

REVIEW ARTICLE

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Electrospun nanofiber-based soft electronics

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Abstract

Electrospun nanofibers have received considerable attention in the field of soft electronics owing to their promising advantages and superior properties in flexibility and/or stretchability, conductivity, and transparency; furthermore, their one-dimensional nanostructure, high surface area, and diverse fibrous morphologies are also desirable. Herein, we provide an overview of electrospun nanofiber-based soft electronics. A brief introduction of the unique structure and properties of electrospun nanofiber materials is provided, and assembly strategies for flexible/stretchable electronics are highlighted. We then summarize the latest progress in the design and fabrication of representative flexible/stretchable electronic devices utilizing electrospun nanofibers, such as flexible/stretchable conductors, sensors, energy harvesting and storage devices, and transistors. Finally, a conclusion and several future research directions for electrospun nanofiber-based soft electronics are proposed.

Introduction

Flexible and stretchable electronics have attracted worldwide attention in recent years due to their promising features, such as thinness, lightness, flexibility, stretchability, conformability, and compatibility^{1,2}. Accordingly, they have been applied widely in the development of flexible displays^{3,4}, healthcare monitoring^{5,6}, sensors^{7–9}, artificial skins^{10,11}, energy harvesting and storage devices^{12,13}, and implantable bioelectronics^{14,15}. Tremendous effort has been applied to improve the performance of soft electronics by designing flexible/stretchable structures and/or finding intrinsically soft materials^{16–19}. To this end, several advanced nanomaterials (e.g., graphene, carbon nanotubes (CNT), nanoparticles, nanowires (NW)/nanofibers, nanomembranes) have been developed to manufacture soft electronics due to their large specific surface area and excellent mechanical and electrical properties^{20–22}. There are several configurations to fabricate nanomaterial-enabled soft electronics, such as wavy, prestrain, composite, and transfer^{23,24}. Considering repeatability and large-scale fabrication, it is becoming obvious that both physical and economic factors

will limit their further advances with the aforementioned techniques, in addition to the complex procedures required during the fabrication of nanomaterials^{25,26}. However, electrospinning provides a low-cost, efficient, and large-scale method for preparing one-dimensional nanofibers with exciting mechanical and electrical characteristics, including high porosity, surface area, conductivity, transparency, and ultrahigh flexibility (Fig. 1). In addition, diverse, viable nanofiber assemblies are pivotal in devising thin, soft, lightweight, breathable, and conformable electronic devices. Moreover, the applicability of biocompatible polymers makes electrospinning ideal to fabricate substrate materials for flexible/stretchable electronic devices that directly interface with tissues, organs, or cells^{27–29}.

With the rapid development of electrospinning techniques, significant progress has been made toward functional applications of electrospun nanofibers in soft electronics (Fig. 1)^{30–33}. Organic polymers, small molecules, colloidal particles, composites, organic/inorganic, and organic/organic systems have been successfully electrospun into nonwoven mats through modified electrospinning techniques^{34–36}, further extending the application of electrospinning for flexible/stretchable electronic devices. Although electrospun nanofibers have been widely investigated for several decades, their use as building blocks for flexible and stretchable electronics is still at the early stage despite their advantageous

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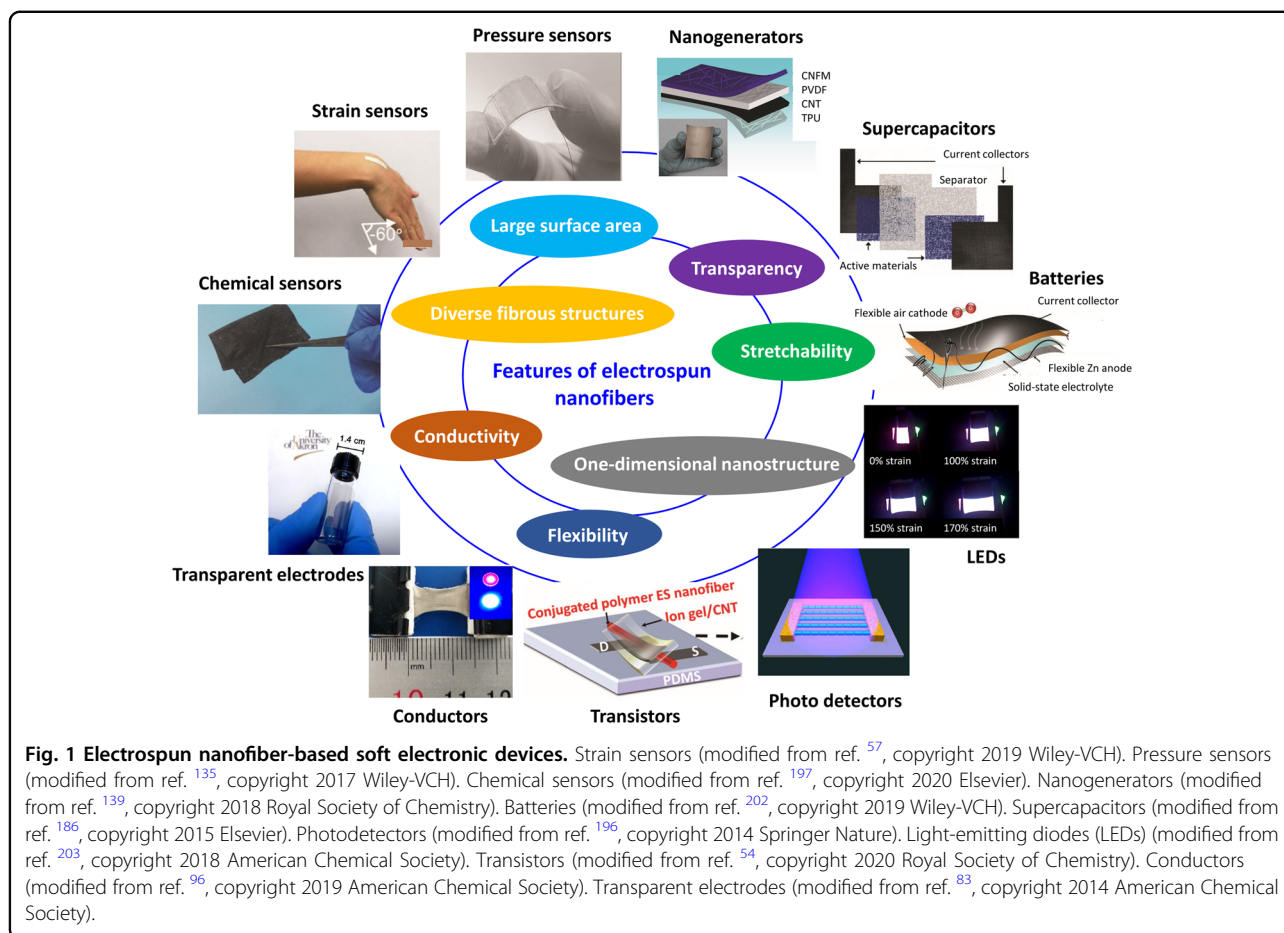
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features^{37,38}. This review aims to introduce readers to the current development of electrospun nanofiber-based soft electronics and to suggest possibilities for several exciting developments. State-of-the-art applications in soft electronics, including flexible/stretchable conductors, transparent electrodes, strain sensors, pressure sensors, energy harvesting and storage devices, transistors, and optoelectronics, are presented in detail. Finally, an outlook on future prospects is also provided.

Electrospinning

Among the different methods for producing one-dimensional nanostructures, electrospinning is the simplest and most economically viable; furthermore, electrospinning has been a commercially successful process for the large-scale production of nanofibers with adjustable diameters (from nanometers to micrometers)^{39,40}. Electrospinning was first studied in the 1930s and began to draw noticeable attention from the late 20th century due to growing industrial and academic interest⁴¹. To date, more than 100 different types of organic polymers, both natural and synthetic, have been successfully explored for solution electrospinning to directly produce continuous nanofibers²⁷.

Technically, electrospinning is a process that uses a strong electrical field to draw a polymer solution or melt into fine filaments. The basic electrospinning setup, shown in Fig. 2, consists of a needle nozzle, high-voltage power supply, container for the spinning fluid, and an electrode collector. When a viscous fluid is charged at a high voltage, the electrostatic force draws the fluid into a liquid jet. Because of the interaction between the jet and the external electric field and the charge repulsion inside the jet, the charged jet undergoes bending or whipping instability to stretch it thinner. Solvent evaporation from the filaments results in solid fibers^{42,43}. In most cases, the as-spun fibers deposit randomly on the electrode collector, forming a nonwoven, entangled nanofiber mat. While the diameters and morphologies (e.g., parallel and crossed fiber arrays, helical or wavy fibers, twisted fiber yarns, patterned fiber webs, and 3D fibrous stacks) of these electrospun nanofibers can be well adjusted by a variety of parameters, such as the intrinsic properties of the solution (e.g., the type of polymer and solvent, the conformation of the polymer chain, viscosity/concentration, elasticity, electrical conductivity, and the polarity and surface tension of the solvent), the operational conditions

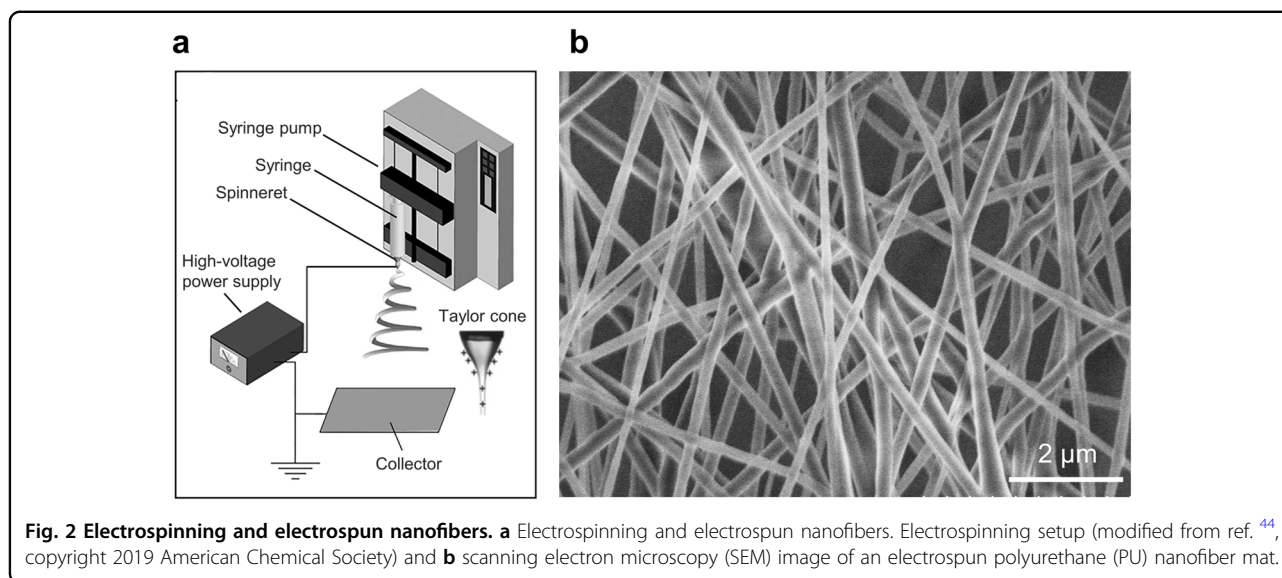


Fig. 2 Electrospinning and electrospun nanofibers. **a** Electrospinning and electrospun nanofibers. Electrospinning setup (modified from ref. ⁴⁴, copyright 2019 American Chemical Society) and **b** scanning electron microscopy (SEM) image of an electrospun polyurethane (PU) nanofiber mat.

