CHEMICAL REVIEWS

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Review

Functional Fiber Materials to Smart Fiber Devices

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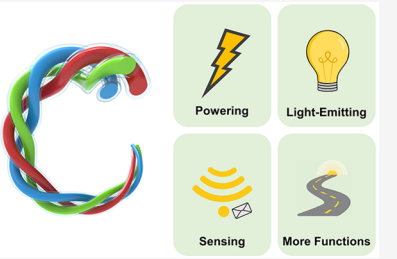
ABSTRACT: The development of fiber materials has accompanied the evolution of human civilization for centuries. Recent advances in materials science and chemistry offered fibers new applications with various functions, including energy harvesting, energy storing, displaying, health monitoring and treating, and computing. The unique one-dimensional shape of fiber devices endows them advantages to work as human-interfaced electronics due to the small size, lightweight, flexibility, and feasibility for integration into large-scale textile systems. In this review, we first present a discussion of the basics of fiber materials and the design principles of fiber devices, followed by a comprehensive analysis on recently developed fiber devices. Finally, we provide the current challenges facing this field and give an outlook on future research directions. With novel fiber devices and new applications continuing to be discovered after two decades of research, we envision that new fiber devices could have an important impact on our life in the near future.



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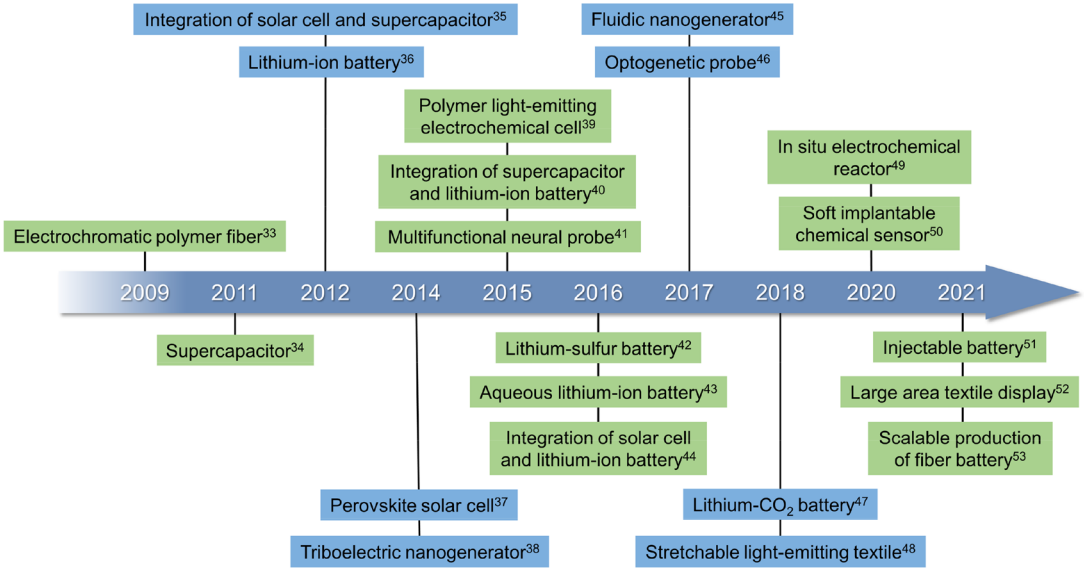

Figure 1. Timeline of key developments in fiber devices in the recent decade, categorized mainly according to their functions.[33−53]

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1. INTRODUCTION

The need for personalized electronics has been greatly increased during the past two decades.[1−4] These electronic devices are developing toward a general direction, including being miniaturized, multifunctional, self-powered, and user-friendly. Electronic devices with a spectrum of functions such as energy harvesting, energy storing, displaying, health caring, and computing have been discovered and designed as wearable or implantable systems for close contacts with our bodies to offer various applications.[5−10] Many pioneer prototypes of such electronic devices have been first developed in planar devices, including solar cells, electronic skin, and neural sensors.[11−15] Future devices need to be better integrated into our daily lives, e.g., as actual wearable textiles or long-term implantable devices.[16] As a result, new fiber materials and electronic devices have emerged to meet the need of next-generation electronics.[17−19]

Fiber represents one of the most used shapes in preparing materials. The evolution of human civilization is always accompanied by the development of new fiber materials.[17] Fibers with a large aspect ratio have been mainly used to weave clothes to keep people warm since ancient times. Importantly, they have been given new functions to work as electronic devices mainly in the recent decade.[20] The bending stiffness of a device is determined by the device dimension and geometry a fiber device is expected to demonstrate higher flexibility compared to its planar counterparts.[21] With proper design and modification, the unique one-dimensional shape also offers multiple functions and good tissue compatibility.[19,22−24] As a result, they have become particularly promising for applications as wearable and implantable devices during the last few years.[20,25−27]

No accurate definition has been made for fiber electronics/fiber devices yet. However, there are some criteria to meet for fiber devices. First, we could not call any one-dimensional device a fiber device. In other words, although fiber devices are one-dimensional, one-dimensional devices are not always fiber devices. If one-dimensional devices are too large in diameter, typically >1 mm, they may be better classified as cable-type devices.[28] Fiber devices generally range from submicron to hundreds of micrometers, so they have enough mechanical strength to be used in our daily life.[29] Second, besides the diameter requirement to bear external forces during applications, fiber devices should be long enough with aspect ratios of >1000, as they are typically woven into textiles.[30,31] Third, an electronic device is typically composed of two electrodes and an active material. Finally, fiber devices are

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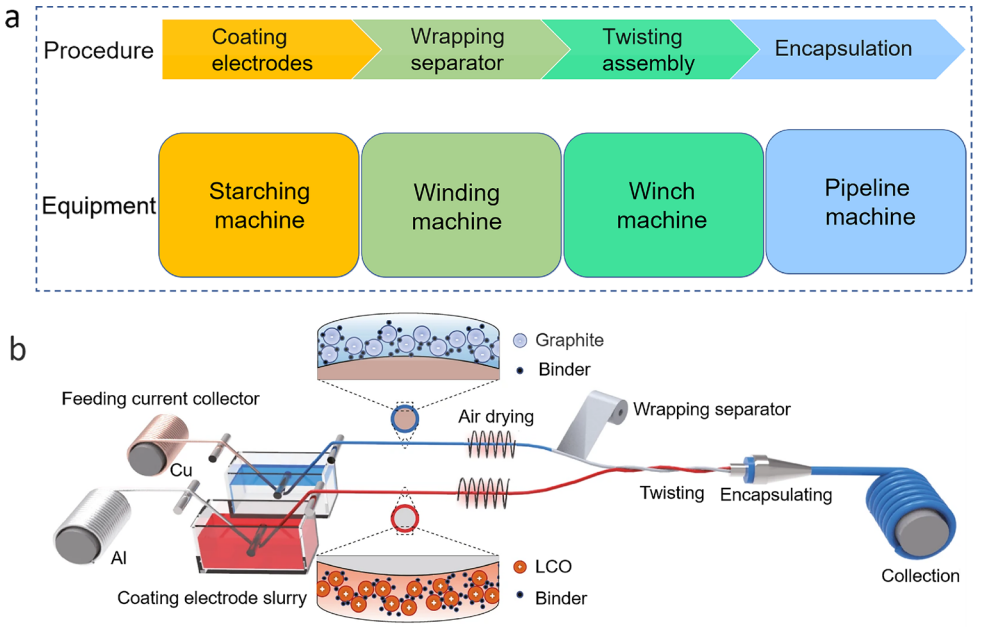

Figure 2. (a) Typical fabrication procedure and required equipment of sequential-deposition method. Four main steps are included, i.e., coating active material on the fiber substrate, winding a separator on the anode fiber, twisting the anode and cathode fibers, and sealing the fiber device. (b) Schematic illustration of industrial fabrication of a fiber battery via the sequential-deposition method. Reprinted with permission from ref 53. Copyright 2021 Springer Nature.

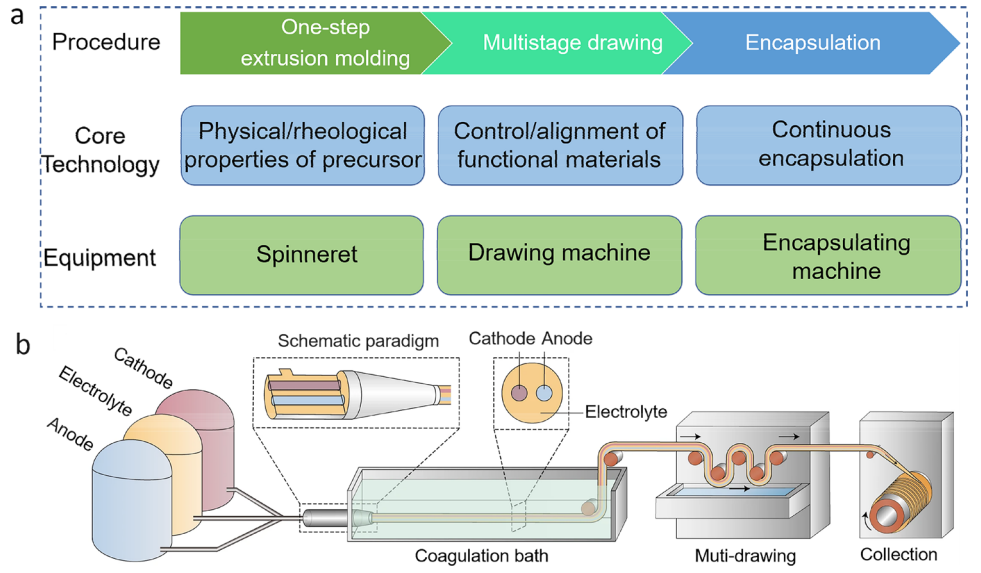

Figure 3. (a) Typical fabrication procedure, core technology, and equipment required for the one-step extrusion method. Three main steps are included, i.e., extruding all materials, drawing the fiber device, and sealing the fiber device. (b) Schematic illustration of continuous production of a fiber battery via the one-step extrusion method. Reprinted with permission from ref 58. Copyright 2022 Springer Nature.

generally flexible and washable because they may be scaled up for practical applications in the format of textiles.[32]

To this end, it remains unavailable to conclude when fiber electronics exactly started. At the early stage, fiber substrates are deposited with optoelectronic materials to enhance their properties. Fiber electronics started to boom mainly in the past decade, with combined efforts from scientists with different backgrounds across chemistry, materials science and engineering, energy science and technology, electrical engineering, and textile technology. Biomedical scientists also contributed to this direction starting from just recent few years. Figure 1 summarizes the main development of fiber devices in a timeline according to their functions.

It should be noted that the performances of fiber devices have been gradually improved through recent efforts. For example, the energy densities of early fiber lithium-ion batteries were <1 Wh/kg, making it difficult to realize practical applications, while their planar counterparts could provide energy densities >270 Wh/kg.[54] However, with the growing efforts to understand the charge-transfer mechanism, scientists were able to optimize material components and interfaces within the fiber batteries, leading to improved energy densities.[55] By using a highly conductive fiber electrode and

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fine control of the structure parameters of the fiber battery, which allows a better host of active materials and facilitates smooth charge transfer, the energy densities of fiber batteries could be improved to 85 Wh/kg.[53] On the other hand, almost all functions in conventional planar electronic devices have been achieved for fiber devices in the lab.[17] It is also good to see that some products of fiber devices have appeared at the market. To the best of our knowledge, light-emitting fibers and battery fibers already have been commercialized.[56,57]

Although it is possible to fabricate some fiber devices via a continuous process, the breakthroughs were mainly made just recently in 2021.[52,53,58,59] The continuous production of a fiber device is critical as it is closely rated to the low-cost, large-scale application. Previously, fiber devices had been typically produced by using protocols mimicking their planar counterparts, and they were limited at the lab scale. The fabrication requirements for fiber devices differ from planar devices. For instance, the modification of an intact active material layer on fiber electrodes is a key step that is easy for planar devices based on the current techniques.[60,61] However, completing this process on thin and flexible fiber electrodes is challenging or impossible with a high yield. There are a few notable attempts to fabricate fiber devices at a large scale.[62,63] For example, Figure 2a shows a typical sequential-deposition method for continuous production of fiber devices that involves four major steps, including the coating active material on a fiber substrate, winding the separator on an anode fiber, twisting the anode and cathode fibers, and sealing the fiber device. As a result, it is crucial to design corresponding critical facilities including coating, winding, twisting, and pipeline machines. Such a method can be used to fabricate a fiber battery at a large scale (Figure 2b).[53] The one-step extrusion method could also be used for large-scale fabrication. The precures including predissolved materials were extruded through a spinneret to form a two-phase solvent/nonsolvent mixture and then coagulate to form fibers. The typical procedure, core technology, and equipment required for this method are illustrated in Figure 3a. The key for a successful fabrication via the one-step extrusion method depends on the well-dispersed precures, proper excursion speed, and fine control of the coagulation rate (Figure 3b).[58]

Unquestionably, fiber materials are the basis of fiber devices, so this review starts with fiber materials. We then focus on configurations of fiber devices, fiber energy-harvesting devices, fiber energy-storing devices, fiber light-emitting devices, fiber sensors, and integrations of fiber devices, with an emphasis on the design principle. Finally, we try to present the remaining challenges and possible solutions of this booming direction of fiber electronics.

2. FIBER MATERIALS

The development of a new fiber material usually indicates the birth of a new class of fiber devices or brings new functions to fiber devices. Currently, fiber materials can be mainly cataloged into the following types: natural fibers, metal wires, polymer fibers, and carbon-based fibers, according to the discovery sequence by humans throughout history.[12,16,18,64] This section centers on the electrical, chemical, and mechanical properties of these fiber materials, as these properties are keys to design high-performance fiber devices.

2.1. Natural Fibers

Natural fiber materials including cotton, wool, linen, and silk, mainly from plants or animals, were the first used fiber materials in human history, and they have been used throughout human civilization.[65,66] For example, these natural fibers have been used to weave into clothes to keep people warm and away from harm. Natural fiber materials can be collected from animals or plants, and they are in fact a type of renewable and environmental friendly material. However, only recently have these materials been rediscovered as building components of fiber devices, including supercapacitors, batteries, and sensors, which will be detailed later.[18,67] Natural fibers typically demonstrate desirable mechanical properties for constructing human-interfaced devices. However, most natural fiber materials are not electrically conductive, thus requiring additives to enhance their electrical properties for the design of device.[68] On the other hand, using natural materials to make conductive fibers could offer a general and effective solution to this problem. For example, a composite fiber with a carbon nanotube (CNT) core and a silk sheath showed both high mechanical (tensile strength of 1.1×10^3 MPa) and electronic (electrical conductivity of 3.1×10^4 S/m) properties, which could be used as building blocks for textile electronics.[69] Due to the advantages of easy availability, biofriendliness, and renewability, many natural fiber-based devices have been developed.[19,70,71] For example, the natural fiber can be welded electrode yarns for knittable textile supercapacitors.[72,73] The surface roughness of the natural fiber materials can offer them high surface areas, thus allowing for high loading densities of active materials.[74]

One major advantage of natural fibers is their biocompatibility, primarily due to their natural origin.[75] These natural fiber-based electronics can be in close contact with our bodies.[76,77] To this end, a class of natural fiber-based devices, especially implantable devices, have been developed with high biocompatibility and controlled degradability.[78−80] Even with drawbacks such as low electrical conductivities, natural fibers are expected to play an essential role in the design of future fiber devices.

2.2. Metallic Wires

Metallic wires are necessary in our daily life, mainly as the connecting parts of many electric devices. They had been considered as the first choice in the early fabrication of fiber devices, as they were commercially available and also cheap.[81,82] Compared to the other fiber materials, the most distinguishing property of metallic wire lies in their high electrical conductivities.[83] They can be used as conductive substrates in various fiber devices, such as electrodes for solar cells and fiber sensors.[84,85] The mostly investigated metallic wires are made from copper, titanium, gold, platinum, and their alloys.[17] However, metallic wires are usually heavier than the other types of fibers, which is a problem for fiber devices because they are widely proposed for applications in portable and wearable electronics.

2.3. Polymer Fibers

Synthetic polymer fibers such as polyamide, polyvinyl chloride, and polyester have infiltrated modern society.[86] Polymer fibers usually have easy structure control, low density, and high degrees of flexibility.[87−89] However, synthetic polymer fibers are mainly used as clothes or textiles rather than electronic devices. One primary reason is that the electrical conductivities of such fibers are low.[90] Thus, they are not suitable to directly

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construct electronic devices, and incorporating conductive materials such as CNTs and metals can help improve their electrical conductivities for electronics.[91−93]

On the other hand, synthetic polymer fibers can be used as important building blocks for flexible or stretchable fiber devices; by winding an energy-storage fiber device on a stretchable synthetic polymer fiber, the resulting device can function as a wearable device.[94] Another interesting direction of synthetic polymer fibers is to design actuators, such as artificial muscles. Such a device demands the fiber to have better mechanical properties than superior electronic properties.[95,96]

2.4. Carbon-Based Fibers

Carbon-based fiber electrodes are now one of the most explored fiber electrodes for fiber devices, mainly because they simultaneously demonstrate remarkable electrical, electrochemical, mechanical, and thermal properties.[97,98] They have been generally prepared from CNTs, graphene, and carbonized organic fibers. The CNT fiber shows high specific surface area and aligned structure, and it is also highly flexible.[99] Therefore, the CNT fiber represents an ideal electrode for a fiber device, but it is difficult to synthesize and relatively more expensive.[100] The graphene fiber can be more easily synthesized with lower cost, but it is less flexible.[101] The carbon fiber is already available in the market. However, it has a solid structure with a low specific surface area, is rigid, and also displays poor electrical conductivity, so we may find some applications for them in fiber solar cells and limited applications in batteries.[85,102] They are further carefully analyzed and compared below, from a viewpoint of applications for fiber devices.

2.4.1. Carbon Nanotube Fibers. CNTs can be aligned and assembled into continuous fibers, the aligned structure has been found to effectively extend the remarkable properties of individual CNTs to macroscopic fibers.[99] For instance, a CNT fiber may show tensile strengths of several gigapascals and electrical conductivities on the level of 10^3 S/cm.[103−105] In addition, it is flexible with a bending stiffness close to that of soft tissues after further hierarchical and helical assembly.[50] In addition, active materials can be easily and efficiently incorporated into aligned CNTs with high loading densities. Therefore, CNT fibers have been mostly investigated to fabricate a variety of high-performance fiber devices.[97]

There are mainly three methods to synthesize CNT fibers. First, CNT powders can be dispersed in solvents, followed by a solution-extrusion process to produce fibers.[106] The shear force during preparation may align building CNTs along the axial direction.[107] It is relatively easy to scale up the production process, but the second phase is often required for this method, while bare CNT fibers should be typically used for the fabrication of fiber devices. Second, CNT arrays can be synthesized typically on silicon substrates by chemical vapor deposition, and CNTs then can be spun from the arrays to form continuous fibers. However, it remains difficult to synthesize CNT fibers by this dry-spinning process because the synthesis is time-consuming with a low yield.[108] Third, a modified floating catalyst chemical vapor deposition method could provide a high yield; however, the produced CNT fibers may contain a lot of residual catalysts inside, which is harmful for their applications in many fiber devices.[109]

The building of the CNT inside can be further modified with various functional groups on their surfaces and connected with a variety of organic molecules, so the CNT fibers may be tuned for many properties such as hydrophobic or hydrophilic, depending on the specific types of fiber devices. The fabrication, modification, and post-treatment of CNT fibers have been extensively illustrated in a few review articles.[110,111]

2.4.2. Graphene Fibers. Graphene has high tensile strength (∼130 GPa), elastic modulus (∼1 TPa), electrical conductivity (10^6 S/cm), carrier mobility (200 000 $cm^2/V·s$), and ampacity (1−2 GA/cm^2).[112] Graphene and its derivatives are promising for applications ranging from fundamental condensed matter physics research to materials science and drug delivery.[113] Graphene fibers integrate the excellent properties of individual graphene sheets into macroscopic ensembles that also possess the common advantages of fibers, such as high flexibility for textiles and a small footprint for implantable devices.[114] The fabrication of graphene fibers can be achieved via wet-spinning of graphene oxide liquid crystal phases, hydrothermal treatment, chemical vapor deposition (CVD), and spontaneous reduction and assembly strategy. The functions of graphene fibers can be tailored by the intercalation of functional components, such as adding stimulus-responsive materials and polymer composites for actuators or functional textiles.[101,115]

2.4.3. Carbon Fibers. Carbon fibers that usually show diameters of 5−10 μm are mainly composed of carbon atoms. The carbon fiber was first produced in the 1860s and used for light bulbs, but only recently have they been used for smart devices due to both high mechanical and electrical properties.[97,116] Carbon fibers have several advantages, including high stiffness, high tensile strength, low weight-to-strength ratio, and high chemical resistance.[117] The preferred orientation of the graphite crystallite structure along the carbon fiber axis offers high strength and modulus along the fiber axis. The tensile strength and modulus of a carbon fiber reach 7 and 900 GPa, respectively, and it is considered one of the strongest engineering materials.[118,119] Carbon fiber is usually composited with other materials to form a composite and is thus useful for aerospace, engineering, and sensor applications.[120,121]

2.4.4. Liquid Metal Fibers. Liquid metal fibers are newly emerging fiber materials with advantages of high electrical conductivities (10^4 S/m), good mechanical stretchability (>1000%), and superior flexibility of liquid metals.[122] Liquid metal fibers usually require a core−sheath structure with the liquid metal as the core and the polymer as the protective shell layer. The liquid metal fibers could be prepared by wet-spinning or melt-processing methods and have demonstrated promising applications in wearable self-powered sensors and smart fabrics.[123−126]

2.5. Fabrication of Fiber Devices

After obtaining fiber electrodes, the functionalization of the fiber electrode with active materials can be achieved via dip-coating or in situ chemical reactions. For the dip-coating strategy, active materials are first dispersed in solutions, followed by dipping the fiber electrodes in and withdrawing them from that solution at a particular speed. Later, the coating layer is evaporated to obtain a dry thin film of active materials. The dip-coating method is facile, low-cost, and compatible with a variety of active materials. The quality of the active-material layer can be influenced by the precursor density, viscosity, surface tension, immersion time, electrode-withdrawing speed, coating cycle, fiber surface property, and drying condition.[127] The other typical strategy to functionalize fiber

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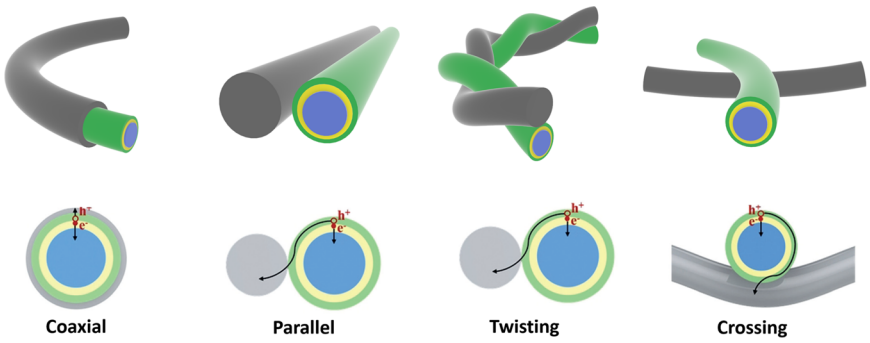

Figure 4. Routes for charge transfer in fiber devices with coaxial, parallel, twisting, and crossing configurations. Reproduced with permission from ref 20. Copyright 2019 Wiley-VCH.

electrodes is via in situ chemical reactions. For example, carbodiimide cross-linker chemistry was widely used for fiber electrodes to be functionalized with enzymes, while an in situ electrochemical reductive reaction can easily deposit the electrodes with a variety of functional nanoparticles.[50] The other fiber electrode modification technologies, including physical adsorption, thermal evaporation, and CVD thin-film deposition, are also widely used for fabricating fiber devices.[128] Note that 3D printing and thermal drawing have been further developed with one-step fabrication of functional fiber electrodes, eliminating the need for post-treatments of fiber electrodes.[129,130]

The assembly and encapsulation of fiber electrodes are sequentially followed to produce fiber devices. To obtain functional fiber devices, fiber electrodes are usually assembled with electrolytes by designing several configurations (including coaxial, parallel, twisted, and crossing). Later, an encapsulating layer is often required to ensure a stable environment for the electrode and electrolyte to work and to protect them from breaking and leaking to the surroundings. Such a process can be completed with the help of machineries such as spindle rotor, electric motor, or custom-designed industrial assembly line.[17,131]

3. CONFIGURATIONS AND WORKING MECHANISMS OF FIBER DEVICES

The charge-separation and -transport processes are indispensable for electrochemical or optical electronics.[132] To understand the working mechanism of a fiber device, it is crucial to know how the earlier-discussed processes occur in the fiber device. Taking a fiber battery or fuel cell as an example, efficient charge transport lowers the internal resistance of the fiber device and ensures higher output voltage and current. The one-dimensional shape offers a unique charge-separation and -transport route (Figure 4).[20] This section first presents a detailed analysis of various configurations of fiber devices and then discusses how the configuration affects charge separations and transports by understanding their working mechanisms.

