Nature-inspired Structural Materials for Flexible Electronic Devices

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Abstract

Exciting advancements have been made in the field of flexible electronic devices in the last two decades and will certainly lead to a revolution in peoples' lives in the future. However, due to the poor sustainability of the active materials in complex stress environments, new requirements have been adopted for the construction of flexible devices. Thus, hierarchical architectures in natural materials, which have developed various environment-adapted structures and materials through natural selection, can serve as guides to solve the limitations of materials and engineering techniques. This review covers the smart designs of structural materials inspired by natural materials and their utility in the construction of flexible devices. First, we summarize structural materials that accommodate mechanical deformations, which is the fundamental requirement for flexible devices to work properly in complex environments. Second, we discuss the functionalities of flexible devices induced by nature-inspired structural materials, including mechanical sensing, energy harvesting, physically interacting, and so on. Finally, we provide a perspective on newly developed structural materials and their potential applications in future flexible devices, as well as frontier strategies for biomimetic functions. These analyses and summaries are valuable for a systematic understanding of structural materials in electronic devices and will serve as inspirations for smart designs in flexible electronics.

Contents

1. Introduction

Since the 1950s, the rise of the modern electronics industry, which is based on semiconductor materials and micro/nano-engineering processes, has dramatically accelerated the development of technology and completely changed the life-style of human beings. Humans have chosen another route for evolution that is entirely different from the natural style through which we have evolved for three million years. Through decades of development, the modern electronics industry is currently based on complementary metal oxide semiconductor (CMOS) systems, which are constructed by metals and semiconductors with high moduli and excellent electric properties. However, the rise of flexible devices in the new century brings new challenges for widely utilized electronic materials and technologies.

Next-generation flexible electronics have been colorfully and fantastically conceived in fictional novels and movies. Such electronics have drawn increasing attention and have become an attractive and fashionable topic in the science and business worlds due to their potential to bring forth yet another revolution in the human way of life.[1-4] However, most traditional materials with good electronic properties show poor sustainability in complex stress environments, which make them difficult to directly involve in flexible and stretchable device construction. Thus, much research must be conducted towards the design of structures and materials with mechanical characteristics to realize flexible and stretchable devices that can accommodate large strain and geometrical deformations, such as bending, twisting, folding, and stretching. In the long history of evolution, living organisms have developed various environment-adapted structures and materials through natural selection. Thus, nature can serve as guide for how to resolve the limitations of materials and engineering techniques for the coming electronics revolution.[5] This review focuses on the structural materials that have been designed through inspiration from natural creatures and have been subsequently utilized in electronic flexible devices to enable mechanical accommodations and functional improvements.

1.1 Nature-inspired artificial structural materials with deformability

Flexible electronic devices are designed with excellent mechanical deformability for wearable applications as well as enhanced smart functions such as sensitive environmental responsiveness, efficient human-machine interaction, and intelligent processing.[6] One of the most critical concepts for flexible electronics is the maintenance of good performance under physical deformations, as well as adaptability in real environments comprising complex strains. Thus, the development of material design and engineering should focus on the realization of stretchability and flexibility.[7-9] In general, two complementary routes are used to achieve these characteristics:

a) The design and synthesis of intrinsically flexible and stretchable materials with good electronic performance to replace traditional semiconductors.
b) The endowment of rigid high-performance semiconductors with a “soft” nature though engineering.

Electronic devices have been developed for decades based on intricate and efficient device structures and high-moduli electronic materials, such as metals and inorganic semiconductors. Although newly developed intrinsically flexible and stretchable materials show potential applications in flexible electronics, their performances and functionalities remain incomparable with that of the traditional electronic materials due to their insufficient behaviors in electronics.[10-15] Thus, the processing of traditional high-performance materials through appropriate geometrical designs for flexibility and stretchability constitutes a valuable strategy and has drawn increasing attention in flexible device engineering.[16-21] Through a long evolution of optimization, natural architectures provided many fantastic and wonderful examples of mechanical accommodation that can guide us to construct various architectures despite the limited choices of existing conductors and semiconductors. For example, by mimicking the hierarchical and laminated substructures in bone, metastability-assisted multiphase steels containing nanolaminated microstructures had been developed to achieve superior fatigue resistance though reducing crack initiation and propagation (Figure 1a,b).[22] Spider webs exhibit outstanding mechanical adaptability via the organization of two types of silk with different stiffness properties (radial and spiral threads),[23-26] while artificial web structures can also realize high mechanical strength by a hierarchically defined nanofibrillar assembly (Figure 1c,d).[27] Moreover, artifacts in our daily lives can also inspire wonderful designs in constructing flexible electronic devices (Figure 1e,f).[28] Thus, the essential characteristics of natural structural materials, which serve as guidance in the construction of structural materials in flexible electronics, can be described as:

a) *The combination of "rigid" and "soft" aspects.* Natural materials, especially organisms, comprise hard and soft phases that enable macro performance through the interplay of strength and toughness.[29,30] In constructing flexible and stretchable devices, this inspired us to endow the conventional "rigid" materials, such as metals and semiconductors, with an incorporated "soft" nature via an appropriate geometrical design to achieve mechanical compatibility with elastomeric substrates, such as polydimethylsiloxane (PDMS) and Ecoflex.
b) *Hierarchical architectures spanning from the nanoscale to the macroscale.* Natural structural materials exhibit complex mechanical properties that benefit from their 3D or 2D hierarchical structures through long term evolution, which constitutes an efficient path to combine multiple desirable mechanical characteristics, including light weight, flexibility, strength, and stretchability.[31-33] Thus, in the construction of flexible devices, architectural design on the

nano- and micro-scale may constitute a strategy that enables rigid electronic materials to accommodate the macro-mechanical strain prevalent in complex environments, which benefit from the development of nanotechnology.[34]

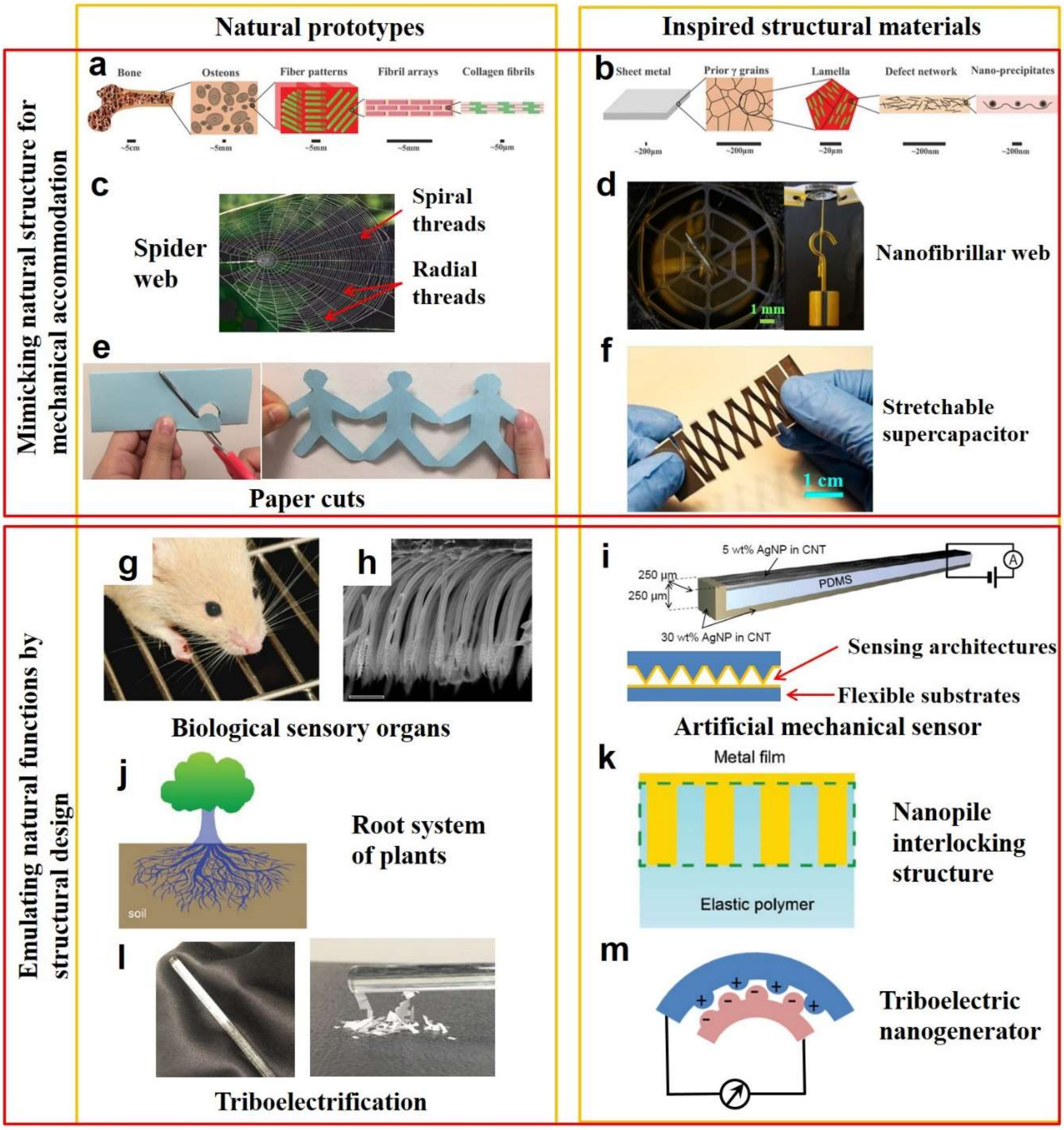


Figure 1. Artificial structural materials mimicking natural structure and emulating natural functions. a,b) Comparison of the hierarchical structures of natural bone and bone-like steel. Reproduced with permission from ref. 22. Copyright 2017 the American Association for the

Advancement of Science. c,d) Optical image of two configurations of thread in a spider web and a nanofibrillar web supporting a point load. Reproduced with permission from ref. 24 and 27. Copyright 2009 National Academy of Sciences, and 2017 Nature Publishing group. e) Photographs of papercuts. f) A stretchable supercapacitor based on kirigami structure. Reproduced from ref. 28. Copyright 2016 American Chemical Society. g,h) Photograph of the whiskers of a mouse and SEM image of the setae in beetle *Clytus arietis*. Reproduced with permission from ref. 35 and 36. Copyright 2007 Elsevier, and 2006 COMPANY OF BIOLOGISTS LTD. i) Schematic illustration of an electronic whisker constructed by a patterned CNT/AgNP composite film. Reproduced with permission from ref. 37. Copyright 2013 United States National Academy of Sciences. A schematic illustration of a pressure sensor with a structural sensing layer. j,k) The root system of plants inspired a nanopile interlocking structure. Reproduced with permission from ref. 38. Copyright 2016 John Wiley and Sons. l) Triboelectrification on a glass rod. m) Illustration of a triboelectric nanogenerator.

1.2 Artificial structural materials emulating natural functions

Due to well-established evolutionary structures, natural creatures are not only adaptable to complex strain environments but also can perform smart biofunctions, such as mechanical sensing, reversible locking and dry adhesion. For example, sensory hairs on the body surface of insects and arrays of vibrissae in the faces of cats and rodents perform highly sensitive responses to physical contact and airflow, which inspired the fabrication of artificial electronic whiskers for sensing and transducing mechanical forces (Figure 1g,h,i).[36,37,39-42] By mimicking the root system of plants, a nanopile interlocking structure was designed to enhance the adhesion between rigid metal electrodes and elastic substrates (Figure 1j,k).[38] Moreover, inspired by triboelectrification, a common physical phenomenon, various triboelectric nanogenerators have been realized with designed nano/microscale architectures (Figure 1l,m).[43] Based on transferring relative architectures in device fabrication, structure-based functions bring new opportunities in flexible devices to realize attracting and promising functions.

Recently, various nature-inspired structures have been introduced to rigid electronic materials for the construction of flexible devices to achieve diverse functionalities. The utilization of structural materials in flexible devices can be divided into two main approaches:

a) *Mechanical accommodations*: Engineering conventional high modulus materials, such as metals, semiconductors, and some polymers, into various 2D or 3D structures to accommodate mechanical deformations by shape changes for the realization of flexibility and stretchability in electronic devices while maintaining their electronic performance.
b) *Structure induced functionalities*: Mimicking natural existing structural-based phenomenon in the fabrication of flexible devices to introduce corresponding functions, which can enhance the basic electronic performance and also bring new concepts in electronic applications.

This review will summarize the different designs for realizing mechanical accommodation in electronic materials (Section **2**), as well as endowing smart functions in flexible and stretchable devices (Section **3** and **4**). The sorting and analysis of these remarkable works are expected to inspire more intelligent ideas and new insights into achieving electronic devices with excellent adaptability and wonderful functions.

2. Structural materials for accommodating deformations

Flexible devices are required to function in practical conditions comprising complex strains. For example, wearable flexible devices would have to maintain their performance while stretching and compressing along with human skin.[44-46] To address the mismatch between the rigid nature of conventional electronics and the requirements for flexible and stretchable electronics, scientists and technicians have turned to rational structural design,[19,47-52] a principle which involves releasing the applied strain through geometry changes without incurring physical damage to the electronic materials.[53-59] Inspired by architectures that exist in nature for the accommodation of mechanical stress, electrodes and functional electronic units with 2D and 3D structures have been designed and developed to maintain the conductivity and electronic performance of flexible and stretchable devices, which will be discussed in this section.

2.1 3D structure design for stretchable electronics

Through rational design, high moduli materials with a modest flexibility are engineered into various spatial structures with the ability to release incoming strain through shape changes.[48,56] One of most widely used designs is buckled (also known as wavy or wrinkled) films or stripes with a wave-like structure, which has been proven to be an effective strategy for endowing rigid materials with good stretchability. While applying an external force on these type of films, the buckling structure can flatten out to accommodate the strain. In 1998, George M. Whitesides and coworkers first fabricated a buckling metal film with a uniform wavelength of 20–50 micrometers on a thermal treated PDMS substrate, which showed potential for applications in stretchable devices.[60] The compressive stress on the polymer substrate generated during the cooling process is the reason for generation of the buckling structure in the metal films.[60,61]

A general and efficient strategy with good operability has been designed for the fabrication of buckling metal or semiconductor films, which is to deposit a continuous film with brittle materials on a pre-stretched elastic substrate that would generate compressive stress to the film after releasing the pre-strain, thus resulting in the formation of wavy structures.[62] Detailed experimental and theoretical studies had been conducted to understand the mechanism for the formation and characteristics of the buckling structure.[63-70] The typical process is shown in Figure 2a, which demonstrates the fabrication of wavy Si ribbons on a PDMS substrate.[71] Arrays of silicon ribbons are fabricated by selective etching on a Si-on-insulator (SOI) wafer coated with a ribbon structured resist layer via photolithography. After removing the photoresist and etching the SiO_2 layer, the released Si ribbons are conformally contacted with a pre-stretched PDMS film. Finally, Si ribbon arrays with well-defined waves can be obtained by peeling the PDMS film with the Si ribbons off from the wafer and releasing the pre-strain, which will induce a compression in the ribbons to form the wavy structure. The wavy structure can accommodate mechanical deformations through geometrical changes in the wavelength and amplitude.[71] It can also be further regulated and orientated through chemical and physical modification of the surface of the elastic substrate, leading to intricate and ordered patterns over large areas.[64,72-75]

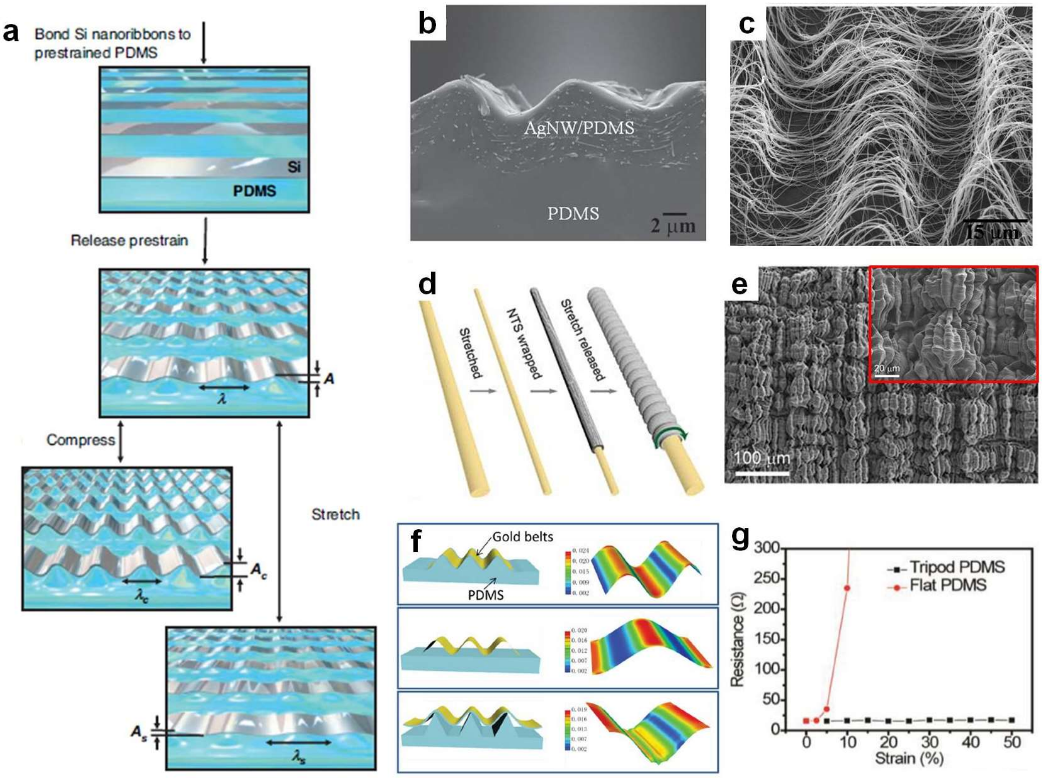


Figure 2. Stretchable conductors with wavy/buckling structures. a) Schematic illustration of the fabrication process for the wavy Si ribbons and the corresponding response of the structure when strain is applied. Reproduced with permission from ref. 71. Copyright 2007 National Academy of

Sciences. b) Cross-sectional SEM image of wavy structure of AgNW embedded in a PDMS surface formed by a strain relaxation process. Reproduced with permission from ref. 76. Copyright 2012 John Wiley and Sons. c) SEM image of the buckling CNT ribbon on PDMS. Reproduced with permission from ref. 77. Copyright 2012 John Wiley and Sons. d) Fabrication process of the sheath-core conducting fiber and e) SEM images of the buckling structure of the sheath-core conducting fiber subjected to 100% strain. Reproduced with permission from ref. 78. Copyright 2015 the American Association for the Advancement of Science. f) Strain distribution analysis using the FEM method for wrinkled (top), suspending (middle) and tripod (bottom) pre-stretched structures. g) Plotting shows the resistance change of the electrodes with flat or tripod PDMS structures as a function of strain after repeated stretching/relaxing cycles. Reproduced with permission from ref. 79. Copyright 2015 John Wiley and Sons.

