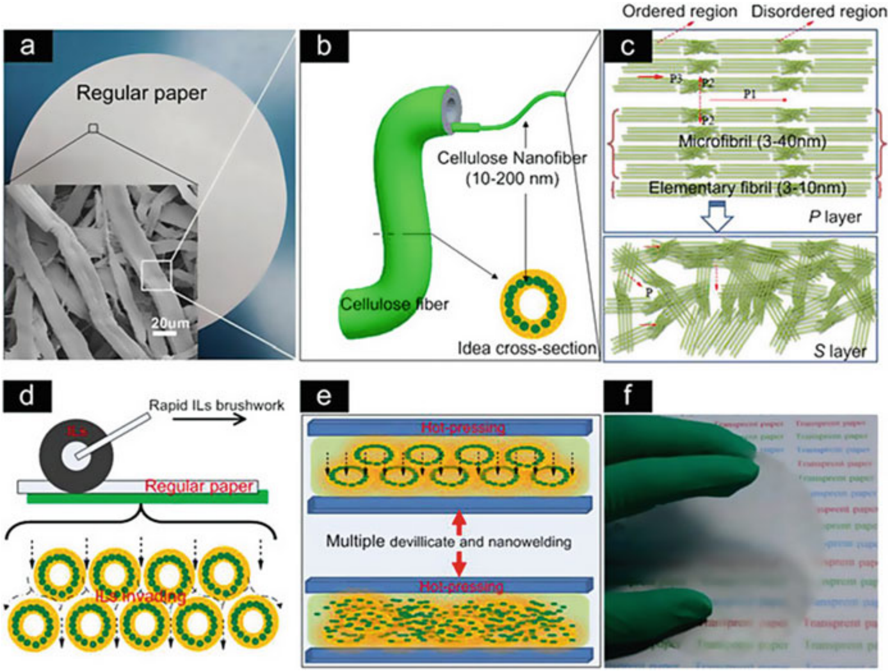

Fig. 10 Transparent nanopaper prepared directly from an ordinary paper by the fast ILs-polishing process. (**a**) Regular paper. Inset SEM image of the plain paper. (**b**) The ideal structure of individual CNF. Inset magnification map a single CNF cross-section. (**c**) Invasion pathways of various ion channels in cell wall P and L layers. The S-layer indicates the disordered region, which exists in the ordered region of blank microfibrils and crystalline basic fibers. (**d**) The intrusion model of cellulose fiber matrix in hot pressing process. (**e**) Formation and nanowelding process of cellulose nanofibers. (**f**) Digital image of nanostructure paper [64]. (Copyright 2017, Nature)

biodegradable and flexible cellulose nanofiber papers. Figure 11 outlines the procedure for manufacturing GaInP/gallium arsenide (GaAs) heterojunction bipolar transistors (HBTs) on CNF substrates via schematic illustrations [65].

A highly flexible nonvolatile memory as one of the most important competent of flexible electronic devices is an important flexible circuit competent. The smallest curvature of existing nonvolatile memory is limited to millimeters due to the largely decreasing memory properties while bending. Nagashima, Kazuki et al. [66] proposed ultra-flexible resistive nonvolatile memory using Ag-decorated cellulose nanopaper, which has been bent down to the radius of 350 μm without degradations in memory performance (Fig. 12). In order to test the flexibility of Ag-decorated cellulose nanopaper, first, they put the Ag-decorated cellulose nanopaper onto Alfoil as shown in Fig. 12a, b and then they used Ag-decorated cellulose nanopaper device to wrap around a5mmglass rod (Fig. 12c). The clear resistive switching events and good retention were confirmed for the present Al foil-based memory devices.