3.1. Configurations of Fiber Devices

3.1.1. Coaxial Configuration. A coaxial configuration is simple and common for fiber devices. A coaxial-fiber device first uses a fiber electrode as the substrate, followed by coating with active or functional materials. Because of the close interface between fiber electrodes and active materials, the coaxial-fiber devices have several advantages such as rapid charge transports.[133] Maintaining a stable interface between the substrate electrode and the active material requires mechanical compliance within the fiber device, as each layer of the device should deform uniformly under external strain. A family of coaxial-fiber devices has been fabricated via extrusion, dip-coating, and 3D-printing methods.[134−137] The coaxial configuration is preferred for energy-harvesting/storing devices and biomedical sensors, which simultaneously require rapid mass transfer, charge separation, and charge transport.[133,138,139] It is not strange that the coaxial configuration first appeared under inspiration of typical planar devices. The challenges lie in the realization of an active layer and another electrode layer as they are required to be both thin and uniform on a highly curved fiber surface.

3.1.2. Twisting Configuration. The twisting configuration involves two fiber electrodes that are twisted together with a particular angle and helical pitch.[140] Compared to the coaxial configuration, the contact area in a twisting-fiber device is much lower, which may affect the charge-transfer process inside the fiber device. Meanwhile, the active materials should be modified on the two fiber electrodes in the twisting configuration.[17] This configuration lowers the difficulty of fabricating multiple-layer fiber devices with higher production speeds compared with the coaxial configuration.[141] For instance, it is challenging to sequentially deposit thin and uniform photoactive layers on a curved fiber substrate in a coaxial configuration in fiber solar cells. It may be more difficult to realize a continuous and repeatable production for scaling up the fabrication.[18] In contrast, we may much more easily twist two fiber electrodes with one coated with a photoactive layer into a fiber solar cell at a large scale.[85] The multiple contact points within the twisting configuration facilitates the mass transfer between the two fiber electrodes.[70] To this end, the twisting angle significantly impacts the device performance, as the closer contact provides better charge transfer between two electrodes, so it is important to reveal the optimal twisting angle for a specific type of fiber device.[142] A variety of fiber energy-harvesting/storing devices, such as solar cells, supercapacitors, and batteries, have been developed from the twisting configuration, as illustrated later in sections 4 and 5.

3.1.3. Parallel Configuration. The parallel configuration requires two fiber electrodes to be assembled in parallel. As a special organization of the two-electrode configuration, the parallel configuration has been widely adopted in fiber devices because it is simple and easy to fabricate. Many fiber devices, including supercapacitors, batteries, sensors, and light-emitting devices, can utilize this configuration.[143,144] The charge transfer typically needs an intermediary electrolyte, as the two fiber electrodes have no physical contacts. Compared with

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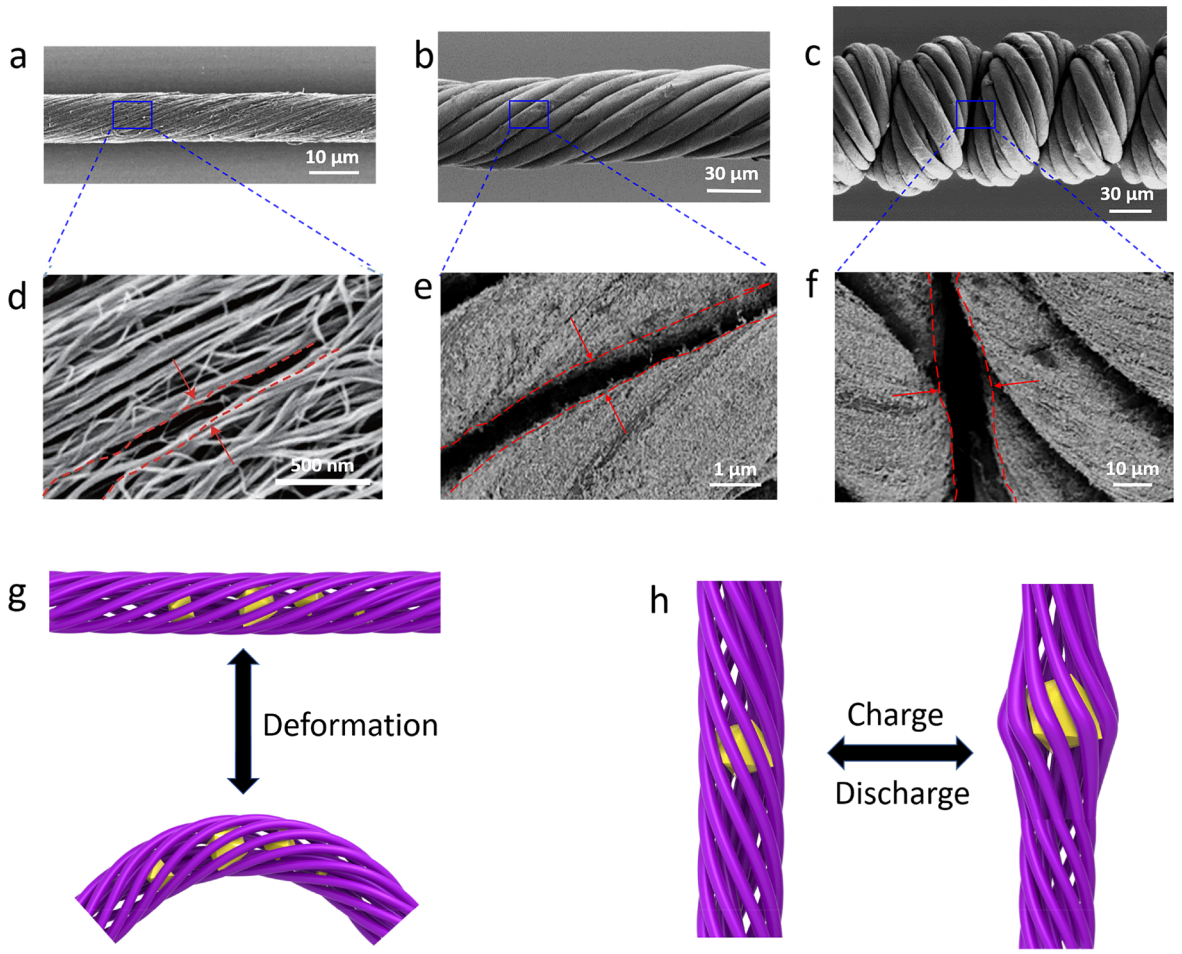

Figure 5. (a–f) Scanning electron microscope (SEM) images of a hierarchically assembled helical fiber electrode and the multiple scale gaps within and between primary fibers. Reprinted with permission from ref 150. Copyright 2015 Springer Nature. (g, h) Hierarchically assembled helical fiber electrode that can stably confine the active materials, providing a stable active material/electrode interface. Reprinted with permission from ref 151. Copyright 2020 Science China Press.

the typical twisting configuration where two fiber electrodes can be effectively stabilized with each other by mechanical forces, the two parallel fiber electrodes may easily contact to produce short circuits in liquid electrolytes. The use of semisolid or solid electrolytes with careful designs may solve this problem to certain degrees.[145,146] Because of the simplicity of the parallel configuration, the parallel fiber devices are considered to be favorable for large-scale fabrication with simple procedures and low requirements.

3.1.4. Crossing Configuration. It is easy to understand that a crossing configuration may be used for the situation where fiber devices are made into electronic textiles. Typically, two fiber electrodes are crossed with each other by a weaving method, and the contacting point of the two fiber electrodes contributes to the realization of the designed functions, such as emitting light.[52] Here the electroluminescent units have been formed only at the crossover points of two interlacing, electrically conducting fibers. Similarly, the contacting points may also serve as miniature solar cells if photovoltaic materials are used to replace the light-emitting materials.[147,148] Compared to the other configurations, it is generally applicable to optoelectronic devices in the fiber shape. The crossing configuration offers a high degree of controllability, and it is particularly promising for the production of optoelectronic textiles at a large scale.

3.2. Interfaces within Fiber Devices

Usually, two interfaces determine the properties of fiber devices during working. The first is the interface between the active material and the fiber electrode, which is crucial for transmitting physical or chemical changes from the surrounding environment to the electrode. Usually, a uniform coating layer of an active material on the electrode is required to construct this interface. However, due to the small size and high curvature, we have previously discussed that it is challenging to make uniform coating layers of active materials. Such an interface remains a challenge to build, particularly in the development of new fiber devices for which there are no accumulated research studies. In addition, the fiber devices may suffer from constant deformations, and the active-material layer may form cracks and be peeled off from the electrode.[141] For fiber batteries, the active materials may undergo significant volume changes (e.g., ~400% for silicon) during the charge/discharge process, also leading to unstable active material/electrode interfaces.[149] A solution to address this problem is to use a hierarchically assembled helical fiber electrode.[42] For example, by twisting multiwalled CNTs into primary fibers and then twisting the primary fibers together to assemble a hierarchically helical fiber electrode, the nanoscale gaps among the CNTs and micrometer-scale gaps formed among the primary fibers could provide anchor sites for active materials

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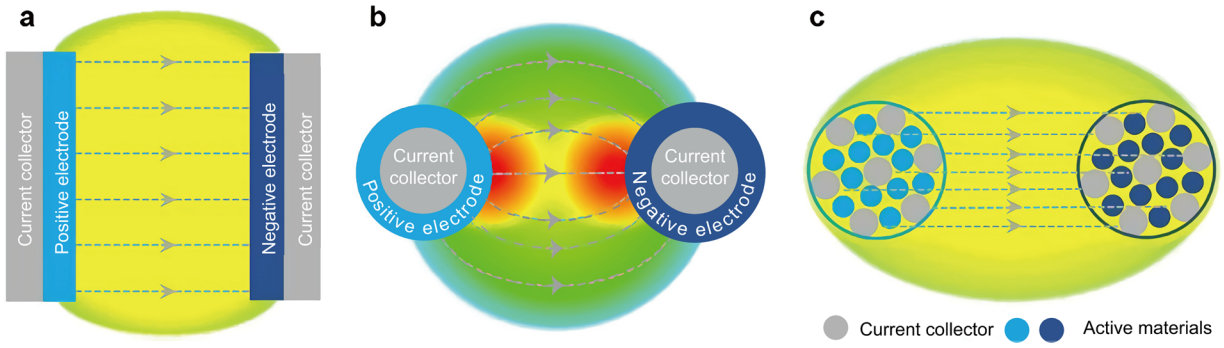

Figure 6. Electric field form in (a) a planar device, (b) fiber devices with two solid electrodes, and (c) fiber devices with two porous electrodes. Reprinted with permission from ref 17. Copyright 2020 Springer Nature.

(Figure 5a–f).[150] The gaps could confine the active materials and prevent them from detaching the electrode surface during deformations or volume expansions, hence providing stable active material/electrode surfaces (Figure 5gh).[151] The other solutions, including using soft, premodified electrodes or optimized fabrication processes, can also help in obtaining stable interfaces. For example, experimental results demonstrated that pretreated CNT electrodes could offer proper surface roughness and functional groups to anchor active materials via physical absorptions or chemical reactions for stable interfaces.[152]

The second interface is that between the active material and the surrounding environment. The successful infiltration of an electrolyte into the active material/electrode surface is crucial for the mass or signal exchange between the fiber device and the surrounding environment. Modifying fiber electrodes with functional materials might cause a change of the surface property of the fiber device, e.g., changing the surface property from hydrophilic to hydrophobic, resulting in unstable active material/electrolyte interfaces and subsequently sluggish mass transfers.[153] It should be noted that the one-dimensional shape of the fiber devices provides a 360° interface with the surrounding environment, making it favorable to receive chemical and physical stimuli from all directions, which should also be considered when designing a fiber device.[20]

3.3. Charge Transfer Inside Fiber Devices

Charge transfer is a key step for fiber devices. For example, an effective electron transfer indicates a lower inner resistance of the fiber device, so there are less charge losses while working with higher voltage or current outputs.[154−156] For biosensors, the electrical signals collected from biological recognition events are usually low, and the losses of signals decrease their responses and sensitivities.[157,158] Therefore, facilitating the charge transfer in the fiber device is vital for better device performances. Ideally, such a process can be optimized by improving the electrical conductivities of fiber electrodes via pretreatment of the electrodes with functional nanomaterials.

3.3.1. Electron Transfer. An electron is the primary charge carrier in fiber devices. The building materials of fiber electrodes are critical to electron transfer. Electron transfer in a metallic wire is straightforward, and electrons are delocalized from metal atoms and move in a particular direction to form a current.[159] Electron transfer in carbon-based fibers is more complicated and is determined by the structure of the fiber material.[160] For example, aligned CNT fiber electrodes have been proven to facilitate electron transfer even though the basal planes in the CNT are supposed to slow down the electron transfer.[161] A hopping mechanism could explain the reason behind this phenomenon. The charges can rapidly hop from one CNT to neighboring CNTs based on the three-dimensional hopping conduction model, leading to effective transport along the axial direction of the CNT fiber.[162] On the other hand, it is known that low-modulus fibers usually exhibit more rapid electron-transfer kinetics than high-modulus fibers due to a smaller fraction of exposed edges, also making aligned CNT fibers ideal candidates for electrodes.[161]

Fibers based on conducting polymers are another important group for fiber devices. They rely on functional groups on the polymer backbone to conduct electrons. These materials can also store charges in the electro double layer through a rapid faradaic charge transfer.[163] The presence of noncovalent bonds among polymer chains makes the conducting polymers favorable for ion transports.[164] Post-treatment is also an effective way to alter the electron-transfer process for fiber materials. For example, by doping with noble metal nanoparticles or electrodeposition of metallic layers on carbon-based or polymer electrodes, the electron-transfer kinetics can be further improved.[165] The surface roughness, specific surface area, and functional group are also important factors for the electron transfer in fiber devices, which should be taken into consideration when designing fiber devices.[166]

3.3.2. Ion Transfer. Ion transfer is another important class of charge transfers for many devices, including energy-harvesting/storing devices or biosensors.[167] The ion transfer in such devices typically involves three processes: ion migration from the bulk electrolyte to the electrode surface, ion adsorption, and desorption from the electrode surface. In the absence of an external driving force, ions in an electrolyte are under random motions, described by Fick's law of diffusion.[168,169] When a potential is applied, the ion migration in bulk is a process that is affected by the ion itself (e.g., ionic radius and charge), the electrolyte, and the applied potential.[170] When the ion is approximately within the electrode, the second ion-transfer process begins to take effect. Depending on the properties of the electrode surfaces, such as specific surface area, porosity, and wettability, the adsorption and migration processes of ions are varied on the electrode surface.[171]

These processes also apply to the ion transfer among fiber electrodes, driven by the electric field built between two electrodes. However, unlike their planar counterparts, which could establish uniform electric fields perpendicular to the electrodes and efficiently drive the ion transfer between the two electrodes (Figure 6a), the fiber electrodes normally

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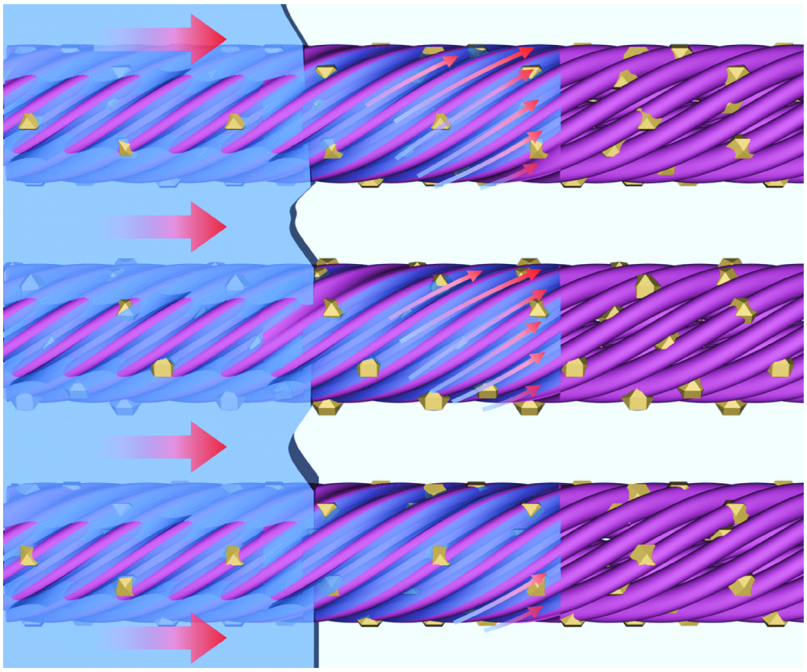

Figure 7. Electrolyte infiltration in a hierarchically assembled helical fiber electrode. The nanoscale gap within the primary fibers was first infiltrated, followed by the micrometer gaps between the primary fibers (indicated by the red arrows). Reprinted with permission from ref 151. Copyright 2020 Science China Press.

generate insufficient electric fields between the electrodes, resulting in unutilized electrode surface area and poor performance (Figure 6b).[17] A possible solution is to use hierarchically helical fiber electrodes, so the active materials could be evenly distributed inside the porous fiber electrode, thus building a uniform electric field between the two electrodes for ion transfer (Figure 6c).[17] The porous nature of this electrode also provides larger electrode−electrolyte interfaces and allows more rapid infiltrations of electrolytes into the electrodes via the produced multiscaled gaps (Figure 7).[151]

3.4. Chemical Reactions on Fiber Electrodes

Depending on specific functions, chemical reactions on fiber devices are vastly different. Fiber energy-storage and -conversion devices, such as batteries and fuel cells, require a pair of oxidation and reduction reactions on two electrodes to complete an electric circuit, while on fiber biosensors, the chemical reactions on the working electrode are the major research of interest. In general, active materials modified on fiber electrodes are responsible for chemical reactions, while the fiber electrodes are designed to efficiently complete the charge-transfer process. As a result, the smooth mass transfer between electrode and electrolyte is crucial for rapid and reliable chemical reactions and requires functionalization and modification of fiber electrodes. In addition, it is also of great significance to find proper fiber electrodes to host active materials and well-maintain their structures to be both mechanically and chemically stable in real applications, which will be discussed in detail in each later section.

4. FIBER ENERGY-HARVESTING DEVICES

4.1. Fiber Solar Cells

Solar cells can directly convert light to electrical energy through the photovoltaic effect, first discovered in 1839.[172] The first solar cell was reported by Bell Laboratories in 1954.[173] Since then, the research of solar cells has boomed because solar energy is renewable, clean, sustainable, and readily available, making it a promising alternative to fossil fuels.[174−176] In general, solar cells are classified into three generations, including wafer-based first generation (e.g., amorphous silicon), thin-film-based second generation (e.g., amorphous silicon and CdTe solar cells), and emerging third generation (e.g., dye-sensitized solar cells (DSSCs), polymer solar cells (PSCs), and perovskite solar cells (PeSCs)).[141,177−179] The first two generations of solar cells have already been commercialized because they can power portable electronic devices with power conversion efficiencies (PCEs) of >20%.[179] However, the third-generation solar cells can offer higher PCEs, lower manufacturing costs, longer lifetimes, and improved mechanical properties (such as flexibility and stretchability). While many successful examples of the third-generation solar cells have been demonstrated as planar devices, fiber solar cells have additional advantages as wearable devices because they can accept light irradiation from all directions, potentially improving their performances.[180−182]

4.1.1. Fiber Dye-Sensitized Solar Cells. Fiber solar cells inherit the third-generation design of solar cells, which can be classified into fiber DSSCs, PCSs, and PeSCs.[17] The fiber DSSC is the most extensively studied type due to its simple design, robust mechanism, and excellent performance.[183] A fiber DSSC consists of a fiber photoanode, a fiber counter electrode, an electrolyte, and a dye.[182−184] The device configuration and the chemical process of a typical DSSC are demonstrated in Figure 8. To make a DSSC work, photons absorbed by the dye molecules create charges, and the electrons are injected into the conduction bands of the photoanode and transmitted to external circuits. Meanwhile, the oxidized dye molecules are reduced by I^- while I^- is oxidized to I_3^-. Then, the I_3^- diffuses to the counter electrode and is reduced to I^- to complete the photoelectrochemical

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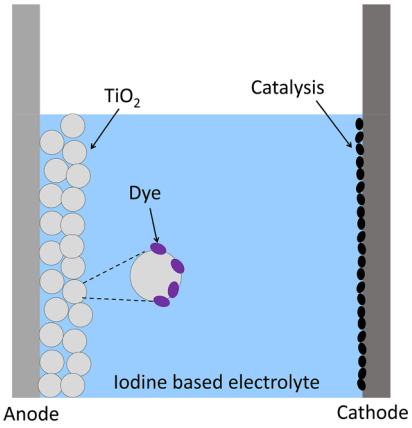

Figure 8. Device configuration of a typical DSSC.

cycle.[185,186] To accomplish this photoelectrochemical process with high efficiency, a few basic requirements are raised to design fiber DSSCs. (1) It must be easy for the surface of the fiber electrode to be immobilized with photoanode materials (usually Ti materials) and catalysts on the counter electrode (e.g., Pt). (2) The fiber electrodes should show excellent electrical conductivities and both high chemical and mechanical stabilities. (3) The device configuration should be favorable for light absorption and charge transfer.

Fiber Photoanode. The early fiber DSSC was designed using metallic fiber electrodes, such as stainless steel or Ti wire as the anode and Pt wire as the counter electrode, because of their high availability and excellent electrochemical performances. For example, a fiber DSSC with a TiO_2-coated stainless steel wire as the photoanode and a protective polymer-coated Pt wire as the counter electrode was reported.[187] The two fiber electrodes were twisted together, as shown in the SEM image in Figure 9, and demonstrated an open voltage, current density,

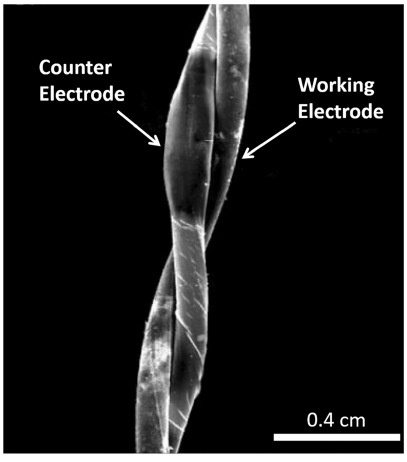

Figure 9. SEM image showing a twisted fiber DSSC. Reprinted with permission from ref 187. Copyright 2008 Wiley-VCH.

and fill factor of 0.61 V, 1.2 mA/cm^2, and 0.38, respectively. Later, to further improve the PCE of the fiber DSSCs, the TiO_2-coated stainless steel wire photoanode could be replaced with a P25 TiO_2 nanoparticle-modified Ti wire electrode. The fiber electrodes were twisted in the electrolyte solution and sealed in a capillary, and the resulting fiber DSSC demonstrated an improved PCE of 5.41%.[188] To further improve the PCE of the fiber DSSCs, the same group developed a grooved microreflector that could help collect the diffusion light and thus enhance the performance of the fiber DSSCs. The PCE of the DSSCs could be improved to 7.02% due to their ability to collect light from all directions.[189]

Although a metal anode has been successfully used in fiber DSSCs, they are inherently heavy, rigid, and susceptible to corrosion, and they cannot endure repeated deformation either, thus having limited applications as wearable devices. To this end, various flexible nonmetal anode materials, such as carbon fibers, CNT fibers, and graphene fibers, were developed as photoanodes.[182] For instance, a CNT fiber-based DSSC was designed to offer both high mechanical strength and flexibility.[190] The CNT fiber can host both photoactive dye-loaded TiO_2 nanoparticles and function as the catalytic counter electrode, showing a PCE of 2.94%. Later, by replacing TiO_2 nanoparticles with aligned TiO_2 nanotubes on the anode surface, the PCE of the fiber DSSC was enhanced to 4.6%.[191] The highly ordered TiO_2 nanotube arrays on the CNT fiber anode improved charge-collection efficiencies via the one-dimensional electric channel, enhanced light scattering on the electrode surface, and showed a large internal surface area to host more dye molecules.[192−194] In addition, specific device configuration design is also an effective way to enhance the fiber DSSC performance. For example, the PCE could be enhanced to 5.4% by using five anodes assembled around a signal counter electrode, while the PCE was only 2.8% when using a single photoanode.[195]

Fiber Counter Electrode. The counter electrode is crucial because the DSSC needs catalytic reactions on this electrode to complete the photoelectrochemical circuit.[196] To avoid reactions on this electrode becoming the limiting step for the power-conversion process and improve the overall fiber DSSC performance, many efforts have been devoted to improving the catalytic and mechanical performances of the counter electrode.[197] For example, compared to early metal wire or bare CNT fiber electrodes with either limited mechanical or electrochemical performance, a Pt nanoparticle-modified graphene oxide fiber was designed.[198] The fiber counter electrode offered high electrochemical activity, and a 8.45% PCE was reached upon coupling the counter electrode with a dye-absorbed TiO_2-modified Ti photoanode. Rather than using novel fiber materials, designing proper electrode and electrolyte interfaces could also improve the performance of the fiber DSSC. For example, by engineering the surface properties of the counter electrode, e.g., by designing a hydrophobic aligned CNT core to provide high electrical conductivity and a hydrophilic aligned CNT sheath to offer sites to the active materials, the fiber DSSC could achieve a PCE of 10.00%.[199]

On the other hand, while Pt has been demonstrated as a reliable active material for the electrochemical reaction on the counter electrode, the use of the expensive noble metal limits the large-scale application of fiber DSSCs.[197,200] To this end, many efforts have been devoted to finding proper substitutes of Pt without compromising the performance of the fiber DSSC.[201,202] For instance, with excellent catalytic performance and a large surface area on the counter electrode, a $Co_{0.85}Se$-modified carbon fiber electrode showed a PCE of 10.28% when assembled as a fiber DSSC with TiO_2 nanotubes grown on the Ti wire as the photoanode.[202] Other catalytic materials, including carbon materials, metal oxides, transition metals, and conductive polymers, were developed as Pt-free materials on the counter electrode.[203] Such strategies can improve photo-

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electronic performances, lower the fabrication cost, and accelerate the commercialization of the fiber DSSCs. In addition, by wrapping a spring-like working electrode around a CNT sheet modified elastic counter electrode, a flexible and stretchable fiber DSSC could be produced (Figure 10a and b).