(e.g., the strength of the applied electric field, the distance between the spinneret and collector, and the feeding rate of the polymer solution), as well as the humidity and temperature of the surroundings^{44,45}. Therefore, the electrospinning process should be optimized in regard to the above parameters to obtain a uniform, long, continuous fibers with a desired diameter, structure, composition, morphology, porosity, and orientation for use in developing a variety of electronic, environmental, and biomedical applications^{41,46,47}.

Strategies for electrospun nanofibers in soft electronics

Diverse nanomaterials, such as organic polymer nanofibers, carbon nanofibers, metal nanofibers, ceramic nanofibers, and inorganic hybrid nanofibers, have been fabricated directly or indirectly by electrospinning. We have seen their increasingly growing applications in soft electronics, including transparent electrodes, conductors, transistors, optoelectronics, sensors, and energy devices⁴⁸. To this end, different methods have been employed to engineer electrospun nanofibers as passive components (such as functional substrates, templates, and precursors) and active components (such as friction layers for nanogenerators, electrolyte separators for energy storage devices, and sensing layers for chemical sensors) for flexible/stretchable electronic devices⁴⁹. In the following, we will introduce six main strategies to assemble electrospun nanofibers in soft electronic systems (Fig. 3).

Single semiconducting nanofiber

Single electrospun semiconducting nanofibers have been used as channel materials for flexible/stretchable transistors due to their high carrier mobility and good

mechanical flexibility. Compared to conventional film-type semiconducting materials, one-dimensional nanostructures from conjugated polymers exhibit the following promising advantages: (1) a large contact area with a dielectric layer, (2) inherent flexibility, (3) the capability to engineer wrinkled or buckled structures for better stretchability, (4) easier patternability via direct printing, and (5) efficient and easy scale-up fabrication. A variety of nanofibers, such as poly(3-decylthiophene) (P3DT)/poly(3-hexylthiophene) (P3HT) copolymer⁵⁰, poly(3,3''dido-decylquarterthiophene) (PQT-12)/poly(ethylene oxide) (PEO) copolymer⁵¹, P3HT⁵², thiophene diketopyrrolopyrrole (FT4-DPP)/PEO copolymer⁵³, and core-shell P3HT/PEO copolymer⁵⁴, have been fabricated as single nanofiber flexible/stretchable transistors.

Entangled porous nanofiber sheet

Flexible/stretchable electronic devices are generally fabricated by depositing electrical materials onto elastomeric substrates or by embedding them into elastomeric matrixes^{16,55}. The elastomeric materials used for soft electronics are mainly polydimethylsiloxane (PDMS), polyimide (PI), polyethylene terephthalate (PET), Ecoflex, Parylene, latex rubber, paper, etc.¹⁷. However, these polymeric substrates have inherent mechanical mismatches with commonly used electrical materials such as metals, metallic nanomaterials, and carbon-based materials¹⁸. In addition, the limited gas/liquid permeability will restrain their application in on-skin electronics for long-term continuous monitoring⁵⁶. In contrast, elastomer substrates prepared from electrospun nanofiber sheets can not only provide notable mechanical flexibility but also breathability for wearable skin electronics. Furthermore, the large surface area and

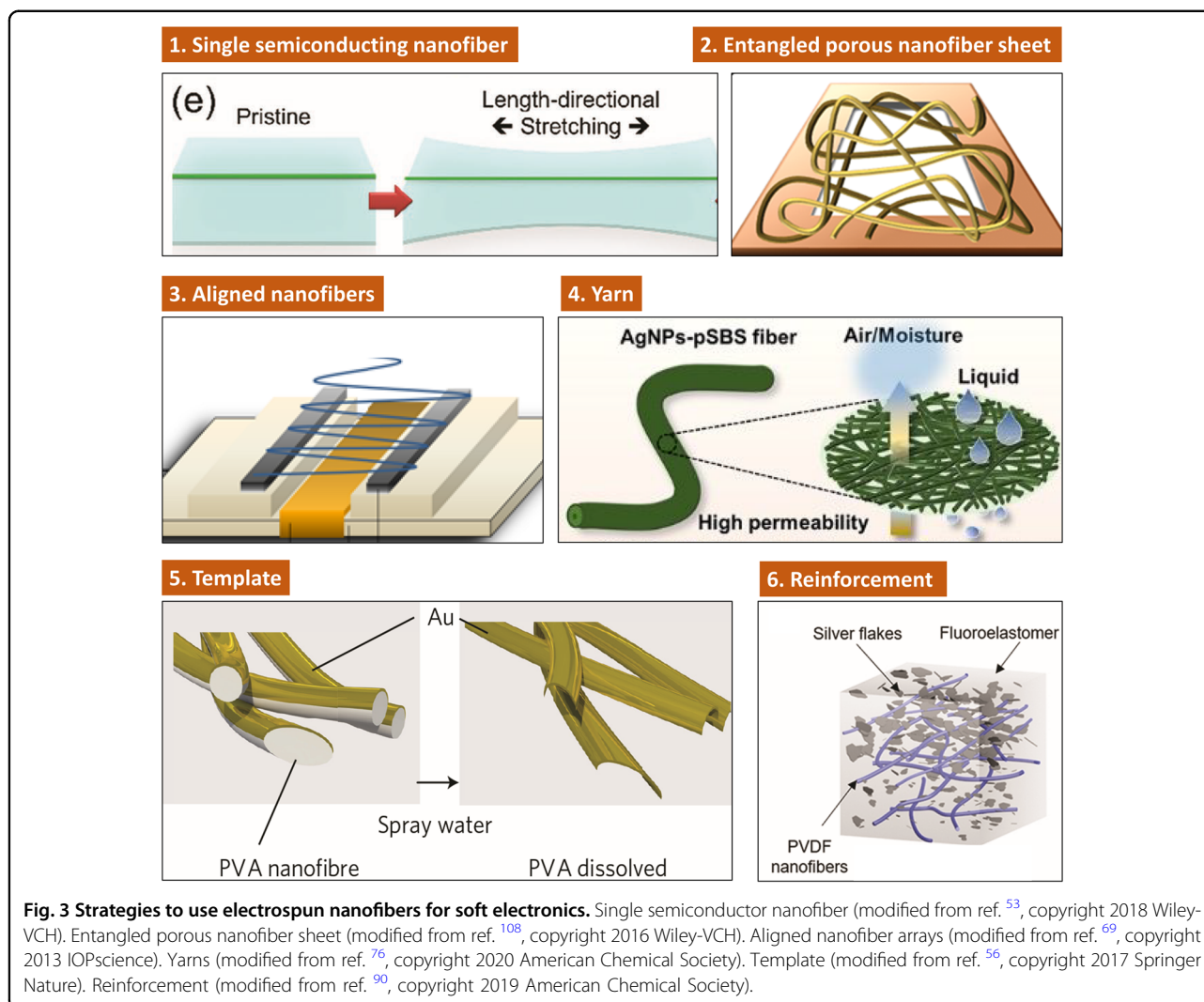


Fig. 3 Strategies to use electrospun nanofibers for soft electronics. Single semiconductor nanofiber (modified from ref. ⁵³, copyright 2018 Wiley-VCH). Entangled porous nanofiber sheet (modified from ref. ¹⁰⁸, copyright 2016 Wiley-VCH). Aligned nanofiber arrays (modified from ref. ⁶⁹, copyright 2013 IOPscience). Yarns (modified from ref. ⁷⁶, copyright 2020 American Chemical Society). Template (modified from ref. ⁵⁶, copyright 2017 Springer Nature). Reinforcement (modified from ref. ⁹⁰, copyright 2019 American Chemical Society).

high pore volume of nanofiber-based substrates can benefit the loading of large amounts of electrically active materials⁴⁴, resulting in highly sensitive devices. Wearable electronic devices can be lightweight, conformable, highly sensitive, flexible, and/or stretchable and breathable while using nonconducting electrospun nanofiber sheets as supporting materials^{57,58}. Recently, a high-adhesion, biocompatible, ultrathin PDMS nano/microfiber mat was developed by using electrospun core-sheath PDMS-polyvinylpyrrolidone (PVP) nanofibers. The PDMS nano/microfiber mat was suggested to be an ideal candidate as a supporting substrate for epidermal electronics⁵⁹. In addition, nonconducting electrospun nanofiber sheets can be used as other components in soft electronic devices, such as electrolytes and separators for batteries and insulating/passivation layers^{60,61}.

Conducting (e.g., carbon, polyvinylidene fluoride (PVDF), composite) nanofiber sheets are applied as active functional

materials in flexible/stretchable electrodes⁶², conductors⁶³, and sensors⁶⁴. Semiconducting, e.g., P3HT, metal oxide, and composite, nanofiber sheets are employed as channel materials in flexible/stretchable transistors⁶⁵, active materials in flexible/stretchable sensors⁶⁶, and friction layers in flexible/stretchable nanogenerators⁶⁷.

Aligned nanofiber array

Highly aligned nanofibers can be easily achieved by adjusting conditions regarding the flow rate and collector during the electrospinning process⁶⁸. Apart from the single semiconducting nanofibers previously mentioned in “Single semiconducting nanofiber”, aligned nanofibers from conjugated polymers have also been used as channel materials. It has been reported that the maximum on-current values can be manipulated by the number of aligned P3HT electrospun nanofibers connecting the source (S)/drain (D) electrodes⁵². Specifically, a transistor based on a single P3HT nanofiber has an on-current value

of 0.7 mA, and the value increases to 3.8 mA when there are 10 aligned nanofibers. Therefore, this approach is promising for fabricating transistor arrays with high density.

A stretchable platinum-based transparent electrode has been made with a sheet resistance of $131 \Omega/\text{sq}$ at $T = 90\%$ by using aligned electrospun nanofibers as templates⁶⁹. Ultrafine and well-aligned PVDF-based nanofibers can be utilized to fabricate high-performance piezoelectric pressure sensors due to the enhanced piezoelectric coefficient^{68,70,71}. Moreover, a multidirectional strain sensor was developed by orthogonally stacking two aligned carbon nanofiber layers with a remarkable selectivity of 3.84⁷².

Yarn

Notably, e-textiles are of great importance for wearable electronics because of their potential use in portable electronics, smart fabrics, and wearable displays⁷³. Though other spinning methods (dry, force, melting, and wet spinning) have been developed to produce nano/microfibers for e-textile applications, they are not suitable for large-scale manufacturing^{74,75}. To this end, diverse three-dimensional porous microfibers or yarn-shaped flexible/stretchable conductors have been prepared by wrapping conductive membranes based on electrospun nanofibers. Typically, entangled nanofibers are loaded with electrically conducting materials, such as CNTs, graphene, and silver nanoparticles (AgNPs)^{76–79}. The as-prepared yarns are generally used as highly sensitive strain and pressure sensors with a large maximum strain (>50%) and good conductivity^{79–82}.

Temporary template

A typical approach to fabricate transparent electrodes is to use electrospun polymer nanofiber mats as temporary templates^{69,83,84}. After the removal of nanofibers by annealing or dissolving in solution, the porous mesh structures of the conductive metal layers are maintained. Characteristics such as the transparency and sheet resistance of the transparent electrode can be well tuned by changing the morphology of the electrospun nanofiber sheet. Furthermore, a substrate-free, breathable, on-skin conductor can be readily obtained by dissolving polyvinyl alcohol (PVA) nanofiber sheets with a water mist after attachment^{56,85}.