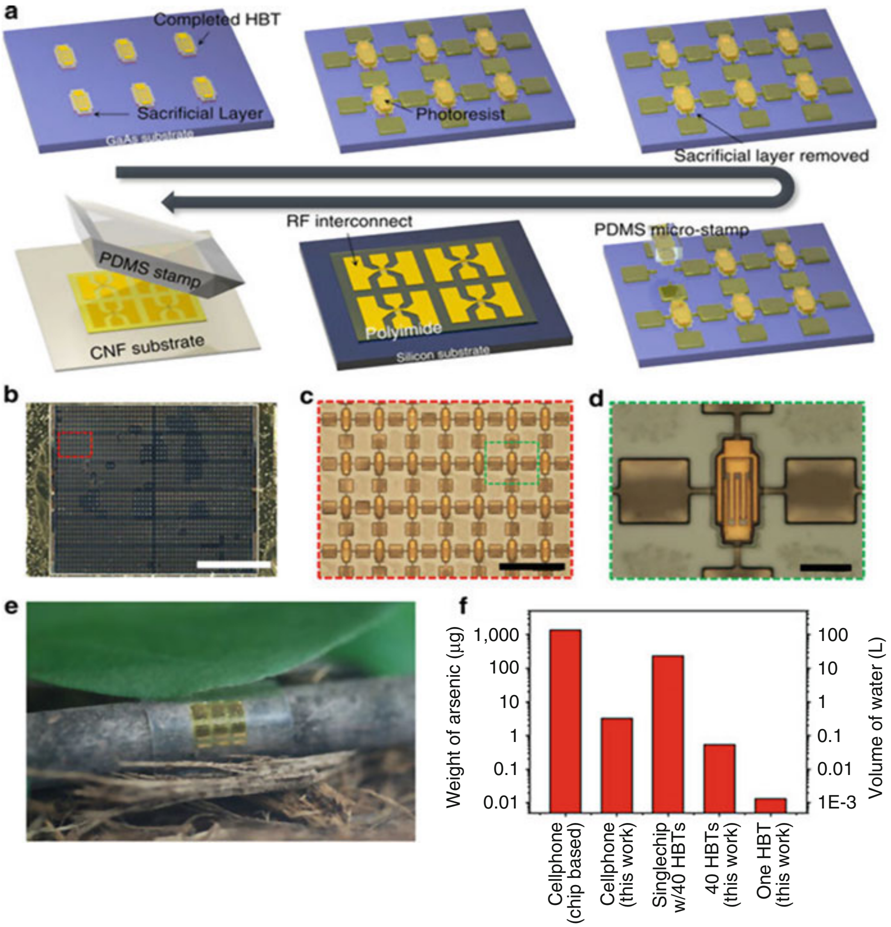

Fig. 11 High-performance green flexible electronics based on biodegradable cellulose nanopaper. (**a**) The fabrication process of GaInP/GaAs HBTs on a CNFs substrate. (i) HBTs are grown on sacrificial layers of GaAs substrates and released by photoresist protection anchors. Each HBT is picked up with a polydimethylsiloxane (PDMS) microstamp and printed on a temporary Si substrate. The equipment is printed with PDMS stamp to the CNF substrate after radio frequency (RF) interconnection metallization. (**b**) Optical image shows the 1500 release of the HBT in the 5 × 6 size of 2 mm GaAs substrate intensive arrangement. Scale bar, 2 mm. (**c**) Magnified image of the array. Scale bar, 200 µm. (**d**) Removable HBT is anchored to the substrate and photoresist. Scale bar, 30 µm. (**e**) Photo array substrate for the CNFs-HBTs around 3 mm radius stick. (**f**) A comparison chart shows the amount of arsenic corresponding to each type of device/transistor, and the amount of arsenic calculated in accordance with the Environmental Protection Agency (EPA) standard in this device/transistor [65]. (Copyright 2015, Nature)