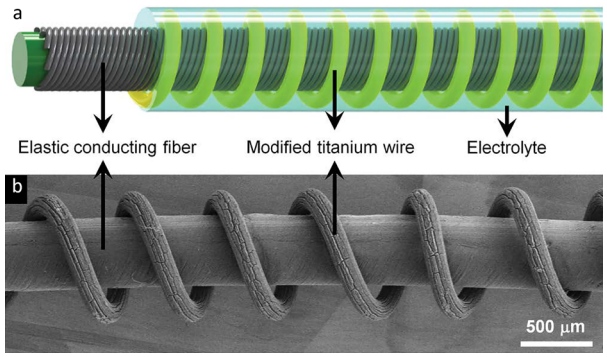

Figure 10. (a) Design of the flexible and stretchable fiber DSSC. A spring-like working electrode was wrapped around an elastic conductive fiber counter electrode to produce the fiber DSSC. (b) SEM image of the fiber device. Reprinted with permission from ref 204. Copyright 2014 Wiley-VCH.

It demonstrated a PCE of 7.13% and could be maintained under deformation, proving that the use of an elastic fiber counter electrode coupled with a spring-like working electrode can help the development of fiber DSSCs.[204]

Electrolyte. First, the electrolytes in fiber DSSCs should have high charge mobility to facilitate the charge-transfer process between the electrolyte and the electrode surface, which is a crucial step to complete the photoelectrochemical circuit. Second, the electrolyte should form a stable interface with the electrode. In addition, the light absorption of the electrolyte could decrease the PCE of the fiber DSSC.[17] The early fiber DSSCs are mainly based on liquid electrolytes.[85] Despite the progress, the liquid electrolytes may cause problems such as difficulties in packing and integrating into wearable platforms. Although reports indicated that the liquid electrolytes could be maintained on the electrode surface via a capillarity reaction, such an unstable interface may cause low PCE or device failure.[188,205] The liquid electrolytes also increase the risk of leakage during repeated deformation and safety issues to humans. Therefore, quasi-solid-state and solid-state fiber DSSCs were developed with novel gel electrolytes and solid-state electrolytes.[133,206−208] For example, a solid-state electrolyte was created using a hybrid of a LiTFSI electrolyte and 4-hydroxy-2,2,6,6-tetramethylpiperidin-1-oxyl with high catalytic oxidant characteristics. As a result, the fiber DSSC could provide a PCE of 6.16% and survive 500 bending cycles and 10 washing cycles with 92% of the initial PCE.[209]

Overall, fiber DSSCs have been developed rapidly during the past decade. The findings of new electrode materials, active materials, and electrolytes have evolved from experimental prototypes toward real platforms to power electronic devices. However, a few problems remain in the field. First, the PCE of the fiber solar cells needs to be further improved. Achieving this goal requires substantial research efforts to understand better the charge-separation and -transport processes on the different electrode surfaces or in the electrolyte. For example, the thickness and morphology of the active materials play an essential role in determining the performance of the fiber DSSCs.[210,211] Specifically, a thick and porous layer of active materials may increase the amount of hosted dye molecules and be better at trapping light on the electrode surface.[212] However, it may also decrease the charge-transfer efficiency due to the long transport pathway.[213] On the other hand, a thin layer of the active material could lead to efficient charge transport but may cause a short circuit between two fiber electrodes. How to balance this trade-off has been a long-standing problem in the field.

Second, better solid-state electrolytes are still highly demanded to facilitate the application of fiber DSSCs. However, a high-performance solid-state electrolyte with excellent carrier mobility is not available yet. The PCEs of the fiber DSSCs are usually lower for the use of solid-state electrolytes. Third, the large-scale fabrication of a high-performance fiber DSSC is challenging and requires a convenient electrode-modification method with low cost and careful design of the device structure and packing strategy.[214]

4.1.2. Fiber Polymer Solar Cells. PSCs do not require liquid electrolytes and could be fabricated via the solution-based method, and they thus may be suitable for large-scale fabrication.[85] The fiber PSCs usually consist of two electrodes, an electron-transport layer, a photoactive-polymer layer, and a hole-transport material.[215,216] The working mechanisms of fiber PSCs are similar to those of their planar counterparts. The photoactive layer was responsible for absorbing light to generate excitations, and the charges were separated and transported to the electrode surface to generate currents; thus, finding proper light-absorbing and charge-carrying materials is crucial to the operation of fiber PSCs.[85]

For example, a prototype of a fiber PSC was constructed by coating a fiber core with an indium tin oxide as an anode, a hole-transporting poly(3,4-ethylenedioxythiophene)/poly-(sodium-*p*-styrenesulfonate) layer, a photo-absorbing poly(3-hexylthiophene)/1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6)C_{61}, and an outer LiF/Al layer as the cathode. Such a fiber PSC could provide PCEs up to 1.1%.[217] However, although the indium tin oxide layer with a low resistance (∼10 ohm/cm^2) is widely used for planar PSCs and early fiber PSCs, they still suffer from the loss of the photocurrent (under 1 sun irradiance) when the transport distance is >10−15 mm, which lowers their PCEs and limits their applications as fiber PSCs.[218] Thus, a new fiber PSC was proposed using two fiber electrodes, which could eliminate the use of an indium tin oxide layer and improve the PCE.[218] The stainless-steel wire electrode was deposited with a TiO_x electron-transport layer, a poly(3-hexylthiophene)/1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6)C_{61} photoactive layer, a poly(3,4-ethylenedioxythiophene) layer, and an outer silver layer. Each layer could be deposited via a solution-based method by dipping the wire electrode into different precursors, and the thickness of each layer was carefully controlled to ensure smooth charge transport. Then, a secondary counter wire electrode was wrapped around the photoactive electrode with tension before both were sealed in transparent cladding (Figure 11) with a PCE of 3.27%.

To improve the flexibility and stretchability of fiber PSCs, new soft electrode materials and configurations should be researched. For instance, using flexible metallic Ti and CNT fiber as the fiber electrodes, the resulting fiber PSC could maintain ∼80% PCE after bending for 1 000 cycles.[219] Further, a flexible fiber PSC was obtained by wrapping the coaxial-fiber electrode around an elastic fiber and then coated with CNT

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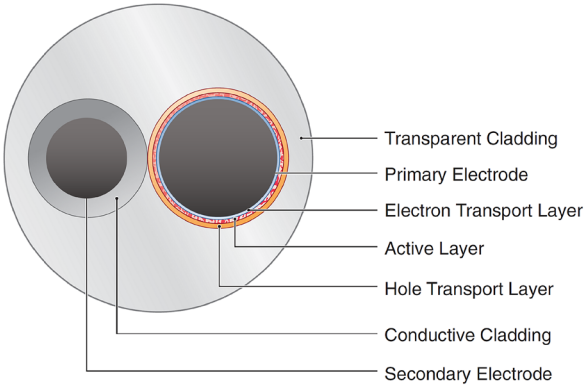

Figure 11. Schematic diagram showing the structure design of a fiber PSC. Multiple functional layers were coated on the primary electrode before cladding in a transplant polymer. Reprinted with permission from ref 218. Copyright 2009 AAAS.

sheets as the cathode. The fiber PSC could maintain the PCE by 88% for strains up to 40% and offer flexibility and stretchability upon weaving into textiles.[220]

4.1.3. Fiber Perovskite Solar Cells. Perovskite solar cells have attracted widespread attention due to their high light-harvesting ability, high charge-carrier mobility, and low cost.[221−223] Inspired by these advantages, fiber PeSCs have been developed during the past few years. The first fiber PeSC was proposed in 2014.[37] The coaxial-fiber PeSC consists of a metal fiber core, a compact blocking layer, a perovskite $CH_3NH_3PbI_3$ sensitizer, a hole-transport layer, and a CNT sheet as the cathode. The obtained all-solid-state device had a PCE of 3.3% and maintained 95% of the initial PCE after being bent for 50 cycles. Later, to improve the performance of the fiber PeSC, a Ti/c-TiO_2/meso-TiO_2/perovskite/hole-transport layer layer/Au was designed (Figure 12).[224] The

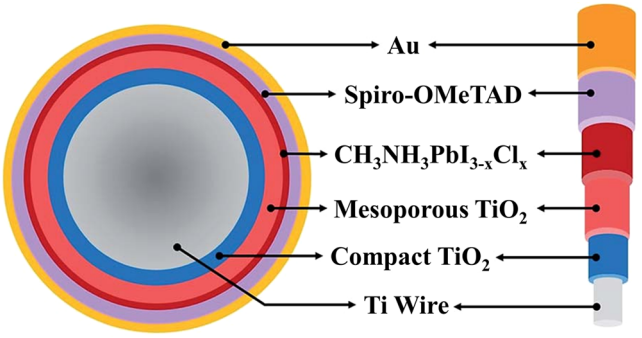

Figure 12. Schematic diagram showing the structure of a coaxial-fiber PeSC. Reprinted with permission from ref 224. Copyright 2016 Royal Society of Chemistry.

semitransparent Au cathode could facilitate light harvesting and charge collection. As a result, the fiber PeSC demonstrated a PCE of 5.3%. By further optimizing the fabrication process of the perovskite layer and TiO_2 layer via electrochemical deposition, the obtained functional layers could uniformly cover curved electrode surfaces, leading to a higher PCE of 7.1%.[225]

To improve the flexibility of the fiber PeSC, CNT fibers were used to form a twisted structure.[142] The obtained all-solid-state fiber PeSC exhibited a maximal PCE of 3.03% and maintained stability for >96 h in ambient conditions with a protective poly(methyl methacrylate) layer. The high flexibility allowed them to bear bending for >1000 cycles without significant performance degradation. In addition, wrapping the anode on an elastic fiber to form a spring-like structure made a fiber PeSC with high stretchability, and it maintained 90% initial PCE after enduring a strain of 30% for 250 cycles.[226]

Although fiber PSCs and PeSCs are promising as practical wearable devices due to their all-solid state, easy fabrication, and low cost, a few remaining problems have limited their further development. First, the PCEs of these devices need to be further improved. The delicate control over the interfaces among each functional layer could help accomplish this goal, as the charge transport among these interfaces plays an important role in determining the PCE. Further, a long operational lifetime of the fiber PSCs and PeSCs is required for real-life applications. However, these devices suffer from undesired degradation even under ambient conditions.[227−229] The stability of these devices could be improved by developing new functional layers or designing a more efficient packing strategy. Third, even with the all-solution-based fabrication process, the large-scale production of these devices is still difficult, and their performance may deteriorate with the increasing fiber lengths.[230] To understand this phenomenon, it is crucial to reveal how the inner resistance of the fiber solar cell varies with different device lengths and configurations, which requires a considerable amount of basic research in the field.

4.2. Fiber Nanogenerators

The nanogenerators can harvest and convert mechanical energy to electricity, which has recently attracted much research attention.[231−233] The human body, in constant motion, is a reliable energy source. Nanogenerators could be designed as wearable or implantable devices to collect mechanical energy from walking, breathing, or heart beating. The major types of nanogenerators are based on piezoelectric or triboelectric effects, and the detailed mechanisms of these two types of nanogenerators have been reviewed.[234] Briefly, piezoelectric nanogenerators (PENGs) use the piezoelectric effect to convert the stress to electricity, and triboelectric nanogenerators (TENGs) use electrification and electrostatic impact for collecting electricity.[235,236]

4.2.1. Fiber Piezoelectric Nanogenerators. The early fiber nanogenerator was developed with an Au/ZnO nanowire-modified Kevlar fiber wrapped around a ZnO/Kevlar fiber. The relative displacement between the two fibers caused deformation of the ZnO nanowire array, leading to the piezoelectric potential of the fiber electrode.[237] However, this device requires displacement between two twisted fiber electrodes. It cannot work as a wearable device because it is challenging to achieve reversible motions between the two twisted electrodes. To solve this problem, a coaxial-fiber PENG was developed (Figure 13). The device consists of a carbon fiber core and a ZnO thin film as the piezoelectric material. Upon applying pressure on the device, the piezoelectric effect in the ZnO layer could produce an output peak voltage of 3.2 V and an average current density of 0.15 $\mu A/cm^2$. The device was sensitive to pressure change and could be driven by a heart pulse.[238] Later, more efforts were devoted to improving the performance of the fiber PENG, including using new electrode materials to enhance charge collection and using new piezoelectric material compositions to enhance energy-conversion efficiency.[239−241] Further, the piezoelectric layer

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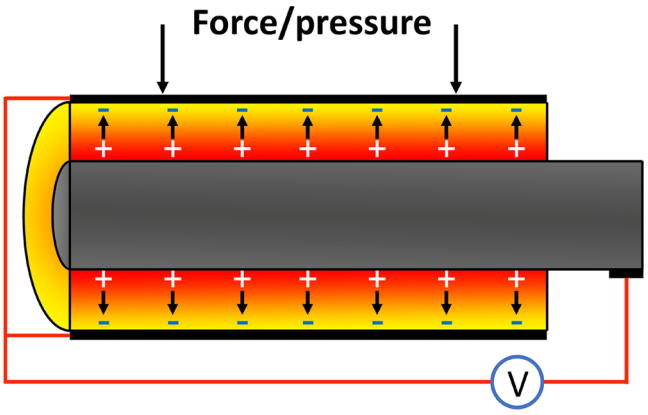

Figure 13. Schematic of a coaxial-fiber nanogenerator based on a carbon fiber coated by a ZnO layer. Reprinted with permission from ref 238. Copyright 2011 Wiley-VCH.

on the fiber surface is vulnerable to deformation, and damage of this layer may hinder the performance. To solve this problem, fiber PENGs were developed with soft polydimethylsiloxane encapsulation and piezoelectric poly(vinylidene fluoride) protection layers.[242,243]

4.2.2. Fiber Triboelectric Nanogenerators. PENGs require specific piezoelectric materials. In contrast, the triboelectric effect could be observed on two different materials or even on the same material, so TENGs have a much more comprehensive material selection. A more straightforward device design offers advantages toward large-scale applications.[244] In addition, the working principle for a TENG combines contact electrification and electrostatic induction, which could offer more significant high energy-collection efficiencies than PENGs. Thus, fiber TENGs have been extensively explored in recent years.[38] For example, a fiber TENG could be fabricated by coating polydimethylsiloxane or silicone rubber on a conductive fiber core. This simple design could offer devices with power densities ranging from 31.39 to 85 mW/m^2, depending on the material selection and device optimization.[245,246]

Fiber nanogenerators have been rapidly developed, and they could directly power small electronic devices.[247−249] However, the output currents from these devices are still low, usually under a few hundred microamperes, limiting their applications. Besides continuously searching for new electrode/active materials and further optimizing device configurations, coupling them with other fiber energy-harvest or -storage units may be elegant solutions.[244,250,251]

4.3. Fiber Biofuel Cells

The human body contains a lot of chemicals, many of which can undergo oxidation or reduction, such as oxygen, glucose, and lactic acid. The working mechanism of biofuel cells is shown in Figure 14, where the oxidation occurs at the anode and the reduction occurs at the cathode.[252] Glucose and lactic acid are the most common fuels at the anode due to their abundant existence and chemical reducibility. Enzymatic or inorganic catalysts should be delicately modified on the electrode to achieve high-efficiency fuel oxidization.[252−254] In addition, small-molecule or polymer mediators are often used as electron shuttles to help the electron transfer from the redox center of the enzyme to the electrode.[255] Oxygen reduction is the common choice at the cathodes, which are also modified with enzymes (e.g., bilirubin oxidase) or inorganic catalysts (e.g., platinum nanoparticles). Because the concentration of oxygen is usually lower than that of glucose or lactic acid, the size of the cathode should be more significant than that of the anode for practical use to avoid the performance limitation due to the oxygen-reduction reaction.[256,257]

The electrode materials are decisive for biofuel-cell performance, while the choices of active materials (enzymes or inorganic catalysts) and mediators are relatively narrow. To construct a high-performance biofuel cell, the electrode materials should be highly conductive, easily modified, nontoxic, and chemically and mechanically stable.[74,258,259] An early example of a fiber biofuel cell that used a 7-μm-diameter carbon fiber as the electrode was reported.[260] By modifying glucose oxidase and an electron-conducting redox polymer on the anode and laccase on the cathode, such fiber biofuel cells could be operated in buffer solutions with power densities of 64 and 137 μW/cm^2 at 23 and 37 °C, respectively. More advanced fiber electrodes, such as CNT and graphene materials with enhanced electrochemical and mechanical performances, have been developed.[97,261] Since these pioneer works, biofuel cells have gradually made their way into working in a physiological buffer,[260,262−264] living plants,[265−267] and finally in animals[268−279] as implantable devices.

Although biofuel cells have demonstrated their ability as power sources for electronics or can be directly used as sensors,[280−287] brain stimulators,[288] or drug-release systems,[289,290] the applications in vivo for biofuel cells have still been hindered, partly due to the unstable performance of such devices in the body. Biofouling is a notorious phenomenon for blocking the active surface on implantable devices, occurring immediately after devices are in contact with biofluids.[291−295] A porous CNT fiber electrode was developed as an antifouling biofuel cell electrode to overcome this problem.[278] The porous structure could efficiently prevent the cells and proteins in body fluids to adsorb on the electrode surface, thus ensuring effective electrocatalytic reactions on the biofuel cell electrode. As a result, the device could maintain almost 100% performance after being implanted in living organisms, and a

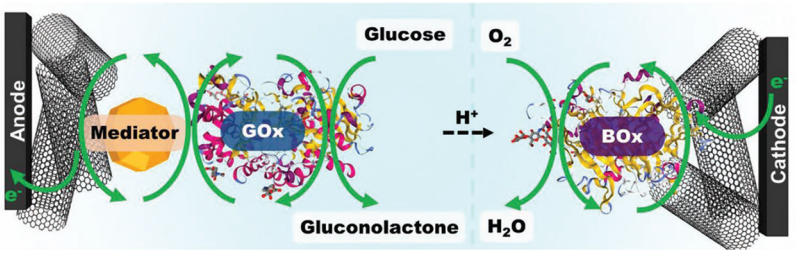

Figure 14. Schematic showing the design of a typical glucose/O_2 biofuel cell. Reprinted with permission from ref 252. Copyright 2020 Wiley-VCH.

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maximal output power of 76.6 mW/cm^3 could be achieved in vivo.[278]

Although the fiber biofuel cells have advanced dramatically, most of their applications remained only at the lab. This may be attributed to a few unmet challenges in the field. First, the lifetime of the biofuel cells is limited due to the reduced enzyme activity over time and the strict storage conditions (usually below 0 °C). These problems make it difficult and economically justified to design and fabricate textile biofuel cells at a large scale. Second, as a device that is in close contact with humans (to have access to the biofuels), biocompatibility is a significant issue that should be considered. The use of improper fiber electrode materials, exogenous enzymes, and toxic mediators may also raise safety and ethical concerns. Third, the smooth and uninterrupted access to a biofuel might be challenging for both wearable and implantable biofuel cells. However, with continuous efforts in the field, we believe that using fiber biofuel cells is an elegant way to power many bioelectronics in the near future.

5. FIBER ENERGY-STORAGE DEVICES

A family of fiber energy-storage devices, including supercapacitors and batteries, emerged in the past few decades, enabled by the fast development of fiber materials.[97,162,296] One major development trend of energy devices is to have close contact with human interfaces while being lightweight and portable and having good flexibility and stretchability.[297] Planar energy devices were first raised to address these needs, and a class of soft materials and specific structures (e.g., island bridge) were developed for designing planar wearable and implantable devices.[298] However, the practical use of these devices is challenging due to their bulky structure, which makes them uncomfortable for the users. Specifically, the flexibility of a planar device is usually limited, whereas for various human-interfaced devices arbitrary bending and deformation are constantly involved during use.[299] Additionally, planar devices are relatively bulky, and thus it is not easy to integrate them into fabrics in a large area to take full advantage of wearable devices.[141] Therefore, developing flexible, lightweight, and wearable high-performance energy-storing devices has become a major challenge.