Reinforcement

Inspired by reinforcement wires in concrete, numerous nanotube or nanowire-based materials have been widely used as reinforcement materials in composites to enhance mechanical performance^{86,87}. Upon incorporating electrospun nanofibers into an elastomer matrix, the as-received elastomer substrate becomes a skin-like substrate even though the thickness is on the micrometer scale^{88,89}. The good mechanical properties originate from the existence of

randomly distributed nanofibers in the polymer matrix. In addition, the fiber reinforcing effect enhances the toughness and suppresses crack growth due to the embedded nanofibers in the composite conductor⁹⁰.

Soft electronic applications of electrospun nanofibers

Because the impressive properties of electrospun nanofibers in one-dimensional nanostructures, namely, the porosity, flexibility, conductivity, transparency, and diverse fibrous morphologies are vital to designing high-performance soft electronics, a number of recently developed flexible/stretchable electronic devices are discussed and summarized in the following sections, including conductors, transparent electrodes, strain sensors, pressure sensors, nanogenerators, supercapacitors, batteries, transistors, and other applications.

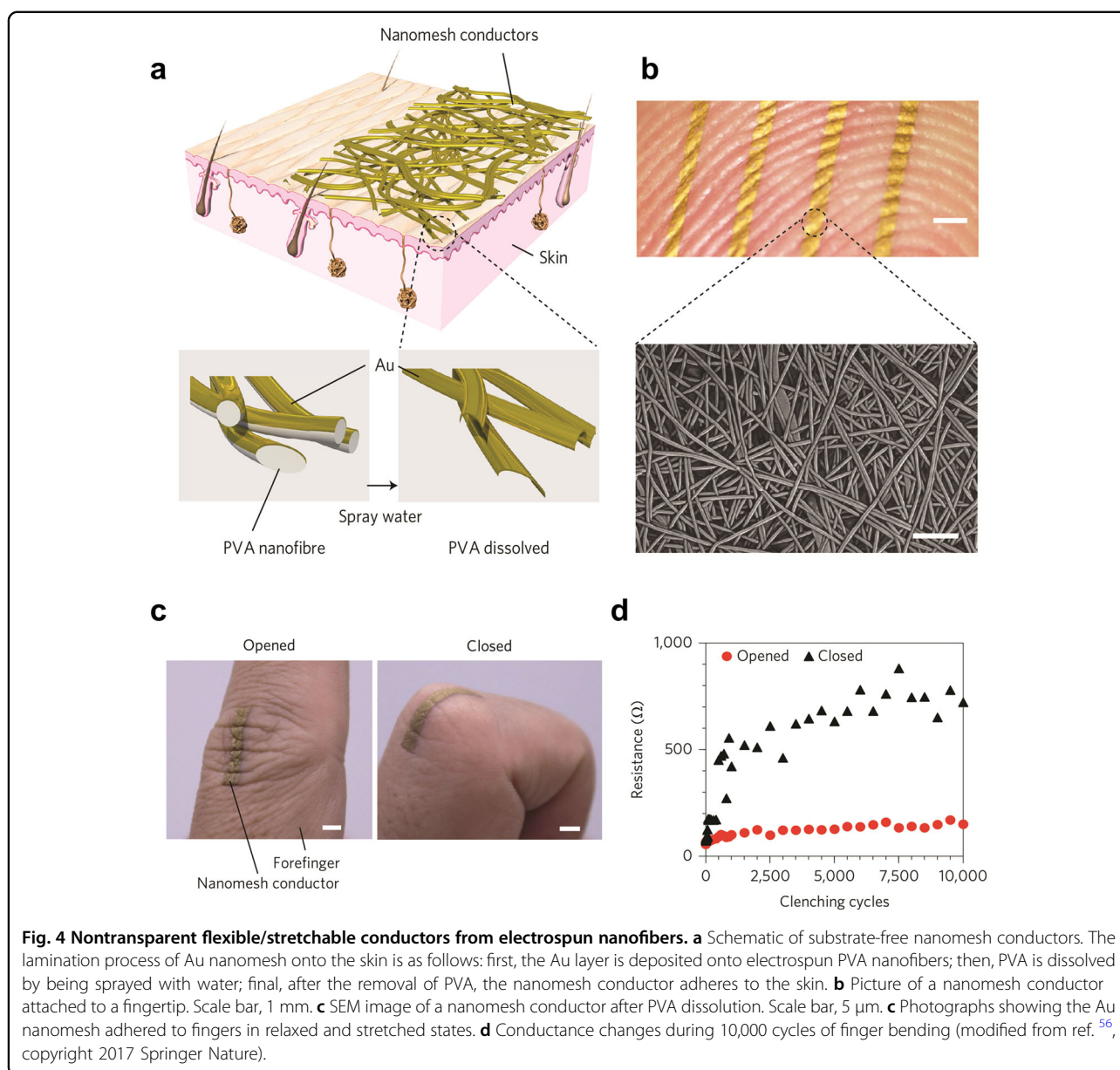
Conductors

Flexible and/or stretchable conductors are indispensable components for soft electronic integrations to develop next-generation wearable electronics and soft robotics^{20,91,92}. In this section, we introduce flexible/stretchable nontransparent conductors and transparent electrodes made from electrospun nanofibers.

Nontransparent conductors

The basic feature of a flexible and/or stretchable conductor is that it retains high conductivity under certain mechanical deformations^{93–95}. Thanks to the excellent flexibility of the nanofiber-networked porous structure from electrospinning, a plethora of flexible/stretchable conductive films or yarns have been fabricated^{63,76–78,96,97}. For example, carbon nanofiber (CNF)/graphene nanosheet (GNS) composite paper can be directly prepared by electrospinning after adding GNS to the electrospun polymer precursor. The flexible paper shows a specific capacitance up to 197 F/g, which is 24% higher than that of pure CNF paper, revealing its promise for application in high-performance flexible capacitors⁹⁸.

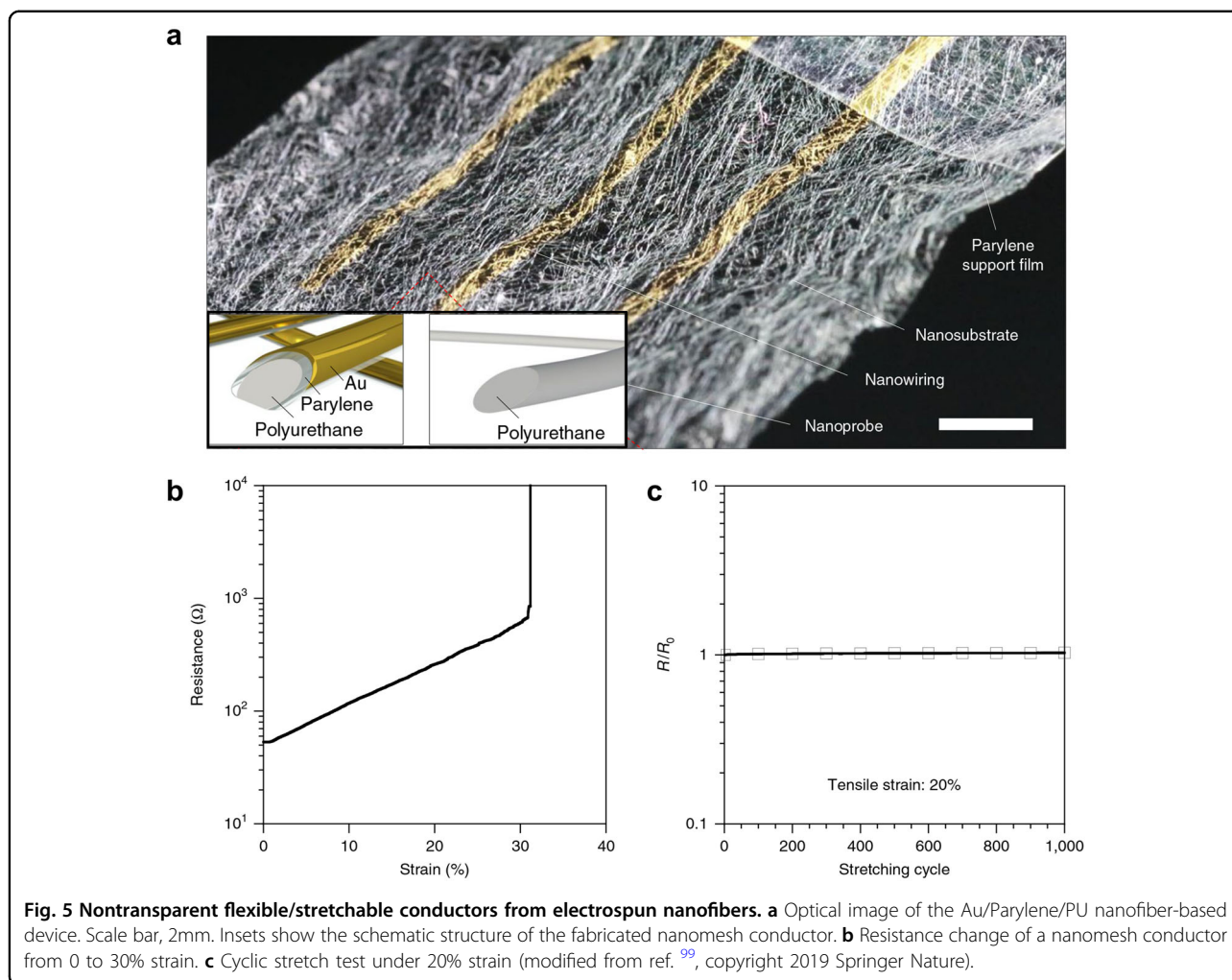
By using water-dissolvable PVA electrospun polymers, Someya et al. reported substrate-free on-skin electronics^{56,85}. The fabrication process was simple and effective. In brief, a PVA nanofiber sheet was prepared by electrospinning. Then, a 70–100-nm-thick Au layer was deposited on top using a shadow mask. When the Au/PVA conductors were placed on skin and sprayed with water, the PVA nanofibers easily dissolved, and the Au nanomesh conductor adhered to the skin (Fig. 4a). Figure 4b shows the fine ridges of the human finger, demonstrating the good conformability of the material due to the ultrathin adhesive layer produced by the dissolved PVA. Compared to conventional planar substrates, substrate-free nanomeshes offer notable gas permeability and a significant decrease in



stiffness, rashes, and other inflammatory skin reactions. Furthermore, these nanomesh conductors can survive 10,000 cycles of finger bending ($\sim 40\%$ strain) (Fig. 4c, d). A recently published work has shown that with heat-press treatment at 180°C , the resistance of the nanomesh conductors decrease by 76%, and the stretchability increases from 25 to 80% strain⁸⁵.

