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Stretchable electronics: functional materials, fabrication strategies and applications

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ABSTRACT

The primary developing trends in flexible and stretchable electronics involve the innovation of material synthesis, mechanical design, and fabrication strategies that employ soft substrates. The biggest challenge is that the entire electronic system must allow not only bending but also stretching. Therefore, stretchable conductors become a crucial construction unit for the connection of working circuits of various stretchable devices. Owing to the success of stretchable conductors, various stretchable electronic devices are fabricated with the help of multiple manufacturing strategies, including stretchable heaters, stretchable energy conversion and storage devices, stretchable transistors, sensors and artificial skin. The continuous development of stretchable electronics has led to the new functionality of transparency, and the fabrication of related materials, fabrication strategies and various applications of stretchable electronics. It focuses on the fundamental structural design, mechanisms, and tactics, as well as on challenges and opportunities in the manufacture of stretchable electronic devices on the fundamental structural devices and their various applications.



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1. Introduction

As a newly developed technology, flexible and stretchable electronics is emerging and achieving a great variety of applications, because its components can be compressed, twisted and conform to complex non-planar surfaces [1]. Currently, wearable electronics applications have positive effects on various aspects of daily life, leading to economic growth and the rapid development of stretchable electronic devices and related manufacturing technologies. Flexible, soft and stretchable forms of electronic devices enable next-generation wearable electronic

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Figure 1. Various applications of stretchable electronics.

applications, opening up various applications for healthcare, energy and military purposes.

The interest in and fabrication strategies for stretchable electronics have been driven by the huge demand for wearable, intelligent, and integrated electronics systems during the past decade (Figure 1). Therefore, strenuous efforts have been made to realize the stretchability of electronic devices and reduce the total fabrication cost via sophisticated material design and novel device configuration. Moreover, devices that exploit bio-inspired designs or require intimate integration with the human body demand curvilinear shapes and/or elastic responses to large strain deformations [2,3]. However, mechanical compliance is critical in the manufacture of stretchable electronic devices, and devices should not incur physical damage or change their performance under the bending or stretching states. A number of important discoveries and breakthroughs in the fundamentals of this technology and fabrication strategies to face these challenges and achieve the above-mentioned targets are summarized in this review.

Various strategies are used to make the electronic devices stretchable, including directly utilizing the intrinsically stretchable materials [4] or constructing structure design strategies [5], including wavy structure configuration, island-interconnect configuration, mesh structure, fractal design approach, origami and kirigami structural configurations. We begin with a brief overview of these structure design strategies. However, stretchable conductors and electrodes are the fundamental building blocks for the final stretchable electronic devices, and hence the following section summarizes the advances in this field. The compliant, deformable and stretchable qualities of stretchable

conductors have updated the traditional ideas that people must use non-flexible silicon-based electronics and opened up a new direction of next-generation flexible electronics [6]. The understanding of nanoscale phenomena, functional materials, and soft devices has progressed to a point where substantial advances in stretchable electronics applications are realistic [7]. How to pattern the stretchable conductors is another challenge in the fabrication of stretchable electronic devices, especially using direct printing techniques [8]. Over the last several years, studies demonstrated the possibility of stretchable conductors, resulting in the development of more complicated and advanced stretchable electronic devices. The content that follows highlights the fabrication methods and application of stretchable electronics in energy conversion and storage, including solar cells, supercapacitors, lithium-ion batteries (LIB), Zn-based batteries and nanogenerators (NGs). Subsequently, we focus on how to fabricate the stretchable transistors, sensors and artificial skin. Furthermore, adding the transparent properties onto stretchable electronic devices will make it possible to obtain more applications, such as stretchable transparent electrodes (STEs) and transparent and stretchable electronic devices (TSEDs), where high levels of both optical transparency and stretchability are required for conformal placement of devices on the human body or any arbitrary surface [9]. This review highlights these advances, with an emphasis on underlying methods and engineering strategies in stretchable device construction. Furthermore, various applications of stretchable electronics are presented.

2. Manufacturing methods and mechanism

In general, stretchable systems are often achieved by engineered shapes and elastic substrates that are intrinsically stretchable, which offer a foundation for applications that exceed the field of common wafer and circuit board techniques because of their unique capability to combine with flexible materials and curvilinear surfaces. Various methods have been applied to manufacture stretchable systems, and the most common method is the use of intrinsically stretchable materials. Stretchable elastomers are often used as soft substrates in many electronic devices, such as natural rubber (NR), styrene butadiene rubber (SBR), ethylene-propylene-diene monomer (EPDM), polyurethane (PU), thermoplastic polyurethane (TPU) and predominant poly(dimethylsiloxane) (PDMS), etc., which can reversibly endure high deformations (>200%) [10]. However, this method often results in low electrical mobility and high electrical resistivity of electronic devices. Hence other

methods that involve wavy structural configuration, fractal design, and mesh, interconnected island, origami or kirigami structures were developed to make the entire system stretchable. These tactics may either be used to improve the tensile capacity of intrinsic stretchable conductors or enable the utilization of common conductive bulk metals in stretchable electronic devices.

2.1. Wavy structural configuration

Mechanical buckling method (MBM) is often used to realize the wavy structural configuration on the surface of elastomeric substrates, making the stiff thin-film layer flexible and stretchable (Figure 2). The wavelength (λ) and peak amplitude (A) are key factors for the deformation ability and electronic properties of the final devices. Because λ and A values can be determined, the stretchability and compressibility of the buckling systems can be calculated [11]. According to the energy method, when smaller applied strain the is than 5%, $\lambda_0 = 2 |\pi h(ar{E}_{film}/3ar{E}_{substrate})^{1/3}$, and $A = h(rac{arepsilon_{pre}}{arepsilon_{min}}-1)^{1/2}$, in which ε_{pre} is the prestrain, ε_{\min} is the required minimum strain to generate buckling, $\varepsilon_{\min} =$ $0.25(3\bar{E}_{substrate}/\bar{E}_{film})^{2/3}$, *h* is the thickness of stiff thinfilm layer, and \overline{E} is the plane-strain modulus. When large strain is used, the λ and A of a compliant substrate become nonlinear, and geometrical deformation and a nonlinear constitutive model for the substrate should be considered [12]. Generally, brittle semiconductor materials (such as Si, GaAs, InP, lead zirconate titanate) and metals are made into wavy shape through MBM to achieve stretchability [13–15].

Recently, wave structures have been developed and extended from the traditional MBM film to arched structures. For example, a highly stretchable LIB based on an arched structure could effectively accommodate large strain, but the areal capacity was only 0.11 mA h cm⁻². The arched electrode could bear a 400% strain and exhibited stable electrochemical performances after



Figure 2. Schematic of the MBM thin films on a compliant substrate, forming wavy structures with different amplitudes and wavelengths.

500 cycling tests, which are used in the fabrication of stretchable LIBs, and the final LIBs could also bear a 400% strain. The stretchability is nearly three times the maximal value of the stretchable devices based on the previous wavy methods [16]. As shown in Figure 3, a simple method is used to fabricate fully stretchable LIBs based on a wavy shape at the battery device level, where all the components (include cathode, anode, separator and current collectors) and packaging were stretched equally for the first time. These stretchable LIBs showed a stretchability of 50%, and a high areal capacity of 3.6 mA h cm⁻² [17]. The wavy structural stretchable device concept can be extended to new classes of flexible devices, representing promising directions for future research.

2.2. Island-interconnect configuration and mesh structure

Another common and effective strategy for the manufacture of stretchable electronics is to develop islandinterconnect configurations and mesh structures for enhancing the stretchability of functional devices. In this structural design, islands (fabricated from rigid materials) are interconnected by metal interconnections or other flexible bridges to achieve large and reversible deformation for strains applied on certain axes (Figure 4). These stretchable interconnects can be prepared by utilizing highly malleable/compliant electronic materials, such as low-temperature liquid metals [18–20], or by designing the interconnections to mitigate local strains through out-of-plane deformations (e.g. buckled devices [21], pop-up interconnections [22], and serpentine-shaped interconnection configurations [23]). The shortcoming of out-of-plane structure is that it is difficult to implement in a rapid manufacturing process, such as roll-to-roll and printing [24]. Therefore, an alternative method for the fabrication of stretchable interconnects is to use in-plane geometries for the conductors, e.g. stretchable line or zigzag, pulse, and horseshoe geometries. Recently, printed stretchable spiral interconnects have been fabricated by using reactiveink chemistry; the printed 2D geometries could be changed as free-standing stretchable interconnects [25].

Moreover, the stretchability could be effectively enhanced by the rational design of stretchable interconnects. For example, the stretchability of two serpentine-based and one spiral-based interconnects are comparative studied under the limitations of the same in-plane areas and contour lengths rooted from the same areal coverage and electrical resistance. The results revealed that the entanglement within a spiral helps to avoid elevation above the substrate; the spiral-based interconnects are much more stretchable than serpentinebased interconnects and can be stretchable up to 250% under elastic deformation and 325% without



Figure 3. (a) Schematic of a stretchable LIB based on device-scaled wavy structure and elastic-sticky separator. (b) Electrochemical performance of the wavy battery in the released and stretchable states at 50% strain. (c) Cycling performance and Coulombic efficiency for the wavy battery under releasing and stretching states (50% strain) (Reproduced with permission [17]).



Figure 4. Illustration of the stretchable system concept using rigid islands and stretchable interconnects/substrate.

failure [26]. The serpentine design of interconnects has been widely used for a variety of stretchable electronic systems, including epidermal electronics, skin-like temperature sensor and heater, stretchable battery [27–29], because serpentine structure design can enhance the capability to stretch interconnects in electronic systems [30]. Further stretchability enhancement of the serpentine structures can be realized by allowing the structure to deform and buckle in the out-of-plane directions.

Based on the interconnect island mechanism, a good mesh structure can achieve a large and reversible level of stretchability, and the stretchability is often larger than 100% [31]. For instance, materials and mechanical mesh design strategies offer extremely high stretchability for classes of electronic circuits, allowing accommodation of even demanding configurations such as corkscrew twists with tight pitch (e.g. 90° in ≈1 cm) and linear stretching to 'rubber-band' levels of strain (e.g. up to ≈140%) [32]. In the future, progress will be made in mesh design, and materials using mesh and associated fabrication technologies will enable stretchable electronic devices with unconventional formats, with useful functions.



Figure 5. Conventional fractal-inspired layouts.

2.3. Fractal design of stretchable interconnects

The utilization of fractal-inspired structural design is a very important method to realize the conductive interconnect configurations, which could simultaneous achieve higher stretchability and large-area characteristics for stretchable electronic applications. Moreover, this structure often undergoes intricate nonlinear deformations under external stress, because of the highly complex and diverse microstructures inherent in high-order fractal patterns [33,34]. Generally, fractal designs could transform inextensible sheets into highly tensile and super-conformable materials with various conceivable shapes and patterns via introducing a series of simplex cuts in a multilevel hierarchy with different motifs, as shown in Figure 5 [35]. The simple transformation hierarchical cuts and motifs would cause the changing of macroscopic shapes and final stretching performances [36]. Therefore, the innate character of the fractal design is to introduce cuts into a flexible substrate, dividing the material into rotating units prescribed by their cutting patterns and motifs.

The rigid thin films with fractal motifs are often combined to a stretchable substrate, enabling unusual mechanics with a large range of implications in stretchable devices. For example, Peano, Greek cross, Vicsek and other fractal designs are often introduced and used to fabricate space-filling structure of hard electronic materials (such as sensors, actuators, and radio frequency antennas), and their function of different fractal constructs are determined [37]. The geometries of the fractal designs play a critical role in its final stretchability performance.