The aforementioned strategy was popular and widely utilized for constructing stretchable electrodes with various conductive materials, which are indispensable units for realizing device circuits in flexible devices. Although some rational strategies had been developed to realize wavy structured electrodes, such as the template-based method,[80,81] the strategy based on strain relaxation in an elastic substrate is still dominant in constructing buckling structures due to its universality, versatility, and easy operation. The principle of this method is the deposition and transfer of conductive materials, such as metals,[82-87] carbon materials,[77,88-91] and conductive polymers,[92-95] on a pre-stretched elastic polymers, followed by releasing the strain (Figure 2b,c). For conductive polymers, a typical example would be the wrinkled polypyrrole (PPy) electrodes fabricated through *in situ* deposition of PPy from pyrrole solution onto a prestrained polyurethane elastomer film via chemical reduction.[92] For the fabrication of metal or alloy films on elastic substrates, conventional electronic techniques, such as vacuum evaporation or transfer printing, are still commonly used.[84,85,96] A cost-effective solution-processing approach has also been introduced to deposit Cu on pre-stretched elastic substrates with the surface modification of polyelectrolyte brushes.[87] The fabricated Cu-coated conductive rubber with a buckled structure can reach tensile strain values larger than 300% and a stable conductivity of c.a. 1×10^5 $\mathrm{Scm^{-1}}$. Metal nanostructures are also promising candidates for the construction of buckling films with outstanding conductivity and stretchability.[76,97-100] For instance, ultra-long Ag nanowire-based wavy electrodes, fabricated by transferring a uniform thin film of the AgNW percolation network onto a pre-stretched Ecoflex substrate, can reach 460% strain with a low sheet resistance.[98] The stretchability relies on the combined effect of the deformability of buckled structure and the nano-welded network of ultra-long Ag nanowires (which will be discussed in Section **2.2.2**). In addition to the aforementioned materials, carbon materials such as carbon nanotubes and graphene can also be used to construct stretchable electrodes via the buckling strategy. Prior to forming a coating on pre-strained substrates, the CNTs were engineered into different types of aggregates, such as spray-coated CNTs films[101] or uniformly aligned CNTs films, ribbons and fibers,[77,89,90,102] to realize practical utility in transparent electrodes and mechanosensors. In some cases, nanoparticles were incorporated onto the surface of CNTs to improve the conductivity and functionality of the stretchable electrodes.[77,103-105] Large-scale graphene films can also be fabricated in the buckled structure to accommodate stretching strain for practical usage in stretchable and transparent electrodes. The graphene films were first fabricated by chemical vapor deposition on wafer surface with a thin Ni film as a sacrificial layer.[88] After etching the Ni film with an aqueous iron (III) chloride solution, the graphene films can be transferred onto a pre-strained PDMS film by simple contact methods to realize stretchability.

Beyond the basic wavy structure, hierarchical buckling structures have also been developed to endow better mechanical properties through modification of the aforementioned fabrication process, such as the deposition of functional materials on multiaxially stretched 2D substrates[106-108] or 1D elastic fibers.[78,109,110] Carbon nanotube sheets (NTS) can be wrapped on pre-stretched elastic rubber fibers with orientation in the fiber direction (Figure 2d).[78] After releasing the strain, the resulting NTS sheath structure exhibited periodic and distinct hierarchical buckling in the axial and belt directions (Figure 2e), which can enable ultra-high stretchability (up to 1320%) with a small resistance change (less than 5%). Furthermore, by controlling fabrication parameters, such as the sheath-core structure and the layers of the NTS, the conducting fibers can function as highly sensitive strain sensors and electrically powered torsional muscles.[78] Inspired by various biofilms with hierarchical 3D architectures regulated via mechanical/biochemical coupling,[111-114] such as the cerebral cortex of mammalian brains[115] and fingerprints,[116] a freestanding and conductive PPy film with a hierarchical wrinkled structure has been fabricated via chemical oxidation polymerization of pyrrole on a pre-stretched PDMS film, followed by strain release and substrate removal.[94] The obtained wrinkled PPy films with two-scale wavelengths contain intertwined stripes (λ~ 0.65 μm) and periodical labyrinth patterns (λ~ 3.48 μm), which can be attributed to the self-reinforcing effect and tuned by synthesis parameters, such as the modulus of the substrate and film thickness (controlled by reaction time). Although the wrinkled structure provides good stretchability through out-of-plane bending, the large strain concentration at the peaks and valleys of the buckled films and the moduli-mismatch between the conductive material and the elastic substrate would limit its stretchability, mechanical stability and conductive performance during mechanical deformations.[66] To overcome these difficulties, a suspended wavy conductor was developed through rational structural design of the substrate surface.[79] Gold belts were transferred on a pre-strained PDMS substrate with a unique out-of-plane tripod structure that provides enough space to facilitate the shape change of the nanobelts. Upon releasing the pre-strain, wavy gold belts would be formed, which are characterized by a smooth sinusoidal structure without sharp deformations. These gold belts would be suspended rather than directly attached to the PDMS substrate (Figure 2f), which would significantly reduce the stress caused by physical contact. The resulting stretchable electrodes exhibit large stretchability of 130% and good stability (> 10000 cycles) without any obvious change in resistance during stretching (Figure 2g).

Beside electrodes, various electronic materials with high moduli, including polymeric and inorganic semiconductors,[73,74,117-119] can also be adapted for this suspended buckling strategy to realize stretchability while maintaining electronic functions to construct stretchable devices, such as energy harvesting devices,[95,120-123] sensors,[78,101,118,124] energy storage device,[106,125-131] and information storage devices.[132,133] Using the aforementioned suspended wavy structure, the same group developed stretchable supercapacitors based on buckled graphene films, which can maintain energy storage performance during strain (Figure 3a-d).[129] A stretchable piezonanogenerator has also been developed with the piezoelectric ceramic lead zirconate titanate (PZT, $Pb[Zr_{0.52}Ti_{0.48}]O_3$), which exhibits an outstanding piezoelectric coefficient but a high modulus of 49 GPa and a maximum tensile strain of 0.2%.[134,135] The fabrication process involves the patterning of PZT ribbons on a magnesium oxide (MgO) layer before being transferred onto a pre-stretched PDMS film by etching with phosphoric acid to obtain the buckled structure (Figure 3e).[121] The resultant piezonanogenerator can not only accommodate physical strain but also enhance the piezoelectric effect by up to 70% (Figure 3f,g). This can be attributed to the large location-dependent strain gradient on the wavy ribbons and the reduction of substrate clamping in the elevated buckles.[136] Similarly, a stretchable memory device has been developed based on buckled poly(methyl methacrylate): poly(3-butylthiophene) (P3BT:PMMA) and graphene film.[132] P3BT:PMMA was first spin-coated on a conductive graphene/copper foil to obtain a freestanding hybrid film upon etching of the copper substrate. After transferring the P3BT:PMMA/graphene film onto the pre-strained PDMS substrate and thermally depositing the top electrodes, the fabricated buckled memory device showed stable resistance switching performance even under up to 50% strain (Figure 3h-j).

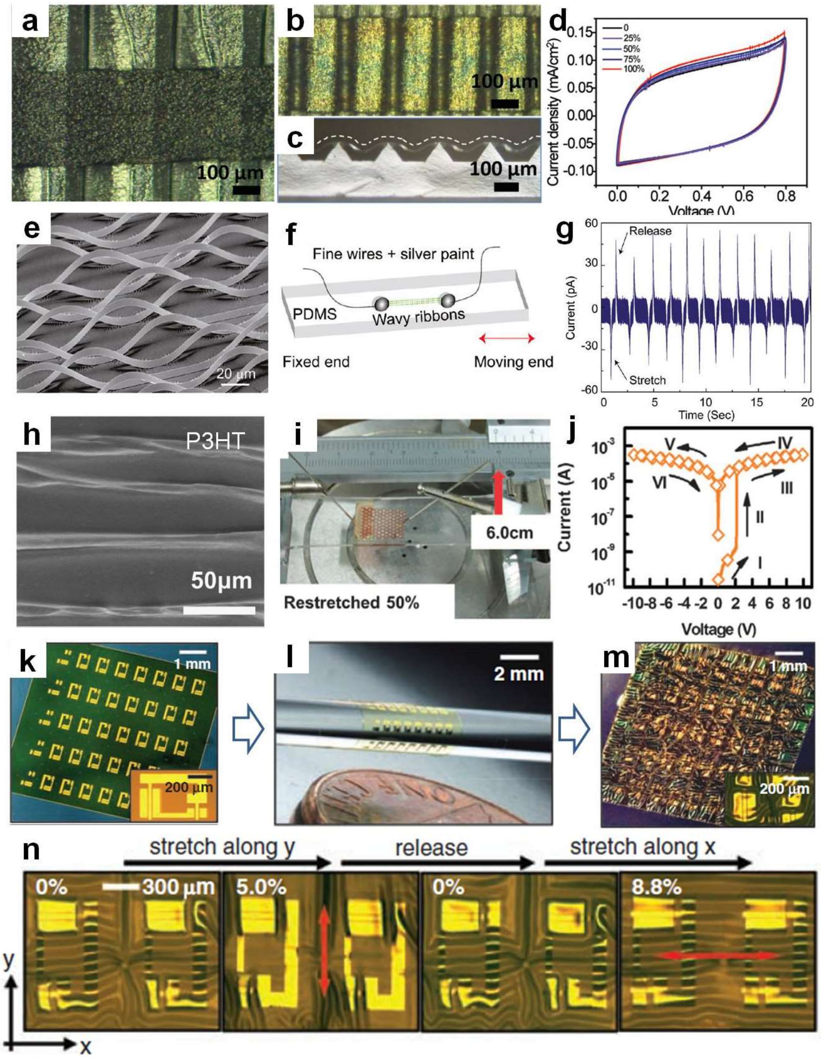


Figure 3. Stretchable devices based on wavy/buckling structures. Digital photographs of a graphene microribbon-based microsupercapacitor in a) stretched state (100% strain), b) relaxed state and c) cross-sectional image. d) Cyclic voltammetry characterization of the microsupercapacitor at various strains. Reproduced with permission from ref. 129. Copyright 2015 John Wiley and Sons. e) SEM imaging of the PZT ribbon buckling structure. f) Schematic illustration of the energy harvester using buckled PZT ribbon and g) the current flow generated by stretching the lead zirconate titanate (PZT) ribbons under periodic stretching and relaxing cycles. Reproduced from ref. 121. Copyright 2011 American Chemical Society. h) SEM image of the

stretchable memory device with a wrinkle structure using a polymer complex as active layer. i) Optical image and j) resistance switching behavior of the memory device at 50% strain. Reproduced with permission from ref. 132. Copyright 2014 Nature Publishing Group. Optical images of flexible silicon integrated circuits k) on a carrier substrate (inset: magnified imaging of a single CMOS inverter), l) wrapped on a thin rod, m) on PDMS. n) Images of stretching the CMOS inverter in x or y directions. Reproduced with permission from ref. 137. Copyright 2008 the American Association for the Advancement of Science.

Beyond the individual devices, flexible and wearable electronics for health monitoring and biomedical treatments require not only outstanding mechanical deformability but also integrated electronic functions, which include sensing, actuating, information processing and communication. Thus, strategies have been developed to transfer intricately designed circuits comprising electrodes and functional units onto pre-stretched substrates to achieve high stretchablility.[137-139] For example, metal-oxide semiconductor field effect transistors (MOSFETs), CMOS logic gates, ring oscillators, and differential amplifiers can be integrated to construct a stretchable and foldable circuit exhibiting electrical performances comparable with those of conventional systems built on wafers (Figure 3k-m).[137] To obtain the buckling structural configurations, planar layouts comprising aligned arrays of nanoribbons of single crystalline silicon and patterned metal electrodes were fabricated on a sacrificial layer, and the resultant ultrathin flexible CMOS circuit are released and transferred onto a pre-stretched PDMS film. The resulting stretchable integrated circuit inverter shows reliable performances under various tensile and uniaxial applied strains (Figure 3n). Although a great extent of the flexibility of materials is necessary and required, the advantage of the buckling strategy exhibiting easy integration with conventional manufacturing methods and simple transferring process make it a valuable and widely used technique in obtaining flexible and stretchable electronic modules.

Other 3D structures have also been designed to endow devices with deformability and stretchability to maintain performance under mechanical strain. Functional devices, such as lithium-ion batteries (LIB), which comprise brittle materials and delicate planar structures that are difficult to be replaced or modified into spatial structures, would have to be designed via a strategy of paper folding (origami) and cutting (kirigami) to achieve system-level deformability. An approach has been developed based on origami, an ancient art of paper folding,[140] to transform two-dimensional device sheets into compact and deformable three-dimensional architectures though high degrees of folding along designed creases, thus enabling high mechanical deformability on the system level, including folding, unfolding, twisting and bending.[141-147] Using this approach, conventional planar LIBs (Figure 4a) were first constructed by layered materials of current collectors, anode, cathode, separator and packaging. Next, stretchable LIBs were realized by folding these layers into two specific origami patterns according to the difference in angles between adjacent creases such that the LIBs are almost completely compressible in one direction (Figure 4b) or collapsible in two directions (Figure 4c). Thus, the strain caused by deformations can be released by the folding and unfolding processes of the creases, while the faces remained in rigid configurations and thus maintained the device performance.[148]

Similarly, great stretchability can also be achieved with architectures via kirigami patterns, which is another form of paper art that involves cutting and folding.[149,150] For example, a cut-N-shear pattern constructed by designed cutting and folding methods can accommodate applied strain by rotations occurring at the cuts (Figure 4d). The deformability mechanism of the LIBs relies on rotations at the cuts. With this design, LIBs produced according to battery manufacturing standards can accommodate over 150% strain while maintaining the energy storage function.[151] Other designs with different kirigami patterns in planar sheets have also been developed to exhibit remarkable deformability by strain-induced out-of-plane deformations (Figure 4e-h).[28,152-156] This strategy, which integrates the art of paper folding and cutting with materials science and functional devices, provides a valuable and instructive paradigm to construct 3D architectures for flexible electronics with exceptional mechanical characteristics and functionalities.

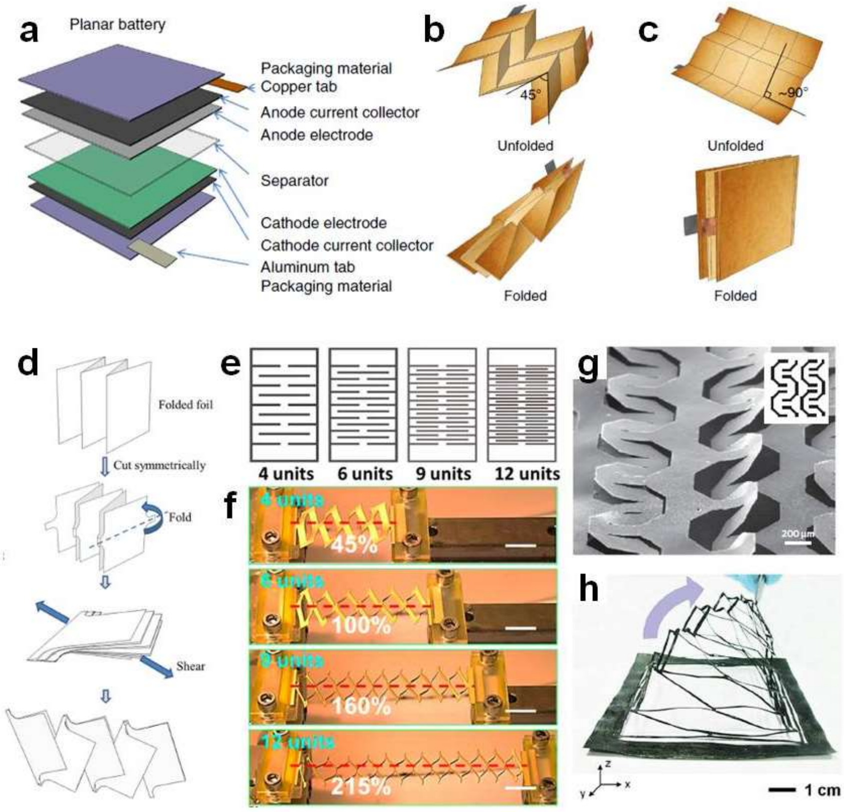


Figure 4. Stretchable conductor and energy storage devices based on origami or kirigami designs. Schematic illustrations of a) a planar lithium-ion battery with multilayer structures and b,c) two different origami lithium-ion battery configurations. Reproduced with permission from ref. 148. Copyright 2014 Nature Publishing Group. d) Illustration of a cut-N-shear kirigami pattern. Reproduced with permission from ref. 151. Copyright 2015 Nature Publishing Group. e) Design of kirigami-based electrodes for supercapacitors with different geometries and f) the corresponding stretching behaviors. Reproduced from ref. 28. Copyright 2016 American Chemical Society. g) SEM image of microscale kirigami patterns formed in GO-PVA nanocomposites. Reproduced with permission from ref. 152. Copyright 2015 Nature Publishing Group. h) Optical image of a

pyramid-shaped CNT film with outstanding out-of-plane deformability designed for a three-dimensionally stretchable supercapacitor. Reproduced with permission from ref. 154. Copyright 2016 Royal Society of Chemistry.

A coil spring is an elastic object that can store mechanical energy through compression or stretching which is widely used in our daily life. For its structural characteristics, the helically coiled spring configuration was introduced in constructing stretchable devices.[157-162] Since 2013, Peng's group has developed stretchable, fiber-shaped energy storage devices based on the coiled spring structure.[163-168] The key concept involves the wrapping of aligned carbon nanotube (CNT) sheets, which serve as electrodes and functional units, on an elastic fiber, which would confer stretchability in the resulting supercapacitors. The spring-like structure of the fibers wrapped with CNT sheet electrodes can maintain the aligned structure and capacitance during stretching.[165] A stretchable lithium-ion battery can be realized by wrapping two types of fibers on a PDMS fiber in the coiled spring shape as anode and cathode (Figure 5a). The anodic fiber comprises CNT sheets incorporated with $Li_4Ti_5O_{12}$ (LTO)), while the cathodic fiber consists of $LiMn_2O_4$ (LMO).[164] Several CNT fibers can also be overtwisted together to form a hierarchical spring-like fiber with coiled loops aligning along the fiber axis (Figure 5b,c) such that a freestanding elastic LIB can be achieved without the elastic fiber core, with two spring-like fibers serving as anode and cathode. Such LIBs can maintain specific capacities at 100% strain.[163] In addition to spring structures on the nano or micro scale, macrostructures with helical or spiral shapes have also been introduced in stretchable device fabrication, which show good compatibility with existing techniques (Figure 5d).[169-171] For example, by engineering a PDMS substrate into spring structure with a straw as the template, the stretchability of a conductive Cu nanowire film coated on the PDMS can be dramatically enhanced (Figure 5e).[169] Considering that fiber-shaped devices may supplement the missed function in MEMS while at the same time extending stretchability, this coiled spring design provides a general inspiration in constructing flexible and stretchable fiber-shaped devices.

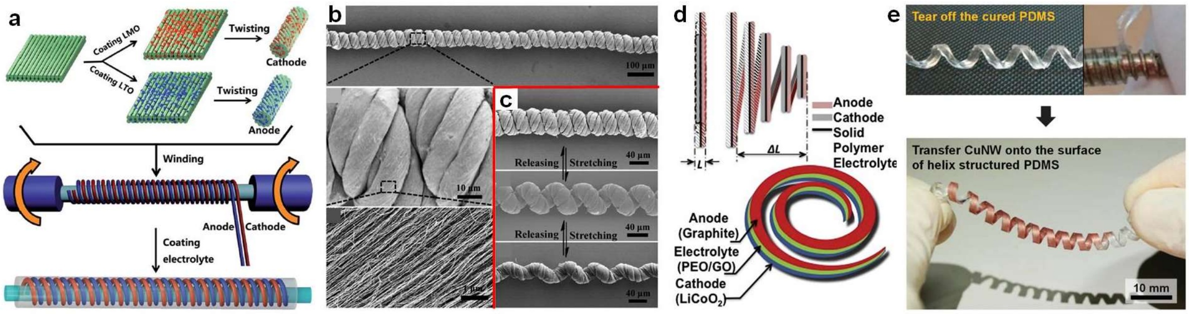


Figure 5. Stretchable conductors and devices with coil spring structures. a) Fabrication of a CNT-based composite fiber with a spring-like structure as cathodes and anodes. Reproduced with permission from ref. 164. Copyright 2014 Royal Society of Chemistry. b) SEM images of a CNT fiber with a coil spring configuration. c) SEM imaging of the fiber at different stretching states. Reproduced with permission from ref. 163. Copyright 2014 John Wiley and Sons. d) Schematic illustration of a lithium-ion battery with a spiral configuration. Reproduced with permission from ref. 170. Copyright 2016 Elsevier. e) Fabrication process of a CuNW electrode with a helical structure. Reproduced with permission from ref. 169. Copyright 2014 Nature Publishing Group.

