Fiber-shaped Supercapacitors: Advanced Strategies toward High-performances and Multi-functions

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Abstract  Fiber-shaped supercapacitors (FSSCs) show great potential in portable and wearable electronics due to their unique advantages of high safety, environmental friendliness, high performances, outstanding flexibility and integrability. They can directly act as the power sources or be easily integrated with other flexible devices to constitute self-powered and sustainable energy suppliers, providing excellent adaptability to irregular surfaces. This review mainly summarizes the recently reported works of FSSCs including preparation methods of various fiber electrodes, construction strategies of FSSCs and multi-functional device integrations, exploration of reaction mechanisms and strategies to improve the electrochemical performance and provision of suggestions on further designing and optimization of FSSCs. Meanwhile, it shares our perspectives on challenges and opportunities in this field, shedding light on the development of high-performance fiber-shaped supercapacitors with multi-functions.

Keywords  Fiber-shaped supercapacitor; Performance improvement; Multi-function device; Integrated electronics

INTRODUCTION

In the era of information explosion, there is an increasing demand for flexible electronic devices in both military and civilian domains (e.g. wearable smart electronics, smart skins and implantable sensors). For promoting the development of small-sized flexible smart wearable electronics,\textsuperscript{[1−3]} it is urgent to make some explorations on light-weight, flexible, miniaturized and integrated power accessories.

As a special family member of the energy-storage systems, supercapacitors (SCs) possess advantages of high power density (> 10\textsuperscript{5} W kg\textsuperscript{-1}), outstanding reversibility and ultralong cycle life (> 10\textsuperscript{5} cycles),\textsuperscript{[4,5]} and gain popularity in consumer electronics, renewable energy storage systems,\textsuperscript{[6−8]} and even hybrid electric vehicles in the past few decades.\textsuperscript{[9,10]} However, it hardly catches up with the increasing demand for consumer electronic gadgets, which put forward more stringent requirements for high energy density, ultra-long operation life, mini-size, lightweight, and mechanical flexibility. Besides, conventional SCs use toxic and corrosive liquid electrolyte posing a potential risk to human health and environmental safety. Therefore, FSSCs based on gel electrolyte sprang up attributed to the unique features of linear-structure, lightweight, high safety, high flexibility, small volume, and knitability as yarn.\textsuperscript{[11]} These unique properties are adaptive to different mechanical motions, such as bending, curling and twisting, making up for the uncovered areas of the conventional bulk/planar energy storage with rigid structure and accelerating the application process of mini-sized electronic devices.

Inspired by the advantageous features of one-dimensional (1D) geometry, the first FSSC in twisting form was fabricated by adopting CNT fiber and it was weaved into textile by Baughman and coworkers in 2003.\textsuperscript{[12]} An advanced hallmark was achieved until the successful fabrication of the first co-axial FSSC,\textsuperscript{[11]} which exhibited more mechanical flexibility to accommodate various deformations and shortened ion diffusion distance to the advanced electrochemical performance. Reviewing the development history of FSSCs, it is found that single-functional energy storage device becomes more and more difficult to meet the overwhelming demand of consumer electronics on durable and stable energy supply. As a result, multi-functional and self-powered energy systems gradually stepped into the spotlight, which stood a profound milestone on the way to practical applications.

So far, significant progress and creative explorations on FSSCs have been achieved from stiff devices to flexible ones,\textsuperscript{[11]} further to stretchable,\textsuperscript{[13]} transparent,\textsuperscript{[14]} self-healing,\textsuperscript{[15]} shape-memory,\textsuperscript{[16]} photo-detective\textsuperscript{[17]} and electrochromic gadgets,\textsuperscript{[18]} and even from single-functional energy storage units to multi-functional self-powered energy sys-
Electrostatic double-layer capacitors (EDLCs)
EDLCs have higher electrostatic double-layer capacitance, where charge separation in a Helmholtz double layer occurs at the interface between the positive/negative electrode and electrolyte (Fig. 1a), and are assembled with electrodes based on carbon or carbon derivatives.[26] Motivated by external currents, the electrons gradually accumulate at the near-surface region of the electrode and are simultaneously counterbalanced by anions or cations in the electrolyte, thus triggering the capacitance stored in the electrostatic field. The capacitance performance totally relies on the amounts of the stored electrons, which is closely related to the physical properties of the electrode such as particle size, specific surface area and morphologies of the electrode materials. Due to the superior reversibility of the surface adsorption-desorption process, the EDLCs show good cycling stability even in a long-term test and ultrahigh rate capability.

Pseudocapacitors
The capacitance of pseudocapacitors mainly arises from highly reversible redox electrochemical reactions of the electrode concentrated in metal oxide or conducting polymers.[27] Reversible electrochemical reaction takes place at the surface or near-surface of the electrode/electrolyte interface as shown in Fig. 1(b) and the capacity depends on the amounts of converted electric charges of the electrodes,[27] thereby presenting 10−100 times energy per unit volume or mass higher than that of EDLCs.[28] The speed of redox reaction process involving charge transportation and ion diffusion is the rate-determining step for rate capability of pseudocapacitors.

Configuration of FSSCs
Considering the prevailing miniature electronic devices especially for the implanted and portable/wearable electronics, stringent requirements are raised for the next-generation fibrous energy storage devices, including the mechanical flexibility to accommodate various deformations and the compatibility with irregular surface of energy storage and

Fig. 1  Schematic diagrams of different energy storage mechanisms: (a) EDLCs and (b) redox pseudocapacitance (RPCs) (Reprinted with permission from Ref. [26]; Copyright (2016) The Royal Society of Chemistry). Schematic diagrams of FSSCs in different structures: (c) conventional format (Reprinted with permission from Ref. [29]; Copyright (2016), American Chemical Society), (d) parallel format, (e) twisting format, and (f) coaxial format (Reprinted with permission from Ref. [30]; Copyright (2015), Springer).
superior electrochemical performances including the energy/power density, cycling stability and wide operation temperature. Different from the conventional planar SCs (Fig. 1c), the fabrication of the fiber-shaped electrodes and selection of suitable gel electrolytes lay the foundation for construction of FSSCs. At the same time, how to design and assemble the configuration of FSSCs is another key issue for achieving high performance.

As reported, several configurations of FSSCs including the parallel, twisting and coaxial forms have been achieved to satisfy given requirements and operating situations (in Figs. 1d–1f). Specifically, the parallel structure is composed of two electrodes in parallel, which is easy to assemble but the electrochemical performance is not in the best optimization to some extent due to the limited contact area of two electrodes. Compared to the parallel structure, the twisting one has a larger contact area between two electrodes and benefits better electrochemical performances and mechanical adaptability. It is worth noting that FSSCs in either parallel or twisting form might suffer somewhat materials shedding, structure damages and performance degradations due to incompact structures. Correspondingly, a landmark breakthrough in assembling FSSCs is adopting coaxial configuration with the inner electrode, gel electrolyte and outer electrode in one fiber. Coaxial FSSCs possess a better integration with higher electrode contact areas, larger accommodation to multiple deformations and convenience to achieve the multifunctionalities such as self-powered devices. Moreover, the coaxial FSSCs display special advantages in highly efficient utilization areas and short ion diffusion pathways, which is expected to advanced electrochemical properties especially for fast charge/discharge performances.