Fractal-inspired layout designs trigger many new applications for stretchable electronics, such as biomedical systems and artificial skin. For instance, advanced material and fractal design concepts were used to construct a 3D conformal electronic platform, which was used in cardiac electrotherapies, and its device geometry and strain distribution in the undeformed state, with 20% uniaxial stretching and with 15% biaxial stretching, respectively [38]. Ultrathin monocrystalline Si nanoribbons with fractal motifs are used to fabricate strain, pressure and temperature sensor arrays, after integrating with humidity sensors, electroresistive heaters and stretchable multi-electrode arrays, resulting in the emergence of artificial electronic skin, which could be used for nerve stimulation and operated at ~20% strain [39].

2.4. Origami and kirigami structural configurations

Origami is an ancient art of paper folding, in which the key is to form strategically designed creases, and hence origami structural electronic devices fit the requirement of foldable electronics. Foldable electronics is a key technology for next-generation portable displays and wearable electronics. The principle of the origami method is that when the paper or other soft materials are folded, their mechanical properties depend on the folded shapes and patterns. Researchers can therefore use different fold patterns to achieve the stretchable properties that they need. For instance, the principle of origami designs are studied and used to fold reprogrammable mechanical metamaterials [40].

The conventional methods for preparing origami electronics is the fabrication of devices directly on origami substrates [41,42]. Origami structures are often used to manufacture foldable conductors [43],

stretchable LIBs, and supercapacitors. For example, foldable antenna could be fabricated by mask printing of Ag nanowire pastes on paper substrates [44]. The origami LIBs are prepared by slurry coating of electrodes onto paper current collector, and then packaging in standard materials, followed by folding using the Miura patterns. The final LIBs exhibited outstanding linear deformability (1340%) and areal deformability (1670%), large twistability and bendability [45]. An alternative approach yielded deformable silicon solar cells through origami design [46]. The recent boom in miniaturized electronics requires more flexible, stretchable, and higher-performance portable energy storage systems. Therefore, the growth of energy storage devices with innovative origami structures has accelerated in the field of flexible and stretchable electronics. Recently, a controlled, compressive buckling of a 2D origami precursor was exploited to achieve the dimensional changing in underlying elastomer supports. This novel approach offers broad versatility in the selection of materials (from polymers to device grade semiconductors), feature sizes (from cm to nm scale), topological forms (open frameworks to closed form polyhedra) and shape controllability (dynamic tuning of shape), thereby opening up a promising avenue to autonomic assembly of complex 3D systems [47].

Kirigami is essentially a variation of origami, and is a combination of folding and cutting. Therefore, kirigami can transform inextensible substrates into highly tensile ones, like mechanical springs, just by adding parallel cuts, dividing it into an array of thin strips with short cross connections [48]. Kirigami structure demonstrates an unusual mechanical response and appears very promising for a wide range of engineering applications. In addition, the unique properties of kirigami structures offer a broad range of innovative technical solutions for stretchable electronic and optoelectronic devices, among other application possibilities. Indeed, kirigami structures can significantly improve the stretchability of materials [49-52]. Compared with the initial soft sheet without line cuts, the kirigami sheet presents an out-of-plane mechanical deformation, and the sheet is highly stretchable [53]. For example, a network of kirigami structural notches in rigid nanocomposites and other inextensible sheets was prepared by top-down patterning technologies, which avoided the occurring of uncertain local failures and increased the ultimate strain of the sheets from 4 to 370% [54]. In addition, kirigami patterns were used in the fabrication of LIBs by standardized battery fabrication procedures, and significantly enhanced the stretchability (over 150%) [55]. Another traditional kirigami structure was introduced into the manufacture of stretchable triboelectric NGs and showed promising prospects in both power generation and self-powered sensor applications [56]. A simple kirigami patterning approach enables the

Table 1. The pros and cons of existing fabrication techniques.

| Methods | Merits | Limitations |
|----------------|---|---|
| Drop casting | Very simple | • Limitations in large area coverage |
| | No waste of material | Thickness hard to control |
| | | Poor uniformity |
| Spin coating | Good uniformity and reproducibility | Waste of material |
| | Good control on thickness | No large area |
| | | Film dries fast |
| Doctor blading | Large area | Micrometric precision of blade regulation |
| | No waste of material | |
| | Good uniformity | |
| Dip coating | Quite good uniformity | Waste of material |
| | Very thin layers | Time consuming |
| | Large area coverage | Double side coverage |
| Spray coating | Adjustable layer thickness | Homogeneity of the film |
| 1 / 5 | Large area coverage | 5 , |
| | Independence on substrate topology | |
| | | |

heater to be extremely stretchable (>400%) while stably retaining its excellent performance [57]. It is necessary for kirigami design and structures to be scaled down to the micro- and nanoscale for a broad scope of potential applications in energy storage devices, foldable and bendable microrobots, and optoelectronic devices.

Based on the above mechanism, various fabrication techniques have been developed, including direct printing methods, solution coating, and deposition. Printing is a powerful technique to enable the production of large-scale, low-cost electronic devices and systems, and includes contact printing (such as screen printing and gravure printing) and non-contact printing (such as inkjet printing, aerosol jet printing and electrohydrodynamic printing) [8]. For instance, a flexible and stretchable circuit has been fabricated by printing an Ag NWs/PDMS composite. Randomly oriented Ag nanowires (NWs) were buried in PDMS to form a conductive and stretchable electrode [58]. The resolution of contact printing is typically limited to the development of microscale electronic devices, and the resolution of non-contact printing is mainly limited by the size of the printer nozzle. For the non-contact printing method, avoiding the risk of nozzle clogging and maintaining the structural integrity of the printable functional materials during the printing process are the main challenges.

Solution coating and deposition methods include drop casting, spin coating, doctor blading, dip coating, and spray coating. Drop casting involves placing a droplet onto the substrate and waiting for the solvent to evaporate; the obtained film thickness depends on the concentration of solution. It is neither repeatable nor produces uniform films, it is limited to small samples and it takes a lot longer. Spin coating/ casting is highly repeatable and produces fairly uniform films over large areas. In doctor blading, a constant relative movement is established between the blade and the substrate; the paste or inks are spread on the substrate to form a thin sheet which results in a gel-layer upon drying. In the dip coating, the substrate is dipped into the solution and then withdrawn at a controlled speed. Thickness is determined by the balance of forces at the liquid-substrate interface. In the spray coating method, the substrate is hit by a vaporized solution flux. Spraying solution through printing techniques like electrohydrodynamic atomization can produce a uniform thin film. These methods have all been applied to prepare stretchable electronic devices, and the advantages and disadvantages are given in Table 1.

3. Stretchable conductors and electrodes

Conductors and electrodes are materials that allow the flow of an electrical current in one or more directions, and are the basic element of all electronic devices. Therefore, a wide range of soft and stretchable conductive materials could be made either by making electrical conductors more stretchable or by adding to intrinsic stretchable materials as conductive fillers. In this section, the conventional stretchable conductors, stretchable printed tracks and stretchable heaters are discussed.

3.1. Stretchable conductors

There are a number of techniques for producing stretchable conductors. Existing stretchable conductors include electronic conductors, e.g. metal nanoparticles (NPs), Ag NWs [59], Ag flakes, fractal Ag nanostructures [60], Cu NWs [61], carbon nanotubes (CNTs) [62,63], graphene [64], serpentine-shaped metallic wires, conductive polymers [65] and their composites [66]. These conductive components are often used as fillers and arranged in the elastomer matrix while combining the structure design. For instance, we fabricated a flexible and stretchable conductor by embedding fractal-structured Ag particles into a PDMS substrate, which could stretch up to 100% [67]. Two stretchable conductors of PU containing Au NPs are prepared by a layer-by-layer assembly method and a vacuum-assisted flocculation method, respectively. High conductivity and stretchability are

observed in both composites [68]. Liquid metal-based interconnects embedded into flexible polymer substrates are used as stretchable and highly conductive interconnects. The fabrication process is very simple and the as-prepared active devices showed a high fill factor [22,69,70]. Ordered zigzag structures are used to prepare stretchable conductive tracks; their high stretchability comes from the synergistic effect of interpenetrating conductive networks of polymer gel and Ag NPs, and the zigzag structures [71].

As one of the most important conductive materials, Ag NWs have recently attracted a lot of attention for fabrication of stretchable conductors. Zhu's group carried out a pioneering work in this field [72]. Ag NWs are drop-casted onto the surface of pre-cleaned substrates, then the liquid PDMS or other elastomers are casted on top of the Ag NWs film, and finally the PDMS or other elastomers are when it is cured [48]. Moreover, Ag NWs are widely used as a thin film with percolation network embedded in elastic substrates. For instance, a photolithography process enables the fabrication of complex 3D interconnected patterns of Ag NWs networks embedded in PDMS, achieving stretchable microelectrodes with tailored electrical properties, low sheet resistances (down to 0.6 Ω / sq), controllable gauge factors (ranging from 0.01 to 100), and good stretchability (above 50% uniaxial strain) [73]. The aspect ratios of Ag NWs play a critical role in the final electronic performance of flexible or stretchable electrodes [74]. For instance, a highly stretchable metal electrode were fabricated by the solution-process method. The Ag NWs ware very long (>100 µm), and hence the low-temperature sintering process ensured the formation of conductive networks. The resulting electrode simultaneously exhibited a high electrical conductivity (~9 Ω/sq) and mechanical compliance (strain > 460%) (as shown in Figure 6) [75]. Microscale structural design strategy is another effective method to improve the performance of Ag NWsbased stretchable electrodes, such as binary network structure design [76], constructing highly stretchable conductive fiber [77], etc.

CNT is another conventional nanomaterial to fabricate stretchable conductors by using tens of μ m- to mm-long bundles and ropes of singlewalled CNTs (SWCNTs) and multi-walled CNTs (MWCNTs) [78]. Similar to the Ag NWs, CNTs are often used as filler in the conductors; for example, SWCNTs were uniformly dispersed in a fluorinated rubber to construct elastic conductors [79], and MWCNTs were homogeneously dispersed in the poly[styrene-b-(ethylene-co-butylene)-b-styrene] triblock copolymers (SEBS) matrix to achieve superior stretchable conductors with a stretchability of more than 600% [80]. However, with a few exceptions, CNT-based stretchable conductors provide modest conductive performance, require high concentrations of CNTs, are opaque, and their electrical conductivity declines significantly under stretching state [81]. Multiple strategies are developed to realize CNTs-based stretchable conductors, such as wavy ribbons of CNTs [82], buckling of aligned CNTs [83], and well-aligned CNT ribbons embedded in PDMS [84].

Additionally, carbon materials and metal nanomaterials are also formed as hybrid composites to fabricate stretchable conductors [85]. For instance, the resistance of hybrid Ag flakes/MWCNTs composite-based stretchable conductors is 5710 S cm⁻¹ at the beginning and 20 S cm⁻¹ at 140% strain [86]. The normalized resistance (R/R_0) of Ag NWs/graphene composites is nearly invariable under 10% stretching, and then gradually increases to 1.5 under 20% stretching, to 2.5 under 40% stretching [87]. In addition, CNTs and graphene may be composited and used in the fabrication of stretchable conductors [88,89].

Obviously, considerable time and efforts have been devoted to developing simple, scalable and low-cost approaches for manufacturing of stretchable conductors or electrodes. Methods to achieve superior conductivity, stretchability, and compatibility with existing preparing/patterning techniques are very important emerging issues in this field. Stretchable conductors will become invaluable in stretchable and wearable electronics, artificial skin, and stretchable energy conversion and storage.