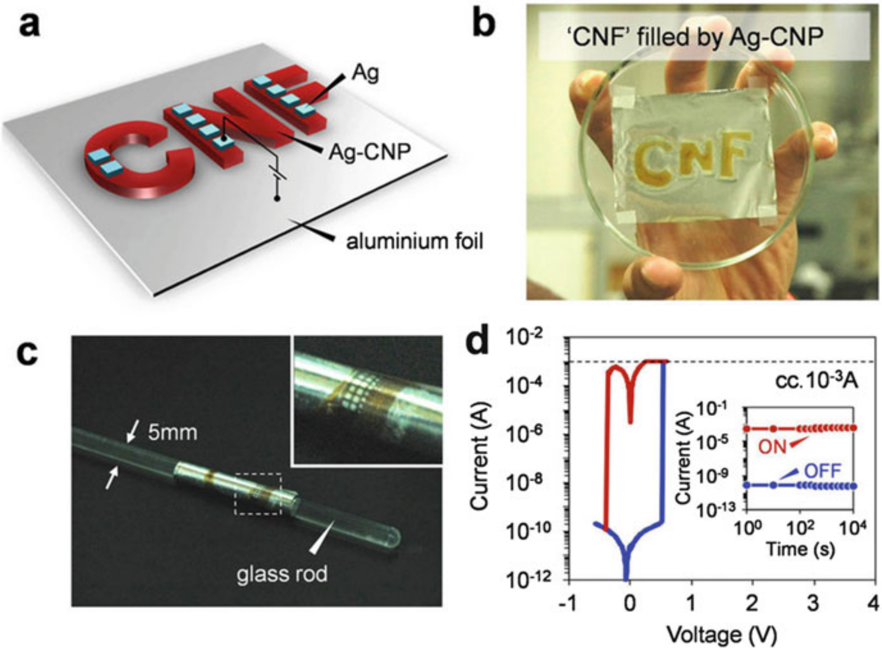

Fig. 12 Cellulose nanopaper as an ultra-flexible nonvolatile memory: (**a**) Schematic illustration of flexible nonvolatile memory based on Ag-decorated CNFs paper. (**b**) Photograph of the Ag-decorated CNFs paper on Alfoil. (**c**) Ag-decorated CNFs paper device wrapped around the glass rod with a diameter of 5 mm. The magnified image near the device stack is shown in the right-upper insert. (**d**) I-V characteristics of the Ag-decorated CNFs paper device on Al-foil when the Ag-decorated CNFs paper was platen again. The inset shows data retention taken at 0.1 V in the condition of room temperature and atmospheric pressure [66]. (Copyright 2014, Nature)

Water Treatment and Air Filtration

While the rapid development of industry increases human living standard, it also causes serious environmental pollution such as air pollution and water contamination. According to the characteristics of water contaminants, it can be divided into three categories: organic pollution, ionic pollution, and solid particle pollution. Air pollutants are mainly consisting of particles and poisonous gas. Cellulose nanofibers as renewable, degradation, and green materials are promising materials that can be used in industrial filtration without any by-product pollutants, due to their high surface to volume ratio and high strength.

Today's energy industry is largely dependent on petroleum-based oils. Because of the frequent oil spill and increasing oil pollution in industrial wastewater, oil and water emulsion separation has become a worldwide urgent need. Crude oil needs to be separated from water before using. Nevertheless, it is a big challenge on efficient oil-water separation technology due to the high stability and viscosity of water/oil emulsions. Because of the aggregation of surfactants and the formation of molecular networks, surfactants on the water/oil interface are believed to produce stable water/oil emulsions.

Rohrbach et al. [67] have reported that nanocellulose-based filters can improve current membrane technology by increasing efficiency and reduce clogging with environmentally friendly, renewable cellulose material. It is shown that there is a filter layer with a layer of CNF hydrogel by an impregnation and drying process using hydrophilic behavior of hydrogels. It creates a hydration layer by the filter absorbing water to saturation. The three-phase interface layer of hydrogel, water, and oil improves the properties of oil dispersion. The significant difference between the surface energy of oil and water makes it impossible for oil to penetrate the filter. If there is no coating, both oil and water can penetrate filter. The moisturizing coating produces hydrophobic hydrophilic behavior while retaining the absence of conventional functional groups. Hydrophilic/hydrophobic properties are the unique creation of cellulose with different properties and structures in macro scale and without introducing any harmful chemicals to scale.