5.1. Fiber Supercapacitors

5.1.1. Basic Information on Supercapacitors. Supercapacitors are a class of typical energy-storage devices that consists of two electrodes separated by an electrolyte and sealed by packaging materials. Supercapacitors use either an electrostatic double layer or electrochemical pseudocapacitor mechanisms to store and release electric energy.[300] In the electrostatic double layer supercapacitors, the current is generated via ion migration in the electrolyte and an adsorption/desorption process on the surface of the two electrodes. No chemical reactions occur during the charging and discharging of the devices, and thus only nonfaradaic currents are generated. The electrochemical pseudocapacitors contain electrochemically active materials such as metal oxides, metal hydroxides, or conducting polymers on the electrodes. During the charging and discharging of the devices, both nonfaradaic and faradaic currents (the dominating current) are generated on the electrodes.[301] Typically, electrostatic double layer supercapacitors do not involve chemical reactions, and they thus could transfer ions more rapidly than electrochemical pseudocapacitors, showing higher power density and cycle performance. However, the latter could load additional active materials on the electrode surface, thus having higher energy density than the electrostatic double layer supercapacitors (up to as much as 10 times).[81] Supercapacitors are generally categorized as symmetric and asymmetric ones based on electrode configuration or electrolyte composition. A symmetric supercapacitor typically comprises two identical electrodes and an electrolyte without redox activity, leading to facile material synthesis and device fabrication. In comparison, an asymmetric supercapacitor usually includes two kinds of different electrodes or has redox-active electrolytes; they are credited for broader stable voltage windows and higher energy densities compared with symmetrical ones. The performance of a supercapacitor could be evaluated by power density, energy density, specific energy density, and specific capacitance. The specific capacitance is often used as a key parameter, illustrated as follows:[302]

$$C_{sp} = \frac{2i}{m\Delta V/\Delta t}$$

where i, ΔV, and Δt represent the charge/discharge current, voltage window, and testing time, respectively, and m is the mass of the active materials on the electrodes. For fiber supercapacitors, C_{sp} could be measured by

$$C_{sp} = \frac{C_{WE}}{B_i}$$

Here, C_{WE} is the capacitance of the working electrode measured by a three-electrode electrochemical cell, and B_i is the geometric configuration of the electrode in the form of mass (g), length (cm), area (cm^2), or volume (cm^3). It should be noted that the measurement of fiber supercapacitor performance is not standardized, and readers are guided to other reviews throughout this issue.[81,303] However, it is well-recognized that a higher specific capacitance (C_{sp}) and operating voltage window (V) and a lower cell equivalent series resistance (R_{ESR}) are desired in a fiber supercapacitor. In general, C_{sp} and V require better electrode materials and proper electrode structure (e.g., a higher electrode surface area is favorable for increased C_{sp} and V), while a more conductive electrode and electrolyte and better interface between them could lower R_{ESR}.[304] A primary research direction of fiber supercapacitors is to achieve these goals.[305]

5.1.2. Materials for Fiber Supercapacitors. The choice of electrode is usually the first and vital step for designing fiber supercapacitors. The electrode properties, especially electrical conductivity, surface morphology, and functional group, are the dominating factors for obtaining high energy density and high C_{sp}.[133,306] Early fiber supercapacitors concentrated on designing and/or fabricating high-performance devices.[81] For example, metal wires were used as current collectors because of their high mechanical strength and electrical conductivity. The fiber supercapacitors were obtained by depositing various capacitive materials on the metal wire, such as metal oxides, conductive polymers, and carbon materials.[307]

Conducting Polymers for Electrodes. Conducting polymers are pseudocapacitive materials that have also attracted interest as electrodes of fiber supercapacitors. Polyaniline and polypyrrole with high electrical conductivities of ~1−100 S/cm in doped states are easily fabricated at a large scale with low cost, and they thus have been widely used as pseudocapacitive electrode materials for supercapacitors.[81,308,309] The theoretical

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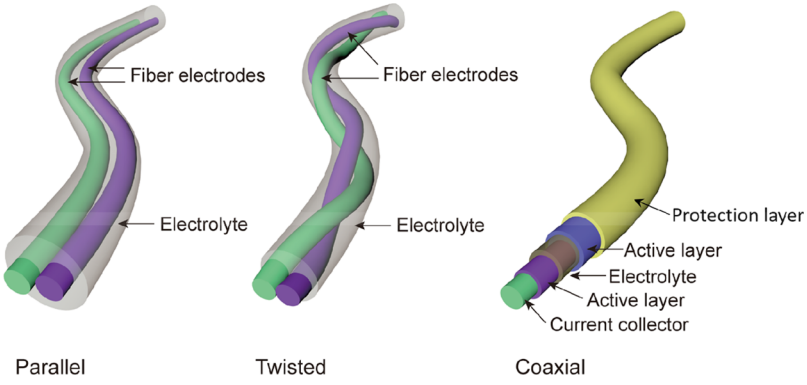

Figure 15. Schematic illustration of the detailed compositions of three typical configurations for fiber supercapacitors.

charge/discharge capacity of polyaniline and polypyrrole could reach 1284 and 480 F/g, respectively.[17] It has also been demonstrated that the supercapacitors could achieve great capacitance retentions of ~95 and ~85% after 10 000 cycles by using polyaniline and polypyrrole as electrodes, respectively, making them attractive materials for fiber supercapacitors.[310]

Carbon Materials for Electrodes. Carbon materials, such as CNT and graphene, have also been developed as electrodes for fiber supercapacitors. The sp^2 hybridized bonds in these materials offer large specific surface areas and excellent electrochemical and mechanical properties, making them ideal for fiber supercapacitors.[311−313] For example, by using biscrolled CNT yarn and metal wire current collector as fiber electrodes, the volumetric capacitance of the fiber supercapacitor could be up to 179 F/cm^3.[314] With the development of fiber supercapacitors, the pursuit of soft human-interfaced devices raised the search for mechanically flexible electrodes. CNT fiber electrodes thus emerged as a solution due to their inherent mechanical flexibility resulting from the helical assembly of CNT bundles. The supercapacitors based on CNT fiber electrodes could be used as wearable devices due to their weavable and flexible features, achieving a high specific capacitance of 323.8 F/g that can be further maintained by 95.5% after repeated bending processes for up to 10 000 cycles.[315]

Hybrid Materials for Electrodes. Hybrid electrodes use more than one type of electrode material in a fiber supercapacitor, aiming at harvesting the best of all the materials.[316] For example, MXenes are a class of two-dimensional inorganic materials with high electrical conductivities of transition metal carbides and hydrophilicity due to their hydroxyl- or oxygen terminated surfaces, which are favorable for high-performance supercapacitors.[317,318] By hybridizing $Ti_3C_2T_x$ MXene nanosheets and poly(3,4-ethylenedioxythiophene)−polystyrenesulfonate (PEDOT:PSS), the fiber electrode exhibited a high electrical conductivity of 1489 S/cm and an excellent volumetric capacitance of ~614.5 F/cm^3. The superior strength and flexibility of the hybrid fibers also allowed them to be wrapped on a silicone elastomer fiber to achieve 96% capacitance retention under 100% strain.[319] The hybrid electrodes, such as metallic/CNTs has also been reported with attractive performances.[320]

Electrolytes. An electrolyte is another important sediment in a fiber supercapacitor. As discussed earlier, the proper function (charging and discharging of the electrodes) of supercapacitors relies on fast ion transfer (electrostatic double layer model) or chemical reactions (pseudocapacitance model). To achieve this goal, the electrolyte needs to have a stable interface with the electrode and it should also be conductive to facilitate charge transfer.[305,321−323] The most prominent application scenario of fiber supercapacitors is for wearable or implantable devices, and thus the safety of the electrolyte raises another concern. Three types of liquid electrolytes, including aqueous electrolyte, organic liquid electrolyte, and room-temperature ionic liquid electrolyte, were widely used for fiber supercapacitors.[324] However, these electrolytes were challenging to be packed in fiber devices, limiting their human-interfaced applications.[325] In comparison, solid and gel electrolytes with high ion-migration rates are more attractive for these applications for fiber supercapacitors. They could eliminate undesirable short circuits of the two electrodes without the need for a membrane separator and offer convenience for packing the device.[323,326] For example, a poly(vinyl alcohol) (PVA)-based proton-rich gel electrolyte was widely used for fiber supercapacitors due to its high chemical stability, high ionic conductivity, expansive potential windows, low toxicity, and low cost.[325,327]

5.1.3. Configurations for Fiber Supercapacitors. Fiber supercapacitors usually use three basic configurations: parallel, twisting, and coaxial (Figure 15). The parallel fiber supercapacitors used two aligned electrodes immersed in the electrolyte and packed by a plastic tube or polymer layer. This configuration is often used when the diameter of the electrodes is relatively large (e.g., on the millimeter level). However, this configuration is not stable during deformations because the two fiber electrodes may deform differently under strain, causing the risk of electrolyte leakage and device damage.[328] To overcome this problem, fiber supercapacitors with a more mechanically stable configuration by twisting the two electrodes were developed.[36] The two twisted fiber electrodes were packed with electrolytes, making it easier to maintain the structure during deforming. The coaxial configuration is another major design of fiber supercapacitors. To better understand the working mechanism of coaxial supercapacitors, it could be regarded as a variant of a planar supercapacitor.[94] Similar to a rolled-up planar supercapacitor, the coaxial supercapacitor consists of a core fiber electrode, a separator or solid/gel electrolyte, and an outer electrode layer. The coaxial configuration provides a more efficient interface between two electrodes than the other two configurations, and it is also structurally stable upon deformations.[329,330] However, the continuous deposition of capacitive materials on a coaxial-fiber electrode is difficult, so a large-scale application of these supercapacitors is more challenging.

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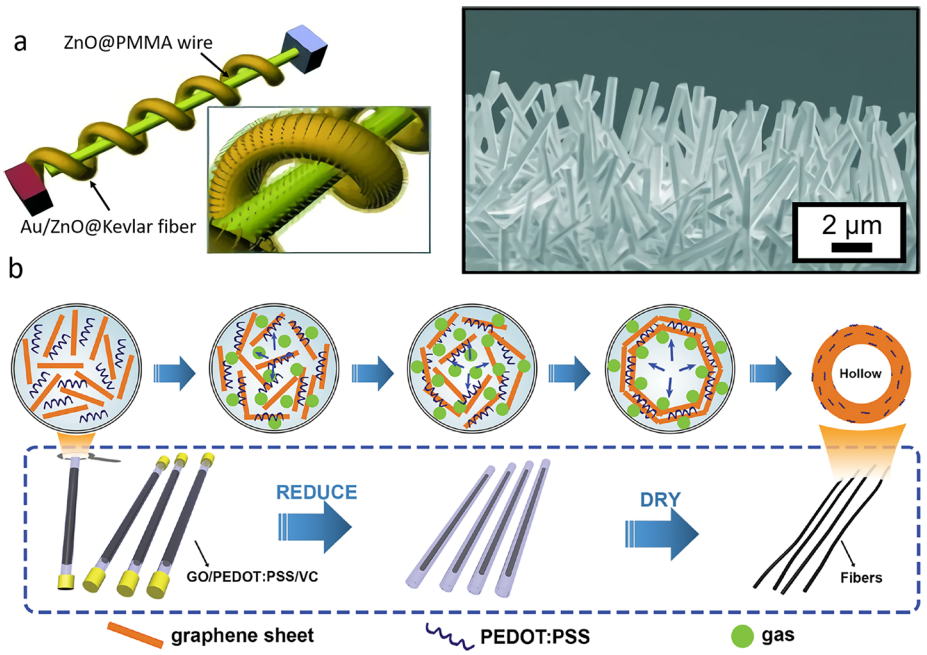

Figure 16. (a) Schematic illustration of a fiber supercapacitor, using a ZnO@PMMA wire wrapped around an Au/ZnO@Kevlar fiber to form the fiber supercapacitor. The closed-up SEM image of ZnO is provided in the right panel. Reprinted with permission from ref 34. Copyright 2011 Wiley-VCH. (b) Fabrication process of a hollow graphene/conducting polymer fiber electrode. Such electrodes could provide additional surfaces so that the fiber supercapacitor may achieve a larger specific capacitance. Reprinted with permission from ref 331. Copyright 2016 Wiley-VCH.

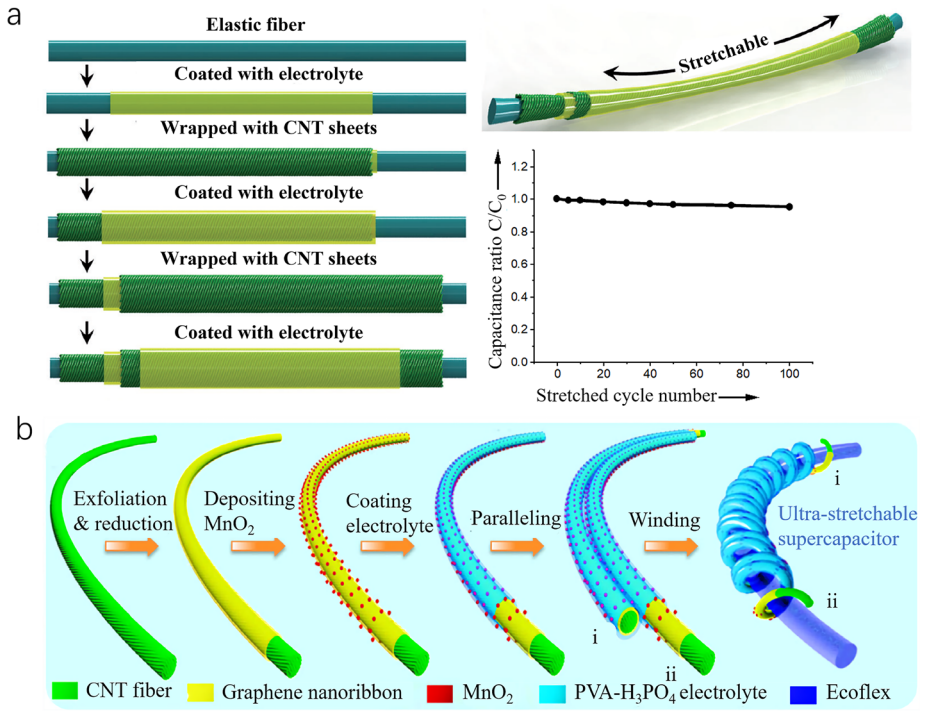

Figure 17. (a) Illustration of fabrication of a highly stretchable fiber supercapacitor with a coaxial structure using an elastic fiber core. The fiber supercapacitor could stably work after being stretched for 100 cycles. Reprinted with permission from ref 94. Copyright 2013 Wiley-VCH. (b) Design and fabrication of an ultrastretchable fiber supercapacitor. Two flexible CNT@graphene@MnO_2 fibers were helically wound around a superelastic core fiber to produce the fiber supercapacitor. Reprinted with permission from ref 335. Copyright 2018 Springer-Verlag GmbH.

5.1.4. Toward High Performances and Multiple Functions. Supercapacitors were mainly used in the automobile industry, such as regenerative braking or burst-mode power delivery.[17] In recent decades, they have attracted attention in wearable devices due to their simple design and reliable performance, which enables them to power wearable sensors and smart textiles. Early works on wearable supercapacitors were based on a planar configuration.[325] Fiber supercapacitors later emerged due to being lightweight, weavable, and flexible with good electrochemical performance. An early fiber supercapacitor that was introduced used a ZnO nanowire-modified Kevlar fiber as an electrode, along with a

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thin layer of gold deposited to improve the charge-collection capacity (Figure 16a).[34] The fiber supercapacitor demonstrated high specific capacitances of 2.4 mF/cm^2 in the PVA/H_3PO_4 gel electrolyte. Since this early prototype of fiber supercapacitor, new fiber supercapacitors have been developed using new electrode systems or different structures. Table S1 in the Supporting Information lists a few notable progresses in this field over the past decade.

Other than finding new electrode materials, exploring novel microstructures of fiber electrodes is also an efficient way to improve the performance of fiber supercapacitors. For example, a hollow rGO/conducting polymer composite fiber was designed (Figure 16b) to reach a high specific areal capacitance up to 304.5 mF/cm^2, which corresponded to an energy density of 27.1 μW/h cm^2 at a power density of 66.5 μW/cm^2.[331] The excellent electrochemical performance was attributed to the additional surface area created by the hollow structure of the fiber electrode, which could be maintained by 96% after 10 000 cycles of charging and discharging.

Another important research direction is to develop multiple functions of these devices and eventually push their applications one step closer to real life. As mentioned earlier in this section, a significant challenge for fiber supercapacitors is to improve their mechanical flexibility and stability to construct an intimate interface for humans. Soft electrodes, safe capacitive materials, and reliable packing of the supercapacitors were required to achieve this goal. Among them, finding a soft electrode with high stretchability and flexibility is a crucial step. A simple method to obtain such electrodes is to modify elastic fibers with functional materials. For example, aligned CNT sheets simultaneously have high tensile strength, flexibility, electrical conductivity, and mechanical stability. By modifying the CNT sheets with a H_3PO_4−PVA gel electrolyte on an elastic fiber (Figure 17a), the fiber supercapacitor could maintain a high specific capacitance of ∼18 F/g after being stretched by 75% for 100 cycles.[94]

The use of a prestretched fiber electrode could also make fiber supercapacitors superstretchable. Researchers have realized fiber supercapacitors that could maintain 97% capacitance after being stretched by 350% by using a prestretched fiber electrode.[332] Later, the performance of a fiber SC was improved to maintain 79.4 F/g after being stretched at 300% for 5 000 cycles using aligned CNT/polyaniline composite sheets as electrodes.[333] Another example showed that, by wrapping CNT sheets on a fully stretched coiled rubber fiber, the capacitance of the supercapacitors decreased by <7.4% during reversible stretching by 600%.[334] By helically winding around an elastic core with a CNT@graphene@MnO_2 fiber (Figure 17b), the supercapacitor could maintain 82% of the specific capacitance after 1 000 stretching and releasing cycles with strains of 700%.[335] These results showed that stretchable supercapacitors could operate well under different deformation conditions.

However, even with the high stretchability of fiber supercapacitors, breaking may still occur under repeated bending or other deformations during use, leading to device failures. A self-healing fiber supercapacitor was proposed to overcome this problem by wrapping electrically conducting nanomaterials around a self-healing polymer fiber. This device demonstrated 92% of the initial specific capacitance after breaking and healing cycles.[336] Later, researchers found that incorporating magnetic Fe_3O_4 particles into a fiber supercapacitor could assist the alignment of the breaking fiber electrode. Thus, all of the mechanical properties of the fiber supercapacitor could be well-maintained, restoring 71.8% of the specific capacitance after four breaking/healing cycles.[337] Other than searching for self-healing electrodes, other self-healing supercapacitors were developed by using a dual cross-linked polyelectrolyte and a notch-insensitive supramolecular hydrogel electrolyte, which enabled the device to undergo 50−60 breaking/healing cycles. Such results showed that the electrolyte design is essential for the self-healing ability of fiber supercapacitors due to the need for a stable electrode/electrolyte interface during the breaking/healing cycles, providing insight into an exciting research direction of this field.[338,339]

The development of fiber supercapacitors in real-life applications has been an essential direction of the field. Early examples tried to incorporate dye particles into CNT fiber supercapacitors. The resulting fluorescent fiber supercapacitor could show colors under ultraviolet light, paving the way to construct wearable devices in dark environments.[340] Further attempts combined fiber supercapacitors with other energy-harvesting devices, such as solar cells and nanogenerators.[341,342] Such devices could simultaneously convert solar energy or mechanical energy to electricity and then store it for future use. This integration strategy offered the promising prospect for scaled-up practical applications by weaving the fiber supercapacitors into functional textiles. In addition, fiber supercapacitors are also promising as implantable devices to power medical devices in vivo. For example, a biscrolled PEDOT:PSS/ferritin/MWNT fiber supercapacitor was developed with an actual capacitance of 32.9 mF/cm^2 in phosphate-buffered saline, which could be maintained by >90% after being implanted in mice for 8 days.[343] The results showed that fiber supercapacitors might be able to power devices in vivo and hold promise for self-reliant medical systems.

To conclude, fiber supercapacitors have been developed rapidly in the past decade. Their electrochemical performance has increased with the exploration of new electrode materials and new microstructures, even though fiber supercapacitors with higher charge-storage capabilities are still desired.[344,345] The functions of the fiber supercapacitors have also been expanded. However, most of the applications are still at the early stage. We anticipate that the fiber supercapacitors could be integrated into wearable or implantable electronic systems as energy-storing devices, such as to power practical medical devices like pacemakers or implanted biosensors. The large-scale fabrication of fiber supercapacitors is essential for broad applications. However, it has been challenging because manufacturing fiber devices at a large scale usually accompanies problems such as insufficient electrode modification and a poor electrode/electrolyte interface, unavoidably leading to device failures. Although pioneering work has successfully demonstrated the fabrication of fiber supercapacitors >100 m long via the thermally drawn technology, more efforts should be devoted to this direction to enhance the performance of fiber supercapacitors, lower the cost, and improve the reliability of such industrial production.[346]

5.2. Fiber Batteries

5.2.1. Necessity of Fiber Batteries. The battery is an essential part of modern life since it has been used for portable power devices, medical electronics, and automobiles. A battery requires a cathode, an anode, an electrolyte, and a separator to work.[347] The traditional battery shares a bulky three-dimen-

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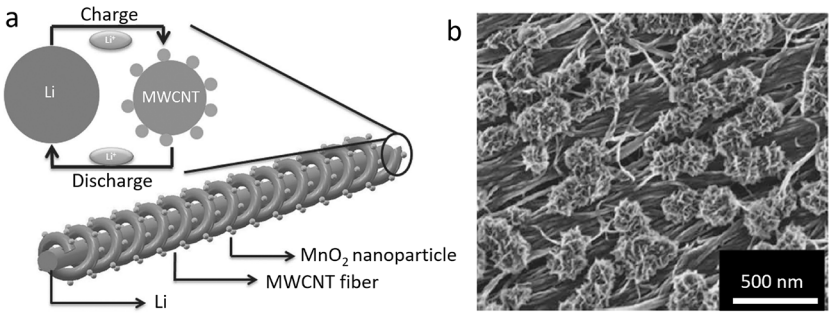

Figure 18. (a) Design and structural demonstration of a fiber battery using CNT fiber/MnO_2 as a cathode and Li wire as an anode. (b) SEM image of deposited MnO_2 particles on the fiber electrode. Reprinted with permission from ref 36. Copyright 2013 Wiley-VCH.

sional shape, but the development of current wearable devices requires them to be miniaturized, flexible, and stretchable.[348−350] The early wearable batteries had been made into thin films, which were attractive for powering wearable sensors and other devices.[351] Following this trend, a few commercial wearable battery companies have emerged. However, the film-like batteries are still relatively bulky and rigid, and particularly they may cause discomfort to the users due to the airtight interfaces with the environment.[17,352] To this end, fabricating fiber batteries is recognized as a promising strategy to accommodate various deformations including bending, stretching, and twisting. Fiber batteries are usually composed of two fiber electrodes embedded in the electrolyte with diameters from tens to hundreds of micrometers. With the unique one-dimensional configuration, the fiber batteries are expected to stably operate upon various serious deformations. Moreover, they are also easily woven into smart textiles to power a spectrum of wearable electronic devices.

5.2.2. Difficulties in Fiber Batteries. *High-Efficiency, Uniform Electrode Modification.* The high curvature and small diameter of fiber batteries make it difficult for active materials to form robust and uniform layers on fiber electrodes. Such a problem is significantly exacerbated when large-scale fabrication is involved.[53] To overcome this issue, the fiber electrodes should have large specific surface areas to host active materials. The modification method also plays an important role in this process. Many methods have been developed to ensure successful electrode modification, such as dip-coating, spray/cast-coating, in situ electrochemical reaction, thermal drawing, and solution extrusion.[20,353]

Configuration Integrity. Similar to fiber supercapacitors, fiber batteries also use three typical configurations, i.e., parallel, twisted, and coaxial. A fiber battery consists of fiber electrodes, active materials, electrolytes (also functioning as separators in most cases), and packing materials.[162] Encapsulating such a long, thin device and maintaining the device structural integrity under various deformations are tricky.[53] A short circuit and electrolyte leaking resulting from structural failure could cause a disastrous outcome for the device itself or the users, which should be carefully considered during the design of fiber batteries.[58]

Mechanical Performance. The applications of fiber batteries raise unique requirements for their mechanical performances.[354] As a part of wearable or implantable electronics, fiber batteries should have decent mechanical strength, flexibility, and stretchability. This challenge could be tackled by using soft and stretchable fiber electrodes, such as CNT and graphene fibers.[355] With the development of new fiber electrodes, we can expect more options when designing fiber batteries with suitable mechanical properties.

Safety Concern. It is known that conventional planar batteries face many safety concerns (e.g., short circuit, burning, or explosion) owing to the intrinsic characteristics of battery materials (e.g., dendrite growth, electrolyte combustion, and electrolyte leakage) and inappropriate operation (e.g., over-charge, exposure to high-temperature environments, and component aging). These problems become more serious for fiber batteries because they always experience continuous deformations and changeable application environments. Toward the important wearable applications, the safety issues must be especially considered in the material choice and structural design of fiber batteries. Currently, many fiber batteries have been designed and constructed using flammable organic solvent electrolytes and active Li/Na metal anodes to achieve better electrochemical performances. These device designs provide good inspiration from the perspective of basic research but also pose potential safety risks in practical applications, especially considering the failure of encapsulating materials, which may lead to the leakage of electrolytes and active materials.[51] Many efforts have aimed at addressing these concerns.[356] For example, aqueous fiber batteries have been developed because the commonly used organic electrolyte in fiber batteries could be toxic to humans.[43] Quasi-solid or all-solid fiber batteries are safer than traditional liquid electrolytes because they avoid the electrolyte leakage and short-circuit issues. The biocompatibility of the electrolyte still needs improvement for practical applications. In the long term, the package of fiber batteries must be improved considering the severe deformation and usage environment.

5.2.3. Fiber Lithium-Ion Batteries. Fiber electrodes serve as substrates to host active materials, and an ideal fiber electrode should have a large specific surface area, low electric resistance, high chemical stability, and good mechanical strength.[141] To meet the requirements of wearable devices, the fiber electrodes should also be flexible, biofriendly, and capable of being produced and processed at a large scale.[19] A group of carbon-based fiber electrodes, such as CNT and graphene fibers, have been utilized for fiber batteries. Fiber batteries are evolving with the development of electrode materials, as electrode materials dominate the electrochemical and mechanical properties. Table S2 summarizes a few notable examples of the achievements.[357]

An early fiber lithium-ion battery was built in 2012 using a CNT/MnO_2 fiber as the cathode and a Li wire as the anode, as shown in Figure 18a.[36] The MnO_2 particles were decorated via electrochemical deposition on the CNT fiber electrode (Figure 18b) to achieve low electrical resistance and high mechanical

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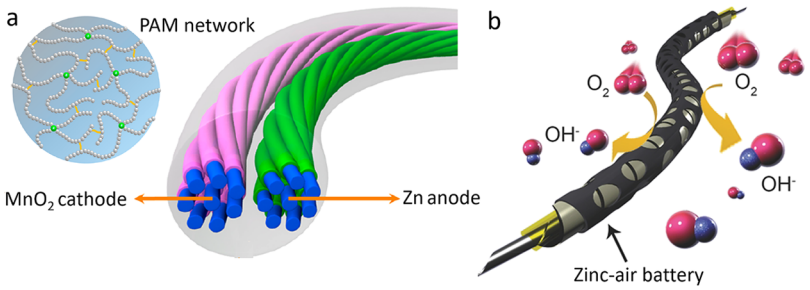

Figure 19. (a) Schematic illustration of a zinc ion fiber battery with MnO_2 and Zn modified helical fiber electrodes. A polyacrylamide polymer matrix was used as an electrolyte. Reprinted with permission from ref 366. Copyright 2018 American Chemical Society. (b) Design of an all-solid-state fiber zinc−air battery. Reprinted with permission from ref 28. Copyright 2015 Wiley-VCH.

strength. The CNT electrode also offered good flexibility, allowing the electrode to be bent repeatedly without causing electrode deterioration. When coupled with a Li wire anode, the fiber battery showed a specific capacity of 218.32 mAh/g and an energy density of 92.84 mWh/cm^3. Although this work represents an essential step in fiber batteries, a Li wire was still a problem for building safe and user-friendly fiber batteries due to the reactive chemistry nature of Li metal.[354,358]

Si has been proved to be a safe anode material for batteries with a high theoretical capacity of 4 200 mAh/g, which may be a potential substitute for Li anodes.[359−361] Si has been used for anode materials for decades, but the reversibility of Si anodes is limited due to the pulverization process during charging/discharging.[358] Finding an ideal electrode material to host Si has been a challenge due to the huge volume change during the Li^+ intercalation and deintercalation, which may cause battery damage and dysfunction.[362] To solve these problems, a thin layer of Si was coated on the CNT fiber electrode to form a core−sheath Si/CNT fiber anode.[363,364] This anode could maintain the high specific capacity of the Si and the high electrical conductivity of the CNT, while the nanoscale pores in the twisted CNT fiber could effectively endure the volume change of the Si component. The core−sheath structure and aligned feature of the Si/CNT composite nanotubes display high specific capacity and cyclic stability, providing insights into developing safe, efficient, and reliable fiber anodes.

The earlier-described examples demonstrated a few attempts toward safe, highly efficient, and mechanically durable fiber batteries.[363] However, there is still plenty of room to improve further the overall performance of fiber batteries, especially their electrochemical and mechanical performances and safety issues, by choosing fiber electrodes, active materials, and electrolytes.[18,99] High-performance fiber batteries could be designed by incorporating different active materials, including Si, MnO_2, $Li_4Ti_5O_{12}$, $LiMn_2O_4$, Fe_2O_3, or MoS_2, into highly conductive fiber electrodes such as a CNT fiber and rGO fibers.[55] For example, a coaxial lithium manganate/CNT fiber/lithium-ion manganese oxide cathode was developed to improve both the safety and working voltage of fiber batteries. Lithium-ion manganese oxide was a widely used cathode material due to its high voltage and structural stability and the capability to be easily processed on various electrode materials.[365] When coupled with a CNT fiber/Si fiber anode, the coaxial-fiber full lithium-ion batteries exhibit a linear capacity density of 0.22 mAh/cm or a linear energy density of 0.75 mWh/cm, demonstrating a successful step toward high-performance wearable energy-storage devices. Another work aimed at improving the electrolyte, as the low elasticity, low ionic conductivity, and poor mechanical strength of a solid or gel electrolyte may limit the applications of fiber batteries as wearable devices.[366] A cross-linked polyacrylamide polymer electrolyte was developed to host two MnO_2 and Zn coated CNT double-helix yarn electrodes to assemble a zinc-ion battery, as shown in Figure 19a. The quasi-solid-state fiber battery demonstrated excellent knittability, good stretchability (up to 300% strain), and waterproof capability owing to the polyacrylamide matrix and CNT fiber electrodes.[366]

5.2.4. Metal−Air Fiber Batteries. Metal−air batteries have a theoretical energy density that is much higher than those of lithium-ion batteries (~3600 Wh/kg for Li−air batteries).[367] Fiber metal−air batteries have additional advantages because the one-dimensional shape offers 360-degree gas and ion transport interfaces for improved electrochemical performances.[368,369] Unlike the commonly twisted or parallel structured fiber Li-ion batteries, the common configuration for metal−air fiber batteries is coaxial because it could provide a high active surface area for the air cathode. Flexible coaxial-fiber Li−air batteries were designed with a gel polymer electrolyte and an aligned CNT sheet air electrode.[370] The gel electrolyte served as an ionic conductor and a protector against oxygen diffusion to the lithium metal electrode. The reported fiber Li−air batteries showed a high discharge capacity of 12 470 mAh/g at 1 400 mA/g and a long lifespan of 100 cycles with a cutoff capacity of 500 mAh/g. Even though the gel polymer electrolyte can offer partial protection for the metal anode, the development of Li−air batteries is still challenging due to the unstable Li metal and dendrite problem, especially for the human-interfaced wearable devices.[296] The development of Li alloy anodes, such as M_xLi (M = Si, Ge, Sn, and Al), offers promising solutions to improve the stability of the anodes.[371] A coaxial Li−air battery was developed with a lithiated Si/CNT hybrid fiber as an inner anode, a polymer gel as a middle electrolyte, and a bare CNT sheet as an outer cathode. The fiber battery showed a high energy density of 512 Wh/kg that could be effectively maintained after bending for 20 000 cycles. Such a design can avoid dendrite formation, improve the safety of the lithium metal, and be suitable for large-scale applications as battery textiles.[354]

While most metal−air batteries use oxygen for cathode reactions, metal−CO_2 batteries have also attracted research attention, as they could justify the advantage in specific applications such as space exploration and CO_2 fixation.[372] A flexible fiber Li−CO_2 battery was constructed with a Mo_2C nanoparticle-coated CNT-based electrode.[47] The incorporated Mo_2C nanoparticles assisted in stabilizing the intermediate discharge product of $Li_2C_2O_4$ via a coordinative electron transfer, leading to a lower overpotential. The assembled fiber

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Li−CO_2 battery had a low charge potential below 3.4 V and a high energy efficiency of >80%, and it could be reversibly discharged and charged for 40 cycles. The as-fabricated flexible fiber Li−CO_2 battery could also keep working at increasing bending angles from 0° to 180°, showing the potential for powering wearable devices.[47] In addition, considering the ultralow temperatures involved in space exploration (e.g., approximately −60 °C on Mars), a Li−CO_2 battery that could work in such a harsh environment was developed by using a 1,3-dioxolane-based electrolyte and an iridium-based cathode. The battery demonstrated a discharge capacity of 8 976 mAh/g and a lifespan of 150 cycles at −60 °C.[373] Further, by integrating light-responsive semiconductor photocathodes, a series of fiber Li−CO_2 batteries were constructed with high round-trip efficiencies and cycling stability under illumination.[374,375]

Besides Li−air fiber batteries, Zn−air and Al−air fiber batteries are also promising due to their high safety, eco-friendliness, and high theoretical energy densities (1086 Wh/kg for Zn and 2769 Wh/kg for Al).[376] In addition, Zn and Al are relatively stable in air and can be assembled under ambient conditions, which is favorable for large-scale fabrication.[377] The early fiber Zn−air battery consists of a spiral Zn anode, an air electrode with a Fe/N/C catalyst, and a freestanding gel polymer electrolyte, as shown in Figure 19b.[28] The Fe/N/C catalyst used in the air electrode contributed to a high catalytic activity for the oxygen-reduction reactions. The catalyst-loaded electrode showed a higher voltage plateau of 0.9 V and a longer duration of 10 h. However, such a Zn−air battery was not flexible due to the rigid fiber electrode.[28] The CNT air electrodes are promising to solve this problem because of their large specific surface area, high electrical conductivity, good mechanical stability, and flexibility. Through the use of a RuO_2 hydrate layer coated onto the gel electrolyte as a catalyst for the oxygen-evolution reaction, the fabricated fiber Zn−air battery exhibited excellent discharge/charge performances at 1 V and 1 A/g. It could maintain stability after bending and stretching for 100 cycles.[378] Al−air batteries could be designed via a similar route. For example, a fiber Al−air battery based on a silver nanoparticle-coated CNT air cathode and a spring-like Al anode was developed with a high specific capacity of 935 mAh/g and a high energy density of 1 168 Wh/kg at 0.5 mA/cm^2. As a proof of concept, two tandem fiber batteries could be used to charge a digital watch, demonstrating their applications as wearable devices.[379]

However, despite the achievements in fiber metal−air batteries, several challenges remain, including parasitic reactions, poor cyclability in air, risk of electrolyte leakage, demand for more effective electrode modification, and difficulty in fabrication at a large scale.[369] Such problems can be gradually resolved with the innovation of novel electrodes, active materials, electrolytes, and new electrode-modification methods, which should be addressed in future studies.[380]

5.2.5. Aqueous Fiber Batteries. Aqueous batteries use an aqueous solution as an electrolyte to facilitate the charge transfer between electrodes.[381,382] Most fiber batteries use toxic and flammable organic electrolytes, but they may suffer electrolyte leakages and safety concerns during arbitrary deformations. Compared to nonaqueous batteries, aqueous batteries use nonflammable aqueous electrolytes and have low chances of explosion.[383,384] An early fiber aqueous lithium-ion battery was developed utilizing a polyimide/CNT hybrid fiber as the anode, a $LiMn_2O_4$/CNT hybrid fiber as the cathode, and a Li_2SO_4 aqueous solution as the electrolyte. The battery outputted a power density of 10 217.74 W/kg and a high energy density of 48.93 Wh/kg, which exceeded those of most supercapacitors. It could be woven into flexible energy textiles, holding promise as a safe and high-performance power system.[43]

Later, aqueous zinc-ion and sodium-ion aqueous fiber batteries were also developed with higher theoretical energy densities, higher safety, and lower costs compared to aqueous fiber Li-ion batteries. Aqueous Zn/MnO_2 batteries with $ZnSO_4$/$MnSO_4$ electrolytes are regarded as one of the most promising candidates for flexible Zn-ion batteries due to their broad electrochemical windows, cost-effectiveness, eco-friendliness, high specific capacities, and simple fabrications.[385] For example, a rechargeable Zn/MnO_2 fiber battery was designed with a graphene oxide-embedded poly(vinyl alcohol) hydrogel electrolyte via the synergy of graphene oxide and $ZnSO_4$/$MnSO_4$ salting out. The as-constructed battery showed stable cyclability exceeding 500 h while maintaining 98.0% capacity after 1 000 cycles. The fiber battery could be seamlessly integrated into a multifunctional e-textile, providing a stable energy power for continuous and simultaneous health management.[386] To solve the limited power-supply problem of wearable devices, a high-capacity aqueous Zn-ion fiber battery with air-recharging capability was constructe comprising a Zn anode and a freestanding V_6O_{13}/aligned CNT fiber cathode. The cathode offered abundant active sites for Zn-ion storage and could experience a spontaneous redox reaction with air at its discharged state to recover capacity. The resultant battery delivered high specific capacity (371 mA h g^{-1} at 200 mA g^{-1}) and long cycling life (91% capacity retention after 5 000 cycles at 5 A g^{-1}), and it could be efficiently recharged to ∼60% upon exposure to air. The fiber Zn-ion battery showed advantages of simultaneously high flexibility, capacity, and safety, and it had been integrated with sensors for various wearable applications.[387] To prevent the peeling of cathode active materials during deformations, a durable protective layer was developed to stabilize the incorporated cathode materials for flexible aqueous Zn−MnO_2 fiber batteries with both high integrity and durability. The resulting fiber battery demonstrated high durability and stable energy output under varying deformations and delivered a long cycle life of up to 4 000 cycles at 2 A g^{-1}.[388]

Aqueous fiber Na-ion batteries are promising to power various implantable electronics. For example, a biocompatible and rechargeable fiber battery was constructed with CNT hybrid fibers as electrodes. The soft fiber battery could be injected into all regions of the body by a mini-invasive syringe and showed a power density of 78.9 mW cm^{-3} in vivo, which is enough to drive various implanted electronic devices. The injectable fiber battery formed stable interfaces with tissues and showed high performances in the brain, heart, and subcutis.[51] Further, a biocompatible battery was proposed by designing biodegradable fiber electrodes with polydopamine/polypyrrole as the anode, MnO_2 as the cathode, chitosan as the separator, and body fluid as the electrolyte. The fiber battery can be directly injected into the body mini-invasively and can well-integrate with biological tissues without inducing immune responses.[389]

On a fundamental level, such aqueous fiber batteries could alleviate the safety concern of nonaqueous fiber batteries and demonstrate excellent electrochemical performances.[387,390−392] However, the low output voltage, electrode corrosion, and

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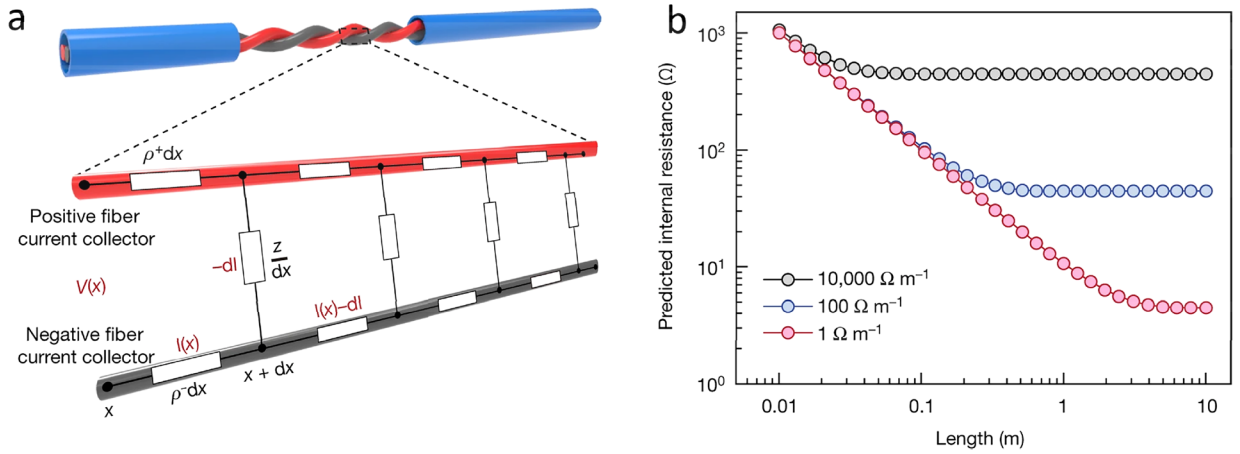

Figure 20. (a) Equivalent circuit diagram of a twisted fiber lithium ion battery including resistance units connected in series and parallel. The symbols ρ^{+}dx and ρ^{-}dx represent unit resistances of the positive and negative fiber current collectors, respectively. (b) Predicted internal resistances of current collectors with different resistances, using internal resistances with 1, 100, and 10 000 Ω/m as examples. Reprinted with permission from ref 53. Copyright 2021 Springer Nature.

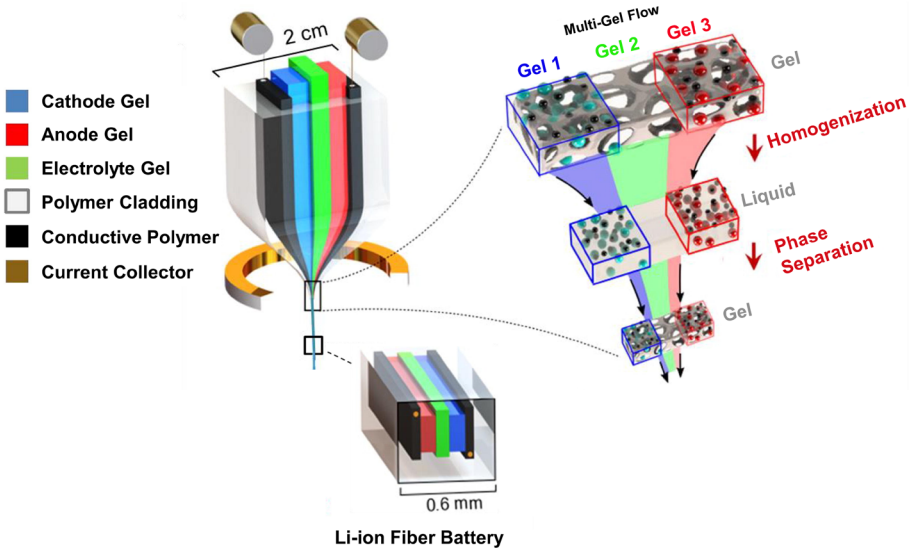

Figure 21. Schematic illustration of producing a continuous fiber battery using a thermal-drawing technique. Multiple components could be homogenized at the elevated temperature and then phase-separated at room temperature to form a fiber battery with a controllable configuration. Reprinted with permission from ref 59. Copyright 2022 Elsevier.

risky side reactions limit their further advances.[393] It should be mentioned that aqueous fiber batteries do not face these problems alone. It will benefit the evolution of all aqueous batteries family once these issues are addressed.

5.2.6. Self-Healing Fiber Batteries. Even though reliability and flexibility are partly realized for fiber batteries, along with the safety and bio/environmental compatibility being improved over the past few years, the breaking of fiber batteries may still cause safety concerns.[394] Self-healing ability is thus important to fiber batteries and has attracted increasing attention.[395,396] To solve this problem, a self-healing fiber battery was proposed with a porous rGO fiber filled with SnO_2 quantum dots as the anode and a $LiCoO_2$-modified spring rGO fiber as the cathode.[397] The lithium-ion battery could be assembled by the as-prepared cathode, anode, and gel polymer with a self-healing protective shell (e.g., carboxylated polyurethane). The fibrous anode and cathode had diameters of 750 and 250 μm, respectively, which could be reconnected with visual observation. The developed flexible and self-healable lithium-ion battery reached a capacity of 82.6 mAh/g under a series of deformations and could retain 50.1 mAh/g after the fifth healing process at a current density of 0.1 A/g.[397] Besides the self-healing polymer, fiber batteries could also be designed using self-healing gel electrolytes. For example, a self-healing fiber battery was designed utilizing in situ polymerization of calcium ion cross-linking sodium polyacrylate and sodium alginate hydrogel electrolyte. Such a fiber battery could self-heal for 8 cycles of complete breaking/healing with 68% capacity retention.[398]

5.2.7. Large-Scale Fabrication. *Inner Resistance Dilemma.* The large-scale fabrication of fiber batteries is limited by the uniform electrode modification and efficient packing. More importantly, for a decade, the internal resistances of fiber batteries were thought to increase with their increasing lengths.[55,162,399,400] Recently, such a hypothesis was overturned by a discovery that the internal resistance of such fibers has a hyperbolic cotangent function relationship with fiber length.[53] It first decreases then levels off as the fiber electrode length

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increases. The equivalent circuit diagram of a twisted fiber lithium-ion battery and predicted internal resistance of current collectors are shown as parts a and b of Figure 20, respectively, while the internal resistance R of a fiber lithium-ion battery with a length L can be expressed as follows,

$$R = \frac{[(\rho^+ + \rho^-) \times Z]^{1/2}}{\tanh(kL)}$$

where ρ^+ and ρ^- represent the unit resistances of positive and negative fiber current collectors, respectively. Z is the polarization resistance of a fiber lithium-ion battery measured by electrochemical impedance spectroscopy analysis, k is a parameter determined by ρ and Z, and L represents the fiber length. Encouraged by this result, an optimized, scalable, and industrial process was designed to produce high-performance fiber batteries on a meter scale. For example, Li-ion fiber batteries were fabricated via industry-standard equipment, where active materials and separators could be successively coated and wrapped onto the fiber current collectors.[53] The fiber electrodes were then twisted together and encapsulated in an encapsulation tube. To achieve high-loading, robust, and uniform coating of active materials on fiber electrodes, polyvinylidene fluoride binders were added to the positive slurry, and sodium carboxymethyl cellulose and styrene-butadiene rubber emulsion were mixed into the negative slurry to improve such interfacial adhesion. The as-prepared mass-produced fiber batteries displayed a high energy density of 85.69 Wh/kg and could maintain capacity by 90.5% after 500 charging/discharging cycles and 93% at a 1 C rate. The fiber batteries were soft and safe under various conditions so that they could be woven into textiles to power cellphones, fiber sensors, and textile displays.

Another approach to fabricate fiber batteries at a large scale was reported recently using a thermal drawing method.[59] Electroactive gels, particles, and polymers within flexible protective cladding were mixed and thermally drawn to form a fiber battery, as illustrated in Figure 21. Such a strategy allows for the fabrication of arbitrarily long lithium-ion fiber batteries. As a proof of concept, a 140-m-long fiber battery was shown with a discharge capacity of ~123 mAh and a discharge energy of ~217 mWh.

A solution-extrusion method that could produce continuous fiber batteries in a single step at an industrial scale was also reported recently.[58] The electrodes and electrolyte of a fiber battery were simultaneously extruded and combined through a three-channel industrial spinneret with high production rates. During the extrusion and drawing process, the shear force condensed and aligned the nanomaterials in the inks, resulting in a meter-long fiber battery. The aligned microstructure was favorable for charge transport and thus could help to improve the performance of the obtained fiber batteries. Such a technology can be generalized to fabricate Li-ion, Zn−Mn, and Na-ion fiber batteries using optimized cathode inks. In addition, the diameter of the extruded fiber batteries could be finely tailored to suit the application requirements. Those advances have established the first attempts in the large-scale fabrication of fiber batteries, while future efforts need to be devoted to further improving the electrochemical performance, safety, and ability to adapt to various application scenarios.

6. LIGHT-EMITTING FIBERS

Fiber light-emitting devices have attracted much attention due to their applications in disease therapy and textile display. Compared to traditional planar light-emitting devices, light-emitting fibers are more flexible and more easily integrated into functional textiles.[401,402] More importantly, the light-emitting spectrum independent of the observation angle make the light-emitting fibers competitive as bioelectronics and wearable displays.[403] A basic light-emitting fiber is composed of conductive fiber electrodes and functional materials. Conducting polymers, CNTs, and silver nanowires have been used as fiber electrodes.[138,404] Although indium tin oxide (ITO) is one of the most common options for a planar light-emitting device, the inherent brittleness limits its applications in fiber devices. The functional materials on light-emitting fibers are responsible for luminescence, including conjugated luminescent polymer, luminescent electrolyte, and emissive phosphor particles.[39,405−407] Depending on the specific type of light-emitting mechanism, an encapsulation layer can also be necessary to protect the device from oxygen and moisture.

6.1. Fiber Inorganic and Organic Light-Emitting Diodes

Light-emitting diodes (LEDs) have achieved wide applications in lighting, displaying, and bioelectronics.[408] Developing soft and stretchable LEDs for wearable and implantable applications has become an increasing popular research topic.[409,410] For example, μLEDs can be used as optoelectronic probes for a fundamental neuroscience study.[411] Recently, a thermal drawing process has been developed to fabricate diode fibers.[412] Specifically, light-emitting diodes and photodetecting p−i−n diodes in micron sizes are integrated into fibers with a uniform orientation using tungsten or copper as conductive wires (Figure 22a). Such a thermal-drawing process could fabricate hundreds of diodes in parallel inside a single fiber, paving the way toward large-scale application of fiber LEDs (Figure 22b).

Since their discovery in 1987, organic light-emitting diodes (OLEDs) have been widely used in television screens, computer monitors, and portable systems.[413] OLEDs have a typical two-dimensional structure consisting of a stack of thin organic layers sandwiched between an anode and a cathode layer.[414] Fiber OLEDs can be designed using a similar structure. For example, a fiber OLED was reported with organic charge transport and emission layers sandwiched between a metallic anode and cathode.[403] These layers were deposited on a 480-μm-thick polyimide-coated silica fiber via vacuum thermal evaporation. However, using the vacuum thermal evaporation to deposit organic layers is not suitable for the large-scale fabrication of fiber OLEDs. Thus, a solution-based strategy was later developed.[407] The conducting polymer poly(3,4-ethylenedioxythiophene)/polystyrenesulfonate was used as the cathode; zinc oxide nanoparticles, polyethylenimine, and Super Yellow were then dip-coated on the fiber; and the MoO_3 and Al layers were finally deposited via atomic layer deposition. The fiber OLEDs have a high luminance and current efficiency of >10 000 cd/m^2 and 11 cd/A, respectively. They could be weaved into textiles, showing promise for large-scale application (Figure 23). To optimize the power consumption and brightness of fiber OLEDs, the same group recently proposed phosphorescent organic light-emitting diodes based on a modified dip-coating method.[415] These fiber OLEDs exhibited excellent optoelectronic performance,

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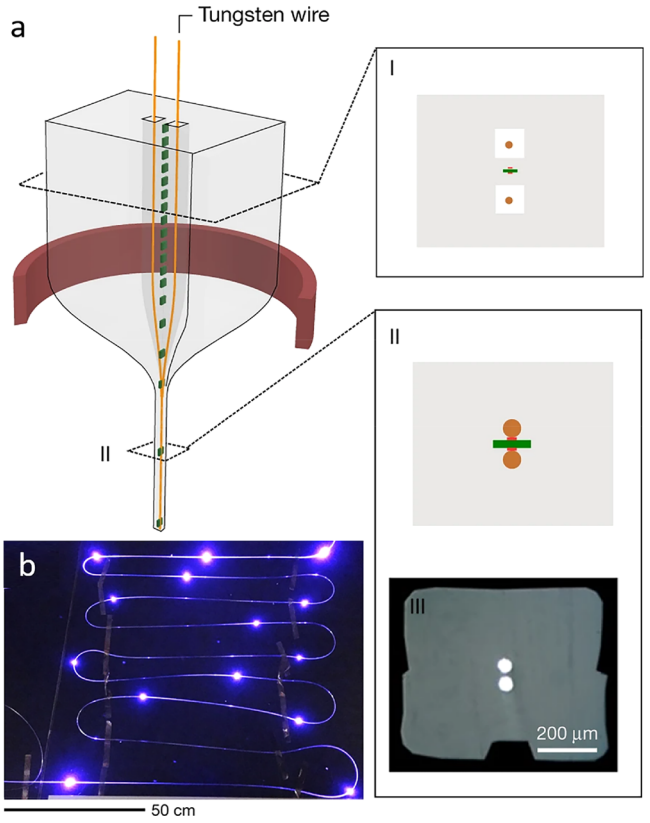

Figure 22. (a) Illustration of the light-emitting diode fabricated by the drawing process. The metallic wires were fed and then were heated and drawn (red ring). The metallic wires and devices were then embedded and packaged as fiber devices. The three insets show the cross-sectional schematic illustration (I and II) and image (III) of the device configuration. (b) Photograph of a light-emitting fiber containing multiple InGaN blue-colored LEDs. Reprinted with permission from ref 412. Copyright 2018 Springer Nature.

showing high current efficiency values of 16.3, 60.7, and 16.9 cd/A for red, green, and blue colors, respectively.