3.2. Stretchable printed tracks

As a simple and cost-effective approach, printing techniques could enhance current methods of constructing a patterned surface for nanomaterials and offer opportunities for developing all-printed stretchable circuits and electronic devices, especially offering the possibility of ubiquitous lowcost and flexible devices [90-92]. The patterning of electrically conductive materials on stretchable elastic substrates can used to create innovative electronic devices with extraordinary mechanical properties, allowing bending, folding, stretching, or conforming applied environments. to their Various printable functional materials have been developed and optimized for traditional printing methods to achieve stretchable patterning, including screen printing, inkjet printing, and roll-to-roll (R2R) printing [93].

In the last decade, studies on elevating the stretchability of conductive lines on elastic substrates have made remarkable progress, but designing and printing patterns on stretchable substrates directly remain a big challenge. For example, PDMS is a hydrophobic and chemically inert material. Printing inks do not



Figure 6. Normalized resistance as a function of strain, photographs and schematics of macroscopic and microscopic surface morphology of an electrode on a pre-strained Ecoflex during stretching process (Reproduced with permission [75]).

adhere well on PDMS without pretreatments, and therefore it is difficult to print conductive tracks directly on PDMS [94]. Some tactics have been developed and used to enhance the bond strength between conductive inks and elastomer surfaces, including spraying with several coats of primer or printing a glue layer, changing the binders in functional inks and surface modification [95-97]. Recently, a plasma treatment associated with a printing approach was used to prepare deformable conductive patterns or devices directly on the surface of PDMS [98]. The obtained printed tracks indicated strong mechanical stability and exhibited a high electrical conductivity under bending, twisting, and stretching conditions [99]. As an alternative strategy, Ag-PDMS tracks are fabricated onto the surface of PDMS substrate by screen printing, which could be stretched to 20% over 1000 cycles and the bonding is stable without any mechanical failure [100]. As shown in Figure 7(a), different stretchable electrodes, contact pads, and interconnect patterns are screen

printed onto Ecoflex substrates, which are used to fabricate the all-printed stretchable electrochemical devices (which can withstand severe tensile strains, as high as 100%, without any significant effects on their performance) [101]. As shown in Figure 7(b), low-cost and scalable screen printing technology is used to deposit Ag/AgCl conductive inks onto the surface of TPU film. Its heat lamination onto knitted fabrics and subsequently protective packaging layers yield tensile interconnections, which demonstrated stretchability of over 100% strain and fatigue life of 1000 cycles at 20% strain [102]. In addition, researchers found that both Ag NPs and Ag NWs could serve as metallic inks to achieve the patterning of conductors for stretchable electronics. However, Ag NPs inks can spray through a stencil mask for large-scale patterns and rapid manufacture with limited resolution while other finer-resolution routes, such as stamping and ink-jet printing techniques, are not very suitable to Ag NWs inks [103]. Recently, research demonstrated water-based Ag NWs ink could be used to



Figure 7. (a) Schematic (top left) of screen-printing of a stretchable array of electrochemical sensors using tailored functional inks, and photographs showing different stretching states (Reproduced with permission [101]); (b) Stretchable structure used to connect an electrocardiography (ECG) sensor (top left), inner side of as-prepared ECG T-shirt and recorded ECG signals (Reproduced with permission [102]); (c) Chemical formulas of stretchable Ag flakes-based functional ink, printed elastic conductor and demonstration of the stretchability, and a comparison of this work with other conductive stretchable conductors (Reproduced with permission [105]).

fabricate stretchable conductors and wearable thinfilm transistors (TFTs) by screen printing [104]. As shown in Figure 7(c), the resistance of Ag flakesbased printable stretchable conductor is 738 S cm⁻¹ at 0% strain and 182 S cm⁻¹ at 215% strain [105]. More recently, an electrohydrodynamic printing method has been used to directly print Ag NWs on a pre-strained PDMS; the resistance increased a little with the increasing number of cycles under 30% strain (i.e. 10.6% increase in 50 cycles) [106]. These results demonstrate that Ag is an important ink material for achieving stretchable printed tracks and its morphology plays a very important factor for improving the stretchability.

Besides direct printing, other printing-related techniques or methods are also used to realize the patterning of stretchable tracks. For example, conventional printed circuit board (PCB) production technologies combined with stretchable circuit designs (especially meander-shaped conductors) are also used to manufacture stretchable tracks [107–109]. For stretchable circuits fabricated by PCB technology, the stretchability comes from the shaped structure of Cu track (horseshoe-shaped meanders). The functional conductive components are embedded into PDMS and utilized as a circuit carrier [110]. Based on this technique, stretchable PCB (SCB) and stretchable mold interconnects (SMI) can be fabricated [111]. In addition, direct ink writing techniques offer competitive alternatives to meet the demanding of design rules and form factors required for flexible conductors in printed electronics and optoelectronics. This method is also used to fabricate stretchable conductors. For example, flexible, stretchable, and spanning Ag microelectrodes were fabricated by the direct ink writing method with the use of tailored Ag nanoparticle inks [112]. Writable particle-free inks are being developed for fast preparation of stretchable circuits with high conductivity. These functional inks are based on soluble Ag salts and adhesive polymers [113]. This direct ink writing method can be used to fabricate stretchable electronic devices [114,115].

3.3. Stretchable heaters

Electrically driven resistive heaters possess a wide range of applications, such as temperature maintenance in industrial process, defogging in cold climates, thermal therapy and even painting conservation. In essence, stretchable heaters are an extension and expansion of stretchable conductors. In flexible heaters, the temperatures are often controlled by the joule heating of electrodes and resistors. Stretchable heaters are expected to be particularly valuable for personal thermal management and healthcare supports, but their implementations bring extra needs that are very challenging to satisfy all at once. To date, there are several studies reporting on functional materials and designs of flexible and stretchable electrodes that could be employed as thermal units based on the joule heating, as shown in Table 2.

These examples include stretchable metal NWs electrodes, ultralong percolated CNTs embedded in elastomers, metal mesh, and electrospun rubber fibers with percolation of metal NPs. Among various materials, metal NWs play a key role in the fabrication of stretchable heaters [135]. For example, as shown in Figure 8(a), highly stretchable and transparent electrical heaters are prepared via constructing a partially embedded Ag NWs percolating network on a tensile elastomer [136]. As shown in Figure 8(b), the conformal lamination of flexible and stretchable heaters is prepared via constructing serpentine-mesh patterns. The SBS/Ag NWs-SBS/SBS sandwich structures can achieve effective heat transfer even during motion, and can operate at relatively lower input voltage because the Ag NWs/SBS mesh possesses a high conductivity [137]. Besides the metal nanomaterials, carbon nanomaterials and conductive polymers are also used in the fabrication of stretchable heaters. For example, a novel soft strip-shaped thermochromic resistive heater (STRH) is prepared via incorporating an aligned carbon nanotube sheet and a thermochromic silicone elastomer. The resulting textiles woven from the STRHs are flexible, stretchable, and breathable, and they can stably work under various deformations such as twisting [138]. As shown in Figure 8(c), highly stretchable fabric heaters with low resistivity are successfully prepared by utilizing a conductive PEDOT:PSS polymer modified with sodium dodecyl sulfate (SDS), which showed reversible electrical behavior with cyclic loading of stretching strains > 80% [139]. Recently, the challenges in stretchable heaters are how to achieve uniform and stable temperature distributions over a large area, how to realize rapid heating and cooling, how to maintain long-term operation at low-enough input powers, and finally how to enable all the aforementioned are achieved while simultaneously maintaining stretchability. For instance, uniform and

stable heating of stretchable heaters was recently developed because the ligand exchange of Ag NWs allows the nanowires to be homogeneously dispersed in the elastomeric media. The joule heating characteristics remained stable even under applied strain of ~30% for 5000 cycles [140]. Moreover, Lee et al. attained uniform and stable temperature distribution of a CNT-based heater owing to the high degree of nanotube alignment. The surface temperature fluctuations were as small as 5 °C [141]. Meanwhile, non-toxicity, together with stable electrical and thermal properties under mechanical disturbance, is also required for a wearable heater.

4. Stretchable energy conversion and storage devices

The energy source plays a key role in the construction of independent electronic systems. Although several approaches are available for stretchable conductors and electrodes, an urgent technical problem is in soft power sources that have similar mechanical performance, enabling them to be used in conjunction with other stretchable electronic devices. Owing to a sustained requirement of compatible tensile power sources, especially for some emerging biomedical electronic devices, stretchable energy conversion and storage systems have attracted increasing attention in the past few years [142]. To realize energy storage systems with stretchable properties, two strategies are often used: one is employing the intrinsic stretchable functional materials directly, and the other is utilizing the aforementioned structural designs that are able to operate under mechanical strain. Despite recent progress in stretchable electrodes, fully stretching energy conversion and storage devices consisting of electrodes, separator, and sealing material, remain a great challenge. There have been significant research efforts in stretchable energy conversion and storage devices, including stretchable solar cells, stretchable supercapacitors, stretchable LIBs and other batteries, and stretchable nanogenerators.

4.1. Stretchable solar cells

Stretchable solar cells, especially thin film solar cells (TFSCs) and dye-sensitized solar cells (DSSCs), are attracting increasing interest as wearable power sources, which are important for portable electronic devices. However, the complicated manufactureing process, high cost and lower stretching properties limited their potential applications. Much work has been done on the development of structural engineering to obtain stretchable inorganic crystalline solar cells, e.g. wavy, serpentine, stiff island, and cross-buckled structures [143]. A competitive method to achieve stretchability is the origami strategy: the

