

Bioinspired Self-healing Soft Electronics

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Keywords: bioinspired electronics, self-healing, soft electronics, polymer, hydrogel

Inspired by nature, various self-healing materials which can recover their physical properties after external damage have been developed. Recently, self-healing materials have been widely used in electronic devices for improving durability and protecting the devices from failure during operation. Moreover, self-healing materials could integrate many other intriguing properties of biological systems, such as stretchability, mechanical toughness, adhesion, and structural coloration, providing additional fascinating experiences. All of these inspirations

have attracted extensive research on bioinspired self-healing soft electronics. In this review, a detailed discussion on bioinspired self-healing soft electronics is presented. Firstly, two main healing mechanisms are introduced. Then, four categories of self-healing materials in soft electronics, including insulators, semiconductors, electronic conductors, and ionic conductors are reviewed and their functions, working principles, and applications are summarized. Finally, human-inspired self-healing materials and animal-inspired self-healing materials as well as their applications, such as organic field-effect transistors (OFETs), pressure sensors, strain sensors, chemical sensors, triboelectric nanogenerators (TENGs), and soft actuators, are introduced. It is believed that this cutting-edge and promising field will stimulate more excellent cross-discipline works in materials science, flexible electronics, and novel sensors, accelerating the development of applications in human motion monitoring, environmental sensing, information transmission, etc.

1. Introduction

Electronic devices have dramatically revolutionized way people's daily life since their inception. Today, the development of electronic devices such as computers and mobile phones is moving towards miniaturization and flexibility. In addition, wearable and flexible soft electronics have attracted extensive attention due to the increasing emphasis on human health, the diversification of entertainment options, and the development of the Internet of Things technologies.^[1] For instance, Matsuhisa et al. reported a stretchable polymer diode and implemented an on-skin strain sensor and wireless display system,^[2] Lu et al. fabricated a flexible neural probe using AgNW-coated fiber,^[3] and Liu et al. constructed a soft TENG for energy harvesting based on a hybrid hydrogel-elastomer.^[4] Compared with traditional electronic devices, soft electronics can be seamlessly integrated with the human body or shape, which greatly expands their applications in healthcare monitoring,^[5] electronic skins,^[6] human-machine interface,^[7] and other fields.^[8,9] However, one concern in soft electronics is physical damage, such as cracks caused by external forces and aging fractures caused by long-term use. Even ductile polymer materials be damaged and fractured under cutting by sharp objects or large deformations. So functional materials are expected to be designed and developed to solve this problem. The emergence of self-healing materials provides a solution to this thorny problem. During the long evolutionary process in nature, animals and plants have acquired self-healing abilities. For example, the leaves of plants and the skin of animals can repair themselves after injury, which allows them to survive in harsh environments. Inspired by nature, self-healing materials that are capable of recovering their physical properties after being subjected to external damage could be adopted to improve the service life of soft electronics.^[10,11] This

special functional material was proposed in the 1980s and subsequently drew tremendous attention.^[12] The self-healing process is achieved through dynamic bond exchange or supramolecular interactions in the polymer chain segments, or by the release of encapsulated healing reagents. Research on self-healing materials has long been focused on mechanisms, fabrication and characterizations.^[13] Recently, the self-healing property has been integrated into soft electronics, blazing a promising trail in the field of self-healing soft electronics.^[14-17] These self-healing soft electronics including OFETs, TENGs, capacitors, and electronic skins have been developed for various scenarios such as strain sensing,^[18-20] human motion monitoring,^[21-24] energy harvesting,^[25-29] and wound healing.^[30,31]

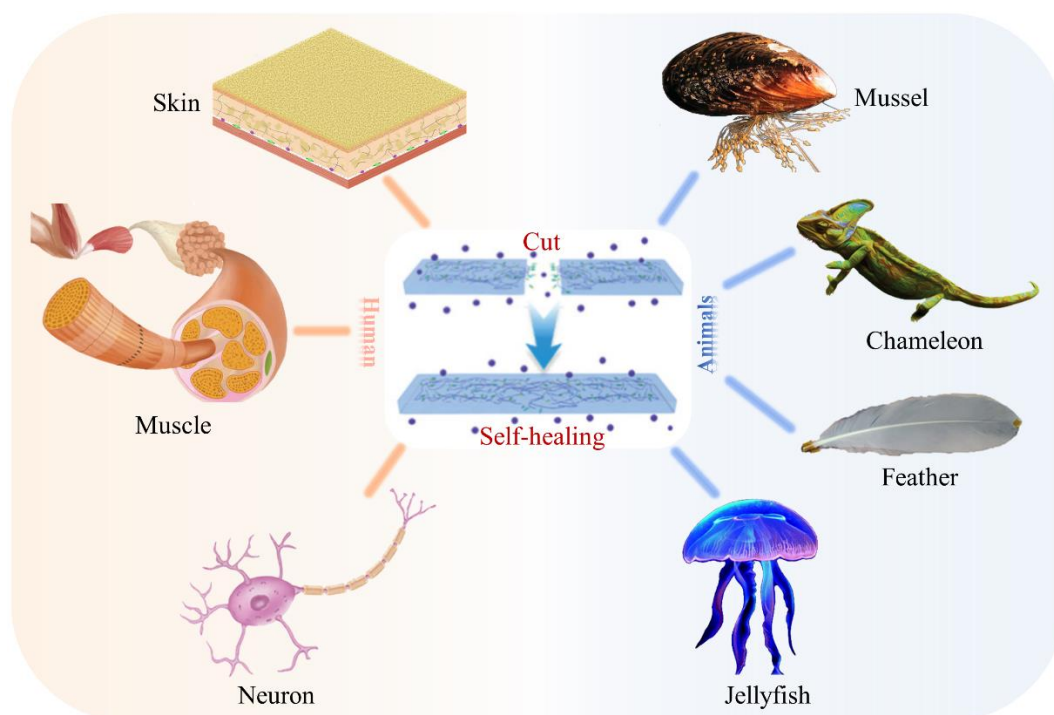


Figure 1. Schematic diagram of self-healing substances in nature depicted in this review. Self-healing of ionic gel. Reproduced with permission.^[32] Copyright 2021, Wiley-VCH GmbH. Stretchable human skin with a multilayer structure. Reproduced with permission.^[33] Copyright 2022, American Association for the Advancement of Science. Tough human muscle with fibrous structure. Reproduced with permission.^[19] Copyright 2019, American Chemical Society. The basic unit of the nerve: the neuron. Reproduced with permission.^[34] Copyright 2022, Wiley-VCH GmbH. Adhesive mussel byssus composed of hundreds of threads. Reproduced with permission.^[35] Copyright 2017, Company of Biologists Ltd. Chameleon with adjustable skin color. Reproduced with permission.^[36] Copyright 2020, Wiley-VCH GmbH. Feather with cascade slide-lock structure. Reproduced with permission.^[27] Copyright 2021, Wiley-VCH GmbH. Fluorescent jellyfish. Reproduced with permission.^[37] Copyright 2021, Royal Society of Chemistry.