Recently, the hollow fiber electrodes have offered additional advantages. For instance, the empty core of a hollow fiber could better accommodate different functional materials, and the cylindrical geometry of the hollow fiber could effectively suppress the wave-guided light loss at the electrode/substrate interface, resulting in a higher luminance efficiency.[416] Attributed to these advantages, the hollow-fiber OLEDs could achieve a luminance of 6300 cd/m^2 and a current efficiency of 11 cd/A. Despite the huge progress, there remains room to further enhance fiber OLED performance by designing new electrode configurations.

6.2. Fiber Polymer Light-Emitting Electrochemical Cells

Polymer light-emitting electrochemical cells (PLECs) have achieved rapid development since their first appearance in the mid-1990s.[417] A PLEC consists of two electrodes and an active layer with a light-emitting material, ionic conductor, and dissolved salt. When an external bias is applied to the PLEC, the ion migration in the emission layer leads to the formation of a p−i−n junction. Then the electrons and holes move to the middle intrinsic junction and recombine to form excitons, leading to light emission.[418] Such a robust and simple light-emitting mechanism offers many advantages for PLECs, including simple device structure, low requirement for the electrode surface smoothness, and energy-level matching.

A color-tunable fiber PLEC was reported using all-solution-based processes.[39] The fiber PLEC used a coaxial configuration with a ZnO-modified metal wire as the cathode and a CNT sheet as the anode, and the electroluminescent polymer layer (e.g., blue light-emitting polymer PF-B, ethoxylated trimethylolpropane triacrylate and lithium trifluoromethanesulfonate) was sandwiched between them. As expected, the luminance was independent of the observation angle. The brightness of fiber PLECs could be maintained by >90% of its maximum after bending with a radius of curvature of 6 mm for 100 cycles. The all-solution-based fabrication process in the air promised the device the possibility of large-scale production. Besides conjugate polymers, an ionic transition-metal complex could also be adopted as an emission layer. A coaxial electroluminescent fiber was developed with Galistan liquid metal as the cathode, an ionic transition-metal complex as the functional layer, and an ITO thin-film coating as the anode.[419] The luminescence from the device could be detected by a charge-coupled device (CCD) camera at a turn-on voltage of 4.2 V and observed by the naked eye at 5.6 V in N_2, showing their possibilities for applications in optoelectronic textile.

6.3. Fiber Alternating Current Electroluminescent Devices

Alternating current electroluminescent (ACEL) devices that can emit light under alternating current have recently attracted much attention. A typical ACEL device consists of two electrodes, an insulator, and emissive phosphor particles.[420] The light-emitting mechanism for ACEL is relatively simple. The charge carriers are accelerated into high energy under the alternating electrical field, and then the excited and ionized luminescent center generates electron−hole pairs. The luminescence occurs due to the radiative recombination of charge carriers.[421] Such electric field-driven devices only require spatial contacts between electrodes and the luminescent layer to illuminate, making them intrinsically suitable and durable as highly flexible fiber ACELs in coaxial, twisted, or parallel configurations. For example, a coaxial ACEL fiber was

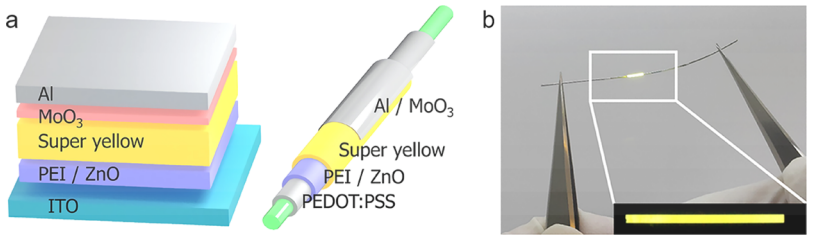

Figure 23. (a) Schematic showing a typical planar device of ITO glass-based OLEDs (left) and the design of a coaxial-fiber OLED fabricated by a solution process. (b) Photograph of the fabricated fiber OLED. The inset shows the enlarged light-emitting unit. Reprinted with permission from ref 407. Copyright 2018 American Chemical Society.

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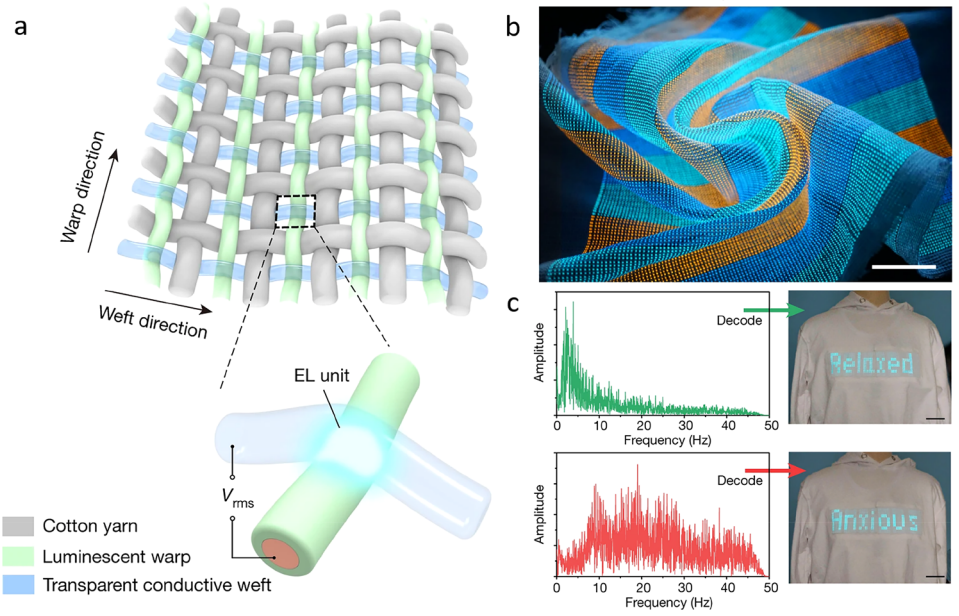

Figure 24. (a) Schematic showing the structure design and electroluminescence mechanism of the light-emitting textile, where the cross point emits light to function as a pixel. (b) Photograph of a three-color display textile under twisting. The blue and orange colors in the textile were achieved by doping ZnS with Cu and Mn, respectively. Scale bar, 2 cm. (c) Information related to brain wave being shown on the light-emitting textile. Reprinted with permission from ref 52. Copyright 2021 Springer Nature.

constructed using AgNW-based electrodes, a ZnS phosphor layer, a silicone dielectric, and an encapsulation layer.[138] Such ACEL fiber could be obtained via an all-solution-based fabrication protocol, showing an angular-independent luminance up to 202 cd/m^2 at 195 V and 2 kHz alternate current excitation. Considering the application scenarios of light-emitting fibers, a soft and stretchable ACEL fiber was proposed later.[406] The electrode was fabricated by wrapping a CNT sheet on a prestretched elastic fiber, and the luminescent layer was prepared by combining a light-emitting phosphor with elastomers. The as-prepared ACEL fiber could be stretched by >200% without compromising its illuminant property. A parallel ACEL fiber was developed with two hydrogel electrodes and a phosphor electroluminescent layer via a continuous one-step extruding method.[48] The ACEL fiber could emit light efficiently with a luminance of >240 cd/cm^2. Due to the high elasticity of the hydrogel and the electroluminescent layer, the fiber luminance could be fully recoverable at 300% strain, remaining stable after 100 cycles, and the ACEL fiber could even be stretched by up to 800%.

Despite the rapid progress of light-emitting fibers/textiles, the textiles could only display predetermined patterns rather than a dynamic display, of which the resolution depends on small, narrowly spaced illuminating units. Display textiles have not been achieved so far because it is challenging to obtain the small and yet durable illuminating units that could be easily assembled over a wide area of textile. An interwoven strategy of luminescent fiber was proposed to address this problem.[52] Rather than the traditional sandwiched structure, the electroluminescent unit was constructed directly at a fiber crossover point of luminescent warp and transparent conductive weft (Figure 24a). The structure allows the large-scale fabrication of textile displays with a decent resolution (Figure 24b). The conductive weft fibers were produced by melt-spinning ionic-liquid-doped polyurethane gel and luminescent warp fibers by coating commercially available ZnS phosphor on a silver-plated conductive yarn. The electric fields at the contact points/over the crossover points were stable and relatively uniform, enabling the textile to display stably. A 6-m-long and 25-cm-wide display textile containing 5 × 10^5 electroluminescent units could be achieved by this method. Such a display textile was flexible and breathable and could withstand repeated machine washings, showing practical applications in navigation or healthcare displays. For example, the device could help express mental states of a user by decoding representative electroencephalogram signals (Figure 24c).

6.4. Fiber Mechanoluminescent Devices

Mechanoluminescence is the emission of light in response to a mechanical force or stress imposed on a material, which was promising for the self-powered light-emitting device without the requirement of an external power source.[422] The first mechanoluminescent phenomenon was observed over 400 years ago, according to either stress-induced or triboelectricity-induced luminescence mechanism.[423] Mechanoluminescence can be observed when mechanoluminescent materials deform elastically or plastically or are fractured, corresponding to elastico-mechanoluminescence, plastico-mechanoluminescence, and fracto-mechanoluminescence, respectively. Elastico-mechanoluminescence is the most widely used phenomenon for functional devices because continuous light emission can be realized under cyclic elastic deformation.[423] Therefore, a simple mechanoluminescent fiber was constructed by coating a poly(dimethylsiloxane) fiber with ZnS phosphors particles.[424] When the fiber was deformed, the strain of the elastomer caused mechanical stress on the ZnS particles, and the piezoelectric potential generated by ZnS crystal deformation promoted carrier recombination to emit light. The mechanoluminescent intensity linearly increased with the increasing content of ZnS/Cu particles or applied strain, and the highest luminance reached 15.14 cd/m^2. The emitting color (green or orange) mainly depended on the doping elements or the ratio of emitters. The soft poly-

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(dimethylsiloxane) fiber allowed for the retained optical performance of the fiber after 10 000 cycles of stretching and releasing. Such a successful design of mechanoluminescent fibers allows for the development of force-sensitive, light-emitting fabrics that could be applied as wearable strain sensors to monitor and visualize human motion in real time.

7. FIBER SENSORS

Sensors could capture and translate targeted signals into other forms of easily observed or detected signals, and they are booming with the development of personalized digital devices in the Internet of Things era.[13,425,426] Generally, wearable sensors need to contact our bodies directly to provide accurate real-time physiological status and environmental information around humans.[427−431] While many sensors are designed in a planar configuration, fiber sensors also have been rapidly developed in the past decade.[432−437] The one-dimensional shape renders fiber sensors a few major advantages: (i) better flexibility to adapt to the human body for stable human−device interfaces, (ii) easier integration into textiles, (iii) easier implantation into deep tissues without causing traumatic damage to the tissue, and (iv) higher sensitivity for harvesting signals from all directions. Fiber sensors include physical (strain, light, electric signal, etc.) and chemical sensors. The sensors generally convert the target signals to electric signals and then deliver the signals to analyzing and monitoring systems. A wide range of wearable and implantable sensors are available on the market, such as Apple Watch and continuous glucose-monitoring devices, with the function to provide information on vital physical signals, blood glucose levels, or COVID-19 symptoms. However, they are relatively bulky and may cause discomfort to the user, which limits their applications. In contrast, fiber sensors could work in the form of textiles or miniaturized implants, could provide better interfaces with human users, and could be more readily blended into our daily life. As a result, there is a shift of research interests toward fiber and textile sensors.[426,438,439]

7.1. Fiber Physical Sensors

7.1.1. Fiber Strain Sensors. Strain sensors can transduce physical deformations into measurable electric signals, rendering them useful for applications in health and motion monitoring.[440−444] As an important subclass of strain sensors, fiber strain sensors could be classified into resistive, capacitive, piezoelectric, and triboelectric sensors according to the working mechanism.[27,445,446] The resistive and capacitive strain sensors are widely studied due to their simple structures and excellent performance. A fiber resistive sensor consists of a single electrode that changes its resistance with the applied strain.[27,447,448] In comparison, a fiber capacitive sensor contains two electrodes, where the capacitance changes with the variation of electrodes geometry or distance.[446,449] A fiber strain sensor should have an effective detection range over the stretching/contracting range of human motions of 55%, high stability, and low hysteresis to monitor human motion.[445,450]

The resistances of many fiber materials, such as CNT fibers, graphene fibers, conducting polymers, and liquid metals, change with the applied strain.[437,451−453] The equivalent circuit diagram of a CNT fiber sensor is illustrated in Figure 25.[454] Each resistor is composed of multiple CNT fibers in parallel, in which the distance among the CNT fibers enlarges with the applied force, leading to the increasing resistance of the sensor. The coiled CNT fiber-based strain sensor can

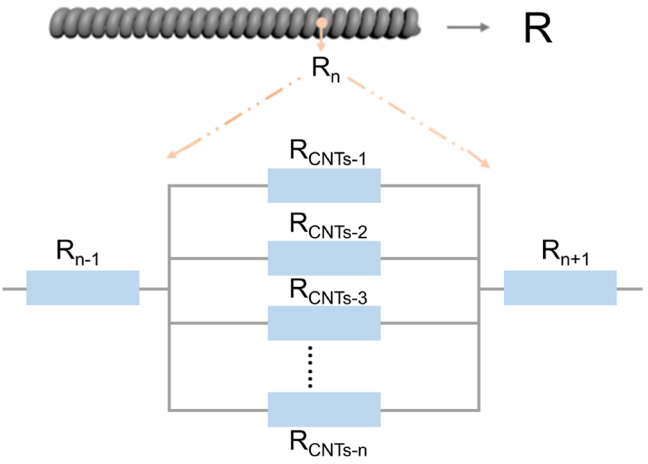

Figure 25. Working mechanism of a resistive strain sensor. The simulative circuit scheme showing the circuit of the CNT-conductive network consists of numerous resistors in series. Reprinted with permission from ref 454. Copyright 2020 American Chemical Society.

experience an extensive sensing range up to 500%, with a positive gauge factor of 0.13% and high cyclic stretching/releasing stability. Building a strain sensor with a hierarchical structure change is a simple method with large-scale production potential.[455,456] However, the low sensing range and sensitivity limit their applications.

Another resistive fiber sensor consists of an elastic fiber core and conductive coating materials.[456] Adopted from planar strain sensors, this design offers higher stretchability and sensitivity than the bare coiled conductive electrodes because the elastic substrate allows a higher degree of mechanical deformation and the conducting network is more susceptible to the external strain. A stretchable fiber strain sensor with controllable sensitivity was developed using commercial yarns as a core electrode and graphene nanoplatelets/PVA as a piezoresistive layer. The as-prepared fiber strain sensor showed high stretchability up to 150% and could detect diverse human motions such as arm bending or chewing.[457]

Another fiber strain sensor consists of a highly elastic polyurethane core fiber with a helically winding polyester fiber modified by a graphene oxide sheet; it achieved high sensitivity to tensile strain deformation (detection limit of 0.2% strain) and an extensive sensing range (up to 100% strain).[437] The resistance variation of the fiber device mainly depends on the gap numbers produced by the winding numbers of polyester fibers. With deliberate design, the performance of the fiber strain sensor could be further improved. For example, a fiber strain sensor with high sensing range (200% strain) and detection limit (0.01% strain) was developed using an elastic rubber fiber core and CNT coating. Helical gaps in micrometers and nanometers of the CNT/polymeric fiber composite formed on the electrode surface produced larger structure changes under small strains and contributed to the excellent sensing performance.[458]

It is more difficult to fabricate the fiber capacitive strain sensor due to the complex structure of two electrodes and one dielectric layer. The fiber capacitive sensor has lower hysteresis and better long-term cyclic stability because the capacitance change is generally more reversible than the resistance mechanism.[445] A highly stretchable fiber strain sensor was developed by wrapping CNT sheets oriented in the fiber direction on stretched rubber fiber cores. This buckling

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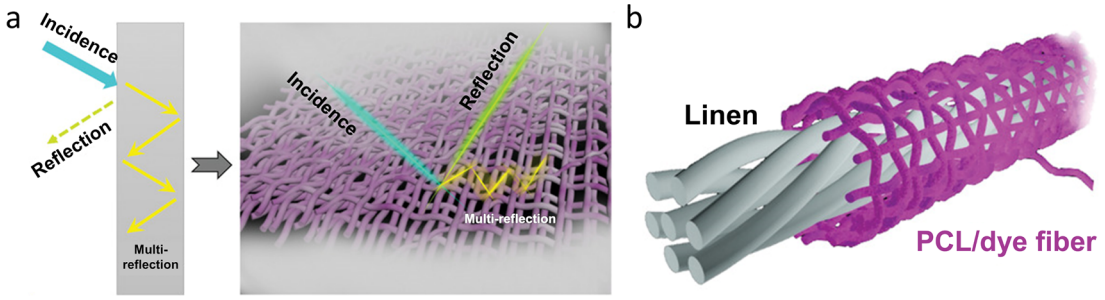

Figure 26. (a) Schematic of the ultraviolet-sensing fabric enabling multireflection via the fiber network. (b) Schematic of a linen core−PCL sheath yarn. Reprinted with permission from ref 472. Copyright 2020 Wiley-VCH.

structure enabled 860% capacitance change upon applied 950% strain, while the resistance varied by <5%.[459] Owing to the flexible fiber core, such a fiber sensor had a linear response and high cyclic stretching−releasing stability over 500 cycles at 950% strain. Later, another fiber strain sensor was produced by sandwiching a rubber fiber core between two buckled functionalized CNT electrodes. This sensor design gave the sensor high structural, mechanical, and electrical stabilities against bending, stretching, and twisting deformations, with 115.7% capacitive changes upon 200% stretching.[460] The weavability and excellent electrochemical stability (over 95% performance retention under stretching and twisting) made the fiber device promising in wearable strain sensors. On the other hand, natural fibers are biocompatible materials with low costs for human-interfaced sensors because of their natural origin. In this regard, a variety of natural fiber-based sensors, including active-material-modified cotton fibers or carbonized silk fabrics, can be explored as wearable strain sensors.[77,461,462] For example, carbonized silk fabrics can offer a wide sensing range (from 0 to >500% strain), rapid response (<70 ms), and high durability (10 000 cycles), showing potentials as mass-produced sensors due to the availability of natural silk materials.[77]

Strain sensors could monitor the deformation of organs and provide vital information on the health status of the user. Many pioneering studies have demonstrated the in vivo applications in cardiac and orthopedic systems.[463,464] Fiber strain sensors are more easily implanted and form stable interfaces with tissues, leading to more significant applications as implantable devices. Recently, an implantable fiber strain sensor was built with two double-helical stretchable conductive fibers and a protection layer of Ecoflex coating.[465] The fiber sensor had a sensing range of 15.0−27.5% and a sensitivity of ∼12, and its performance could be modulated by adjusting the double-helical structure. The sensor was implanted in the knee of a porcine leg to monitor strains on the patella ligament and remained stable for 3 weeks without causing severe inflammation or fibrosis.

Over the past few decades, fiber strain sensors have advanced dramatically, but some problems remain. One major challenge is device stability. As human motions usually occur repeatedly, the stability of strain sensors is important to obtain accurate and reliable signals. The existing fiber strain sensors can only endure thousands of repeating motions, while long-term applications have rarely been reported, possibly due to the fragile mechanical and electrochemical performance of electrodes in complicated environments in vivo. Searching for flexible materials and designing bendable structures could help solve this problem. The second challenge is the safety concern of such wearable and implantable devices. As the sensors need to contact our bodies closely, the unsafe encapsulating layer and active materials exposure after the mechanical failure of these devices could harm the users. Third, the large-scale production of fiber strain sensors with ideal performances has not yet been achieved. Although the large-scale fabrication could be achieved by directly modifying commercial textiles or depositing active materials on carbonized cotton fabric, obtaining high-performance textile sensors with a cost-effective strategy remains to be developed.[466,467]

7.1.2. Fiber Ultraviolet Sensors. Ultraviolet light is known to associate with skin cancer.[468,469] A major ultraviolet exposure source for humans is the sun; thus, a wearable ultraviolet sensor may help to alarm the user of ultraviolet exposure damage in daily life.[470,471] An ultraviolet sensor usually operates based on photoelectric, resistance changes, or photochromic effects.[472] According to these mechanism, various fiber ultraviolet sensors have been developed. For example, a fiber ultraviolet sensor was developed by wrapping a CNT fiber on a p-CuZnS/n-TiO_2-modified Ti wire electrode. The photogenerated electrons and holes were collected separately by the Ti and CNT electrodes, respectively, generating a photocurrent of 13 mA. The device design also endows a fast response time with the rise and decay times being <0.2 s at 0 V under 320 nm ultraviolet light. The fiber sensor was also flexible enough to be integrated into wearable devices for real-time ultraviolet sensing.[473] Another wearable fiber ultraviolet sensor used ultraviolet-sensitive single-walled CNT-coated cotton thread as a detection unit. The adsorbed O_2 molecules detached from the CNT surface upon exposure to ultraviolet radiation, leading to the resistance change of the device (4.76% and 0.76% per mW/cm^2 for 254 and 365 nm ultraviolet light, respectively).[470]

Although fiber sensors based on photoelectric effect or resistance change are reliable in many scenarios, it remains challenging to distinguish the electric signal change from other products, such as motion-induced resistance changes in connecting networks. Fiber ultraviolet sensors using a photochromic effect could avoid this problem because the detection results could be directly observed by the naked eye or image-analysis software.[474] For example, such a concept could be achieved by doping organic dyes in the electrospinning of polycaprolactone to produce photochromic fibers (Figure 26). The ultraviolet-detecting fibers could be coated on commercial linen fabric and woven into different patterns as wearable ultraviolet detectors.[472]

7.1.3. Fiber Electrophysiology Sensors. Electrophysiology reveals the activity of living cells and the working mechanism of neural circuits, which is a fundamental tool in

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neuroscience study.[475−478] Two major types of electrodes, Utah electrode (planar structure) and Michigan electrode (fibrous structure), have been developed to monitor electrophysiology signals in living animals.[479,480] The design of electrophysiology electrodes is simple, and the most important influential factors of the electrode performance are the electrode size and impedance. The larger surface exposition on the electrode tip results in lower electrode impedance and activity detection from many neurons at a relatively larger scale. A fiber sensor with a smaller exposed electrode surface was suitable for identifying the function of single cells. Usually, the impedance of electrophysiology electrodes falls into the range 5−20 MΩ when measured by an alternating current at 50 Hz.[481]

Fiber electrodes have particular advantages as electrophysiology electrodes. First, their small diameter allows them to enter a deeper region in the brain than those planar devices (limited to the cortex). Second, the mechanical advantage of fiber sensors plays an important role, especially in chronic studies because they could better adapt to the constant movement of target animals and provide a stable tissue/device interface. Early fiber electrophysiology electrodes used thin metal wires as recording electrodes.[482] With the fast development of fiber materials, a new family of neural electrodes has been proposed during the past two decades, including carbon fibers, CNT fibers, graphene fibers, and conducting polymers.[21,22,483−485] For example, carbon fiber microelectrodes have been widely used to record extracellular/intracellular neural activity. However, they fail to operate in large primates chromatically because they need to be encapsulated in fused silica tubes or glass capillaries, which increase the size and moduli of the device.[486] To solve this problem, an integrated composite electrode with a carbon fiber core electrode, poly(*p*-xylylene)-based thin-film dielectric barrier, and poly(thiophene)-based recording site was developed.[487] The composite electrode could record neural signals in the rat brain stably for 5 weeks, showing improved mechanical compliance. Other than carbon fibers, the other fiber materials, such as CNT fiber, graphene fiber, and polymer electrodes, have been reported to record neural signals with improved tissue compatibility and signal-to-noise ratios.[21,23,24,483,488]

While many efforts have been devoted to improving the tissue compatibility and performance of neural electrodes for long-term detection, another vital research trend emerged recently by expanding the functions and capabilities of neural electrodes, also enabled by the rapid development of new fiber materials.[489] For example, it is important to simultaneously monitor the neural activity in multiple sites because it may help to reveal the working mechanism of neural circuits.[25,490,491] Traditionally, it is achieved by a Utah array covering a large surface. However, the Utah array is mainly used in the cortex because it is difficult to implant them into a deeper region. Fiber probes thus have been discovered with advanced fabrication methods such as photolithography with multiple recording sites incorporated into the electrode tip (with a diameter of a few micrometers). Recently, a multichannel neural probe, named Neurobasal, was developed via a thermal drawing of molten polyethylene glycol and a subsequent electrocapillary self-assembly process.[492] A 16-channel Neurobasal could record 121 neurons in the mouse brain and was ready to integrate with optical fibers for optogenetic stimulation. Due to its small size and high flexibility, such a device could build chronically stable interfaces with the nervous systems and record neuron signals in mice for 6 weeks.