| ethylenedioxythiophene | e) polystyrene sulfonate, PL | JA for polyure | ethane acrylate, FC for | fluorocarbon, | ITO for indiu | m tin oxic | le, FTO for fluorine-doped tin oxide, | CPI for colorle | ss polyimide, PEN fo | r poly |
|--|------------------------------|----------------|-------------------------|---------------|---------------|------------|--|------------------------------------|----------------------|--------|
| (ethylene naphthalate), | WPU for waterborne poly | urethane, an | d rGO for reduced gra | phene oxide). | | | | | | |
| Materials | Method | Substrate | Area (mm²) | Voltage (V) | Temp. (°C) | t (s) | Flexible/stretchable stability | $R_{\rm S}~(\Omega~{\rm sq}^{-1})$ | Application | Ref. |
| Ag NWs | Vacuum filtration | PDMS | 10×15 | 10 | 150 | 60 | 50% (5 cycles) | 30 | Human wearable | [136] |
| CNT | Paving | Textile | 10×13 | 8 | 95 | 100 | 100% (500 cycles) | Z | Human wearable | [138] |
| Ag NWs | Template | SBS | 10×13 | - | 40 | 60 | 30% (5000 cycles) | 0.02~0.07 | Thermotherapy | [137] |
| Ag NWs | Spin coating | PVA | 10×10 | 5 | 74 | 20 | 1 mm (10,000 cycles) | 20 | Thermotherapy | [116] |
| PEDOT:PSS | Spin coating | PET | 25×25 | 12 | 140 | 140 | 10 mm (10 cm $	imes$ 10 cm 500 cycles) | 57 | Defrost | [117] |
| CuNi micromesh | Transfer printing | PUA | 23×19 | 6 | 225 | 60 | 4 mm (1000 cycles) | 16.2 | Defrost | [118] |
| Pt mesh | Electron-beam evaporator | PET | 7 × 11 | 5 | 86 | 15 | 3.8 mm (1000 cycles) | 69.1 | Deice | [119] |
| CuZr nanothrough | Transfer printing | PDMS | 20×20 | 7 | 180 | 70 | 50% (15,000 cycles) | 3.8 | Defog | [120] |
| Ag nanofibers | Electrospinning | PI/PET | 50×65 | 4.5 | 249.5 | <10 | 70 μm (10,000 cycles) | 0.5 | Human wearable | [140] |
| PS@Ag | Screen printing | PDMS | 20×40 | m | ~40 | | 30% (30 cycle) | | | [121] |
| FC/Ag/FC/ITO/SiO ₂ | R2R sputtering | PET | 100×100 | 10 | 110 | 90 | 3 mm (10,000 cycles) | 4.5 | Defrost | [122] |
| Au mesh | Spin coating | PET/glass | 0.7×0.7 | 1.5 | 100 | ٢ | | 10 | Thermotherapy | [123] |
| Ag NWS/clay | Rod coating | PET | 50×75 | 7 | ~110 | 70 | 10 mm (50 cycles) | 10 | Defrost | [124] |
| Ag NWs | Vacuum filtration | CPI | 10×10 | 2.2 | 75.7 | | 60° angle | | | [125] |
| GO/Ag NWs | Vacuum filtration | Quartz | 25×25 | 15 | 210 | 110 | | 27 | Defrost | [126] |
| Ag NWs | Spray | PEN | 25×25 | 7 | 55 | 100 | | 35 | Thermochromics | [127] |
| CNT/Ag NWs | R2R | PET/glass | 40×40 | 15 | 105 | 15 | 1.5 mm (1000 cycles) | 30 | Thermochromics | [128] |
| AI | Kirigami | Ecoflex | 10×30 | 1.2 | 43 | 60 | 300% (1000 cycles) | 0.075 | Thermotherapy | [129] |
| FTO/NICr/Ag NWs | Scattered | PET | 40×30 | 9 | 162 | 30 | 8 mm (10,000 cycles) | 9.9 | | [130] |
| Cu wire/Al ₂ O ₃ /Pl | Transfer printing | PDMS | 15×20 | 2 | 45 | 60 | 30% (100 cycles) | <10 | | [131] |
| WPU/PEDOT:PSS/rGO | Drop casting | | 10×20 | 5 | 100 | 180 | 30% (200 cycles) | 18.2 S cm ⁻¹ | Thermotherapy | [132] |
| Cu NWs | Spray | PU | 15×15 | 7 | 102 | >120 | 10% (1000 cycles) | 4.7 | Glove | [2/4] |
| Cu nanofibers | Transferring | Ecoflex | 5×25 | 2.5 | 80 | | 300% (1000 cycles) | 0.058 | | [133] |
| PE microfibers/Cu NWs | Dip coating | Ecoflex | L80 mm \times r0.5 mm | 6 | 150 | 20 | 100% | | Thermotherapy | [134] |

Table 2. Noteworthy examples of flexible and stretchable heaters (SBS stands for solid bleached sulfate, PVA for polyvinyl alcohol, PET for polyethylene terephthalate, PEDOT:PSS for poly(3,4-



Figure 8. (a) Temperature distributions by joule heating under different strain (Reproduced with permission [136]); (b) Stretchable heater consisting of SBS/Ag NWs-SBS/SBS sandwich structures and IR image after voltage application (Reproduced with permission [137]); (c) Joule heating behaviors as a function of different applied strains at an input voltage of 12 V, and photograph and IR images of the heater before and after bending fingers at an input voltage of 6 V (Reproduced with permission [139]).



Figure 9. (a) Photographs of origami-like silicon solar cells, folded and unfolded (Reproduced with permission [46]); (b) Schematic illustration of the fabrication process of stretchable organic solar cells (Reproduced with permission [150]).

origami-enabled silicon solar cells could reach 644% areal compactness while preserving high photovoltaic performance upon 40 times cyclic folding/unfolding (Figure 9(a)) [46]. In addition, the transfer printing technique is often used in manufacturing of stretchable inorganic crystalline solar cells. For example, stretchable inorganic amorphous TFSCs are prepared by using a transfer printing approach with a watersoluble germanium oxide sacrificial layer; the performance of the stretchable solar cell will be maintained even after stretching to 50% [144]. Novel structures are also designed and used to prepare stretchable inorganic crystalline solar cells. For example, stretchable DSSC textile is prepared from tensile and conductive fibers as counter electrodes and spring-like TiO₂ wire as the working electrodes. The energyconversion efficiencies of as-obtained DSSCs could reach 7.13%, which could be well preserved under stretching at 30% strain and after 20 cyclic stretching tests [145].

Although organic photovoltaics (OPVs) have a lower performance and poor environmental stability compared to that of inorganic solar cells, they are still good candidates for stretchable solar cells because of their mechanical compliance. Organic semiconductor-base solar cells are potentially inexpensive alternatives to the aforementioned stretchable device made of crystalline and polycrystalline silicon and other thin-film materials. The hope is to achieve more flexible, lightweight, and stretchable OPVs than with other technologies [146-149]. For example, Zhenan Bao et al. developed the first intrinsic tensile organic solar cells through depositing the used materials in conventional OPVs on the pre-stretched rubber substrates that they then allowed to compress (as shown in Figure 9(b)). The buckling waves in these organic thin film layers made them stretchable. The as-obtained solar cells could be well maintained under stretch lengthwise by up to 27% [150-152]. The stretchable polymer solar cells are fabricated by attaching the ultrathin polymer solar cell onto a prestretched elastomeric substrate and then releasing the prestrain to form random buckling, and the power conversion efficiency (PCE) could reach 5.8% under 70% tensile strain. The stretchable OPVs exhibit small fluctuations in performance after 400 stretching-releasing cycles [153]. Recent literature demonstrated the polymer solar cells were semitransparent and could be stretched like a rubbery film by as much as 100% strain. The measured PCE was 3.48%, which was increased to 3.67% after one cycle of stretching to 50% strain and decreased to 2.99% after 100 stretching cycles [154]. Because these soft organic thin film materials generally possess intrinsically deformability and flexibility, the deliberate selecting of functional materials on the basis of their mechanical properties (not just charge-transport properties) is still required for various applications with mechanically demanding form factors, such as those that exist in the field of wearable electronics [155]. Developing new methods or new materials in accordance with most studies on stretchable OPVs would contribute to improving the efficiency, cutting the cost, extending the lifetimes, and increasing the reliability of the devices.

Lifespan and durability are other important factors in developing stretchable solar cells. Many reports evaluated the durability of stretchable solar cells via cycling tests, but the lifespan of those solar cells was not provided and should be examined using a longterm accelerated aging experiment under accelerated soaking visible-light in successive research. Understanding the aging mechanism is very important. By modifying the current prevailing solar cell designs and the electrolyte, the lifetime of solar cells can be extended by ten times. For the stretchable solar cells, the long-term accelerated aging experiment should also combine the mechanic durability test. Elastic solar cells would be more resistant to fracture under tensile and bending strain than nonstretchable devices, and might increase the lifetime of OPV devices against mechanical failure [150]. Note that the lifespan of a modern solar panel is far longer than the 20 years that we use to calculate costs and earnings. It still remains to be seen whether it will be possible to extend the lifespan of stretchable solar cells from several hours or weeks to several tens of years.

4.2. Stretchable supercapacitors

Electrochemical capacitors have been widely studied in energy storage fields because of their higher specific capacitances and power densities. Moreover, supercapacitors exhibit many merits including rapid charging/ discharging abilities, higher safety and long cycling life, leading to much brighter prospects for various applications. However, compared with studies on flexible or bendable supercapacitors [90], it is more challenging to manufacture tensile ones since stretching typically induces much larger arbitrary deformations, including bendability, stretchability, compressibility, twistability and other capabilities [156].

To manufacture stretchable supercapacitors, sandwich structure is usually applied, in which the solidstate electrolyte and separator are assembled into the two electrodes. Two conventional strategies are developed to realize full stretchable supercapacitors. The first method relies on stretchable substrates (such as PDMS, Ecoflex, and PU) to provide stretchability [157–161]. For example, tensile crumpled-graphene paper was prepared and used as supercapacitor electrodes; the final supercapacitors exhibited a combination of high stretchability (e.g. linear strain ~300%, areal strain ~800%), highly specific capacitance of ~196 F g⁻¹, and



Figure 10. (a) Schematic illustration of the structure of supercapacitor, its electrode is the crumpled-graphene-papers, and the polymer electrolyte gel is used as electrolyte and separator. Also shown are the corresponding image, CV and galvanostatic charge/discharge curves of the final assembled device (Reproduced with permission [162]); (b) Photographs of the supercapacitor arrays integrated on the stretchable substrate, CV curves and normalized capacitance measured under different types of deformations (Reproduced with permission [167]); (c) Normalized electrical resistance versus tensile strain and electrical resistance variation of pristine and buckled CNT films subjected to various types of tensile loadings, and photos of buckled CNT film at various stretching states (Reproduced with permission [170]).

good stability (e.g. over 1000 stretching/relaxing cycles) (Figure 10(a)) [162]. We proposed a simple and efficient fully-printing approach to fabricate stretchable asymmetric supercapacitors by using Ag@PPy@MnO₂ cathode electrode (PPy = polypyrrole) and activated carbon anode electrode, which displayed an ultra-high energy density of 0.0337 mW h cm⁻² at a high power density of 0.38 mW cm^{-2} . Its capacitance retention reached 90.8% after 5000 cycles and 86.2% after 40% stretching strain [163]. In addition, the pre-stretched substrates are often used to fabricate stretchable supercapacitors for improving the stretchability [164]. Strenuous efforts have been made to develop carbon materials-based stretchable supercapacitors with different structure features and electrochemical performances, such as conventional planar/textile, wearable fiber-shaped, transparent, and solid-state devices onto stretchable polymer substrates with aesthetic appeal [165].

Another strategy is to deposit electrochemical active polymers such as PPy on the stretchable electrolytes [156]. For example, a highly stretchable H₃PO₄-PVA polymer electrolyte was synthesized with a low resistivity of 3.4×10^{-3} S cm⁻¹ and a high fracture strain at

410% elongation. Then a stretchable supercapacitor was fabricated and showed only a small capacitance loss of 5.6% at 30% strain, and could preserve 81% of the initial capacitance after 1000 cyclic stretching tests [166]. As shown in Figure 10(b), tensile supercapacitor arrays are fabricated by using a gel-type electrolyte of poly(methyl methacrylate)–propylene carbonate–lithium perchlorate. A dry-transferred supercapacitor array on a specially designed tensile elastomer displayed stable electrochemical performances under different types of deformations, including bending, twisting, both uniaxial and biaxial stretching up to 50%, and winding around the curvilinear substrates [167].

Recently, omnidirectional stretchable high-performance supercapacitors have become a new research focus in stretchable supercapacitor fields [168]. For example, omnidirectional stretchable, all-solid-state supercapacitor based on graphene–CNT layered structure are developed [169]. As shown in Figure 10(c), isotropic buckled acid treated CNT@polyaniline (PANI) electrodes exhibited high specific capacitance of 1147.12 mF cm⁻² at 10 mV s⁻¹. The as-prepared supercapacitors could withstand the omnidirectional stretching up to 200% strain, which is twice the maximum strain of CNT-based bi-axially tensile supercapacitor reported in previous literature [170].