Flexible/stretchable conductors have also been fabricated when electrospun nanofibers are used as supporting scaffolds for electrical materials. Someya et al. presented an ultrasoft Au/Parylene/PU conductor and employed it to monitor dynamically pulsing cardiomyocytes⁹⁹. The PU nanofiber (diameter: 800–900 nm) sheet was ultrathin and flexible and had a coverage of 10–20%. The Parylene layer

(200-nm-thick) was deposited via chemical vapor deposition to uniformly cover the PU nanofibers. Finally, the conductor was completed by depositing a 100-nm-thick Au layer on top (Fig. 5a). The effective Young's modulus of the PU nanosubstrate was estimated to be 0.274 ± 0.039 MPa, which was two to three orders of magnitude lower than that of a uniform pure Parylene layer. The Au/Parylene/PU nanomesh conductor could be stretched to 30% strain while remaining highly conductive ($211.6 \pm 63.4 \Omega$) (Fig. 5b). Moreover, this conductor exhibited stable electrical responses under repetitive stretching (up to 20% strain) and releasing despite their ultrasoftness (Fig. 5c). To increase stretchability and conductivity, electrospun polymer nanofibers can also



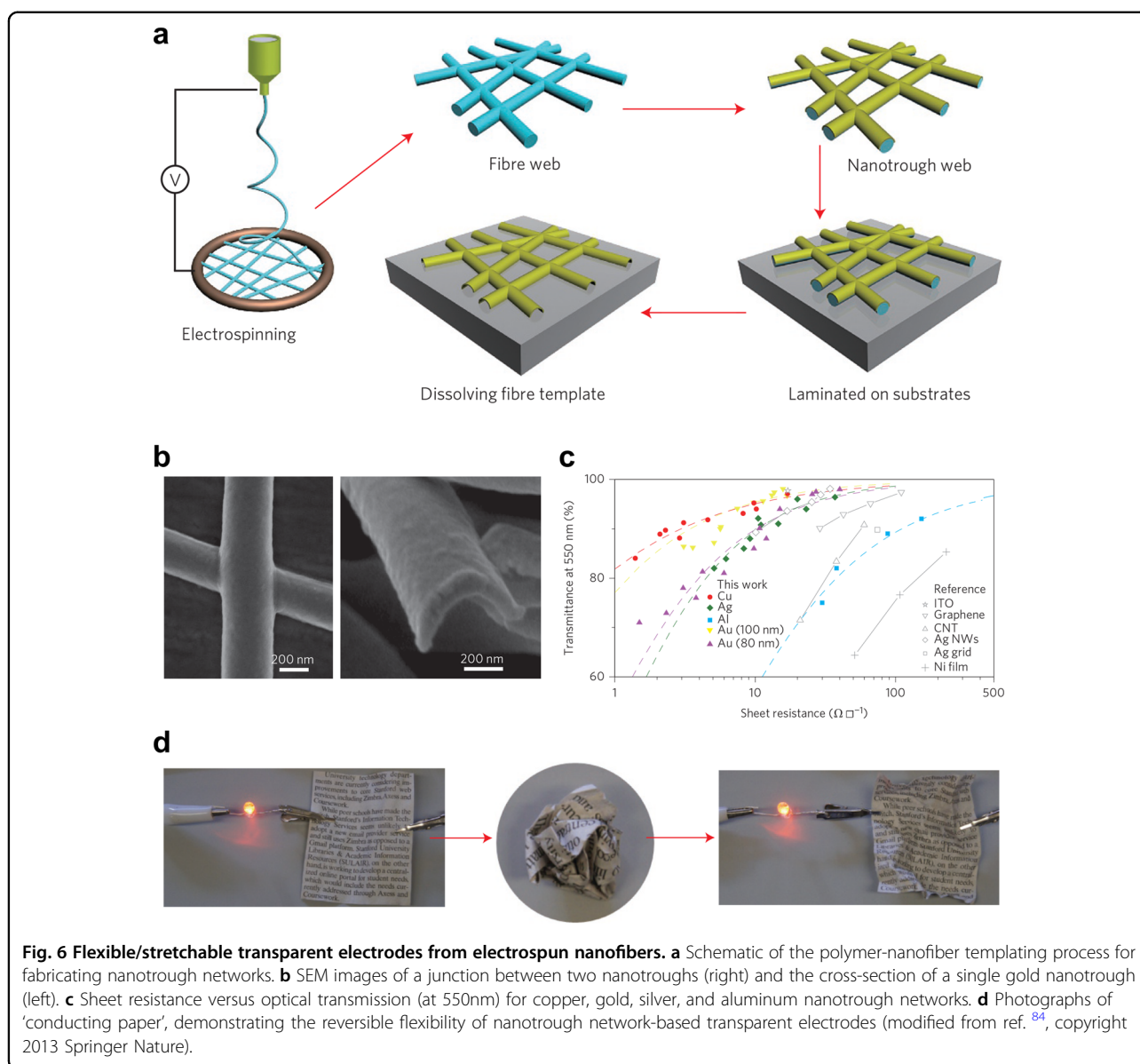
function as reinforcements^{57,90}. A porous AgNW/PU nanofiber elastic conductor can be prepared with a layer-by-layer structure of nanofibers/nanowires due to interfacial hydrogen bonding⁵⁷. The resultant nanofiber-based conductor exhibits a conductivity of up to 9190 S/cm, good stretchability of 310% strain, and high cycling durability at 70% strain. The as-prepared conductor also demonstrates good breathability for continuous on-skin electrophysiological signal recording.

Transparent electrodes

Flexible transparent electrodes, which combine high electrical conductivity, optical transparency, and mechanical flexibility, play a fundamental role in soft electronic applications, including transparent heaters, photovoltaics, supercapacitors, light-emitting diodes, and batteries¹⁰⁰. Although transparent conductive oxides are universally used as conducting materials, their brittleness and required high-pressure vacuum processes during fabrication limit their applications in flexible/stretchable electronics¹⁰¹.

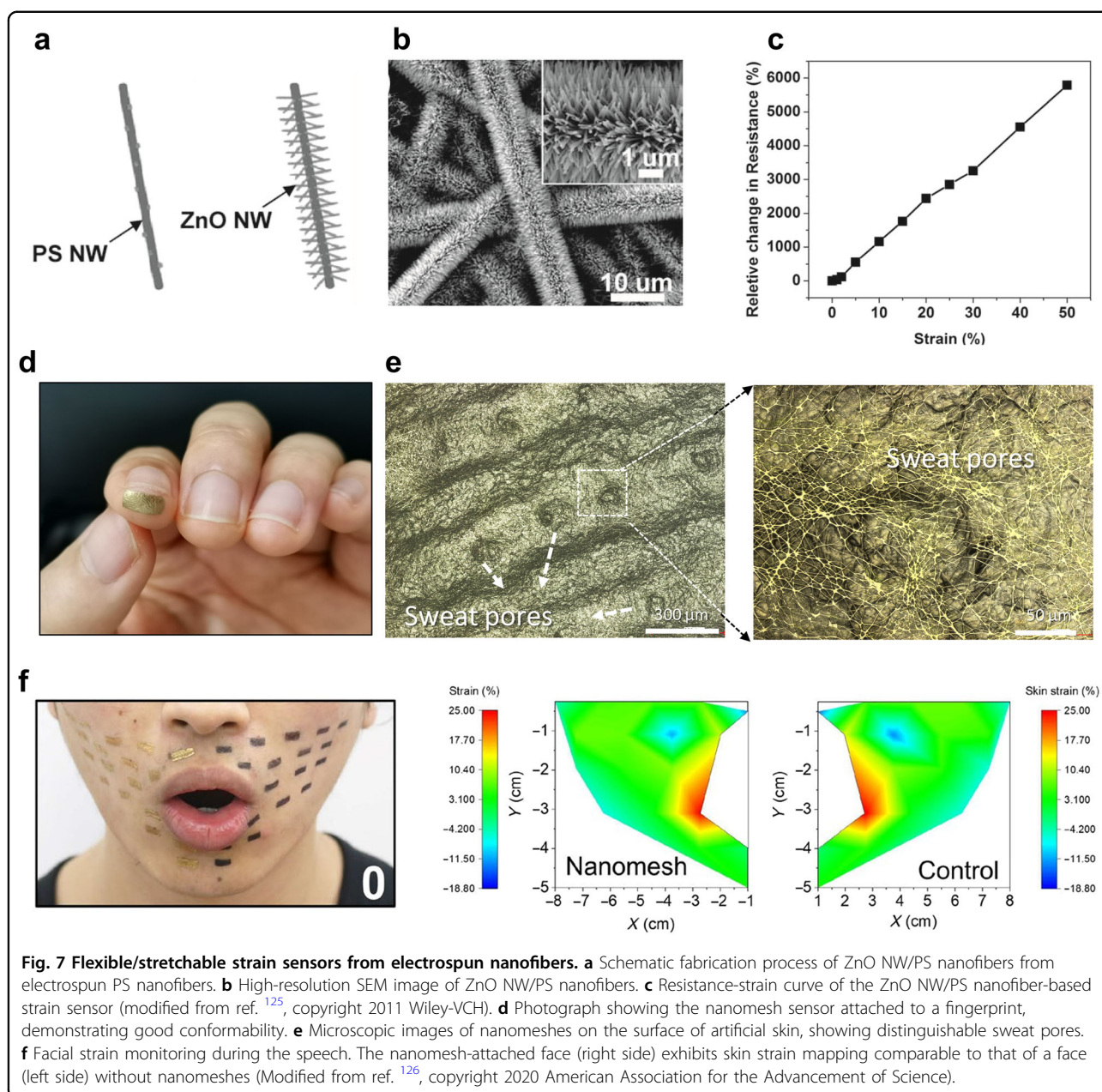
Considerable efforts have been directed toward manufacturing flexible transparent electrodes with one-dimensional nanowires or nanofibers owing to their outstanding physical properties^{48,102}. Owing to electrospinning, a low-cost and high-throughput method, diverse materials such as metals, polymers, and composites have been manufactured directly or with other techniques to construct flexible/stretchable transparent electrodes^{69,83,103–106}.

A representative example is a metal nanotrough network-based transparent electrode developed by Cui et al.⁸⁴. First, water-soluble PVA and PVP are electrospun into polymer nanofibers as templates. Then, a thermal evaporator is employed to deposit metals onto the porous nanofiber sheet. Finally, the transparent electrode is obtained by dissolving the underlying polymer nanofiber support (Fig. 6a). Various materials have been used via this approach to fabricate transparent electrodes, including silicon, indium tin oxide (ITO) and metals, such as gold, silver, copper, platinum, aluminum, chromium, nickel, and their alloys. After the polymer



nanofiber is removed, the nanotroughs are found to be connected at junctions and exhibit concave cross-sections (Fig. 6b). The sheet resistance (R_s) values of copper nanotrough networks have the best performance: $2 \Omega/\text{sq}$ at $T=90\%$, $10 \Omega/\text{sq}$ at $T=95\%$, and $17 \Omega/\text{sq}$ at $T=97\%$, comparable to that of state-of-the-art device-grade ITO (Fig. 6c). Furthermore, the nanotrough networks show good adhesion after attaching to flexible substrates such as PET plastic and paper (Fig. 6d)⁸⁴. After introducing an in-plane sinusoidal wavy structure, an Au nanotrough networks can be stretched to more than 300% strain¹⁰⁷. This buckled Au nanotrough electrode exhibits fatigue-free (>100% strain) properties and presents high optical transparency of 91% with a sheet resistance of $10 \Omega/\text{sq}$.

Transparent self-junctioned metal electrodes can also be made by seeding metal ions or nanoparticles onto electrospun polymer nanofibers and subsequently performing electroplating or oxidation^{105,108–110}. In addition to thermal evaporation and electroless deposition, the direct coating of functional materials onto electrospun polymer nanofiber scaffolds is also effective in creating percolation networks for transparent electrodes^{106,111,112}. After immersion in a highly concentrated dispersion of single-walled CNTs, the electrospun nylon 6 membrane becomes a conductive transparent film with a sheet resistance of $2100 \Omega/\text{sq}$ at $T=77.4\%$ ¹⁰⁶. These flexible or stretchable transparent electrodes have been demonstrated in applications such as solar cells^{109,111,113}, flexible touch screens⁸⁴, batteries¹¹¹, electrophysiological sensors¹¹², and LEDs^{110,114,115}.



Sensors

Various highly sensitive flexible/stretchable sensors have been developed using electrospun nanofibers owing to their high surface area and ultraflexibility. Strain sensors and pressure sensors, the two most studied sensors using electrospun nanofibers, are reviewed in this section.