**Basic Performance Evaluation for FSSCs**

These main parameters including capacitance, energy, and power density are the indication of SCs performance and the guiding evaluation for screening materials and designing configuration.

**Capacitance**

Specific capacitance is a term to evaluate the ability of a device to store and release charges, calculating its potential energy density. A SC assembled in a two-electrode configuration, where the charge accumulation occurs on both positive and negative electrodes, can be equivalent to two single capacitors in series connection. Thus, the whole device capacitance of the SCs \( C_{\text{cell}} \) can be calculated using Eq. (1),

\[
1/C_{\text{cell}} = 1/C_p + 1/C_n
\]  

(1)

where \( p \) and \( n \) mean the positive and negative electrodes, respectively. For a symmetric SC with two identical electrodes, \( C_p = C_n = 2 \times C_{\text{cell}} \). Coaxial FSSCs is usually measured by the galvanostatic charge-discharge (GCD) test of SCs, according to Eq. (2),

\[
C_{\text{cell}} = Q/U = i \times dt/du
\]

(2)

where \( i \) presents the galvanostatic discharge current, and \( dt/du \) is a constant obtained from the slope of the galvanostatic discharge curve for EDLCs. Alternatively, \( C_{\text{cell}} \) can also be calculated from the cyclic voltammetry (CV) curve, which is another measurement with a characteristic of constant \( du/dt \) value (Eq. 3),

\[
C_{\text{cell}} = Q/(2 \times U) = \frac{1}{2UV} \int U(i(U)du)
\]

(3)

where \( Q \) is the total voltammetric charge obtained from the area integral of the positive and negative sweeps (\( \int U \) is the current) in a CV curve, \( v \) is the scan rate, and \( U (U = U_p, U_n) \) represents the scanned potential window.

Besides, the \( C_{\text{sp}} \), specific capacitance \( (C_{\text{sp}}) \) is an intuitive performance metric showing the potential for electrode materials in SCs, and used as the standard comparison between various materials even in different forms, which can be calculated from Eq. (4):

\[
C_{\text{sp}} = C_s/B_x = 2 \times C_{\text{cell}}/B_x
\]

(4)

where \( x \) refers to the capacitance per unit mass (\( M \)) for mass-specific capacitance (\( F \cdot g^{-1} \)), length (\( L \)) for length-specific capacitance (\( F \cdot \text{cm}^{-1} \)), area (\( S \)) for area-specific capacitance (\( F \cdot \text{cm}^{-2} \)), or volume (\( V \)) for volume-specific capacitance (\( F \cdot \text{cm}^{-3} \)) of one electrode \( (B_x) \) or two electrodes \( (2B_x) \). The surface area of fibrous electrode is calculated from \( A = \pi \times D \times L \) or \( A = D \times L \) depending on the equivalent area. The volume of fibrous electrode is calculated from \( V = \pi/4 \times D^2 \times L \), where \( D \) is the diameter of the fibrous electrode and \( L \) is the length of the overlapped portion of two electrodes wrapped by the electrolyte. The specific capacitance \( (C_s) \) is calculated based on a single fibrous electrode.

**Energy density and power density**

The energy stored in an assembled SC \( (E) \) and energy per unit mass/length/area/volume (specific energy density, \( E_y \)), power of SCs \( (P) \), and power per unit mass/length/area/volume (specific power density, \( P_y \)) are the performance metrics for the total SCs device. The difference is that \( E_y \) is used to describe the ability of releasing the energy of electrode materials, while \( P_y \) to describe the number of released energy in an instant and is a prominent advantage for FSSCs. Besides, optimized assembly and packaging methods can improve \( E_y \) or \( P_y \) values to some extent. The calculation equations are as following:

\[
E = 0.5 \times C_{\text{cell}} \times U^2
\]

(5)

\[
E_y = E/B_x = 0.25 \times C_{\text{cell}} \times U^2 / B_x
\]

(6)

\[
P = E/t_{\text{discharge}} = 0.5 \times C_{\text{cell}} \times U^2 / t_{\text{discharge}}
\]

(7)

\[
P_y = E_y/t_{\text{discharge}} = 0.25 \times C_{\text{cell}} \times U^2 / (B_x \times t_{\text{discharge}})
\]

(8)

where \( t_{\text{discharge}} \) is the discharge time and \( B_x \) is similar to the above mentioned, which results in gravimetric \( (W \cdot h \cdot kg^{-1}) \), volumetric \( (W \cdot h \cdot cm^{-1}) \), areal \( (W \cdot h \cdot cm^{-2}) \), and areal \( (W \cdot h \cdot cm^{-2}) \) energy density \( (E_y/power density (P_y)) \), respectively. It should be noted that \( x \) corresponds to the mass/area/volume value of a single electrode, and \( y \) corresponds to the one of a device only containing two electrodes while sometimes corresponds to the value of the whole SC device including two active electrodes, separators, and electrolytes.

Besides, for FSSCs with great application prospects in wearable/portable electronics, the consumer and designers are inclined to focus on the comfort, aesthetics and functionality. Thereby, it is really significant to seek a balance between performance and functionality. The exploration and discussion on related industry requirements and uniform standards should be put forward to cater to the application demands.
MATERIALS AND METHODS OF FSSCS

For FSSCs, active electrode materials in fiber format can be divided into three families: carbon-based fiber, conducting polymers fiber, and transition metal oxides/sulfides fiber supported by 1D substrate. To prepare the fibrous electrodes, it is supposed to possess characteristics of excellent conductivity, exceeding electrochemical activity and superior mechanical properties to guarantee the normal operation. Up to now, several typical strategies and methods are adopted to fabricate fiber-shaped electrode materials, as discussed in the following parts.

Carbon-based Fiber

Carbon-based fibers with unique properties of lightweight, resistance to chemicals, extraordinary mechanical performances and high electrical conductivity can work with favorable stability even under a harsh environment (e.g. cold temperature or under extreme stress). Based on the inherent characteristics of different carbon materials, several synthesis methods including dry spinning, wet spinning, electrostatic spinning and capillary template have been utilized to fabricate carbon-based fibers.