Ferric hexacyanoferrate, also known as Prussian blue (PB), shows excellent properties for selective capture of radioactive cesium ions; however, it has not been used successfully in trapping open-field radioactive cesium (Cs) decontamination applications due to its high tendency to form stable colloids in water. Vipin et al. [55] reported that CNFs backboned PB is highly tolerant to water. This unique water-insoluble property is highly desirable for radioactive Cs elimination. In consideration of the overcapacity in dealing with low levels of radioactive pollution, they used two spongiform adsorbents to absorb CNF/PB, by which they could control the amount of CNF/PB, depending on the radioactive contamination level. One of the sponge form materials is polyurethane (PUF) which is extensively employed in spongiform (Fig. 13a), and the other is polyvinyl alcohol (PVA) (Fig. 13b). Then, they observed the porosity of the CNF/PB/PVA spongiform by SEM as shown in Fig. 13d, e. There is a significant decline in combined radioactivity (Cs-134 and Cs-137) which from 27,146 Bq/Kg ($n = 5$) before decontamination to 14,816 Bq/Kg ($n = 5$) after restoration. The radiation cameras about changes in the amount of radiation are shown in Fig. 13h, i [55]

Multilayer filtration membranes inspired by nature filtration system have extensively attracted many researchers' attention because of their unique properties such as high-water throughput, high filter efficiency, and low-pressure drop. However, the design of multilayer membrane structures in nanoscale is still a big challenge. Mushtaq et al. [68] designed hybrid nanocomposite materials composed of gold nanoparticles and a cellulose acetate membrane to remove radioactive iodine under continuous in-flow conditions by a vacuum filter system. Several composites with various concentrations of nanoparticles have been made, and the nanomaterials were incorporated stably on the cellulose nanofibrils.

