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REVIEW

Functionalization of Fiber Devices: Materials, Preparations and Applications

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Abstract

Conventional electronic devices with bulky and rigid features cannot fully meet the requirements of flexibility and wearability in wearable applications. Fiber-shaped electronic devices have been intensively pursued in the past decade attributed to their excellent flexibility, weavability and wearability. The innovation of novel functions has been widely recognized as an emerging direction of fiber-shaped electronic devices, pursuing a better adaptability and longer lifetime in practical applications. In this Review, we summarize the recent advances of functional fiber devices, focusing on the preparation of functional fiber electrodes and electrolytes, as well as the formed interfaces. Fiber devices with a variety of novel functions are systematically introduced, including but not limited to stretchability, healability, shape memory and electrochromism. The remaining challenges and opportunities are also discussed to propose future directions for functionalization of fiber electronics.

Keywords Fiber devices · Functionalization · Functional fibers · Wearable electronics

Introduction

The modern society has witnessed the rapid development of wearable devices in the past decade, which shows huge potentials in a variety of important applications such as artificial intelligence, internet of things, smart healthcare and aerospace technology [1–4]. Bulky and planar devices that are generally rigid cannot be fully integrated with daily clothes to fit the highly irregular surface of the human body. To this end, electronic devices in a novel fiber format have emerged as promising candidates for wearable electronics. For instance, their lightweight and flexibility can guarantee the stability of functional devices when encountering complex deformation such as twisting and stretching. Most importantly, they can be woven into flexible, breathable and wearable textiles that fit human body to meet the requirements in various wearable and portable applications [5–7]. Up to date, a broad range of fiber-shaped devices have been developed and optimized, including but not limited to energy harvesting devices (solar cells, nanogenerators, photodetectors) [8–15], energy storage devices (supercapacitors and rechargeable batteries) [16–21], sensors [22–24] and actuators [25–27]. Very recently, long fiber-shaped lithium-ion batteries based on industrial production equipment and process have provided insightful inspiration for mass-production of fiber-shaped devices with high performances [28]. However, one of the key challenges lies in the functionalization of fiber electronic devices, that is, introducing various functions to fiber devices to enhance the adaptability in complex and harsh environment [29–32]. In the past decade, great efforts have been made to realize a variety of functional fiber devices as shown in Fig. 1.

The functionalization of fiber-shaped devices is mainly motivated by the potential problems in practical applications. For instance, fiber-shaped devices are expected to accommodate the frequent strain and stress in wearable applications, which gives birth to stretchable fiber-shaped supercapacitors in 2013 [31] (Fig. 1). Additionally, delicate fiber-shaped devices may encounter structural damage in wearable applications, which requires rational designs to realize the healing of mechanical and electrical properties. Thus, self-healable fiber-shaped supercapacitors were developed to meet the above requirements in 2014 [32] (Fig. 1). In both cases, the incorporation of functional components

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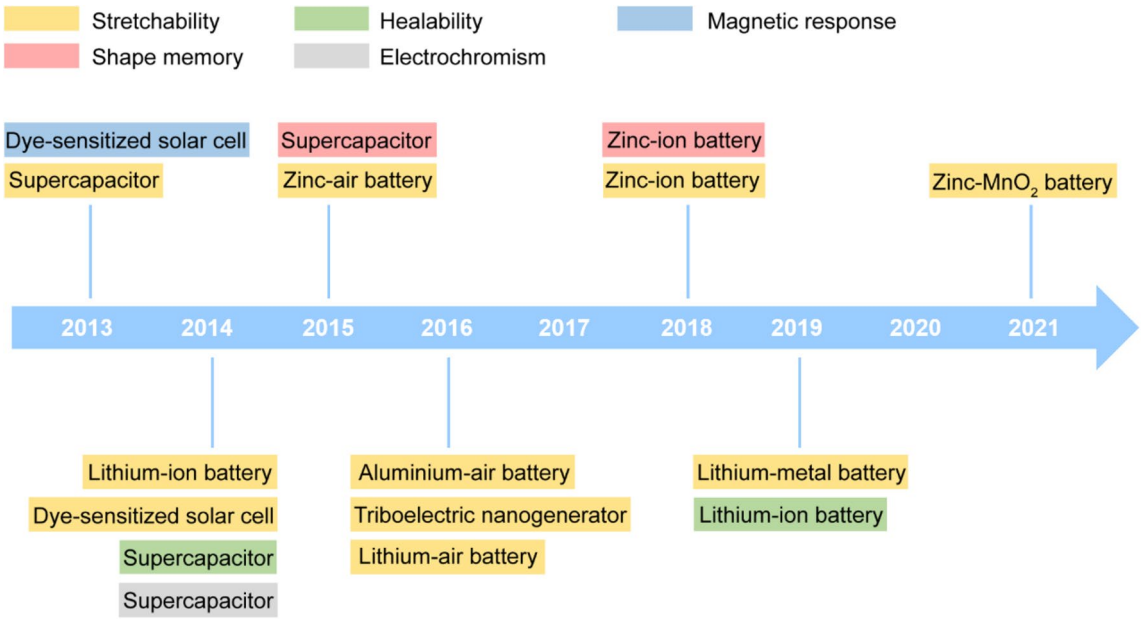

Fig. 1 Timeline of developments in functional fiber devices including stretchability, healability, shape memory, electrochromism and magnetic response

with fiber electrodes represents a key issue, and the formed interfaces play an indispensable role in the functionality and stability of the obtained devices.

In this Review, the latest advancements on the functionalization of fiber-shaped devices are introduced. An emphasis has been made on the incorporation of functional guests into various fiber electrodes, focusing on different fiber electrode materials and configurations, as well as the interfaces between different components contributing to high flexibility and stability. Various functional fiber devices are introduced including but not limited to stretchability, shape memory, healability and electrochromism, mainly demonstrating the motivation, evolution and application of each function. The challenges and perspectives of functional fiber-shaped devices are discussed in the end, proposing opportunities and future directions of this emerging field.

Functional Fiber Electrodes and Novel Interfaces

Intelligent wearable electronics are expected to be multifunctional to accommodate complex and harsh application scenarios [33]. Functional fiber electrodes are considered as the building blocks for production of functional fiber-shaped devices. It is particularly crucial to incorporate functional and conductive components into fiber electrodes without sacrificing the device performances, such as power conversion efficiency, energy density and cycling stability. Therefore, a variety of fiber electrodes have been employed as the skeletons for preparing functional fiber electrodes with several efficient approaches.

Metal wires are widely recognized as promising fiber electrodes of fiber-shaped devices, due to their high tensile strength and remarkable electrical conductivity [34, 35]. However, the high density and limited flexibility have hindered their applications in fiber-shaped devices to some degree. To this end, conductive materials have been widely incorporated with various fiber substrates serving as flexible and lightweight fiber electrodes [36–41]. For example, polyaramid/polypyrrole/Cu composite fibers could achieve high electrical conductivity via sequential deposition of polypyrrole and Cu on a polyaramid fiber [39]. In this process, the chemisorption of palladium on the polypyrrole layer was proved to be necessary for uniform growth of the Cu layer. With an optimal deposition time of 240 s, a 25 cm^2 composite textile showed a resistance of 5 Ω.

In addition, other materials such as MXene, carbon black and silk can be spun into flexible nanofibers with increased surface areas, which can be coated onto various fiber substrates for improvement of the electrochemical performances [42–44]. For instance, a nanofibers-based fiber electrode was prepared by uniformly winding MXene/polyethylene oxide nanofibers on the polyester fiber via a modified electrospinning process, in which the thickness of the nanofiber coating could be controlled by the spinning time [44]. Nanofiber coating could offer large surface area to improve the interface contact between the electrodes and electrolytes. Polyester fiber provided composite fibers with good mechanical and flexible properties. On

the basis of nanofibers coated fiber electrode, the wearable supercapacitors demonstrated a high areal capacitance of 18.39 mF/cm^2 at 50 mV/s and remained 98.2% of original specific capacitance after 6000 cycles, superior than the pure MWCNT (4.28–8.66 mF/cm^2) and graphene fiber-based supercapacitors (0.44–1.7 mF/cm^2).

However, the conductive coatings are prone to peel off from fiber substrates due to the instable interfaces based on physical adsorption, and the tensile strength of the obtained composite electrodes cannot fully compete with metal wires. As a solution to the above challenges, fiber electrodes based on carbon nanomaterials such as carbon nanotube and graphene have attracted broad attentions in the past decade, due to their high strength, high electrical conductivity, large specific surface area and stable interfaces [45–50]. For instance, aligned multi-walled carbon nanotube (MWCNT) fiber is considered as an ideal fiber electrode on the basis of its excellent mechanical and electrical properties, e.g., remarkable tensile strength and electrical conductivity as high as 5.5 GPa [51] and 1.09×10^7 S/m [52], respectively. As a result, aligned MWCNT fibers have been widely used as the conductive skeleton for incorporation of functional components, and are mainly introduced in this review.

Wrapping aligned MWCNTs onto the surface of a functional fiber represents a rational strategy for preparing functional fiber electrodes. The functionality is realized by the functional fiber at core, and the aligned MWCNTs serve as the conductive components at shell. For instance, a stretchable electrode could be fabricated by wrapping MWCNT sheets on the rubber fiber through a "rotation-translation" method (Fig. 2a) [31]. The stretchability was realized by the stretchable rubber fiber at core, and the aligned MWCNTs served as the conductive and energy storage components

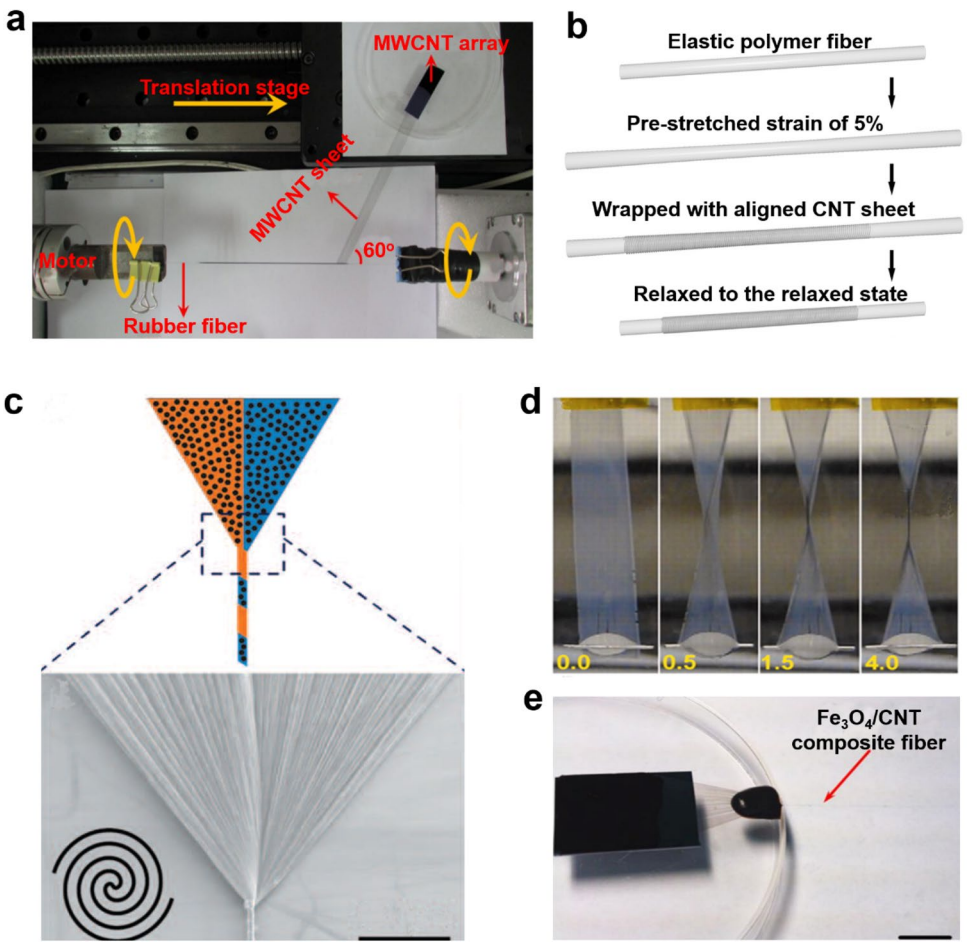

Fig. 2 **a** The "rotation-translation" equipment for wrapping aligned MWCNT sheets onto a stretchable rubber fiber [31]. Reproduced with permission. Copyright 2013, Wiley–VCH. **b** The pre-strain process for production of a stretchable fiber electrode [53]. Reproduced with permission. Copyright 2015, Wiley–VCH. **c** Functional guests and aligned MWCNT sheets stacked together (scale bar: 250 μm). **d** The preparation of functional fiber electrode using a biscrolling method. The numbers at the left bottom corner represent the twisting turns [54]. Reproduced with permission. Copyright 2011, AAAS. **e** A continuous process for production of Fe_3O_4/MWCNT fibers with magnetic response (scale bar: 1 cm) [55]. Reproduced with permission. Copyright 2013, Wiley–VCH