The conventional pure carbon fiber is prepared from natural cellulose, synthetic polyacrylonitrile (PAN), and pitch (Fig. 2a), which undergoes carbonization and/or graphitization at high temperatures to eliminate most of the non-graphic components and finally forms the special amorphous or graphitic structure. However, the pyrolysis process of organic precursors introduces a negative influence on the capacitance because of the widely existing stacks and vast grain boundaries as well as confined polycrystalline during this process. Aimed at alleviating these issues and promoting the performance, our group reported the hierarchical three-dimensional (3D) interconnected porous carbon fibers with outer omnidirectional pores and inner hierarchical pores by two-step electrospinning and hard template methods as shown in Fig. 2(b). Polyacrylonitrile (PAN) nanofibers film was initially obtained from electrospinning with in situ nitrogen (N) doping and tailored pores after removing the implanted SiO$_2$ nanoparticles, and then this film was rolled into a microfiber (Figs. 2c and 2d). PAN microfiber was coated with polyaniline (PANI) nanopillars array in order to boost its pseudocapacitance (Fig. 2d). Such hierarchical 3D interconnected porous structure with outer- and inner-omnidirectional pores and channels could provide higher specific surface for electrolyte penetration and charge storage/transportation, resulting in ultrahigh specific capacitance of 339.3 F·g$^{-1}$ (85.1 mF·cm$^{-1}$), good cycling stability with retention of 74.2% after 3000 cycles, and high energy density of 11.6 Wh·kg$^{-1}$ (3.0 mWh·cm$^{-1}$).

The research on new carbonaceous fibers has surged up since the discovery of carbon nanotubes (CNT) and graphene. These newly emerging fibers can be prepared from one- and two-dimensional independent carbon blocks, respectively, as shown in Fig. 3(a), complying with the “bottom-up assembly” strategy (Figs. 3b and 3c). Connected with intrinsic covalent sp$^2$-hybridized carbon atoms and unique assembly structure, they present exceeding perform-

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Fig. 2  (a) The precursor of carbon fiber (Reprinted with permission from Ref. [33]; Copyright (2015) Elsevier). (b) Schematic illustration of the fabrication processes for the PANI-N-doped porous carbon nanofibers fiber scroll (PANI-HCNFs), (c) the SEM image, (d) the cross-section SEM image, and (e) the enlarged SEM image of HCNFs fiber scrolls (Reprinted with permission from Ref. [35]; Copyright (2017) Elsevier).

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Graphene fiber (GF) is another promising carbonaceous material, and is fabricated by aligning graphene sheets continuously into uniaxial direction following wet-spinning process, as illustrated in Fig. 4(a). It is hard to directly bind the separated sheets together and roll large dimensional graphene sheets into continuous fiber. So, bridging materials such as linear or hyperbranched polymers were utilized to guarantee spatial continuity. With persistent efforts, Gao’s team successfully prepared pure GF consisting of ultrahigh quality graphene without adding any binder.\(^{43}\) Adapting the similar technology, the soluble, chemically oxidized graphene or graphene oxide (GO) sheets could form chiral liquid crystals, and by wet-spinning, the GO and montmorillonite (MMT)-GO suspensions (Fig. 4b) in a twist-grain-boundary phase-like model with simultaneous lamellar ordering and long-range helical frustrations were assembled into fiber shape. Herein, macroscopic graphene fibers with a length of more than ten meters can be continuously prepared by spinning the aqueous GO liquid crystals and subsequent reducing to achieve high conductivity and superior electrochemical performance. Besides its excellent electrochemical performance, it is worth mentioning that the outstanding flexibility makes it able to be knitted into designed patterns and directly woven into conductive textiles.

Additionally, GF can also be prepared by a facile template synthesis method without complex dispersion and toxic chemical coagulation bath. Our group utilized capillary as the fiber template to assemble GF. As shown in Fig. 4(c), graphene oxide and the conducting polymer PEDOT:PSS (poly(3,4-ethylenedioxythiophene):polystyrene sulfonate) mixture solution was injected and sealed into the glass fiber with reducing agent of vitamin C (VC).\(^{46}\) In the reduction process, the reduced graphene oxide (rGO) sheets were overlapped and self-assembled with PEDOT:PSS into a certain orientation to form continuous fiber relying on the n-n interaction between them (Figs. 4d and 4e). The prepared composite fiber exhibited a hollow structure with high mechanical strength by lifting up objects above 120 g, which was thousand times heavier than that of the hollow fiber (Fig. 4f). During GO reduction, gases such as carbon dioxide that originated from the functional groups on GO sheets may be formed and released from the solution and accumulated into the sealed pipe, further forming a subminiature airflow from the solution interior to the void space, ultimately resulting in hollow fiber. It was inclined to form lyotropic nematic liquid crystals, and the hydrophilic functional groups in PSS facilitated the diffusion of rGO sheets to promote the formation of hollow structure. Compared with wet-spinning, this one-pot method can form fiber-shaped rGO and rGO composite electrode simultaneously without complex post-treatment processes. And it is very convenient to control the diameter and length of fiber by adjusting the size and length of the template. Furthermore, this method provided an option for in situ doping heteroatoms to promote performance. In our following work, N-doped rGO fibers were designed and synthesized by capillary template with the addition of N-containing agent such as urea into the GO solution precursors in the reducing process (Fig. 4g).\(^{47}\) The N-doped rGO exposed more active sites and improved the whole conductivity of the fiber, thereby facilitating the improved electrochemical performances.
Conducting Polymer Fiber

Except for carbon and graphene fibers, some conducting polymers have also been manufactured into fiber materials for FSCs owing to the long chain/network structure and excellent spinability in specific orientation. Some typical methods including the wet-spinning, electrospinning, and melt-spinning have been applied to prepare the polymer fiber. Among them, the wet-spinning is a widely accepted method for large-scale production and for polymer fibers with versatile functionalization and morphologies. Following the typical wet-spinning process, the polymer solution is injected into the coagulation bath, where bilateral diffusion happens between the coagulation bath and the fibrous precursor. In this process, small molecules and solution solvent are removed, and the residual polymers form the continuous fiber relying on the precipitation in the insoluble solvent. The as-spun fiber can be continuously prepared to a desirable length and show high flexibility even after bending, twisting or knotting deformations. It should be noted that the physical parameters of spun fiber such as strain/tension, microstructure, diameter and morphology are closely related to the properties of the spinning solution and coagulation bath. So the components, concentration, treating time, and treating sequences of the coagulation bath and spinning precursors should be optimized according to the characteristic of the specific polymer.