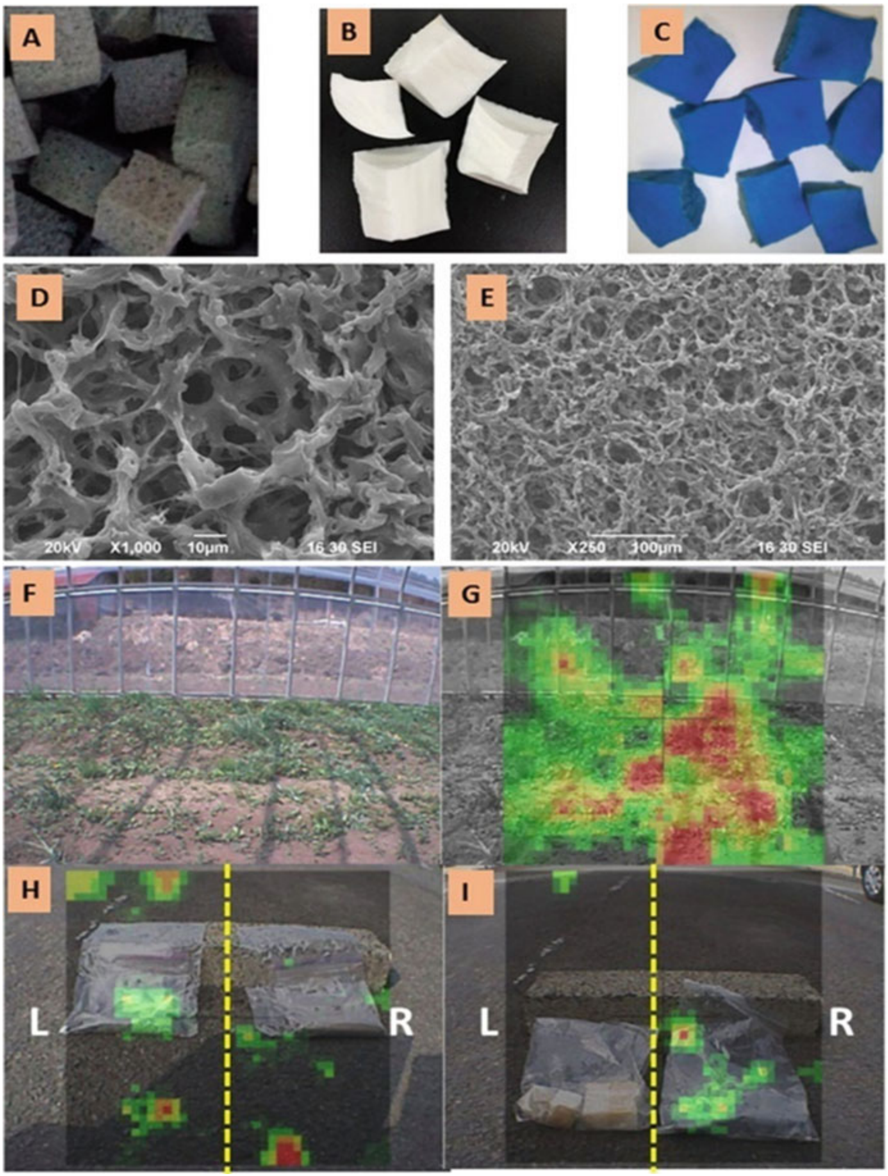

Fig. 13 Cellulose nanofiber (CNF) backboned Prussian blue (PB) nanoparticles as powerful adsorbents for the selective elimination of radioactive cesium (Cs). (**a**) CNF/PB nanoparticles/polyurethane (PUF) sponge, (**b**) Blank polyvinyl alcohol (PVA) sponge, (**c**) CNF/PB/PVA sponge; (**d**, **e**) SEM images of sponges at different magnification. (**f**) The subject area needs to be decontaminated; (**g**) Radiation camera images of soil radiation before decontamination. (**h**) Radiation camera images of soil radiation after decontamination by polyurethane (PUF) sponge (l) and the CNF/PB/PUF sponge (R). (**i**) Radiation camera images of pure PUF sponge (L) and CNF/PB/PUF sponge (R) after adopting radioactive Cs [55]. (Copyright 2016, Nature)

Biomedical and Healthcare Applications

The biodegradability, mechanical strength, and biocompatibility make CNF an ideal candidate for a range of applications including drug delivery, biomedical applications, wound dressings, and tissue engineering scaffolds. The ability to modify cellulose surface properties has been exploited recently to produce cellulose surfaces with antimicrobial properties [69]. CNFs also can be used for bioimaging and biomedical materials, antimicrobial films (protective coatings), and components of electronic devices [28].

Coronary artery bypass surgery is one of the most common treatments for cardiovascular disease, which is the blood supply to the heart by proper vascular substitutes having a good mechanical strength (up to 880 mm Hg blasting pressure) and blood compatibility [70]. It is possible to develop nanocellulose especially bacterial cellulose (BC) as a potential replacement for small artificial materials (<4 mm) or large (greater than 6 mm) size of the vascular graft [71]. The first research organization to study and apply artificial vascular substitutes from BC is the team of Dieter Klemm (University Jena and Polymer Jena, Germany) [72]. It is shown in Fig. 14 that the application of BC as blood vessel replacement in some publications. A clinical product named bacterial synthetic fiber is described, which has high mechanical strength in the wet state, large water retention property, and low roughness on the inner tube surface. In recent years, various properties and biological evaluation of BC tubes as vascular substitutes have been studied, involving BC inducing biomaterials coagulation, cell adhesion, proliferation, survival and invasion, hemodynamic analysis, and evaluation of the microcirculation.

Cellulose-Based Catalytic Membranes

Metallic nanoparticles with unique properties of high surface-to-volume ratio and distinctive surface electronic properties are particularly attractive for the catalysis research community, and they show excellent catalytic performances in various chemical processes.

However, on account of the small size and high surface energy, their direct introduction in the reaction system readily results in aggregation, which dramatically lowers their catalytic activities. In order to address issues mentioned above, making metallic nanoparticles immobilized or dispersed in various solid matrices like polymeric materials and metal oxides is one of the approaches. Among all the approaches, metallic nanoparticles immobilized in free-standing porous membranes show better catalytic performances. However, the preparative processes of such free-standing porous membranes are so complicated that the practical application is limited.