at shell. The resulting fiber electrode could stably retain its electrical resistance (change less than 5%) at a maximal strain of 100%. In another case, the pre-strain strategy that was widely used in stretchable planar electrodes was also adopted to realize stretchable fiber electrodes. An elastic polymer fiber (Ecoflex 30) was first stretched and wrapped by aligned MWCNT sheets, and the wrinkle structure could form when the fiber was released to the relaxed state [53] (Fig. 2b). The highly stretchable polymer fiber and the wrinkle structure offered a significantly enhanced stretchability of the resulted fiber electrode, delivering a super stretching property with an impressive strain of 500%. The electrical resistance of this fiber electrode was increased by less than 70% when the strain was below 400%.

Apart from the core–shell structure, functional fiber electrodes with other configurations were also developed. As early as 2002, Lima et al. showed that the functional guests could be incorporated with aligned MWCNT sheets via a facile "biscrolling" method [54]. In detail, the functional guests and aligned MWCNT sheets were stacked together (Fig. 2c), followed by twisting the two ends along opposite directions to obtain a functional fiber (Fig. 2d). Based on the lightweight MWCNT sheets with an ultralow areal density of about 1–3 μg/cm^2, this method could afford high loadings of various functional components up to 95%, providing a powerful platform for preparing various functional fiber electrodes. However, the biscrolling method still suffered from several drawbacks, e.g., continuous production was difficult because the MWCNT sheets were required to be fixed before twisting into a fiber. In addition, in order to avoid the shrinkage of MWCNT sheets in solvents, the functional guests had to incorporate with MWCNT sheets via solvent-free methods such as thermal evaporation.

To solve the above issues, Sun et al. developed a continuous preparation strategy of functional fiber electrodes. The functional components such as Fe_3O_4 nanoparticles were dispersed in water, and the aligned MWCNT sheets were scrolled and drawn through the dispersion (Fig. 2e). The Fe_3O_4 nanoparticles were readily incorporated into MWCNTs as verified in scanning electron microscope (SEM) [55]. The introduction of Fe_3O_4 nanoparticles was also proved by the magnetic response capability of the obtained hybrid fibers. Importantly, continuous functional fibers with lengths of hundreds of meters could be prepared via this efficient approach.

Wet-spinning is another effective method for continuously preparing functional fiber electrodes. For instance, a representative work reported by Xu et al. proposed a wet-spinning method for producing graphene oxide (GO) fibers [56]. GO liquid crystal dispersions were extruded to a coagulation bath to obtain continuous GO fibers. Subsequently, chemical reduction could be performed to produce reduced graphene

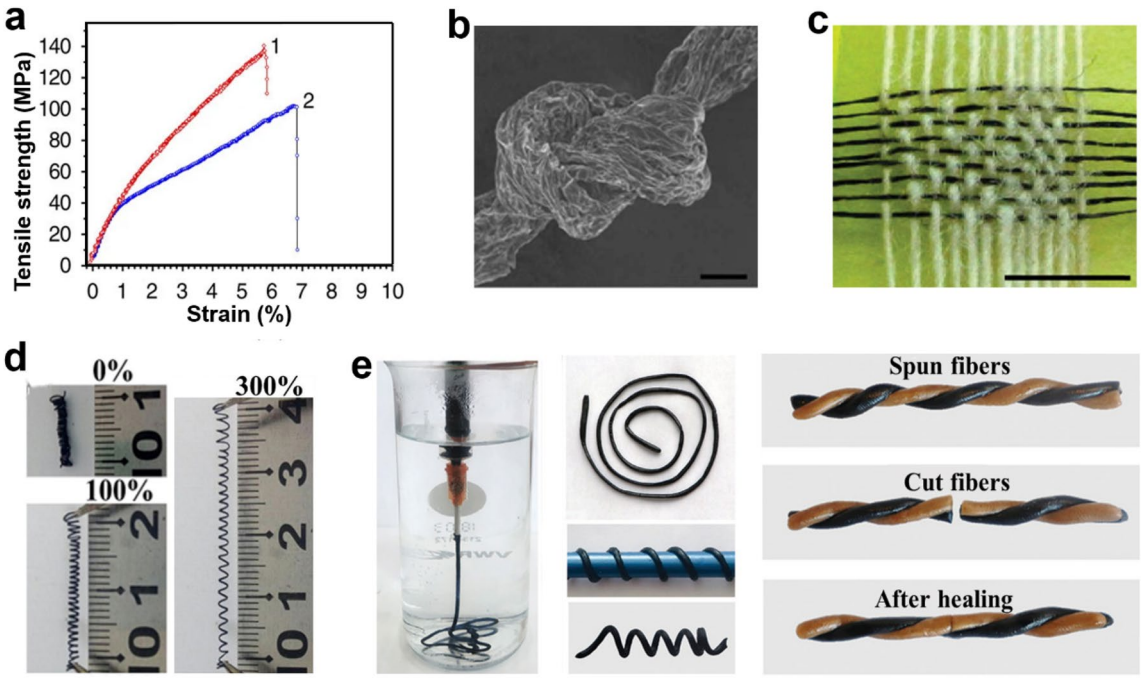

Fig. 3 **a** The tensile curves of graphene fibers (1) and GO (2). **b** The tighten knot of a graphene fiber (scale bar: 50 μm). **c** Several graphene fibers (horizontal) knitted with cotton threads (vertical) (scale bar: 2 mm) [56]. Reproduced with permission. Copyright 2011, Nature Publishing Group. **d** Stretchability demonstration of an rGO-based fiber spring [57]. Reproduced with permission. Copyright 2017, American Chemical Society. **e** CareGum fiber prepared via a wet-spinning method with self-healing ability [58]. Reproduced with permission. Copyright 2021, Wiley–VCH

oxide (rGO) fibers, which showed impressive mechanical properties (140 MPa in tensile strength, 5.8% in elongation) (Fig. 3a) and electrical conductivity (2.5×10^4 S/m). The graphene fiber could be made into tightened knots and knitted in cotton network (Fig. 3b, c), indicating its excellent flexibility and knittability. Wang et al. further designed a rGO-based fiber spring electrode based on GO, MWCNTs, sodium dodecyl sulfate and vitamin C [57]. The mixed components were injected into a pipe to form a composite fiber, followed by thermal reduction of the GO at 90 °C. A thin layer of polypyrrole (PPy) was further electrodeposited on the fiber. The resulted rGO-based fiber was twisted into a spring-shaped electrode, which demonstrated a high stretchability with a maximal strain of 300% (Fig. 3d).

Despite of the impressive achievements on various functional fiber electrodes, it is still challenging to realize multiple functions in a single fiber electrode. Recently, A novel fiber electrode named "CareGum" has been developed on the basis of silk, calcium chloride, tannic acid and rGO [58]. The CareGum showed a variety of functions including self-healing, adhesive and sensing capabilities derived from the hierarchical bonding scheme consisting of hydrogen and electrostatic bonds. Additionally, continuous fibers with millimeter-sized diameters were obtained via a wet-spinning method, indicating the application potentials in wearable electronic displays, artificial skin and stain sensors (Fig. 3e).

The introduction of functional components ineluctably creates new interfaces which play important roles in the functionality and stability of the obtained fiber devices. This is relevant to the unique curved configuration of the fiber electrode, as well as the mechanical mismatch of different components. Particularly, the interfaces between the structural and functional components play critical roles in the stability and functionality of the resulted fiber devices. For instance, delamination is prone to occur during stretching or bending, which results in performance degradation of the obtained fiber electrodes and devices. Therefore, a layer of gel electrolyte comprised of polyvinyl alcohol (PVA) was coated on the elastic fiber to enhance the contact between aligned MWCNT sheets and elastic fiber substrate. Subsequently, the vacuum post-processing was performed to efficiently improve the infiltration of electrolyte into aligned MWCNT sheets. These strategies resulted in enhanced interfacial stability of the fiber-shaped supercapacitors during stretching (Fig. 4a), which retained over 90% of the original specific capacity after 1000 stretching cycles at a strain of 75% [31].

In addition, the rational interface optimization between the conductive and functional components has been proved to be important for achieving high performances. For instance, aligned MWCNT sheets could be uniformly and stably wrapped around a self-healing polymer fiber comprised of elastic rubber with abundant hydrogen bonds. The van der Waals force contributed a reliable interfacial interaction between aligned MWCNT sheets and self-healing polymer (Fig. 4b), resulting in the formation of a "bridge" structure between the broken ends after healing (Fig. 4c), which was vital for the recovery of the electrical properties (electrical resistance increased by 8.5% after healing) [32]. As a comparison, the control group using randomly dispersed Ag nanowires could not fully rebuild the broken conductive networks due to the instable interface formed with the self-healing polymer, leading to a significant electrical resistance increase of 191.6%.

The interface between the electrolyte and fiber electrode also has a vital influence on the device performances. For example, fiber electrodes were prepared via a 3D printing technology using a highly viscous polymer ink, which comprised of poly(1,1-difluoroethylene) (PVDF) and MWCNT. Then, a layer of poly(vinylidene fluoride-co-hexafluoropropene) (PVDF-co-HFP) gel was coated on the surface (Fig. 4d), on which the porous structure was beneficial for electrolyte absorption and ion transport [59]. The resulting fiber-shaped lithium-ion battery exhibited a high specific capacity (110 mAh/g) and cycling stability (81% of capacity retention over 30 cycles), indicating the importance of the stable electrolyte/electrode interface. In addition, the optimized interface between active materials and MWCNTs is favorable for effective charge transport in fiber devices. For instance, polyaniline was electrochemically deposited on the aligned MWCNT, between which the π–π interaction contributed to a strong adhesion. The orientation of polyaniline induced by the alignment of MWCNTs further enhanced the charge transport, thus leading to good electrochemical performances (Fig. 4e) [60].

The favorable interfaces are also dependent on the working mechanisms of different devices. For instance, efficient charge transfer at the electrolyte–electrode interface is particularly important for energy storage devices. Hence, the rational interface optimization is important for fiber-shaped supercapacitors and batteries to enhance the infiltration and ion diffusion of the electrolyte. For example, the interfacial electrical resistance derived from polyacrylamide-based hydrogel electrolyte and double-helix MWCNT fibers showed no significant changes after 300 cycles at 2 A/g. As a comparison, the control group using conventional liquid electrolyte (2 mol/L $ZnSO_4$ and 0.1 mol/L $MnSO_4$) demonstrated significantly increased interfacial electrical resistances. The resulted solid-state zinc-ion fiber battery using polyacrylamide-based hydrogel electrolyte exhibited a high capacity retention of 98.5% over 500 cycles [61].