Porous structures with flexible materials can accommodated strain via a geometric change of volume involving a soft skeleton, such as in networks,[172,173] textiles[174-179] and sponge structures,[180-187] which have also been introduced for endowing the functional materials and substrates with flexibility and stretchability for realizing flexible devices. A stretchable pressure and thermal sensor can be achieved through the incorporation of a network structure design based on a non-stretchable polyimide and poly(ethylenenaphthalate) film as the substrate and a conductive rubber as the sensing unit (Figure 6a,b).[173] Using a sugar cube as the template, a 3D PDMS sponge exhibiting compressibility and stretchability can be obtained by the facile drop-casting method (Figure 6c-f). By filling electrode materials, such as active materials ($LiFePO_4$ (LFP) and LTO), carbon black, and polyvinylidene fluoride binder in the sponge-like PDMS scaffolds, the obtained

3D porous LIBs can accommodate a large stretching strain of 80% while maintaining good energy storage performance.[185]

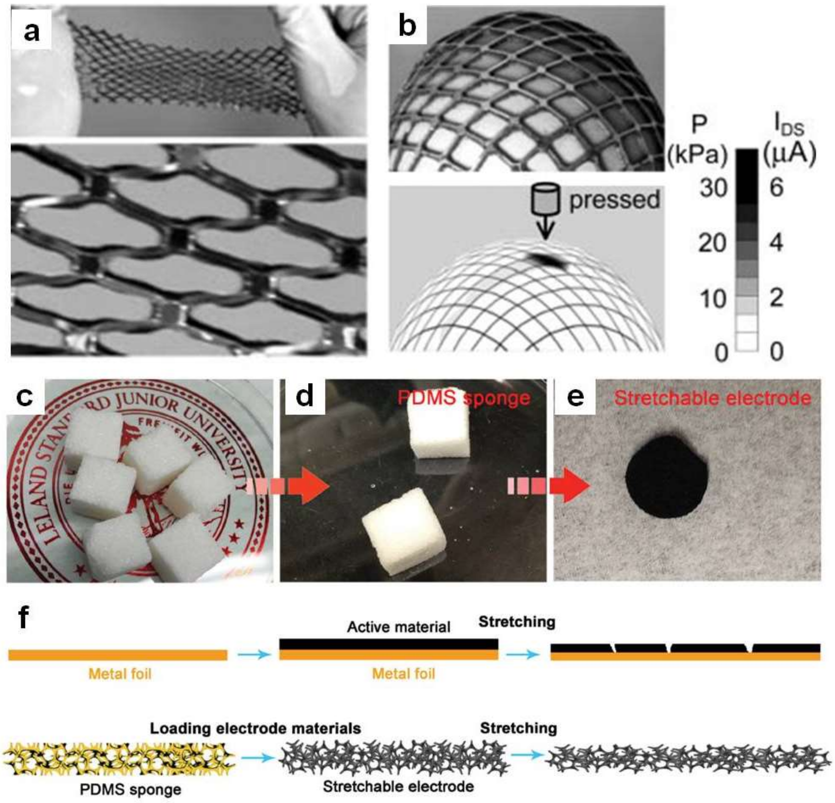


Figure 6. Stretchable devices with porous structures. Optical images of a) a pressure sensor with a network structure based on an organic semiconductor as well as b) the pressure sensor network mounting on an egg and the corresponding pressure distribution. Reproduced with permission from ref. 173. Copyright 2005 National Academy of Sciences. Photographs of c) sugar cubes, d) PDMS sponges prepared by using the sugar cube as a template and e) the stretchable electrode produced by filling the PDMS sponge with active materials. f) Illustration comparing the stretching behavior between a conventional metal foil-based electrode and the PDMS sponge-based electrode. Reproduced with permission from ref. 185. Copyright 2016 John Wiley and Sons.

2.2 Stretchable design with 2D structure

Metal electrodes constitute one of the essential units in circuit construction, be it in CMOS systems or flexible and stretchable devices. However, well-processed metal electrodes with flat continuous structures on elastic substrates, which are widely used in traditional circuit design, exhibit poor sustainability on elastic substrates under external strain.[188-191] Thus, with rational design, patterned electrode structures in 2D shapes will be an efficient solution for constructing stretchable circuits. Unlike the aforementioned buckled structure in Section **2.1**, in which the undulations are added in the z-direction, 2D patterned electrodes can easily enable the building of planar circuit structures due to their compatibility with traditional well-developed techniques, such as transfer printing,[192-196] screen printing,[96,197,198] lithography, patterning,[88,188,199,200] among others.

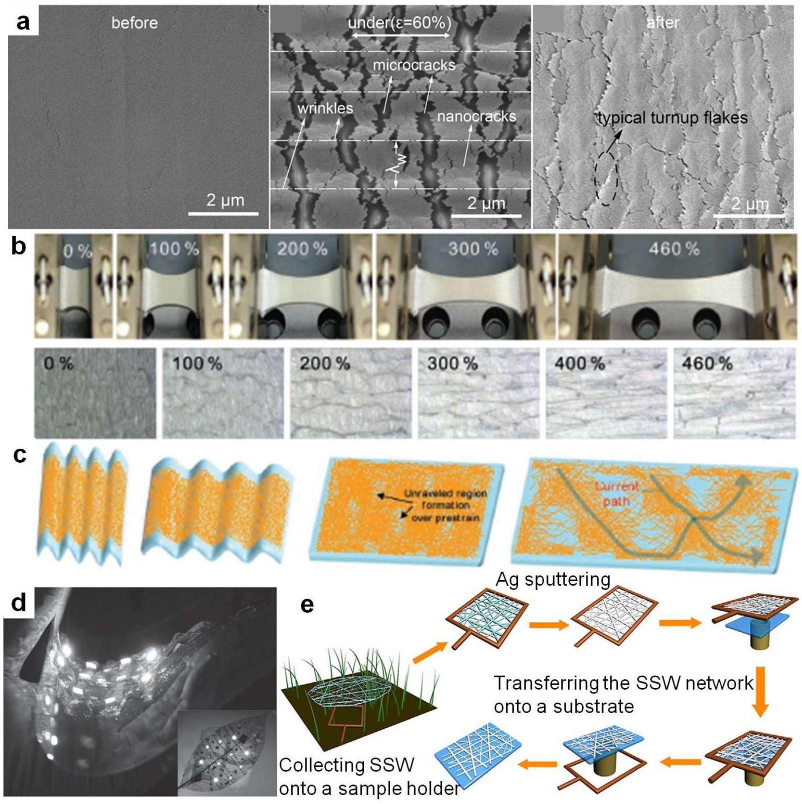


Figure 7. Crack-based stretchable conductors. a) SEM images of gold films with micro/nano-cracks on a PDMS substrate before, during and after stretching. Reproduced from ref. 201. Copyright 2014 American Chemical Society. b) Photographs and microscopic images of an ultra-long Ag NW electrode on pre-strained Ecoflex substrate under various stretching states. c) Illustration of the mechanism of the electrode maintaining conductivity during stretching. Reproduced with permission from ref. 98. Copyright 2012 John Wiley and Sons. d) Optical images

of flexible LED arrays fabricated by leaf vein-based transparent electrodes. Reproduced with permission from ref. 202. Copyright 2015 John Wiley and Sons. e) Illustration of a stretchable silver network fabricated by metalizing a silk spider's web. Reproduced with permission from ref. 203. Copyright 2014 Nature Publishing Group.

2.2.1 Crack-based stretchable electrodes

Metal films, especially gold films, have been widely used as electrodes in the fabrication of conventional planar circuits. To extend their applications to flexible and stretchable electronics, the microcrack structure was introduced to improve the accommodation of physical deformations in these metal films.[83,85,204] While applying external strain, randomly patterned cracks ranging from the nano or micro scale will form in the conductive film to release the stress (figure 7a), while the continuous pieces of conductive materials will work as a percolating pathway to maintain electrical conductivity.[38,201,205] The morphology of the Au film fabricated via thermal or electron beam evaporation can be controlled by evaporation parameters to obtain a microcrack structure, such as the gold thickness, deposition temperature, elastic modulus of substrate, adhesion layer thickness, surface properties of the substrate (for example, O_2 plasma treatment), and so on.[206-210] Thus, by adjusting the gold and the elastic substrate, stretchable electrodes with large-scale reversible elasticity can be achieved.[211,212] This strategy can also be used for films formed by other conductive materials, including other metals[213-215] and conductive polymers[216-219] (such as PEDOT:PSS, P3HT), via modifying the adhesion on elastic substrates. Moreover, directly structuring the surface of conductive films can be another approach to generate cracks during applying strain.[220,221] For example, existing natural structural surfaces would work as effective templates to help fabricate crack-based stretchable electrodes. By a one-step soft lithography replication process, petals of yellow roses containing pentagonal and hexagonal micropapillae can be used as molds to generate a PDMS film with biological topographic structures on the surface, showing continuous 3D microscale crater-like architectures.[220] When followed by polymer-assisted deposition of copper thin film on top,[222-224] the resulting electrodes showed remarkable stretchability, which can be attributed to the space-confined formation of cracks in the valley that can accommodate applied strain, while the sharp ridges would maintain the conductive pathway.

2.2.2 Networks of 1D conductors

Natural network systems provide excellent mechanical properties and functionalities.[225-227] For example, spider webs show a combination of flexibility and roughness,[26] while leaf venation and river systems exhibit efficient mass transportation,[225,226,228,229] all of which inspire researchers to fabricate nanomaterials into networks with mechanical accommodation and electronic conductivity for applications of stretchable electrodes.[230-233] A simple approach for constructing conductive networks is to directly spread or embed 1D conductive materials, including carbon nanotubes or nanofibers,[105,234-236] metal nanowires[98,237-242] and polymer fibers,[243,244] on the surface of or in elastic substrates, generating random network structures via dense connections among long nanostructures. Upon applying external strain, the random networks of 1D conductors can release the stress by geometrical deformation without physically breaking, while the long 1D nanostructures bridged with each other will retain the electrical conductivity via percolation transport pathways (figure 7b,c).[245-247] The conductivity and deformability of the random networks rely on the length of the 1D conductor,[98,242] the density of nanostructures,[248,249] the interaction in the junctions,[238,245,250] and the adhesion between networks and substrates.[241,251] By optimizing these parameters, highly stretchable electrodes with low resistance and excellent stability can be realized.[98,250] To further enhance the deformability of conductor networks, buckled 1D nanostructures were introduced in the formation of network structures, which involves the aforementioned buckled strategy.[101,252] By transferring a network of Au-coated electrospun PVP nanofibers onto a biaxially pre-strained PDMS substrate, the network of in-plane buckled nanofibers was formed after releasing the prestrain, which can accommodate mechanical strain by straightening and geometrically changing the network structures.[252] Moreover, network structures usually exhibit good optical transparency, which makes these 1D material-based electrodes suitable for applications in transparent circuits and devices.[253-255] Beside random patterns, networks with regular morphologies were designed and utilized for stretchable electrodes, including aligned structures[248,256-258] and nanomeshes with rhombic or honeycomb structures.[259-261] For example, carbon nanotubes patterned into parallel or vertical alignment show good accommodation of incoming strain, which is based on the conductive networks formed by the connecting bundles of carbon nanotubes.[262,263] Moreover, conductive films with other mechanical behaviors, including high strength and toughness, can also be realized by capillary splicing of aligned carbon nanotubes into nano-architecture textiles.[264]

Beyond various networks formed by interconnected 1D conductors on elastic substrates, natural network systems can also provide inspiration and directly participate in fabrication towards the achievement of highly stretchable electrodes.[202,203] For example, leaf venation constitutes a network structure for the efficient delivery of nutrients in leaves under low lighting.[226,228] Based on evolution over a long time, natural optimization processes make leaf venations exhibit a quasi-fractal structure, which is quite adaptable for building flexible and transparent electrodes.[225,227,229,265] Several methods have already been developed to fabricate conductive networks using the leaf venation pattern, such as direct physical deposition of metals on an etched leaf venation as electrodes, polymer-assisted metal deposition on veins (figure 7d), and replication of the network structure by stamp printing or photolithography with the leaf venation as a template.[202,203] Additionally, silk spider webs, another common natural network system, have also been used to fabricate conductive electrodes with high transparency and stretchability by simply forming a metal coating and subsequently transferring it onto an elastic substrate (figure 7e).[203]

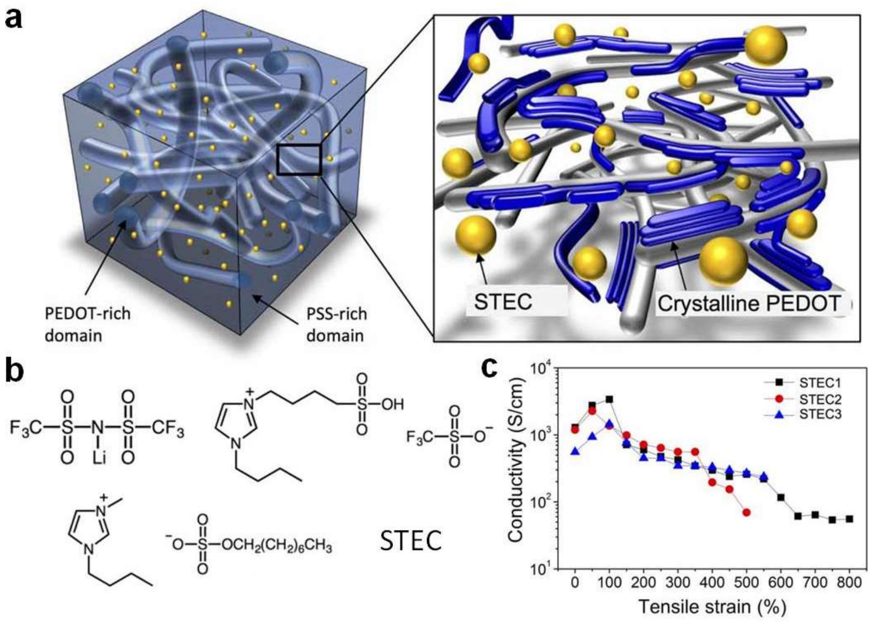


Figure 8. Stretchable conductor based on a 3D network structure. a) Illustration of a stretchable PEDOT film incorporated with a stretchability and electrical conductivity (STEC) enhancer. b) Chemical structures of typical STEC enhancers. c) The conductivity change under various strains of a PEDOT film doped with different STEC enhancers. Reproduced with permission from ref. 266. Copyright 2017 the American Association for the Advancement of Science.

This design has been extended to the spatial engineering of 3D networks based on electronic material fibers for improving strain tolerance.[266-269] For example, a method of chemically induced structure operation has been developed to endow PEDOT:PSS films, one of most widely used conductive polymers with a fracture strain of only 5%,[217] with high stretchability through chemically induced structure operation (Figure 8a).[266] By doping with ionic additive-assisted stretchability and electrical conductivity (STEC) enhancers, the obtained PEDOT:PSS films exhibit a combination of high conductivity and stretchability, with higher than 4100 S/cm under 100% strain and a fracture strain of 800% (Figure 8b,c).[266] The high stretchability and enhanced conductivity can be attributed to the morphological change of PEDOT, which leads to increased crystallinity in the PEDOT region and more interconnected nanofibrillar networks in the matrix. At the same time, the charge screening effect of the ionic STEC additives in the more disordered regions of the film further softens the material.

2.2.3 Structural pattern of electrodes

Unlike the patterned structure of conductive nanomaterials discussed above, rational and delicate design in the geometry of conventional electrodes has also been developed and studied by calculation and simulation, showing outstanding performance in deformability and compatibility in constructing flexible electronic circuits (figure 9a,c).[190,270-273] A typical strategy is the 2D spring-shaped structure inspired by the common helical spring, which had been designed to realize stretchable electrodes in 2004.[188] Spring-shaped gold wires with 2D oscillations were fabricated by standard lithography techniques and embedded in PDMS. Compared with traditional straight electrodes, the stretchability of these structural electrodes can reach a relatively high strain, which can be modulated by controlling the geometric parameters of the spring structure, including wire width, amplitude, wavelength, and so on.[274-277] Additionally, systematical calculations and simulations of the mechanical properties of this structure have been deeply studied to optimize its ability in accommodating deformations.[276,278-281] Compared with the crack-based electrodes, the gold bridge with a spring-shaped structure can be further stretched with a minimal change in the conductivity because deformability of the twist can be used to accommodate larger strain without obvious physical damage on the conductive stripe (figure 9b).[279,282] Another advantage of this 2D spring shaped design is that the fabrication process can be easily involved in constructing various planar layouts by conventional and well-developed techniques, such as photolithography,[282,283] printing,[96,284-286] direct writing,[287-290] and so on. As a universal strategy, other conductive materials, including metals,[274,291] metal nanostructures,[292] polymers,[96] carbon materials[285,293,294] and metal oxides,[277] have also been manufactured into this stretchable structure.

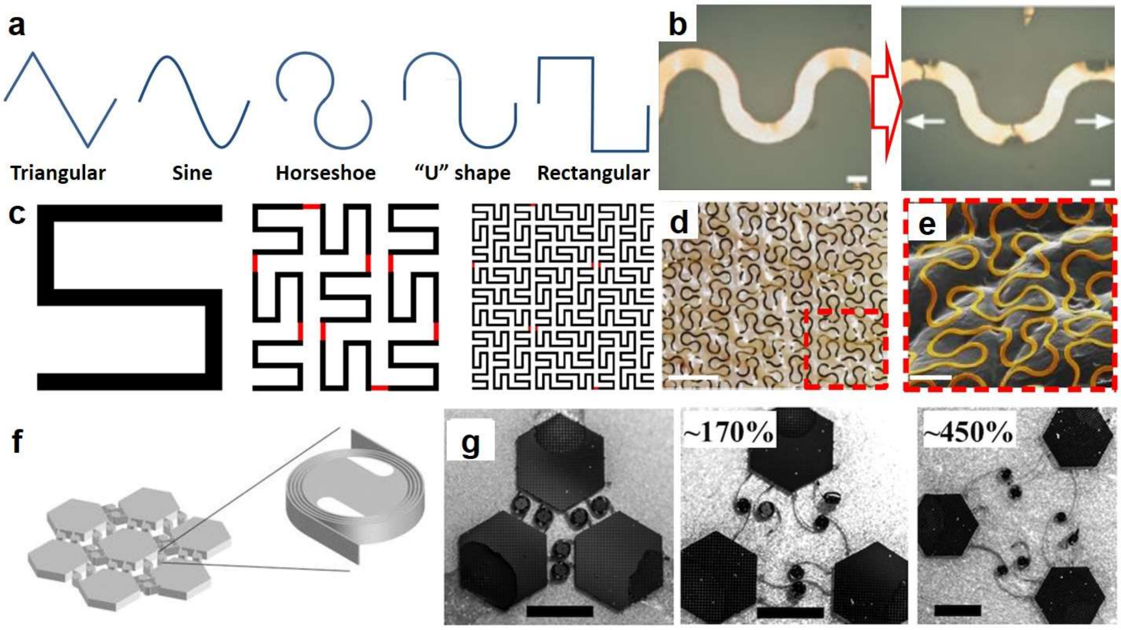


Figure 9. Stretchable conductors based on 2D structures. a) Various 2D shapes for stretchable electrodes. b) Microscope images of gold wire on a PDMS substrate with a 2D spring structure before and during stretching. Scale bar: 100 µm. Reproduced with permission from ref. 188. Copyright 2004 John Wiley and Sons. c) Illustration of 2D Peano curves in different iterations. d) Digital photograph and e) SEM images of Peano-patterned metal wires attached to skin and a skin replica. Scale bar: d) 2 mm, e) 500 µm. Reproduced with permission from ref. 272. Copyright 2014 Nature Publishing Group. f,g) Illustration images and optical images of a silicon-based array of islands connected by spiral springs. Scale bar: 1 mm. Reproduced with permission from ref. 295. Copyright 2014 AIP Publishing LLC.

Based on the different geometrical structures of the twists, various architectures of 2D spring-shaped electrodes have been designed and simulated for accommodating mechanical strain, including elliptical, “U” shape, horseshoe, trapezoidal, triangular, and so on (figure 9a).[188,278,291,296-298] John Rogers’ group has deeply investigated this type of electrode (also known as the serpentine structure) and opened wide applications in constructing deformable devices and circuits.[48,51,56] Based on the serpentine structure, even integrated modules composed of functional electronic units have also been realized, which will be discussed in the following sections.