Cellulose nanofibers are abundant and low-cost materials, possessing intrinsic unique features of high porosity, good flexibility, robust mechanical strength, as well as chemical stability. Niu, Xu, Xiao, and Huang (2014) reported that cellulose-based catalytic membranes can be fabricated by deposition of Au-nitrophenols on Titania

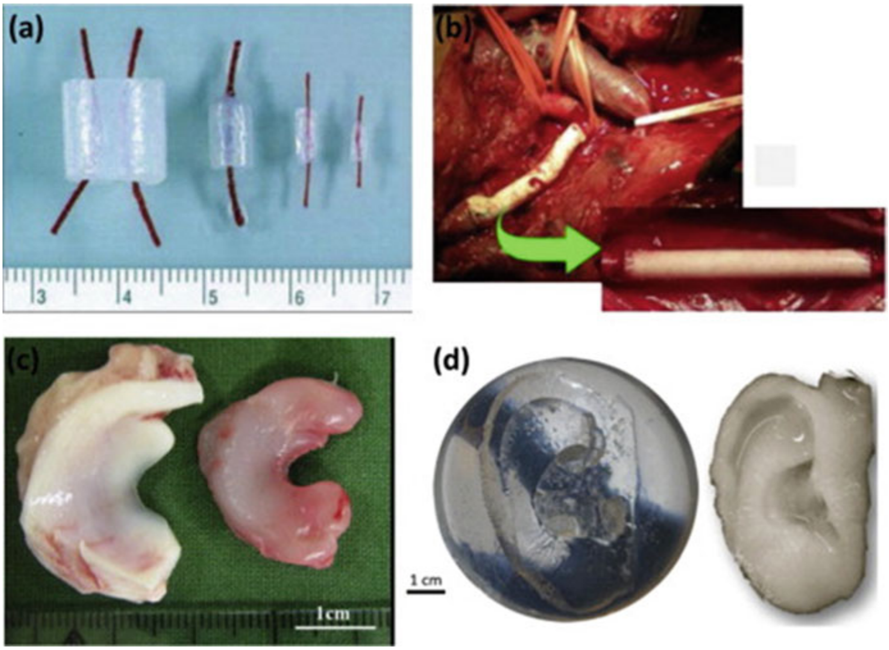

Fig. 14 Examples of substitutes from nanocellulose. (**a**) Bacterial cellulose (BC) with different dimensions. (**b**) Vascular prostheses made of CNF-polyurethane placed between the brachiocephalic trunk and the right common carotid artery in the male patient. (**c**) Comparison between pig meniscus (left) and BC hydrogel (right). (**d**) Negative silicone mold with the large-scale features of the external ears used to direct bacterial culture (left); and 3D BC implant prototypes in which effective cellulose content is 1% generated throughout the external ear shape obtained by 3 T magnetic resonance scanning technique (right) [72]. (Copyright 2014, Elsevier)

gel film pre-coated cellulose nanofibers of filter paper, which exhibited splendid catalytic activities towards the reduction of 4-nitrophenols to 4-aminophenol through a facile filtration process [73].

Conclusions and Future Outlooks

Cellulose nanofibers possess unique properties such as renewability and biodegradability with satisfactory mechanical and barrier properties when incorporated into other natural and synthetic polymers. The above discussion shows that cellulose nanofibers have an enormous potential in applications such as reinforcement of polymers, batteries, supercapacitors, air filtration, ultrafiltration membranes, sensing devices, fileable electronics, scaffolds, and so on. They will contribute to solve the environmental issues and create recycle-based and sustainable societies. Even though cellulose nanofibers have exhibited great advantages over conventional media in many applications, there remain a large number of challenges. The challenges comprise the mass production schemes of high-quality cellulose nanofibers and nanofiber-based composites. Besides, further research of a lot of other potential applications of cellulose nanofibers is also needed. Without question, cellulose nanofibers will be further explored for many different emerging applications because of their unique properties.

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