For photovoltaic devices, the photovoltaic conversion efficiency highly relies on the electron transport process at the interface between the photoactive material and semi-conductor skeleton. Perpendicularly grown TiO_2 nanotube arrays on a Ti wire were used for dye adsorption, and numerous

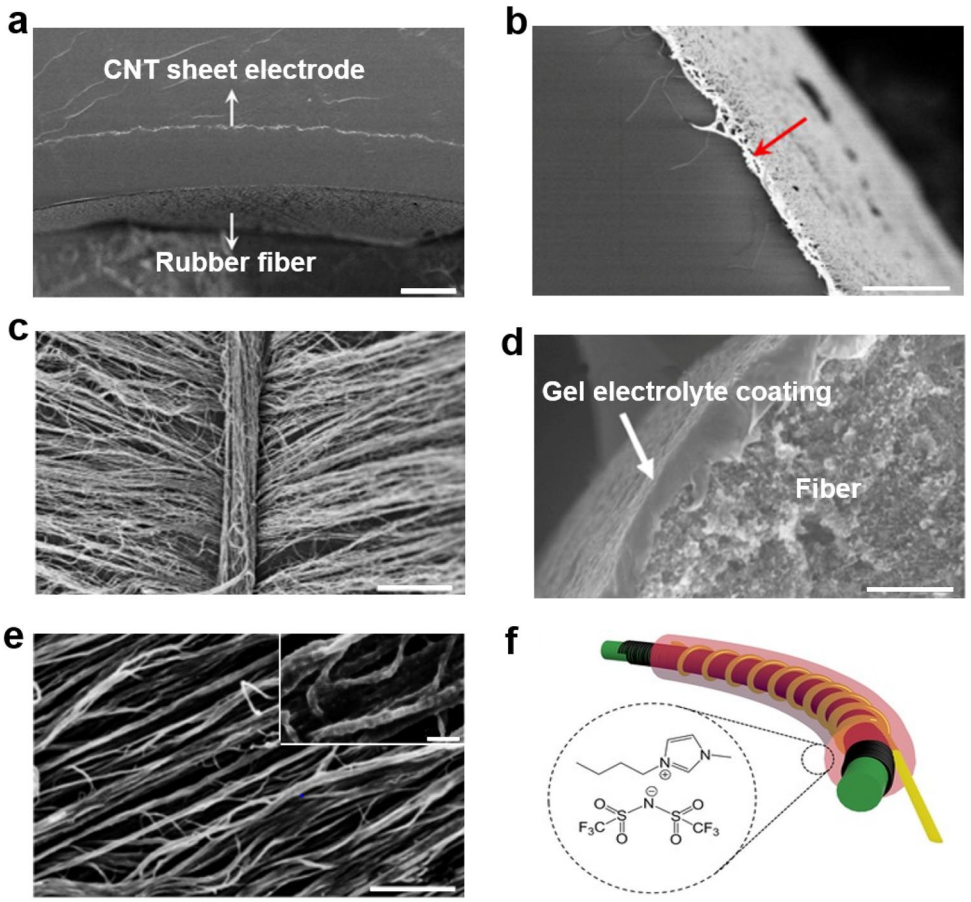

Fig. 4 **a** Cross-sectional SEM image of the stretchable fiber-shaped electrode with a core–shell configuration (scale bar: 10 μm) [31]. Reproduced with permission. Copyright 2013, Wiley–VCH. **b** The cross section of a self-healing fiber showing the interface between aligned MWCNT sheets and self-healing polymer (scale bar: 3 μm). **c** The "bridge" structure formed between the broken ends after healing (scale bar: 5 μm) [32]. Reproduced with permission. Copyright 2014, Wiley–VCH. **d** Cross-sectional SEM image of a 3D printing fiber electrode coated with a PVDF-co-HFP gel (scale bar: 25 μm) [59]. Reproduced with permission. Copyright 2017, Wiley–VCH. **e** SEM images of bare aligned MWCNTs (scale bar: 500 nm) and MWCNT/PANI composites (inset, scale bar: 100 nm) [60]. Reproduced with permission. Copyright 2013, The Royal Society of Chemistry. **f** The structure of fiber-shaped dye-sensitized solar cell and the chemical structure of the ionic liquid gel electrolyte [63]. Reproduced with permission. Copyright 2015, Wiley–VCH

V-shaped voids were formed among these arrays [62]. The dye molecules and electrolyte could more effectively infiltrate into the aligned TiO_2 nanotubes compared with conventional TiO_2 nanoparticles that formed a dense coating layer. The charge transport along the aligned TiO_2 nanotubes was significantly improved compared with TiO_2 nanoparticles due to the improved charge transport pathway with less boundaries and defects. As a result, when the length of the TiO_2 nanotubes increased from 10 to 30 μm, the short circuit current density increased from 9.34 to 17.11 mA/cm^2 due to the enhanced charge transport process.

The safety issue of fiber-shaped devices should be taken in serious consideration due to the close contact with the human body. Conventional electrolytes on the basis of carbonate or ether solvents are generally flammable, suffering from unfavorable safety concerns when used in fiber-shaped energy harvesting and storage devices. Therefore, developing instinct non-flammable electrolyte becomes a pivotal direction for enhancing the safety of fiber-shaped energy devices. For instance, polymer-ionic liquid (IL) gel electrolyte was prepared using PVDF-co-HFP copolymer and 1-butyl-3-methylimidazolium bis(trifluoromethanesulfonyl) imide (BMImTFSI) IL (Fig. 4f) [63]. The non-flammability and outstanding thermal stability of the IL significantly enhanced the safety and long-term stability of the obtained fiber-shaped dye-sensitized solar cells, retaining 90% of the original power conversion efficiency after 30 days. In addition, the device showed good stability in humid environment due to the hydrophobic nature of PVDF-HFP co-polymer and F-based IL.

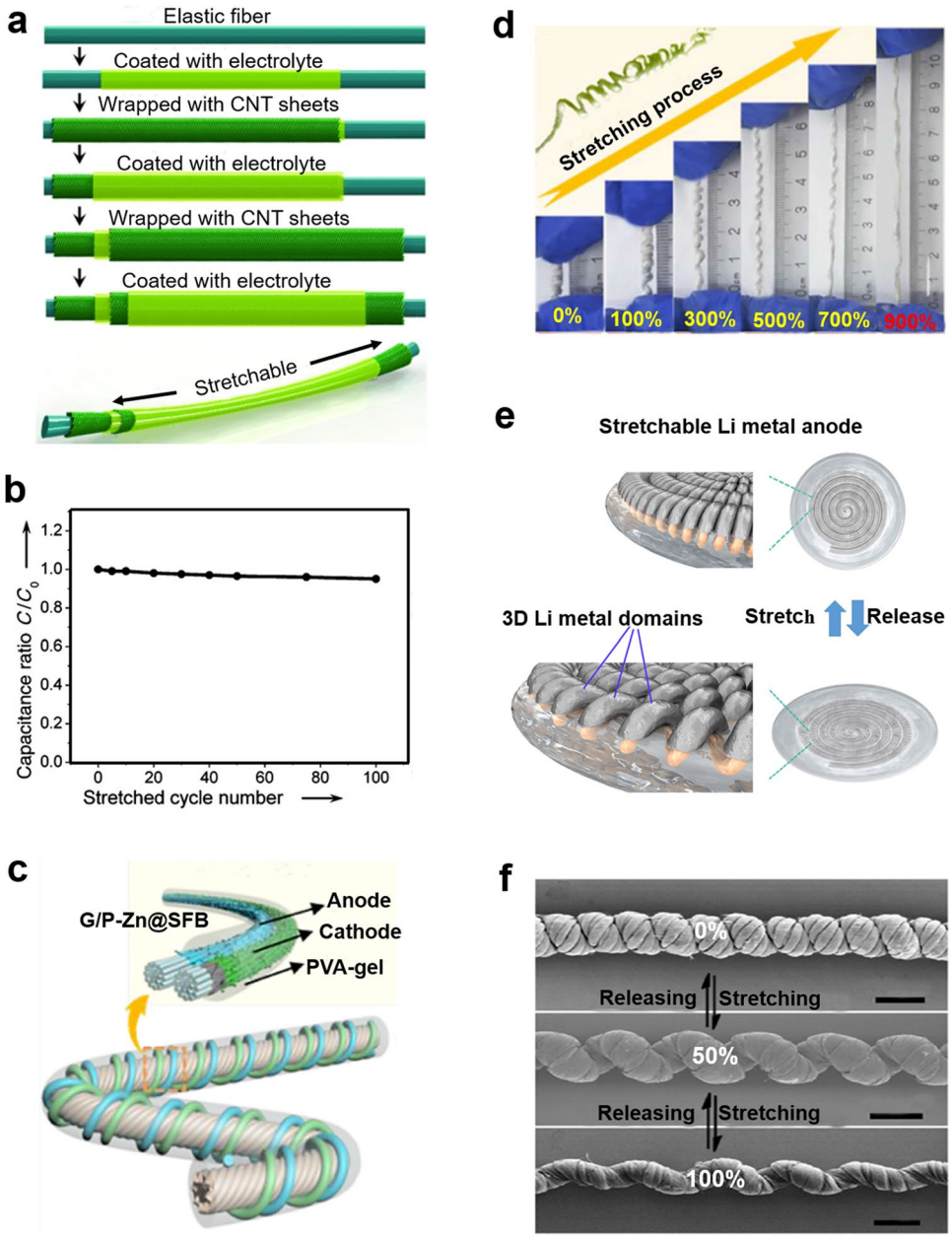

Fig. 5 **a** The fabrication process of a stretchable fiber-shaped supercapacitor with a core–shell configuration. **b** Capacitance retention on various stretching cycle number at a strain of 75% [31]. Reproduced with permission. Copyright 2013, Wiley–VCH. **c** Schematic illustration of the fiber device parallel-twisted around a spandex fiber to realize stretchability. **d** The stretchability demonstration of the fiber-shaped Zn-ion battery [64]. Reproduced with permission. Copyright 2021, American Chemical Society. **e** Schematic illustration of the stretchable Archimedean spiral-like Li metal electrode [67]. Reproduced with permission. Copyright 2018, Elsevier. **f** SEM images of a MWCNT fiber spring at different strains of 0%, 50% and 100% (scale bar: 40 μm) [70]. Reproduced with permission. Copyright 2014, Wiley–VCH

As a brief summary, the functional guests and the derived interfaces play critical roles in the functionality and stability of the obtained fiber-shaped devices. Novel fiber electrodes, going beyond flexibility and stretchability, can realize a variety of novel functions such as healability, magnetic response, shape memory, electrochromism etc. On the basis of these functional fiber electrodes, various functional fiber-shaped devices have been developed with impressive adaptability to complex application conditions.

Stretchable Fiber Devices

Stretchability is of great significance to fiber-shaped devices due to the inevitable deformation in wearable applications. As a result, efforts have been made towards developing stretchable fiber-shaped devices to accommodate the dramatic deformation in practical applications. A pioneering work reported by Yang et al. showed stretchable fiber-shaped supercapacitors with a core–shell configuration [31]. In detail, the stretchable fiber supercapacitor was prepared through winding aligned MWCNT sheets and coating PVA gel electrolyte layer-by-layer onto a rubber fiber (Fig. 5a). Particularly, a novel "rotation-translation" method was developed in order to precisely control the helical angle and thickness of the aligned MWCNT sheets. The structural stretchability was realized by the rubber fiber at core, while the electrical conductivity could be retained during stretching on the basis of the helical MWCNTs. As a result, the fiber-shaped supercapacitor exhibited specific capacities of 42–80 F/g, which could be maintained over 90% after 1000 stretching cycles at a strain of 75% (Fig. 5b).