The mathematical concept of the fractal space-filling curve, with the key feature of "self-similarity", is a continuous curve filled in a 2D plane or 3D space through periodically repeated structures (figure 9c).[299] In 2013, the Rogers group introduced this concept in the fabrication of stretchable electrodes to improve the serpentine structure for high system level stretchability and low interconnect resistance.[300,301] Due to its outstanding mechanical deformability and high areal capacity, this strategy has already been extended into various applications, such as heating, mechanical sensing, health monitoring, among others.[297,302-306] A typical fractal electrode shown in the figure contains repeated spring-shaped motifs at multiple length scales (figure 9c-e).[272] By increasing the fractal order of the space filling structure, the electrodes with this design can reach even higher stretchability values relative to that of the serpentine structured strategy.[272,307] For example, the ‘half-and-half’ Peano layout, containing spring-like gold stripes as unit cells with alternating orientations, exhibits stretchability values along the x- and y-axis of 16% and 13%, respectively, while the third order layouts will yield 32% and 28%.[272] Based on rationally optimized hierarchical structure, the maximum principal strain of a computational model with a 4th order fractal structure can reach as high as 2300%.[307] This improvement of the stretchability in a high-order fractal structure relies on the process of ordered unraveling, which can be attributed to the geometric scaling of the arc sections and an increase in the length of the electrodes.[308-311] In addition to this spring-shaped structure, other regular 2D structures have also been designed to engineer stretchable electrodes, such as spiral-based designs (figure 9f,g),[295,312-314] which show ultra-stretchability (approximately 1000 %) based on the unwrapping of the coiled spiral-shaped arms.[314]

2.3 Stretchable circuits with "rigid and soft" hybrid structures

Although significant efforts had been made to design structures that endow traditional electronic materials with stretchability and flexibility, many functional devices containing brittle active materials and delicate structures with susceptibility to shape change still remain incompatible with these strategies. Thus, an engineering concept was introduced to achieve the integration of high performance and large stretchability by technically combining "rigid" and "soft" electronic modules in a single circuit design. The essential strategy is to spatially separate a stretchable chip into two types of domains: one is mechanical neutral plane for brittle and functional modules; the other is flexible and deformable connection of the stretchable conductor.

2.3.1 Island and interconnection design

Via patterning or buckling strategies, separately patterned rigid device islands can be connected by stretchable electrodes to realize stretchable circuits forming integrated systems. Typical strategies are shown in figure 10, in which the stretchable interconnections are constructed by aforementioned structural electrodes with out of plane or in plane designs, including buckling,[315] serpentine[316] and fractal structures.[300] Meanwhile, non-stretchable functional devices, which are usually fabricated by conventional materials with excellent electronic performance, would be connected by these stretchable electrodes. During stretching, the "soft" interconnections with in-plane or out-of-plane structures can accommodate the applied strains, while the relatively "rigid" islands would effectively isolate the active but brittle components from physical deformations that could lead to function failure.[83,317-321]

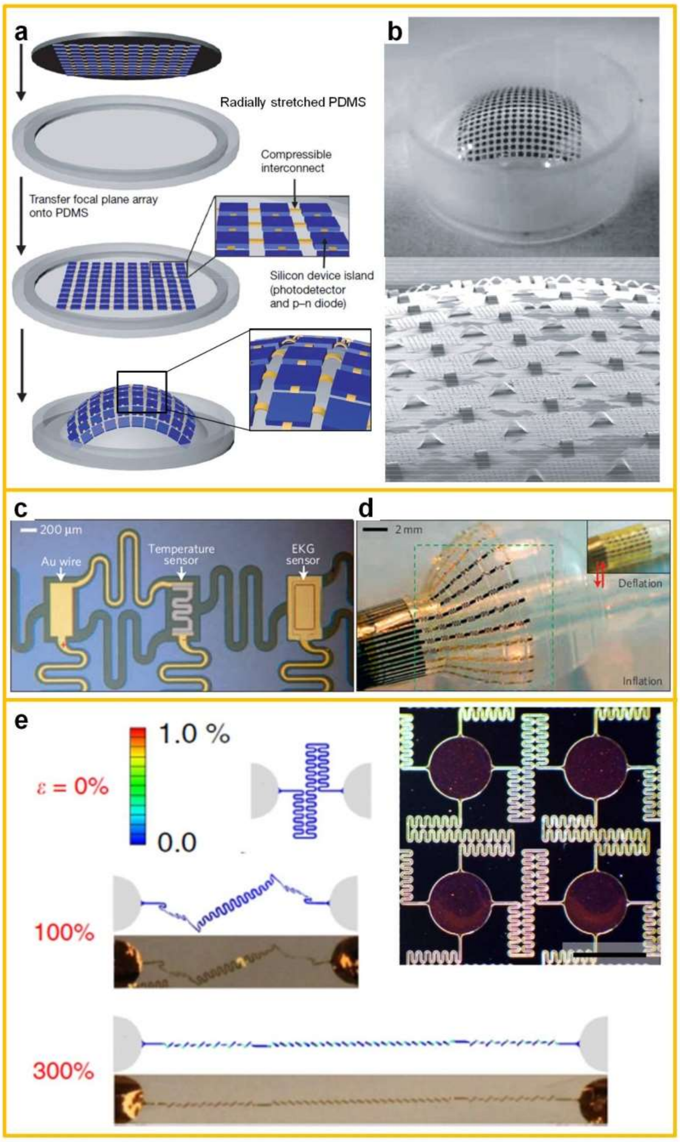


Figure 10. Stretchable devices with the interconnection design. a) Illustration of the fabrication process for hemispherical eye cameras. b) Optical and SEM images of the hemispherical PDMS transfer component with compressible interconnections. Reproduced with permission from ref. 315. Copyright 2008 Nature Publishing Group. c) Magnified optical image of the integration of various sensors in a circuit with serpentine interconnection d) Images of the integrated circuit mounted onto a balloon catheter in both inflated and deflated (inset) states. Reproduced with

permission from ref. 316. Copyright 2011 Nature Publishing Group. e) Comparison of experiment and simulation for the stretching behaviors of the circuit with serpentine interconnections designed for lithium-ion battery. Inset shows the image of copper pad electrodes with interconnects. Scale bar: 2 mm. Reproduced with permission from ref. 300. Copyright 2013 Nature Publishing Group.

The device configuration of non-stretchable electronics on flexible or conventional rigid substrates is typically based on well-established technologies with planar natures, including patterning, etching, transfer printing, materials deposition and growth.[9,192,199,322-328] Due to the adaptability with rational island and interconnection design, these methods are still widely used in fabricating stretchable circuits. First, a two-dimensional configuration of patterned device islands connected by structural electrodes fabricated on a planar substrate. Next, stretchable circuits can be realized by transferring the planar layouts onto a pre-stretched elastic substrate, forming a deformable "islands and interconnections" structure to accommodate complex deformations on a curvilinear surface or under applied strain.[293,329-331] Figure 10a presents a typical fabrication process of an island-interconnected system comprising stripe-shaped metal electrodes and a square array of CMOS devices.[315] After fabricating an array of single-crystalline silicon photodiodes and current-blocking p–n junction diodes connected by metal (chromium–gold–chromium) interconnects on a planar substrate, the matrix layout was then transferred on a radically stretched PDMS substrate. The resulting buckled electrodes between the device with islands show good tolerance to deformations on curvilinear surfaces (Figure 10b). Through the combination of imaging optics and hemispherical housings, the as-fabricated curvilinear optoelectronics would function as electronic eye imagers that are comparable to the human eye.[315]

To further enhance the accommodation to large strain and curvilinear deformation, serpentine structured electrodes can be introduced as interconnectors, which show even larger stretchability than that of the straight bridge design due to the deformability of the large twist structures.[286,298,332-337] An example shown in Figure 10c,d is an “island and interconnection” design with microscale light-emitting diodes, sensor electrodes, temperature detectors and other components connected by gold electrodes of the serpentine structure. The design exhibited good compliance with the soft, curvilinear surfaces of the body, as well as reversible stretchability with reliable electronic performance.[316] Moreover, circuit designs containing serpentine structures can be transferred on a pre-stretched substrate to obtain buckled serpentine interconnections with enhanced deformability, which show two types of strategies for accommodating strain via out-of-plan and in-plane architectures.[338-340]

Furthermore, the introduction of the fractal structure to the interconnections can efficiently improve the mechanical stretchability while maintaining the areal capacity of active devices. For example, by connecting a rechargeable lithium ion battery array with fractal interconnection structures, a stretchable battery can be realized with stretchability up to 300% (Figure 10e), while maintaining capacity densities of ~1.1 mAhcm^{-2}.[300] This strategy of combining the conventional well-established device units with stretchable conductive pathways has opened up a new methodology for realizing stretchable electronics that can accommodate nearly any type of mechanical deformations while retaining high performance.

2.3.2 Mechanical hybrid substrate

Although the ‘island and interconnection’ design has efficiently extended the opportunities for rigid electronic modules involved in stretchable circuits, this combination of “rigid” and “soft” design based on functional materials for device performance often suffers from large strain concentrations at the rigid-to-soft transition zones, which could result in mechanical failure and thus limit the long-term performance of the stretchable circuit.[270,341]

Due to this limitation, another strategy has been developed to engineer the elastic substrate into a mechanically heterogeneous structure comprising patterned rigid and soft domains for fabricating electronic circuits with systematical stretchability. The key strategy is to locate the electronic devices with delicate structures and brittle materials on top of rigid device islands that can protect them from physical damage. Meanwhile, the elastic domains among the device islands serve as stretchable interconnections to accommodate the applied strain.[341-344] A typical process consists of embedding SU-8 photoresist platforms (E = 4 GPa) in a PDMS substrate (E = 1 MPa) as device islands, and brittle materials patterned on the PDMS surface above the SU-8 platforms will not suffer physical damage during stretching, thus enabling repeated mechanical loading steps.[341] Due to the simple fabrication process, this strategy shows efficient practical usage for endowing functional devices with stretchability, including transistors, energy storage devices, and even integrated circuits.[345-353] For instance, high performance biaxially stretchable microsupercapacitor arrays can be realized based on the hybrid substrate with patterned polyethylene terephthalate (PET) films embedded in Ecoflex substrate.[346] The capacitors, with layered structures containing Au electrodes, MWNTs films and a patterned ionic gel electrolyte, were fabricated on top of PET islands, which are connected by stretchable liquid metal embedded in the Ecoflex substrate between the islands. The obtained device arrays showed high energy and power density (25 mWh/cm^3 and 32 W/cm^3) as well as stability under repeated deformations (100% strain uniaxial and 50% biaxial).

2.4 Stretchable electronic modules with integrated functions

In practical applications, various electronic units need to cooperate as a circuit to exhibit enhanced performances and rationally designed functions. Assisted by the aforementioned strategies, stretchable circuits containing multiple electronic elements can be realized as multifunctional modules for practical utilities.[303,354-356] These achievements would push forth the development of stretchable electronics, especially for applications in the wearable electronics industry, such as healthcare monitoring, medicine delivery and robotic control.

Based on this strategy, various multiple sensory systems have been well-designed and developed, exhibiting promising performances with high efficiency in human-machine interaction.[303,355,357-360] An "epidermal electronic system", which exhibits conformal adhesion on the skin's soft and curvilinear surface, has been systemically constructed through the integration of multiple electronic elements by optimized geometrical designs on an elastic polymer sheet, such as the power supply (i.e., solar cells and wireless power coils), multifunctional sensors (i.e., electrophysiological, temperature, and strain sensors), light-emitting diodes (LEDs), active/passive circuit elements (i.e., transistors, diodes, and resistors), and devices for radio frequency communications (i.e., high-frequency inductors, capacitors, oscillators, and antennae) (Figure 11a).[357] Active materials, such as silicon and gallium arsenide, are fabricated into serpentine nanoribbons or micro- and nanomembranes to function as electronic elements, which are connected by serpentine Au electrodes, to accommodate the deformations on human skin. Thus, this ultrathin, stretchable, and conformal device can be attached to the skin surface as a temporary transfer tattoo by soft contact and exhibit high-quality performances in electrophysiological recording, including electrocardiography (ECG), electromyography (EMG), and electroencephalography (EEGs).

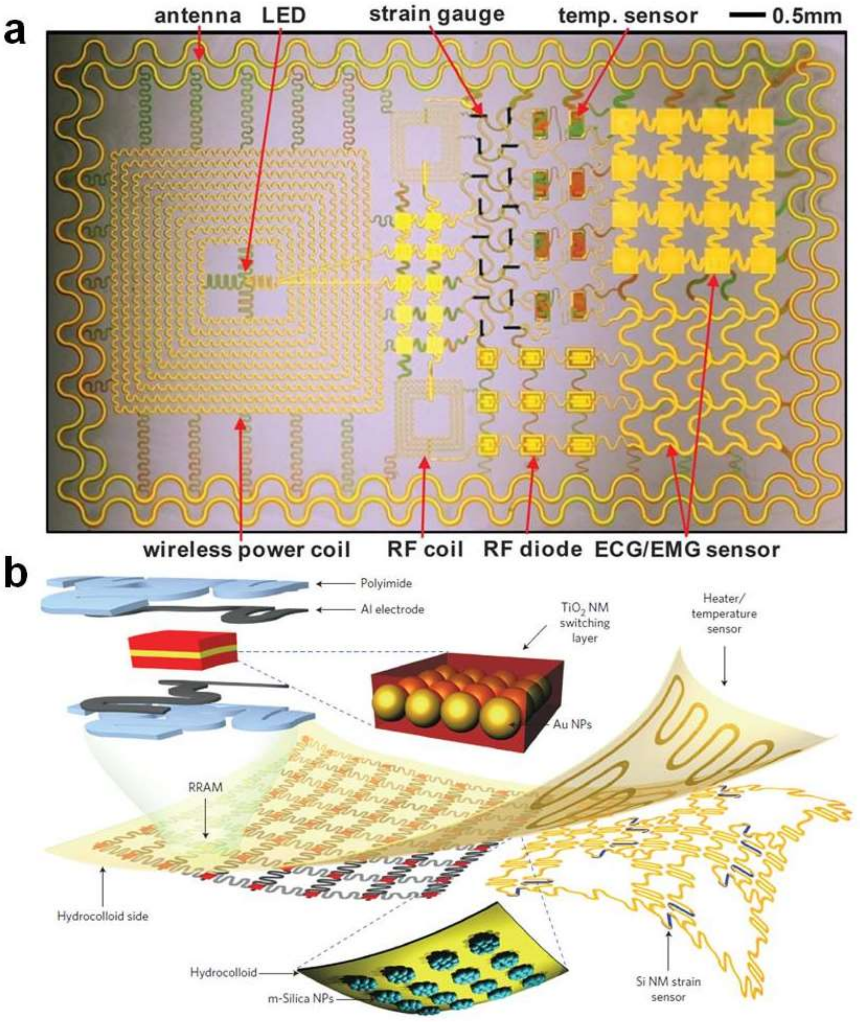


Figure 11. Stretchable electronic modules with integrated functions. a) Image of the epidermal electronics system consisting of power supply, multifunctional sensors (strain, temperature, electrophysiology), LEDs and wireless communication devices. Reproduced with permission from ref. 357. Copyright 2011 the American Association for the Advancement of Science. b) Illustration of a wearable system integrated with physiology sensors, memory module and actuating device for medical diagnostic and therapy. Reproduced with permission from ref. 361. Copyright 2014 Nature Publishing Group.

Beyond the development of various sensory systems for efficient information collection of physiological activities, the function-induced smart integration of different electronic elements showing intelligent response to incoming stimuli has been extensively proposed.[316,356,361,362]

Wearable systems with the integration of physiological sensors, non-volatile memory and drug-release actuators show intelligent behavior in monitoring muscle activity, storing data and delivering feedback therapy, which is highly desirable for personalized medicine and healthcare (Figure 11b).[361] The island-interconnection design was fully utilized to accommodate deformations on human skin. Through attachment to the surface of the human body, the tension and compression of skin can be detected via silicon nanomembrane based strain sensors, and the monitored data can be stored in integrated gold nanoparticle RRAMs. Through this, motion-related neurological disorders, such as tremors, which are detected and stored in memory devices, will be further analyzed and categorized into specific disease modes. Finally, the corresponding feedback of drug delivery from mesoporous-silica nanoparticles will proceed transdermally through thermal stimuli by the electroresistive heater. This skin-wearable system constitutes a valuable and promising approach towards the realization of intelligent medical therapy and interactive healthcare.

2.5 Self-healing devices accommodating mechanical damage

Although aforementioned strategies constitute solutions for flexible devices to survive mechanical deformations, the situations in real environments are more complex. When flexible devices containing soft and rigid materials suffer from large external forces, physical damages would occur once the applied strain exceeded the ability of the devices to accommodate mechanical deformations, which would usually destroy the device structure and cause function failure. To address this problem, inspiration was drawn from the healing mechanisms in living things after physical injury, and the self-healing property was thus introduced to flexible devices to survive mechanical damages and enable the recovery of device performance.[363-367]

The principle strategy for realizing self-healing devices relies on the combination of active materials, which are discretely separated to avoid macro physical damage, and self-healing polymers that comprise flexible chains and dynamic interactions.[368-370] In the first examples, active materials were engineered through rational structural design, such as in the form of surface-modified nanowires,[371-373] nanopieces,[374,375] and nanoparticles,[376] to perform electronic functions in a physically separated state to avoid being destroyed by mechanical damages as well as to enable the recovery of the original functions. The self-healing device with electronic properties can then be achieved by directly layering the structural materials on the surface or embedding them into the self-healing polymer substrates. The recovery of device performance is based on the re-association of structural electronic materials with each other induced by the self-healing of the polymer matrix under physical contact via applied external stimulus, including light,[377,378] magnetism,[379] heat,[373,380] and chemical treatments.[371]

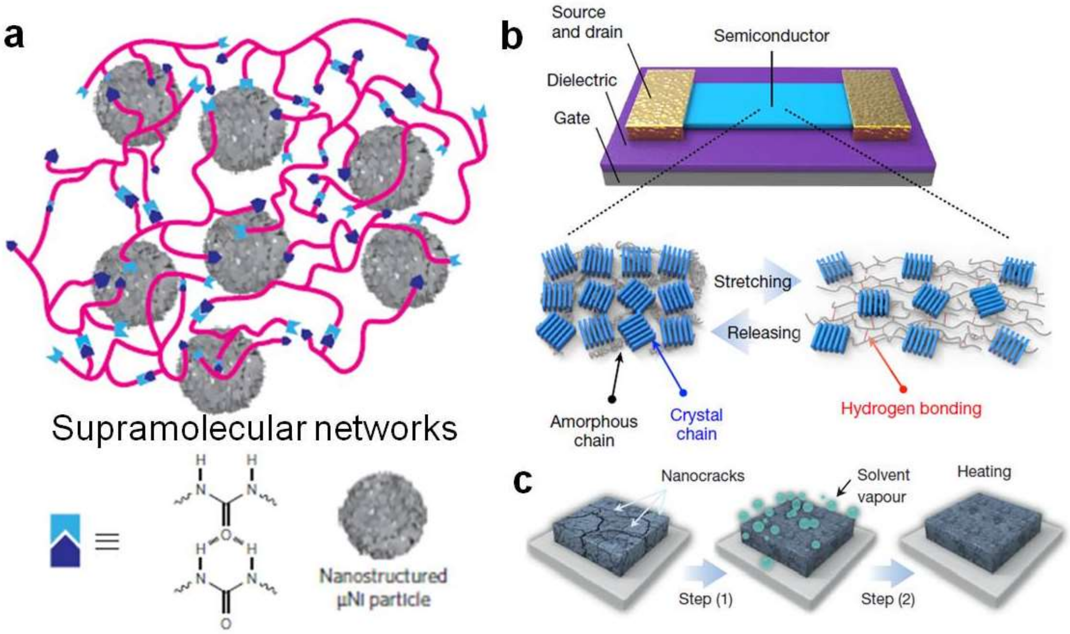


Figure 12. Electronic devices with self-healing properties. a) Illustration of a self-healing composite composed of crosslinked polymer network with micro-nickel particles. Reproduced

with permission from ref. 376. Copyright 2012 Nature Publishing Group. b) Schematic illustration of the configuration of the OTFT transistor and the mechanism for realizing stretchability. c) Illustration of steps to enable a self-healing process. Reproduced with permission from ref. 381. Copyright 2016 Nature Publishing Group.