CNT fibers and polymers are nonferromagnetic and thus compatible with magnetic resonance imaging. The CNT fiber integrated with magnetic resonance imaging provides basic and applied neuroscience studies.[23] Besides, multiple-function neural fiber devices could be fabricated through the thermal drawing of polymers, in which multiple channels were produced with different functions (Figure 27a−c).[41] The soft

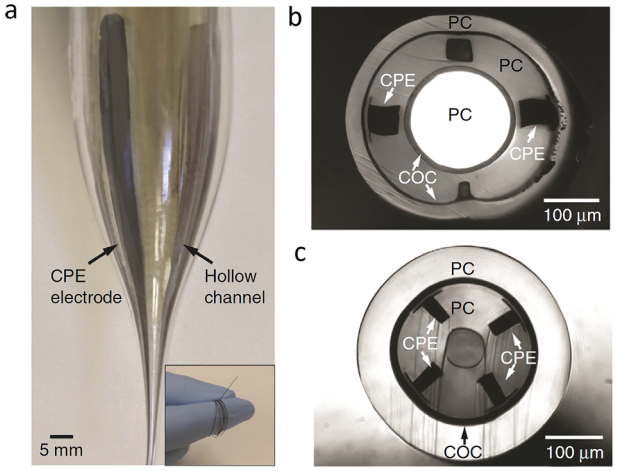

Figure 27. (a) Photograph of the thermal-drawing process of multimodality fiber electrodes. (Inset) Photograph of the drawn fiber wrapped around a finger. (b, c) Cross-sectional optical images of two different multimodality probe tips. Reprinted with permission from ref 41. Copyright 2015 Springer Nature.

fiber device could form stable brain−machine interfaces in mice for 2 months and simultaneously perform electrophysiological recording, optogenetic stimulation, and drug delivery.

However, the soft fiber probes were hard to be directly implanted into the brain due to their low bending stiffness, and rigid materials are required to assist the implantation. The auxiliary method can cause additional tissue damage and chronic interface recovery.[493,494] To solve this problem, a modulus-alterable fiber neural probe was developed. The fiber device consisted of a CNT fiber as a core electrode and calcium cross-linked sodium alginate as a sheath layer. The elastic moduli of the probe changed from ∼10 GPa to ∼10 kPa after implantation due to the calcium cross-linked sodium alginate layer to absorb water, which softened with elastic moduli close to brain tissue. The modulus-alterable fiber eliminated the requirement of assisting materials and could monitor neuron signals stably for 4 weeks in vivo.[495] Another challenge is that the neural electrodes were usually limited to the signal site, whereas the study of neuroscience requires the detection of neuron activity in multiple sites. A spatially expandable fiber-based probe was designed to solve this problem. To disperse the fiber sensors in different sites of the brain region via single implantation, a helical scaffolding fiber was developed (Figure 28).[496] Many functional fiber electrodes (e.g., optogenetic stimulation, electrical recording, and drug delivery) could be inserted in the helical scaffold and implanted into the brain. Then a further extruding process could be applied to position the multifunctional fiber arrays through the scaffold by a calculated length until they reached their targeted locations. This work demonstrated the ability for

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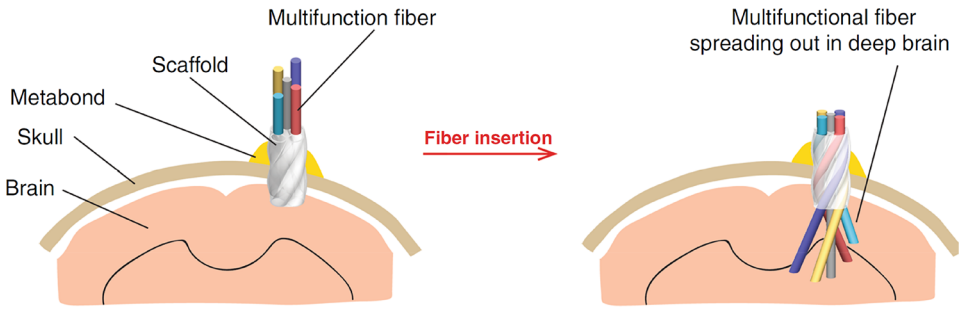

Figure 28. Schematic of the implantation process of the spatially expandable fiber electrodes. The scaffold is first inserted into the brain, and the functional fiber probes are further inserted into the brain tissue through the helical scaffold and reach the different regions in the deep brain. Reprinted with permission from ref 496. Copyright 2020 Springer Nature.

three-dimensional manipulation of implanted fiber electrodes in vivo and achieved mapping of brain activities across distant regions in the deep brain.

7.2. Fiber Chemical Sensors

Human activity is a sum of various chemical processes.[497,498] Constant and precise monitoring of the chemical changes in the human body is vital for disease therapy and health management. For example, a glucose sensor is indispensable for diabetic patients, and the market size of the global health expenditure on diabetes is expected to reach US$825 billion by 2030.[499] Besides glucose, other vital chemicals such as dopamine, cardiac troponin, and vitamins can reveal the human health condition. Thus, many fiber chemical sensors have been developed to interface with different human tissues to provide real-time information.[428] According to the working mechanism, fiber chemical sensors generally include electrochemical and nonelectrochemical sensors.

7.2.1. Fiber Electrochemical Sensors. An electrochemical sensor is an important research field bridging materials science and analytical chemistry that has been utilized widely from laboratory analysis to real-time health management.[500] With proper device design, the electrochemical detection method is fast, cheap, reliable, highly sensitive, and selective. There are many excellent comprehensive reviews on electrochemical sensors.[2,501] Briefly, two critical steps are involved in the operating process of an electrochemical sensor. First, the target chemicals are converted to electric signals at the active sites on the electrodes. Then, the generated charges can be transferred to the electrodes and detected by the back-end instruments.[502] Therefore, a fiber sensor needs to meet a few basic requirements, including being (1) biocompatible with skin or tissue without harming human users; (2) highly conductive and chemically stable; (3) flexible, miniaturizable, and conformable as wearable or implantable devices; and (4) easily modified with active/functional materials.[503,504]

Fiber electrodes have helped to develop electrochemical sensors for decades due to the low-cost, broad sources, and easy functionalization.[505] The most widely used fiber electrodes for electrochemical sensors include metal wires, polymer fibers, and carbon fibers.[20] As electrochemical devices emerged toward the wearable and implantable platform, as demanded by personal health care management, many human-interfaced fiber electrochemical sensors were developed in the past decade.[500]

Fiber Electrochemical Sensors for Textile Systems. Wearable fiber electrochemical sensors mainly target human sweat analysis. Sweat contains many important chemicals, including hormones, electrolytes, and glucose, and it is readily available for wearable devices.[506] For instance, a wearable electrochemical fiber sensor was developed using CNT fiber electrodes modified with sensing components, as shown in Figure 29. These functional layers include an enzyme, ion-to-

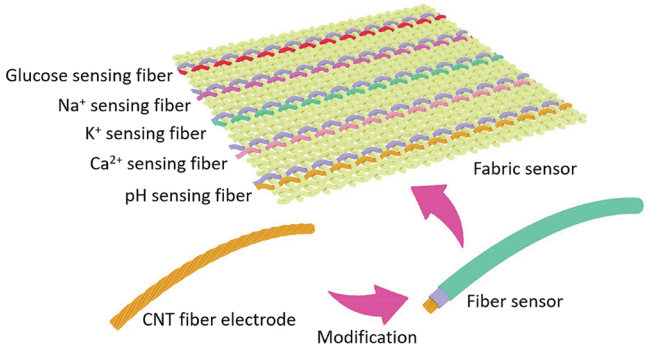

Figure 29. Schematic illustration of the designed wearable fabric sensor, using functionalized CNT fiber as sensor units. Multiple sensors could be fabricated by modifying the electrode with different active materials. Reprinted with permission from ref 507. Copyright 2018 Wiley-VCH.

electron transducer, and conjugated polymer, which were sensitive to metabolites, ions, and pH, respectively.[507] The obtained fiber sensors were woven into a fabric as a real-time sweat-analysis platform, showing promise for intelligent health care management. In addition, another wearable chemical sensor was constructed using stretchable gold fibers fabricated by coating an elastomer-bonded gold nanowire on polymeric substrates (such as polyaniline). The as-prepared fiber sensor could be stretched by 100% and sense pH changes with a sensitivity of 60.6 mV/pH.[508] Recently, a fiber cortisol sensor capable of detecting sweat cortisol was proposed. The anticortisol antibody was immobilized on a ZnO-modified carbon fiber electrode. The recognition layer of the fiber surface could identify cortisol levels as low as 0.098 fg/mL in the sweat sample and had a broad linear detection range from 1 fg/mL to 1 μg/mL, with a sensitivity of 2.12 μA/g/mL.[509]

However, the research of wearable fiber electrochemical sensors is still at an early stage, as some challenges are yet to be solved.[503] First, electrochemical sensors are sensitive to surface area change, which may cause fluctuating electrochemical signals. This problem may be solved by carefully designing the fiber electrode, such as exposing a particular surface area on the fiber electrode and insulating the rest of the area to avoid the

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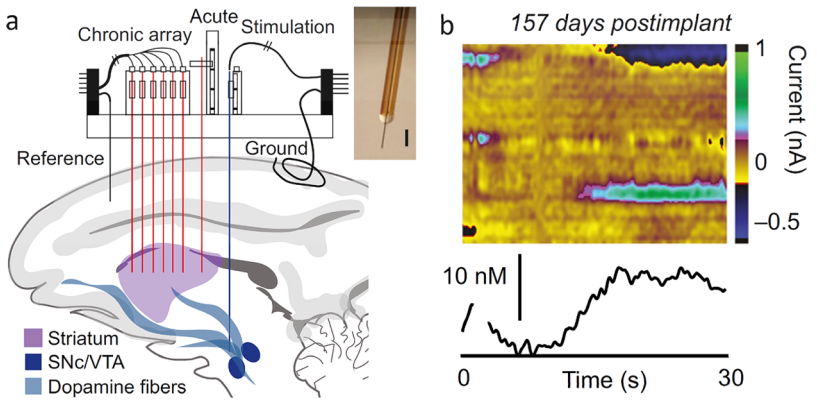

Figure 30. (a) Schematic illustration of the implantable fiber sensor for chronic dopamine detection. The inset shows the optical image of an encapsulated fiber sensor before implantation. (b) Detected signals of an implanted fiber sensor after 157 days via the fast-scan cyclic voltammetry method. Reprinted with permission from ref 515. Copyright 2017 National Academy of Sciences.

formation of a new surface. A recently developed stretchable, strain-insensitive fiber electrode might help to provide insight into a solution for this problem.[510] However, such a problem might not affect sensors based on a potentiometric detection mechanism, which is less sensitive to the area change of the electrode surface.[511] Second, in the wearable system, the stability of the human–device interface is difficult. The fiber/textile may detach from skin constantly, causing the changing volume of the detected sweat sample. This challenge could be addressed by incorporating microfluidic channels with fiber/textile sensors.

Fiber Electrochemical Sensors Implanted into Bodies. Compared to fiber electrochemical sensors for textile systems, implantable fiber electrochemical sensors are poised to provide more accurate chemical information from the deep tissue. For example, for real-time analysis of neurotransmitters in neuroscience, fiber sensors have been extensively applied in animals such as rodents and primates for decades.[512] For example, carbon fibers have been widely adopted to analyze the concentration of neurotransmitters in the brain, usually coupled with fast-scan cyclic voltammetry.[513]

However, most implantable fiber sensors focus on the sensitive detection of chemicals, while chronic monitoring is more important in some applications, such as diabetes management. It is difficult to be realized because of biofouling (the adsorption of macromolecules and cells on the electrode surface), inflammation, and scarring around the device, which slows the mass transfer between the target medium and the sensor surface.[514] Further, the instability of the functional layers, such as enzyme inactivation and mediator leakage, brings additional difficulties.

To solve these problems, some insights have been realized with the fast-scan cyclic voltammetry detection method, which is less susceptible to the area change of the sensor surface. For instance, a long-term dopamine-monitoring system in the brain was developed with carbon fiber electrodes via fast-scan cyclic voltammetry. The carbon fiber electrodes were integrated into a modular platform to monitor the dopamine fluctuations in the striatum in real time, as shown in Figure 30.[515] Because of the ultrasmall size and the stability of the carbon fiber electrode (7-μm tip diameter), the sensor was found to work for >100 days in vivo.

However, many electrochemical sensors (e.g., amperometry sensors) are vulnerable to the surface area change. To avoid unwanted inflammation and scarring around the sensor surface, another effective way is to design flexible fiber sensors with mechanical properties that are compatible with soft tissues. Specifically, the implanted device needs to bear the continuous movements of animals to reduce the relative movement between the tissue and the device. Recent work demonstrated that, by assembly of CNT bundles into fibers, the fiber sensor could adapt to the range of bending stiffnesses for most tissues, such as muscles and blood vessels, as shown in Figure 31a and b.[50] This allows the fiber sensor to be implanted into living animals with negligible immunoreactions, thus enabling them to perform stably for the long term in vivo. As a result, the fiber sensors detected calcium ions and glucose in the venous blood of cats for 28 days. In addition, by adjusting the structure of multiple sensor arrays, the fiber sensor could provide spatial resolutions from different sites, as

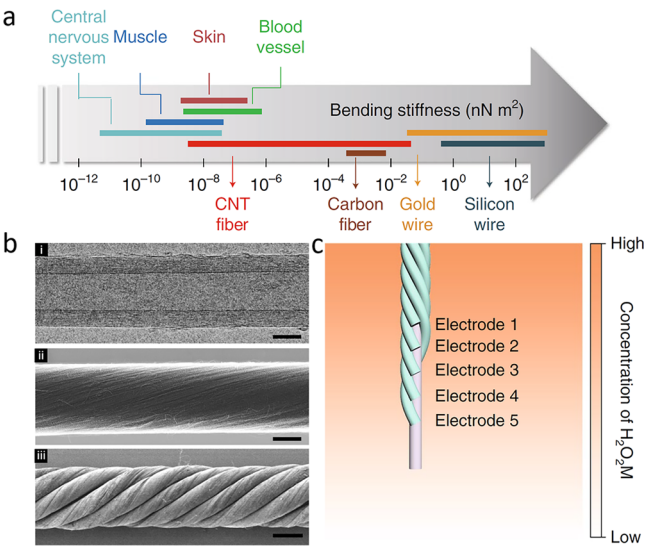

Figure 31. (a) Schematic showing the bending stiffnesses of assembled CNT fibers that could match a range of soft tissues. (b) Transmission electron microscopy image of multiwalled CNT ((i) scale bar, 3 nm), SEM images of the primary CNT fiber ((ii) scale bar, 6 μm), and assembled hierarchically helical CNT fiber ((iii) scale bar, 20 μm). (c) Schematic illustration of five fiber sensors assembled for analysis in an H_2O_2 concentration-gradient environment. Reprinted with permission from ref 50. Copyright 2020 Springer Nature.

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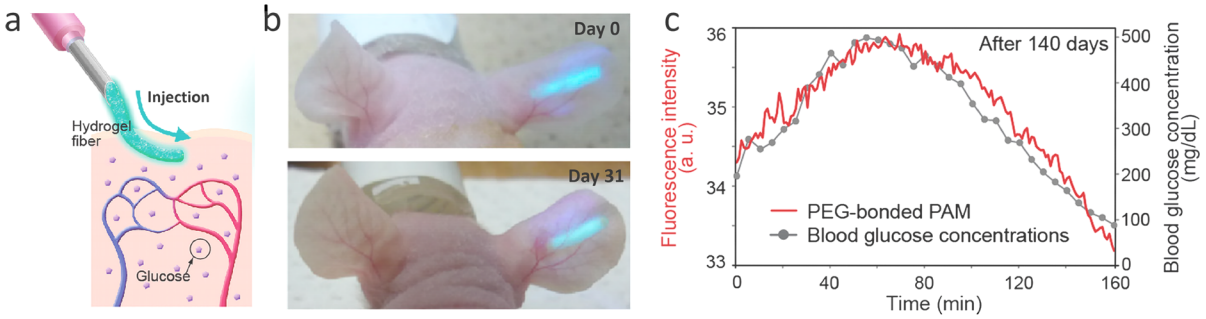

Figure 32. (a) Schematic illustration of the injection process of a glucose-sensing fluorescent fiber. (b) Glucose sensor emitting light after implantation at 0 and 31 days. (c) Detected glucose concentrations after being implanted for 140 days, compared to the blood glucose concentrations. Reprinted with permission from ref 26. Copyright 2011 National Academy of Sciences.

shown in Figure 31c. Such a design may provide an insight for the development of multiple-function sensors.

Although implantable fiber electrochemical sensors have been rapidly developed during the past decade, there is still a long way to go before they can be readily applied in our daily life. For example, there is always room to further improve the sensor safety, selectivity, sensitivity, and long-term operational stability and lower the cost.[503] On the other hand, we also observed an emerging trend of using implantable fiber electrochemical sensors with other research techniques. For example, the implantable fiber electrochemical sensors could be coupled with optogenetics or drug delivery for further comprehensive studies on neuron functions.[516] We believe many efforts will be continuously devoted to this direction.

7.2.2. Other Fiber Chemical Sensors. Besides electrical signals, the transformation of optical signals from chemicals is another widely accepted mechanism for fiber chemical sensors. Using optical signals has several advantages, e.g., optical signals can be observed by the naked eye, making it feasible to obtain the detected result. A fluorescence hydrogel fiber glucose sensor was developed using this mechanism. The fiber glucose sensor was composed of polyethylene glycol-bonded polyacrylamide with high biocompatibility, and then a glucose-responsive fluorescent monomer was immobilized in the hydrogel. The glucose-responsive fluorescent fiber could be injected into the targeted area and excited by ultraviolet light, as shown in Figure 32a and b, where the fluorescent intensity corresponds to the glucose concentration. The soft fiber sensor could continuously glow in response to blood glucose for up to 140 days, as demonstrated in Figure 32c.[26]

7.3. Other Fiber Bioelectronics

Besides sensing, fiber bioelectronics also have applications such as drug delivery, optogenetics, neural modulation, and disease therapy.[517] The small size and mechanical flexibility allow them to be easily implanted into the target site and work stably at long term. For example, a fiber device integrated with recording electrodes, optical waveguides, and microfluidic channels was implanted into the target site in one step and performed stably for 3 months.[46] Another example was a drug-delivery fiber, which could be used for neuron modulation by in situ electrochemical reactions and was stable for 2 months in vivo.[49] In addition, implanted fiber electrodes could also be utilized to treat tumors via toxic chemicals generated through in situ electrochemical reactions, showing the promise for simple, low-cost disease treatments. The functions of fiber bioelectronics could be readily expanded according to the demand. Thanks to the stable interface formed with human tissue, implantable fiber devices will continuously find applications in various disease models and fuel the fundamentals of medical research.

8. OTHER FIBER DEVICES

8.1. Fiber Artificial Muscles

Muscle tissue is composited with multiple hierarchically assembled fiber bundles and is responsible for movements; almost all movements in the body are the results of muscle contractions.[518] Recently, there has been growing interest in mimicking muscle-like motion using synthetic fiber materials. The resulting devices, named artificial muscles, could find applications in soft robotics, prosthesis, sensors, and smart textiles.[519,520] Responsive polymers, liquid metal, and carbon materials have been successfully used to construct artificial muscles.[519] The fibers can be twisted or coiled to store mechanical energy, and the multiple scale gaps on the fiber surface can go through changes upon exposure to stimuli, including temperature, pressure, solvent, electric current, or biofuel, leading to contraction, expansion, or torsional motion depending on the fiber structure.[96,150,521−523] For example, an artificial torsional muscle composited with a twisted CNT fiber could provide a 15 000° rotation and 590 rpm due to the charge injection into an electrochemical double layer on the fiber surface. The motion could be highly reversible if both fiber ends were tethered to prohibit end rotation.[524] Recently, a new application in smart textiles for dynamic temperature and humidity regulation had been proposed based on responsive fiber deformations. A silk yarn was fabricated by twisting electrospinning aligned silk nanofibers, which rotated upon contact with sweat and rotated in reverse without the sweat. The deformation of the sweat-responsive fiber enabled dynamic regulation of moisture and heat on the skin, enabling it to work as a self-regulated air-conditioning textile.[525]

However, instead of giving a detailed development history of fiber artificial muscles, as many excellent reviews have already covered,[140,526,527] we here mainly discuss how artificial muscle research affects the development of various other fiber devices, which has rarely been covered previously. The key for artificial muscles is using stimuli to produce considerable deformations of fiber materials, and the primary research goal is to use less energy to make as much mechanical deformation as possible (toward higher energy-conversion efficiency) and to achieve different motions via simple and robust design of a fiber structure. However, the fiber electrode structures for artificial muscles are also prevalent for fiber devices (e.g., twisted multiple fiber bundles). Even with decent encapsulating and

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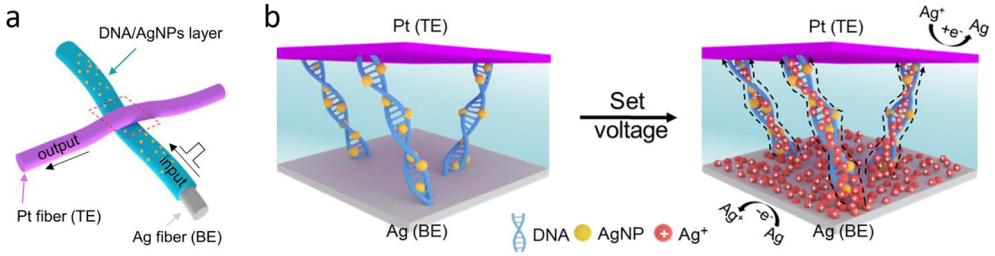

Figure 33. (a) Scheme showing the design of a DNA-bridged fiber memristor. The memristor unit is formed upon interlacing with a Pt fiber electrode. (b) Mechanism of the memristive switching process. Ag ions use DNA bridged ionic pathways to form conductive filaments. Reprinted with permission from ref 544. Copyright 2020 Wiley.

packing, the fiber electrodes of these devices are directly exposed to stimuli such as thermal changes, biofluids (containing water and electrolyte), or electrochemical stimulations (less than a few volts). This raises three concerns. First, for the fiber devices (especially for fiber sensors), will the microstructure of the fiber electrode change under these stimuli? If it does, how will this deformation affect their performance? For example, the active material layers on chemical sensors, such as enzymes and mediators, have a risk of detaching or leaking from the electrode surface upon deformation. Second, the newly generated electrode surface may contribute to an increased faradaic/nonfaradaic current, causing the need for more frequent calibrations for biosensors.[512] Our third concern is more specific for the implantable sensors. Artificial muscles are highly efficient, and they could have an energy density >50 times that of skeletal muscles and generate significant motions and forces.[528] As implantable fiber sensors, if the device could generate motion under stimuli, it is not likely to be reversible for motions, and the exposed electrode surface is limited (a hundred micrometers to a few millimeters). However, will this process cause additional damages to surrounding tissues? More research should be devoted to understanding how fiber devices behave upon receiving environmental stimuli. Also, it should be noted that other fiber devices, such as energy-harvesting and -storing devices, may also face the same problem. Fiber sensors are expected to give precise signals, and they are thus more vulnerable to the electrode surface and microstructure changes.[512,529] After all, the research on artificial muscles sheds light on how a fiber material is responsive with different structures for various stimuli. It could benefit the development of robotics and may help optimize the performance of fiber devices.