PEDOT:PSS has been proven as an ideal candidate among...
various conductive polymers for FSSCs, which owns the properties of good film-forming, high pseudocapacitance, high visible light transmissivity, good conductivity, chemical stability, and excellent aqueous distribution. However, in previous works, highly concentrated PEDOT:PSS solution was mostly injected into the toxic coagulation bath (e.g. acetone and isopropanol) with potential safety risks to the human health and environment.\[40] It was discovered by our group that meter-long PEDOT:PSS fiber could be successfully fabricated by injecting the polymer solution into environment-friendly aqueous calcium chloride (CaCl₂) coagulation solution as presented in Fig. 5(a). When polymer immersed in coagulation solution, the interaction between the ions and the polymer may happen. To be specific, the Cl⁻ and Ca²⁺ ions may bind with the PEDOT chains and PSS chains, respectively, facilitating the formation of PEDOT fibers. The PEDOT:PSS fibers showed high mechanical characteristics with strength and breaking strain of 80 ± 5 MPa and 17% ± 2%, respectively. Meanwhile, their mechanical properties and flexibility can be further augmented by forming the biscalloped yarns with the distinctly increased specific electrical conductivity up to 38 S·cm⁻¹.\[49] In order to greatly improve the conductivity of the fiber electrode, an effective strategy was exploited by our group through solvent treatment. After the solvent treatment, most of the insulated PSS was removed and the PEDOT chains were self-assembled to a highly ordered and crystalline structure (Figs. 5b–5e) so that fiber conductivity and mechanical properties were greatly improved without sacrificing its outstanding electrochemical property.\[50] The unique structure of highly ordered crystalline conductive polymer makes PEDOT-S fiber present superior electrochemical performance and outstanding flexibility, showing great potential in the real application.

Meanwhile, from the perspective of increasing active sites for energy storage, composite fibers have been made to improve the specific surface area of the wet-spun PEDOT:PSS fiber. The composite fibers of GO and PEDOT:PSS with different surface morphologies were prepared by adjusting the adding amount of GO into the PEDOT:PSS solution in Fig. 5(f).\[51] With the increased amount of GO from 5% to 20%, the surfaces of the corresponding fibers showed much more wrinkles (Fig. 5g) which accelerated the electrolyte wettability and increased the interfacial capacitance activity. Recently, a new exciting member of 2D nanomaterials family, transition metal carbides and nitrides so-called “MXene”, has stepped into the spotlight because of exceeding electrical conductivity, specific 2D structure, and hydrophilic nature. However, it is difficult to assemble pure MXene fiber without additional binders or additives due to its large lateral sheet size (> 1 mm) lacking sufficient inter-sheet strength. Interestingly, Ti₃C₂Tx/PEDOT:PSS hybrid fiber reported by Razal’s group using the wet-spinning method with 70 wt% MXene

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**Fig. 5** (a) Schematic illustration of the fabrication processes for PEDOT:PSS fiber (Reprinted with permission from Ref. [49]; Copyright (2016) The Royal Society of Chemistry), (b)–(e) SEM images of PEDOT:PSS and PEDOT-S:PSS fibers after different sulfuric acid treating time, (d) SEM image of a tied PEDOT-S:PSS fiber, and (e) selected area electron diffraction (colored in light green) of the PEDOT-S:PSS fiber (Reprinted with permission from Ref. [50]; Copyright (2018) Elsevier), (f) Schematic illustration of the fabrication processes for PEDOT:PSS fiber mixed with GO (10 wt%) and (g) SEM images of GO-PEDOT:PSS fiber with more wrinkles (Reprinted with permission from Ref. [51]; Copyright (2018) Elsevier), (h) Schematic illustration of the fabrication processes for MXene-PEDOT:PSS fiber, (i) the photograph, (j) SEM image and (k) structural schematic of obtained MXene-PEDOT:PSS fiber (Reprinted with permission from Ref. [52]; Copyright (2019) Wiley).
loading realized a high conductivity of $\sim 1489$ S cm$^{-1}$.\[52\] Moreover, the FSSC based on Mxene/PEDOT:PSS fiber was constructed and showed good electrochemical performances and flexibility, demonstrating the device energy densities up to $\sim 7.13$ Wh cm$^{-3}$ and excellent mechanical strength.

**Metal Oxides and Sulfides Based Fiber**

As for metal oxides and sulfides such as RuO$_2$,\[53\] Co$_3$O$_4$,\[54,55\] VO$_x$, MnO$_2$,\[56,57\] NiO,\[58,59\] and CoS$_2$, it is also hard to connect each other to form a continuous and soft fiber due to the lack of the interaction between their small molecules. Therefore, one of the universal strategies adopted is to anchor them on the surface or incorporate them into the interior of fiber substrates including metal wires, elastic wires, carbon fibers, and conducting polymer based fibers. Metal wires such as Ti, stainless,\[60,61\] Ni,\[62,63\] and Cu\[64,65\] possessing inherently fibrous structure, excellent conductivity and mechanical properties are preferentially selected as the 1D substrates. In addition, their surfaces are easily modified via chemical or physical treatment, acting not only as the current collector but also as the buffer layer for tight adhesion of active materials especially for metal oxides and sulfides.

Ti wire is widely accepted as a great option with exceeding properties of low density, high mechanical strength, excellent conductivity, and high melting point. Furthermore, the sluggishness both in acid and alkaline solutions and the TiO$_2$ formation on the surface providing indispensable buffer sites for the subsequent growth of active materials provide more irreplaceable advantages than other candidates. Based on the above analysis, our work introduced a thin TiO$_2$ buffer layer on Ti surface via annealing in order to assist the hydrothermal growth of MoS$_2$ NSs on Ti fiber (Fig. 6a)\[61\] Introducing the TiO$_2$ buffer layer can not only increase surface roughness and integrated stability of the MoS$_2$ NSs, but also enhance the whole electrode integration even at multiple deformations (Figs. 6b–6d). A solid-state FFSC was designed and fabricated using Ti/TiO$_2$/MoS$_2$ fiber electrodes in poly(vinyl alcohol)
(PVA)/H$_2$PO$_4$ gel electrolyte, exhibiting high electrochemical performance and mechanical properties. The results show that TiO$_2$ modification is vital for uniform growth of active materials, guaranteeing improved and stable performance. Thus, the sulfur-doped TiO$_2$ nanotube array (S-TiO$_2$ NTA) was prepared by electrochemical anodization (Figs. 6e–6h), introducing a large surface as a buffer layer to assist electro-polymerization of PANI, and building short ion diffusion path for improving high rate capability.\cite{86} The FSSC based on PANI/S-TiO$_2$ NTA/Ti exhibited ultra-stable, long cycle lifetime with capacitance retention of 93.78% after 1.2 × 10$^4$ cycles.

Active materials are prone to be anchored on the modified surface of metal substrate to form fiber electrode via adopting interfacial engineering strategies such as annealing, hydrothermal synthesis,\cite{67} electrochemical anodization, electrodeposition\cite{68} and electro-polymerization. However, the non-negligible weight of metal substrates does lower specific capacity and they are not soft and uncomfortable enough to be worn and implanted into fabrics. An alternative fibrous carrier is elastic wires that inherits outstanding bending, stretching ability, and negligible weight, which is tailor-made for the device with a great requirement for the tensile property. However, it is hardly possible for sweat and airflow to penetrate. Thus, metal fibers and elastic wires are replaced by less-weight carbon-related fiber. Commercial carbon fiber, carbon nanotube fiber, graphene fiber, and conducting polymer fiber treated with or without carbonization are more preferable competitors. They usually provide —OH and —COOH functional groups and tough surfaces assisting active material growths. In most cases, commercial non-functional carbon fiber makes less contribution to capacitance due to tight carbon sheets stacking. Another strategy is mixing active materials with other materials common in the solution state, performing electronic spinning/wet-spinning/temple treatment, and finally embedding them into the inner of fiber for advanced capacity. It is notable that the proportion of parasitic materials should not be beyond the reasonable range, otherwise they might sacrifice the integrity and pose risk to softness.