A repeatable, self-healing conductor under room-temperature can be achieved by dispersing micro-nickel particles with nanoscale surface structures into a supramolecular polymeric hydrogen-bonding network with a glass transition temperature below room temperature (Figure 12a).[376] The conductivity relies on inter-particle quantum tunneling, which arises from the nanostructure of the Ni particles and shows a positive correlation to the concentration of Ni particles in the composite. At high Ni particle concentration (31% volume fraction), the composite serves as an electrode, while at the percolation threshold (15% volume fraction), it serves as a mechanosensor to tactile pressure and flexing. Most importantly, upon bifurcation by physical damage, the conductivity can be recovered by gently putting the two ends in contact at room temperature, thus enabling the movement of the molecular chains and the reformation of the hydrogen bonds among them. This would allow the Ni particles to be closely packed again at the healing interface. This work sets an effective example for the fabrication of self-healing electronic elements and devices with dispersed structural active materials.[128,374,382-384]

Moreover, by chemically controlling the molecular structure of the copolymer, a stretchable and self-healing semiconductor can be achieved by combining a semiconducting polymer containing rigid crystalline 3,6-di(thiophen-2-yl)-2,5-dihydropyrrolo[3,4-c]pyrrole-1,4-dione (DPP) units with non-conjugated and amorphous 2,6-pyridine dicarboxamide (PDCA) moieties with dynamic hydrogen bonding systems to enable mechanical deformability and the self-healing property (Figure 12b,c).[381] The polymeric semiconductor showed high stretchability as well as the efficient and complete recovery of field-effect mobility after physical damage as a result of the amorphous PDCA structure with dynamic bonding assisted by a solvent and thermal healing treatment. Overall, rational structural design of multifunctional materials is the essential task for achieving high deformability and self-healing properties for the adaptability of future flexible devices in real complex environments.

2.6 Soft conductive materials in flexible devices

The principle of using structural design to accommodate mechanical deformation is to release strain through shape transformation, which requires electronic materials to present basic flexibility. As discussed in section 2.1, 2.2, bulk metal or semiconductor materials, such as silicon or gold, which usually exhibit rigidity, are engineered into sufficiently thin architectures to present a “soft” nature on elastic substrates. However, this type of material would always have the limitation and show failure at the ultimate strain. Thus, conductive materials with an intrinsically soft nature, such as a liquid or gel, would be valuable candidates for constructing flexible and stretchable devices.

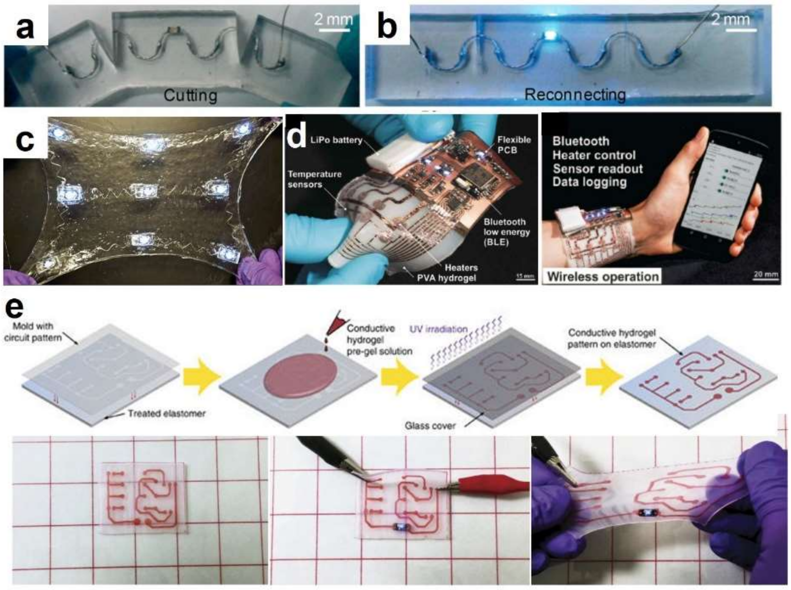


Figure 13. Electronic devices based on soft materials. a,b) Physical cut and reconnection of the LED-integrated galinstan circuit. Reproduced with permission from ref. 385. Copyright 2016 Royal Society of Chemistry. c) A hydrogel electronic device that encapsulates an array of LED lights connected by stretchable electrodes. Reproduced with permission from ref 386. Copyright 2015 John Wiley and Sons. d) Photograph of a flexible hydrogel electronic skin with an integrated circuit on a PVA substrate, which can be controlled and read out continuously via mobile phone. Reproduced with permission from ref 387. Copyright 2017 the American Association for the Advancement of Science. e) Schematic illustration of fabrication procedure for the conductive PAAm-alginate hydrogel patterned on an Ecoflex substrate. The hydrogel circuit can maintain its function under mechanical deformation. Reproduced with permission from ref 388. Copyright 2016 Nature Publishing Group.

Gallium and its alloys show low viscosity and high conductivity, making them promising conductive elements in stretchable electronics.[389-391] Various techniques, including lithography, injection, 3D printing, among others, were developed to pattern liquid metals on elastic substrates to construct flexible circuits, which have been comprehensively discussed in recent reviews.[391,392] By patterning with rational structural design, liquid metal-based conductive elements present excellent mechanical accommodation during physical deformations.[393-399] Galinstan, a commercially available liquid metal alloy, can be inkjet printed onto a stretchable substrate, forming a serpentine-shaped conductor which shows not only a steady conductivity under mechanical deformations but also a self-healing behavior after physical injury (Figure 13a,b).[385] Furthermore, many flexible devices with various functions based on liquid metals have been fabricated, such as pressure or strain sensors, antennas, memory devices, diodes, and capacitors.[400-407] For example, stretchable silicon rubber matrices were embedded with microchannels in designed patterns. When filled with Eutectic Gallium-Indium (eGaIn), the elastic matrix can serve as a sensing layer. By the assembly of three sensing layers, respectively, for x and y-axis strain and z-axis pressure sensing, the obtained highly deformable artificial robotic skin showed a multimodal sensing capability of detecting strain and contact pressure simultaneously.[408] Moreover, a similar strategy can be used to fabricate tactile keyboards with entirely soft materials.[409]

Gels are soft materials constructed by crosslinked polymers or supramolecular polymers that can hold large quantities of solvents.[410-412] Hydrogels, which contain water in gel networks, can easily mimic the elastic modulus of mammalian tissue and are compatible with human bodies.[413-415] By rational molecular design, hydrogels with mechanical properties display good consistency with aforementioned mechanical accommodating strategy and show outstanding biodegradability, biocompatibility and responsiveness to environmental changes.[416-419] For instance, circuits with island-interconnection designs consisting of serpentine-shaped electrodes can be either embedded in or attached to a tough hydrogel matrix, displaying both excellent deformability and stable electrical properties (Figure 13c).[386] In addition, a wearable integrated circuit can also be achieved by adhering a flexible PCB patch with imperceptible sensor/heater electronics to the top of a pre-stretched hydrogel/VHB sheet, which serves as a conformal hydrogel electronic skin (Figure 13d).[387]

By incorporating conductive ions in gel networks, such as solvated salts or ionic liquids, conductive gels can be realized with ionic conductivity as well as mechanical characteristics including toughness, stretchability and self-healing properties.[418,420-424] With rational structure engineering, this new class of material has already been employed in the fabrication of various flexible devices, such as mechanical sensors,[425,426] actuators,[427,428] stretchable electroluminescent displays,[429-431] microphones,[432] and touch panels.[433] For example, an ionically conductive PAAm-alginate hydrogel circuit can be patterned on top of an Ecoflex substrate by an in-situ gelation process, which can maintain reliable conductivity under large deformations (Figure 13e).[388] Based on such materials, all-soft matter devices with rational structural design exhibit promising applications in conformal, implantable, and edible electronics.[434,435]

3. Structural design in mechanical sensors

Mechanical sensors are designed to detect applied physical strain. One widely used sensing mechanism relies on the geometry deformation of the sensing element under extra mechanical forces, which can be further converted into an electrical signal for detection. Through architectures that are sensitive to deformation under mechanical forces, various sensory devices have been developed for sensitive and fast responses to pressing or stretching, spatial morphology recognition, and integrated multisensory systems. As a consequence of long-term evolution, a rich diversity of biosensors with efficient transduction of incoming mechanical information into bio-signals help living things survive in complex environments.[41,436] Thus, nature-inspired structural materials show attractive advantages in improving the sensing ability of devices as well as introducing new approaches for sensory device manufacturing.

3.1 Pressure sensor

In the early stage of research in tactile sensors, especially pressure sensors, conductive rubbers were used as pressure sensing elements, which were fabricated by the incorporation of conductive fillers in an insulating rubber matrix.[437] Due to the high electrical conductivity, carbon materials[438-442] and conductive polymers[443,444] were typically used as fillers to fabricate conductive rubbers with good sensitivity. The conductive rubber can exhibit mechanosensory properties via the detection of resistance changes caused by shape deformation upon the application of external forces. For example, a graphite-containing rubber pressure sensor with an organic field-effect transistor (OFET) as the readout element has been designed with good flexibility and stretchability and is capable of pressure imaging different objects.[173,445] However, due to the viscoelastic behavior, mechanosensors based on conductive rubbers usually do not exhibit satisfactory performances with high sensitivity in the low-pressure regime (<10 kPa) or fast responsiveness.[446,447] In addition, temperature-dependent changes in resistance caused by the large thermal expansion of polymer chains would disturb the analysis of strain sensing.[448-450] Thus, geometrical designs of the sensor structures have been developed and introduced to enhance the sensitivity and reduce the response time, leading to the effective transduction of external stress into electrical signals.[451,452] Compared to traditional sensors based on conductive rubbers, new types of pressure sensors containing 2D polymeric surfaces with microstructures show higher sensitivity, faster response and relaxation times, as well as lower detection limits. Based on different types of electronic signals arising from applied mechanical forces, several approaches for realizing flexible and sensitive pressure sensors have been developed, including piezoresistivity,[453,454] capacitivity,[101,455,456] piezoelectricity,[457-460] triboelectricity,[461,462] among others.

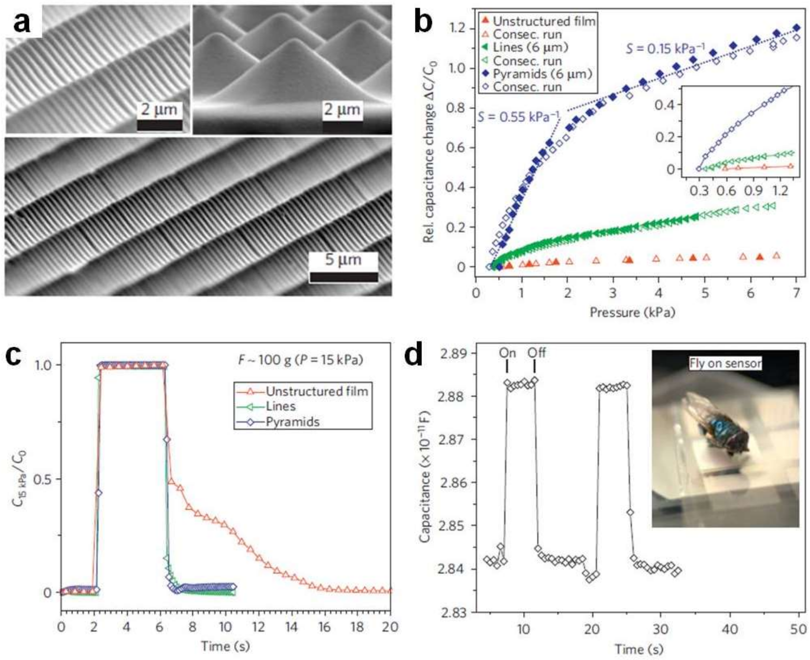


Figure 14. Pressure sensor based on microstructured PDMS. a) SEM images of the PDMS films with different microstructures. Comparison of performance for sensors with various (or without) microstructures: b) sensitivity c) hysteresis and steady-state. d) Demo of detecting the pressure of a fly. Reproduced with permission from ref. 455. Copyright 2010 Nature Publishing Group.

Zhenan Bao's group has developed a type of flexible pressure sensor with a structured-rubber dielectric layer as the key sensory element, which enables the enhancement of pressure sensitivity.[455] Their study proved that pressure sensors based on a pyramidal microstructure exhibit an extraordinary improvement in sensory performance, including substantially higher sensitivity in both the medium and low-pressure regimes as well as fast response and relaxation times (<< 1 s) (Figure 14b,c). The essential component in this design is the micro-structuring of the PDMS surfaces. Assisted by a designed wafer mold, large-area PDMS films with regular and uniform pyramid-structure arrays can easily be fabricated and can elastically deform under external pressure (Figure 14a). Thus, with the addition of another flexible conductive film (ITO on PET sheets) as the counter-electrodes, a small variance in the shape of the microstructure would enable capacitive sensing for pressure detection (Figure 14d). By modifying the microstructures of the

PDMS surface, the sensitivity and working pressure range of the pressures sensors can be tuned for different applications.[463] Furthermore, by integrating the micro-structured PDMS films into organic field-effect transistors, the highly sensitive and fast-response sensing of an applied pressure can be achieved by monitoring the output current on the drain and source in OFET devices.[455] Overall, the structural design of thin elastomeric films can significantly alter the mechanical properties of film surfaces, which is highly promising for highly sensitive and fast-response pressure sensors.

Since then, various nature-inspired nano/microstructures have been designed and utilized in the construction of pressure sensors with excellent performance for practical usage.[452-454,464-467] Some typical works are listed in Figure 15.

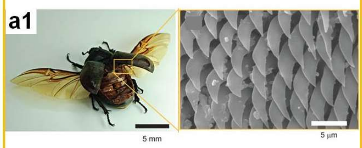
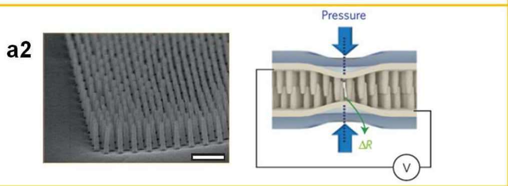
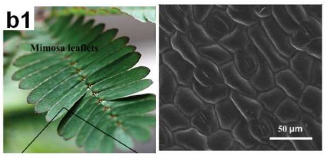
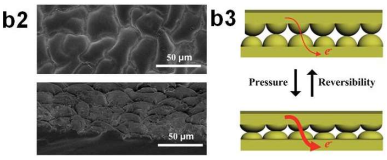
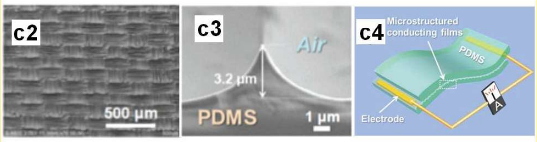
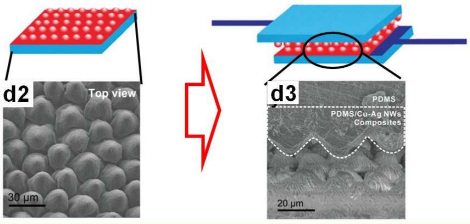
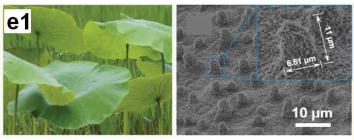
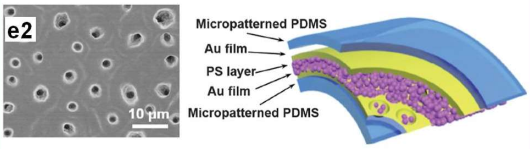
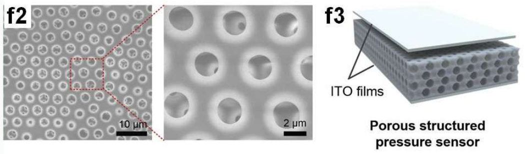
	Natural structure	Morphology of sensing unit and corresponding device structure	Performance (Sensitivity& response time&stability)
a			11.45 (GF) milliseconds range <10,000 cycles (Piezoresistivity)
b			50.17 kPa $^{-1}$ <20 ms >10,000 cycles (Piezoresistivity)
c			1.80 kPa^{-1} 10 ms 67,500 cycles (Piezoresistivity)
d	 Petals of Rose		1.35 kPa^{-1} 36 ms >5,000 cycles (Piezoresistivity)
e			0.815 kPa^{-1} 38 ms (capacitivity)
f			0.63 kPa^{-1} milliseconds range >10,000 cycles (capacitivity)

Figure 15. Nature-inspired structural materials for pressure sensing. a1) Photograph and SEM image of the interlocking structures found in beetles. Reproduced with permission from ref. 468. Copyright 2011 John Wiley and Sons. a2) SEM image of the nanohair structure constructed by Pt-coated polymer fibers and the illustration of the pressure sensing mechanism. Scale bar: 1 μm. Reproduced with permission from ref. 469. Copyright 2012 Nature Publishing Group. b1) Photograph of the leaf of a mimosa plant and SEM magnification of the microstructure of the leaflet. b2) SEM images (top and side views) of the PDMS replica. b3) Illustration of the pressure

sensing process. Reproduced with permission from ref. 470. Copyright 2014 John Wiley and Sons. c1) Photograph of a silk textile. c2) Top view and c3) side view of SEM imaging of the patterned PDMS using silk as a mold. c4) Structural illustration of the sensor device. Reproduced with permission from ref. 471. Copyright 2013 John Wiley and Sons. d1) Photograph of the petal of a rose. Configuration of the sensor and SEM image of the PDMS replica with the petal microstructure: d2) top view d3) cross-sectional view. Reproduced with permission from ref. 472. Copyright 2015 Royal Society of Chemistry. e1) Photograph and SEM image of a lotus leaf and the corresponding surface microstructure. e2) SEM image of the PDMS replica and the configuration of the capacitive pressure sensor. Reproduced with permission from ref. 473. Copyright 2016 John Wiley and Sons. f1) Photograph of a spongia officinalis. f2) SEM images of the PDMS replica with similar porous structures. f3) The configuration of the capacitive sensor. Reproduced with permission from ref. 474. Copyright 2016 John Wiley and Sons.

The mechanotransduction systems found in living organisms exhibit remarkable sensitivity in detecting mechanical deformation, which are usually based on the behaviors of hair-like structures under external forces, such as distortion and interlocking.[475-480] Typical examples of natural interlocking structures that give rise to the reversible binding property are the wing-to-body locking device in beetles[481] and the hooks and loops in burdock seeds, on which the design of the fabric Velcro[482] and nanostructured reversible adhesive surfaces were based.[468,483,484] Inspired by these biological systems (Figure 15a1), various sensing units with interlocking structures have been developed for mechanical sensing.[469,485-488] For instance, a nanoscale mechanical hair-to-hair interlocking architecture with two layers of Pt-coated ultraviolet-curable polyurethane acrylate (PUA) nanohair arrays was utilized in constructing a high performance sensor (Figure 15a2).[469] When external loads are applied on the sensor, the compressive deformations will generate tiny distortions in the uniformly paired hair-to-hair interlocking of regularly ordered, high-aspect-ratio nanohair arrays, which can be converted into a change of resistance for detection. Moreover, different mechanical loadings, including pressure, shear and torsion, can be detected and distinguished by this interlocking-based sensor via the unique and discernible magnitude or pattern of the measured gauge factors (GF). Thus, multiplex outputs from a mechanical stimulus can be achieved by analysis of the specific GFs (~11:45 for pressure, ~0.75 for shear and ~8.53 for torsion), which demonstrates the ability to decouple arbitrary contact with the device. The high sensitivity and the fast response allow the sensor to detect the dynamic motion of a small bouncing water droplet (20 μL).