Strain sensors

Stretchable strain sensors have been extensively developed over the past two decades and have been employed in various applications, such as human activity monitoring and soft robotics^{116,117}. The working mechanism is that mechanical deformations induce electrical responses in

terms of resistance, capacitance or voltage^{118,119}. Electrospun nanofiber-based strain sensors exhibit lightness, flexibility/stretchability, and gas permeability, all of which are significant in wearable electronic applications. An effective way to fabricate strain sensors from electrospun nanofibers is to decorate or deposit functional materials onto nanofiber scaffolds due to their high surface area^{79,120–124}. A typical example is the use of a hybrid ZnNW/polystyrene (PS) nanofiber network as the functional material of a strain sensor¹²⁵. The ZnNW/PS nanofiber porous structure is fabricated by attaching ZnO seeds onto electrospun PS nanofibers and subsequently growing ZnO NWs. The fabricated sensor can be

stretched up to 50% strain with a gauge factor of ~ 116 (Fig. 7a–c).

To continuously monitor skin strain without disturbing natural skin motions, Someya et al.¹²⁶ developed a breathable, ultrathin, and soft nanomesh skin-strain gauge. The nanomeshes were fabricated by reinforcing electrospun PU nanofibers formed by an electrospinning method with extremely thin PDMS. The sensor was ultralightweight (0.012 mg/cm^2) and extremely thin (430 nm), which allowed it to adhere to the skin and detect minute skin strains (Fig. 7d). The ultraconformability was also demonstrated by the distinguishable sweat pores shown in Fig. 7e while laminating the nanomeshes onto the surface of artificial skin. The as-fabricated sensor exhibited high mechanical durability (a resistance degradation of $<3\%$ after 5000 repetitive stretching and releasing cycles under 60% strain) and excellent sustainability (under 40% strain for 12 h with a <0.053 electrical drift error). The reliable electrical responses were independent of the stretch frequencies, and its thin geometry and softness provided minimum mechanical interference on natural skin motions. Hence, the skin strain could be quantitatively measured without affecting the natural movement of the skin. By attaching the developed nanomesh sensor to the face, they succeeded in measuring

mapping facial strains during speech (Fig. 7f). The measured strain from the attached sensors on the right side of the face was asymmetric to that of the left side of the face with black markers. They also succeeded in mapping facial skin strains during a speech even after 3.5 h of wearing the device. This nanomesh sensor is expected to be used in smart facial masks that can accurately recognize facial microexpressions such as joy, anger, sadness, and fear.

Other strategies to fabricate stretchable strain sensors include the following: (1) directly electrospinning polymer nanofibers and carbonizing^{72,127}; (2) directly electrospinning functional nanofibers, such as PVDF nanofibers^{64,71,128}; and (3) adding functional materials to the polymer solution for electrospinning⁸⁰. When any of these three strategies are used, the alignment, density, and diameter of the electrospun nanofibers are tuned to obtain the best performance of the stretchable strain sensor. Table 1 illustrates examples of electrospun nanofiber-based stretchable strain sensors using the aforementioned strategies.

Pressure sensors

The measurement of small normal pressures is critical to accurately evaluate external stimuli on curvilinear and

Table 1 Summary of electrospun nanofiber-based stretchable strain sensors.

Electrospun NFs	Functional materials	Gauge factor	Stretchability	Durability	Mechanism	Refs
TPU	GNRs	7.9–35.7	160%	5000 cycles (100%)	Resistive	204
PU	CNTs	114–720	220%	10,000 cycles (30%)	Resistive	79
PU	GO/PEDOT	10.1–193.2	$\sim 550\%$	4 cycles (160%)	Resistive	120
PAN	CNFs	5–72	300%	8000 cycles (10%)	Resistive	127
TPU	Graphene	3–21	100%	1000 cycles (50%)	Resistive	205
PA6	CNTs	2.8–12	40%	100 cycles (20%)	Resistive	124
PAN	Graphene/CNFs	277–1700	2%	300 cycles (2%)	Resistive	80
TPU	CNTs	1.24–1.67	100%	2000 cycles (50%)	Resistive	81
PU	SiO ₂ /graphene	1.1–5.9	110%	600 cycles (50%)	Resistive	121
TPU	RGO	11–79	100%	6000 cycles (50%)	Resistive	122
PVDF-HFP	PVDF-HFP NFs		100%	300 cycles (100%)	Triboelectric	128
PAN	Aligned CNFs	0.3–180	30%	2500 cycles (25%)	Resistive	72
PU	CNTs	46.4–980	44%	1000 cycles (20%)	Resistive	82
PS	ZnO NW	116	50%	17 cycles (30%)	Resistive	125
P4VP	AuNPs	3.67–11	17%	30 cycles (3%)	Resistive	123
PVDF	PANI/PVDF	0.045–0.84	110%		Resistive	64
PVDF	Aligned PVDF NFs	4.29 mV/%	10%	8 cycles (10%)	Piezoelectric	71
PU	Au	7.26–46.3	150%	5000 cycles (100%)	Resistive	126

NFs nanofibers, TPU thermoplastic PU, PA6 polyamide 6, PAN polyacrylonitrile, HFP hexafluoro propylene, P4VP poly(4-vinylpyridine), PANI polyaniline, GNR graphene nanoribbons, RGO reduced graphene oxide, PEDOT poly(3,4-ethylenedioxythiophene).

Table 2 Summary of electrospun nanofiber-based pressure sensors.

Electrospun NFs	Functional materials	Sensitivity	Detection limit	Durability	Mechanism	Refs
TPU	Dielectric layer	1.21 kPa ⁻¹	0.9 Pa	10,000 cycles (600 Pa)	Capacitive	132
Silk	CNFs	1.16–34.47 kPa ⁻¹	0.8 Pa	10,000 cycles (2.5 kPa)	Resistive	135
PU	CNTs	12.3 N ⁻¹	0.001 N	0.001–5 N	Resistive	79
ZnO/PVDF	PVDF	0.33 V/kPa		5000 cycles (4.67 kPa)	Piezoelectric	147
ZnO/SiO ₂	ZnO/SiO ₂	~0.32 kPa ⁻¹		5 cycles (32.6 kPa)	Resistive	206
P(VDF-TrFE)	P(VDF-TrFE)	60.5 mV/N		12 cycles (0.05 N)	Piezoelectric	207
PVDF, EC	PVDF	1.67–0.20 V/kPa		7200 cycles (7200 MΩ)	Triboelectric	133
P(VDF-TrFE)	P(VDF-TrFE)	1.1 V/kPa	0.1 Pa	7200 cycles (Bending)	Piezoelectric	68
PVA-co-PE, POE	PPy at PVA-co-PE NFs	1.24 kPa ⁻¹	1.3 Pa	50 cycles (6.5, 653 Pa)	Capacitive	208
PAN	PAN-C/BTO	1.44 V/N	0.15 N	60,000 cycles (600 Pa)	Piezoelectric and triboelectric	209
P(VDF-TrFE)	P(VDF-TrFE)	141.7 mV/kPa		5000 cycles (75 kPa)	Piezoelectric	210
P(VDF-TrFE)	P(VDF-TrFE)/MWCNT	540 mV/N		~1300 cycles (2.5 N)	Piezoelectric	70
PANF	Ag NWs/GR/PANF	0.7–134 kPa ⁻¹	3.7 Pa	8000 cycles (2 kPa)	Capacitive	211

P(VDF-TrFE) poly[(vinylidene fluoride)-co-trifluoroethylene], *EC* ethyl cellulose, *PVA-co-PE* poly(vinyl alcohol-co-ethylene), *POE* polyolefin elastomer, *PPy* polypyrrole, *PAN-C/BTO* carbonized polyacrylonitrile/barium titanate, *MWCNT* multiwalled carbon nanotube, *PANF* polyamide nanofiber, *GR* graphene.

dynamic surfaces such as human skin and natural tissues^{129–131}. Electrospun nanofiber mats have high tolerance for repetitive external pressing due to their porous structures and high flexibility. Therefore, a large number of wearable pressure sensors have been constructed using electrospun nanofibers. Among these sensors, there are four types of working mechanisms: capacitive¹³², resistive⁶⁶, triboelectric¹³³, and piezoelectric¹³⁴. Based on this, electrospun nanofibers are applied as dielectric layers, conductive layers, or electrode materials in pressure sensor assemblies. For example, Zhang et al.¹³⁵ used carbonized silk nanofibers as a conductive layer and fabricated a resistive pressure sensor. The developed pressure sensor had a rapid response time of <16.7 ms, a low detection limit of only 0.8 Pa, and a sensitivity of up to 34.47 kPa⁻¹. These outstanding performances were attributed to the unique N-doped silk nanofiber mats. To optimize piezoelectric responses, a highly aligned P(VDF-TrFE) nanofiber array has been fabricated into a flexible and freestanding sheet by electrospinning on a fast-rotating collector⁶⁸. A flexible pressure sensor can be simply formed by adding two electrical contacts at the ends of the as-fabricated sheet, and the resulting pressure sensor can detect pressure signals that are as low as 0.1 Pa. In addition, the device exhibits an excellent response under an extremely low-pressure regime, which suggests its potential utility as a sensor-energy harvesting component for applications in human motion detection and robotics⁶⁸. Table 2 provides a summary of recently reported electrospun nanofiber-based flexible/stretchable pressure sensors.

Energy harvesting and storage devices

Wearable energy generation and storage devices are prerequisites to implement independent wearable electronic platforms. In this section, we present the development of soft energy harvesting devices (e.g., nanogenerators, batteries) and storage devices such as supercapacitors based on electrospun nanofibers.

Nanogenerators

There has been an increasing demand for renewable energy due to the rapid consumption of fossil fuels as well as environmental issues. Hence, mechanical energy harvesting systems that can convert small-scale energy from vibrations and motions into electrical power have been developed to solve this problem⁴⁹. To date, there are two major types of nanogenerators: piezoelectric nanogenerators (PENGs) and triboelectric nanogenerators (TENGs). PENGs have attracted extensive attention since 2006 and are mainly fabricated from PVDF-based materials due to their large piezoelectric coefficients¹³⁶. TENGs were first demonstrated in 2012 by Wang et al., in which they coupled the triboelectric effect and electrostatic induction^{137,138}. With the introduction of piezoelectric, triboelectric, and/or pyroelectric electrospun nanofibers, a flexible/stretchable electronic system can realize self-powered operation with attractive mechanical and electrical performances^{139–144}.

For example, a breathable, all-nanofiber TENG has been fabricated for effective mechanical energy harvesting and whole-body physiological signal detection¹⁴⁵. The device is composed of a top polylactic-co-glycolic acid (PLGA) triboelectric layer, a bottom PVA nanofiber substrate, and an

AgNW layer in between. This one-electrode TENG shows a high power density of 130 mW/m^2 and pressure sensitivity of up to 0.011 kPa^{-1} owing to the high surface area provided by the three-dimensional micro-to-nano hierarchical pores. TENG-based e-skin can be used to monitor whole-body physiological signals with good breathability, biodegradability, and antibacterial properties¹⁴⁵. By anchoring piezoelectric barium titanate nanoparticles onto P(VDF-TrFE) nanofibers, the as-achieved PENG exhibits a significantly enhanced output of 6 V and $1.5 \mu\text{A}$. The values for the PENG made from only P(VDF-TrFE) membranes are only 1.25 V and $0.6 \mu\text{A}$ ¹⁴⁶. Moreover, various pressure/tactile sensors have been developed by piezoelectric and/or triboelectric electrospun nanofibers for manufacturing wearable energy-sustainable electronic devices for monitoring human motion^{103,134,140,147–152}.