Another strategy for achieving stretchability is to attach non-stretchable fiber devices onto stretchable fiber substrates, realizing the stretchability via a helical configuration design [41, 64–66]. This strategy avoids the preparation of stretchable fiber electrodes, thus shows a simplified fabrication process. For instance, a silver fiber deposited with graphene nanosheets and polyaniline (PANI) nanowires served as the fiber-shaped cathode, and another silver fiber was coated with zinc nanosheets as the anode [64]. The two electrodes and PVA/$ZnSO_4$ gel electrolyte were encapsulated with a polyurethane (PU) film, followed by parallel-twisted around a spandex fiber to form a double helix structure (Fig. 5c). On the basis of the high stretchability of the spandex fiber, the resulting fiber-shaped Zn-ion battery exhibited a remarkable strain up to 900% (Fig. 5d). Additionally, Archimedean spiral-like structure was designed to endow Li metal electrode stretchability (Fig. 5e) [67]. A doubly coiled Cu wire was wrapped into an Archimedean spiral-like pattern, which was then embed into poly(styrene-ethylene-butylene-styrene) (SEBS) matrix. Li metal was electrochemically deposited onto the exposed Cu coil surface to obtain a stretchable electrode. Due to the joint effect of Archimedean spiral-like structure and highly elastic SEBS, this Li anode could be stretched up to 60% with well-retained capacity (90% of original value) after 100 stretching cycles.

It is worth noting that the above two strategies have their own pros and cons. For instance, fabricating intrinsically stretchable fiber electrodes and devices can realize impressive stretchability, representing a promising pathway towards stretchable fiber devices with high performances [31, 53, 68, 69]. However, the preparation of stretchable electrodes generally involves complicated assembling processes of the conductive and stretchable components. Furthermore, coating a uniform layer of conductive materials on a highly curved stretchable fiber remains to be challenging. Designing stretchable device configuration, that is, attaching non-stretchable electrodes onto stretchable substrate, extends the compatibility of the device category and simplifies the fabrication processes [64–66]. However, the use of stretchable supports significantly increases the volume of the fiber device, which no doubt sacrifices the device performances, and a short circuit is more likely to occur during the stretching process. Up to date, the former strategy is still the mainstream for production of stretchable fiber electrode and devices, although the advancement of new production techniques is still in urgent need.

The presence of elastic fiber substrate makes no contribution to the energy conversion or energy storage performances, resulting in rather limited power conversion efficiency or specific capacity. Therefore, efforts have been made to prepare intrinsically stretchable fibers with improved energy storage capacity. For instance, a MWCNT fiber spring was prepared by Zhang et al. using a bundle of overtwisted MWCNT fibers [70]. The overtwisting strategy had been verified as an effective approach to realize intrinsically stretchable fiber electrodes, and the MWCNT was an ideal material for supercapacitors and rechargeable batteries. The obtained stretchable supercapacitors could be stretched up to 100% (Fig. 5f), simultaneously delivering a specific capacity of 1587 mF/cm^3 that was over 145 times compared to elastic fiber. With $LiMn_2O_4$ and $Li_4Ti_5O_{12}$ incorporated into MWCNT fiber springs, the resulted fiber-shaped rechargeable batteries were developed, exhibiting a linear and mass specific capacity of 2.2 mAh/m and 92.4 mAh/g, respectively. Compared with previous stretchable fiber supercapacitors and rechargeable batteries, this work could achieve a significantly increased energy density due to the absence of stretchable fiber that contributed no energy storage capability. In another trail, fiber electrode with a supercoil structure was proposed inspired by the configuration of deoxyribonucleic acid (DNA) [71]. By over-twisting a straight fiber electrode followed by coiling again, the obtained electrode showed 29% of the initial length. Based on the supercoil electrodes, the resulting supercoil Zn/MnO_2 battery displayed high stretchability (a maximal strain of 800%) and an excellent linear capacity (2.9 mAh/m).

Shape-Memory Fiber Devices

Shape memory materials can be defined as stimuli-responsive materials capable to "memorize" a macroscopic shape. When manipulated and fixed to a temporary shape, they can

relax to the original, stress-free shape under specific stimulations such as heat, electrical field or other environmental commands [72]. Shape-memory fiber-shaped devices are expected to accommodate complex deformation adapting to non-standard surface and recover to the original shape under stimuli as required. As generally adopted, conductive components were incorporated with a shape-memory fiber substrate to obtain a shape-memory electrode. The shape-memory functionality could be realized by the fiber substrate at core, and the conductive component on the surface could be further modified to realize energy harvesting, energy storage or other capabilities. Up to date, several high-performance shape-memory fiber devices have been developed based on shape-memory alloys and polymers.

Shape-memory effect of shape-memory alloys (including NiTi-based, Cu-based, Fe-based, and intermetallic compounds) is based on the phase shift between austenitic and martensite crystal structures [73, 74]. When heated to the transition temperature, the plastic deformation could recover to the original state. For example, a shape-memory fiber electrode was fabricated by repeated brushing porous carbon dodecahedra mixture onto the NiTi alloy wire [75]. Deformed fiber electrodes could immediately recover to the initial shape within 1 min when the surrounding temperature reached 35 °C. A tensile strength of 95.8 MPa was achieved on the basis of the NiTi alloy wire. A shape-memory fiber supercapacitor was then fabricated using the shape-memory electrodes, which displayed a high volumetric energy density (8.9 mWh/cm^3) and power density (1080 mW/cm^3) based on porous carbon dodecahedra with high porosity and large surface. As a demonstration, a sleeve knitted with these shape-memory supercapacitors could recover to the original shape in a warm environment (about 35 °C), exhibiting the advantages in wearable electronic devices.

On the other hand, shape-memory polymers such as polyurethane/polyurea, polyethylene, polyisoprene, polyester, and styrene-butadiene copolymers can be employed for shape-memory fiber devices towards lightweight and stretchability [76, 77]. For instance, a shape-memory fiber electrode was produced by wrapping aligned MWCNT sheets onto a shape-memory thermoplastic polyurethanes (TPU) fiber [29]. The functionality and conductivity could be realized by TPU substrate and outside MWCNT, respectively. The electrical resistance was about 0.55 kΩ with a wrapping helical angle of 60°, and increased by about 0.5 kΩ when stretched up to 100%. The fiber-shaped supercapacitor was fabricated by using the above shape-memory electrodes and PVA gel. It could be manipulated in both bending and elongating shapes or even more complex architectures (Fig. 6a), and recovered to its original shape when the motion of the molecular chain was activated under the thermal transition temperature. This fiber-shaped supercapacitor was able to be stretched up to 100% and maintained elongated, simultaneously reserving 80% capacity (Fig. 6b). No significant decrease on the electrochemical property was observed after 500 stretching-recovering cycles at a strain of 50%, suggesting the excellent shape-memory reversibility of the obtained fiber devices. However, the relatively low tensile strength, slow recovery rate and high triggering temperature still need further enhancement towards practical applications.

Self-Healing Fiber Devices

Considering the daily wearing scenario, fiber devices are prone to encounter mechanical damage during use, which would result in the failure of the device or even the entire module. Inspired by the self-healing capability of our skin, self-healing fiber devices have been developed to enhance the adaptability and lifetime in wearable applications.

A pioneering work on self-healing fiber-shaped supercapacitor was presented in 2014 (Fig. 6c), using a self-healing fiber electrode with a core–shell configuration [32]. Briefly, a self-healing fiber containing abundant hydrogen bonds was synthesized via a modified Leibler method, which was capable of recovering the mechanical properties upon breaking at room temperature. MWCNT sheets were uniformly wrapped on the healable rubber fiber as a conductive sheath, resulting in a mechanically and electrically healable fiber electrode. The fractured interface of self-healable fiber and oriented MWCNT sheets reconnected due to the present of hydrogen bonds and van der Waals forces. The tensile strength of this fiber could be maintained by 72.5% after five breaking-healing cycles, and the electrical resistance showed no significant change after healing. In comparison, the composite fiber based on random Ag nanowire or MWCNT showed obviously increased electrical resistances (increased by 115 Ω and 5.0×10^4 Ω, respectively), due to the insufficient conductive network derived from different conductive building blocks. The obtained self-healing fiber-shaped supercapacitor with aligned MWCNT sheets could maintain 82.6% of the original specific capacity after five breaking-healing cycles, indicating an improved lifetime when structure damage occurred.

In another example, rGO fiber electrode in a spring shape was encapsulated in a PU shell that could be self-healed based on the reversible formation of hydrogen bonds. The healing process was performed by connecting the two breaking ends together, and the recovery on conductivity was thus highly dependent on the accurate connecting of the rGO network. The obtained supercapacitor showed a 54.2% capacity retention after three breaking-healing cycles (Fig. 6d) [57]. Obviously, the healing efficiencies were greatly affected by the connecting of the broken conducting network. To realize efficient and accurate recombination of the broken

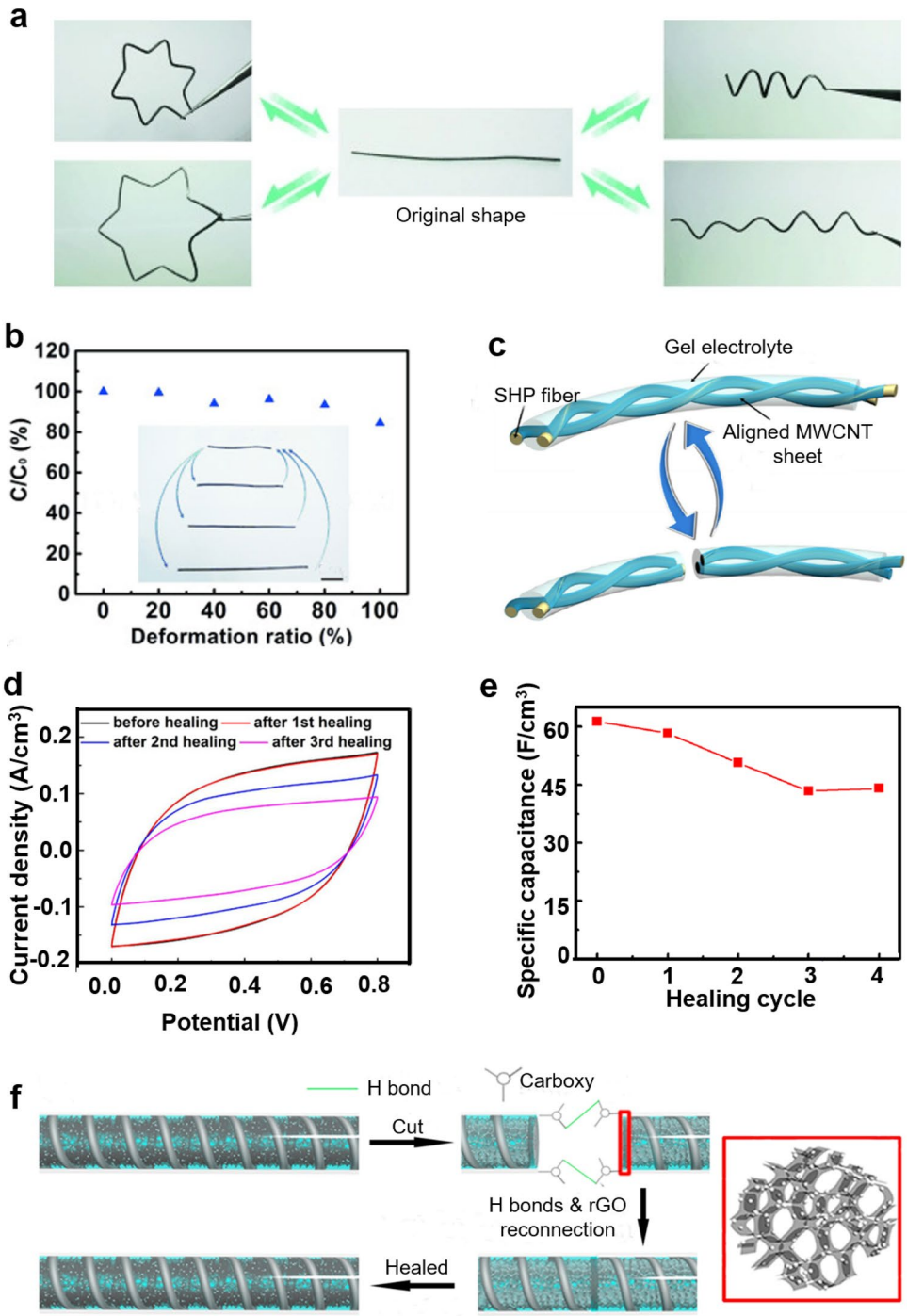

Fig. 6 **a** Photographs of a fiber-shaped shape-memory supercapacitor with different shapes. **b** Variation of the capacitance on different deformation ratio of a fiber-shaped shape-memory supercapacitor (scale bar: 2 cm) [29]. Reproduced with permission. Copyright 2015, Wiley–VCH. **c** Schematic illustration to the self-healing process of a fiber-shaped supercapacitor [32]. Reproduced with permission. Copyright 2014, Wiley–VCH. **d** Cyclic voltammetry curves of self-healing supercapacitors with rGO fiber electrode [57]. Reproduced with permission. Copyright 2017, American Chemical Society. **e** The specific capacitance retention of a magnet-assistant self-healing supercapacitor at different healing cycles [78]. Reproduced with permission. Copyright 2015, American Chemical Society. **f** Schematic illustration of the self-healing mechanism of a fiber-shaped lithium-ion battery [80]. Reproduced with permission. Copyright 2018, Elsevier

networks, self-healing process assisted by magnetic attraction was performed [78]. To prepare magnetic response fiber electrodes, Fe_3O_4 particles were anchored on the fiber by microwave-assisted hydrothermal method followed by an annealing process to enhance the attachment. In addition, a layer of electro-depositing PPy was incorporated to protect the magnetic particles, and provide pseudocapacitance to enhance the energy storage performances. A self-healable fiber-shaped supercapacitor was assembled by wrapping the magnetic response fiber electrodes with a self-healing carboxylated PU shell. The specific capacity could restore 71.8% of the initial value after four breaking-healing cycles based on the synergistic effect of the self-healing PU shell and magnetic force (Fig. 6e). To further enhance the self-healing capability of the fiber supercapacitor, self-healing electrolyte was also developed to accommodate the structural damage during use. For example, an asymmetric fiber supercapacitor based on PPy/Ni_3S_2-CF electrodes could maintain the structural integrity without any obvious "scar" at the electrolyte/electrode interface, due to the hydrogen bond formed by the diol-borate in the electrolyte [79].

Besides self-healing fiber supercapacitors, the healing capability was also realized in fiber-shaped lithium-ion batteries aiming at a higher energy density. For instance, on the basis of a rGO/SnO_2 fiber anode and a spring-shaped rGO/$LiCoO_2$ fiber cathode, a self-healing

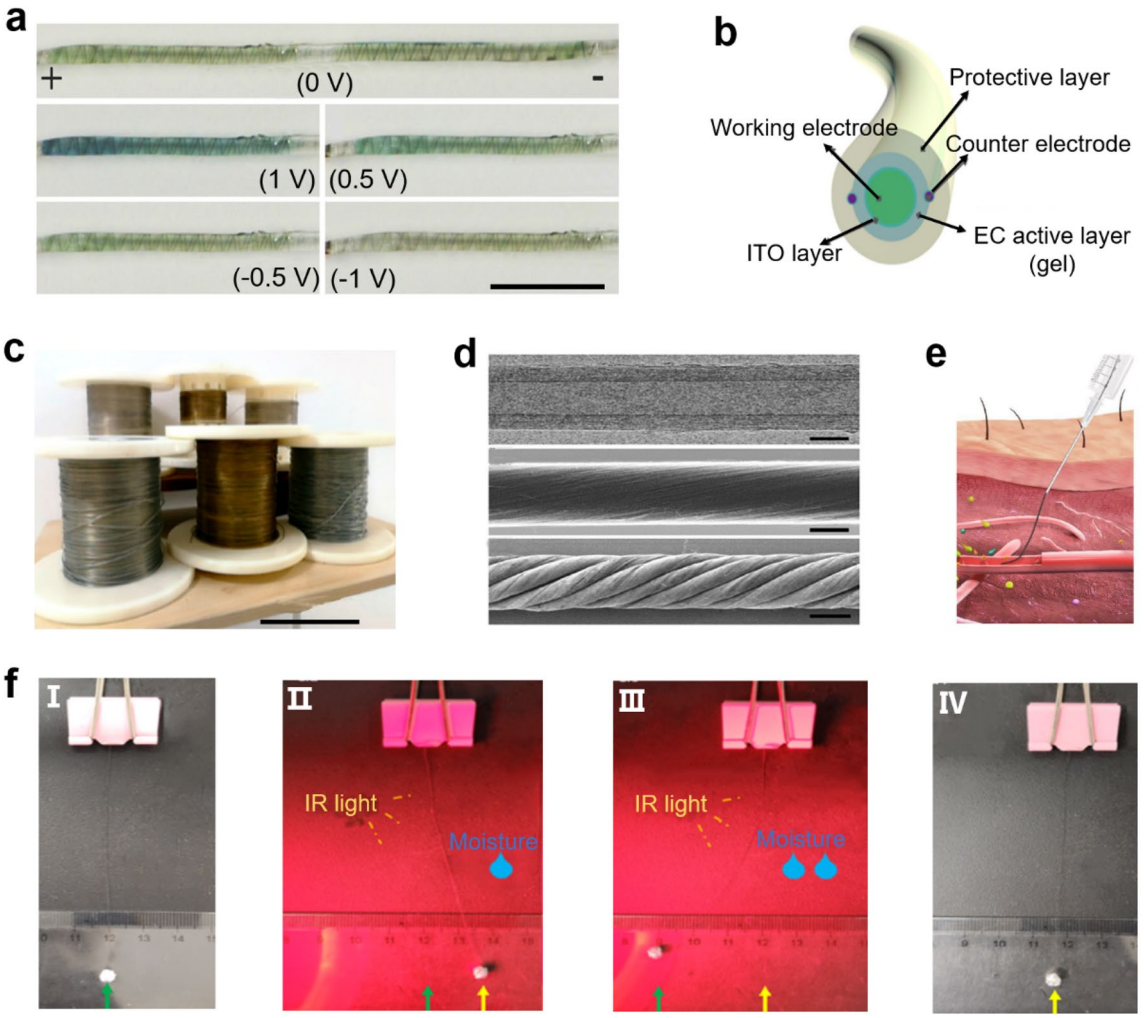

Fig. 7 **a** Chromatic transitions of the fiber-shaped supercapacitor during a charge–discharge cycle (scale bar: 5 mm) [30]**.** Reproduced with permission. Copyright 2014, Wiley–VCH. **b** Schematic illustration of an electrochromic fiber based on the parallel dual-counter-electrode structure. **c** Photograph of long electrochromic fibers with lengths of hundreds of meters (scale bar: 10 cm) [83]. Reproduced with permission. Copyright 2020, American Chemical Society. **d** TEM image of a MWCNTs (scale bar: 3 nm); SEM images of a single-ply fiber (scale bar: 6 μm) and multi-ply sensing fiber bundle (scale bar: 20 μm). **e** Injection of the fiber sensor into blood vessel [91]. Reproduced with permission. Copyright 2020, Nature Publishing Group. **f** Spherical load produced a synergistic response of the forward and backward movement with the stimulation of infrared radiation light and moisture [99]. Reproduced with permission. Copyright 2021, American Chemical Society

lithium-ion battery fiber was prepared using a healable PU shell (Fig. 6f) [80]. The obtained fiber device afforded a specific capacity of 99.5 mAh/g, and retained a specific capacity of 50.1 mAh/g (50.3% of the original capacity) after five breaking-healing cycles, suggesting their potentials in wearable applications with promoted adaptability and lifetime.

Electrochromic Fiber Devices

Electrochromism refers to the reversible optical property change that occurs when a certain material is electrochemically oxidized [81]. Particularly, the incorporation of electrochromism with energy storage/harvesting devices into one single fiber can visually display the real-time working states of the devices, which drew broad attentions in the past decade towards intelligent wearable devices.

Based on a coaxial structure, an electrochromic fiber supercapacitor using PANI as the electrochromic material could display different colors including light yellow, blue and green at different voltages (Fig. 7a) [30]. MWCNTs were first wound onto an elastic fiber, followed by electrodeposition of PANI on MWCNTs serving as the electrochromic component to provide visible information on the working status of the fiber device. The specific capacity of 255.5 F/g (0.189 mF/cm) and a strain of 100% could be achieved. Fiber devices composed of parallel coil electrodes could realize multicolors due to the stacking effect of two electrochromic electrodes. For instance, two helix Au electrodes were prepared by a template method, in which Au layer was sputter-coated on a polyvinyl chloride (PVC) fiber with two adhesive tapes as masks [82]. After removing the tapes, electrochromic supercapacitors were assembled by depositing various electrochromic materials such as tungsten oxide and poly(3-methylthiophene) onto the helix electrode using $LiClO_4$/poly(methyl methacrylate) as a solid electrolyte. The resulted devices could realize multicolor changes based on the stacking effect of two helix electrodes, leading to reversible electrochromism with fast response and various colors (i.e., dark green, gold, and dark red).

Furthermore, it is attractive to realize continuous processing and effective protection of the electrochromic fibers. For example, with viologen as the active material, electrochromic fibers based on a parallel dual-counter-electrode structure were continuously fabricated via home-made equipment (Fig. 7b, c) [83]. Due to the effective protection of electrochemical anticorrosive layer and outer polymer protective layer, the electrochemical and environmental stabilities were significantly improved. This electrochromic fiber could be woven into flexible textiles that realized proof-of-concept camouflage military clothes. Although several impressive electrochromic devices have been achieved, the electrochromic materials with superior chromogenic performances are still needed. The recently developed poly(ionic liquid)s with available structural design could be a promising direction to explore [84].

Other Functional Fiber Devices

Apart from the above functions, several other functions have been also realized to meet the specific requirement in potential applications. For instance, the magnetic response could realize remote collection, movement and fixing of delicate fiber devices. A novel magnetic response fiber electrode was developed via the incorporation of Fe_3O_4 nanoparticles into MWCNT fiber via a dry-spinning method [55]. With an Fe_3O_4 weight percentage of 16.6%, a high saturation magnetization of 17.9 emu/g was achieved. On the basis of the magnetic response capability of Fe_3O_4/MWCNT electrode, the resulted photovoltaic wire could be attached onto and detached from a substrate using magnetic field. Such magnetic responsive fiber devices could be particularly promising for some special application scenarios such as aerospace when conventional operations such as separation and movement are not fully compatible.

Flexible fiber sensors with compliant mechanical properties can convert environmental stimuli into electronic signal to provide related information. Up to date, a broad range of physical signals such as pulse beat, temperature and humidity can be detected by fiber sensors [85–87]. Efforts have been devoted to developing multifunctional electronic textiles with multidimensional position recognition capability and property improvement such as sensitivity, selectivity, detection limit, response time etc. For example, an intelligent Kevlar/MXene wearable fabric has been fabricated with temperature-responsive, respiratory monitoring and liquid molecular recognition capabilities via a continuous wet-spinning method [88]. It exhibited ultrafast responsiveness (90 ms), resilience (110 ms) and precise recognition ability. In another example, Wang et al. [89] have fabricated a tactile sensor unit without any substrates by using ionic gel as glue at intersection point of MXene/cotton electrodes, showing a high sensitivity of 9.62 k/Pa (0–40 kPa), an ultralow detection limit of 0.884 Pa, and a short response time of 24 ms.

In addition, monitoring physiological signals in vivo (such as disease biomarkers, ions, glutamate, lactic acid, etc.) represents a promising direction in smart healthcare [90]. Inspired by the hierarchical structure and helical assembly of muscle, Wang et al. prepared a multi-ply sensing fiber bundle by twisting multiple single-ply sensing fibers (Fig. 7d) [91]. This sensing fiber could be implanted in vivo by simple syringe injection with no inflammation (Fig. 7e). Multiple disease biomarkers such as H_2O_2, calcium ions and

glucose would be monitored, providing a powerful platform for in-vivo disease detection. Furthermore, high sensitivity and stability are of significance for in-vivo detection. Taking advantages of the local amplification effect of organic electrochemical transistors (OECTs), a fiber-shaped organic electrochemical transistor had been designed for detection of several types of physiological signals. The obtained fiber-shaped OECT showed a high sensitivity (detection limit of 100 nM), dynamical stability (maintained for 7 days in brain) and anti-interference capability [92].

Fiber actuators can realize complex movement by automatically changing their shapes in response to environmental stimulus [93, 94]. For example, a hybrid helical multi-ply composite fiber composed of MWCNT fiber and thermoplastic PU could produce a tensile actuation of 13.8% stroke and a contractive stress of 33 MPa via a joule heating process [95]. The excellent performance was benefited from the thermal volume expansion of the thermoplastic PU resin. However, the actuation driven by temperature usually encounters several problems including poor heat dissipation, easy oxidation, and lower energy conversion efficiency. To this end, moisture or humidity stimuli were also used for efficient actuation. For instance, oxygen plasma treatment enabled MWCNTs to possess a hydrophilic surface [26], based on which the actuator could rapidly shrink and rotate in response to water and humidity. Recently, other materials, e.g., linen fiber [96], spider dragline silk [97], lotus fiber [98] have been developed to fabricate the fiber actuations in respond to water or humidity. The exploration on other stimulations can be further conducted to promote the intelligence of fiber actuators.

Up to date, most of the fiber actuators can only respond to one single type of stimulation, i.e., they can hardly realize synergistic responses to multiple stimulations. Aiming at the above challenge, a fiber actuator with an intermittent spiral structure was constructed using sodium alginate and sodium alginate/graphene oxide [99]. This actuator could perform a controllable and programmable "forward-stagnation-reverse" synergistic response under the stimulations of infrared radiation (IR) light and moisture. As shown in Fig. 7f, when fixing one end of the fiber actuator, a spherical load was connected to the other end (Fig. 7f, I), which offset 17 mm to the right under IR stimulation and then maintained balance with the presence of moisture (Fig. 7f, II). Furthermore, the spherical load could be offset 30 mm to the left with the increase of the moisture (Fig. 7f, III). When both stimulations were removed, the fiber actuator and spherical load could return to the original position (Fig. 7f, IV). However, the scalability and cost issues limit the application of artificial fiber actuator. Meanwhile, solving the response hysteresis is also indispensable considering the real-world applications.

In recent years, smart wearable electronic devices have attracted intensive attention. Endowing fiber devices with various responsive functions would be an important step towards enhanced intelligence. As for shape-memory and self-healable functions, non-thermal triggers including electrical, magnetic and optical responses can be further pursued to meet the requirements in special application

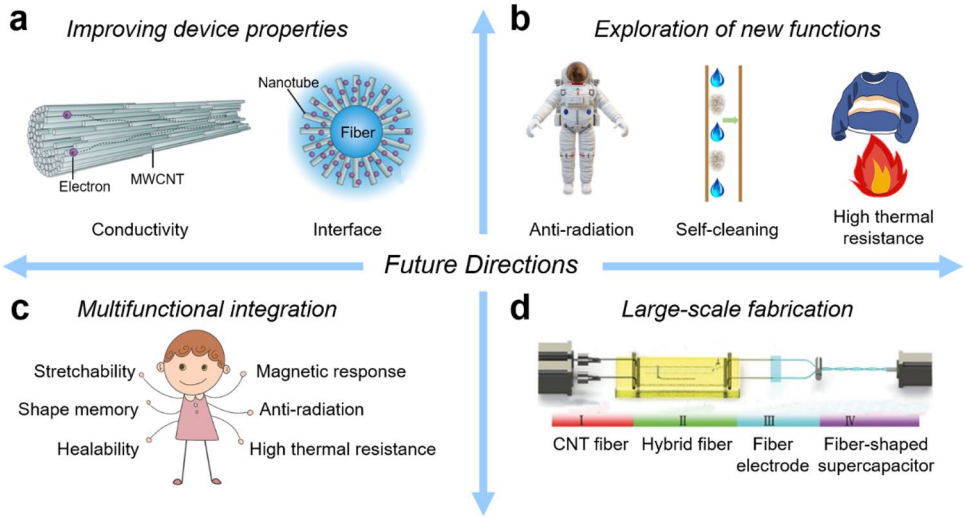

Fig. 8 Future directions of functional fiber devices. **a** Improving device properties via material and interface optimization [5]. Reproduced with permission. Copyright 2017, Nature Publishing Group. **b** Exploration of new functions such as anti-radiation, self-cleaning and thermal resistance. **c** Integration of multiple functions into one fiber device. **d** Large-scale fabrication of functional fiber devices based on a continuous production strategy [104]. Reproduced with permission. Copyright 2015, Wiley–VCH

scenarios, such as in aerospace and abysmal sea. Although the main parameters such as sensitivity and detection limit have been greatly improved, more efforts should be dedicated to improving the safety of flexible sensors, promoting their practical applications especially in terms of implanted devices.

Conclusion and Perspectives

In this Review, the recent advancements of functional fiber devices are mainly introduced and discussed. Several representative preparation methods of functional fiber electrodes are introduced in detail, followed by underlining the formed interfaces that are indispensable to the functionality and stability of the obtained devices. Although, much enthusiasm has been devoted to pushing forward the advancement of functional fiber devices, their fate is intertwined with the following challenges.

First, it is still needed to optimize the properties and processability of conductive materials towards scale-up production. As mentioned above, fiber electrodes based on carbon nanomaterials such as MWCNT and graphene are widely investigated as promising candidates for producing fiber devices, due to their remarkable strengths, electrical conductivities and specific surface areas. The electrical conductivity requires further optimization, which is of critical importance for reducing the inner resistances of long fiber devices (Fig. 8a). In addition, new processing techniques for continuous fiber production are also needed. Both wet and dry spinning methods have been developed for MWCNT fibers, but the low electrical conductivities and high costs are the core issues. The wet-spinning of graphene oxide fiber represents a promising strategy for continuous fabrication. However, the chemical or thermal reduction treatment are expected to be optimized towards a more efficient, economic and environment-friendly way. Encouragingly, it should be noted that the prices of carbon nanomaterials have dropped significantly (0.1–25 $/g) in recent years with the intensive efforts of both academic and industrial communities [100, 101]. This paves the way for mass production of fiber devices using carbon nanomaterials.

Second, the robustness of functionalized fiber-shaped devices should be taken into serious consideration when employed in practical applications. Frequent friction and torsion are common problems during use or washing, which can severely undermine the device performances. Thus, it is required to enhance the resistance on abrasion and torsion. For instance, the recently reported triboelectric fibers based on Fermat Spirals have provided an efficient strategy for improving the durability of wearable electronics [102]. In addition, the stretchability of fiber-shaped devices needs further improvement, accommodating the dramatic deformation in practical applications. As mentioned, both stretchable electrode and configuration are applicable strategy towards stretchable fiber devices. However, the introduction of elastic polymer substrates inevitably occupies the volume of the device without contributions to electrochemical performances. Thus, the balance between stretchability and electrochemical performances needs to be addressed. Particularly, the delamination is a tricky issue that leads to rapid degradation of the electrochemical performances (Fig. 8a). Therefore, realizing a strong interaction at the interface is a promising strategy. For example, the formed hydrogen bonds between eutectic gallium indium oxide and polymethacrylate can greatly enhance the adhesion, which makes a huge contribution to the stability of fiber-shaped devices [17, 103].

Third, the exploration on new functions is still required to enhance the device intelligence for different service environments (Fig. 8b). For instance, to avoid the possible peeling of the active materials from the fiber substrate during washing, it would be interesting to realize the self-cleaning function by introducing related components on the surface of fiber devices. In addition, other useful functions can be further developed to open up new possibilities in harsh conditions, e.g., high pressure, high temperature and radiation, etc. Moreover, the safety issue should be taken into serious consideration due to the close contact of fiber devices with the human body in all scenarios. Therefore, developing non-flammable electrodes and electrolytes could be a promising direction to pursue in the near future.

Fourth, the integration of various functional fiber devices remains to be challenging while promising direction. For instance, the integrated textiles which were woven from fiber-shaped devices with different functions could afford responses to a variety of different stimulations, which is certainly an exciting goal to pursue (Fig. 8c). However, the integration strategies of numerous fiber devices are still rather limited, mainly hindered by the challenges in connecting delicate fiber electrodes via a stable and integrated way. Up to date, although very few successes have been achieved towards this direction, the combination of functionalization and integration can open up a new path of fiber-shaped electronic devices.

Last but not the least, the scale-up production of functional fiber devices can be further explored. For instance, as a unique fabrication method of fiber devices, the rotation and translation process can significantly enhance the production efficiency of stretchable fiber electrodes. However, it is only compatible with spinnable MWCNTs on the basis of a dry-spinning method. More efforts can be devoted to accommodating more materials (metals, conductive polymers and other carbon materials) and techniques. For instance, a continuous fabricating process was developed to integrate fiber electrode preparation, electrolyte coating, device assembling and encapsulation into a continuous process [104] (Fig. 8d).

With the incorporation of functional components into this process, the functionalization of fiber devices can be readily achieved, although the device configuration and interfaces require further optimization. Particularly, it is crucial for developing industrially available equipment and process to fully meet the requirements on technology and cost towards real-world applications. We believe that with the in-depth and extensive investigation and collaboration throughout the world, fiber devices and the derived textile electronics will greatly change our lifestyle in an unexpected and fascinating way.

Declarations

Conflict of Interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

References

1. Lee GH, Moon H, Kim H, Lee GH, Kwon W, Yoo S, Myung D, Yun SH, Bao Z, Hahn SK. Multifunctional materials for implantable and wearable photonic healthcare devices. *Nat Rev Mater* **2020**;*5*:149.
2. Zeng W, Shu L, Li Q, Chen S, Wang F, Tao XM. Fiber-based wearable electronics: a review of materials, fabrication, devices, and applications. *Adv Mater* **2014**;*26*:5310.
3. Hashemi SA, Ramakrishna S, Aberle AG. Recent progress in flexible-wearable solar cells for self-powered electronic devices. *Energ Environ Sci* **2020**;*13*:685.
4. Zhu S, Ni JF, Li Y. Carbon nanotube-based electrodes for flexible supercapacitors. *Nano Res* **1825**;*2020*:13.
5. Sun H, Zhang Y, Zhang J, Sun XM, Peng HS. Energy harvesting and storage in 1D devices. *Nat Rev Mater* **2017**;*2*:1.
6. Kang M, Kim TW. Recent advances in fiber-shaped electronic devices for wearable applications. *Appl Sci* **2021**;*11*:6131.
7. Wang X, Jiang K, Shen G. Flexible fiber energy storage and integrated devices: recent progress and perspectives. *Mater Today* **2015**;*18*:265.
8. Jin L, Xiao X, Deng W, Nashalian A, He D, Raveendran V, Yan C, Su H, Chu X, Yang T. Manipulating relative permittivity for high-performance wearable triboelectric nanogenerators. *Nano Lett* **2020**;*20*:6404.
9. Chen G, Li Y, Bick M, Chen J. Smart textiles for electricity generation. *Chem Rev* **2020**;*120*:3668.
10. Shen S, Xiao X, Chen J. Wearable triboelectric nanogenerators for heart rate monitoring. *Chem Commun* **2021**;*57*:5871.
11. Zhang S, Bick M, Xiao X, Chen G, Nashalian A, Chen J. *Leveraging triboelectric nanogenerators for bioengineering Matter* **2021**;*4*:845.
12. Xiao X, Chen GR, Libanori A, Chen J. Wearable Triboelectric nanogenerators for therapeutics. *Trends Chem* **2021**;*3*:279.
13. Zou D, Wang D, Chu Z, Lv Z, Fan X. Fiber-shaped flexible solar cells. *Coord Chem Rev* **2010**;*254*:1169.
14. Casadio S, Sangiorgi N, Sangiorgi A, Dessì A, Zani L, Calamante M, Reginato G, Mordini A, Sanson A. Highly efficient long thin-film fiber-shaped dye sensitized solar cells based on a fully organic sensitizer. Sol Energy Mater Sol Cells. 2021;224:110986.
15. Dong YH, Zou YS, Song JZ, Zhu ZF, Li JH, Zeng HB. Self-powered fiber-shaped wearable omnidirectional photodetectors. *Nano Energy* **2016**;*30*:173.
16. Chu X, Chen GR, Xiao X, Wang ZX, Yang T, Xu Z, Huang HC, Wang YH, Yan C, Chen NJ, Zhang HT, Yang WQ, Chen J. Air-stable conductive polymer ink for printed wearable micro-supercapacitors. *Small* **2021**;*17*:2100956.
17. Duan M, Ren Y, Sun X, Zhu X, Wang X, Sheng L, Liu J. EGaIn fiber enabled highly flexible supercapacitors. *ACS Omega* **2021**;*6*:24444.
18. Mo F, Liang G, Huang Z, Li H, Wang D, Zhi C. An overview of fiber-shaped batteries with a focus on multifunctionality, scalability, and technical difficulties. *Adv Mater* **2020**;*32*:1902151.
19. Shi FY, Chen CH, Xu ZL. Recent advances on electrospun nanofiber materials for post-lithium ion batteries. *Adv Fiber Mater* **2021**;*3*:275.
20. Chong WG, Huang JQ, Xu ZL, Qin X, Wang X, Kim JK. Lithium–sulfur battery cable made from ultralight, flexible graphene/carbon nanotube/sulfur composite fibers. *Adv Funct Mater* **2017**;*27*:1604815.
21. Ouyang Y, Zong W, Wang J, Xu Z, Mo L, Lai F, Xu Z-L, Miao Y-E, Liu T. Multi-scale uniform Li regulation triggered by tunable electric field distribution on oxygen-functionalized porous framework for flexible Li-S full batteries. *Energy Stor Mater* **2021**;*42*:68.
22. Feng JY, Chen CR, Sun XM, Peng HS. Implantable fiber biosensors based on carbon nanotubes. *Accounts Mater Res* **2021**;*2*:138.
23. Wang Y, Zhou Z, Qing X, Zhong W, Liu Q, Wang W, Li M, Liu K, Wang D. Ion sensors based on novel fiber organic electrochemical transistors for lead ion detection. *Anal Bioanal Chem* **2016**;*408*:5779.
24. Wu R, Ma L, Hou C, Meng Z, Guo W, Yu W, Yu R, Hu F, Liu XY. Silk composite electronic textile sensor for high space precision 2D combo temperature–pressure sensing. *Small* **2019**;*15*:1901558.
25. Leng XQ, Hu XY, Zhao WB, An BG, Zhou X, Liu ZF. Recent advances in twisted-fiber artificial muscles. *Adv Intell Syst* **2021**;*3*:2000185.
26. He S, Chen P, Qiu L, Wang B, Sun X, Xu Y, Peng H. A mechanically actuating carbon-nanotube fiber in response to water and moisture. *Angew Chem Int Ed* **2015**;*54*:14880.
27. Chen P, Xu Y, He S, Sun X, Pan S, Deng J, Chen D, Peng H. Hierarchically arranged helical fibre actuators driven by solvents and vapours. *Nat Nanotechnol* **2015**;*10*:1077.
28. He J, Lu C, Jiang H, Han F, Shi X, Wu J, Wang L, Chen T, Wang J, Zhang Y, Yang H, Zhang G, Sun X, Wang B, Chen P, Wang Y, Xia Y, Peng H. Scalable production of high-performing woven lithium-ion fibre batteries. *Nature* **2021**;*597*:57.
29. Deng J, Zhang Y, Zhao Y, Chen P, Cheng X, Peng H. A shape-memory supercapacitor fiber. *Angew Chem Int Ed* **2015**;*54*:15419.
30. Chen X, Lin H, Deng J, Zhang Y, Sun X, Chen P, Fang X, Zhang Z, Guan G, Peng H. Electrochromic fiber-shaped supercapacitors. *Adv Mater* **2014**;*26*:8126.
31. Yang Z, Deng J, Chen X, Ren J, Peng H. A highly stretchable, fiber-shaped supercapacitor. *Angew Chem Int Ed* **2013**;*52*:13453.
32. Sun H, You X, Jiang Y, Guan G, Fang X, Deng J, Chen P, Luo Y, Peng H. Self-healable electrically conducting wires for wearable microelectronics. *Angew Chem Int Ed* **2014**;*53*:9526.
33. Zhang X, Lin H, Shang H, Xu J, Zhu J, Huang W. Recent advances in functional fiber electronics. *SusMat* **2021**;*1*:105.
34. Wang L, Fu X, He J, Shi X, Chen T, Chen P, Wang B, Peng H. Application challenges in fiber and textile electronics. Adv Mater. 2020;32:e1901971.
35. Fakharuddin A, Li HZ, Di Giacomo F, Zhang TY, Gasparini N, Elezzabi AY, Mohanty A, Ramadoss A, Ling J, Soultati A,

Tountas M, Schmidt-Mende L, Argitis P, Jose R, Nazeeruddin MK, Yusoff AB, Vasilopoulou M. *Fiber-Shaped Electron Devices Adv Energy Mater* **2021**;*11*:2101443.
36. Hou SC, Lv ZB, Wu HW, Cai X, Chu ZZ, Yiliguma, Zou DC. Flexible conductive threads for wearable dye-sensitized solar cells. *J Mater Chem*. **2012**; *22*:6549.
37. Kim B, Koncar V, Devaux E, Dufour C, Viallier P. Electrical and morphological properties of PP and PET conductive polymer fibers. *Synth Met* **2004**;*146*:167.
38. Kaynak A, Najar SS, Foitzik RC. Conducting nylon, cotton and wool yarns by continuous vapor polymerization of pyrrole. *Synth Met* **2008**;*158*:1.
39. Gasana E, Westbroek P, Hakuzimana J, De Clerck K, Priniotakis G, Kiekens P, Tseles D. Electroconductive textile structures through electroless deposition of polypyrrole and copper at polyaramide surfaces. *Surf Coat Technol* **2006**;*201*:3547.
40. Abbasi AR, Akhbari K, Morsali A. Dense coating of surface mounted CuBTC metal–organic framework nanostructures on silk fibers, prepared by layer-by-layer method under ultrasound irradiation with antibacterial activity. *Ultrason Sonochem* **2012**;*19*:846.
41. Zhao Y, Dong D, Wang Y, Gong S, An T, Yap LW, Cheng W. Highly stretchable fiber-shaped supercapacitors based on ultrathin gold nanowires with double-helix winding design. *ACS Appl Mater Interfaces* **2018**;*10*:42612.
42. Tebyetekerwa M, Xu Z, Li W, Wang X, Marriam I, Peng S, Ramkrishna S, Yang S, Zhu M. Surface self-assembly of functional electroactive nanofibers on textile yarns as a facile approach toward super flexible energy storage. *ACS Appl Energy Mater* **2018**;*1*:377.
43. Yin Z, Jian M, Wang C, Xia K, Liu Z, Wang Q, Zhang M, Wang H, Liang X, Liang X. Splash-resistant and light-weight silk-sheathed wires for textile electronics. *Nano Lett* **2018**;*18*:7085.
44. Shao W, Tebyetekerwa M, Marriam I, Li W, Wu Y, Peng S, Ramakrishna S, Yang S, Zhu M. Polyester@ MXene nanofibers-based yarn electrodes. *J Power Sources* **2018**;*396*:683.
45. Yang QY, Xu Z, Gao C. Graphene fiber based supercapacitors: strategies and perspective toward high performances. *J Energy Chem* **2018**;*27*:6.
46. Wu JX, Hong Y, Wang BJ. The applications of carbon nanomaterials in fiber-shaped energy storage devices. *J Semicond*. **2018**; *39*:011004.
47. Jang Y, Kim SM, Spinks GM, Kim SJ. Carbon nanotube yarn for fiber-shaped electrical sensors, actuators, and energy storage for smart systems. *Adv Mater* **2020**;*32*:1902670.
48. Benzigar MR, Dasireddy VDBC, Guan XW, Wu T, Liu GZ. Advances on emerging materials for flexible supercapacitors: current trends and beyond. *Adv Funct Mater* **2020**;*30*:2002993.
49. Meng WS, Nie MY, Liu ZY, Zhou J. Buckled fiber conductors with resistance stability under strain. *Adv Fiber Mater* **2021**;*3*:149.
50. Zhang T, Zong W, Ouyang Y, Wu Y, Miao Y-E, Liu T. Carbon fiber supported binary metal sulfide catalysts with multidimensional structures for electrocatalytic nitrogen reduction reactions over a wide pH range. *Adv Fiber Mater* **2021**;*3*:229.
51. Wang JN, Luo XG, Wu T, Chen Y. High-strength carbon nanotube fibre-like ribbon with high ductility and high electrical conductivity. *Nat Commun* **2014**;*5*:3848.
52. Oh E, Cho H, Kim J, Kim JE, Yi Y, Choi J, Lee H, Im YH, Lee KH, Lee WJ. Super-strong carbon nanotube fibers achieved by engineering gas flow and postsynthesis treatment. *ACS Appl Mater Interfaces* **2020**;*12*:13107.
53. Zhang Z, Deng J, Li X, Yang Z, He S, Chen X, Guan G, Ren J, Peng H. Superelastic supercapacitors with high performances during stretching. *Adv Mater* **2015**;*27*:356.
54. Lima MD, Fang SL, Lepro X, Lewis C, Ovalle-Robles R, Carretero-Gonzalez J, Castillo-Martinez E, Kozlov ME, Oh JY, Rawat N, Haines CS, Haque MH, Aare V, Stoughton S, Zakhidov AA, Baughman RH. Biscrolling nanotube sheets and functional guests into yarns. *Science* **2011**;*331*:51.
55. Sun H, Yang Z, Chen X, Qiu L, You X, Chen P, Peng H. Photovoltaic wire with high efficiency attached onto and detached from a substrate using a magnetic field. *Angew Chem Int Ed* **2013**;*52*:8276.
56. Xu Z, Gao C. Graphene chiral liquid crystals and macroscopic assembled fibres. *Nat Commun* **2011**;*2*:571.
57. Wang S, Liu N, Su J, Li L, Long F, Zou Z, Jiang X, Gao Y. Highly stretchable and self-healable supercapacitor with reduced graphene oxide based fiber springs. *ACS Nano* **2017**;*11*:2066.
58. Kadumudi FB, Hasany M, Pierchala MK, Jahanshahi M, Taebnia N, Mehrali M, Mitu CF, Shahbazi MA, Zsurzsan TG, Knott A, Andresen TL, Dolatshahi-Pirouz A. The manufacture of unbreakable bionics via multifunctional and self-healing silk-graphene hydrogels. *Adv Mater*. **2021**;*33*:e2100047.
59. Wang YB, Chen CJ, Xie H, Gao TT, Yao YG, Pastel G, Han XG, Li YJ, Zhao JP, Fu K, Hu LB. 3D-printed all-fiber Li-Ion battery toward wearable energy storage. *Adv Funct Mater* **2017**;*27*:1703140.
60. Cai ZB, Li L, Ren J, Qiu LB, Lin HJ, Peng HS. Flexible, weavable and efficient microsupercapacitor wires based on polyaniline composite fibers incorporated with aligned carbon nanotubes. *J Mater Chem A* **2013**;*1*:258.
61. Li H, Liu Z, Liang G, Huang Y, Huang Y, Zhu M, Pei Z, Xue Q, Tang Z, Wang Y, Li B, Zhi C. Waterproof and tailorable elastic rechargeable yarn zinc ion batteries by a cross-linked polyacrylamide electrolyte. *ACS Nano* **2018**;*12*:3140.
62. Yang Z, Sun H, Chen T, Qiu L, Luo Y, Peng H. Photovoltaic wire derived from a graphene composite fiber achieving an 8.45% energy conversion efficiency. *Angew Chem*. **2013**;*125*:7693.
63. Li HP, Guo JJ, Sun H, Fang X, Wang DH, Peng HS. Stable hydrophobic ionic liquid gel electrolyte for stretchable fiber-shaped dye-sensitized solar cell. *Chemnanomat* **2015**;*1*:399.
64. Li M, Li Z, Ye X, Zhang X, Qu L, Tian M. Tendril-inspired 900% ultrastretching fiber-based zn-ion batteries for wearable energy textiles. *ACS Appl Mater Interfaces* **2021**;*13*:17110.
65. Guo K, Ma Y, Li H, Zhai T. Flexible wire-shaped supercapacitors in parallel double helix configuration with stable electrochemical properties under static/dynamic bending. *Small* **2016**;*12*:1024.
66. Wu J, Wang Z, Liu W, Wang L, Xu F. Bioinspired superelastic electroconductive fiber for wearable electronics. *ACS Appl Mater Interfaces* **2019**;*11*:44735.
67. Liu K, Kong B, Liu W, Sun YM, Song MS, Chen J, Liu YY, Lin DC, Pei A, Cui Y. Stretchable lithium metal anode with improved mechanical and electrochemical cycling stability. *Joule* **1857**;*2018*:2.
68. Marriam I, Wang X, Tebyetekerwa M, Chen G, Zabihi F, Pionteck J, Peng S, Ramakrishna S, Yang S, Zhu M. A bottom-up approach to design wearable and stretchable smart fibers with organic vapor sensing behaviors and energy storage properties. *J Mater Chem A* **2018**;*6*:13633.
69. Yu S, Wang X, Xiang H, Zhu L, Tebyetekerwa M, Zhu M. Superior piezoresistive strain sensing behaviors of carbon nanotubes in one-dimensional polymer fiber structure. *Carbon* **2018**;*140*:1.
70. Zhang Y, Bai WY, Cheng XL, Ren J, Weng W, Chen PN, Fang X, Zhang ZT, Peng HS. Flexible and stretchable lithium-ion batteries and supercapacitors based on electrically conducting carbon nanotube fiber springs. *Angew Chem Int Ed* **2014**;*53*:14564.
71. Lee JM, Chun S, Son W, Suh D, Kim SH, Kim H, Lee D, Kim Y, Kim YK, Lim SK, Choi C. DNA-inspired, highly packed supercoil battery for ultra-high stretchability and capacity. *Nano Energy*. **2021**;*85*:106034.

72. Jahid MA, Hu J, Zhuo H. Stimuli-responsive polymers in coating and laminating for functional textile. *Smart Textile Coat Lamin*. **2019**;155.
73. Brinson LC. One-dimensional constitutive behavior of shape memory alloys: thermomechanical derivation with non-constant material functions and redefined martensite internal variable. *J Intell Mater Syst Struct* **1993**;*4*:229.
74. Song G, Ma N, Li HN. Applications of shape memory alloys in civil structures. *Eng Struct* **2006**;*28*:1266.
75. Shi M, Yang C, Song X, Liu J, Zhao L, Zhang P, Gao L. Recoverable wire-shaped supercapacitors with ultrahigh volumetric energy density for multifunctional portable and wearable electronics. *ACS Appl Mater Interfaces* **2017**;*9*:17051.
76. Berg GJ, McBride MK, Wang C, Bowman CN. New directions in the chemistry of shape memory polymers. *Polymer* **2014**;*55*:5849.
77. Xia Y, He Y, Zhang F, Liu Y, Leng J. A review of shape memory polymers and composites: mechanisms, materials, and applications. *Adv Mater*. **2021**;*33*:e2000713.
78. Huang Y, Huang Y, Zhu M, Meng W, Pei Z, Liu C, Hu H, Zhi C. Magnetic-assisted, self-healable, yarn-based supercapacitor. *ACS Nano* **2015**;*9*:6242.
79. Wu XM, Meng L, Wang QG, Zhang WZ, Wang Y. A self-healable asymmetric fibered-supercapacitor integrated in self-supported inorganic nanosheets array and conducting polymer electrodes. *Chem Eng J* **2018**;*352*:423.
80. Rao JY, Liu NS, Zhang Z, Su J, Li LY, Xiong L, Gao YH. All-fiber-based quasi-solid-state lithium-ion battery towards wearable electronic devices with outstanding flexibility and self-healing ability. *Nano Energy* **2018**;*51*:425.
81. Chen K-C, Hsu C-Y, Hu C-W, Ho K-C. A complementary electrochromic device based on Prussian blue and poly (ProDOT-Et_2) with high contrast and high coloration efficiency. *Sol Energy Mater Sol Cells* **2011**;*95*:2238.
82. Zhou Y, Fang J, Wang HX, Zhou H, Yan GL, Zhao Y, Dai LM, Lin T. Multicolor electrochromic fibers with helix-patterned electrodes. *Adv Electron Mater* **2018**;*4*:1800104.
83. Fan H, Li K, Liu X, Xu K, Su Y, Hou C, Zhang Q, Li Y, Wang H. Continuously processed, long electrochromic fibers with multi-environmental stability. *ACS Appl Mater Interfaces* **2020**;*12*:28451.
84. Yang BG, Yang GJ, Zhang YM, Zhang SXA. Recent advances in poly(ionic liquid)s for electrochromic devices. *J Mater Chem C* **2021**;*9*:4730.
85. Zhang Y, Wang H, Lu H, Li S, Zhang Y. Electronic fibers and textiles: recent progress and perspective. *iScience*. **2021**;*24*:102716.
86. Zhao HX, Zhou YL, Cao ST, Wang YF, Zhang JX, Feng SX, Wang JC, Li DC, Kong DS. Ultrastretchable and washable conductive microtextiles by coassembly of silver nanowires and elastomeric microfibers for epidermal human-machine interfaces. *ACS Mater Lett* **2021**;*3*:912.
87. Zhong W, Liu C, Xiang C, Jin Y, Li M, Liu K, Liu Q, Wang Y, Sun G, Wang D. Continuously producible ultrasensitive wearable strain sensor assembled with three-dimensional interpenetrating Ag nanowires/polyolefin elastomer nanofibrous composite yarn. *ACS Appl Mater Interfaces* **2017**;*9*:42058.
88. Cheng B, Wu P. Scalable fabrication of kevlar/$Ti_3C_2T_x$ MXene intelligent wearable fabrics with multiple sensory capabilities. *ACS Nano* **2021**;*15*:8676.
89. Wang P, Li GX, Liu J, Hou ZR, Meng CZ, Guo SJ. Flexible, freestanding, ultrasensitive, and iontronic tactile sensing textile. *ACS Appl Electron Mater* **2021**;*3*:2195.
90. Wang JJ, Wang LY, Feng JY, Tang CQ, Sun XM, Peng HS. Long-term in vivo monitoring of chemicals with fiber sensors. *Adv Fiber Mater* **2021**;*3*:47.
91. Wang L, Xie S, Wang Z, Liu F, Yang Y, Tang C, Wu X, Liu P, Li Y, Saiyin H, Zheng S, Sun X, Xu F, Yu H, Peng H. Functionalized helical fibre bundles of carbon nanotubes as electrochemical sensors for long-term in vivo monitoring of multiple disease biomarkers. *Nat Biomed Eng* **2020**;*4*:159.
92. Wu XY, Feng JY, Deng J, Cui ZC, Wang LY, Xie SL, Chen CR, Tang CQ, Han ZQ, Yu HB, Sun XM, Peng HS. Fiber-shaped organic electrochemical transistors for biochemical detections with high sensitivity and stability. *Sci China Chem* **2020**;*63*:1281.
93. Yang Y, Wang H, Zhang S, Wei Y, He X, Wang J, Zhang Y, Ji Y. Vitrimer-based soft actuators with multiple responsiveness and self-healing ability triggered by multiple stimuli. *Matter* **2021**;*4*:3354.
94. Naciri J, Srinivasan A, Jeon H, Nikolov N, Keller P, Ratna BR. Nematic elastomer fiber actuator. *Macromolecules* **2003**;*36*:8499.
95. Song Y, Zhou S, Jin K, Qiao J, Li D, Xu C, Hu D, Di J, Li M, Zhang Z, Li Q. Hierarchical carbon nanotube composite yarn muscles. *Nanoscale* **2018**;*10*:4077.
96. Wang Y, Miao MH. Helical shape linen artificial muscles responsive to water. *Smart Mater Struct*. **2021**;*30*:075031.
97. Dong L, Qiao J, Wu Y, Ren M, Wang Y, Shen X, Wei X, Wang X, Di J, Li Q. Programmable contractile actuations of twisted spider dragline silk yarns. *ACS Biomater Sci Eng* **2021**;*7*:482.
98. Wang Y, Wang Z, Lu Z, Jung de Andrade M, Fang S, Zhang Z, Wu J, Baughman RH. Humidity- and water-responsive torsional and contractile lotus fiber yarn artificial muscles. *ACS Appl Mater Interfaces*. **2021**;*13*:6642.
99. Zheng QC, Jiang ZL, Xu XT, Xu CX, Zhu M, Chen C, Fu FF. Bio-inspired bicomponent fiber with multistimuli response to infrared light and humidity for smart actuators. *ACS Appl Polym Mater* **2021**;*3*:3131.
100. Zhang Q, Huang JQ, Qian WZ, Zhang YY, Wei F. The road for nanomaterials industry: a review of carbon nanotube production, post-treatment, and bulk applications for composites and energy storage. *Small* **2013**;*9*:1237.
101. Lin L, Peng H, Liu Z. Synthesis challenges for graphene industry. *Nat Mater* **2019**;*18*:520.
102. Zhang D, Yang W, Gong W, Ma W, Hou C, Li Y, Zhang Q, Wang H. Abrasion resistant/waterproof stretchable triboelectric yarns based on fermat spirals. Adv Mater. 2021; 2100782.
103. Chen G, Wang H, Guo R, Duan M, Zhang Y, Liu J. Superelastic EGaIn composite fibers sustaining 500% tensile strain with superior electrical conductivity for wearable electronics. *ACS Appl Mater Interfaces* **2020**;*12*:6112.
104. Wang B, Fang X, Sun H, He S, Ren J, Zhang Y, Peng H. Fabricating continuous supercapacitor fibers with high performances by integrating all building materials and steps into one process. *Adv Mater* **2015**;*27*:7854.

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