Existing stretchable supercapacitors are often assumed to have complicated manufacturing procedures, expensive and lower stretchability features, restricting their potential applications. Next generational stretchable supercapacitor systems should offer significant improvement over existing methodologies for stretchable electronics in terms of stability, multimode deformation, and density of capacitance.

4.3. Stretchable batteries

4.3.1. Stretchable LIBs

Rapid development and substantial achievements have been shaping the field of wearable electronic devices, resulting in the requirement for stretchable and flexible LIBs. Stretchable and flexible LIBs are generally LIBs that can operate within the normal elastic range. When the external forces are unloaded, it can completely restore its initial state without any remaining plastic deformations. Various strategies are used to fabricate stretchable LIBs, such as the aforementioned wave structure configuration [17], and kirigami structure configuration [55]. As shown in Figure 11, unusual 'self-similar' interconnect structures are designed and used in rechargeable LIBs, which can withstand reversible tensile strains, as high as 300%, while preserving capacity densities of ~ 1.1 mAh cm⁻², and very low loss in capacity for up to 20 cyclic recharges [28]. The Cui group developed stretchable LIBs by using stretchable Li₄Ti₅O₁₂ anodes and LiFePO₄ cathodes, which can sustain 80% stretching strain because of the utilizing of 3D interconnected porous PDMS sponge based on sugar cubes. Capacity retentions of 82% and 91% for anodes and cathodes are realized after 500 cyclic stretching/releasing tests [171]. Recently, other stretchable LIBs have been prepared by coupling LiMn₂O₄ (LMO)/CNT film (in situ growing of LMO nanocrystals inside 3D CNT film networks) with a wrinkled MnOx/ CNT film anode, which exhibited a high average specific capacity of ≈ 97 mA h g⁻¹ and significant stability after more than 300 cyclic stretching tests under 100% strain [172]. Sufficiently stretchable LIBs are still underdeveloped, which is one of the biggest challenges in realizing fully deformable power sources.

4.3.2. Stretchable Zn-based batteries

Zn-based batteries might be the most promising candidates for mass manufacturing by traditional highthroughput printing technologies due to their air stability, resulting in the prospect of stretchable batteries that integrate the inexpensive and simple 2D and 3D printing techniques in the future, which is a key advantage compared to LIBs for stretchable electronics applications. The mechanics of stretchable Zn-based batteries are still achieved by the aforementioned structural designing of hard and soft components. For example, a soft matter based rechargeable alkaline manganese battery consisted of elastomer, functional gel and textile mesh electrode, and sustained over 700 cyclic mechanical stretching/relaxing tests under 25% strain, with an average cell capacity of 6.5 mA h [173]. As shown in Figure 12(a), stretchable Zn-MnO2 alkaline cells based on off-the-shelf compliant Ag fabric embedded with MnO₂ and Zn particles are fabricated. The functional fabrics are used as current collectors and mechanical supports for the electrochemical-active particles. The embedded fabric electrodes with a PGE electrolyte were packaged inside an elastomeric pouch, and the cells preserved its capacity during stretching up to 100% strain [174]. As shown in Figure 12(b), a highly stretchable Zn-Ag₂O battery is prepared by incorporating polystyrene-block-polyisoprene-block-polystyrene (SIS) as a hyperelastic binder for custom-made printable inks. The remarkable mechanical properties of the SIS binder lead to an all-printed, stretchable Zn-Ag₂O rechargeable battery with a ≈ 2.5 mA h cm⁻² reversible capacity density even after multiple iterations of 100% stretching [175].

As a mature battery, Ag-Zn batteries show high performance and are a green solution to power stretchable devices, which are inherently safe because of the utilization of aqueous electrolytes and they are free from the flammability problems that have plagued LIBs. Stretchable Ag-Zn batteries based on Ag NWs were embedded into tensile PDMS matrixes, offering unique dual functionalities and preserving their performances even under high stretching states (80% strain) [176]. As shown in Figure 12(c), an integrated approach was developed to fabricate mechanically robust and intrinsically safe Ag-Zn batteries. The fractal design includes helical spring and serpentine current collectors, which are utilized as structural supports and backbones for all battery components. Wire-shaped batteries are prepared by using the helical spring design-based current collectors, which are resilient to fatigue and maintain electrochemical parameters after more than 17,000 cyclic bending tests at a 0.5-cm flexure radius. Serpentineshaped batteries could stretch with tunable degrees and directions while preserving their specific capacity [177]. Developing newly flexible and stretchable Zn-based batteries with high mechanical stability and power safety that is on par with commercial standards is still a major challenge in the future.

4.4. Stretchable nanogenerators

Nanogenerators have attracted much attention because they can convert the mechanical energies (such as body or muscle tensile movements), vibrational energies (such as acoustic or ultrasonic waves),



Figure 11. (a-c) Schematic illustration of the structure of stretchable LIBs, which integrate the multiple layers configuration and fractal design of stretchable interconnects. (d) Photographs of the Al and Cu electrode pads and self-similar interconnections on silicon wafer. (e) Photographs of the LIBs lighting a red light-emitting diode under different types of deformations, including biaxial stretching to 300%, folding, twisting, and mounting on the human elbow (Reproduced with permission [28]).

and hydraulic energies (such as body fluid flows) into electrical energy. Various studies demonstrated NGs could be used to power micro-/nanoscale devices without traditional batteries, and play a key role in the development of self-powered systems [178,179]. According to the mechanism, piezoelectric NGs (PENGs) harvest the mechanical energy, and the triboelectric NGs (TENGs) utilize triboelectrifications and electrostatics induction to convert ambient mechanical energy into electrical energy.

The development of tensile energy generation devices is indispensable for achieving stretchable, self-powered electronic systems. Stretchable TENGs are highly dependent on their constituent materials, and materials with intrinsic elasticity like PDMS and rubber are necessary [180–182]. For example, a flexible TENG based on wavy-structured Kapton films and serpentine electrodes on tensile elastomers was presented, achieving a maximum tensile strain of 22% [183]. A fabric-structured TENG was prepared and

used the highly stretchable 2D fabrics by weaving fibers. The fiber mainly consists of Al threads and PDMS tubes with a high-aspect-ratio nanoscale textured surface with vertically aligned NWs, which exhibited high robustness behavior even after 25% stretching [184]. The traditional origami or kirigami structural configurations are also used in the fabrication of stretchable TENGs. As shown in Figures 13(a, b), a novel type of highly stretchable TENGs was prepared, where the stretchability comes from the mechanically design of interlocking kirigami structures. Owing to the shape-adaptive thin film design, this TENG can harvest energies from various types of motions, including stretching, pressing, and twisting. The stretching limit of PET and Kapton TENG can reach up to 60% strain. The pressing mode can be easily realized by hand clapping, the TENGs can generate a maximum open-circuit voltage V_{oc} = 115.49 V and a maximum transferred charge $Q_{\rm tr}$ = 39.87 [56].



Figure 12. (a) Polarization, electrochemical impedance and discharge curves, and photographs of stretchable batteries at 0, 50, and 100% strain (Reproduced with permission [174]); (b) Screen-printing steps of a Zn-Ag₂O battery on a stretchable textile using a SIS binder and photographs of sealed battery while being 0% stretched, twisted, indentation strains, 100% stretched, and biaxial stretched (Reproduced with permission [175]); (c) Electrochemical and mechanical performance of the stretchable Ag-Zn batteries (Reproduced with permission [177]).



Figure 13. (a) Schematic structure and (b) photograph of paper-based stretchable TENG under different tensile strains (Reproduced with permission [56]); (c) Schematic illustration and (d) photograph of the stretchable PENG device, (e) piezoelectric output voltage of the device under stretch-release condition, and pyroelectric output voltage under a thermal gradient (heat and cool), and (f) photograph of the device at various locations on human body (Reproduced with permission [186]).

Recent studies have promoted the development of micro-/nanoscale PENGs with high efficiency on soft substrates and in unconventional structure for bendable energy harvesting. However, the introduction of stretchability is still relatively difficult because the applied strains could exceed the fracture limitations of the most efficient piezoelectric crystals. Polymeric polyvinylidene fluoride (PVDF) materials are often used in the PENGs, which are naturally soft and tensile, accommodating a maximal strain of 2% or higher [15]. To realize the stretchable PENGs, modified PVDF and the classic strategies should be integrated. For example, a novel stretchable transparent PENG was prepared, and the device had a sandwich structure. Organic piezo-electric material of poly(vinylidene fluoride trifluor-oethylene) [P(VDF-TrFE)] was loaded between the mobility-modified CVD-grown graphene electrodes by ferroelectric polarization into P(VDF-TrFE), and PDMS substrate was used for achieving the stretchabil-ity [185]. As shown in Figures 13(c–f), higher tensile



Figure 14. (a) Fabrication process, photographs and optical transmittance of a TFT array (Reproduced with permission [195]); (b) Sequential transfer process, photographs of the device at initial state, twisting state, stretching state (100%) and devices electrical performance (Reproduced with permission [200]).

hybrid NGs were prepared, consisting of micro-patterned P(VDF-TrFE), PDMS-CNT composites, and graphene nanosheets. Individual NG can harvest both the mechanical and thermal energy. They exhibit a high electrical performance even after 30% stretching and yield very stable piezoelectric and pyroelectric power outputs because of micro-patterned structures [186].

Various fabrication methods have enabled the design of high-performance PENGs and TENGs on flexible and stretchable substrates [187]. In fact, the introduction of stretchability for NGs is beneficial to increase the energy outputs. However, the maximum challenge would be the integration of stretchable NGs with a conventional energy storage unit to construct a self-charged power system. How to use the individual stretchable PENG and TENG to drive the wearable electronic devices, and replacing traditional batteries to build high-efficiency self-power systems, are the emerging issues in the future.

5. Stretchable transistors, sensors and artificial akin

5.1. Stretchable transistors

With the most exciting recent developments of printable organic electronic devices and flexible/stretchable electronic devices, many novel methods have been developed to design conjugated polymer semiconductors with new properties such as molecular stretchability and self-healing [188,189]. Various polymer materials have been developed for TFTs making them highly stretchable [190–192]. For example, organic transistors with elastic conductors and dielectrics could be stretched up to 250% strain while preserving the transistor characteristics [193].

Currently, representative examples of stretchable transistors usually use thin films of inorganic nanomaterials as the semi-conductive and conductive layer. An early report demonstrated that monolithic graphene transistors exhibited hole and electron

mobilities of 1188 \pm 136 and 422 \pm 52 cm² V⁻¹s⁻¹, respectively, with stable operation at up to 5% strain even after 1000 or more cycles [194]. As shown in Figure 14(a), intrinsic stretchable and transparent TFTs based on printable Ag NWs, CNTs and an elastomeric dielectric have been developed. The manufacturing processes are solution-based approaches and involve directly fabricating onto the stretchable substrate. The final TFTs exhibited a mobility of ~30 $cm^2 V^{-1} s^{-1}$, on/off ratio of $10^3 - 10^4$, switching current >100 μ A, and transconductance >50 μ S, which could be operated at lower input voltage and higher stretching state (as high as 50% strain). After 500 cyclic stretching tests (20% strain), the electrical performance is stable without significant loss [195]. Indeed, the TFTs on soft and elastic substrate require tensile gate materials. Ion gels (gelation of an ionic liquid) are one of the best candidates for such materials, and they have drawn wide attention owing to their good printability, higher ionic conductivity, and large specific capacitance [196].