Batteries

Lithium (Li)-ion batteries play a pivotal role in energy storage systems for fast-growing demands in hybrid/full electric vehicles, portable devices, and power tools^{153,154}. A Li-ion battery consists of a cathode, electrolyte, separator, and Li anode¹⁵⁵. The separator is sandwiched between the two electrodes and immersed in the electrolyte. Li is the lightest metal in electrochemical energy storage and has the lowest reduction potential and an excellent theoretical specific capacity of 3860 mAh/g ⁶⁰. Owing to their large surface area, high porosity, and good conductivity, electrospun nanofibers have shown great potential for the development of flexible/stretchable Li-ion batteries⁴⁴. To date, various electrodes, separators, and electrolytes have been developed using electrospun nanofibers to improve the electrochemical, mechanical, and safety performances of Li-ion batteries^{156–160}.

Free-standing Li_2S paper electrodes based on electrospun carbon nanofibers have been fabricated for flexible high-energy Li-ion batteries¹⁶¹. The free-standing conductive electrode also largely lowers the cost and manufacturing complexity of Li_2S cathodes. The flexible paper electrode is composed of N-doped carbon nanofibers (NCNFs) decorated with ultrafine Li_2S nanoparticles (Fig. 8a–c). These mechanically robust paper electrodes allow a high content of Li_2S loading through layer-by-layer stacking. This highly opened but microscopically confined design enhances structural stability, reaction kinetics, and areal specific capacities. In addition, high performance with a high capacity of 520 mAh/g (4.68 mAh/cm^2) is maintained with a stable capacity retention of over 65% for 200 cycles when utilizing a very high Li_2S loading of 9.0 mg/cm^2 (Fig. 8d); additionally, an approximately 100% coulombic efficiency is achieved. Electrodes with a Li_2S loading of 6.0 mg/cm^2 can deliver high capacities of 650, 540, 505, and 490 mAh/g (3.9, 3.24, 3.03, and 2.94 mAh/cm^2 , respectively) at current

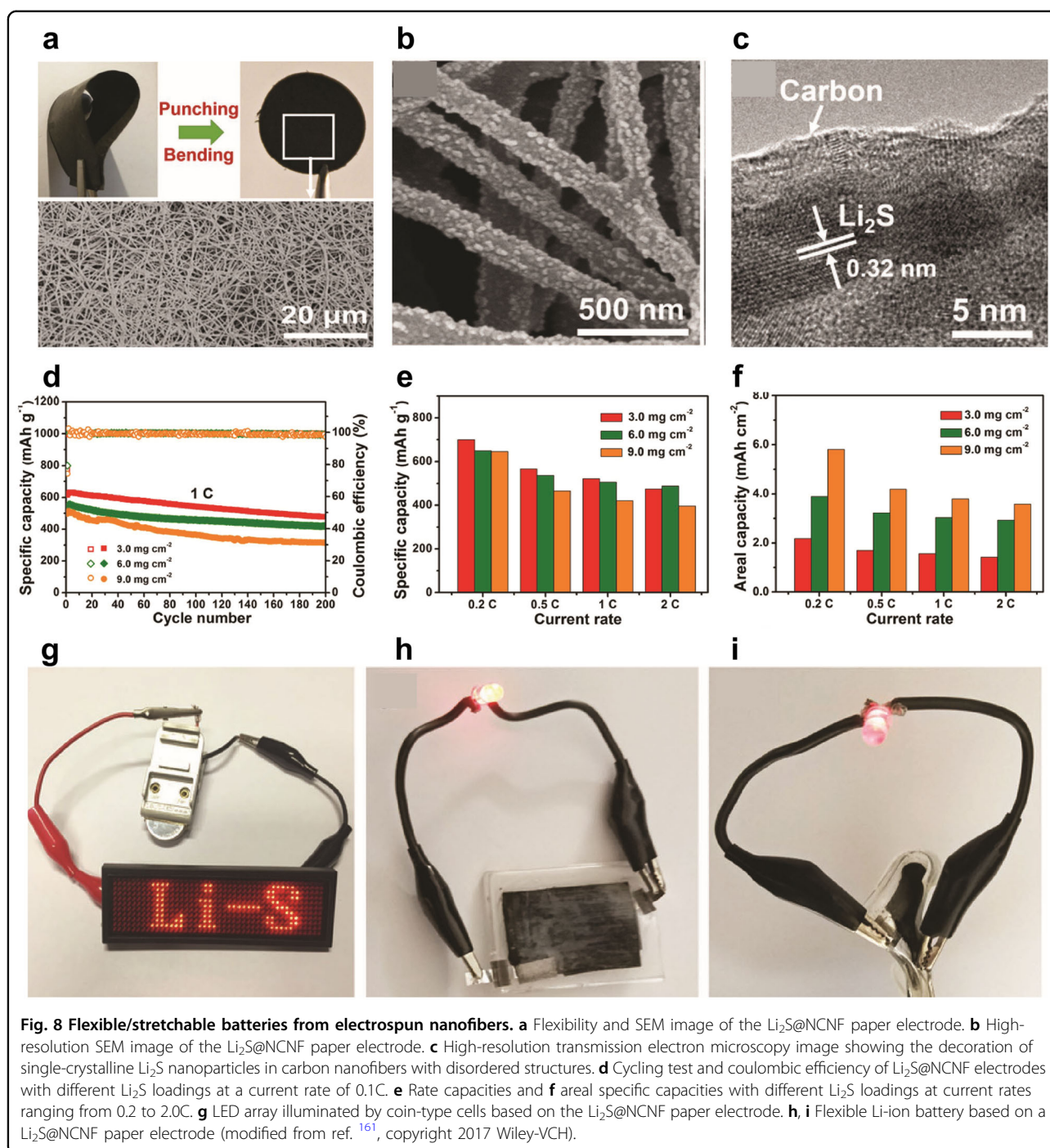
rates of 0.2, 0.5, 0.1, and 2.0C, respectively. The high rate capacity is retained with multilayered $\text{Li}_2\text{S}@ \text{NCNF}$ electrodes (Fig. 8e, f). The high initial capacity of $\text{Li}_2\text{S}@ \text{NCNF}/\text{Fe}_3\text{O}_4$ full cells can power an LED array consisting of 11×44 diodes (Fig. 8g). Figure 8h, i shows that the as-obtained prototype battery exhibits excellent flexibility while powering an LED¹⁶¹. Recently, Jeong et al. reported an intrinsically stretchable Li-ion battery that exhibited a high capacity of 91.8 mAh/g at 100% strain using an electrospun poly(styrene-*b*-butadiene-*b*-styrene) (SBS) block copolymer nanofiber mat as the separator membrane¹⁶². The developed stretchable rechargeable battery was successfully used to power stand-alone intrinsically stretchable electronic device platforms.

In addition to Li-ion batteries, other batteries, such as sodium-ion and zinc-air batteries, can also be achieved with electrospun nanofibers^{163–165}. For example, by loading $\text{S}_{1-x}\text{Se}_x$ on porous carbon nanofibers, a flexible cathode electrode is achieved, which delivers a high reversible capacity of 840 mAh/g after 100 cycles at 0.1 A/g for Li-S batteries and 762 mAh/g after 100 cycles at 0.1 A/g for Na-S batteries¹⁶⁶.

Supercapacitors

Supercapacitors, i.e., electrochemical capacitors, are energy storage devices that possess advantageous properties, such as fast charging/discharging capacities, high power and energy densities, long-term cycling, and safe operation^{167,168}. A supercapacitor consists of two electrodes with a sandwiched ion-permeable electrolyte layer/separator^{169,170}. To integrate a supercapacitor into a soft electronic system, it should maintain high performance under repeated mechanical deformations^{171,172}. Following this target, the development of flexible/stretchable supercapacitors mainly focuses on designing highly flexible/stretchable capacitive materials for electrodes^{173,174}. As a novel class of electrode materials, porous electrospun capacitive nanofibers are able to provide (1) mechanical flexibility, (2) a large surface area accessible to the electrolyte, (3) efficient ion transport kinetics, and (4) structural stability^{62,160}. Consequently, high capacitive performance can be readily realized by engineering the pore size distribution of interconnected nanofibers^{175,176}.

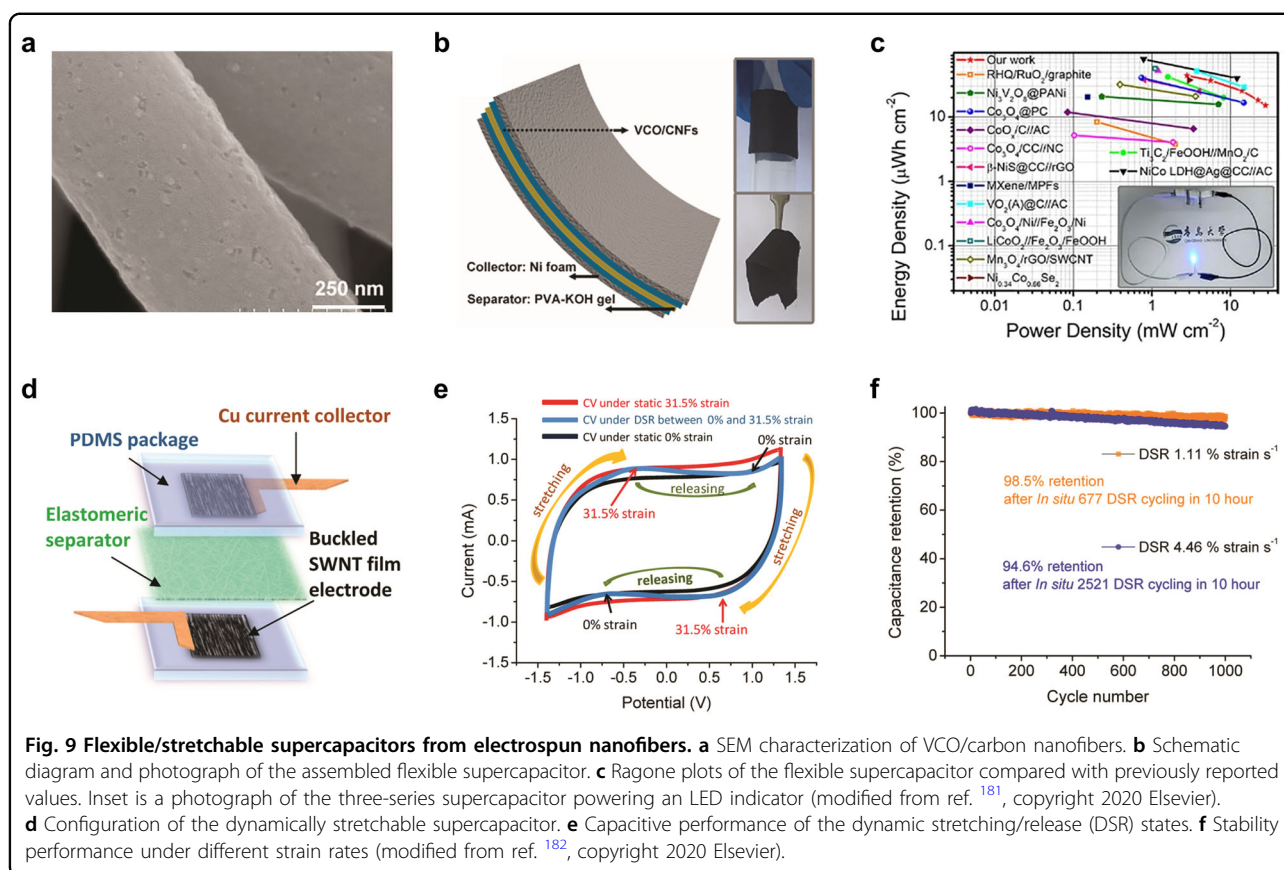
Electrospun carbon nanofiber-based electrodes are one of the most important components of supercapacitors because of their large specific surface area and good conductivity and chemical stability¹⁷⁷. Strategies such as dopants, hollow structures, and composites have been applied to enhance electrochemical activity^{178–180}. For instance, vanadium (V)/cobalt monoxide (CoO) has been introduced with conductive carbon nanofibers to produce extra reaction sites, accelerate electron transfer, and stabilize the electrode structure with good mechanical



flexibility and accessibility for real applications¹⁸¹. VCO/carbon nanofibers are achieved by electrospinning PAN, which is subsequently annealed (carbonization process) (Fig. 9a). The above electrode yields a high areal capacitance of 1.83 F/cm^2 at a current density of 8 mA/cm^2 . A binder-free, nonpolar asymmetric flexible supercapacitor is then assembled with PVA/KOH gel as the electrolyte and separator between the two binder-free VCO/carbon

nanofiber electrodes (Fig. 9b). The supercapacitor yields an areal energy density up to $44.2 \mu\text{Wh/cm}^2$ at a power density of 2.8 mW/cm^2 (Fig. 9c). The device also exhibits good mechanical flexibility and is able to survive a high bending angle of 180° with a small capacitance decay of 5.5%¹⁸¹.