This interlocking design can be further improved by incorporating nanostructures on each polymer micropillar to form hierarchical architectures, which would introduce highly sensitive and ultrafast responsiveness with minimal hysteresis. This can be achieved because the large surface area amplifies the change in contact resistance under compressive deformations. The piezoresistive sensor was constructed by orderly patterned PDMS micropillars decorated with ZnO nanowire arrays, followed by Pt coating on top.[487] Based on the interlocked geometry of the hierarchical structures from the nano- to micro- scale, the flexible piezoresistive sensor shows a high pressure sensitivity (-6.8 kPa^{-1}) and an ultrafast response time (<5 ms). This enables the detection of minute static pressures (0.6 Pa), vibration levels (0.1 $m\ s^{-2}$), and sound pressures (~57 dB). Moreover, with the decorated interface of the interlocking structure to Ni-coated ZnO NWs and bare ZnO NWs, the piezoelectric mode of the pressure sensor can be realized by conversion of external loads to electrical potential differences, showing high sensitivity for the detection of high-frequency pressure variations. Compared to the piezoresistive mode, the instantaneous generation of piezoelectric signals via the separation of dipoles in piezoelectric mode can enable interlocking sensors to quickly respond to external dynamic pressures, such as real-time sound monitoring.

In addition to pressure sensors based on regular nano- and micro-structures fabricated by traditional lithography techniques, new routes have also been developed for obtaining structural sensing elements by copying the microstructures of natural creatures and artifacts (Figure 15b-e).[470-473,489] In particular, the nano- and micro-structures that widely exist on plant surfaces as a result of billions of years of evolution and natural selection can be directly used as molds for the cost-effective and scalable fabrication of patterned electrodes. For example, the leaves of mimosa that show remarkable responsiveness under external stimuli are natural pressure flexible sensors, which can serve as templates for microstructuring the PDMS surface by a two-step negative/positive molding process.[470] Upon the deposition of gold, structural electrodes with an irregular pattern of microdomains would be formed, which constitute contact-resistance-type pressure sensors with high sensitivity (50.17 kPa^{-1} in the regime of 0-70 Pa) and fast responses (<20 ms) (Figure 15b). Other surfaces of plant organs have also been investigated, such as a PDMS film templated with the inverse microstructures of the rose petal, forming a resistive sensor by coating it with Cu-Ag nanowires (Figure 15d).[472] Another example is the micropatterned flexible PDMS/Au substrate duplicated from the lotus leaf, which forms an essential sensing element in a flexible capacitive tactile sensor (Figure 15e).[473]

In addition to the previously discussed strategies for constructing microstructures on the surface of elastic substrates, other natural materials showing effective deformability under subtle mechanical forces would be valuable candidates for designing mechanical sensors. For example, inspired by *spongia officinalis* with a hierarchically porous structure, a sponge-like structural PDMS dielectric layer was fabricated using a sacrificial template comprising a polymer microbead stack (Figure 15f).[474] After sandwiching the porous PDMS film between two flexible electrodes, a high-performance capacitive pressure sensor was achieved (0.63 kPa^{-1} with 6 μm pore diameter), with a sensitivity that can be tuned by controlling the size of the pores. Porous PDMS films with larger pore sizes would exhibit higher compressibility, which determines the ability to accommodate deformation under external pressure, thus enabling an improvement in sensitivity but simultaneously sacrificing the workable pressure range.

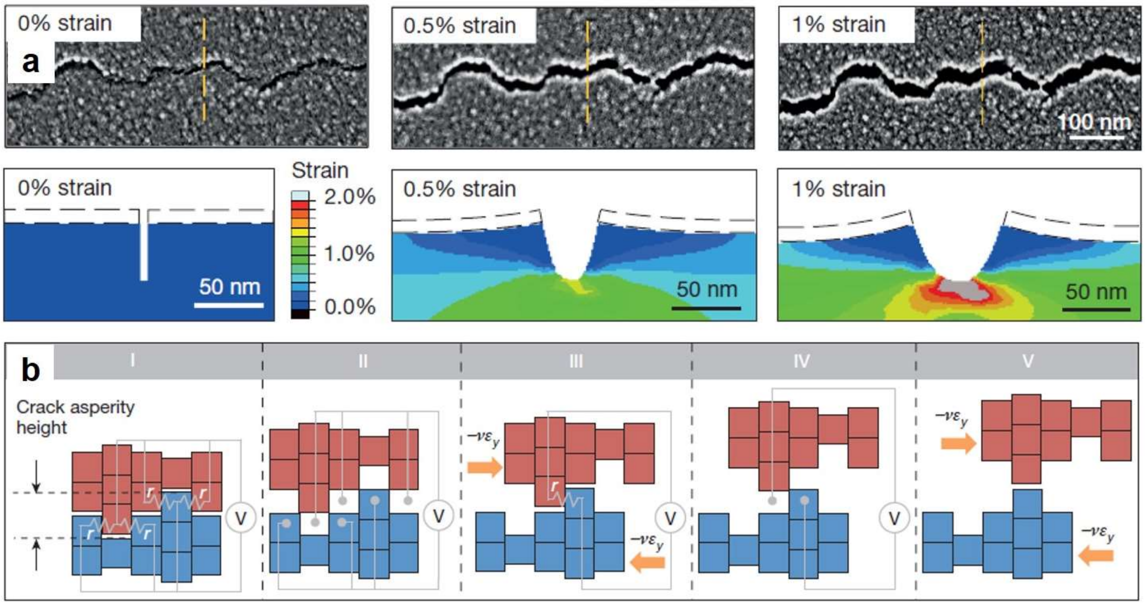


Figure 16. Nanoscale crack-based sensor inspired by spiders. a) SEM images and FEM simulation results of the cracks formed under different strains. b) Illustration of the behaviors of the crack asperity through the process of disconnection and reconnection. Reproduced with permission from ref. 490. Copyright 2014 Nature Publishing Group.

3.2 Strain sensor

In addition to pressure sensors based on structural materials, strain sensors with high sensitivity to physical strains are another significant family in mechanosensing.[491-493] Until now, various strategies, including dimensional changes,[494-498] piezoresistivity,[499-504] and the tunneling effect,[505-508] have been utilized to achieve strain sensors with excellent properties. Recently, spatial separation of conductive elements in conductors, which usually comprise structural nanomaterials, was utilized to realize a highly sensitive strain sensor. One typical example is the aforementioned 1D nanomaterials network that shows strain responsiveness via a resistance change caused by the disconnection of the original overlapped 1D structures under stretching.[76,509-513] Another one is based on cracks, in which the sensory mechanism relies on the mechanical fractures due to the mechanical mismatch between the elastic substrate and rigid conductive films consisting of 2D nano- or micro- sheets, nanowires, nanotubes, and nanoparticles.[354,514-520]

In nature, the long history of evolution brought about various strain sensory systems in living creatures with ultra-high mechanosensitivity, flexibility and durability, which perfectly match their specific biological needs.[41] For example, spiders, one of the most sensitive species to mechanical stress, feed on their entangled insect prey by monitoring the vibrations of the spider web. Their most important strain detectors are the lyriform slit systems embedded in the exoskeleton near the leg joints, which contain parallel crack-shaped slits that can be deformed under minute cuticular strains as low as a few microepsilons (10^{-6}).[521,522] The deformations on this lyriform slit sense organ can be detected and further processed by the nervous system for the perception of external vibrations. Inspired by the geometry of the sensory organs, various crack-based mechanosensors have been designed to exhibit sensitive responses to physical strain based on the formation and propagation of cracks in conductive films. As a typical example, a nanoscale crack sensor based on platinum films showed ultrahigh sensitivity to strain and vibration.[490,523] To mimic the structure of parallel slits, a stiff platinum (Pt) layer on top of a viscoelastic polyurethane acrylate (PUA) was mechanically bent, resulting in nanocracks being formed penetrating the Pt film and the surface of substrate in the transverse direction (Figure 16a).[490] Thus, a tiny strain is applied to these straight transverse cracks, the electrical resistance of the Pt film would experience a sudden jump to an extremely high value, which is the basis for the high gauge factor of the nanoscale crack sensor. At <1% strains, the fluctuation in the process of conductance decreasing along with the increasing strain reveals the dynamic behaviors of gap-bridging steps on the opposite edges of nanocracks under stretching. The stretched substrate with a positive Poisson's ratio results in compression in the transverse direction while being extended in the axial direction, which leads to numerous small steps of disconnections and reconnections at the zigzag edges of the cracks (Figure 16b). This process would continuously occur during uniaxially stretching until the gap distance overcomes the crack asperity height. The resulting ultra-high mechanosensitivity with a strain gauge factor over 2000 and vibration amplitudes of 10 nm made the nanoscale sensor applicable in speech pattern recognition and the detection of physiological signals. Meanwhile, unlike the Pt film, the as-prepared Au film can only exhibit random island-type cracks instead of straight cracks, which would help maintain the conductivity under strain.[85,205,212] However, by modifying the interaction between the metal film and substrate, the Au film can also show high sensitivity in strain detection (which will be discussed in Section **4.2**).[38,524]

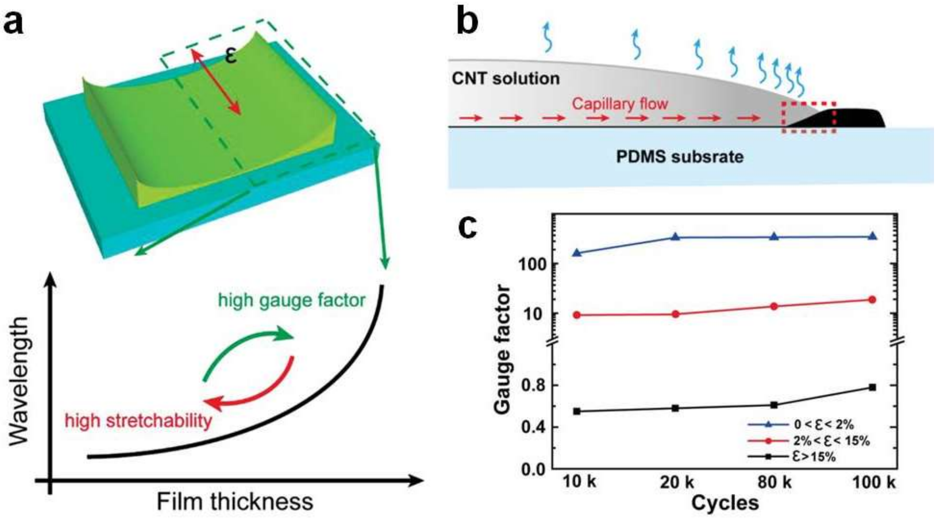


Figure 17. Strain sensor with a thickness-gradient configuration. a) Illustration of the desired gradient structure of the sensor and b) the fabrication process of SWCNT films due to the self-pinning effect. c) Gauge factor of the sensor at various strains and cycles. Reproduced with permission from ref. 525. Copyright 2015 John Wiley and Sons.

However, strain sensory materials with high sensitivity usually show a narrow strain sensing range, which can be attributed to the brittle nature of sensory materials with high gauge factors, such that physical deformations for sensing occur under small strains and totally collapse under large strains. To solve this problem, the gradient structure, a widely existing architecture in nature that can combine materials of different moduli and mechanical properties, was introduced to integrate high sensitivity and large stretchability in a single strain sensor (Figure 17a).[525] Through the self-pinning effect, also known as the coffee ring effect, a SWCNT solution can form a thickness-gradient SWCNT film on an elastic PDMS substrate by a facile dip-casting process with a designed mask (Figure 17b). The thick SWCNT assembly on the edge of the 'coffee ring' would exhibit a brittle nature that enables high sensitivity to strain, while the thin SWCNT film in middle, with a 1D conductive network, would accommodate large strain. The obtained stretchable strain sensors with thickness-gradient structure possesses gauge factors as high as 161 ($\varepsilon < 2\%$), 9.8 (Avg., $2\% < \varepsilon < 15\%$), and 0.58 ($\varepsilon > 15\%$) with isotropic strain of more than 75%, which show potential practical usage in the detection of weak vibrations and the reorganization of sound (Figure 17c).

3.3 Electronic whiskers

The whisker is another family of mechanosensory systems on the body surface of mammals or insects that is capable of tactile detection, such as in the detection of wind, touch and vibration. For example, thin hairs on the exoskeleton surface of insects can detect signals of direct contact or airflow.[526-529] Cats and rodents can perceive spatial information by laterally oriented arrays of vibrissae in the face via active whisking (Figure 1g).[39,530-534] Inspired by this unique structure, hair-like tactile sensors have been investigated to achieve sensing and transduction of various mechanical forces, and been demonstrated for airflow monitoring as well as the spatial mapping of object surfaces.[35,535-544] Recently, the rise of nanomaterial-based mechanical sensory systems has added new concepts in this domain, exhibiting high sensitivity, excellent flexibility and stretchability.[37,42,545-549]

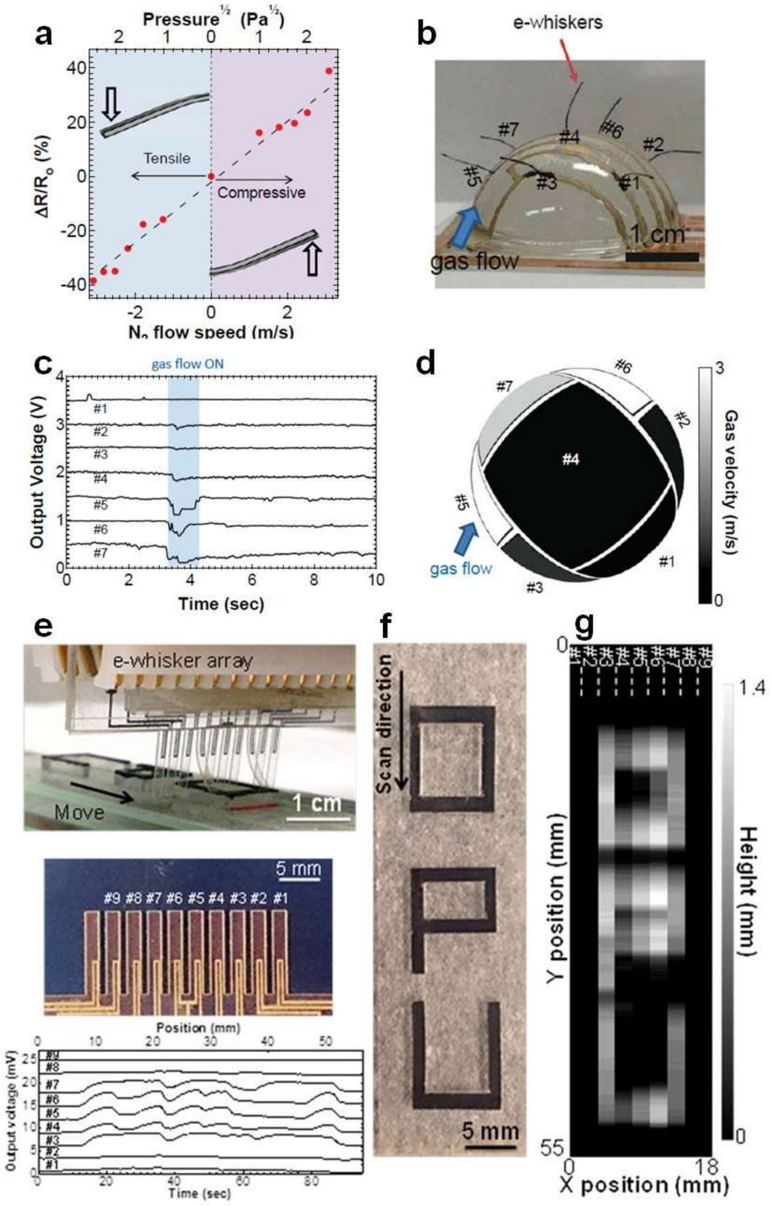


Figure 18. Electronic whiskers. a) Sensitivity of CNT-AgNP nanocomposite film-based e-whisker for the detection of gas flow in both tensile and compressive ways. b) Photograph of an array of e-whiskers for wind mapping. c) Output voltage and d) the corresponding 3D resistance

change mapping in response to gas flow. Reproduced with permission from ref. 37. Copyright 2013 National Academy of Sciences. e) Photographs of a printable e-whisker array for 3D spatial distribution mapping and the mapping configuration. Optical images of f) the mapping target and g) the corresponding mapping result. Reproduced from ref. 545. Copyright 2014 American Chemical Society.

A series of electronic whiskers based on hairlike PDMS substrates coated with CNT–AgNP composite films that exhibit high strain sensitivity have been developed (Figure 1i). The conductive CNT network endows it excellent flexibility, while the AgNPs enhance the conductivity and exhibit high strain sensitivity, the mechanism of which is based on the change in the spacing between the AgNPs upon deformation of the substrate.[37,545] As the two sides of the substrate are always under opposite strains when the e-whisker is bent, a design comprising asymmetrical coating with a CNT-AgNP composite on the top and bottom electrodes is introduced in the device configuration, which is essential to detect tensile and compressive forces. A 5 wt % AgNP composite was used on the top side for strain detection, while a 30 wt % AgNP composite was patterned on the bottom side to serve as an interconnect electrode in order not to affect the reading of resistance change in the whole device during strain sensing. Thus, the electronic whiskers can distinguish between tensile and compressive stress with high sensitivity (Figure 18a). For instance, spatial mapping of wind flow was achieved by an array of whiskers mounted on the desired PDMS hemisphere (Figure 18b).[37] By reading the voltage drop of each whisker, the values of gas velocity at each domain can be obtained and used to construct a three-dimensional airflow map (Figure 18c,d). A similar design of device structures involves a printable multifunctional artificial whisker array for two- and three-dimensional space and temperature distribution mapping.[545] CNT-AgNP films and a (PEDOT:PSS)-CNT composite were patterned on the whiskers to monitor the spatial and temperature information of object surfaces, showing integrated sensing properties beyond the original biofunctions (Figure 18e). For mechano-sensing, the electronic whisker array can realize 3D height mapping of object surfaces by reading the voltage change of each whisker in the array, and the resolution of height differences can reach tens of microns (Figure 18f,g). The biomimetic hair-like electronic whiskers also show high sensitivity in real-time monitoring of environmental and spatial information, which has potential applications in advanced robotics engineering, human–machine interfaces, and wearable health monitoring systems.

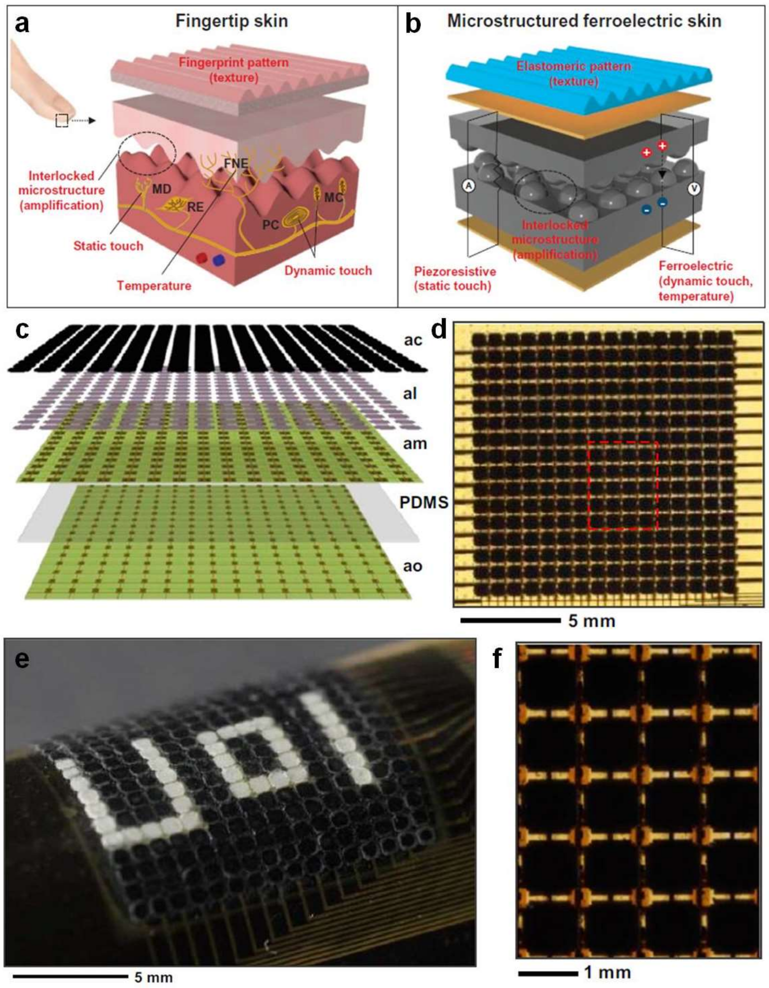


Figure 19. Multifunctional e-skin devices inspired by nature. a) Illustration of the structure and function of human fingertip skin. b) Structure of flexible ferroelectric e-skin mimicking the human fingertip. Reproduced with permission from ref. 550. Copyright 2015 the American Association

for the Advancement of Science. Illustration of the construction of an artificial camouflage skin with multilayer structures: c) exploded view, d) top view. e) Optical image of the device displaying information under bending. f) Magnified image of the area in the red rectangle. Reproduced with permission from ref. 551. Copyright 2014 National Academy of Sciences.