**STRATEGIES TO IMPROVE THE PERFORMANCES**

**Improve the Energy Density**

As mentioned previously, the energy density of SCs is calculated with Eq. (6), and this value is in positive correlation to capacitance and operation voltage window. It is greatly affected by the conductivity of the electrolyte, the electrolyte accessible specific surface area, electron transfer rate in electrodes and the degree of matching property between electrode and electrolyte, as well as the voltage window. Up to now, researchers have devoted great efforts to these two aspects as the following discussion.

**Improve the capacitance**

Based on the principles of EDLC, its capacitance is proportional to the face-to-face area of the two parallel-plates according to the equation of $C = \varepsilon S/4k\pi d$.\cite{80} For fibrillar electrode, from the structural perspective, the effective “face-to-face” area is equivalent to a rectangular projection area that is related to the configuration of devices. However, most of the fiber electrodes are solid that only the outer surfaces can have access to the electrolyte with low interfacial areas. Therefore, hollow and porous materials have been selected to raise the specific surface area to promote the specific capacitance and energy density.\cite{73} The hollow/porous structure can increase electrode specific surface area effectively and provide numerous electroactive sites for efficient electrochemical reactions. On the other hand, such a structure can provide a fast channel for electron and ion transportation due to the relatively high absorbing liquid capability. In order to increase the interior electrochemical active area, a hollow rGO/PEDOTs fiber electrode (donated as HGF) was prepared by our group and assembled in FSSCs.\cite{66} It was found from the cross-sectional SEM images that an obvious interior hole was formed inside the composite fiber (Figs. 7a and 7b) and the rGO sheets were nematic and aligned along the length of the HGF. In addition, the surface of HGF was rough and wrinkled, imposing more electroactive sites and increasing specific areas for charge storage at the electrode/electrolyte interface (Figs. 7c and 7d). It was also found that the ratios of the inner to the outer diameters were gradually increased with the increasing polymer weight percentage from 10% to 33% in the composite fibers. And only a small hole can be observed in the solid pure rGO fiber without adding PEDOTs (Figs. 7e–7h), comparatively. The HCF-based SC exhibited an ultrahigh specific area capacitance ($C_{sp}$) of 304.5 mF·cm$^{-2}$ (143.3 F·cm$^{-3}$ or 63.1 F·g$^{-1}$) at 0.08 mA·cm$^{-2}$, which was also much higher than those in the previous reports at the report time, as shown in Fig. 7(i). Our calculation and experiment results declared that the change rate of the capacitance ($R_C$) should be theoretically equal to the superficial area change rate ($R_S$) of the fiber (Fig. 7i), demonstrating the significant contribution of the hollow structure to increase the specific capacitance.

Since pseudocapacitive materials utilize redox reactions to store/release energy, they possess much higher energy densities than carbon based double-layer capacitors. Therefore, pseudocapacitive materials such as conductive polymers, metallic oxide, transition metal disulfides (TMDs) and many others have been utilized in FSSCs following the as-mentioned fiber electrode method to increase the energy densities of the FSSCs. As studied, a FSSC based on bare rGO fibers showed $C_{sp}$ of 1.2 mF·cm$^{-2}$\cite{72} After incorporation of Mn$_2$O$_3$,\cite{72} MnO$_2$,\cite{73} RuO$_2$,\cite{76} PEDOTs,\cite{80} or MoS$_2$,\cite{74} the $C_{sp}$ was increased to 73.6, 91.5, 107, 131, or 598 mF·cm$^{-2}$, respectively. RuO$_2$ is recognized as the top-ranking material among such category, and mature in commercial application. Recently, RuO$_2$ nanoparticles were incorporated in PEDOTs to form PEDOTs–RuO$_2$ composite fiber by a wet spinning method.\cite{79} As expected, the PEDOTs–RuO$_2$ FSSCs exhibited higher $C_{sp}$ and $E_{A}$ than pure PEDOT FSSCs. Ragone plots of PEDOTs–RuO$_2$@PEDOTs based FSSCs showed an ultra-high $E_{VR}$ of 22.5 μWh·cm$^{-2}$ (based on entire device) with a maximum $P_{max}$ of 750 μW·cm$^{-2}$, which was approximately three times that of PEDOTs fiber (8.3 μWh·cm$^{-2}$, 40 μW·cm$^{-2}$). At the same time, the PEDOTs–RuO$_2$@PEDOTs based FSSCs exhibited an ultra-high $E_{VR}$ of 13.2 mWh·cm$^{-2}$ with a maximum power density of 441.1 mW·cm$^{-2}$. In addition, some redox additives were added into electrolyte to enhance the capacitance of FSSCs.\cite{82} For example, Peng’s group developed a new redox gel electrolyte
via adding 2-mercaptopyridine to PVA-H$_2$SO$_4$ which underwent reversible redox reactions for a high pseudocapacitance. Thus, by using this gel electrolyte, the FSSCs acquired a high areal specific capacitance of 507.02 mF·cm$^{-2}$.[76]

**Widen the electrochemical window**

SCs based on aqueous electrolytes are safer and more environment-friendly than non-aqueous SCs typically employing the toxic and flammable organics as solvent.[77] However, the stable working voltage of aqueous electrolytes is commonly below 1.23 V (the decomposition potential of pure water), which makes the aqueous SCs suffer from a natural disadvantage in energy and power density.[78] In comparison, the operation voltage of non-aqueous SCs can be currently broadened to 3.0 V.[79] In order to extend the operation voltage and increase the energy density of aqueous SCs, many strategies have been proposed and developed recently.