Electrospun nanofiber mats can also be employed as a separator in a supercapacitor^{182,183}. Wei et al. demonstrated

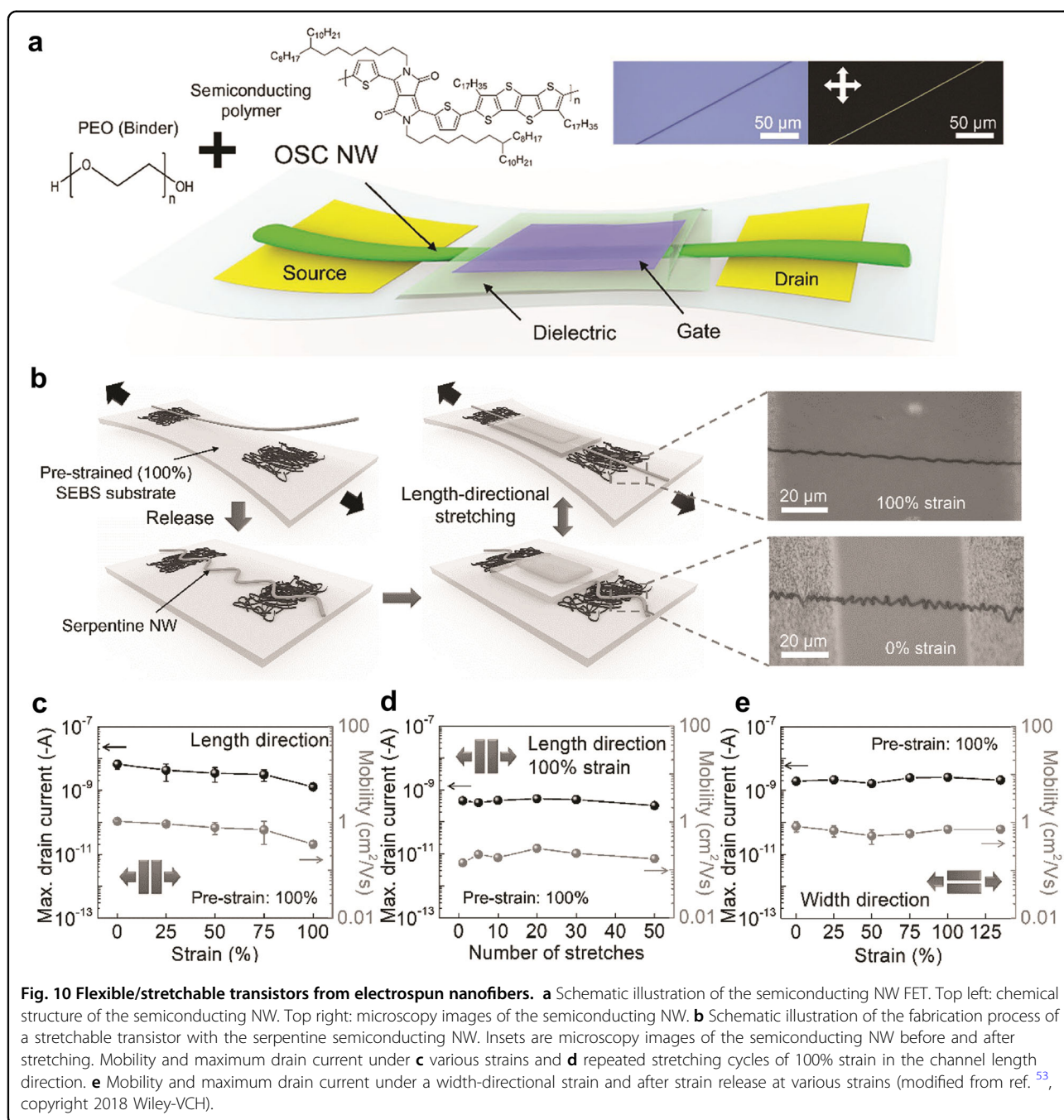


a fully stretchable supercapacitor with an electrospun membrane made of PU nanofibers as a separator, buckled SWNT films as electrodes, and an organic electrolyte consisting of tetraethylammonium tetrafluoroborate in propylene carbonate¹⁸². The resultant supercapacitor exhibited full stretchability up to 31.5% strain and retained 94.6% capacitance even after 2521 cycles of dynamic stretching and release under a high strain rate of 4.46% strain/s (Fig. 9d–f).

In addition, porous entangled nanofibers can enhance the capacitive performance by loading a high content of active capacitive materials due to the high surface area of these materials^{184,185}. For example, an SWCNT-coated electrospun SBS nanofiber mat has been employed to fabricate a high-performance fully stretchable supercapacitor. The as-obtained supercapacitor delivers a volumetric capacitance of 15.2 F/cm³ at 0.021 A/cm³ and can maintain this performance under strains of up to 40%¹⁸⁵. Specifically, an all-textile-based flexible supercapacitor has been fabricated by using electrospun PEDOT nanofibers as electrodes, a PAN electrospun nanofibrous membrane as a separator, and carbon fabric cloths as current collectors¹⁸⁶. The as-prepared supercapacitor is totally flexible and demonstrates high cyclability under ambient conditions.

Transistors

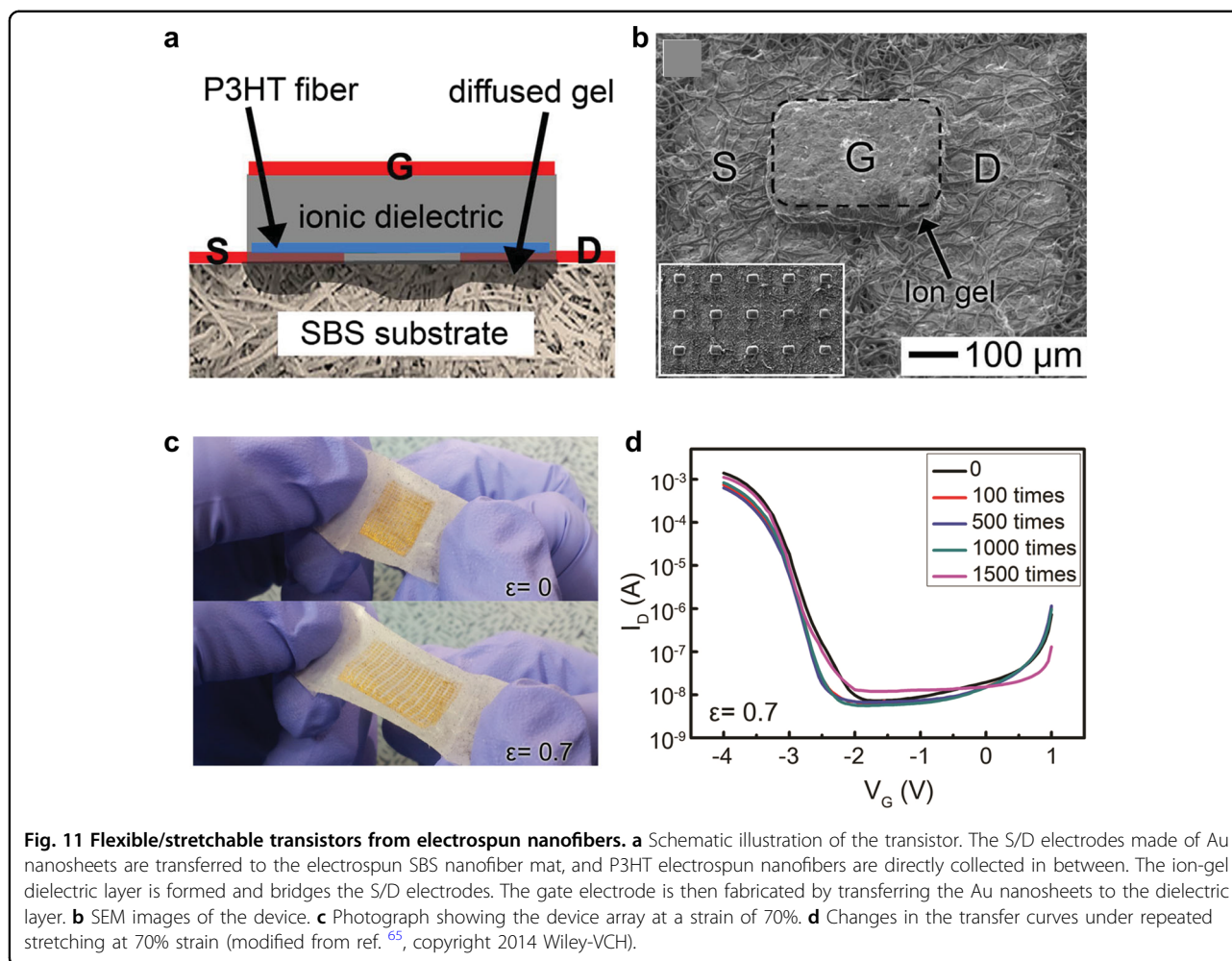
A field-effect transistor (FET) is generally regarded as an electrical switch that uses an external electric field to control the charge transport behaviors in a device. It consists of three electrodes (source, drain, and gate); a semiconductor, i.e., an active channel; and a dielectric utilized as a thin insulating layer between the channel and gate electrode^{187,188}. When a voltage is applied at the gate electrode, charge carriers move from the source to the drain to generate a source-drain current. Therefore, by adjusting the gate voltage, the source-drain current can be well controlled¹⁸⁹. Flexible/stretchable transistors are of high importance in soft electronic integrations, and are capable of effectively manipulating and processing electrical signals^{190,191}. However, polymer semiconductors made from film-type materials or nanofibril composites usually suffer from degraded charge carrier transport and limited flexibility. These issues can be overcome by the application of electrospun organic semiconducting nanofibers, where charge carrier transport can be enhanced by the preferential orientation of the polymer chains along the longitudinal direction of the fibers. It has been reported that the charge carrier mobility of P3HT electrospun nanofibers is dramatically improved by over 3 orders of magnitude¹⁹². In addition, electrospun



semiconductors can avoid complex lithography-based patterning procedures by being directly aligned on polymeric substrates.