3.4 Nature-inspired multifunctional e-skin

The skin of animals is a complex sensory system that comprises an efficient network of sensors for the detection of environmental stimuli, such as pressure, temperature and vibration.[552-554] After sensing, the incoming stimuli are transduced into physiological signals which can be transmitted though the nervous system and finally interpreted and processed in the brain. Recently, electronic skin, an artificial integrated sensory module, has been widely investigated and designed to mimic the architectures and functions of natural skin using electronic devices. This can serve as a multifunctional platform in wearable devices, artificial prosthetics, health monitoring, and smart robots.[555-559]

Natural evolutionary structures play a critical role in the perception of natural skins, which is the inspiration for the construction of high performance electronic skins. During the sensing process, i.e., when a human fingertip touches an object, the fingerprint patterns and the interlocked microstructures between the epidermal and dermal layers can amplify the tactile stimuli and efficiently transfer the incoming signals to cutaneous mechanoreceptors, thus enabling the perception of a fine surface texture (Figure 19a).[560-563] Inspired by the layered structure and diverse functions of the human fingertip, an integrated e-skin with a laminated structure has been fabricated with well-designed fingerprint-like patterns, interlocked microstructures, and the cooperation of various sensors, which showed high sensitivities in the multiple spatiotemporal perceptions of static and dynamic tactile stimuli, vibration, and temperature (Figure 19b).[550] These unique properties were realized by using the piezoelectric and pyroelectric responses of ferroelectric polymer films composed of reduced graphene oxide (rGO) sheets in a poly(vinylidene fluoride) (PVDF) matrix. The resistance of the ferroelectric rGO/PVDF composite films showed a negative temperature coefficient (NTC), which can be used for temperature sensing. Static and dynamic tactile stimuli sensing were achieved by detecting the piezoresistive change based on the interlocked microstructures on the rGO/PVDF composites, which present a larger contact area and stress concentration effect. To mimic the fingerprint patterns on the fingertip, parallel microridges were fabricated on the top of composite films to enhance the vibrotactile signals produced when the e-skins are used to scan a textured surface (Figure 19b). Furthermore, the e-skins have been successfully used in the temperature-dependent pressure monitoring of artery vessels, the precise detection of acoustic sounds, as well as surface textures. Thus, nature-inspired microstructured ferroelectric skins may provide valuable sensory platforms in humanoid robotics and wearable medical diagnostic systems.

In addition to the human skin, some animal skins can exhibit additional functions, for example, chameleons and cephalopods are capable of changing their skin color for camouflage and communication,[564-566] which inspired the construction of artificial materials with smart color-changing abilities.[567-569] In these skin tissues, laminated structures also play an essential role in realizing the color changing process. For cephalopods, a three-layered system endows the skin with rapid, patterned physiological color change, which is the coordinated action of (i) chromatophores (top layer), (ii) iridophores (middle layer), (iii) leucophores (bottom layer), (iv) muscles, (v) central ocular organs, and (vi) distributed opsins.[570,571] By mimicking the layered structure with functional devices analogous to each element, an artificial electronic system that can autonomously sense and adapt to the coloration of their surroundings has been realized.[551] This cephalopod-inspired flexible sheet is constructed by four layers of multiplexed device arrays, including a color-changing element based on a thermochromic dye (analogous to a chromatophore), a white reflective thin layer of Ag (analogous to a leucophore), an actuator made by Si diodes providing local heating (analogous to the muscles), and a light sensor that consist of photodiodes and multiplexing switches (analogous to a functional unit involving opsins) (Figure 19c). All these components constitute unit cells that are patterned into arrays for multilayer stacking (Figure 19d,f). When the devices are exposed to white light, the multiplexed photodetectors activate the Si diodes to generate local heat to control the optical properties of the dye, which shows reversible responsiveness to temperature. Thus, different static geometries and dynamic pattern recognition can be achieved by this functional integration of multilayer device arrays exhibiting black-and-white patterns to match the surroundings (Figure 19e). In addition to mimicking the natural process, rational designs for integrating functional electronic units would achieve more intelligent modules beyond the mammalian functions. For instance, sensor arrays can be combined with other functional electronic units in layered structures, including memory devices, light-emitting diodes, and transistors, to perform integrated and smart behaviors, such as sensory memory systems with digital and optic reading properties.[139,572-576] Thus, the design of such integrations will bring opportunities for future artificial skins with desirable multifunctional performances and adaptability for various specific utilities.

4 Structural material-dominated functionalization in flexible devices

In nature, structural materials usually present remarkable mechanical properties since the permanent evolution, such as the tolerance of physical deformations and effective transfer of mechanical forces. When the architectures are used in flexible electronics, the combination of electronic techniques and the aim for practical utilities will transcend the boundaries of their original functions and bring new concepts for future devices.

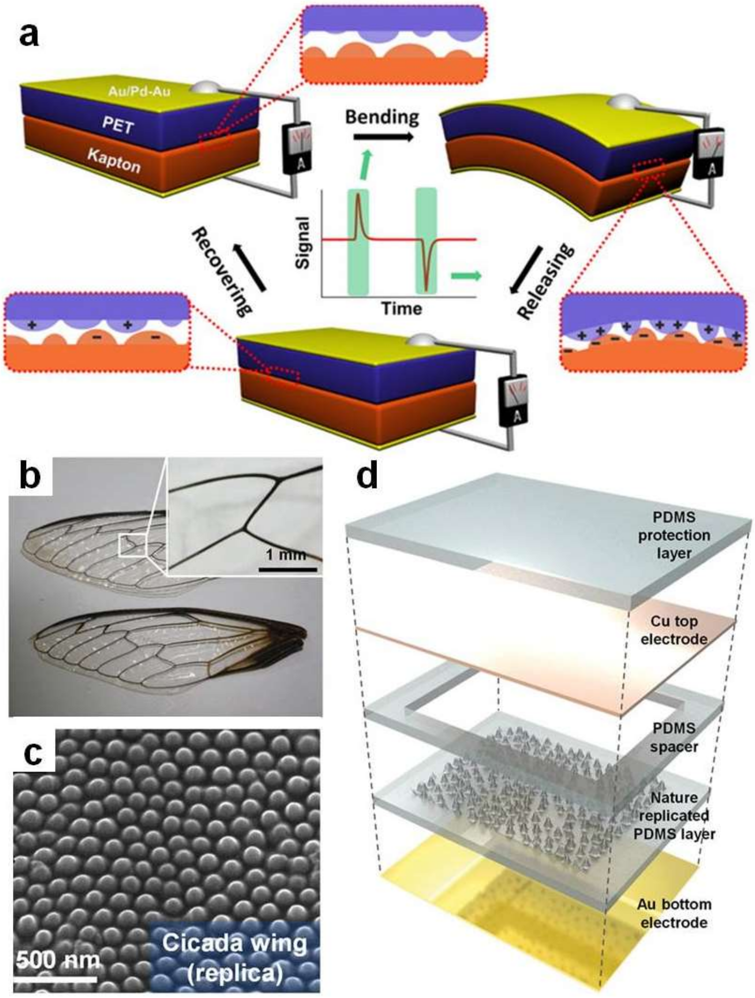


Figure 20. Nanogenerators inspired by nature. a) Schematics of the structure and electrical measurements of a triboelectricgenerator during bending and releasing tests. Reproduced with permission from ref. 577. Copyright 2012 Elsevier. b) Photograph of a cicada wing and c) SEM image of the cicada wing replica based on PDMS. d) Illustration of the structure of triboelectric nanogenerator device with nature replicates. Reproduced with permission from ref. 578. Copyright 2014 John Wiley and Sons.

4.1 Nanogenerator

The nanogenerator is a type of device that can convert mechanical energy into electricity, which is a promising power supply and shows potential applications in our daily lives.[5,43] Two key approaches for constructing flexible nanogenerators have been developed, namely that of piezoelectric nanogenerators and triboelectric nanogenerators.[579-583] The power generation process of piezoelectric nanogenerators is based on nanostructural piezoelectric materials, such as ZnO,[584-592] lead zirconate titanate (PZT),[593-597] $BaTiO_3$ (BTO),[598,599] MoS_2,[600,601] and PVDF.[459,602-605] In such materials, the cations and anions of the charge centers can be separated to form an electric dipole under mechanical deformations, thus producing a piezopotential.[606-608] Due to their fast response to external forces, this phenomenon can also be used in the mechanical sensors discussed above.[457-459]

On the other hand, triboelectric nanogenerators are realized by a combination of triboelectrification and electrostatic induction, which is a very common phenomenon in nature but has been ignored as an energy source for electricity.[43,581] The principle relies on friction-induced charge separation on two sheets comprising materials with distinctly different triboelectric characteristics (i.e., electron donor and acceptor).[577] In 2012, the first example of a triboelectric nanogenerator was constructed by stacking a Kapton film and a PET film with a rough surface on the nanoscale. [577] Mechanical forces applied to the device leads to a small degree of friction between the two layers, causing electrostatic charges separated on the surfaces of the Kapton and PET layer, which would generate electric potential for a power supply (Figure 20a). Two main approaches for constructing triboelectric nanogenerators are the vertical contact-separation mode[162,609-611] and the in-plane sliding mode.[612-618] Due to the dominant effect of surface morphology on the friction experienced by the triboelectric nanogenerator, the structure engineering at the interface is essential for enhancing the performance, and various surface structures have been developed and investigated, such as arrays of micropyramids,[462] nanorods,[619] nanowires,[620-622] nanoparticales,[623] nanoflowers,[624] micro/nano hierarchical structures,[625,626] self-assembled nanoscale patterns of polymers,[627] among others. Furthermore, surface morphologies with densely packed hierarchical structures that enlarge the contact area exist widely in nature, such as fingerprints on fingertips that can enhance friction and sensing.[560,561,563] Thus, the inspired fabrication and direct replication of hierarchically structural surfaces in nature would be an effective approach for building triboelectric nanogenerators.[578,628] For example, a cicada wing composed of nanostructures has been introduced as a template for the fabrication of a structural PDMS layer through a two-step negative/positive molding process (Figure 20b,c).[578] Under the contact-separation mode, the fabricated triboelectric nanogenerator can exhibit stronger instantaneous voltages and currents and a 3 times enhancement in total generated energy compared to a flat control structure (Figure 20d). Since first proposed in 2012, the efficiency in output performance of triboelectric nanogenerators has been dramatically enhanced. Developments of structural materials have played an important role in this progress and inspiration from creatures in nature will contribute to the further creation of highly efficient nanogenerators with reliable durability and mechanical adaptability.

4.2 Interfacial interactions in flexible devices

When designing a flexible device, the mechanical interactions, and most of all the adhesion effect, is an important factor that needs to be considered.[629-631] The topic can be divided into two aspects: (1) the adhesion between the materials constructing the devices, which usually endow the layered structure with different elasticities; and (2) the interactions between the fabricated devices and the working substrates, especially for wearable devices that are designed to be adaptable on biological surfaces.

During the mechanical deformation of flexible devices, the interfacial adhesion between rigid and soft materials with huge differences in moduli has always been considered as a crucial engineering problem, especially for mechanical interactions between different layers and rigid metal electrodes on elastic substrates.[632-639] Conventional solutions to improve adhesion rely on the introduction of adhesion materials, such as a thin layer of metals or a chemically activated layer on the surface.[107,241,633,640-645] Inspired by natural creatures, structural modification at interfaces would constitute a valuable and effective solution to this problem.[38,524,646] For example, root systems show remarkable mechanical properties that enable plants survive in complex and hostile environments. Therefore, the structure of root systems can be introduced at interfaces between electrodes and soft substrates to realize desirable stretchability and high adhesion (Figure 21a). Inspired by this, an interlocking layer with biomimetic roots extending into the PDMS substrate under the gold electrode has been designed to enhance adhesion.[38] The fabrication of the gold nanopile array was based on electrochemical reduction in solution with an anodic aluminum oxide (AAO) membrane as a sacrificial template. After coating with the PDMS film, the crack-based gold film, which is discussed in Section **2.2.1**, was thermally evaporated on top of the nanolocking layer to form the electrode. Compared to a crack-based stretchable gold film (CSGF) and a dense nonstretchable gold film (NSGF), the adhesion between the electrode and the substrate with the nanopile-structured gold film showed remarkable improvements (Figure 21c). The adhesive strength showed a positive correlation with the nanopile length due to the contact area between the nanopiles and the substrate. However, compared to the CSGF electrode, the nanopile-structured gold film exhibited low stretchability and a corresponding high gauge factor, which can be attributed to the regulated strain distribution in the metal film that is not advantageous to the formation of homogeneous microcracks (Figure 21b). As a result of the high gauge factor, the high-adhesion stretchable gold films can be used as strain sensors with tunable stretchability.

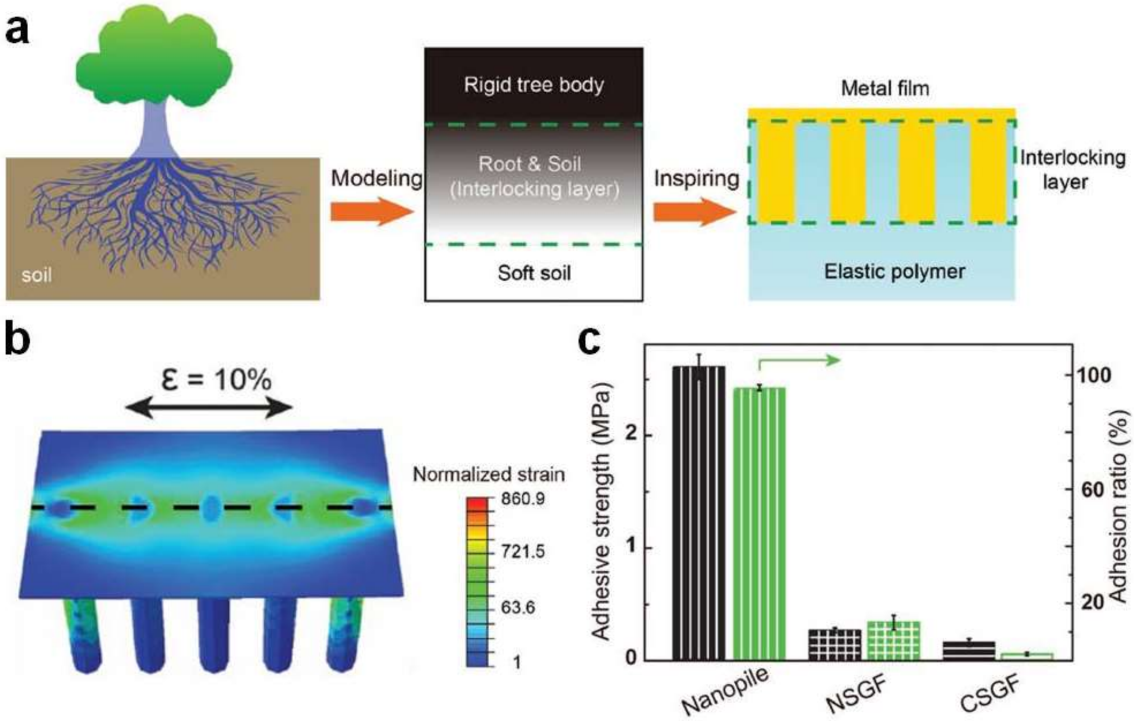


Figure 21. Flexible electrode based on a gold/PDMS complex film with highly effective interlayer adhesion inspired by the root system of plants. a) Illustration of the principle for the nanopile interlocking structure to achieve a combination of "hard" and "soft" materials. b) FEM simulations result of the strain distribution in the film with the nanopiles c) Comparison of adhesion for the nanopile film, a common crack-based stretchable film and a flat non-stretchable film. Reproduced with permission from ref. 38. Copyright 2016 John Wiley and Sons.

Wearable electronics, particularly skin-attachable devices, are designed with hierarchically organized topography to exhibit high deformability on curvilinear surfaces, as well as conformal contact on biological surfaces.[42,647-649] Microhair architectures inspired by gecko foot hairs, which contain hierarchical structures with excellent adhesion,[36,650-653] have been recently introduced as an effective interfacial modification to improve dry adhesion and the transfer of various mechanical forces (Figure 22a).[654-659] For example, mushroom-like micropillar arrays have been designed to mimic gecko foot hairs, which endowed a real-time diagnostic device with good dry adhesion on the human skin (Figure 22b,c).[658] The hair-like structure not only showed effective adaptation to rough human skin but also provided excellent biocompatibility, with respect to sufficient space for air ventilation and minimal contact between the skin and potentially toxic chemicals. The adhesive strength is governed by the geometry of the wide tip, the aspect ratio of the micropillar and the array density, which can be tuned to optimize the performance of the dry adhesive patch.

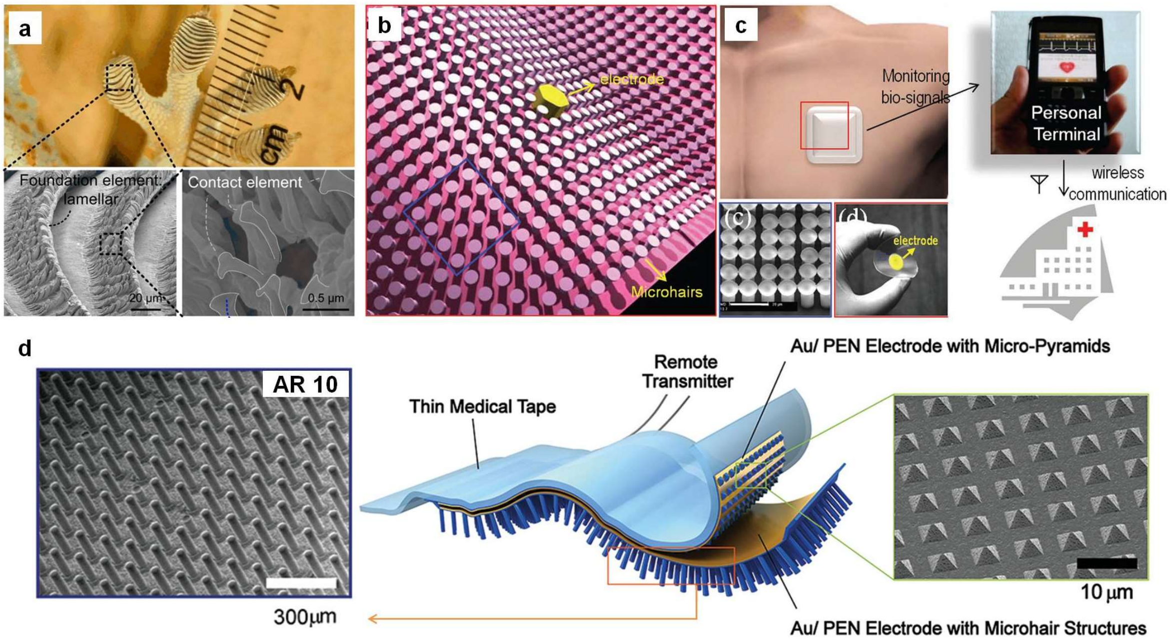


Figure 22. Sensors designed with enhanced adhesion/conformability inspired by nature. a) Optical image of a gecko's toe pads and SEM images of the lamellar skin and the contact element. Reproduced from ref. 659. Copyright 2017 American Chemical Society. b,c) Illustration of the concept of using a dry adhesive in a real-time medical monitoring device. Reproduced with permission from ref. 658. Copyright 2011 John Wiley and Sons. d) Schematics of the multilayer structure of the skin-conformal microhairy sensor (middle) and SEM images of a microhair array

(left) and the pyramid-shaped PDMS layer (right). Reproduced with permission from ref. 42. Copyright 2014 John Wiley and Sons.