8.2. Fiber Transistors and Memristors

As we enter the Internet of Things age, textile-based electronics are expected to have energy-management, computing, and display functions. The transistors are basic units for modern electronics, so developing fiber transistors is vital for wearable electronic devices.[530] As a result, fiber organic electrochemical transistors (OECTs) have drawn attention because of their high mechanical flexibility, simple chemical synthesis, and easy processing, making them ready to be integrated into wearable devices.[531−533] OECTs use the electrolyte medium between the channel and the gate electrode to complete the electrochemical doping/dedoping of the polymer to modulate the channel current.[534] The early fiber OECT was designed using two crossing polymer-coated fibers as substrates, with an electrolyte contact at the junction of the fibers. Such a design allowed the fiber OECT to demonstrate an on/off ratio >1000 with low operating gate voltages between 0 and 1.5 V. The fiber OECTs could be woven into fabric due to the flexibility of the fiber electrode and simplicity of the device structure and integrated as multiplexers for digital logic, showing promises for electronic textiles.[535] Later, the fiber OECTs were explored as biosensors because of their in situ amplification.[536] This characteristic offered the fiber OECTs high sensitivity as biosensors. For example, by using a poly(3,4-ethylenedioxythiophene)/(poly(sodium-*p*-styrenesulfonate)-functionalized cotton fiber as the source and drain electrode, a silver fiber as the gate electrode, and saline solution as the electrolyte, the fiber OECT could detect the change of the saline concentration at the physiological level by showing varying source-drain currents under 0.2−0.4 V gate voltages.[537]

Further research found that the fiber OECTs were easily integrated into wearable systems as biosensors due to their high flexibility and sensitivity. By coating metal and a poly(3,4-ethylenedioxythiophene) multilayer on nylon fiber electrodes, the OECTs maintained stable performance during bending, validating their application potentials in wearable platforms. The fiber OECT could detect glucose, dopamine, or uric acid depending on the modification layers of active materials (e.g., glucose oxidase or uricase−graphene oxide for glucose and uric acid, respectively). Thanks to it being susceptible to different chemicals in the electrolyte, the fiber OECT could achieve a detection limit of $3-10 \times 10^{-8}$ M.[531] More importantly, long-term chemical detection in vivo is difficult because the weak output signals could easily be affected by the unstable device−tissue interface. Hence, a soft implantable fiber OECT was developed using a CNT fiber, which showed matched mechanical properties with soft tissues. The fiber OECT could detect dopamine for 7 days when implanted in the mouse brain, thanks to the amplification effect of the OECT and the excellent mechanical properties.[538]

Because of their unique responsive nature to the changes in the electrolyte, the fiber OECTs could be used as either logic circuits or biosensors. The study of the fiber OECT is still at an early age, and more work may be devoted to improving their performance (such as increasing the signal output, lowering the operation voltage, improving the selectivity as biosensors, and enhancing the stability in real-life scenarios) and expanding their functions by integrating them with other fiber devices.

Memristors were theoretically predicted in 1971, as they may switch resistances upon applying voltage, and they have potentials in storage technology, logic circuits, and neuromorphic systems.[539] Memristors could be fabricated with various functional materials, including metal oxides, chalcogenides, and organic materials.[540−542] The resistive switching of memristors may be attributed to the formation/rupture cycles under the driving electric field.[543] However, most memristors

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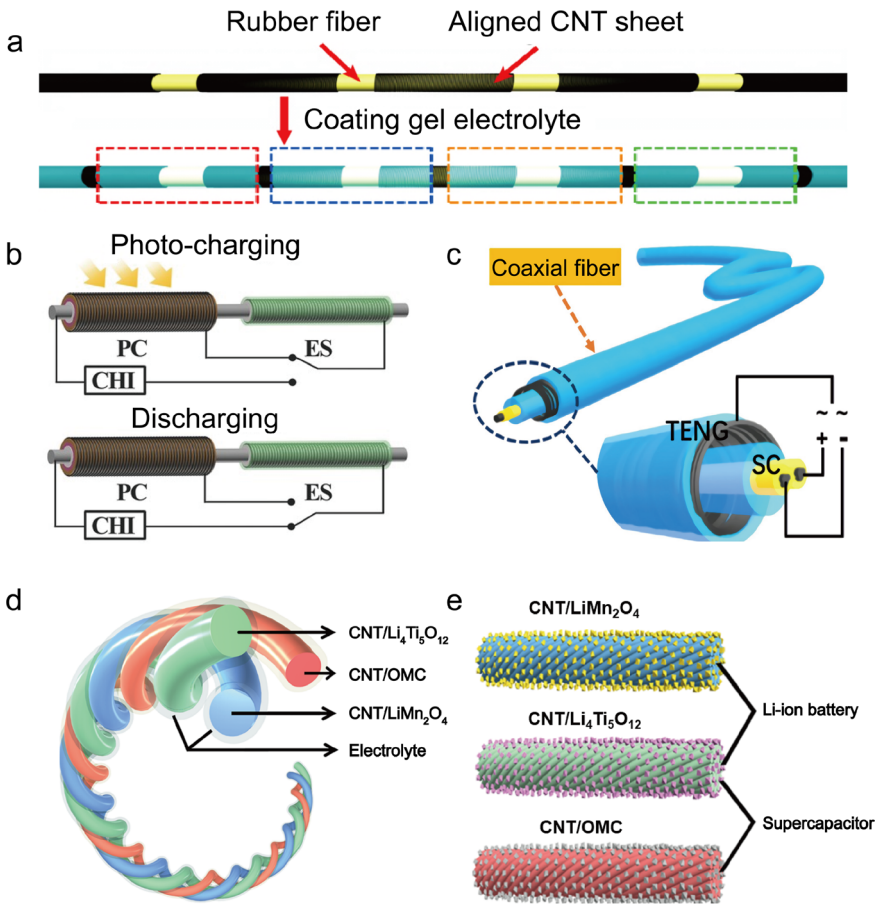

Figure 34. (a) Schematic illustration of the connected supercapacitors in series on a single fiber electrode. Reprinted with permission from ref 550. Copyright 2016 Wiley-VCH. (b) Design of the circuit connection state in the process of charging and discharging. PC and ES represent photovoltaic conversion and energy-storage module, respectively. Reprinted with permission from ref 341. Copyright 2014 Wiley-VCH. (c) Coaxially integrated energy fiber, consisting of inner fiber supercapacitor and outer TENG. Reprinted with permission from ref 342. Copyright 2018 American Chemical Society. (d, e) Schematic illustration of the structure of the twisted integrated fiber Li-ion battery and supercapacitor, respectively. The CNT/$Li_4Ti_5O_{12}$ and CNT/$LiMn_2O_4$ electrodes function as a Li-ion battery, while the CNT/ordered mesoporous carbon and CNT/$Li_4Ti_5O_{12}$ electrodes function as a supercapacitor. Reprinted with permission from ref 40. Copyright 2015 Wiley-VCH.

suffered from low switching speed, high operation voltage, and poor stability due to the lack of proper building molecules to ensure the effective formation/rupture cycles of conductive filaments. To this end, a fiber memristor was made from DNA incorporated with Ag nanoparticles as the active layer; the helical structure and the orientated DNA molecules could offer ideal conductive filaments for ion transport, which altered the resistance of the active layer (Figure 33). By interlacing the Ag fiber with a Pt fiber, the memristor could offer a low operation voltage (0.3 V), low power consumption (100 pW), and high switching speed of 20 ns. By further integrating with power-supplying and light-emitting modules, the device could function as a fabric information-processing system.[544]

9. INTEGRATION OF FIBER DEVICES

The power output provided by a single fiber energy-harvesting/storing device is limited, and the integration of multiple fiber devices offers them the potential to complete tasks that could not be achieved by a single fiber device.[133,545−547] On the other hand, fiber devices with different functions may be integrated. For example, the energy-harvesting/storing fiber devices may directly power various fiber electronic devices, such as sensors or displays, toward truly wearable self-powered systems.[143,240,548] After all, the integrated fiber devices may better satisfy real-life applications as they can provide the advantages of different functional units.

9.1. In-Serial Connection

The in-serial connection of the same type of fiber devices could boost their performances, which represents a practical solution to integrate fiber energy-harvesting/storing devices, such as fiber solar cells, supercapacitors, and batteries, to provide higher output voltages.[331,391,549]

For example, the output voltages of a single fiber supercapacitor using water-based electrolytes are limited by the thermodynamic decomposition potential of water (~1.23 V). By mimicking the in-serial assembly of electrolytes in electric eels, shown in Figure 34a, the integrated fiber supercapacitors could result in output voltages as high as 1000 V.[550]

On the other hand, the in-serial connection of different types of fiber devices could also obtain an improved performance. For instance, the fiber energy harvesters, such as solar cells and nanogenerators, are often integrated with energy-storage devices on a single fiber electrode. The in-serial connection of two types of functional units allows the efficient harvesting of energy when the device is exposed to a proper stimulus, as well as storage of the excessive energy for further use.[133,341]

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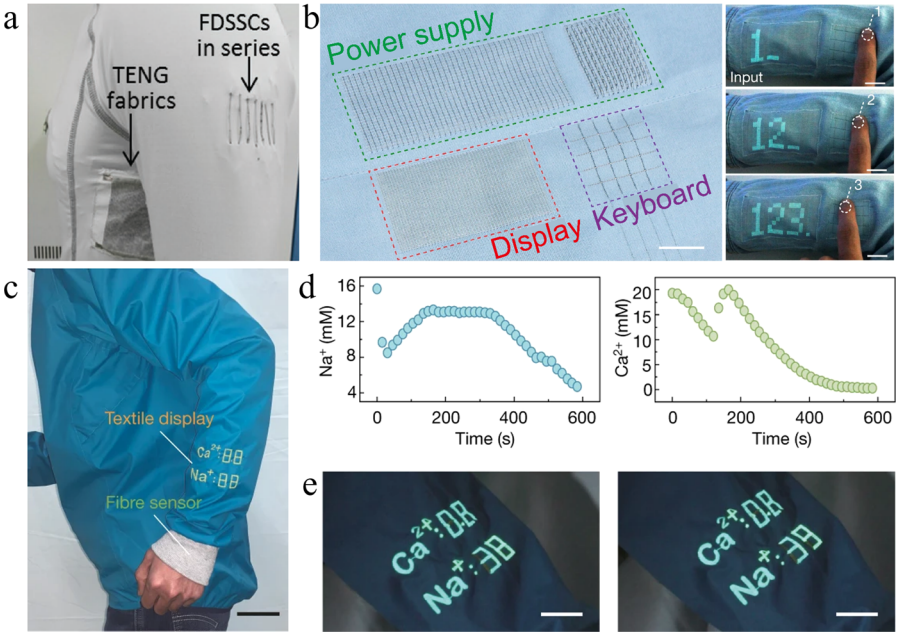

Figure 35. (a) Photograph of an integrated energy-harvesting textile with TENG fabrics underneath the arm and seven fiber DSSCs in series on the shoulder. Reprinted with permission from ref 558. Copyright 2016 Wiley-VCH. (b) Photograph of an integrated textile system consisting of display, power supply, and information input units. The inputted information via the keyboard could be displayed by the textile. Scale bars, 2 cm. Reprinted with permission from ref 52. Copyright 2021 Springer Nature. (c) Photographs of an integrated textile system consisting of textile display and sensors, powered by a fiber Li-ion battery (not shown). Scale bar, 10 cm. (d, e) Fiber sensors detecting Ca^{2+} and Na^{+} in sweat and displaying the results via the electroluminescent module, respectively. Scale bars, 2 cm. Reprinted with permission from ref 53. Copyright 2021 Springer Nature.

Figure 34b shows an example of the integration of a polymer solar cell and an electrochemical supercapacitor on a flexible fiber substrate. The in-serial connection allowed the energy-storage efficiency in the supercapacitor to reach 65.6%.[341] In addition, the in-serial connection of two devices is beneficial for maintaining the structure stability, as the mechanical properties of the whole device are largely determined by the same fiber electrode. This allowed them to better maintain electrochemical performance during the deformations.

9.2. Coaxially Integrated Fiber Devices

Using the coaxial configuration is another way to integrate multiple functions on a single fiber electrode. Although the coaxial or sheath−core configuration inevitably increased the difficulties in the fabrication of fiber devices, a few pioneering studies have successfully utilized this configuration.[551−553] For instance, the TENG and supercapacitor could be integrated coaxially.[342] The TENG was fabricated by winding carbon fiber bundles over the supercapacitor fiber, with a silicone rubber coated outside as a triboelectric material, as shown in Figure 34c. The inner supercapacitor was spontaneously charged upon deformations, and the collected energy could be stored and later used to power electronic products. In addition, other than integrating two fiber energy-harvesting and -storing devices, this strategy could be used to integrate powering fibers with other functional fiber devices that use electricity, such as photodetectors and strain sensors.

9.3. Parallel and Twisted Integrated Fiber Devices

An advantage of integrating fiber devices in parallel is to decrease the inner resistance of the whole device, leading to a boost of performance. For example, by weaving multiple fiber TENGs in parallel, the inner resistance was decreased by an order of magnitude. As a result, the output current of parallel TENGs showed an 11.8-fold enhancement compared to a single long-fiber TENG.[554] Another work demonstrated the parallel connection of fiber solar cells, and the current output of the device could be improved approximately proportional to the number of integrated photoanodes without an obvious negative impact on the voltage output.[555]

Each integrated unit in a parallel integrated system works independently. In contrast, the electrodes in a twisted integration system are in close contact with each other, which may have an impact on their overall performance. For example, the fiber battery and supercapacitor could be integrated in a twisted configuration, as shown in Figure 34d. The hybridized energy-storage device could achieve both high energy and power density, offered by the battery unit and supercapacitor unit, respectively.[40] The two units shared a CNT/$Li_4Ti_5O_{12}$ fiber electrode, and more importantly, the twisted configuration allowed any two of the three electrodes to be closely contacted, benefiting the ionic transport among the electrodes, which is crucial for the high electrochemical performance.

9.4. Integrated Functional Textile Devices

The format of fiber devices makes them mutually promising for integration on textiles, especially with the weaving and looming techniques.[556,557] For example, considering that solar energy is dependent on the weather, the TENG could be integrated with DSSCs on the textile as a complementary power provider (Figure 35a).[558] The resulting output performances of the TENG fabrics achieved a peak power density of 3.2 W/m^2, while the DSSC achieved an average PCE of 6%. The concept of hybridizing two types of energy-harvesting devices in textiles provides an insight into making high-performance powering textiles.

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The fiber devices with different functions could be integrated into textiles, and the resulting textile system could provide multiple functions including displaying, communicating, and health monitoring. Figure 35b shows the integration of power supply, display, and information input units in the textile.[52,53] The integrated textile system functioned as a communication portal, as the inputted information by the user could be spontaneously displayed on the textile. Another recent work demonstrated that the fiber Li-ion battery could power the integrated fiber sweat biosensors and display the outcome via an electroluminescent textile display, as shown in Figure 35c−e.

10. CHALLENGES FOR FIBER DEVICES

To discuss the challenges for the fiber devices, we first need to envision where this field is going and then identify the obstacles blocking the road. The fiber devices are promising to work as human-interfaced smart electronics as power supplies, computation modules, displays, sensors, or therapeutic devices, or to be integrated as an electronic system to provide a combination of these functions. Current fiber devices have shown promise to provide these functions as wearable or implantable devices. However, most of the studies are still at the early stage. There is a long way to go before these fiber devices can reach the market and impact human society. In general, there are a few aspects to consider before fiber devices could be further developed and widely used.

10.1. Safety Concerns

Safety is the first concern when it comes to actual applications. As human-interfaced devices, the fiber devices need to undergo constant deformations and hence are susceptible to mechanical failures and the leakage of encapsulated materials. In any circumstances, toxicity caused by these materials to the human being is not acceptable. The current fiber device is usually composited with two fiber electrodes, an active material, and an electrolyte and then packed with an encapsulating layer. The biocompatibility of fiber electrodes, including metal wires, carbon materials, and polymer fibers, is generally good.[559−563] However, there are still reports concerning the toxicity of these materials at the cellular level, as the materials may interact with living cells and cause cytotoxicity.[564] Natural fiber materials have advantages in this aspect, but they usually require additive materials to enhance their electrical properties as fiber devices. The active materials and electrolytes of fiber devices face a more severe safety problem, as they may use heavy metal elements or toxic organic components.[55] Gradually replacing these materials with compatible materials is a crucial research direction, e.g., developing an aqueous electrolyte and bioabsorbable active materials could help solve the problem.[389,565] Another way to alleviate the problem is to pack the device with a reliable encapsulation layer, which must meet several criteria, including biocompatibility, superior mechanical strength, flexibility, and suitability for continuous fabrication.

Another concern for the safety of fiber devices resulted from the mechanical mismatch between the fiber devices and soft tissues, especially for chronic implanted devices. This problem not only causes the failure of the device (encapsulated by activated immune cells) but also leads to scarring and inflammation of the body.[22] The soft fiber devices rose to address this problem by using intrinsically flexible materials with bending stiffnesses close to those of living tissues. These devices showed a prolonged operation lifetime and were more compatible with living tissues.[495] However, most of the results were obtained from rodent models; how the soft implantable fiber devices impact large primates and humans largely remains unknown. More efforts should be devoted to understanding this point because the micromovement of tissues in large primates and humans is more volatile, which may raise additional mechanical requirements for the implantable fiber devices.

10.2. Performance Improvements

Although the performances of fiber devices have steadily improved during the past decade, their performances are still lower than those of their planar counterparts. The electrode selection, structural design, active-material modification, and device assembly or packing determine the performance of the fiber device; thus, it is important to consider them carefully depending on the application purpose of the fiber device. First, fiber electrodes play a significant role in achieving high-performance devices because they act as conductive substrates and host active materials. In addition, the mechanical performances of fiber electrodes are also significant if the fiber devices are used as wearable or implantable platforms. Thus, an ideal fiber electrode is expected to have a low electrical resistance to facilitate charge transport, large surface area to host active materials, and high mechanical strength and flexibility for human-interfaced applications. The existing fiber electrodes may meet several of these requirements, but we often see trade-offs among these properties. For example, CNT fiber electrodes have excellent mechanical properties and large specific surface areas, but their resistances are higher than those of metal wire electrodes. On the other hand, the metal wire electrodes could support highly efficient charge transport, but their flexibility may be a problem for wearable or implantable applications. In short, the overall performance of fiber electrodes should be further improved. Hybridizing the existing fiber materials or developing new fiber materials are promising research directions.

The active materials, acting as a bridge between the electrolyte and the electrode, determine the specific function of the fiber devices. The current active materials used for fiber devices were mainly borrowed from planar devices.[18,548,566] Although they could provide stable performances in many cases, the stable interfaces between active materials and the electrode or electrolyte are hard to achieve, especially if the fiber device suffers from constant mechanical deformations. A possible route to solve this problem is to design one-dimensional active materials at the molecular level and assemble them seamlessly with the fiber electrode. Such an interface may be more stable and efficient for charge separation and transport, thus establishing high-performance fiber devices.

The encapsulation of fiber devices is another crucial step for obtaining high-performance fiber devices. The encapsulation of fiber devices is 2-fold. (1) It could provide an isolation layer to avoid invasion of water or oxygen into the fiber device and also avoid the deactivation or degradation of the active materials or electrolyte. (2) It could serve as a protection layer to eliminate the leakage of toxic encapsulated materials. An ideal encapsulation is indispensable for fiber devices to work stably. Although the small size and high curvature of the fiber electrodes make the packing of the devices difficult, various materials were found to encapsulate the fiber devices, including polypropylene, polyvinylidene chloride, polydimethylsiloxane, and polymer/graphene composites. These materials could

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provide a low water vapor transmission rate of 0.01−100 g/m^2/day and allow for an operation time of fiber devices ranging from minutes to hours. However, for real-life applications, the performance of the encapsulation layer should be improved (<0.01 g/m^2/day), which could further result in an increased lifetime of the fiber device.[141] In addition, the encapsulation layer should not be the bottleneck of the overall mechanical performance of the fiber device and should be suitable for large-scale fabrication. Research on the encapsulation layer is a less common topic in the field and deserves more attention in future studies.

10.3. Universal Testing Standards and Evaluation Systems

As a newly emerging research field, how to precisely evaluate the performance of fiber devices remains vague. For example, the power density of fiber energy devices could be measured by watt per square meter, watt per cubed meter, or watt per gram, making it difficult for scientists to compare and assess the technology advancement of each work.[34,36,330] In addition, mechanical performance is crucial for fiber devices, mainly when they are used as wearable or implantable platforms. However, the mechanical properties were usually evaluated by standard tensile machines, which may fail to work with the low mechanical strengths of fiber devices.[567] More sensitive stress-sensing systems should be designed and utilized to solve this problem. Further, bending, twisting, and stretching tests were mainly performed by hand or homemade equipment, resulting in the arbitrary mechanical property determination of fiber devices.[55] After all, the lack of a universal testing standard and evaluation system confuses the field and slows down its development.

10.4. Integrations

The integration of fiber devices could provide them with a higher power output or multiple functions, which could not be achieved by a single fiber device. The integration of fiber devices is a major step to push them toward real-life applications. However, the integration of fiber devices on textiles requires reliable electrical connections among various functional parts. Such connections have been typically achieved by using soldering/welding or mechanical gripping of fiber electrodes. However, the soldering/welding of multiple fiber electrodes may lead to a rigid textile network, decreasing their comfortability as wearable devices.[568,569] In addition, the brittle connections of the textile network, formed by either soldering/welding or mechanical gripping, may not survive repeated mechanical deformations, resulting in device failures. Further, integrating multiple fiber devices increases the difficulty in large-scale fabrication. To the best of our knowledge, the integrated fiber/textile devices mainly are demonstrated at the laboratory stage, and the large-scale fabrication of integrated fiber systems remains largely unexplored. We anticipate more work will be done to lower the cost of the integrated system to push them toward real-life applications. Meanwhile, newly emerging technologies, such as artificial intelligence, could also help improve the performance of fiber devices. For example, with the assistance of artificial intelligence, fiber sensors could better analyze data and provide personalized health care suggestions with high efficiencies.

In addition, the power management of integrated fiber devices remains difficult. The integrated fiber devices are expected to use more significant amounts of energy, and thus a reliable and sustainable power supply module in such an integrated system is required. Although fiber energy-storage devices, including fiber batteries and supercapacitors, and fiber energy-harvesting devices, including biofuel cells (BFCs) and nanogenerators, could provide power to the integrated system, they still have some limitations. For example, the power output of a wearable fiber BFC is usually delayed for getting access to sweat, which contains the necessary biofuels for power generation. Meanwhile the nanogenerators could provide power immediately, but their output is usually low. Thus, smart management of the power output could affect the performance of the integrated electronics. The incorporation of multiple energy modules could potentially solve the problem, such as integrating batteries and supercapacitors or BFCs and nanogenerators in one system to compensate the drawbacks of each component.

10.5. Large-Scale Fabrications

The large-scale fabrication of fiber devices is a key step for their broad applications. Currently, most reported fiber devices share a similar fabrication protocol with planar devices, which could only be fabricated with a limited length (centimeters to meters). Many fiber devices depend on delicate and time-consuming handcraft work, industrial installation to continuously fabricate fiber devices is rare. Hence, designing an effective route to process and optimize the fiber devices at a large scale remains an unmet challenge and requires seamless collaboration efforts between academy and industry. The main problem facing the large-scale fabrication of fiber devices should be shared between academy and industry. The small size and high curvature make the uniform and robust modification of the fiber electrode and subsequent device assembly difficult. The one-step thermal-drawning technique has proven to be suitable for fabricating specific fiber devices at a large scale (e.g., fiber battery).[129] However, it requires procuring the molten component materials, limiting their potential as a universal and versatile platform. Further, controlling the drawing stress of each molten phase and viscosity remains difficult, often resulting in low yield and poorly controlled interfaces within the fiber device. On the other hand, initial attempts have demonstrated the large-scale fabrication of fiber devices using wet-spinning and successive coating methods, which could result in fiber energy devices that are hundreds of meters or kilometers long.[53,58] More efforts should be devoted to adapting these methods to fabricate a range of other fiber devices such as sensors or displays, and more dedicated studies should be made to reveal the relationship between fabrication parameters and device performances to further increase the production rates. It should be noted that the newly emerging fabrication technologies could have an impact on the large-scale fabrication of fiber devices. For example, 3D printing has demonstrated its versatility to fabricate a range of materials and devices, ranging from microrobots to biomedical scaffolds, as well as novel fiber devices, including fiber sensors and energy devices.[570−572] The recent 100-m-long supercapacitors fabricated by 3D printing have demonstrated the potential for large-scale fabrications.[346] However, using this fabrication method to fabricate other fiber devices at a large scale has rarely been reported. The key to such a successful fabrication is to obtain low-cost printing materials, accurately control the fabrication speed, and achieve high resolutions during printing, which remains difficult and requires more efforts.

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To summarize, the newly developed fiber devices have demonstrated their capabilities with various functions as human-interfaced electronics. In this review, we comprehensively reviewed the basics from fiber materials to design principles of fiber devices. During the past decade, we have witnessed the development of various fiber devices. Although most studies still stay at the laboratory stage, we do observe a trend to push them toward industrial production and real-life applications. We expect new fiber devices will be continuously discovered, along with the growing efforts devoted to the field. Further, the opportunities and challenges coexist in the field. By addressing the challenges remaining in the field, including improving the safety and performance, enhancing the lifetime, designing more efficient integrated systems, and lowering the fabrication cost toward large-scale applications, fiber devices may be ready to infiltrate our daily life soon.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.chemrev.2c00192.

Examples of recently reported fiber supercapacitors and batteries (PDF)

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Notes

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