For instance, a single electrospun semiconducting NW with a diameter of 675 ± 40 nm has been used as an active channel, which is composed of a fused thiophene diketopyrrolopyrrole-based polymer semiconductor and high-molecular-weight polyethylene oxide as both the molecular binder and deformability enhancer (Fig. 10a)⁵³. SBS elastomer films are applied as stretchable substrates

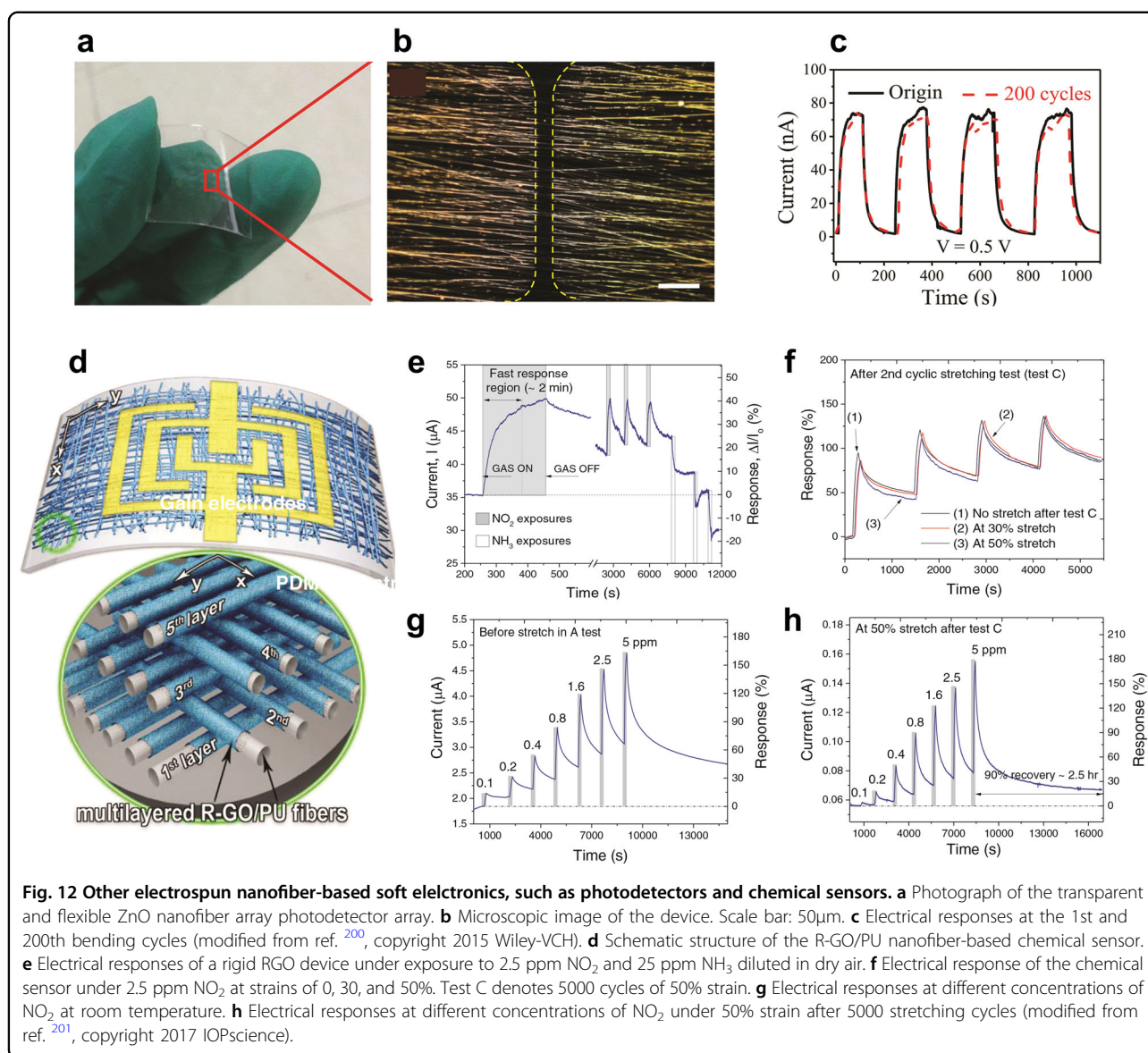
for the transistor. CNT percolating networks are transferred to SBS films to fabricate S/D electrodes. Then, a straight semiconducting NW was transferred on top of the S/D electrodes. The transistor fabrication is completed by subsequently transferring nonionic SEBS dielectric and eutectic gallium-indium gate electrodes on the NW. The original mobility and maximum drain current of the device are 0.78 ± 0.21 cm²/Vs and -6.7 ± 1.88 nA at a gate/drain voltage of -100 V, respectively. When stretching along the channel length structure to



100% strain, the elongated NWs become wrinkled after release. Thus, both the mobility and drain current vary by <20% during cyclic stretching. In contrast, along the perpendicular direction, both the mobility and drain current remain almost the same up to a strain of 100%. The reason is that NWs do not actually experience stress but wrinkle in the channel length direction. By applying prestrain to the NWs to obtain a serpentine geometry, the mechanical durability of the transistor can be largely enhanced (Fig. 10b). The electrical performance can be easily maintained when stretched in both the length and width directions to $\geq 100\%$ strain (Fig. 10c–e). When attached to a rubber balloon, the deformable transistor can survive a three-dimensional volume change of 1735% with a constant current output.

In addition to single nanofibers^{50,51,54,193}, entangled or aligned electrospun semiconductor nanofibers are applicable as channel materials^{52,194}. An entirely stretchable transistor has been developed by using electrospun SBS nanofibers as the substrate, P3HT nanofibers as the active channel, an ion-gel dielectric layer, S/D electrodes,

and gate materials made of Au-nanosheet networks (Fig. 11a)⁶⁵. The SBS nanofiber mat has a thickness of $\sim 500\ \mu\text{m}$, and the average diameter of a nanofiber is $\sim 300\ \text{nm}$. Au nanosheets are first transferred to PDMS and then to SBS membranes to form S/D electrodes with a size of $100 \times 800\ \mu\text{m}$. The active channel is fabricated by directly collecting electrospun P3HT nanofiber (diameter: $2.5\ \mu\text{m}$) sheets between the S/D electrodes. The dielectric layer is also made from stretchable materials, namely, ion gels, and bridges the S/D electrodes. Finally, the Au-nanosheet network is transferred to the ion-gel dielectric layer to form a gate electrode (Fig. 11b). Figure 11c shows that a 15×17 transistor array can be stably operated up to 70% strain. The device mobility experiences a light decrease to $15.5\ \text{cm}^2/\text{Vs}$ after 500 cycles under 70% strain and recovers to $18\ \text{cm}^2/\text{Vs}$ after 1500 cycles of stretching and releasing under 70% strain (Fig. 11d). This decrease may be because of the change in position of P3HT fibers under repeated stretching. Notably, the on-off current and threshold voltage remain the same during the cycling test.



Other applications

Other applications, such as flexible/stretchable heaters¹⁹⁵, photodetectors¹⁹⁶, chemical sensors^{88,197}, optically encoded sensors¹⁹⁸, solar cells¹⁹⁹, and organic LEDs^{114,115}, have also been developed with electrospun nanofibers. For example, a fully transparent and flexible ultraviolet-visible photodetector has been fabricated using the photoelectric effect of electrospun ZnO/CdO (cadmium oxide) nanofibers²⁰⁰. ZnO/CdO nanofibers have been fabricated by fusing ZnO and CdO nanoparticles into individual nanofibers. These highly aligned ZnO/CdO nanofibers, i.e., channel materials, can be exposed to light and induce a change in conductivity. The hybrid photodetector shows a transparency of up to 95% in the spectral range of 400–800 nm and a good mechanical durability of 200 bending cycles, as shown in

Fig. 12a–c. The as-achieved fibrous membranes can be used as chemiresistors for wearable chemical sensing applications by incorporating CNTs or graphene on electrospun fibrous scaffolds^{197,201}. A stretchable and highly sensitive chemical sensor has been manufactured by utilizing multilayered PU nanofiber/RGO networks (Fig. 12d)²⁰¹. The conductance of the device increases upon exposure to NO₂ (oxidizing gas) and decreases upon exposure to NH₃ (reducing gas). Here, RGO is the p-type semiconductor, while NO₂ molecules are the electron acceptors and NH₃ molecules are the electron donors. The device exhibits a responsivity of ~40% toward a 2.5 ppm exposure of NO₂ (Fig. 12e). The current responses to NO₂ exposure show very small changes even after 5000 cyclic stretching/releasing tests at 50% strain (Fig. 12f–h)²⁰¹.

Summary and outlook

In this review, we first introduced the basic electrospinning technique for nanofiber fabrication. The advantageous electrical and mechanical properties of electrospun nanofibers include high transparency, good conductivity, a large surface area, diverse porous structures, excellent flexibility and/or stretchability, a one-dimensional nanostructure, and easy scale-up, all of which provide strong benefits in the development of high-performance flexible/stretchable electronics. Generally, there are six strategies for the design of soft electronic devices: (1) single semiconducting nanofibers, (2) entangled porous nanofiber sheets, (3) aligned nanofiber arrays, (4) yarn, (5) temporary templates, and (6) reinforcement. Both single nanofibers and assembled nanofibers have been proven to be superior candidates for use in soft electronic applications. We then presented recent advances in electrospun nanofiber-based soft electronic devices, such as flexible/stretchable conductors, transparent electrodes, strain sensors, pressure sensors, nanogenerators, supercapacitors, batteries, and transistors. Nevertheless, to approach cost-effective, daily life applications, there are several remaining challenges regarding nanofiber fabrication and assembly, device performance, and system integration.

- (1) It is difficult to electrospin organic conducting and semiconducting polymers, which are commonly used for conventional transistors, energy devices, and optoelectronics. Although some electrospun conducting (e.g., PPy and PANI) and functional (e.g., PEDOT: PSS and P3HT) nanofibers have been utilized for soft electronic devices, spinnable active polymers are still limited. The basic requirements for solution electrospinning are a sufficiently high molecular weight of the polymer and an appropriate solvent. The electrical conductivity of the polymer also influences the electrospinnability. Hence, advances in lowering electrospinning requirements and developing novel polymers and polymer blends are highly desired to achieve electrospinnable organic functional polymers for flexible/stretchable electronic applications.
- (2) It is still crucial to improve the flexibility and stretchability of electrospun nanofibers. Existing electrospun nanofiber-based soft electronics endow a certain degree of mechanical flexibility owing to the inherent flexibility of nanofibers. However, some electrospun nanomaterials, such as carbon nanofibers and metallic oxide nanofibers, have limited mechanical flexibility, resulting in inferior mechanical properties of the as-fabricated devices. It is necessary to precisely control the density, orientation, and porosity of the nanofibers in an effort to improve the flexibility/stretchability while

maintaining the electrical performance of the devices. In addition, different nanofiber assembly methods should be combined, and new device assembly techniques are needed to realize soft electronics with desirable flexibility/stretchability.

- (3) As fundamental breakthroughs in individual electrospun nanofiber-based soft electronic devices have already been achieved over the past 10 years, these successes will lead to particularly promising achievements when combining two or more of the as-developed devices. Due to tremendous advancements in the flexible/stretchable electronic field, it is highly necessary to develop integrated and portable soft electronics. An appropriate combination of sensors and energy harvesting and storage devices can achieve a fully flexible/stretchable system based on electrospun nanofibers. For instance, a textile format, self-powered integrated wearable system can be prepared by using as-fabricated yarn sensors⁸⁰, all-textile flexible supercapacitors¹⁸⁶, and all-nanofiber-based ultralight TENG¹⁵¹. We believe that the assembly of existing components into smart soft systems will bring new opportunities both in the market and in academic research.

Acknowledgements

This work was supported by Japan Science and Technology ACCEL (grant number JPMJMI17F1).

Conflict of interest

The authors declare that they have no conflict of interest.

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Received: 29 August 2020 Revised: 8 October 2020 Accepted: 13 October 2020.

Published online: 8 March 2021

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