Assembling asymmetric SCs is one of the accessible approaches to acquire high working voltage, which combines the different stable potential windows of a positive electrode and a negative electrode, presenting high overpotentials for hydrogen evolution reaction (HER) and oxygen evolution reaction (OER).[80] In recent years, numerous efforts have been made to develop innovative fiber-shaped asymmetric SCs (FSASCs), and great success has been obtained on expanding their operation voltages. Generally, the FSASC is made up of two different electrodes with one Faradic electrode and one capacitor-like electrode, serving as the energy source and the power source, respectively. Zhi et al. developed fiber-shaped asymmetric SCs with PEDOT@MnO$_2$@stainless-steel and C@Fe$_2$O$_4$@stainless-steel as electrodes. The operation voltage reached 2.0 V and presented an excellent $C_d$ of 60 mF·cm$^{-2}$ and an $E_{sc}$ of 33.5 μWh·cm$^{-2}$.[81] Besides asymmetric SCs, the working voltage of the symmetric SCs could also be widened by improving the electrode materials structure and conductivity. Very recently, our group prepared a symmetric FSSC with high voltage window of 1.6 V by employing PEDOT-SPSS fiber as electrode and PVA/H$_2$SO$_4$ as gel electrolyte.[82] The removal of most PSS molecules from PEDOT:PSS with concentrated H$_2$SO$_4$ allowed the rearrangement of PEDOT polymer chains, resulting in a densely packed structure of high-crystalline aggregates owing to the strong $π-π$ stacking of the PEDOT backbone (Figs. 8a and 8b). The structural changes dramatically improved the crystallinity and charge carrier characteristics of the electrode, thereby extending the working voltage from 0.8 V to 1.6 V and imparting high areal and volumetric power densities of 400 μW·cm$^{-2}$ and 320 mW·cm$^{-3}$, respectively (Figs. 8c–8e).

It is reported that pH-neutral electrolyte can improve the kinetic stability of water that inhibits HER and OER compared to the strong acid and base electrolyte (e.g. H$_2$SO$_4$ or KOH).
aqueous solution). For example, an active carbon (AC) based SCs with gold current collector exhibited a surprising working voltage of 2.2 V in 1.0 mol·L⁻¹ Li₂SO₄ aqueous electrolyte. Additionally, high concentration electrolytes (water-in-salt) were also applied in expanding the operation voltages of aqueous SCs. In water-in-salt solution, the number of water molecules is far smaller than in the normal electrolyte solution, making the water molecular effectively segregated and thus decreasing the electrochemical activity. On the other hand, the crystal-like model of water-in-salt electrolyte contributes to a reductive potential of negative ion more positive than the reductive potential of hydrogen evolution. For example, 2.0 and 2.2 V high working voltage of symmetric AC-based SCs were obtained in 0.5 mol·L⁻¹ Na₂SO₄ and 1.0 mol·L⁻¹ Li₂SO₄ aqueous electrolyte, respectively. Recently, our group prepared 2.2 V high performance symmetrical fiber-shaped aqueous SCs which were enabled by "water-in-salt" polymer gel electrolyte (LiCl-in-PVA,
8.96 mol·L\(^{-1}\)) and N-Doped graphene fiber.\(^{[47]}\) The voltage window of this FSSC using highly-concentrated LiCl electrolyte was approximately two times higher than those of the symmetric SCs using acidic or common aqueous electrolytes. The broadened voltage window might be originated from the increase of hydration energy and the physical protection of Li\(^+\) ions on the electrode surface. Benefiting from the unique porous structure and broadened voltage window, the as-fabricated FSASC demonstrated an ultrahigh energy density of 25.6 μWh·cm\(^{-2}\) (Figs. 8f–8i), outperforming most of the symmetric FSSCs reported previously (Figs. 8j–8l).

A novel hybrid electrolyte with salt-to-water-to-acetonitrile molar ratio of 1:1.5:2.4 based on sodium perchlorate (NaClO\(_4\)) salt was proposed by Dou et al. and the hybrid electrolyte showed a high ionic conductivity, excellent wettabili-

ity, and moisture-tolerant and flame-retardant properties, enabling carbon-based SC to operate at 2.5 V with high specific capacitance and an excellent cycling performance over 1.5 × 10\(^4\) cycles.\(^{[83]}\) Moreover, adding redox-active additives into electrolyte can also widen the operation potential range. Wei reported a CNT-based FSSC by adding two redox additives simultaneously, specifically polypyrrole in electrode and hydroquinone in electrolyte (H\(_2\)SO\(_4\)/PVA hydrogel). As expected, the FSSC achieved an operation voltage of 1.2 V and a specific capacitance of 55.7 F·g\(^{-1}\), as well as excellent capacitance retention of 103% after 2000 cycles.\(^{[86]}\) Kaner et al. further extended the operating voltage of activated carbon-based SCs to 2.0 V by adding 0.1 mol·L\(^{-1}\) [Fe(CN)\(_6\)]\(^{3-}/2-\) into the electrolyte of 1.0 mol·L\(^{-1}\) Na\(_2\)SO\(_4\).\(^{[87]}\)

**Improve the Cycling Performance**

As expected for FSSCs, it is designed to be utilized in the portable/wearable smart devices, so cycling stability plays an important role in the long-term application. The irreversible expansion of electrode materials that emerges in the long period of charging-discharging process might block the electrode channel, damage the surface structure, and even lead to the exfoliation of active materials from the electrode surface. Many available approaches have been proposed to solve these issues, such as coating technique and interfacial engineering.

It was found that RuO\(_2\) nanoparticles were easily exfoliated from electrode in the long-term charge-discharge process, dramatically limiting the operation time. To overcome this problem, a core-shell PEDOTs-RuO\(_2\)@PEDOT fiber consisting of a protecting shell was prepared to enhance the structural stability by our group (Fig. 9a).\(^{[75]}\) It was found that the morphology of PEDOTs-RuO\(_2\)@PEDOT fiber had no change after hundreds of cycling because of the PEDOT protection layers (Figs. 9b and 9c). Excellent cycle stability (~80% after 5000 cycles) was successfully achieved under the elaborated core-shell fiber as compared with the PEDOT-RuO\(_2\) fiber-based FSSCs (only 80% after 1000 cycles) (Figs. 9e and 9f). Similarly, Liu et al. improved the quasi-solid-state FSASCs cycling stability by preparation of α-MnO\(_2\) nanorod@δ-MnO\(_2\) nanosheets array cathode, in which the α-MnO\(_2\) nanorod served as an effective shell to protect δ-MnO\(_2\) nanosheets from taking off from Ti wires.\(^{[88]}\) The resulting FSASCs with 2.0 V operation voltage delivered high volumetric energy and power densities (9.53 mWh·cm\(^{-3}\) and 2.272 × 10\(^4\) mW·cm\(^{-3}\), respectively) as well as outstanding cycling stability over 1.0 × 10\(^6\) cycles. Additionally, interfacial engineering is adopted to develop the ultralong cycle lifetime FSSCs. Our group have prepared polyaniline (PANI)/sulfur-doped TiO\(_2\) nanotubes array (PANI/S-TiO\(_2\)) materials via interfacial treatment.\(^{[68]}\) As shown in Figs. 9(f) and 9(g), the S-doping effectively narrowed the bandgap of TiO\(_2\) and promoted the conductivity of TiO\(_2\) nanotubes, as well as enhanced the binding affinity of PANI anchored on the electrode surface through N−S bonding (bonding length of N−S of 1.634 Å larger than N−O of 1.420 Å), leading to the excellent electrode structure stability. The assembled solid-state FSSCs achieved high energy density and ultralong cycle lifetime. A high C\(_A\) of 91.9 mF·cm\(^{-2}\), \(E\_A\) of 3.2 μWh·cm\(^{-2}\) and a capacitance retention of 93.78% after 1.2 × 10\(^4\) charge/discharge cycles were maintained, respectively (Figs. 9h–9j).

**Widen the Working Temperature Range**

Up to now, most of the existing FSSCs can only operate at room temperature. FSSCs working in extremely cold/hot zones are limited by the material properties of electrolytes, electrodes, and their synergetic interactions. Thus, significant methods have been proposed for expanding working temperature ranging from −60−75 °C, especially for aqueous SCs. To date, some strategies such as gel electrolytes, water-organic mixture solvent, and high-concentrated electrolytes have been adopted to widen the working temperature. Our group fabricated an all-climate FSSCs which used PEDOT/RuO\(_2\) fiber as electrode and LiCl/PVA hydrogel as the anti-freezing electrolyte.\(^{[75]}\) By optimization, the FSSCs could operate within a temperature range of −60−75 °C. As exhibited in Fig. 10(a), comparing to other PVA-based gel, the LiCl/PVA gel electrolyte was still flexible at −30 °C and an exothermic peak of about −40 °C emerged in the DSC curve of LiCl/PVA hydrogel, suggesting the lower freezing point (Fig. 10b). Benefiting from the synergistic effects of fiber electrode and aqueous gel electrolyte, the PEDOT/RuO\(_2\) fiber still delivered 78.8 mF·cm\(^{-2}\) at −60 °C (Fig. 10c). Additionally, the capacitance could reach 202.5 mF·cm\(^{-2}\) and maintain 74.3% of its initial capacitance (200.6 mF·cm\(^{-2}\)) after 1000 cycles at 75 °C. Furthermore, even cycled at −60 °C, the PEDOT/RuO\(_2\) fibers based FSSCs could still maintain 43.9% of specific capacitance after 5000 cycles. In such extreme conditions with temperature ranging from −60 °C to 75 °C, a LED could be easily lightened up by two series-connected FSSCs, indicating good temperature-tolerant properties in practical application (Figs. 10d−10f).

**Extend the Functions**

In practical applications, portable/wearable electronics might experience diverse deformations with body bending, stretching and moving, which introduce risks of structural and functional damage because of the long-term stress. In this regard, the elaborated FSSCs with multifunction have been proposed and prepared to mitigate the irreversible destruction, such as stretching SCs,\(^{[31,39]}\) self-healing SCs,\(^{[92]}\) electrochromic SCs,\(^{[93,94]}\) and implantable SCs.\(^{[99]}\) In 2015, Peng’s group, for the first time, realized a shape-memory FSSCs utilizing core-shell CNT/polyurethane (PU) stretchable fiber as electrode and PVA-H\(_2\)SO\(_4\) gel as electrolyte.\(^{[101]}\) Our group firstly developed a new type of “internal tandem” stretchable FSSCs which combined stretchable conducting wires and energy storage in one device based on high crystalline PEDOT polymer fiber (Fig. 11a).\(^{[48]}\) The resulting assemblies consisting of 8 serially
connected cells displayed a high-voltage output of 12.8 V, ultrahigh energy density of 41.1 μWh·cm⁻² at power density of 3520 μW·cm⁻², and remarkable stretchability of up to 400% without obvious capacitance degradation. The tandem FSSC could lighten up microelectronic devices (such as USB light, logo abbreviation and commercial digital watch) under different deformation conditions even at a strain elongation of 400%

Zhi et al. fabricated an electrically and mechanically self-healable FSSC employing magnetic materials as electrodes and wrapping with a self-healing carbonylated PU shell (Figs. 11g). Benefiting from the synergistic effect of the magnetic electrodes and the self-healing shell, the FSSCs exhibited an excellent self-healing property that the specific capacitance could be restored to 71.8% after the fourth self-healing cycle. (Figs. 11b–11f).
Fig. 10  (a) Photograph of H$_3$PO$_4$-PVA, H$_2$SO$_4$-PVA, PVA-H$_2$O, LiCl-H$_2$O and LiCl-PVA electrolytes at −30 °C; (b) DSC curves of LiCl-PVA, PVA-H$_2$O, LiCl-H$_2$O, and H$_2$O; (c) The specific capacitance of PEDOT-RuO$_2$ FSSCs measured at different temperatures at a scan rate of 50 mV·s$^{-1}$; (d) Two PEDOT-RuO$_2$ FSSCs connected in series to lighten up a green LED at the temperature of −60 °C, (e) 25 °C and (f) 75 °C, respectively. (Reprinted with permission from Ref. [75]; Copyright (2018) Elsevier).

Fig. 11  (a) Schematic illustration to the fabrication of PEDOT based FSSCs in series, (b) PEDOT based FSSCs to lighten up a commercial USB light, (c) a logo abbreviation, (d, e) a commercial digital watch, and (f) two series-connected PEDOT based FSSCs to lighten up a green LED under increasing strains from 0 to 400% (Reprinted with permission from Ref. [49]; Copyright (2018) Elsevier). (g) Schematic illustration of the self-healing process of supercapacitor (The magnetic alignment could assist the reconnection of the fibers in the broken yarn electrodes when they are brought together.) (Reprinted with permission from Ref. [96]; Copyright (2019) American Chemical Society). (h) Schematic of mechanism of photo-responsive smart coloration in electrochromic SC system for energy saving, architectural application with inset showing the structure of the smart coloration, energy storage system, and gel electrolyte composition consisting of LiClO$_4$ used for electrochemical stability (Reprinted with permission from Ref. [18]; Copyright (2019) Wiley).

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healing. And, the mechanical properties of the whole device could be well maintained through multiple cutting-healing cycles. In addition, another stretched and self-healing RGO/PU FSSC was prepared by Gao’s group,\(^\text{[97]}\) which could be stretched at high percentages up to 300% and the tiny broken fibers could be reconnected. The FSSC had 82.4% capacitance retention after a large stretch (100%) and 54.2% capacitance retention after the third healing.

The visible indication of operation state of FSSCs is meaningful in practical applications for timely replacement and real-time sensing. Thus, electrochemical materials are introduced into FSSCs not only for energy stores but also for accurate voltage variable indication. Several electrochromic materials including metal oxides (WO\(_3\), V\(_2\)O\(_5\)), conductive polymers (PANI, PEDOT) and others have come into our horizons. Yun et al. designed an electrochromic SCs which was capable of electrochemical and electrochromic reactions.\(^\text{[18]}\) The fabricated device exhibited a high coloration efficiency of 64.8 cm\(^2\)C\(^{-1}\) and the specific capacitance \(C_s\) of 406.0 F·g\(^{-1}\), respectively (Fig. 11h). Kim et al. also developed an all transparent-stretchable electrochromic SCs adopting bi-stacked WO\(_3\) nanotube/PEDOT:PSS as active materials, Au/Ag core-shell nanowire embedded PDMS as elastic substrate, and polyacrylamide (PAAM) hydrogel as the electrolyte.\(^\text{[14]}\) For the transparent-stretchable WO\(_3\)-based electrochromic SCs, the maximum specific capacitance of 471.0 F·g\(^{-1}\) was obtained with a 92.9% capacity retention even after 5.0 × 10\(^6\) cycles. Additionally, a high coloration efficiency of 83.9 cm\(^2\)C\(^{-1}\) was shown due to WO\(_3\) nanotube and PEDOT:PSS thin layer, both possessing coloration and pseudocapacitor double characteristics.\(^\text{[18]}\)

**Integrated Multifunctional Devices**

In recent years, the great growth of portable/wearable electronics ranging from the mobile phone, personal multimedia to medical devices encourages researchers to develop more promoted energy-storage and ultra-long operation technologies. The self-powered technique for FSSCs is becoming a great candidate and numerous efforts have been devoted to designing and fabricating various novel self-powered systems such as photovoltaic,\(^\text{[98]}\) piezoelectric,\(^\text{[19]}\) and hygro voltaic self-charging ones.\(^\text{[99]}\)

The self-charging devices as a power source, composed of electrical energy conversion and storage units, are connected with energy-consuming electronics in series to constitute a complete self-powered integrated system. Our group fabricated a self-powered photodetection system containing a solar cell, an all-solid-state FSSC, and a TiO\(_2\) photosensor.\(^\text{[61]}\) The capacitance of FSSCs was up to 230.24 F·g\(^{-1}\) (70.6 F·cm\(^{-2}\)) and the energy density and power density could reach up to 2.70 Wh·kg\(^{-1}\) (4.98 mWh·cm\(^{-2}\)) and 530.9 W·kg\(^{-1}\) (977.4 mW·cm\(^{-2}\)), respectively. Moreover, this integrated power source was steady and continuous in providing adequate energy for TiO\(_2\) NW UV-photodetector over a long operation time. Zou et al. designed a new integrated device that combined dye-sensitized solar cells (DSSCs) and FSSCs by using an identical bifunctional electrode of PANI-coated stainless steel wire in the whole system, which successfully realized solar energy conversation and storage.\(^\text{[100]}\) For PANI-based DSSC, photoelectric conversion efficiency of 5.41% can be obtained. For energy storage, the as-fabricated FSSCs had a maximum \(C_s\) of 41 mF·cm\(^{-2}\) that was superior to other reported FSSCs at that time. More importantly, in an integrated fiber-shaped self-power device supported by DSSCs and FSSCs, a total energy conversion could reach 2.1%. To further improve the overall conversion efficiency, our group developed a fiber-shaped DSSCs-SCs integrated device by sharing one common electrode.\(^\text{[101]}\) In this kind of fiber-shaped DSSCs-SCs, highly crystalline conductive polymer fibers with high electrocatalytic activity and energy storage efficiency for both energy harvest and energy storage were used as electrodes. Synergistically, the DSSCs-SCs fabricated by sharing one electrode to reduce energy loss in the conversion process and optimize the capacitance compatibility of individual units exhibited a remarkable photoelectro conversion efficiency of 9.3% and high overall conversion efficiency of 5.1%, which are higher than those of reported energy fibers and even close to those of the planar devices in previous reports (Figs. 12a and 12b).\(^\text{[101]}\) An upgrade version was subsequently achieved with higher sensitivity and response rate in a light-driven self-powered system containing DSSCs, FSSCs and UV sensor (Figs. 12c–12e).

It is worth mentioning that body motion energies could also be collected and utilized. As Wang et al. reported,\(^\text{[101]}\) a self-charging textile system simultaneously collecting and storing sunshine and body motion energies was designed to sustainably operate portable/wearable electronics. In such a system, a total power conversion efficiency of 5.64% was achieved, a DSSC unit with \(V_{OC}\) of 0.74 V and a \(J_{SC}\) of 11.92 mA·cm\(^{-2}\) was obtained, and FSSC unit with specific capacitance of 1.9 mF·cm\(^{-1}\) was exhibited. In addition, the self-charging textile system could be easily woven into electronic textiles to fabricate smart clothes and wearable electronic devices. Besides, Sun et al. built a flexible paper-based self-charging power system which was composed of EP-TENGs as energy harvest unit and EP-SCs as energy storage unit (Fig. 12f).\(^\text{[102]}\) A single EP-TENG device showed open-circuit voltage \(V_{OC}\), short circuit current \(I_{SC}\), short circuit charge \(Q_{SC}\), and the maximum area power \(P_{A,max}\) of 98.6 V, 11.3 μA, 31.1 nC, and 18.4 mW·m\(^{-2}\), respectively. And the EP-SC served as an effective energy-storage unit by delivering an excellent \(C_s\) of 150 F·g\(^{-1}\). Three-parallel EP-TENGs were further integrated with three-series EP-SCs to fabricate the flexible paper-based self-charging system which could sustainably power an electronic watch and a calculator.

As known, the FSSCs have a high power density but low energy density compared with batteries. It is reported that FSSCs display only an \(E_{A}\) less than 10 Wh·kg\(^{-1}\), while \(E_{A}\) of the fiber-shaped Li\(^+\) batteries (FSBs) reaches up to 27 Wh·kg\(^{-1}\),\(^\text{[103]}\) and even 297.5 Wh·kg\(^{-1}\) for fiber-shaped Li-O\(_2\) batteries.\(^\text{[104]}\) And it is possible to complement each other by sharing common electrodes to associate them together for achieving unity of power density and energy density in hybrid energy-storage systems.\(^\text{[103]}\) Peng’s group fabricated a fiber-shaped hybrid energy-storage system integrating a FSB and a FSSC via sharing an identical CNF/Li\(_2\)TiO\(_3\) (LTO) electrode (Fig. 12g). The fiber-shaped hybrid energy-storage devices delivered an energy density of 50 mWh·cm\(^{-3}\) (~90 Wh·kg\(^{-1}\)) and a power density of 1.0 W·cm\(^{-3}\) (or 5970 W·kg\(^{-1}\)).
ing the performance of most as-reported FSSCs and FSBs (Figs. 12h–12j).

CONCLUSIONS AND PERSPECTIVE

FSSCs have shown great potential in the next generation of wearable and portable electronics possessing the unique properties of lightweight, high flexibility, knittability and integrability. It is envisioned that FSSCs will permeate into every corner of our life and prevail in the market of wearable, implantable artificial intelligence in the near future for monitoring the body healthy condition, detecting the environmental circumstance changes, and even serving the man-machine interaction system. To date, researchers have devoted intense efforts to designing and fabricating advanced materials and innovative devices to improve the electrochemical and mechanical properties of FSSCs. In this review, we have...
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