Beyond endowing the basic adhesion property, structuring at the interface between wearable devices and biological surfaces can also benefit the transduction of various mechanical forces. The incorporation of microhair interfacial structures in a flexible pressure sensor can enable signal amplification with significant enhancement in the signal-to-noise ratio. This provides the possibility of detecting extremely weak signals from the human body, such as the deeply buried internal jugular venous pulses.[42] The device is comprised of a multiple-layered structure with a pyramidal-shaped PDMS film as sensing layer, as well as a laminated microhair-structured PDMS film as the attachment layer (Figure 22d). Due to the dual-level rough surface of skin presenting a combination of macroscale and microscale roughness, the biopulsation-induced contractions and expansions on the part of the skin with microscale roughness are difficult to detect with a plane surface. This means that conformal contact with skin will improve the efficiency in collecting mechanical deformations on rough surfaces, thus enhancing the signal intensity. Compared to a flat device, the pressure sensor with the microhair structure with a high aspect ratio (i.e., AR=10) showed a nearly 12 times increase in signal-to-noise ratio, which can be attributed to the full coverage on the rough surface. In this way, bio-inspired designs demonstrated unique advantages in achieving ultra-conformity on non-flat surfaces with the substantial enhancement of mechanotransduction, which are important tasks for realizing practical applications of flexible devices in wearable states.

4.3 Electronic plants

Plants typically live in complex environments, and survive a wide variety of mechanical stresses through hierarchical architectures with rigid and soft materials, some of which have already been used as templates to fabricate structural materials for flexible devices, as shown in our previous discussion (Section **2.2**). Moreover, artificial electroactive materials with nanostructures have been directly applied in cells and the vascular systems of living plants to study the effect on plant physiology[660,661] and induced complex functions.[662-664] Inspired by the abundant vascular systems in plants, another strategy, called the electronic plant, has been conceived that advances plants as technological bioelectronic platforms and directly manufactures electronic materials in plant structures as integrated electronic systems.[665-667] For instance, after cutting off the lower part of a stem of *Rosa floribunda* (garden rose), the fresh cross section was immersed in an aqueous solution containing PEDOT-S:H, a PEDOT derivative with a covalently attached anionic side group.[668] A hydrogel-like and continuous PEDOT-S:H wire grew along the xylem channels due to the aqueous-rich environments with divalent cations (Figure 23a). The fabricated PEDOT-S:H wires showed an electronic conductivity of 0.13 S/cm and transistor functionality that can be further used as "xylem logic", with complex xylem-templated circuits in vivo performing NOR logic at the output (Figure 23b,c).[665] Due to the spongy mesophyll structure in rose leaves, PEDOT:PSS combined with nanofibrillar cellulose can also be deposited in the compartments via a solution-processed vacuum infiltration technique, forming a electrochromic display circuit with two-dimensional electrode networks.

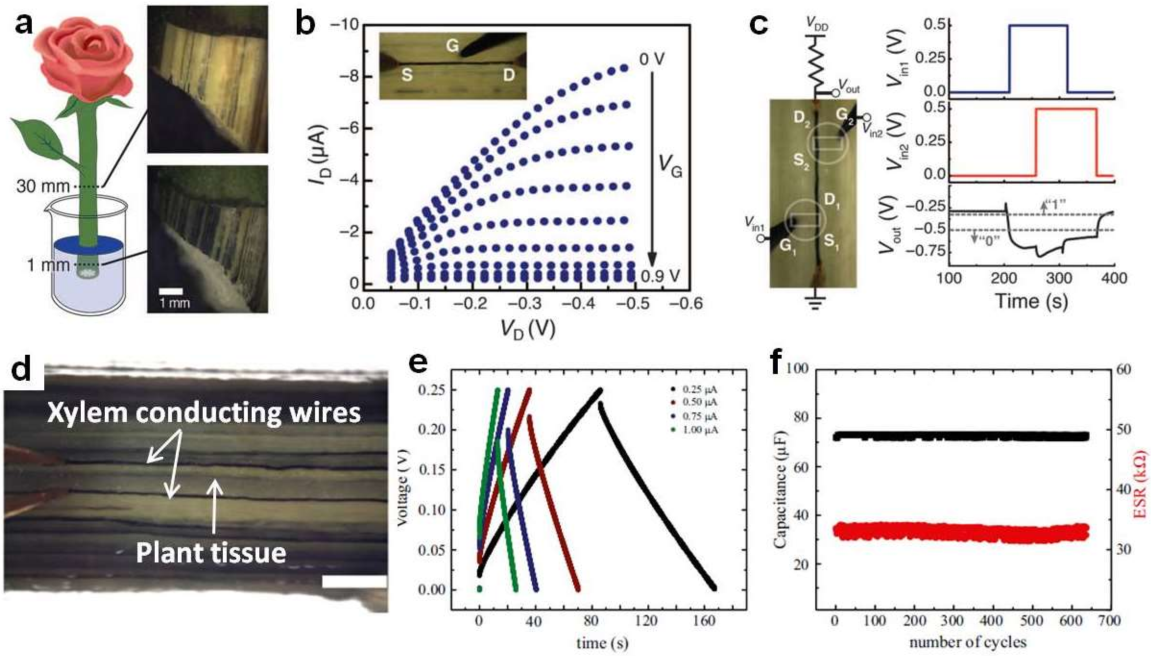


Figure 23. Electronic plant. a) Illustration of the fabrication of PEDOT-S:H wires in the xylem of a rose and microscopic observation of the wire formed 1 mm and 30 mm above the bottom b) Output of the xylem-based organic transistor. Inset shows the source, drain and gate. c) Optical image of the NOR logic gate constructed by the xylem wire, and its NOR function indicated by the voltage traces. Reproduced with permission from ref. 665. Copyright 2015 the American Association for the Advancement of Science. d) Microscope image of a rose supercapacitor (scale bar: 1 mm). Electrochemical characterization of the rose supercapacitor: e) typical charge-discharge curves, f) capacitance and ESR retention. Reproduced with permission from ref. 666. Copyright 2017 National Academy of Sciences.

Furthermore, besides electronic functionality achieved in localized regions, such as the xylem vascular tissue and leaves, a whole plant's structure can also serve as a physical template for the self-organization and polymerization of a water-soluble conjugated oligomer to realize a functional electronic circuit in living plants.[666] By immersing a rose cutting with a fresh cross-section in an aqueous solution containing sodium salt of a 3,4-ethylenedioxythiophene (EDOT) derivative, the oligomers can be distributed in every part of the xylem vascular tissue from the stem to leaves and flowers, then polymerized into long-range conductive wires through catalysis with reactive oxygen species in xylem vessels released from the parenchyma cells.[669] Based on the natural architecture in the stem, supercapacitors are fabricated in vivo with paralleled conducting xylem wires as separate electrodes, in which the surrounding plant tissue comprising cellular domains and extracellular space full of electrolyte will work as the separator, exhibiting the specific capacitance of 20 F/cm^3 (Figure 23d-f). This idea opens up a new strategy in which the physical structure and physiology of living beings can not only serve as a template but also as an integrated section of electronic circuits, which provides variety in electronic engineering and creates possibilities for new device concepts.

5 Outlook

The use of nature-inspired structural materials in flexible devices has already demonstrated remarkable efficiency in revolutionizing the electronic industry from device engineering to functional performance. From the aforementioned discussions, it can be deduced that there are two general strategies for designing structural materials for the realization of device performances. The first involves the engineering of the device structure to endow it with mechanical characteristics such as the ability to accommodate external strains and deformations. The second is the function-oriented structural design for intelligent device performance, especially bio-function mimesis. In the progress of future flexible devices, these ideas will play increasingly important roles due to the requirements of future flexible and deformable device for wearables and smart electronics. There is still room for improvement in the mechanical properties and functionalities of future flexible devices.

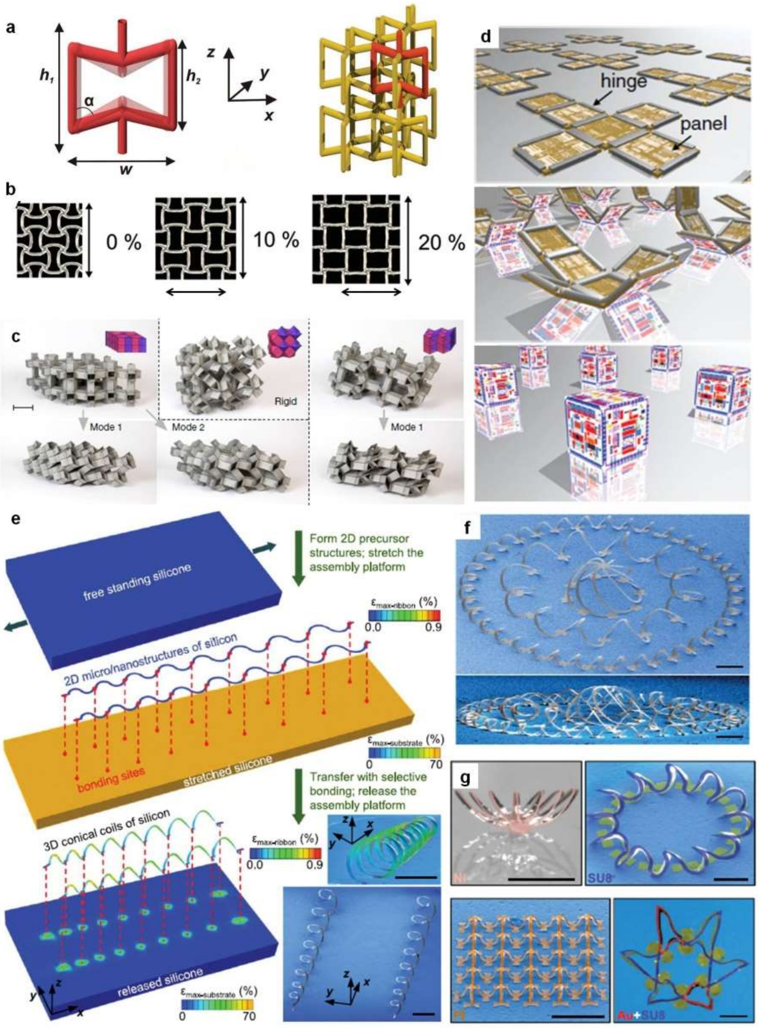


Figure 24. Metamaterials. a) Illustration of the bow-tie element and arranging it into a 3D crystal structure. Reproduced with permission from ref. 670. Copyright 2012 John Wiley and Sons. b) Response of the array of the pattern with a Poisson's ratio value of -0.8 under strains. Reproduced with permission from ref. 671. Copyright 2015 John Wiley and Sons. c) Illustration of 3D prismatic architected materials and their deformation modes. Reproduced with permission from ref. 672. Copyright 2017 Nature Publishing Group. d) Schematic illustration of a self-assembled 3D device: before (top), during (middle) and after assembly (bottom). Reproduced with permission from ref. 673. Copyright 2016 John Wiley and Sons. e) FEA simulation showing the formation of 3D conical helices from 2D silicon ribbons with certain points bonded to a pre-stretched silicone elastomer. Inset presents the SEM images of an experimental result. f) SEM images of complex 3D mesostructures formed from a 2D precursor selectively bonded on a biaxially stretched elastic

substrate (Scale bar: f,g) 400 μm). g) Various 3D mesostructures formed by metal (Ni), polymer (photodefinable epoxy (SU-8) and polyimide (PI)), and heterogeneous combinations of materials (Au and SU-8) (Scale bar: 500 μm). Reproduced with permission from ref. 674. Copyright 2015 the American Association for the Advancement of Science.

5.1 Structural materials for future flexible devices

Architectures that originate from the analysis of natural creatures by structural mechanics and mathematics, as well as from rational deductions, can be used to endow future electronic devices with distinctive mechanical characteristics. Currently, the development of metamaterials, the behaviors of which are governed not by material compositions but well-designed geometrical structures and the consequent interactions,[675-678] has drawn increasing attention in various scientific domains, such as optical metamaterials,[679] acoustic metamaterials,[680-685] auxetic materials,[670,671,686-689] and other mechanical metamaterials.[690-695] Despite this, efforts to incorporate metamaterials into flexible and stretchable electronic devices remain limited. However, metamaterials with unique mechanical characteristics would be valuable and instructive for the construction of functional flexible devices. For example, auxetic materials with a negative Poisson's ratio exhibit the ability to expand in the unstressed direction under uniaxial stretching.[677,687,689] Various models of 2D and 3D auxetic materials had been established and deeply studied[670,671] (Figure 24a,b), and these possess the potential to be promising candidates for the construction of flexible devices with unique mechanical sensing behaviors and stress-induced functions.

Another example of 2D and 3D reconfigurable architectural metamaterials is based on the ancient art of origami, which involves 2D folding patterns along pre-defined creases[696-703] or 3D-assembly from designed structures,[704-706] some of which possess applications in flexible electronic devices, as discussed in Section **2.1**. The incorporation of metamaterial design will enlarge the region of the available design space and extend the applicability towards more complex environments. Recently, constructions of 3D architectures on the nano/micro scale with electronic materials, including polymers, metals, and semiconductors, have drawn increasing attention due to their unique mechanical properties and promising prospects to push conventional 2D electronics into 3D devices (Figure 24d).[673,707-710] One powerful method to achieve the structuring is based on the compressive buckling strategy, which is discussed in Section **2.1**. With spatially selective bonding of planar precursor structures on a stretched elastic substrate, various mesostructures can be realized by strain release (Figure 24e).[674,711-715] The resulting processes suggest new opportunities for the geometric engineering of electronic materials and the realization of future 3D electronics (Figure 24f,g). Furthermore, a 3D reconfigurable architectural material showing multiple responses can be achieved by a periodic space-filling convex polyhedral connected with elastic hinges on polygon faces built by a stiff material.[672] The designed reconfigurable architected materials can perform a wide range of qualitatively different responses and degrees of freedom in different deformation modes by regulating the space-filling assemblies of the units and the nature (rigid or deformable) of the faces of the polyhedra, which are capable of adapting to changing environments as well as of tunable functionality (Figure 24c).

Apart from structural materials in the macro/microscopic world, structural designs on the molecular level would also have potential applications in constructing functional flexible devices.[716-718] Molecular machines, fabricated by molecules with rational structural design, have already been developed to perform various functions on the nanoscopic scale, such as nano-sized automobiles,[719-721] molecular pliers,[722] scissors,[723,724] and elevators[725,726]. The fascinating nature of this nano-sized machinery is the capability to respond to environmental stimuli by conformational changes of molecular structures and further give feedback to the macroscopic world,[727,728] which makes such materials promising candidates in building functional devices. Thus, by the rational integration of structural mechanics, molecular engineering, mathematics and techniques for flexible electronics, well-designed architectures with various mechanical properties would provide opportunities for future flexible devices to become even more advanced in the accommodation of complex strain, ability for transformation, and mechanical sensitivity.

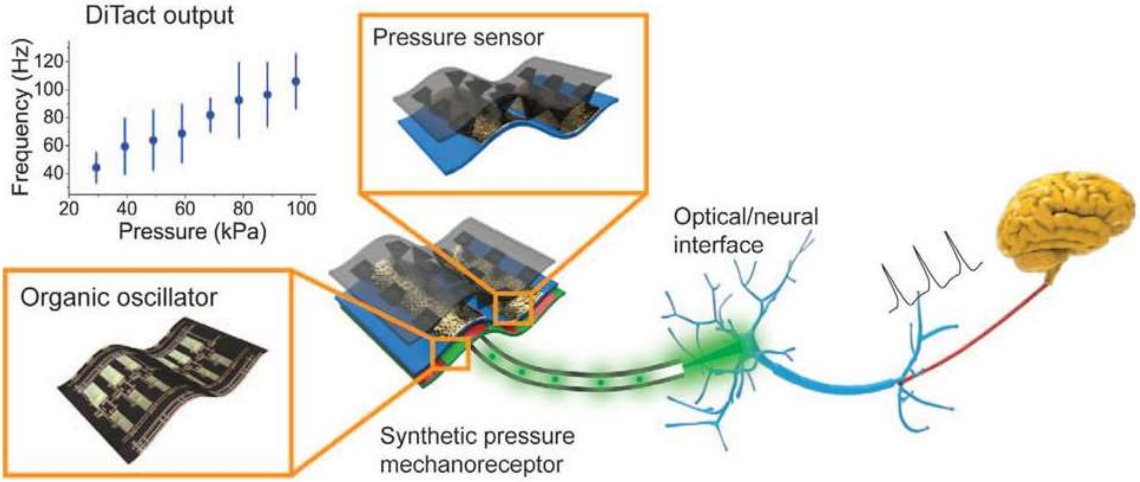


Figure 25. Artificial mechanoreceptor system composed of a pressure-sensitive tactile element and an organic ring oscillator; the optogenetic pulses can be used to stimulate live neurons. Reproduced with permission from ref. 729. Copyright 2015 the American Association for the Advancement of Science.

5.2 Nature-inspired integration of flexible devices

In both electronics and living creatures, realization of functionality is the ultimate objective of constructing and assembling structural materials. In particular, higher animals, including humans, can exhibit complex and intelligent interactions with external environments. Most of the complex behaviors demonstrated by humans are not accomplished by a single organ but by a rational and well-organized cooperation of multiple biofunctional units. For example, the bio-response to external stimuli is a smart organization of sensory, nervous, and locomotor systems that correspond to integrated functions, such as the input, transmission and processing of information, as well as actuation.[730-733] Thus, beyond the performance enhancement of an individual device according to structural design, function-oriented integration of various electronic devices for intelligent applications will become an essential topic for future electronics (discussed in Section **2.4** and **3.4**). To this end, structural design will become a basic and effective tool, not only for the improvement of the mechanical tolerance of a single flexible device but also for integrated and intelligent responses. Over the last several years, the development of intelligent devices that can mimic bioprocesses and exhibit unique responses to external stimuli has entered its infancy period and has drawn increasing attention in the field of flexible electronics.[303,729,734] For example, an artificial mechanoreceptor system that mimics the tactile mechanoreceptors in human skin can be realized via the rational combination of flexible microstructured resistive pressure sensors, voltage-controlled oscillators, and channelrhodopsin engineered specifically to enable optical neuron stimulation.[729] Analogous to the process of the transduction of mechanical strain to oscillating electrical action potentials by a biological mechanoreceptor, the system can detect a tactile pressure and transfer an electronic signal into frequency-encoded LED light, which can be used to stimulate channelrhodopsin to evoke action potentials (Figure 25). There is no doubt that function-oriented design will be an essential task in the construction of intelligent systems for future wearable electronics. To realize this concept in flexible devices, the strategies for structural design should not be limited to the engineering of a single electronic unit but should play an important role in systematic cooperation, showing complex and intelligent interactions with external environments that mimic bioresponses or accomplish even more direct and efficient artificial functions. Future flexible devices can be designed to work on a flexible surface in wearable states to perform smart interactions with physiological and environmental activities, as well as to generate intelligent electronic behaviors. To this end, highly evolved natural creatures constitute an important source of inspiration to enable the rational design of flexible electronics for robotics and biomechanical systems as they comprise numerous smart architectures and functional systems.

Notes

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TOC figure: