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REVIEW PAPER

Cellulose-based fibrous materials for self-powered wearable pressure sensor: a mini review

Miaomiao Zhu · Jichao Zhang · Wenxuan Xu · Ranhua Xiong · Chaobo Huang

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Abstract With the rapid development in wearable pressure sensors, self-powered pressure sensor based on piezoelectric and triboelectric effect have recently attracted great attention to overcome the limitation of conventional hard power sources. In consideration of sustainable development, environmentally friendly and biosafety, cellulose fibrous materials with good biocompatibility and biodegradability are becoming a promising versatile platform for designing and manufacturing self-powered pressure sensor. However, poor hydrophobicity, weak polarity, and insufficient functionalization on cellulose surface partly restricts the development of highly sensitive sensors to a certain extent. Much work is devoted to solving these problems. This minireview provides an overview of cellulose fibrous materials based piezoelectric and triboelectric self-powered pressure sensor. Following a brief introduction to the significance of the cellulose fibrous materials based self-powered pressure sensor, the self-powered sensing mechanism and cellulose based fibrous piezoelectric and triboelectric material for self-powered pressure sensor have been highlighted, including fabrication methods, sensing performance, and its applications. Furthermore, the challenges and future prospects of the cellulose fibrous materials based self-powered pressure sensors are also discussed. Finally, given that some advanced cellulose fibrous piezoelectric and triboelectric sensing materials exist for detecting external pressure, it is believed that these materials will make a significant contribution in intelligent wearable sensing field.

Keywords Cellulose-based fibrous materials · Self-powered pressure sensor · Piezoelectric and triboelectric · Sensing mechanism

M. Zhu · J. Zhang · W. Xu · R. Xiong · C. Huang (✉)
Jiangsu Co-Innovation Center of Efficient Processing and Utilization of Forest Resources, College of Chemical Engineering, Nanjing Forestry University, Nanjing 210037, China
e-mail: chaobo.huang@njfu.edu.cn

M. Zhu · C. Huang
Nanjing Forestry University, Nanjing 210037, China

J. Zhang
Jiangsu Engineering Research Center for Digital Textile Inkjet Printing, Key Laboratory of Eco-Textile, Jiangnan University, Ministry of Education, Wuxi 214122, Jiangsu, China

Introduction

As an emerging technology, wearable electronic devices have attracted wide attention due to their broad application prospects (Choi et al. 2016; Lee et al. 2015), such as human health monitoring, intelligent prosthesis, human-machine interface, smart robots (Dan et al. 2019; Kim et al. 2019; Zhu et al. 2020a). Pressure sensors play an essential role in the human motion detecting, physiological signals monitoring, and the cardiovascular disease prevention

(Meng et al. 2022a; Tan et al. 2020; Zhu et al. 2020b). Conventional metal-based or semiconductor pressure sensors are limited by their fragility and rigidity, which makes it difficult to be applied in flexible wearable devices (Wang et al. 2021a, b). Thus, generous work has been devoted to the development of flexible wearable pressure sensors, which are one of the most significant parts of wearable electronics (Lei et al. 2020; Meng et al. 2022b; Su et al. 2022).

Flexible pressure sensor should be conformably installed on the desired surface to ensure normal operation during deformations (Zhao et al. 2022a, b; Zhu et al. 2021; Su et al. 2021). One of the major obstacles faced by the flexible pressure sensors are dependence on traditional rigid power supply and bulky batteries, which has greatly limited the development of light-weight, conformable and flexible pressure sensor (Lai et al. 2019; Zhu et al. 2022a; Wei et al. 2021). At the same time, it is very inconvenient to periodically recharge or recycle the exhausted batteries in our daily life, largely affecting the long-term independent operation of the wearable pressure sensor. In this context, a promising way to solve this problem is the burgeoning self-powered sensing system, which involves the pressure sensing function and energy harvesting function into one self-sustainable electronics (Lei et al. 2021; Li et al. 2022; Su et al. 2021; Zhao et al. 2022a, b). In addition, using light weight, soft and breathable fibrous materials to construct wearable pressure sensors is another way to achieve high flexibility of electronic devices (Lin et al. 2022; Zeng et al. 2014; Zhu et al. 2022b). Therefore, the development of fiber-based electronics is a compelling approach toward wearable and flexible pressure sensor to maintain wearing comfort (Choi et al. 2016; Lee et al. 2015; Zeng et al. 2014; Liu et al. 2022a, b).

Being an inexpensive, renewable, abundant, biodegradable and versatile material (Wang et al. 2021a, b), cellulose can be widely used to manufacture "green" fibrous materials. Due to the multiple merits of cellulose-based fibrous materials, such as good mechanical performance, appealing piezoelectricity and dielectricity (Fan et al. 2020; Zhao et al. 2021), which is frequently used as a sensor material, substrate and electrode for flexible wearable pressure sensor. This mini review aims at providing the current state-of the-art of cellulose-based fibrous sensor materials for flexible self-powered wearable pressure sensor (Fig. 1). First, the characteristic of cellulose materials and cellulose-based fibrous materials are summarized. Then the self-powered pressure sensing mechanism are introduced. Meanwhile, by analyzing the advantage of cellulose-based fibrous materials, the research progress of cellulose-based fibrous materials in flexible wearable pressure sensor are highlight. Finally, we discuss the current research status and scientific technological challenges coupled with future outlooks for developing cellulose-based fibrous pressure sensor materials for wearable electronics.

Cellulose fibrous materials

To date, cellulose is the most abundant and typical natural fibrous biomaterials and widely concerned in the wearable sensing field (Nie et al. 2020; Pan et al. 2018; Rol et al. 2019), which can be derived into different forms with varied physical morphologies and multifunctionalities. As shown in Fig. 2a, cellulose can be extracted from plants such as trees, cotton, bamboo, and agricultural crops. And it can also be bio-synthesized by some bacteria, algae, and tunicates, as displayed in Fig. 2b (Wang et al. 2021a, b). By removing lignin and hemicelluloses from plants through chemical pretreatments to obtain micro or nano-sized cellulose fibers. Cellulose microfibers (CMFs) and cellulose nanofibrils (CNF) are a bundle of stretched cellulose nanofibers (Miyashiro et al. 2020). Cellulose structure consists of linear β-1,4-linked β-D-glucose rings, and has abundant hydroxyl (–OH) active groups in equatorial positions that can form inter and intramolecular hydrogen bonds within the chain and to neighboring chains (Bethke et al. 2018; Ummartyotin and Manuspiya 2015). These strong hydrogen-bond networks resulting in additional tensile strength. In addition, within the cellulose fibers, there are both crystalline regions with highly ordered macromolecular chains and amorphous regions with disordered macromolecular chains (Fig. 2c) (Zhao et al. 2021). The structure of cellulose relies on the pretreatment approaches and raw materials, which be further processed into various morphologies, such as cellulose nanofibers, cellulose nanocrystals, and cellulose nanosheets (Liu et al. 2022a, b).

In addition to the above nanocellulose, cellulose derivatives including cellulose esters, cellulose ethers and others are one of the main members of cellulose-based materials (Zhang et al. 2021a, b, c). To fully unlock the potential of cellulose-based materials, chemically functionalized cellulose is required, and the introduction of desired functional groups can be achieved by either physical surface interactions or chemical bonding. Cellulose has good chemical reactivity (oxidation, sulfonation, carboxylation, acetylation, silane treatment, polymer grafting, etc.) because of the abundant hydroxy group, which is conducive to the introduction of specific functional groups (Zhang et al. 2021a, b, c). Figure 2d shows common cellulose-based derivatives and their properties (Zhang et al. 2021a, b, c). For example, carboxymethyl cellulose (CMC) is fabricated by alkali reaction of alkali with cellulose, and then etherification reaction with monochloroacetic acid, which usually in the form of sodium salt (sodium carboxymethyl cellulose). As another of the cellulose derivatives, hydroxyethyl cellulose (HEC) is a kind of cellulose ether which prepared by the polymerization reaction between cellulose and ethylene oxide or chlorinated polyethylene during alkaline conditions. Carboxymethyl cellulose, hydroxyethyl cellulose and other functionalized cellulose are the common cellulose derivatives which can be prepared into fibrous materials and widely applied in flexible sensors (Zhang et al. 2021a, b, c).

Self-powered pressure sensing mechanism

With the development of the flexible electronics, there is an increasing need for self-powered sensor system (Aaryashree et al. 2021). Piezoelectric nanogenerators (PENGs) and triboelectric nanogenerators (TENGs) based pressure sensors can directly convert mechanical signals into electrical signals, which also is a kind of burgeoning power generation terminal and have attracted considerable attention (Huo et al. 2022; Pan and Lee 2021). Given the continuous development of PENGs and TENGs based self-powered pressure sensors, which have great application prospects for human motion monitoring, biomedicine, energy harvesting, smart wearable devices, and environmental detection (Zhao et al. 2022a, b).

Piezoelectric pressure sensing mechanism

The PENGs based self-powered pressure sensing mechanism directly depends on piezoelectric effect (Annamalai et al. 2021), which is based on the orientation of dipoles of piezoelectric materials under the mechanical stress (Hosseini et al. 2020). During the sensing process, an external force applied on the asymmetric direction of the dielectric material is accompanied by the generation of a pair of polarized charges on the opposite surface (Peng et al. 2022; Yi et al. 2022). When the external applied force is removed, the piezoelectric material recovers to its initial state. The schematic diagram of detailed piezoelectric pressure sensing process is displayed in Fig. 3a (Chen et al. 2022), which includes four states, i.e., the original state (Fig. 3a-I), the pressing state (Fig. 3a-II), the equilibrium state (Fig. 3a-III), and the releasing state (Fig. 3a-IV) (Huo et al. 2022; Veeralingam et al. 2022).

First, in the original state, in the absence of external force applied to the piezoelectric pressure sensor, the opposite induced charges on the electrodes and the stored charges on the piezoelectric materials are in electrostatic equilibrium. Therefore, there is no electrical voltage or current signals in the circuit (Fig. 3a-I) (Guan et al. 2022; Lee and Yoo 2021). When a force is applied on the piezoelectric pressure sensor, the total thickness of the piezoelectric materials decreases. The total spontaneous polarization degree increases, and the charges are induced significantly, which leads to a potential difference between

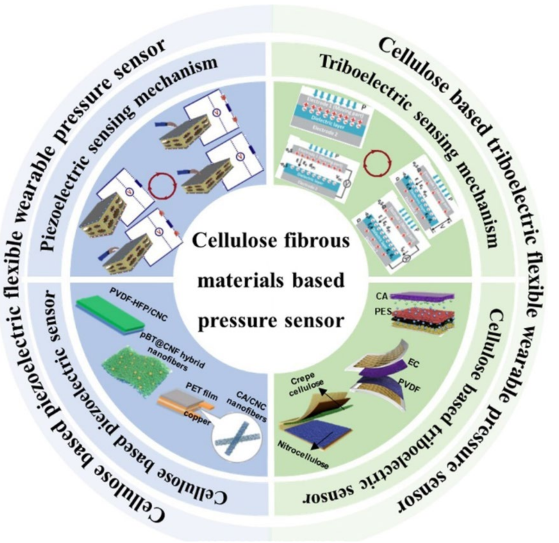


Fig. 1 Cellulose based fibrous materials for self-powered flexible wearable pressure sensor

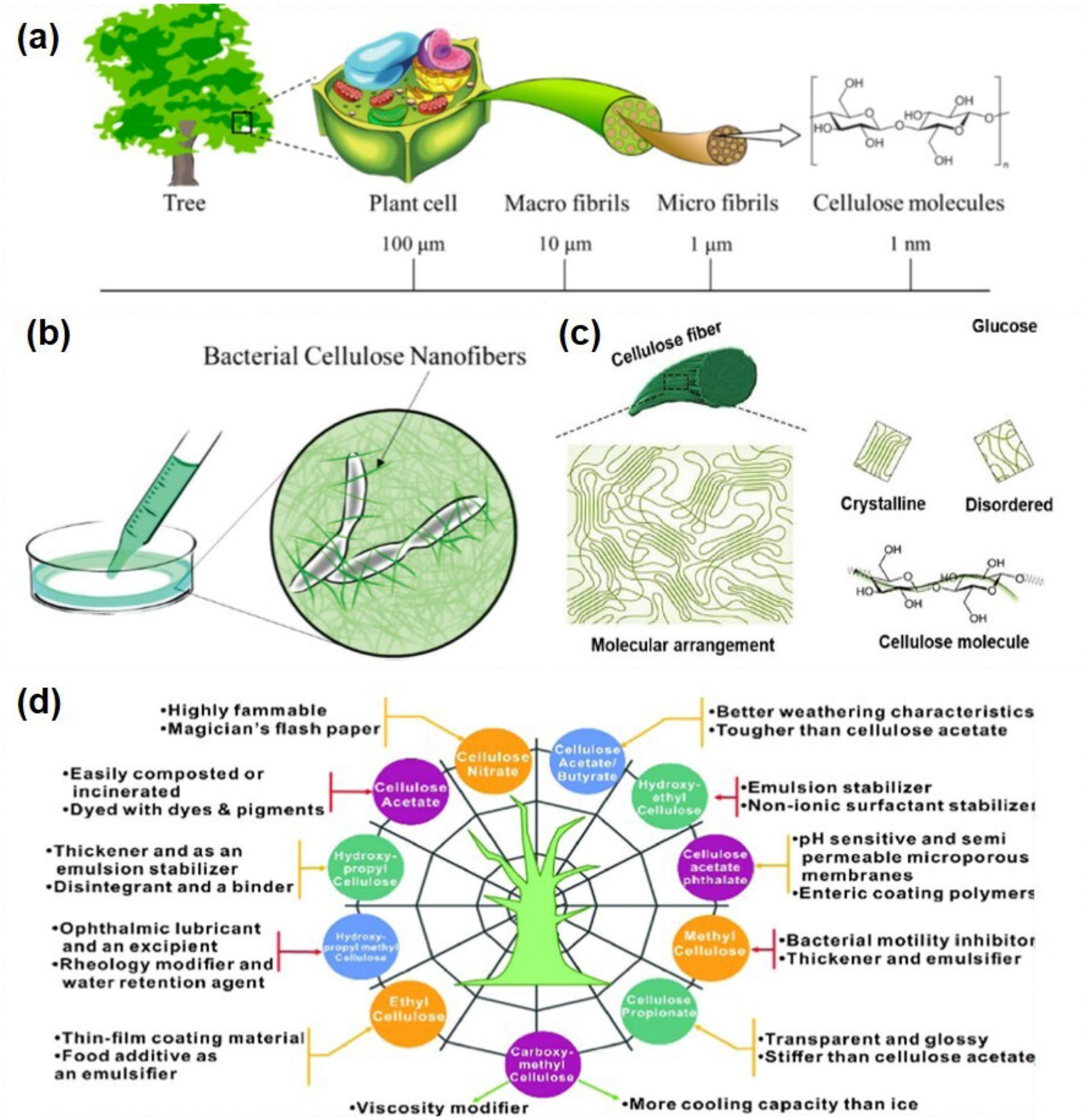


Fig. 2 **a** Hierarchical structure ranging from meters to nanometers of cellulose contained in trees. Reproduced with permission from (Miyashiro et al. 2020). Copyright 2020, MDPI; **b** Bacterial cellulose nanofibers by bacterial culture. Reproduced with permission from (Miyashiro et al. 2020). Copyright 2020, MDPI; **c** Crystalline and disordered regions of the cellulose fiber. Reproduced with permission from (Zhao et al. 2021). Copyright 2020, WILEY-VCH; **d** The classification and fabrication of cellulose derivatives. Reproduced with permission from (Zhang et al. 2021a, b, c). Copyright 2021, Elsevier

electrodes. External free charges will flow from the upper electrode to lower electrode to nullify the electric potential generated, resulting in a positive instantaneous signal (Fig. 3a-II) (Wu et al. 2020; Zhou et al. 2020). In the compressed state, the thickness of the piezoelectric materials decreases to a minimum. The amount of transferred charge and induced charges reaches a maximum, establishing a new electrostatic

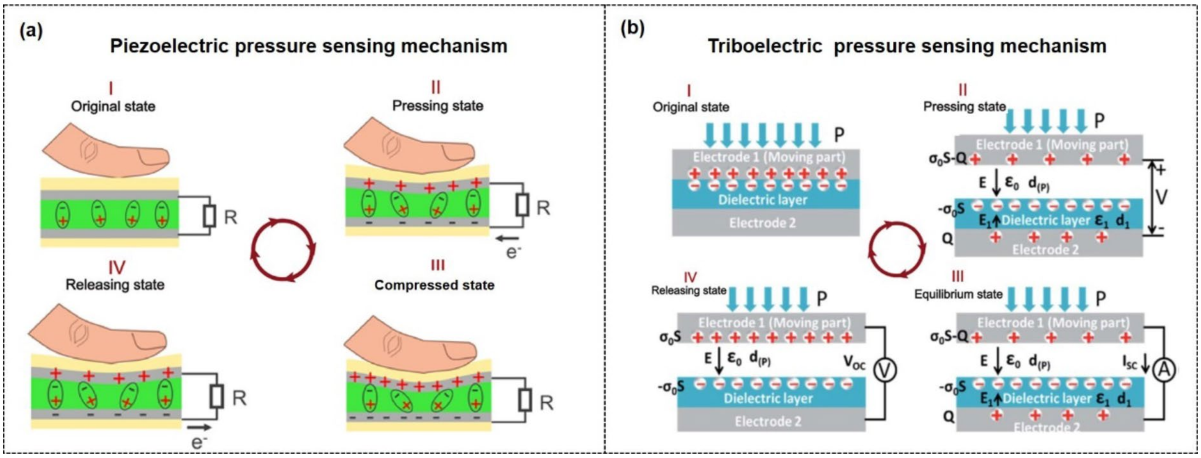


Fig. 3 **a** A schematic diagram of the sensing mechanism of a piezoelectric based pressure sensor. Reproduced with permission from (Chen et al. 2022). Copyright 2022, Springer Nature. **b** A schematic diagram of the sensing mechanism of a triboelectric based pressure sensor. Reproduced with permission from (Lei et al. 2021). Copyright 2021, The Royal Society of Chemistry

equilibrium (Fig. 3a-III) (Annamalai et al. 2021). Finally, in the releasing state, the piezoelectric materials shape will gradually recover, the accumulated charges flow back in the opposite direction due to the disappearance of electric potential, resulting in a negative signal. Therefore, an instantaneous opposite current signal in the external circuit is detected (Fig. 3a-IV) (Zhang et al. 2021a, b, c). When the external force is completely released, the piezoelectric materials return to its original state, completing a sensing cycle (Cao et al. 2021). During the action of periodic external force, the polarized piezoelectric materials will transform the applied external pressure into related current or voltage signals, which realize a self-powered pressure sensing (Tan et al. 2021; Zhu et al. 2020a).

Triboelectric pressure sensing mechanism

On the other hand, the sensing process of triboelectric pressure sensor depends on contact electrification and electrostatic induction coupling effect. Triboelectrification is a common phenomenon, which can generate opposite charges during the friction process of two different materials and be used to effectively transform the frictional contact movement into electrical signals. The fundamental sensing mechanism of TENG based pressure sensor is a sequential process between contact and separation to realize the charge transfer. Similar to the sensing process of piezoelectric based pressure sensor, as elucidated in Fig. 3b, it contains four states, i.e., the original state (Fig. 3b-I), the pressing state (Fig. 3b-II), the equilibrium state (Fig. 3b-III), and the releasing state (Fig. 3b-IV) (Kim et al. 2019; Lei et al. 2021).

When two different materials (positive layer and negative layer) of the TENG based pressure sensor are brought into complete contact state by an applied external force, fully balanced triboelectric charges pairs will be generated on the surface of these two materials due to the contact electrification. An equivalent physical model of the triboelectric based pressure sensor is shown in Fig. 3b-I (Lai et al. 2019). Once the applied force moves away from the triboelectric based pressure sensor, these contacted positive layer and negative layer will separate from each other, resulting an induced potential difference at the top and bottom electrodes due to the different electron affinities of the different dielectric layer, and thus generating an electrical current (Fig. 3b-II) (Zhang et al. 2021a, b, c). When the external force completely leaves the triboelectric based pressure sensor, an electrostatic equilibrium is achieved (Fig. 3b-III) (Wu et al. 2020). When the external force re-approaches the triboelectric based pressure sensor, the equivalent opposite triboelectric charges will be generated on each surface, causing a reversed electrical signal

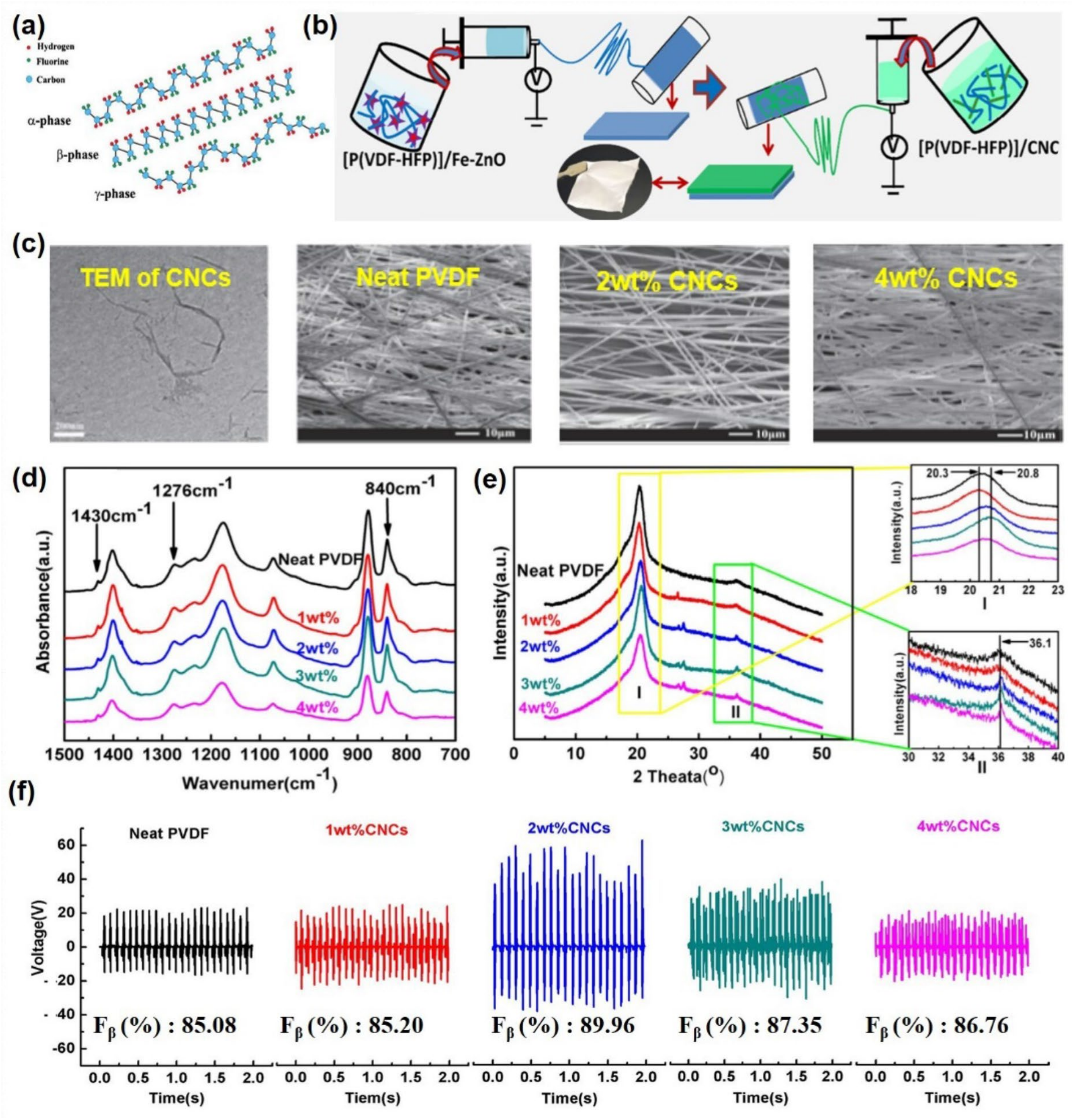


Fig. 4 **a** Diagrammatic sketch for the α, β, and γ-phases of PVDF molecular chains conformation. Reproduced with permission from (Azimi et al. 2020). Copyright 2020, WILEY-VCH. **b** Schematic representation of the fabrication of Fe-ZnO and CNC doped electrospun fiber membranes. Reproduced with permission from (Ponnamma et al. 2019). Copyright 2019, Elsevier. **c** TEM image of CNCs, and SEM images of electrospun neat PVDF, PVDF/CNCs-2 wt% and PVDF/CNCs-4 wt%. Reproduced with permission from (Fu et al. 2017). Copyright 2017, Elsevier. **d, e** FTIR and XRD analysis of PVDF based piezoelectric fibrous membranes with different content of CNCs. Reproduced with permission from (Fu et al. 2017). Copyright 2017, Elsevier. **f** The output voltages and fraction of β phases of PVDF based piezoelectric fibrous membranes with different content of CNCs. Reproduced with permission from (Fu et al. 2017). Copyright 2017, Elsevier

(Fig. 3b-IV) (Lee and Yoo 2021). Under the action of constantly different external forces, the electrical signals including open circuit voltage (V_{OC}) and the short circuit current (I_{SC}) can be recorded (Zhu et al. 2020b), and the magnitude of the electrical signal depends on the magnitude of the applied external force. At the same time, this sensing process of the sensors enable the external forces to be converted into electricity and does not require additional power supply, realizing self-powered sensing (Zhu et al. 2022a).

Cellulose fiber-based piezoelectric pressure sensor

To date, some synthetic ceramics and polymers and natural materials has been proved to have significant piezoelectric effect (Azimi et al. 2020; Bairagi et al. 2020; Scheffler and Poulin 2022). Among them, perovskite-structured ceramic materials such as lead titanate (PT), lead magnesium niobate (PMN), lead zirconate titanate ceramics (PZT), barium titanate ($BaTiO_3$), and alkaline niobates (K, Na, Li)NbO_3, are considered to possess the higher piezoelectric performance (Azimi et al. 2020; Li et al. 2019a, b; Zhu et al. 2020a, b). However, the hardness of these materials limits their application in the field of flexible wearable pressure sensor. In contrast to the inherent brittleness of the piezoelectric ceramics, piezoelectric polymers such as polyvinylidene fluoride (PVDF) and poly(vinylidene fluoride-trifluoroethylene) (P(VDF-TrFE)) (Tan et al. 2021; Ye et al. 2021) are more flexible, stretchable and spinnable (Mo et al. 2019; Tan et al. 2021). Therefore, piezoelectric polymers especially piezoelectric fibrous materials provide the possibility for flexible wearable piezoelectric pressure sensors (Li et al. 2019a, b; Ullrich et al. 2020; Ye et al. 2021). Cellulose based piezoelectric fiber materials have been extensively studied in recent years due to its renewability, biodegradability, and biocompatibility (Rajala et al. 2016). Cellulose has two main roles in piezoelectric materials. The first is that cellulose nanocrystals are added to piezoelectric fibers as nanofillers. The second is that cellulose itself is the main body for preparing piezoelectric fibrous materials (Ullrich et al. 2020; Zhang et al. 2018).

After polarization, the molecular chain configuration of PVDF and its derivatives changes from non-polarized α-phase transformation into polarized β and γ-phase phase, which with good piezoelectric effect, and the crystalline transformation of PVDF molecular chains is shown in Fig. 4a. The β-phase possesses the highest piezoelectric effect due to that all fluorine atoms are pointing to the same side of each unit (Li et al. 2019a, b). Electrospun process combines the electric poling and mechanical stretching, which provides a one-step straightforward approach to fabricate piezoelectric PVDF based fibrous materials (Azimi et al. 2020; Mishra et al. 2019). Moreover, a lot of work introduce some inorganic fillers (e.g., carbon nanotubes, graphene, and two-dimensional materials) into PVDF fiber as nucleating agents to induce PVDF molecular chain to convert from α-phase to β-phase, which improve the degree of polarization to a certain extent and further improve its piezoelectric properties (Mo et al. 2019; Scheffler and Poulin 2022). In addition to the above inorganic fillers, cellulose nanocrystals (CNCs) is a type of electroactive polymer which can also be used as a good nano nucleating agent to induce the conformational transformation of piezoelectric polymers.

Ponnamma et al. designs a polyvinylidene fluoride hexafluoropropylene (PVDF-HFP) piezoelectric electrospun nanofibers, which contains cellulose nanocrystals and the Fe-doped nano ZnO, as shown in Fig. 4b (Ponnamma et al. 2019). This hybrid nanocomposite membranes exhibit maximum output voltage of 12 V, which are respectively higher by 60 times compared to the pure PVDF-HFP fibers. The fabricated pressure sensor can detect the human body movements by attaching with the textile fabrics, such as hand tapping, finger tapping, elbow movements, which are in accordance wearable electronic textiles. In addition, Fu et al. prepared CNCs doped PVDF fibrous membranes by electrospinning, and the influence of CNCs on the piezoelectric properties of PVDF fibrous membranes was further studied and described in detail in their work (Fu et al. 2017). The TEM of CNCs and SEMs of the of different fibrous membranes are displayed in Fig. 4c. Compared with neat PVDF fibers, CNCs doped PVDF fibrous membranes possess higher content of β phase (Fig. 4d). The fraction of β phases (F_β) of PVDF/CNCs fibrous membranes could be calculated by the following equation

$$F_\beta = A_\beta/[(K_\beta/K_\alpha)A_\alpha + A_\beta] \tag{1}$$

where A_α and A_β are the absorbencies of the α and β phases at the wavelength of 763 cm^{-1} and 840 cm^{-1}, K_α and K_β are the absorption coefficients which are 6.1×10^4 cm^2 mol^{-1} and 7.7×10^4 cm^2 mol^{-1}, respectively. It is shown that all of the PVDF/CNCs

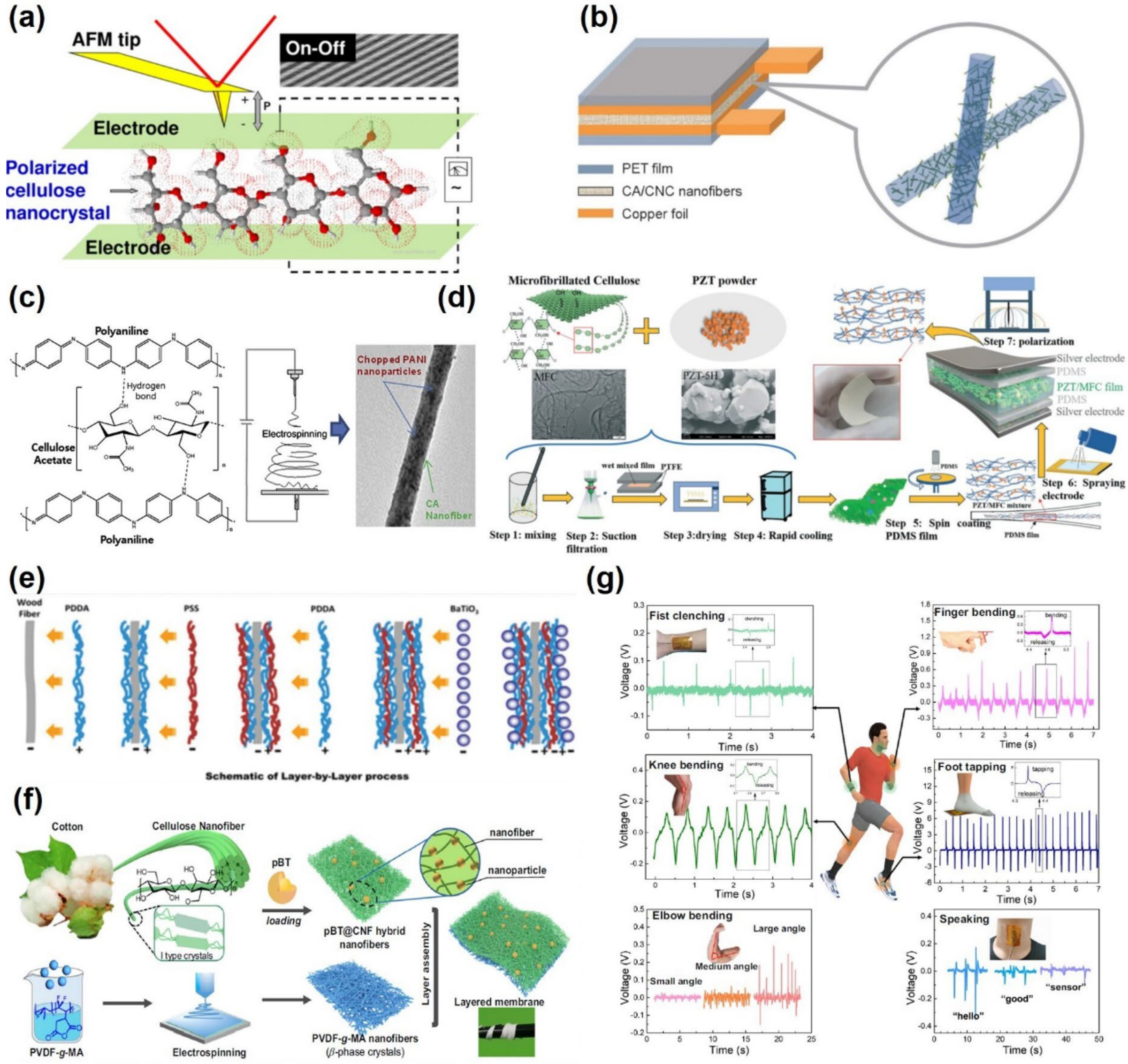


Fig. 5 **a** Schematic representation exhibiting the piezoelectric behavior of CNCs films. Reproduced with permission (Csoka et al. 2012). Copyright 2012, American Chemical Society. **b** Diagrammatic sketch of the CA/CNC nanofibers based piezoelectric device. Reproduced with permission (Sun et al. 2021). Copyright 2021, The Editorial Department of Chemical Research in Chinese Universities and Springer-Verlag GmbH. **c** Schematical illustrations of preparation process for electrospun PANI/CA piezoelectric fibrous membranes. Reproduced with permission (Hong et al. 2013). Copyright 2013, Elsevier. **d** The preparation process of the PZT/MFC piezoelectric composites membranes. Reproduced with permission (Guan et al. 2022). Copyright 2022, The Royal Society of Chemistry. **e** Schematic diagram of the fabrication process of piezoelectric hybrid paper. Reproduced with permission (Mahadeva et al. 2014). Copyright 2014, American Chemical Society. **f** The illustration of the preparation of flexible CPNF membranes. Reproduced with permission (Wang et al. 2022). Copyright 2022, Elsevier. **g** The sensing performance of the CNPF for human fist clenching, finger bending, keen bending, foot tapping, elbow bending, and speaking. Reproduced with permission (Wang et al. 2022). Copyright 2022, Elsevier

composite fibers contain higher Fβ than neat PVDF fibers. And the highest content of β phase is as high as 89.96%, which is achieved by adding 2 wt% CNCs to the PVDF fibrous membranes. As shown in XRD patterns (Fig. 4e), the PVDF fibrous membranes with 2 wt% and 3 wt% CNCs displayed prominent peaks

Table 1 The performance of piezoelectric pressure sensor based on different cellulose materials

Piezoelectric materials	Sensitivity	Durability (cycles)	Applications	References
PVDF-HFP/CNC	–	2500	Hand/elbow movement/shoulder movement sensing	Ponnamma et al. (2019)
CA/CNC	0.248 V N^{-1} (0–2 N)	–	Piezoelectric nanogenerators	Sun et al. (2021)
CA/PZT	27 pC N^{-1}	5000	Plantar sensors	Guan (2022)
pBT@CNF	–	8400	Human motion detecting	Wang et al. (2022)

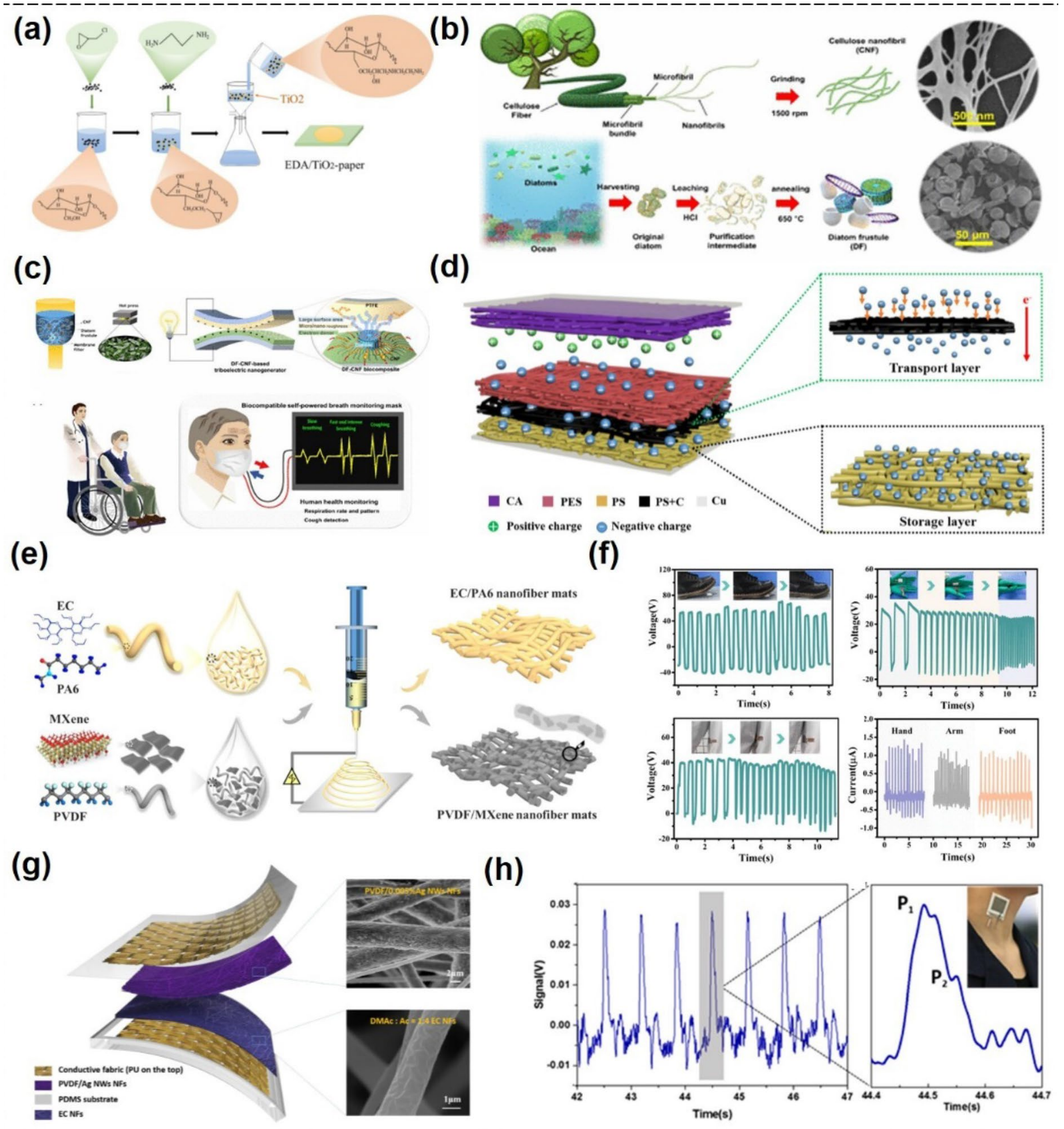
of the β phase at $2\theta = 20.8°$, 36.1° corresponding to the (110), (020) reflections of the phase. Compared with the neat PVDF fibrous membranes, the peak intensities of PVDF/CNCs fibrous membranes at $2\theta = 36.1°$ increased. At the same time, as shown in the Fig. 4f, when the content of CNCs is 2wt%, the CNCs doped PVDF fibrous membrane has the highest output voltage. The output voltage and β phases content of PVDF/CNCs fibrous composites increase along with the increase of CNCs from 1 wt% to 2 wt% but decrease with further increase of CNCs. Consequently, the addition of CNCs to PVDF based fibrous membranes can effectively improve its piezoelectric response.

Furthermore, the cellulose-based biopolymers such as wood, amylase, and starch have good shear piezoelectricity dipolar alignment and confined charges (Rajala et al. 2016), which are considered as a promising material for biomimetic pressure sensor due to some properties including lightweight, low cost, biodegradability, and larger output (Mishra et al. 2019). For example, as shown in Fig. 5a, Csoka et al. used electric field assisted shearing to assemble ultra-thin films of oriented cellulose nanocrystals on mica carriers. And the relationship between polarization gradient and strain mechanics was checked by monitoring the deflection of CNCs film with atomic force microscope operated in contact mode. The piezoelectric response of the aligned CNCs ultrathin films was attribute to the collective contribution of the asymmetric crystalline structure. The effective shear piezoelectric coefficient (d_{25}) of aligned CNCs ultrathin films is 2.1 Å/V, which is comparable to that of a piezoelectric metal oxide (Csoka et al. 2012). Currently, much work has been devoted to the research of cellulose fiber based piezoelectric materials and their application in human motion sensing.

For the piezoelectric fiber membranes, Sun et al. invented an all-cellulose fiber based piezoelectric materials through electrospinning. As illustrated in Fig. 5b, the addition of CNC of fabricated cellulose acetate/cellulose nanocrystal (CA/CNC) nanofibrous membranes enhanced the content of piezoelectric cellulose crystallization and mechanical properties (Sun et al. 2021), and the optimal piezoelectric output voltage is 1.2 V, which exhibited a linear relationship with external force, indicating the great potential in flexible wearable pressure sensors. In addition, Hong et al. developed a biopolymer actuator through electrospun cellulose acetate/polyaniline (CA/PANI) bio-piezoelectric membranes, as shown in Fig. 5c (Hong et al., 2013). The prepared CA/PANI extracellular nanoporous bio-nanofibers membranes possess a good biocompatibility and piezoelectricity, which is suitable for flexible wearable pressure sensor.

Cellulose has attracted widely concerned for piezoelectric pressure sensor. However, the relatively weak piezoelectricity of natural cellulose limits its applications. To achieve high piezoelectric response similar to that of inorganic ceramic materials and meet the demand of wearable devices for material flexibility at the same time, Guan et al. investigated a lead zirconate titanate powder doped microfibrillated cellulose (PZT/MFC) hybrid films for plantar pressure measurements, and the fabrication process and polarization process of this films is illustrated in Fig. 5d (Guan et al. 2022). The maximum piezoelectric coefficient of flexible PZT/MFC films is 31 pC N^{-1}, which can accurately measure the pressure distribution on the sole of the foot and analyze the human body motions including stride frequency, length and speed. This flexible PZT/MFC wearable plantar pressure sensor has broad application prospects in the field of medical rehabilitation training and sports injury detection.

In addition to the relatively toxic PZT, Mahadeva et al. successfully fabricated nanostructured $BaTiO_3$ surface functionalized wood cellulose piezoelectric



fibers. The preparation process is presented in Fig. 5e (Mahadeva et al. 2014), in detail, cellulose fibers was alternately immersed in poly (diallyl dimethylammonium chloride) (PDDA) and poly (sodium 4-styrenesulfonate) (PSS) solution, resulting in a charged surface on the wood fibers. Thus, due to the strong electrostatic interaction, the inorganic piezoelectric $BaTiO_3$ nanoparticles can be attached to the wood fibers. The largest piezoelectric coefficient (d_{33}) of functionalized wood cellulose piezoelectric paper is 4.8 ± 0.4 pC N^{-1}, which is a low-cost and promising substrate materials to construct piezoelectric pressure sensing devices. Wang et al. demonstrate a flexible piezoelectric membrane by integrating cotton cellulose nanofiber (CNF) and high-piezoelectricity maleic-anhydride-grafted polyvinylidene fluoride (PVDF-g-MA) nanofibers. At the same time, using polydopamine@ $BaTiO_3$ (pBT) nanoparticles as interlayer bridges and to synergistically

◂**Fig. 6** **a** Fabrication process of amino modified cellulose fibers. Reproduced with permission (Sheng et al. 2022). Copyright 2022, Springer. **b** Schematic illustration of the fabrication process of mass-producible diatom frustule and cellulose nanofibril from ocean and plants. Reproduced with permission (Rajabi-Abhari et al. 2021). Copyright 2020, American Chemical Society. **c** Illustration showing the preparation process, structure and human respiration sensing of DF/CNF based self-powered pressure sensor. Reproduced with permission (Rajabi-Abhari et al. 2021). Copyright 2020, American Chemical Society. **d** Schematic diagrams of charge transport and storage process in the cellulose based TENG. Reproduced with permission (Li et al. 2018). Copyright 2018, Elsevier. **e** Preparation process of the all-fiber-structured pressure sensor. Reproduced with permission (Huang et al. 2021). Copyright 2021, American Chemical Society. **f** Applications of the self-powered all-fiber-structured pressure sensor for human motion sensing, such as walking, clapping of hands, moving of arms and applying pressure sensor to different body parts. Reproduced with permission (Huang et al. 2021). Copyright 2021, American Chemical Society. **g** Schematic illustration and SEM of the pressure sensor textile. Reproduced with permission (Lou et al. 2020). Copyright 2020, American Chemical Society. **h** Real-time detection of the carotid artery pulse by the self-powered textile. Reproduced with permission (Lou et al. 2020). Copyright 2020, American Chemical Society

improve their piezoelectric properties, as shown in Fig. 5f. The fabricated cellulose based piezoelectric membranes exhibit excellent enhanced piezoelectricity with a maximum piezoelectric coefficient of 27.2 pC N^{-1}. The sensing performance of obtained self-powered cellulose based piezoelectric pressure sensor was further described, which can be used to detect human motions such as fist clenching, finger bending, keen bending, foot tapping, elbow bending, and speaking (Fig. 5g) (Wang et al. 2022). The performance including the cellulose based piezoelectric materials, sensitivity, durability, and applications of aforementioned cellulose based triboelectric pressure sensor are summarized in Table 1.

Cellulose fiber-based triboelectric pressure sensor

Self-powered triboelectric based electronics is a new-fashioned pressure sensor technology proposed by Wang in 2012 (Fan et al. 2012). Through the coupling effects of contact electrification and electrostatic induction, triboelectric pressure sensor can effectively transform external mechanical motion into electric signals generated between two different friction materials surfaces (McCarty and Whitesides 2008). Triboelectric based electronics are mainly consisting triboelectric materials, electrode materials, and substrate materials. Cellulose-based fibrous materials have found promising applications in flexible triboelectric based electronics because of its recyclable, low-cost, good processability, and sustainable features (Niu et al. 2021; Parandeh et al. 2019). Cellulose demonstrates great application potential in the triboelectric materials due to the easily modification characteristics (Zhang et al. 2019). Here, the cellulose material as the triboelectric sensing layer of the self-powered pressure sensor is introduced in detail.

Cellulose generally loses electrons and shows tribo-positivity when during the friction process (Diaz and Felix-Navarro 2004; Zhang et al. 2021a, b, c). Consequently, cellulose fibrous materials have been explored as positive materials in many works (Parandeh et al. 2019; Xia et al. 2018; Zhang et al. 2017). As shown in Fig. 6a, Sheng et al. developed an epoxy chloropropane and ethylenediamine grafted cellulose fibers to overcome the faultiness of the cellulose fibers surface weak polarity (Sheng et al. 2022). Then the epoxy chloropropane and ethylenediamine grafted cellulose fibers as a positive friction layer and a fluorinated ethylene propylene film were assembled together to form a cellulose based TENG. And the output power density of this cellulose based TENG reached 13.78 μW cm^{-2}, which has wide application prospects in wearable electronics. In addition to the chemical modification, some natural products also can be used as modified additive. For example, diatom frustules (DFs) are a kind of tribo-positive possessing 3D hierarchically porous structures and high surface area, which can improve the triboelectric effect of the positive friction layer (Losic et al. 2009; Xu et al. 2017). Abhari et al. demonstrate a diatom bio-silica as a biomaterial additive to enhance the triboelectric effect of cellulose nanofibril (CNF) tribo-positive layer (Fig. 6b), which are assembled with tribo-negative polytetrafluoroethylene (PTFE) film to fabricate a bio based self-powered breath monitoring masks (Fig. 6c) (Rajabi-Abhari et al. 2021). The power density of DF-CNF based sensor is 85.5 mW m^{-2}. Simultaneously, a cytotoxicity and biocompatibility research indicated that the DF-CNF tribo-positive layer was biologically safe.

In addition to nano cellulose, cellulose derivatives can also be prepared into fiber materials for pressure sensing. As illustrated in Fig. 6d, electrospun cellulose acetate (CA) nanofibrous membranes can

Table 2 The performance of triboelectric pressure sensor based on different tribo-positive cellulose materials

Tribo-positive materials	Sensitivity	Durability (cycles)	Applications	References
Amino modified cellulose fibers	–	10,000	Triboelectric nanogenerators	Zhang et al. (2022)
DF-CNF	–	5000	Breath Monitoring	Rajabi-Abhari (2022)
CA	–	6000	Knees bending	Li et al. (2018)
EC	0.665 V N^{-1} (2.3−14.4 N); 0.152 V N^{-1} (14.4−43.1 N)	14,000	Walking/clapping detecting	Huang et al. (2021)
EC	1.67 V kPa^{-1} (0−3 Pa); 0.20 V kPa^{-1} (3−32 kPa)	7200	Motion/Pulse Monitoring	Lou et al. (2020)

be chosen as the tribo-positive friction component due to its intrinsic chemical stability, biodegradability, and excellent positive triboelectric polarity (Li et al. 2018). Cellulose derivatives can co-electrospun with some tribo-positive polymer to enhance the triboelectric effect and mechanical properties. As demonstrated in Fig. 6e, Huang et al. report an all fiber structured flexible TENG based pressure sensor, which consist triboelectric positive electrospun ethyl cellulose/polyamide 6 (EC/PA 6) fibrous membrane and triboelectric negative electrospun PVDF/MXene fibrous membrane (Huang et al. 2021). The fabricated cellulose-based pressure sensor exhibits excellent output performance with a power density of 290 mW m^{-2}, which is capable of human movements self-powered monitoring such as walking, hands clapping, arms moving and different body parts moving (Fig. 6f), providing a promising application prospect in cellulose based flexible wearable electronics. In addition, the specific surface area of the ethyl cellulose fibrous membrane can be improved by adjusting the micro nano structure to further enhance its triboelectric properties. As illustrated in Fig. 6 g, hierarchically rough structured EC and PVDF electrospun fibrous membranes is serve as positive and negative triboelectric layer to manufacture self-powered pressure sensor (Lou et al. 2020). More importantly, the assembled self-powered pressure sensor can be attached on the carotid artery to detect the pulse signals, serving as a reliable approach to reflect human health condition. As shown in Fig. 6 h, the pulse wave consists of two peaks corresponding to the pulse pressure (P_1) and the reflected wave pressure (P_2). The radial artery augmentation index $AIx = P_2/P_1$ is 47.08%, which is within the normal range of a healthy woman under 35 years old. Thus, it can evaluate the healthy status of the human cardiovascular system and physiological diseases (Lou et al. 2020). Table 2 summarize the performance of these cellulose based triboelectric pressure sensor, which include the cellulose based tribo-positive materials, sensitivity, durability, and applications.

Cellulose based materials can also serve as tribo-negative material through functional modification, and further to assemble high performance bio-based wearable pressure sensor. chemical functionalization is employed to adjust the cellulose materials surface polarizability and hydrophobicity (Zhang et al. 2019). Nie et al. take advantage of triethoxy-1 H, 1 H, 2 H, 2 H-tridecafluoro-n-octylsilane (PFOTES) to modified cellulose nanofibrils surface, the preparation process is shown in Fig. 7a. Compared with pure cellulose materials, the fluorine-bearing silane chains grafted CNFs based materials have better surface polarity and hydrophobicity, and the triboelectric charge density is significantly enhanced. The degradable bio-TENGs based pressure sensor retains 70% of the original output voltage and current at 70% humidity, which shows good resistance to ambient humidity (Fig. 7b, c). In addition to the fluorination modification of cellulose, nitrocellulose can also have a capable of obtaining electrons in the process of contact electrification. Chen et al. use commercially available print paper as a substrate, while crepe cellulose paper and a nitrocellulose membrane are used as the tribo-positive and tribo-negative layers, respectively (Fig. 7d) (Chen et al. 2019). The significantly different tribo-polarities of crepe cellulose paper and nitrocellulose membrane yield this all-cellulose based pressure sensor with outstanding triboelectric performance, which can sensitively "feel" the pressing and releasing of external force in self-powered manner

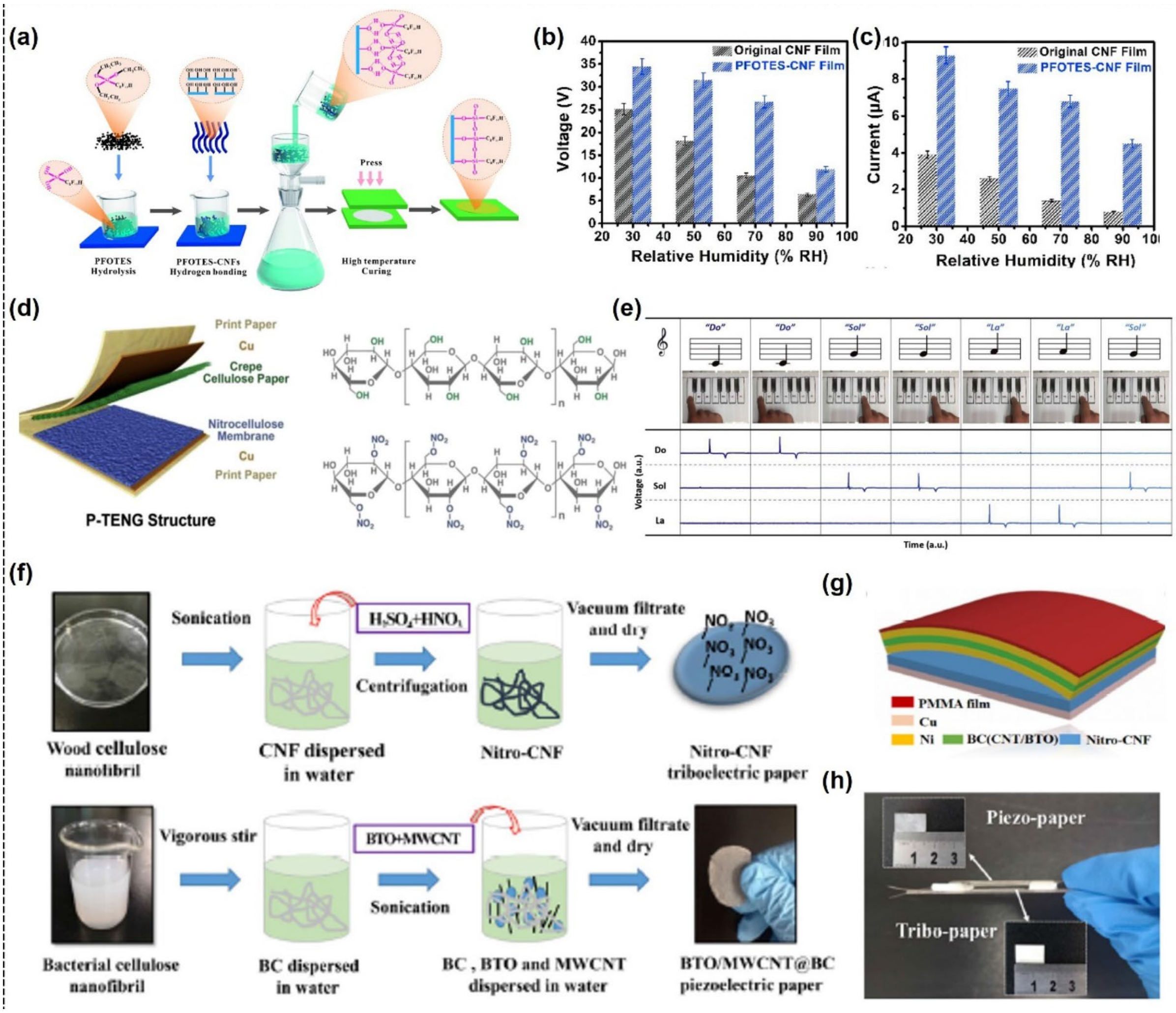


Fig. 7 **a** Fabrication of the PFOTES-CNF film. Reproduced with permission (Nie et al. 2021). Copyright 2021, Elsevier. **b, c** The voltage and current of the CNF based TENG as a function of humidity. Reproduced with permission (Nie et al. 2021). Copyright 2021, Elsevier. **d** The structure of cellulose based self-powered pressure sensor, and the chemical structure of cellulose and nitrocellulose, respectively. Reproduced with permission (Chen et al. 2019). Copyright 2019, Elsevier. **e** Demonstration of the paper piano playing a song of "Twinkle Twinkle Little Star". Reproduced with permission (Chen et al. 2019). Copyright 2019, Elsevier. **f** Schematic illustration of the fabrication process of cellulose nanofibril based triboelectric and piezoelectric materials. Reproduced with permission (Li et al. 2019a, b). Copyright 2019, Springer. **g, h** The schematic illustration and photo of the cellulose based hybrid pressure sensor. Reproduced with permission (Li et al. 2019a, b). Copyright 2019, Springer

Table 3 The performance of triboelectric pressure sensor based on different tribo-negative cellulose materials

Tribo-negative materials	Sensitivity	Durability (cycles)	Applications	References
PFOTES modified CNFs	–	10,000	Triboelectric nanogenerators	Nie et al. (2021)
Crepe cellulose	31.85 V N^{-1} (0.5–6 N)	10,000	human-machine interaction	Chen et al. (2019)
Nitro-CNF	8.276 V cm^2 N^{-1} (0.5–3 N cm^{-2})	6000	hybrid tribo/piezoelectric nanogenerator	Li et al. (2019a, b)

and possess great potential applications in human-machine interfaces. For example, as illustrated in Fig. 7e, the real-time communication between a computer and cellulose paper-based piano can be achieved by a keyboard based on this self-powered all-cellulose pressure sensor array. This work demonstrates a feasible approach to fabricate a sustainable and green wearable electronics.

Moreover, Li et al. also take advantage of nitrocellulose nanofibril paper as a negative triboelectric layer to manufacture cellulose-based hybrid pressure sensor with TENG and PENG component. The piezoelectric layer is prepared from $BaTiO_3$/MWCNT doped bacterial cellulose paper, as demonstrated Fig. 7f (Li et al. 2019a, b). Figure 7 g shows the structure diagram of the cellulose-based hybrid pressure sensor, and Fig. 7 h shows the flexibility of the nitrocellulose nanofibril negative triboelectric layer and $BaTiO_3$/MWCNT doped bacterial cellulose piezoelectric layer. The corresponding maximum power density of piezoelectric and triboelectric component is 1.21 and 10.6 μW cm^{-2}, respectively. The experimental results indicated that the output performance of $BaTiO_3$/MWCNT doped bacterial cellulose piezoelectric layer is around ten times higher than the $BaTiO_3$/polydimethylsiloxane layer. And the output performance of the nitrocellulose nanofibril negative triboelectric layer has considerable condition as fluorinated ethylene propylene, which provides new insights into the design of cellulose-based hybrid pressure sensor with high flexibility and outstanding performance. According to the characteristics of different types of cellulose based triboelectric pressure sensor (Table 3), high performance cellulose based triboelectric pressure sensor may be designed and fabricated to achieve applications in different fields.

Conclusion

Compared with traditional battery technology with limited lifetime, self-powered flexible wearable pressure sensor based on piezoelectric and triboelectric nanogenerators are much green and sustainable. Currently, the introduction of green materials is an essential way to cope with the over proliferation of non-degradable electronic waste. Cellulose have been proven to be an appropriate material to fabricate eco-friendly and flexible self-powered wearable sensors. And cellulose fibrous materials have attracted intensive interest in many researchers in order to incorporate them into self-powered wearable sensors due to their flexibility, lightweight, and easy to be functionalized. At present, a variety of cellulose fibrous based piezoelectric and triboelectric materials have been developed for the preparation of self-powered wearable pressure sensor.

In this feature review, the development of various cellulose based fibrous composite materials for flexible self-powered pressure sensors are described in detail, including the cellulose fibrous materials, piezoelectric pressure sensing mechanism, and triboelectric sensing mechanism. The development of various cellulose fibrous based piezoelectric and triboelectric materials for self-powered pressure sensor and its sensing properties was then reviewed in detail. Cellulose and its derivatives can be fabricated into piezoelectric and triboelectric fibrous composite materials, such as fibrous paper, fibrous membranes and textile. In addition to different cellulose fibrous based materials, various applications of these piezoelectric and triboelectric fibrous materials were also proposed, such as joint bending sensing, walking frequency detecting, and pulse and breath monitoring.

Although great progress has been witnessed in recent years for cellulose based piezoelectric and triboelectric fibrous materials, which is viewed as the promising approach for manufacturing self-powered wearable pressure sensor, there still remains some challenges need to be addressed in practical application. First, in practical applications, the poor mechanical properties of cellulose-based materials are a major drawback, which will cause error signals of sensing process and greatly limits their applications. Second, in the sensing fields, the inherent technical bottlenecks of sensing sensitivity, response time, and detection selectivity are still existed, which still cannot meet the needs of practical applications. In addition, the long-term durability of cellulose fibrous based piezoelectric and triboelectric materials in harsh environments should also be explored. Finally, miniaturized integrated, low cost, recyclable and multi-functional simultaneously detection properties for future practical applications need be carefully considered. With continuous and considerable efforts to overcome these challenges of cellulose fibrous materials based self-powered pressure sensors, which will hold great potential application for improving the human life quality, especially in the field of human motion detecting and healthcare monitoring.

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Declarations

Conflict of interest The authors declare that they have no conflict of interest.

Ethical approval This article is based on previously conducted studies and does not contain any new studies with human participants or animals performed by any of the authors.

Consent to participate Not applicable.

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