Besides the direct fabrication of transistors onto stretchable substrates, transferring the as-prepared electrodes or transistors onto stretchable substrates is another conventional approach to achieve stretchable transistors, such as stretchable ZnO TFTs [197], and SnO₂ NWs-based array of field-effect transistors (FETs) [198]. For example, novel tensile FETs have been developed and illustrated, which could operate at the stretching state as high as 50% strain without significant loss in electronic properties. The MBM approach is used to fabricate stretchable FETs, where the channel is made of a buckled thin film of polyfluorene-wrapped SWCNTs semiconductor, the dielectric layer uses a soft ion gel, and the electrode layer is based on a buckled metal film. These structures enable a high degree of stretchability, and the resulting FETs exhibited a good transistor performance with on/off ratio of $>10^4$, mobility of 10 cm- $^{2}\cdot V^{-1}$ s⁻¹, and could be operated at lower voltage less than 2 V and stretching state over repeated

mechanical cycling, with further strain accommodation possible [199]. As shown in Figure 14(b), mechanically durable and highly stretchable transistors are fabricated by using semiconducting and conducting CNTs. Such transistors could be stretched up to 100% and showed similar strain-dependent behaviors when uniaxially stretched parallel and perpendicular to the charge transport directions. The semiconductor and source/drain electrodes were transferred onto the gate dielectric and TPU substrates [200]. Among various fabrication approaches for stretchable TFTs, the transfer procedure could mitigate some thermal problems (such as deformation, and degradation) associated with plastic or rubber substrates by separating them from conventional annealing procedure.

5.2. Stretchable sensors and actuators

5.2.1. Stretchable physical sensing devices (SPSDs) In the future, a wide variety of flexible electronic devices will be wearable and applied in close contact with our skin. For accommodating multifarious deformations such as twisting and elongation from human motion, these devices should ideally be stretchable. SPSDs will enable a plethora of new applications in multifunctional electronic skin, human motion detection, personalized healthcare monitoring, and human-machine interfaces.

5.2.1.1. Strain sensor. Recently, there has been growing interest in stretchable strain sensors because they can be used in human motion detection. Strain sensors/gauges are devices used to measure strain on an object [201,202]. In particular, stretchable, skin-mountable, and wearable strain sensors are required for a great number of potential applied fields including personal health monitoring, human activity detection, human-machine interface, soft robotics, and so forth [203]. Strain sensors are often categorized into resistive and capacitive strain sensors.

For the resistive strain sensor, the electrical resistance value changes when the object is deforming or stretching. The changing values are often detected by the Wheatstone bridge, resulting in the gauge factors becoming an important index value for the sensor performance. Therefore, stretchable conductors are often used to fabricate the resistive strain sensor via the structure design, and hence Ag NWs, CNTs, graphene and conductive polymers are the mainstream materials for manufacturing stretchable strain sensors [204–206]. The constant conductivity of the stretchable electrode is a key factor in obtaining reliable sensors. As shown in Figure 15(a), the aligned SWCNT-based thin films are used to prepare a novel stretchable strain sensor; this SWCNT film will

fracture into gaps and islands, and bundles bridging the gaps. The resulting sensor could measure strains up to 280%, and exhibited a high endurance, rapid response and lower creep [207]. As shown in Figure 15(b), another sandwich structural stretchable strain sensor is fabricated by using Ag NWs thin film embedded between two layers of PDMS, which exhibits strong piezoresistivity with tunable gauge factors in the ranges of 2 to 14 and can be stretched as high as 70% strain [208]. Various structure and pattern design, and different fabrication strategies are introduced in the fabrication of high-sensitivity stretchable strain sensors, such as embedded 3D printing of strain sensors [209], electrospinning conducting polymer microfibrous arrays [210], synergic conductive networks and sandwich structure (1000% maximum strain) [211], yarn structural graphene strain sensors [212], laser carbonization unidirectional strain sensors [213], and directly printing ring and diamond shaped Ag NWs electrodes [214]. In addition, for graphenebased sensors, the main vibration frequency and electrical conductivity of graphene strongly depends on its topological structures, which could be modulated by applying uniaxial strains, enabling it to be useful for high sensitivity stretchable strain sensing [215]. However, the pure graphene layer could only be stretched to a very limited extent of ca. 6% [64]. As shown in Figure 15(c), the crumpled graphene and nanosized celluloses are embedded into PDMS and finally fabricated as 3D macroporous nanopapers, achieving a high stretchability (up to 100%). The tensile graphene nanopapers were used as strain sensors for monitoring human activities [216].

The rapid development of multidimensional sensors has been intended to surmount limitations of the ordinary single axis-strain sensor. A multidimensional strain sensor was composed of two layers of a pre-strained Ag NW percolation network with decoupled and polarized electrical response in principal and perpendicular directional strain. The information on strain vector is successfully measured up to a 35% maximum strain with large gauge factor (>20) [217]. Another thicknessgradient stretchable strain sensor is developed via employing the self-pinning effect of SWCNT solution, which exhibited large gauge factor, higher durability and highly uniaxial and isotopic stretchability [218]. Compared with the aforementioned single axis-strain sensor, uniaxial and isotopic stretchable strain sensors show a broader potential for application in stretchable electronics and wearable electronics.

For the capacitive strain sensor, which can be assembled by sandwiching elastomer dielectric layers between elastomer electrodes that are filled with conducting components, algorithms for calculating sensor capacitance are based on a lumped-parameter



Figure 15. (a) Schematic of preparation procedures of stretchable SWCNT strain sensor and image of the sensor under stretching. IPA stands for isopropyl alcohol. (Reproduced with permission [207]); (b) Motion detection of index and middle fingers (Reproduced with permission [208]); (c) Schematic of the manufacture procedures for stretchable graphene nanopapers, photographs of nanopaper at different mechanical states and relative resistance changes for five independent strain sensors (Reproduced with permission [216]).

equivalent electrical circuit for the sensor [219]. The electrodes are usually patterned as interdigitated shape to construct a patterned capacitive strain sensor [220]. Generally, the gauge factor of capacitive strain sensor is lower than the resistive strain sensor, which is also can be used to monitor human motion. For example, a highly stretchable capacitive strain sensor was prepared by a metal deposition and laser rasterization method. Conductive electrodes maintained their conductivity up to 250% strain and capacitive soft sensors produced a linear output up to 85% strain because of the electromechanical feature of the conductive area. This capacitive strain sensor was used to monitor human motion and respiration through its integration into a wearable arm sleeve and a thoracic belt, respectively [221].

5.2.1.2. Temperature sensor. If the environment cools, then messages from the skin alert the body's thermostat. The temperature of human body plays a key role in human motion detection and healthcare monitoring, resulting in temperature being one of the most commonly used sensing factors. Stretchable temperature sensing (STS) is one of the critical sensing components for artificial skin. As the normalized current or other electronic signals change as a function of temperature, the stretchable and multifunctional sensor can act as a STS. Much progress has been made to develop the STS, including the thermistor, resistive temperature detectors, and semiconductor-based sensors [222-224]. As shown in Figure 16(a), a soft and biocompatible STS is prepared via the transfer printing method, combining the temperature sensitive materials with semipermeable PU films. An unambiguous temperature signal change on the skin surface caused by flowing air and water drops can also be detected to confirm the precision and dynamical responses [225]. Another scalable and efficient manufacturing approach is adopted to yield a sensor consisting of ZnO NWs and PU fibers; the obtained devices can be used as strain, temperature, and UV multifunctional sensors. The accurate detection range of temperature is between room temperature and 50 °C, and temperature sensitivities are 39.3% °C⁻¹, 26.1% °C⁻¹, 20.1% °C⁻¹, and 16.8% °C⁻¹ versus different strains of 0%, 25%, 50%, and 100%, respectively [226]. Figure 16(b) shows a PANI nanofiber-based STS array combined with an active matrix consisting of SWCNT TFTs. The integrated STS array exhibits stable stretchable performance under biaxial stretching at 30% strain, and has corresponding temperature distribution mapping without any mechanical or electrical degradation [227]. One of the biggest challenges ahead in the STS field is still the fabrication of stretchable-functional electronic components and interconnections, particularly for advanced artificial E-skin.

5.2.1.3. Pressure sensor. The main sensing mechanisms of stretchable pressure sensor (SPS) include resistivity, piezoelectricity, capacitance, and piezoresistivity; depending on the used active materials and device configuration, each of these sensing mechanism has its own features [228,229]. Stretchable substrates, especially pressure sensitive rubber (PSR), are often used to fabricate the force collector type of pressure sensors, which generally use force collectors to detect strains (or deflections) due to applied force over an area (pressure). However, PSR-based pressure

sensors are commonly susceptible to hysteresis [230]. Therefore, various strategies and structure designs are employed to fabricate high-sensitive SPS. As shown in Figure 17(a), a resistive SPS was fabricated by combining the compressible PDMS substrate with micro-pyramidal characteristic arrays and tensile PEDOT:PSS/polyurethane dispersion (PUD) polymer electrode. The pressure induces changes of geometries, and the composite electrode can stretch up to a 40% strain with measured a sensitivity of 10.3 kPa⁻¹ [231]. The stretchable array of highly sensitive SPSs as body-attached devices can simultaneously detect both the pressure and shape of an object under conditions involving body movements such as bending or stretching [232]. For example, an ultrathin activematrix array with resistive tactile sensor was developed, which could be crumpled like paper, withstand stretching as high as 230% on pre-strained elastic substrates, and operate at high-temperature or in aqueous environments. As shown in Figure 17(b), this SPS array can detect both the pressure and the shape of a ring [233].

Optical-type pressure measurements are attracting much attention because of their tremendous commercial prospects. For instance, a novel optical-type SPS was produced, and could serve as a functional substrate because of its high stretchability and transparency. Its sensitivity could be up to 0.2 kPa⁻¹, which demonstrated that weight as low as 1/10 mg could be reliably detected. With a stretching up to a 40% strain the detected sensitivity retains constant at ~0.025 kPa⁻¹ [234]. As shown in Figure 17(c), a stretchable electroluminescent actuator was developed. In this SPS, multifunctional materials with higher stretchability were introduced and served as light emitting sensors to detect the internal and external pressures. This novel SPS could be applied in soft robotics, which illustrated these integrated abilities by stretching and emitting light in motion [235].

Promising approaches towards stretchable conductors and stretchable matrix-type substrates are developing all the time. However, more inexpensive and high-throughput strategies or techniques are still required to produce SPSs with enough sensitivities in both medium- (10–100 kPa, comparable to object manipulation) and low-pressure regimes (<10 kPa, suitable for gentle touch) [236].

5.2.2. Stretchable electrochemical sensing devices Electrochemical sensors are powerful tools that can provide accurate chemical information with a fast response and outstanding sensitivity. For further application in wearable electronics and artificial skin, electrochemical sensors with favorable stretchability will be ideal tools for conforming to cell and tissue deformations, achieving real-time monitoring of various chemical signals.



Figure 16. (a) Configuration of the soft STS on breathable films (Reproduced with permission [225]); (b) Schematic of the preparation processes for the STS array, photographs and temperature mapping of the STS array on the tensile substrate attached onto the right palm where a heart-shaped cold water container (\approx 15 °C) was positioned before and after stretching (Reproduced with permission [227]).

Stretchable electrochemical sensing devices (SESDs) are conceivably a powerful technology that offers crucial chemical information for unravelling soft and curvilinear living body [237]. As shown in Figure 18(a), a fully-printed, low-cost, highly stretchable CNT-based SESD and biofuel cell arrays were developed, which could withstand stretching strain as high as 500% with negligible effects on their structural integrity and electrochemical performance.

Various electrochemical measurements of this SESD reveal that the electrochemical performance is stable without significant loss when the sensor is stretched, torsionally twisted, and indented [238]. A broad range of wearable SESDs and biosensors are designed and fabricated to achieve real-time noninvasive healthcare monitoring, the acquisition electrolyte and metabolite signals as indicator come from the wearers' sweats, tears, or saliva [239,240]. Besides



Figure 17. (a) Schematic of the preparation process of resistive SPS (Reproduced with permission [231]); (b) Schematic of a thin large-area active-matrix SPS array with 12×12 tactile pixels. When the metallic ring was placed on the SPS array, the corresponding drain current image showed its shape (Reproduced with permission [233]); (c) hyperelastic light-emitting capacitor skin endows soft robots with the capability to sense their actuated state and environment and communicate optically (Reproduced with permission [235]).

the traditional electrochemical sensor, other electrochemical mechanism based chemical sensors are also endowed with stretchability to extend their application into wearable electronics, such as gas sensors and pH sensors [241]. For example, as shown in Figure 18 (b), interconnected arrays of miniaturized IrOx pH sensors are prepared through superior material assembly and electrochemical growth techniques. These sensor arrays are placed into the soft and low-modulus elastomer to fabricate a conformal monitoring system, which could be used on the surface of the beating heart because of its non-invasive character. The sensor arrays exhibit excellent Nernstian sensitivities with outstanding uniformities $(70 \pm 2 \text{ mV/pH})$ in *in vitro* testing [242]. Recently, a highly stretchable, transparent gas sensor based on Ag NWs-graphene composite nanomaterials has been fabricated, which demonstrated excellent and stable performance even under mechanical deformations (up to 20% strain). This sensor could integrate Bluetooth systems or inductive antennas, enabling wireless operation (Figure 18(c)) [243].

5.2.3. Stretchable actuators

Actuators control or move things around in a system. An actuator requires an energy source and a control signal which can be in the form of an electric or even mechanical signal. Wearable actuators also require stretchable electronics where the circuit can sustain large deformations without electrical failure or detrimental loss of performance [244]. Stretchable actuators include thermal and electric actuators. Thermal actuators directly convert thermal energy into motion, and the thermal



Figure 18. (a) Image of the stencil employed for printing the stress-enduring stretchable devices, the structure and photograph of SESDs arrays under different stretch states (Reproduced with permission [238]); (b) Photograph of a representative, soft, tensile pH sensor arrays and schematic demonstration of chemical reaction associated with IrO_x electroplating (Reproduced with permission [242]); (c) Schematic illustration of wireless and real-time graphene gas sensor integrated with Bluetooth system (Reproduced with permission [243]).

energy generally comes from flexible heaters. For example, Zhu et al. fabricated low-voltage and extremely flexible electrothermal bimorph actuators, which were made of flexible Ag NWs based heaters on PDMS substrate, which exhibited a fast heating rate of 18 °C s⁻¹ and stable heating performance with large bending. The actuators offered the largest bending angle (720°) or curvature (2.6 cm^{-1}) at a very low actuation voltage (0.2 V sq⁻¹ or 4.5 V) among all types of bimorph actuators that have been reported to date [245]. Electric actuators directly convert electric energy into motion [246-248]. For instance, Yeo et al. present a simple method of integrating a flexible pneumatic actuator with stretchable strain sensor to form a soft sensorized actuator. When the actuator deformed, the strain sensor was stretched, leading to an increase in the normalized resistance. The integration of a pneumatic soft actuator with sensing element enables the measurement of the extent of actuator bending [249]. Dielectric elastomer actuators (DEAs) are flexible lightweight actuators that can generate strains of over 100%, which consist of an electrode-elastomer-electrode stack, placed on a frame. Applying a voltage between the electrodes electrostatically compresses the elastomer, which deforms in-plane or out-of-plane depending on design [250].

Actuator materials convert input energy to mechanical output energy. We must note that in many cases, the actuator does not achieve stretching, and the stretchable actuator often uses a stretchable electrode to achieve the transducing output. Obviously, a key element in stretchable actuators and systems based on elastomer materials are compliant electrodes.

5.3. Stretchable artificial skin

Various sensory receptors in human skin transmit a large number of tactile and thermal signals from the external environment to the brain. Artificial skin is a classical integrated electronics system (IES) and multifunctional deformable sensing platform (MDSP), integrated with stretchable temperature, strain, pressure and humidity sensors [251–255]. Despite advances in our understanding of mechanic and thermal sensation, reproducing these unique sensory features in stretchable artificial skin remains a bigger challenge.

As shown in Figure 19(a), a smart artificial skin is developed by integrating the strain sensor, STS, SPS, humidity sensor, electroresistive heater, and tensile multiple electrode arrays, and is used for nerve stimulations, all the sensors being based on ultra-thin, single crystalline Si nanoribbons. As a multifunctional sensing platform, which can sense and actuate by a broad range of sensory inputs because of various skin deformations, it thus offers enhanced functions and high performance in the emerging fields of smart prosthetics [256]. Another tensile and multiple-forcesensing functional textile has been used as artificial skin, and was weaved by the stretchable coaxial sensor electrodes. Although the stretchable functional textile uses only one kind of sensing unit, it can simultaneously map and quantify the mechanical deformations generated by conventional pressures, lateral strains, and flexion [257]. As shown in



Figure 19. (a) The structure of smart artificial skin, which consists of strain, pressure, and temperature sensor array and the corresponding photographs of this skin covering the entire surface area of a prosthetic hand. The inset demonstrates that the artificial skin is stretching ~20% and the structure of artificial skin and Si nanoribbon sensors (Reproduced with permission [256]); (b) Photographs of artificial skin prototype with soft, stretchable and deformable characteristics (Reproduced with permission [258]).

Figure 19(b), an artificial skin prototype was designed and fabricated from multi-layer tracks in elastomer matrixes, enabling detection of multi-axis strains and contact pressure. The size of this sensor is about 25 mm \times 25 mm, and its thickness is approximately 3.5 mm. This sensor is functional up to strains of approximately 250% [258]. Clearly, artificial electronic skin consists of mechanically deformable and stretchable sensor arrays or networks that can accommodate an irregular surface and spatially map/quantify a broad range of stimuli, such as strain, pressure, and temperature to imitate human somatosensory systems [259].

Currently, the fabrication process of stretchable artificial skin is still time-consuming and complex. Therefore, an easily-fabricated and low-cost manufacturing method is needed, and then next generation artificial skin will become a promising candidate for highly-sensitive IES and multifunctional sensing platform, and further achieving potential applications in advanced robots, human-machine interface devices, prosthetics fields and healthcare monitoring.

6. Transparent stretchable electronics

6.1. Stretchable transparent electrodes

Flexible and stretchable transparent electrodes (STEs) play a key role in the next generation of flexible and wearable electronics. As they can preserve the electrical conductivities and stabilities under large mechanical deformations they are highly desirable for various applications, such as touch

screens, OLEDs, and other portable electronic devices or optoelectronic devices [260,261]. Three kinds of conventional materials are used for achieving STEs, including metal NWs, carbon nanomaterials (graphene and CNTs) and PEDOT:PSS or their hybrid or composite nanomaterials [262,263], as shown in Table 3.

Metal NWs have been intensively studied as promising materials for transparent conducting electrodes because of their simple tunable synthesis and excellent electrical performance. For example, a series of Ag NWs with different aspect ratios in the range of ca. 30 to ca. 1000 could be controllably prepared; these are often synthesized by a simple PVP-mediated polyol approach, using PVP with different average molecular weights that can effectively tailor the aspect ratios of Ag NWs [74]. Ag NWs based STEs consist of Ag NWs networks embedded in the surface layer of a cross-linked poly(acrylate) matrix. The interpenetrated Ag NWs networks and the cross-linked polymer matrix resulted in STEs with high surface conductivity, high transparency, and rubbery elasticity. The sheet resistance of the STEs increases by only 2.3 times at 50% strain [273]. In addition, Cu NWs are much cheaper with a comparable electrical conductivity as Ag, and are more stable under electrical currents. The Cu NWs/PDMS electrodes are easily torn off from the substrate during the stretching process because of the weak adhesion between PDMS and Cu NWs [274]. As shown in Figure 20, metal NWs-based STEs are often fabricated by adding NWs to a stretchable material and then peeled-off from the substrates, and the stretchable substrates

Table 3. Materials, transmittance, electrical property, and stretchability of STEs.

| Materials | Methods | Transmittance | Electronic Properties | Stretchability | Ref. |
|--------------|------------------------------------|---------------|--|---|-------|
| Ag NWs | Drop-casting and peel-off | 80% | 10 Ω /sq at 588 mg/m ² and | $R_{\rm S}$: 10 ² –10 ³ Ω /sq at strains as high as | [264] |
| | | | 88.6 Ω /sq at 147 mg/m ² | 140% | |
| Ag NWs | Spray-deposited | ~80% | ~35 Ω/sq | 30% elongation with less than 40 Ω /sq | [265] |
| Cu NWs | Drop-casting and peel-off | 78% | 22.1 Ω/sq | 50% strain is about 40 Ω /sq | [274] |
| Cu NWs | Curing and peel-off | 84.5% | 56.2 Ω/sq | $R_{\rm S}$ < 100 Ω /sq at tensile strains up to 60% | [266] |
| Au nanomesh | Grain boundary lithography | 82.5% | 21 Ω/sq | $R_{\rm S}$ increased by 3.2 times ($\epsilon = 160\%$) and | [267] |
| | , , , | | • | stable for 1000 cycles ($\epsilon = 50\%$) | |
| Pt nanomesh | Anodized aluminum oxide template | 75.2% | 71 Ω/sq | Resistance unchanged at $\varepsilon = 16.8\%$ | [268] |
| | and metal sputtering methods | | • | 5 | |
| Superaligned | Dry method growth and roll-to-roll | 90% | 208 Ω/sq | 24 Ω/sq, 83.4% | [269] |
| CNTs | setup | | · | r. | |
| Graphene | Transferred film | ~80% | ~280 Ω/sq | Recovers its original resistance after | [64] |
| • | | | · | stretching by ~6% | |
| Graphene/ | Spin-coating | 86% | 68 Ω/sq | Stretching up to the strain of 15% | [270] |
| PEDOT:PSS | 1 | | | 5.1 | |
| AaNW/ | Spin-coating | 94% | 33 Ω/sq | Negligible resistance change within $\varepsilon =$ | [271] |
| graphene | 1 | | | 100% | |
| Aa NWs/ | Spin-coating | 94% | 33 Ω/sq | 100% in tensile strain | [272] |
| graphene | | | | | |



Figure 20. Schematic illustration of drop-casting and peel-off method to fabricate the STEs.

need a curing or casting process; sometimes the metal NWs have been embedded into the stretchable material after drying.

Conductive carbon nanomaterials are conventional materials used to fabricate the STEs, including the popular CNTs and graphene. For example, STEs are prepared by spray-depositing the SWCNTs, which could provide stretchabilities via applying strains along each axis, and then releasing these strains. This method yields spring-like structures in the CNTs, accommodating strain of as high as 150% and a high conductivity up to 2200 S cm⁻¹ in the stretched state [275]. Figure 21(a) shows a templatedriven self-assembly method used to integrate the SWCNTs with 2D rhombic nanomesh films (RNFs), in which the deformations of the rhombic shapes accommodate the strains, greatly enhancing the stretchabilities. The RNFs displayed excellent lower sheet resistance (~10 times) at a similar optical transmittance (78%), greater stretchability (~8 times less resistance increase at 30% strain), and good mechanical durability (~42 times less resistance increase after 500 cyclic stretching tests at a strain of 30%) than those of random-CNT-networks films [276]. Additionally, because of its outstanding optical, electrical and mechanical properties on the microscale,

graphene becomes a promising candidate as the basis carbon nanomaterial in the fabrication of SEs and STEs. However, large-area CVD growth graphene possess a real problem of low stretching ability, which is much lower than that of mechanically exfoliated pristine graphene owning to the yielded intrinsic and extrinsic defects during its preparation, etching-out of the catalytic metals, and the transferring process. This low stretchability becomes a major obstacle to commercially applying the CVD graphene in the field of STEs [277]. The large-scale production of high-quality SWCNTs and monolayer graphene is still a major challenge in the fabrication of carbon nanomaterial-based STEs, two major pathways toward obtaining these carbon nanomaterials separated by electronic structure: selective synthesis and post-synthesis separation.

Despite their discovery decades before the emerging inorganic nanomaterials described, conductive polymers have only been used as STEs in commercial niche applications. In particular, PEDOT and its derivatives became the most successful conducting polymers because of their intrinsically high conductivity and stability. For example, a fluorosurfactant was introduced into the conducting and transparent PEDOT:PSS film and this film was used as an STE.



Figure 21. (a) Schematic of fabrication process, atomic force microscopy images of SWCNT nanomesh films, and the photographs of nanomesh electrode films on different substrates (Reproduced with permission [276]); (b) Schematic of spray-coating EG/PEDOT:PSS hybrid inks onto desired substrate, the corresponding transmittance spectra, scanning electron microscopy image and photographs of as-obtained films (Reproduced with permission [281]); (c) Schematic of the process to fabricate the graphene–metal nano-trough hybrid electrodes (Reproduced with permission [282]).

Four-layer PEDOT:PSS films possessed a sheet resistance of 46 Ω /sq with 82% transmittance (at 550 nm). These films were deposited onto the surface of prestrained and buckled PDMS substrate. This STE exhibited a high and reversible stretchability and excellent stability without a significant loss in sheet resistance after more than 5000 cyclic stretching tests (0–10% strains) [278,279]. Apart from using individual PDMS as conventional stretchable substrate, stretchable conductors based on PEDOT:PSS and PDMS composite polymers are also fabricated via embedding PDMS oligomer and curing agent into a 3D-PEDOT:PSS aerogel [280].

Recently, the integration and hybrid utilization of the aforementioned three materials have attracted much attention because the composite materials may be able to improve the stretchable performance. As shown in Figure 21(b), a new solution preparation method of large-area, high conducting graphene films-based STEs via spray-coating of hybrid inks of exfoliated graphene (EG)/PEDOT:PSS onto PDMS is demonstrated, without significant loss in conductive properties after 50 cyclic tests under stretching to 20% [281]. A recent study compared the stretchability and electronic performance of individual graphene and metal hybrid nano-trough networks. The results demonstrated that composite materials could greatly enhance the uniformity of sheet resistance. The resultant STEs showed a low sheet resistance (1.0±0.1 Ω /

sq), a high transparency (91% in the visible range), and superior stretchability (80% in tensile strain) [282].

The crucial requirements for STEs are high conductivity, high transparency and enough stretchability; however, these features somewhat contradict and restrict each other. From a practical point of view, these materials must be colorless and inexpensive to produce, preferably from non-toxic and environmentally friendly materials. In addition, Park et al. summarizes recent advances in the STEs based on in-plane structures [283], where the structure design could enhance the stretchability of STEs. For example, tensile and transparent graphene interconnections are employed to fabricate microscale inorganic LED arrays on rubber elastomers, and mechanical modeling reveal the strain distributions during the stretching process. As a completely reversible method, the graphene-based STE interconnection integrated LED arrays could withstand more than 100% of strain [284].

6.2. Transparent and stretchable electronic devices

Integration of transparency, flexibility, and stretchability to electronic devices has gained a lot of interest due to the value of wearable electronic systems in practical life. Colorless transparent and stretchable electronic devices (TSEDs) with high deformability



Figure 22. (a) Schematic illustration of stretchable device preparation, graphene/SWCNT TFT preparation and the wrinkled Al_2O_3 layer transferring processes (Reproduced with permission [287]). (b) Schematic of device structures, photographs of the device under different deformation states, transferring and optical transmittance of gated device (Reproduced with permission [290]). (c) Schematic images of the wireless-functional sensor, photographs of the fabricated sensor and attaching to the hand skin, frequency response of the antenna and wireless sensing curves of a sensor (Reproduced with permission [291]).

and conformability are key for wearable sensors or other stretchable electronic devices. Up to now, various TSEDs have been fabricated, such as transistors [197], heaters [136], thin film loudspeakers [285], pressure, strain and touch sensors [286], and artificial skin [234]. As shown in Figure 22(a), highly stretchable and optically transparent FETs are fabricated by integrating graphene/SWCNT electrode and SWCNT-network channel with wrinkled dielectric Al₂O₃ layers. The FETs showed outstanding on/off ratios of ~10⁵, a high mobility of ~40 cm² V⁻¹ s⁻¹ and a low operating voltage < 1 V. Moreover, owing to the wrinkled morphology of Al₂O₃, the transistor preserved its characteristics under strain up to 20% without perceptible leakage current increases or physical degradations. No significant loss in electronic performance was found after more than 1000 cyclic stretching/releasing tests [287]. Recently, transparent stretchable sensors have met the requirements of wearable, patchable, or implantable electronics and became a research focus. They show promising applications in health monitoring and disease diagnostics, and hence various transparent stretchable sensors were designed and fabricated [275,288]. For example, a stretchable, ultra-sensitive, and patchable strain

sensor with high transparency is developed, which consists of a novel sandwich-like stacked piezoresistive nanohybrid film of SWCNTs and a conducting elastomeric composite of PU-PEDOT:PSS. The sensor showed good mechanical stability, higher stretchability (as high as 100% strain), optical transmittance of 62%, and gauge factor of 62, suggesting that when it is attached to the skin of the face, this sensor would be able to detect small strains due to conventional emotional expressions, including laughter, crying and eye movements [289]. As shown in Figure 22(b), another transparent stretchable gated sensor array (TSGSA) showed a high optical transparency, conformality, and high stretchability (the maximum stretchability can be up to 70% strain). This TSGSA possesses a higher responsivity to a temperature change of approximately 1.34%/°C in objects and human skin, and could maintain its response after 10,000 stretching cycles at the strain of 30% [290]. As shown in Figure 22(c), Ag NWs-graphene composite nanomaterials are used to fabricate the field-effect sensors, which exhibited higher mobility (~3000 $cm^2 V^{-1} s^{-1}$) with lower contact resistance. The TSEDs can transfer onto the various surfaces of different substrates. The integration of these sensors for

RLC circuits enables wireless monitoring [291]. Another direction in stretchable electronic devices is integrating power sources, constructing self-powered electronic devices. For example, an Ag NWs/PEDOT: PSS/PU composite materials-based TSEDs was fabricated, because it integrates with both a supercapacitor and a TENG, which becomes a self-powered monitoring system for skin strains with ultra-low energy consumption [292]. Clearly, transparent and stretchable characteristics will enable electronic devices to be applied over a large range, which allows more effective planning in practical applications of TSEDs in various power sources, biomedical techniques, and wearable optoelectronic systems or devices.

Among various TSEDs, if the integrated power sources become a matching device with mechanically stretchable and optical transparent properties, the application area of TSEDs will enlarge and cover many fields besides conventional transparent electronics, ranging from self-powered rolled-up display electronics to self-powered wearable optoelectronic systems. For instance, high-performance all-solid supercapacitors with a good stability were prepared by using highly aligned CNT sheets. The current collectors and active electrodes are also utilized these CNT sheets, and hence the overall device was both transparent and stretchable. It exhibited a transmittance of 75%, a specific capacitance of 7.3 F g^{-1} and could be bi-axially stretched up to 30% of strain [293]. When CNTs were replaced by wrinkled graphene sheets, the resulting supercapacitors could be stretched up to 40% of strain without significant loss in electrical performance after more than 100 cyclic stretching tests [294]. Besides the transparent stretchable supercapacitors, transparent stretchable PENGs [185] and TENGs [187] have also been developed. Yet fabricating other transparent stretchable power sources remains challenging. The main reason is most of the existing electrodes are neither mechanical stretchable nor optical transparent (e.g. metal electrodes), with some of them either stretchable of a low-transmittance (e.g. conductive polymers) or transparent of a low stretchability (e.g. ITO glass, other metal oxide films).

7. Outlooks and perspectives

In summary, stretchable electronics offer a foundation for applications using common flexible electronic techniques because of their powerful capacities to integrate with stretchable functional materials and curvilinear surfaces. Fast development and substantial achievement have been shaping the field of wearable electronic devices, resulting in the persistent requirement for stretchable conductors and stretchable electronic devices. Clearly, recent progress in stretchable electronics has seen the emergence of new technologies, and strenuous efforts have been made to improve their electronic performance under stretching and impart intelligent functions to the surfaces of various soft substrates. Hence, a general summary of recent advances in this field and concrete examples of successful application improvements will be provided.

Two basic principles are often used for manufacturing stretchable conductors and electrodes. A first principle is the use of intrinsic stretchable materials, and another principle is making intrinsic non-stretchable materials stretchable by the aid of out-of-plane design or horseshoe-shaped planar structures to accommodate the applied strains. To obtain stretchable conductors, these conductive components (such as metal nanowires, conductive carbon nanomaterials, and conductive polymers) are often used as fillers and arranged in the elastomer matrix while combining the structure design, including wavy structural configuration, islandinterconnect, fractal design and traditional papercutting. Indeed, more than 25% stretchability is enough for using in smart clothing application and other wearable electronic devices. For fabricating stretchable electronic devices, each component of the device must maintain its performance up to a critical strain. For instance, wearable SPSDs and SESDs have gained a lot of interest because of their tremendous promise for a plethora of applications, resulting from their high performance, small size, and superior stretchability. By virtue of their wide applications in personal electronic devices and industrial monitoring equipment, multifunctional sensors become strong candidates for driving scientific and technological progress in modern society [295]. With continued innovations and resolutions to some key challenges, wearable SPSDs and SESDs are expected to build stretchable IES and MDSP, and finally construct wearable wireless MDSPs and body-sensory networks, which will be able to accomplish much in a broad range of personal healthcare monitoring applications, as well as in sport and military applications. Research on stretchable IES has been dominated by studying their fundamental mechanics, developing their manufacturing techniques, and displaying their capabilities and advantages [296]. To further facilitate the application of stretchable electronics, visual transparency will significantly enhance the advantages of stretchable functional materials and related strategy developments. Furthermore, the different form factors of the various components are still limited to seamless integration with the human skin, giving rise to wearability challenges and signal-to-noise limitations.

However, folding and conformable stretchable IES cannot be realized until soft stretchable power sources that match devices form-factors and power requirements are successfully produced. To achieve stretchable power sources, the present rigid key-components should be either replaced by intrinsically tensile materials or designed as stretchable structures for heterogeneous integration of hard and deformable materials. Another high-value feature for these batteries is rechargeability with high-storage capacity and wireless connection to external power supplies.

The fabrication cost is a very important factor for developing and realizing practical application of stretchable electronics. A simple approach and strategy becomes the key to achieving higher technical level and overcoming challenges. Therefore, the design of dissimilar materials and devices or fabrication process for implementing such a strategy is a considerable challenge. The research progress demonstrated printed electronic techniques would be instrumental in high-throughput manufacturing of these novel stretchable conductors and electronic devices.

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