Besides the self-healing property, various biological self-healing materials in nature could intrinsically own integrate other properties depending on their living environments and functions (Figure 1). Taking the human body as an example, human skin, as the largest sensory organ, is stretchable, resilient, and has the ability to perceive external stimuli.^[33,38,39] Human muscle, on the other hand, is composed of arranged collagen fibers and a hierarchy of bundles, which has high mechanical strength to perform various movements.^[19,40] The neuron is one of the structural and functional units of the nervous system, and its spatial axons can contact >1000 other neurons, enabling a powerful ion circuit.^[34,41] Furthermore, animal structures with self-healing abilities have evolved more characteristics for adapting to their survival environments. For example, mussels are natural adhesives that attach their soft bodies to the wet surface,^[35,42-46] chameleons can change color depending on the environment to protect themselves,^[36] feathers are lightweight and can repair themselves due to their cascade slide-lock system,^[47] while the beautiful jellyfish has green fluorescent protein, and the research on its light-emitting principle has won the Nobel Prize in Chemistry.^[48] All these fascinating properties have inspired researchers to develop unique self-healing materials. In this review, we firstly summarize fabrication strategies and mechanisms such as encapsulation of healing agents and introduction of supramolecular interactions for functional self-healing soft materials. The working principles of different types of self-healing soft electronic devices are introduced, including dielectric and semiconductor-based OFETs, electrical conductor-based TENGs, and ionic conductor-based sensors. Then a few typical bioinspired self-healing soft electronics are discussed in detail in terms of different characteristics, e.g. stretchability, mechanical toughness, adhesion, and structural color. Furthermore, the unique properties, large-scale preparation approaches spanning from films to three-dimensional structures, and potential applications of self-healing soft electronics are summarized and prospected. This review aims to provide a powerful reference for the development of bioinspired self-healing soft electronics that can adapt to different application scenarios. As a comprehensive research field, self-healing soft electronics will inspire more interdisciplinary research works on material synthesis, processing and shaping, device fabrication, and novel sensors.

2. Self-healing Mechanisms

Over the past 20 years, flourish research work have been focused on self-healing materials that can recover their physical properties upon being subjected to external damage.^[10,49] Numerous synthetic polymeric materials have been developed and applied in various fields, such as protective coatings,^[50] sensors,^[51] tissue engineering,^[52] and soft electronics.^[53]

The study on self-healing mechanisms is the basis for the design of numerous materials. Figure 2 depicts the two main self-healing mechanisms, including extrinsic and intrinsic approaches. In extrinsic approaches (Figure 2a), microcapsules containing healing agents such as reactive precursors and catalysts are encapsulated into the materials.^[53] When physically damaged, the liquids flow out to trigger chemical reactions and repair the materials. Obviously, the healing agents are consumed during the self-healing process, which restricts the repair after secondary damage. Intrinsic approaches solved this problem by the incorporation of supramolecular chemistry and dynamic bonds (Figure 2b). Supramolecular chemistry is “the chemistry of the intermolecular bond”, which focuses on molecular assemblies established on weak and reversible noncovalent interactions.^[54] Self-repair occurs through supramolecular interactions between the constituent molecules of the material. In self-healing soft electronics, the most commonly adopted noncovalent interactions are hydrogen bonds,^[25,55,56] metal-coordination bonds,^[57] ion-dipole interactions,^[58] etc. The hydrogen bond is an attraction between an H atom covalently bonded to a strongly electronegative atom such as an N, O, or F atom and another electronegative atom, which is the most widely used due to its abundant choice of materials.^[59] Metal-coordination bond is a strong noncovalent interaction in which a central metallic atom is surrounded by bound molecules or ions.^[60] In contrast to other noncovalent interactions, ion-dipole interaction is an electrostatic interaction between a charged ion and a molecule with a dipole that can be used to construct self-healing materials in aquatic environments.^[32] Figure 2b also shows a typical dynamic bond, the disulfide bond, which will undergo a reversible exchange of dynamic covalent bonds when damaged, allowing the material to self-repair.^[61] Notably, the above-mentioned chemical approaches can be arbitrarily combined to construct self-healing materials with fascinating characteristics.

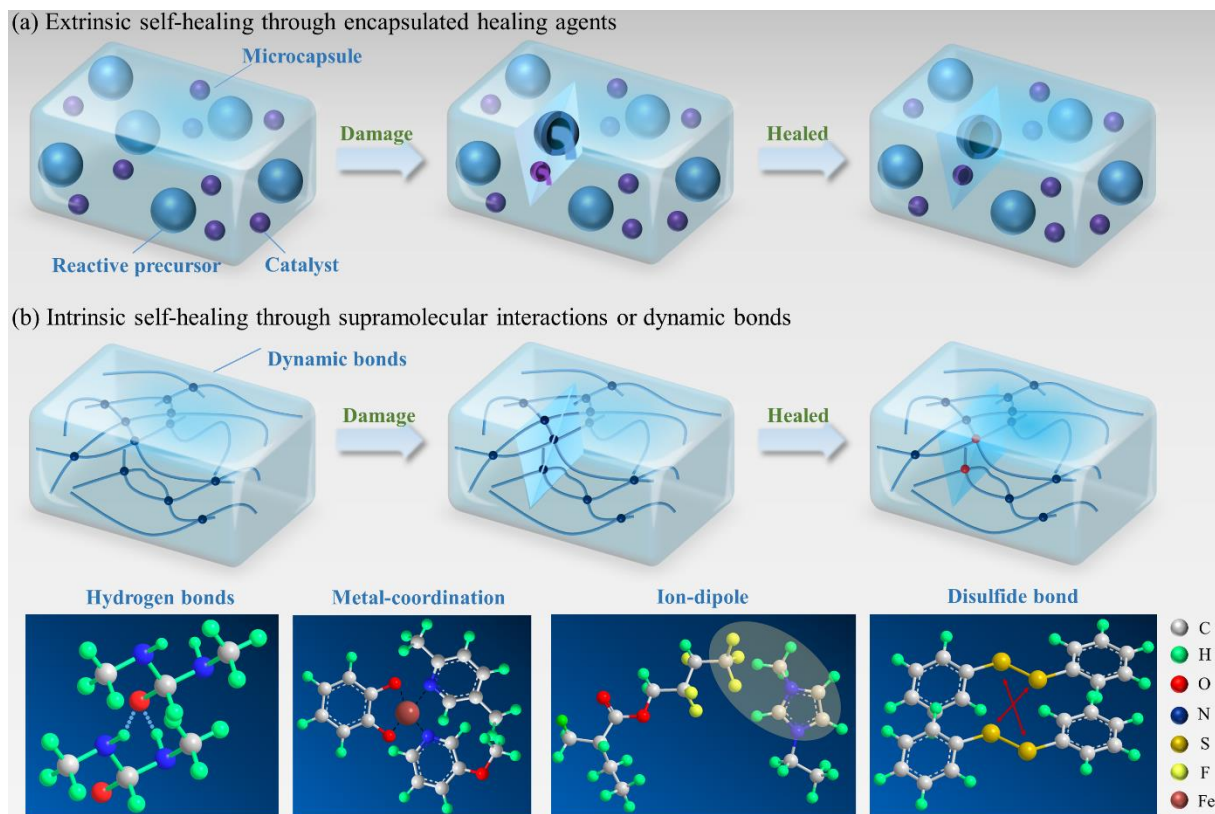


Figure 2. Schematic of self-healing mechanisms. a) Extrinsic self-healing through encapsulated healing agents. Upon damage, the microcapsules rupture and release healing agents to heal the materials. b) Intrinsic self-healing through supramolecular chemistry or dynamic bonds. Reversible supramolecular interactions or dynamic covalent bond exchanges allow the material to be repaired when it is re-spliced after damage.

3. Self-healing materials in soft electronics

Based on the aforementioned self-healing mechanisms, numerous self-healing materials have been prepared and studied. Recently, self-healing materials have been applied to soft electronics to improve device durability and lifetime. The materials in electronic devices are mainly divided into insulators, semiconductors, and conductors, among which conductors are subdivided into electronic and ionic conductors. Ideally, in a multicomponent device, all components should be self-healing, and the components should have compatible mechanical properties and surface chemistry to prevent the device from failing due to delamination in the event of strain. In this section, the materials, representative working mechanisms, and applications of each component are discussed.

3.1. Insulators

Since most polymers are insulating, the development of self-healing insulators actually predates the emergence of self-healing electronics. Figure 3a i) shows a self-healing insulator constructed by dipole-dipole interactions between polymer chains.^[62] This insulator can be used

as an encapsulation layer for an electrical conductor. The conductivity performance of this self-healing conductor is demonstrated through a lighting LED in the circuit. When the severed material reconnects and repairs itself, the circuit becomes conductive again and the LED is lighted up. In addition to encapsulation layers or protective coatings, self-healing insulators can be used as dielectrics.^[49] Dielectrics are insulators that can be polarized by the applied external electric field and the polarization mechanism is schematically shown in Figure 3a ii). Under the action of an external electric field, the positive charge in the dielectric migrates in the direction of the electric field, while the negative charge migrates in the opposite direction. This results in dielectric to be polarized, thus creating a reactive electric field that weakens the electric field inside the dielectric. Rao et al. fabricated a stretchable self-healing polymer through metal-coordination interactions, which was then used as a gate dielectric for an OFET.^[63] The metal not only plays the role of cross-linker but also improves the materials' dielectric constant. As shown in Figure 3a iii), the OFET device can be stretched to 100% strain. However, since the other parts are not self-healing, researchers have only investigated the self-healing ability of the polymer, not the integrated OFET device.

3.2. Semiconductors

Semiconductors play an important role in the fabrication of OFETs, however, fully self-healing semiconductors are difficult to achieve due to the thickness that limits re-splicing in practice (usually <100 nm). As shown in Figure 3b i), Oh et al. reported the first healable semiconducting polymer by incorporating hydrogen bonds into the amorphous polymer segments between crystal-conjugated segments.^[64] Nanocracks formed by fatigue can be healed by heating and solvent annealing to promote the movement of polymer chains. The research group further fabricated a semiconducting polymer that can self-repair without external forces through metal-coordination interactions.^[65] The cut and recovery test of this self-healing semiconductor is conducted by attaching it to a pre-cut PDMS substrate. Breakage was caused by bending the semiconducting film/PDMS stamp, and the self-healing process started after the PDMS was restored to flatness. Figure 3b iii) shows a photograph of the stretchable active-matrix transistor array based on this semiconductor. In short, in order to fabricate self-healing semiconductors, soft segments must be introduced to enhance the mobility of the chains while preserving the charge transport capability of semi-crystalline conjugated chains.

3.3. Electronic conductors

Electronic conductors in self-healing soft electronics are usually constructed by filling conductive nanofillers, such as silver nanowires, silver nanoparticles, carbon nanotubes, and graphene, into self-healing polymers or hydrogels (Figure 3c i).^[66-68] A typical application of

self-healing conductor-based soft electronics is self-healing TENG. As depicted in Figure 3c ii), when a force is applied, the triboelectrically negative material and triboelectrically positive material come into contact, and the charge transfer from the positive material to the negative material, producing equivalent negative and positive charges on both surfaces. When separated, the negative charge on the surface of triboelectrically negative material induces the accumulation of positive charges, and vice versa. The electric current is generated due to the presence of potential difference till potential difference reaches equilibrium. When the mechanical force is applied again, the distance between the two materials decreases, causing the charge to flow in the opposite direction.^[69,70] Figure 3c iii) shows a photograph of a nanocomposite conductor with its conductivity after self-healing confirmed by lighting an LED.^[71] A noteworthy issue is that the incorporation of nanofillers may sacrifice the mechanical strength and self-healing ability of the polymer or hydrogel, but insufficient filling may result in poor conductivity, so it is important to master the balance.

3.4. Ionic conductors

Ionic conductors are usually stretchable and transparent, made from small molecular or polymer gels swollen with ionic liquids.^[72,73] Although ionic conductors typically have higher resistance than electronic conductors, they are still widely studied because the biological systems are often based on the movement of ions rather than electrons. Figure 3d i) is a schematic diagram of an ionic conductor constructed by ion-dipole interactions between the polymer network and ionic liquid.^[58] Kim et al. reported an ionic touch panel constructed by an ionic conductor (Figure 3d iii)), which is soft, transparent, and stretchable.^[74] The working mechanism of the ionic touch panel is shown in Figure 3d ii). When being touched, a closed circuit is formed because the human hand is grounded, and the current will increase by flowing from both ends of the strip to the touch point. The touch panel can be attached to the human arm by a VHB film and then apply to perceive various motions via recording changes in electrical current. Table 1 summarizes typical self-healing materials reported in recent years, including their compositions, self-healing mechanisms, and applications in soft electronics.

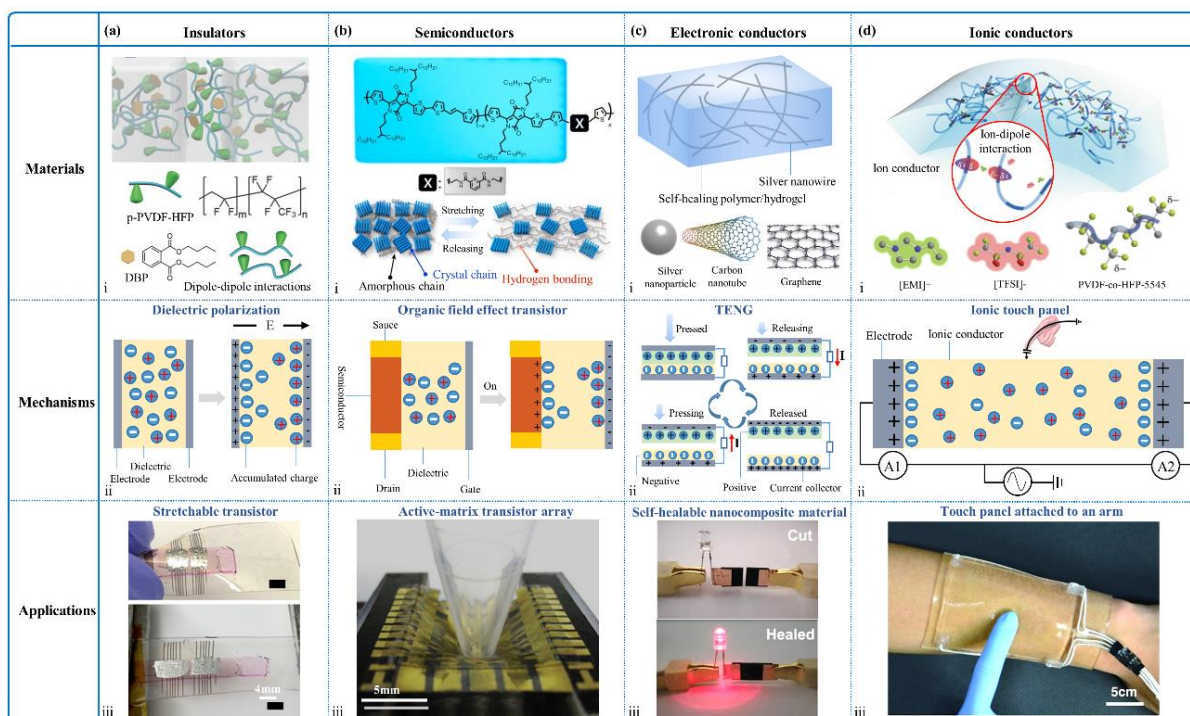


Figure 3. Materials, working mechanisms, and applications of self-healing materials in soft electronics. a) Insulators composed of polymers are used as dielectric layers in OFETs and their dielectric polarization mechanisms are described. Reproduced with permission.^[62] Copyright 2018, Wiley-VCH GmbH. Reproduced with permission.^[63] Copyright 2016, American Chemical Society. b) Semiconductors composed of conjugated polymers are used to construct OFETs, and the working mechanism of OFET is introduced. Reproduced with permission.^[64] Copyright 2016, Springer Nature. Reproduced with permission.^[65] Copyright 2019, American Association for the Advancement of Science. c) Electronic conductors are usually composed of conductive nanoparticle-polymer composite systems, and their working mechanism for TENG application is sketched. Reproduced with permission.^[71] Copyright 2020, Wiley-VCH GmbH. d) Ionic conductors composed of the ionic hydrogel are used as a touch panel, and the working mechanism of the device is presented. Reproduced with permission.^[58] Copyright 2019, Springer Nature. Reproduced with permission.^[74] Copyright 2016, American Association for the Advancement of Science.

Table 1. Typical compositions, self-healing mechanisms, and applications of self-healing materials in insulators, semiconductors, electronic conductors, and ionic conductors.

Function	Composition	Self-healing mechanism	Application	ref.
Insulator	PHPMA/PEI blend polymer	Hydrogen bond	OFET and chemical sensor	[75]
Insulator	poly(urea-urethane) (PUU)	Disulfide bond	Transistor	[76]
Insulator	FeCl ₂ -PDMS	Metal-coordination	OFET	[63]
Insulator	p-PVDF-HFP-DBP complex	Dipole-dipole interaction	Encapsulation layer	[62]
Insulator	PTMEG/HMDI/HEDS elastomer	Disulfide bond and hydrogen bond	Triboelectric skin	[28]
Insulator	DA-PU elastomer	Donor-Acceptor interaction	Dielectric and packing layers	[77]
Semiconductor	DPP/PDCA copolymer	Hydrogen bond	OFET	[64]
Semiconductor	DPP-TVT-PDCA/PDMS-PDCA-Fe elastomer	Metal-coordination	OFET	[65]
Semiconductor	PDPP _{urethane} Copolymer	Hydrogen bond	OFET	[78]
Electronic conductor	μNi particles doped complex	Hydrogen bond	Pressure sensor	[56]
Electronic conductor	PU-DHP/Fe ₂	Hydrogen bond and metal-coordination	TENG	[57]
Electronic conductor	AgNWs embedded MDPB-FGEEDR copolymer	Diels-Alder reaction	Composite conductor	[66]
Electronic conductor	AgNWs embedded PBPUU	Hydrogen bond and disulfide bond	E-skin	[79]
Electronic conductor	CNT embedded PDMS-MPU _{0.4} -IU _{0.6}	Hydrogen bond	E-skin	[67]
Electronic conductor	CNTs-PDA fillers doped PU inverse opal matrix	Hydrogen bond	Human-motion sensor	[36]
Ionic conductor	PNA/PMA	Hydrogen bond	Strain sensing fiber and TENG	[25]
Ionic conductor	Poly[MATAC][TFSI] in [N4111][TFSI] ionic liquid	Ion-dipole interaction	Underwater communication	[32]
Ionic conductor	PVDF-co-HFP-5545 in ionic liquid	Ion-dipole interaction	Pressure sensor	[58]
Ionic conductor	Na ⁺ and (B(OH) ₄) ⁻ ions in PVA	Hydrogen bond	Ionic skin TENG	[69]
Ionic conductor	PVDF-co-HFP-5545/EMIOTf materials	Ion-dipole interaction	Soft actuator	[72]
Ionic conductor	PPBN-hydrogel	Hydrogen bond and metal-coordination	Strain and temperature sensor	[19]
Ionic conductor	Ionic liquids impregnated PU network	Hydrogen bond and dynamic urea bond	Ionic skin	[80]
Ionic conductor	PU scaffold in ionic liquid	Hydrogen bond and disulfide bond	Ionic skin	[81]

4. Human-inspired self-healing soft electronics

In addition to self-healing properties, various biological self-healing materials in nature also have other unique properties according to their living environments and functions. In recent years, biomimetic materials prepared by learning lessons from the biological world have attracted extensive research attention.^[82,83] Humans are one of nature's most amazing creations, with skin that protect the entire body and sense external environment, bones that support the body, muscles that perform movement, and nerves that connect the entire body to transmit signals. In addition, the human body has a strong ability to heal itself and can regenerate after injuries.^[84] This has prompted scientists to explore self-healing materials with stretchability, toughness, sensing capabilities, etc. In this section, human-inspired self-healing soft electronics are presented, these devices are inspired by the human body and are used to improve human life.

4.1. Skin-inspired self-healing soft electronics

As the human body's largest organ, the skin has various beneficial merits such as stretchability, elasticity, self-healing, and superior sensory capabilities.^[85-88] Figure 4d depicts the hierarchical structure of human skin, with a waterproof epidermis that provides protection, a dermis that contains elastic tissue and fibers that give it resilience and toughness, and a hypodermis that deforms to attenuate external pressure.^[81]

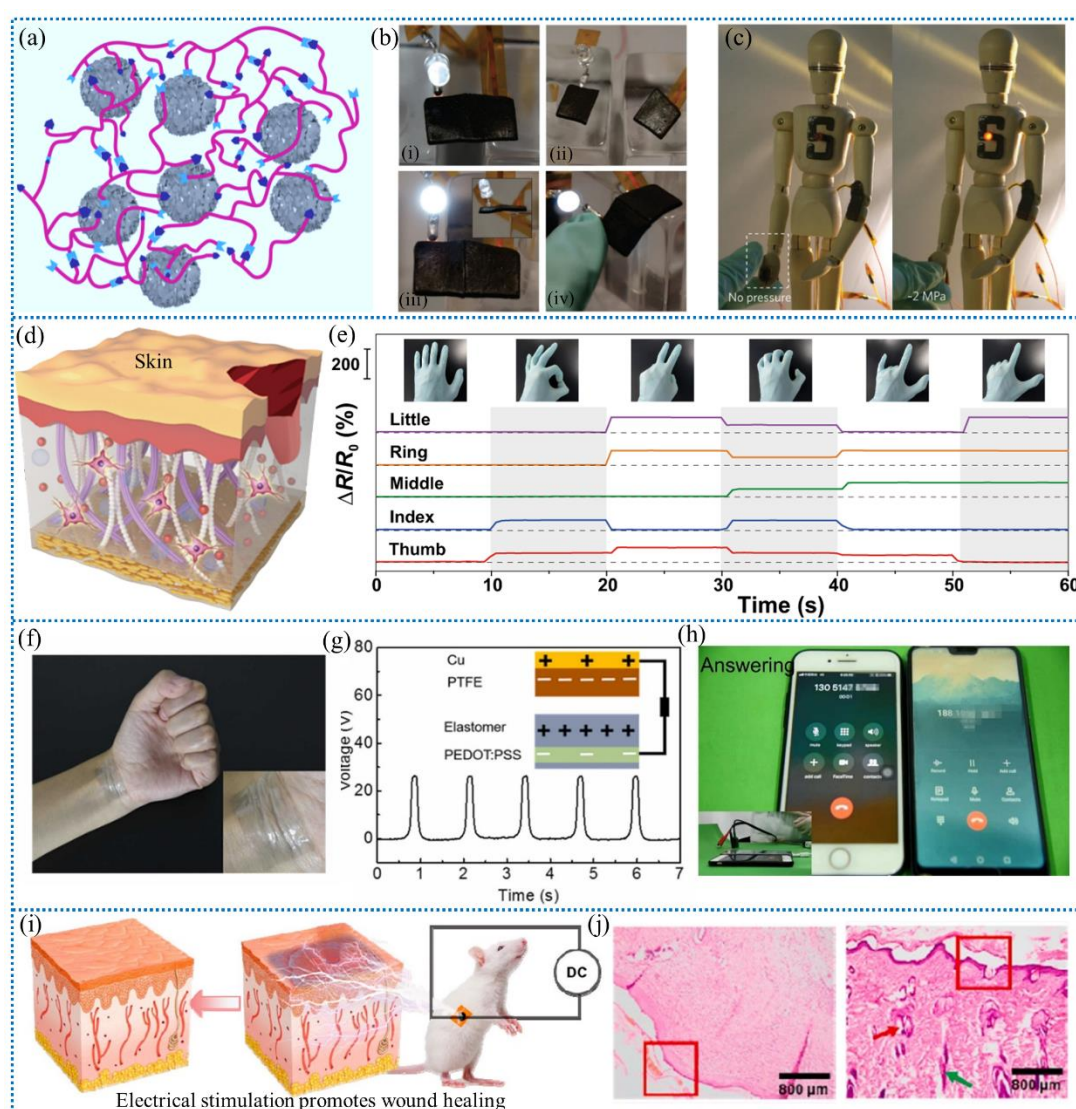


Figure 4. Skin-inspired self-healing soft electronics. a) Schematic of the self-healing electronic skin constructed from supramolecular organic-inorganic composite. b) Photographs of the electric skin with an LED in series at pristine (i), damaged (ii), and healed (iii) states. iv) The flexibility of the healed electric skin. c) The electric skin is used as a pressure sensor. a-c) Reproduced with permission.^[56] Copyright 2012, Springer Nature. d) Schematic of the ion-rich structure of human skin. e) The ionic skin is used for gesture monitoring. d,e) Reproduced with permission.^[81] Copyright 2022, Springer Nature. f) Photo of the transparent self-healing

electronic skin attached to a wrist. g) Voltage of the TENG constructed by triboelectric skin and PTFE/Cu. h) After attaching the TENG to the screen protector, the phone call can be answered by waving a hand above the screen. f-h) Reproduced with permission.^[28] Copyright Year, Publisher. i) Schematic illustration of skin-inspired bio-electronic hydrogel used to promote wound healing after electrical stimulation. j) H&E staining of injured rat skin after 20 days. Left: control group (transparent gelatin hydrogel); right: experimental group (bio-electronic hydrogel with electrical stimulation). Red arrow: blood vessel; green arrow: hair follicle. i,j) Reproduced with permission.^[31] Copyright 2021, Elsevier.

Inspired by biological skins, generous efforts have been invested in developing artificial skins, including electronic skins and ionic skins. Electronic skins can mimic human skin sensation and introduce self-healing features to enhance its durability and longevity. Tee et al. reported the first room-temperature self-healing electronic skin prepared by doping μNi particles in a random branched polymer network connected by hydrogen bonds (Figure 4a).^[56] The electrical conductivity of the organic-inorganic composite can be tuned by the content of μNi particles. Due to the enhancement of mechanical strength by μNi particles, the composite can be compression moulded into thin films. As shown in Figure 4b, a LED in series with the conductive composite is illuminated when the power is turned on. After cutting the composite material, the LED is extinguished, and after 5 minutes of self-healing at room temperature, the LED light up again. The inset of Figure 4b iii) shows that the composite is self-supporting, while Figure 4b iv) displays its flexibility. The researchers then used the composite as a piezoresistive sensor, as shown in Figure 4c. When pressure is applied, the distance between the μNi particles decreased, resulting in a decrease in the resistance of the composite, which ultimately led to an increase in the brightness of the LED connected in series with it.

Ionic skins have also been widely studied and applied in sensing fields.^[21,80,89-91] Recently, Wang et al. fabricated a fatigue-free ionic skin by embedding a polyurethane (PU) scaffold containing disulfide bonds into an ionic matrix containing hydrogen bonds.^[81] The self-healing PU was electrospun and collected to obtain PU nanomesh, and then the PU nanomesh was sandwiched by two silicone spacers, immersed in a pre-gel solution, and evaporated to obtain the hybrid ionic skin. The nanofibrous hybrid structure endows ionic skin with superior mechanical properties, which is both soft and firm like human skin. The ionic skin can withstand fracture stress over 100 MPa and reach a fracture strain of 680%. Due to the concurrent healing processes of nanofibers and ionic matrix, the ion conductivity and mechanical strength of the ionic skin can self-heal. They adopted this ionic skin as a wearable sensor to monitor human hand gestures. As shown in Figure 4e, five ionic skins adhere to five

fingers as independent channels, and finger bending causes the ionic skins to elongate, resulting in an increase in the resistance. Thus, the combined result of the five channels can be used for real-time gesture monitoring. The authors also compared the sensory capabilities of the ionic skin and other common conducting strain sensors, indicating superior sensing performance due to the moisture breathing of the ionic matrix.

In addition to sensors, skin-inspired self-healing electronics have also been used in TENG.^[69,92] Liu et al. synthesized a self-healing elastomer by mixing a hard phase containing disulfide bonds with a soft matrix containing hydrogen bonds.^[28] Different samples with thicknesses ranging from 3 to 100 μm were fabricated by the spin-coating method. Subsequently, a triboelectric skin was fabricated using the elastomer as a triboelectric layer, poly(3,4-ethylenedioxythiophene): polystyrene sulfonate (PEDOT:PSS) as an electrode, and polystyrene nanospheres as an interlayer. Figure 4f is a photograph of the triboelectric skin attached to a human wrist, which can be seen to be ultrathin and transparent. Using PTFE as triboelectrically negative material, the prepared TENG generates an alternating voltage during the contact-separation process (Figure 4g). The TENG output voltages before cutting and outputs after self-healing for 5 minutes are nearly identical, indicating the fast self-healing ability at the device level. Finally, a screen protector with TENG is attached to the phone. Waving the hand above the screen can generate voltage, and by reasonable signal processing and software design, non-contact waving action can answer the phone call (Figure 4h).

Also, skin-inspired self-healing electronics have recently been used for wound healing promotion (Figure 4i).^[31,93,94] Zheng et al. prepared a gelatin-based hydrogel containing PEDOT:PSS and MWCNTs to enhance the conductivity.^[31] The as-fabricated hydrogel can self-heal in 10 min, which benefits from the hydrogen bonds, electrostatic interactions, and π - π stacking interactions. The hydrogel also possesses movement monitoring capability when attached to the human joint, as its resistance increases with elongation. The motion tracking function is considered beneficial for wound healing because it records the epidermal changes in the wound injury area. Thus, hydrogel is used for wound healing promotion. As shown in Figure 4g, a wound treated with the conductive hydrogel and electrical stimulation for 20 days shows new blood vessels and hair follicles compared to the transparent gelatin hydrogel (left) control, indicating that the conductive hydrogel efficiently promoted the wound healing. This enables the hydrogel not only a strain sensor but also a scaffold for wound healing.

4.2. Muscle-inspired self-healing soft electronics

Compared to stretchable skins, which act as a protective layer and sensory organ, muscles have a higher mechanical strength and pull bones by contracting to produce joint movements. Figure

5a displays the structure of a human muscle in which the densely connected epimysium contains many bundles (fascicles) of muscle fibers, giving it robust mechanical properties.^[19] Inspired by the muscle structure, Ge et al. incorporated polyaniline nanofibers (PANI NFs) into poly(acrylic acid) (PAA) hydrogel to fabricate a multifunctional self-healing hydrogel through hydrogen bonds and metal-coordination interactions. The inset of Figure 5b shows a TEM image of PANI NFs with a structure similar to a neuron's matrix. With the addition of glycerol, the PAA-PANI binary networked-hydrogel (PPBN-hydrogel) exhibits excellent freeze resistance and remains conductive at -26 °C. The PPBN-hydrogel can withstand fracture stress over 35 kPa and reach a fracture strain of nearly 1000%. After 6 hours of healing at room temperature, the healed strain can reach 90% of the original strain. This hydrogel can be used as a strain sensor, and its output signal is shown in Figure 5b. The resistance of the PPBN-hydrogel increases exponentially with strain compared to PAA unitary networked-hydrogel (PUN-hydrogel) due to the embedding of PANI NFs to form a porous network. When the cavity apertures are squeezed under large strains, the cross-linked percolation joints rupture, hindering the conductivity path and causing a significant change in resistance. In addition, the resistance of the PPBN-hydrogel decreases with increasing temperature, showing a linear response with high sensitivity and a wide sensing range. This is achieved by using one-dimensional conducting PANI NFs as channel materials. The hydrogel is then attached to the human forehead as a temperature sensor for the "fever indicator" (Figure 5c), and the inset of Figure 5c shows the heat distribution of the object.

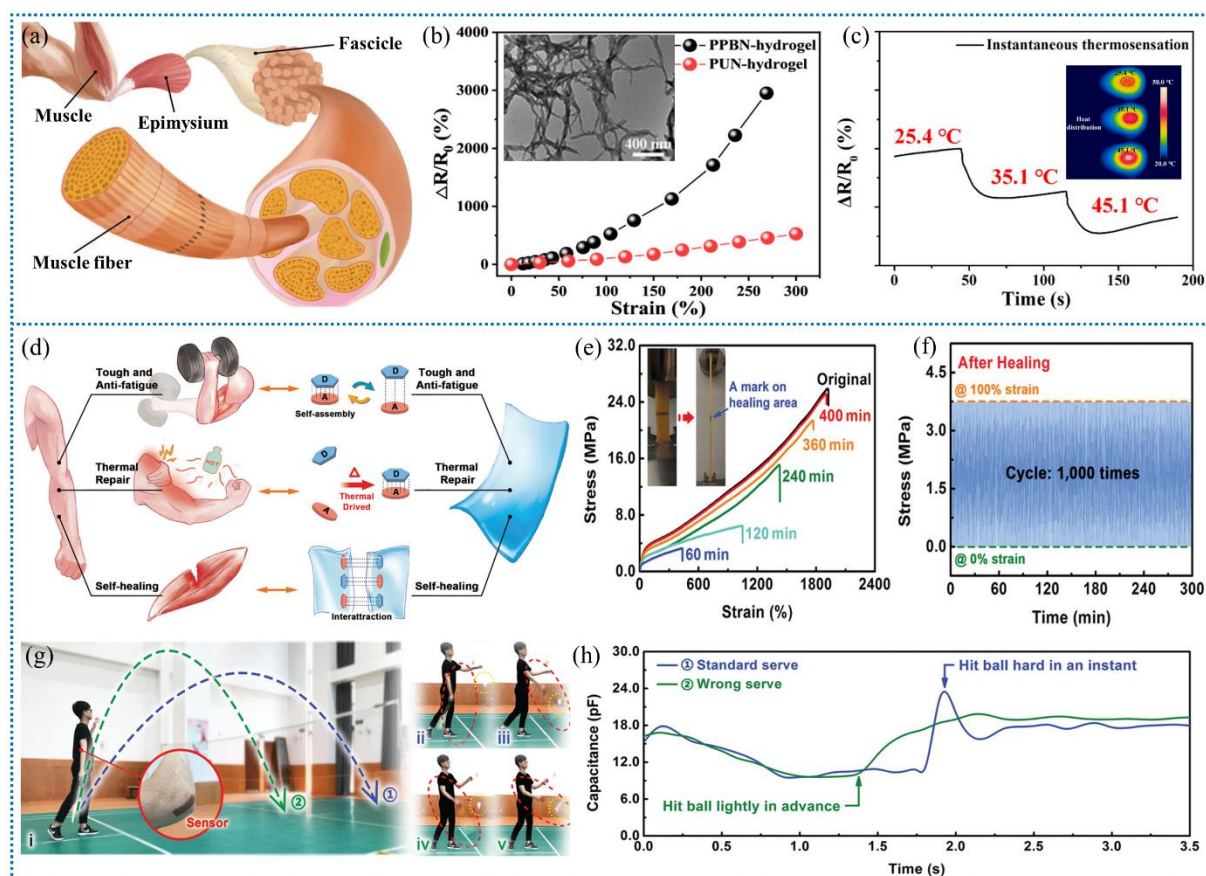


Figure 5. Muscle-inspired self-healing soft electronics. a) Schematic diagram of the human muscle structure, in which the epimysium is tightly connected and contains many bundles (fascicles) of muscle fibers. b) Resistance variation of the PPBN- and PUN-hydrogel to strain. The resistance of the PPBN-hydrogel increases exponentially with strain, while the sensitivity of PUN-hydrogel is lower. Inset: TEM image of PANI nanofibers. c) The resistance of the hydrogel temperature sensor changes with increasing temperature. Inset: Heat distribution of the monitored human forehead. a-c) Reproduced with permission.^[19] Copyright 2019, American Chemical Society. d) Schematic diagram of the muscle-inspired self-healing elastomer. D-A self-assembly endows the elastomer with muscle-like toughness, thermal repair, and self-healing capabilities. e) Stress-strain curves of the elastomers with different healing times. The mechanical properties are almost completely recovered after self-healing for 400 minutes. f) Cycling performance of the elastomer. g) i) Standard and wrong badminton serve. ii-v) Standard (ii, iii) and wrong (iv, v) acts of exerting force. h) Capacitance changes of the elastomer-based sensor in standard and wrong badminton serve. d-h) Reproduced with permission.^[77] Copyright 2021, Wiley-VCH GmbH.

However, due to the inherent properties of hydrogels, the mechanical strength of the PPBN-hydrogel is not ideal, which is inevitably reduced by the presence of solvents such as water and glycerol. Polymer elastomers provide a solution to this problem with stretchability, toughness,

and self-healing properties for durable electronics. Ying et al. reported a self-healing PU self-assembled from a donor-acceptor (D-A) similar to the skeletal muscle protein titin (Figure 5d).^[77] The DA-PU possesses super-tough mechanical properties, which can withstand a fracture stress of ~26 MPa and reach a fracture strain of 1900%. After being cut, the mechanical property can be recovered to 97% after 400 min (Figure 5e). The researchers also conducted cycle tensile tests on the self-healed DA-PU, revealing its superior fatigue resistance (Figure 5f). Finally, a capacitive sensor is fabricated using DA-PU as the dielectric and packing layers, and Cu powder containing GaInSn liquid metal as the conductive layer. When stretched, the distance between the two conductive layers decreased, increasing the capacitance. There is a linear relationship between the capacitance and stretching strain for both original and healed sensor devices. The sensor is then mounted on the human elbow to identify the correct and incorrect movements of badminton players (Figure 5g i)). As shown in Figure 5g ii) and iii), for the standard serve movements, the player keeps the elbow vertical before hitting the ball and fires the ball quickly at impact, and the recorded capacitance curve reaches its highest after balance. For the wrong action (Figure 5g iv) and v)), the elbow is bent in advance and the ball is hit slowly, causing the capacitance curve to stabilize after a slow rise (Figure 5h). This work has made great progress in tough and self-healing soft electronics by employing polymer elastomers.

4.3. Neuron-inspired self-healing soft electronics

Neurons are basic units for transmitting physical signals, and spatial axons allow them to communicate with more than 1000 other neurons. Inspired by this, Chen et al. synthesized a self-healing telechelic polyurea (TP), in which commercial polyether amines (D400 and D2000) were selected as soft segments and connected by interchain quadruple and double hydrogen bonds (Figure 6a).^[41] The three-arm architecture ensures that the molecular chains are not too long, like neuronal units, and that the terminal hydrogen bonds act like spatial axons, which guarantees the high modulus and strength of the synthetic polymer without sacrificing self-healing abilities. Keeping the number of D2000 molecules as 1, Figure 6b shows the stress-strain curves of TPs with a different number of D400 molecules. As the D400 increase, the hard segment content increases, resulting in a higher Young's modulus. Taking TP3 with the best overall mechanical properties as an example, the specimen displays great crack tolerance (Figure 6c). The synthesized polymer can self-heal within 30 minutes and the self-healing performance will improve with increasing temperature due to reversible hydrogen bonds. Benefiting from the neuron-like structure, TP3 also exhibits record-high adhesion and superior triboelectricity, demonstrating great potential for applications in soft electronics.

In addition to neuron-inspired polymer structures, Wang et al. reported an ultra-stretchable and fast self-healing ionic hydrogel (SSIH) with applications inspired by nerves.^[34] The ionic hydrogel is constructed by hydrogen bonds and electronic interactions between the polymers. Figure 6d shows that the transparency and flexibility of the hydrogel increase with Li^+ content, this is due to the breakage of the oriented arrangement of hydrogen bonds caused by Li^+ , which promotes the reconstruction of continuous active dynamic interactions and the diffusion of polymer segments. Thus, SSIH can self-heal in cryogenic environments, which gives it a broader application potential than other self-healing materials. As mentioned above, myelinated axons in neurons help to send ionic signals to organs and tissues quickly. Figure 6e i) depicts the structure of axons in which saline solution is surrounded by an insulating myelin sheath that forms capacitors to transfer nerve impulses. Inspired by this, the researchers fabricated an ultra-stretchable and self-healing artificial nerve fiber (SSANF) by using SSIHs to simulate cytosol as electrolyte and dielectric elastomer VHB to simulate myelin sheath. As shown in Figure 6e ii), when the signal is input, an electrical double layer (EDL) is formed at the interface between the electrode and ionic hydrogel, and then the EDL transmits along with the device to the output ports. Figure 6f displays the information transmission fidelity of SSANF, where V_1 and V_0 represent the output and input voltages, respectively, and $|V_1/V_0|$ remains between 0.975 and 1.025, indicating that the signal is output faithfully. SSANF is further applied in biomimetic intelligent robots, as shown in Figure 6g, where the input port of SSANF is connected to a somatosensory glove and the output port is connected to the robotic hand. By recognizing and encoding the gestures, the processed signals are transmitted by SSANF to the robot hand to decode and perform the same actions. The dry ice proves that SSANF can work in cryogenic environments. The artificial nerve fiber can withstand large strains, which is better than traditional metallic wires.

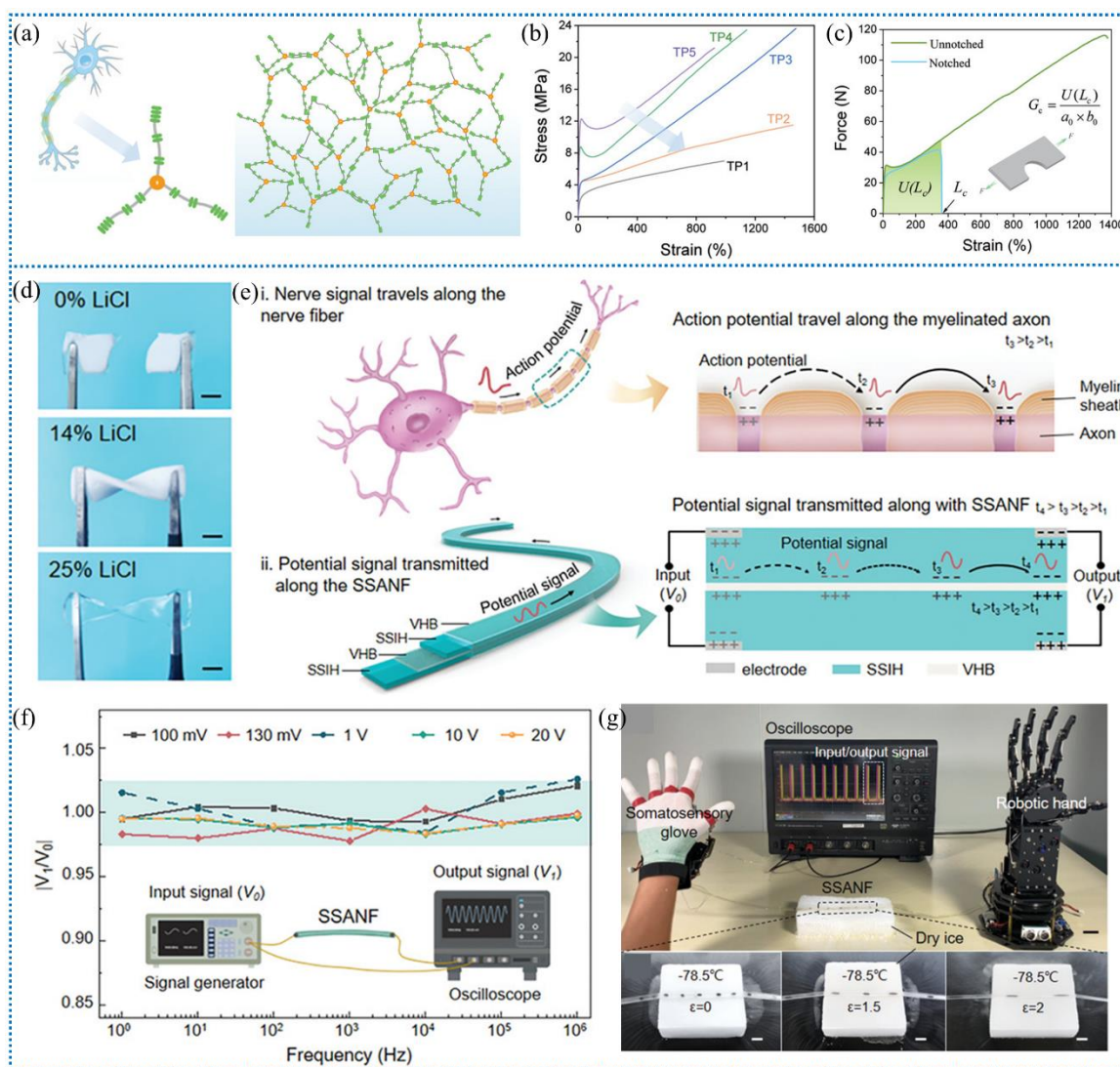


Figure 6. Neuron-inspired self-healing soft electronics. a) Schematic of the neuron-inspired self-healing telechelic polyurea (TP). b) Stress-strain curves of TP_x, with TP3 exhibiting the best overall mechanical properties. c) Force–displacement curves for notched and unnotched TP3 samples, showing great crack tolerance. a-c) Reproduced with permission.^[41] Copyright 2022, Wiley-VCH GmbH. d) Self-Healing ionic hydrogels with different LiCl contents. The transparency and flexibility of the hydrogel increase with Li⁺ content. e) i) Schematic of nerve signal transmission. ii) Diagram of the designed stretchable and self-healing artificial nerve fiber (SSANF). f) $|V_1/V_0|$ varies with frequency and voltage. The value is kept between 0.975 and 1.025, indicating good signal fidelity. Inset: The test equipment. g) SSANF is used to transmit signals in real-time to control the robotic hand in a cryogenic environment. Scale bar: 1 cm. d-g) Reproduced with permission.^[34] Copyright 2022, Wiley-VCH GmbH.

5. Animals-inspired self-healing soft electronics

Nature provides us with a wealth of resources. Various self-healing organisms with unique characteristics, such as mussels, chameleons, jellyfish, and pigeons, have inspired the research on self-healing materials. In this section, animal-inspired self-healing soft electronics will be introduced, along with their unique structures and functions to adapt to their respective living environments.

5.1. Mussel-inspired self-healing soft electronics

Mussels can strongly adhere to almost all surfaces, even to classically adhesion-resistant materials, by forming covalent and noncovalent interactions with substrates via 3,4-dihydroxy-L-phenylalanine (DOPA).^[95-98] Dopamine molecules contain catechol and amine groups, which are considered to be essential for mussels to achieve adhesion.^[18] Inspired by this, Han et al. oxidized dopamine into clay nanosheets to form polydopamine (PDA) and generate sufficient free catechol groups, and then added acrylamide monomer for in situ polymerization. The formed hydrogel possesses good adhesion and superior toughness due to its mussel-like structure (Figure 7a).^[99] The researchers have also used PDA to reduce graphene oxide (GO) following acrylamide monomer polymerization.^[96] The prepared hydrogel is conductive, self-adhesive, and self-healable. After being attached to a human arm, the hydrogel can detect electromyographic (EMG) signals during arm relaxation tension. Compared with the commercial electrode, the hydrogel-electrode maintains stable voltage output during repeated adhesion/peeling processes.

Liao et al. fabricated a self-healing hybrid hydrogel through hydrogen bonds and π - π stacking interactions, and the hydrogel bonds to human skin tightly due to the free catechol moiety of PDA (Figure 7b).^[22] The incorporation of functionalized single-wall carbon nanotube (FSWCNT) makes the hybrid hydrogel conductive, which can light up a LED in series when energized. After cutting the hydrogel, the LED went off, and after reconnecting the two parts to make it self-healing, the LED came on again. It is worth mentioning that the self-healing process can be completed within 2 seconds without external assistance, and the resistance of the hydrogel is restored to 99% of the initial value. Such superior self-healing capability makes the hydrogel a potential material for healable electronics. The conductive hydrogel was then applied to human hand gesture monitoring, as shown in Figure 7c. After adhering to the glove, the hydrogel was stretched and the resistance increased as the bending angle of the finger increased. Similarly, inspired by mussels, Wang et al. synthesized a self-healing conductive elastomer containing AgNPs and applied it to strain sensors.^[42]

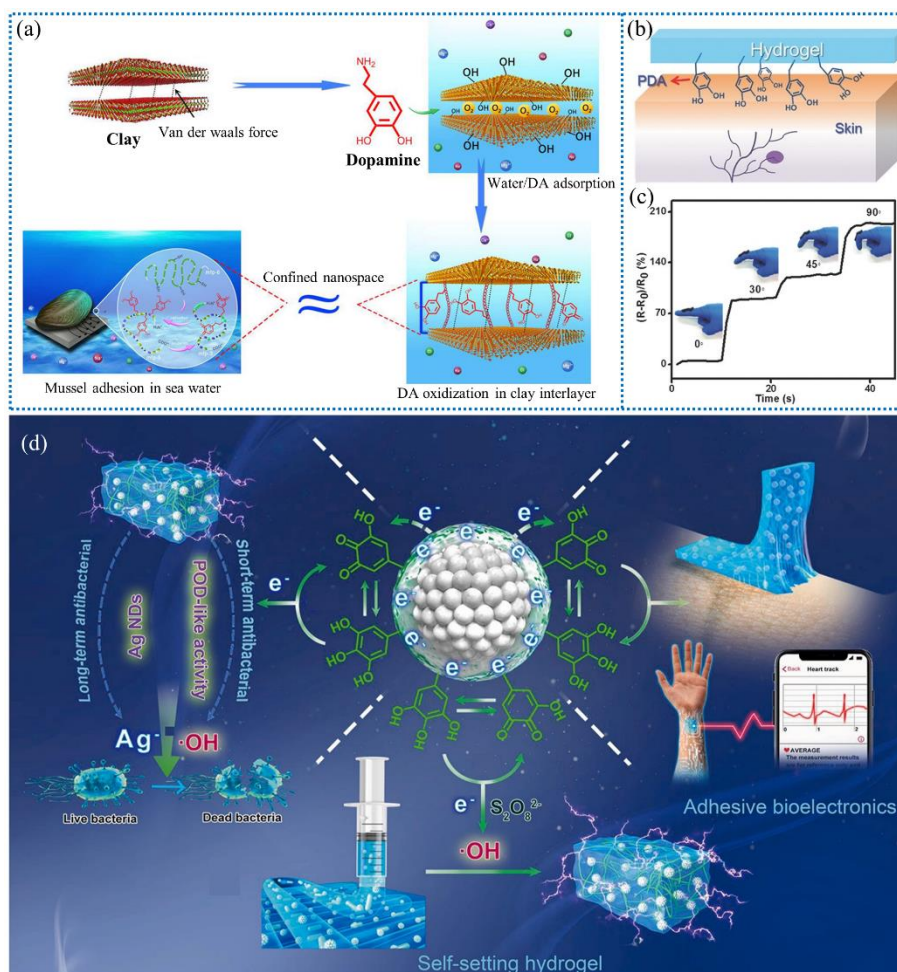


Figure 7. Mussel-inspired self-healing soft electronics. a) A design strategy for the mussel-inspired hydrogel, in which dopamine molecules polymerize in the nanospaces of clay nanosheets. Reproduced with permission.^[99] Copyright 2017, American Chemical Society. b) Schematic illustration of human-hydrogel interaction. c) The resistance of the hydrogel adhered to the glove varies with the bending angle of the finger. b-c) Reproduced with permission.^[22] Copyright 2017, Wiley-VCH GmbH. d) Schematic of mussel-inspired nanozyme catalyzed self-setting hydrogel. The hydrogel can adhere to human skin as bioelectronics for EMG signal detection. Reproduced with permission.^[45] Copyright 2021, Royal Society of Chemistry.

Jia et al. synthesized a mussel-inspired ultra-small tannic acid chelated Ag (TA-Ag) nanozyme catalyzed hydrogel.^[45] Figure 7d shows the structure of the nanozyme, in which the abundant phenolic hydroxyl groups ensure the adhesiveness of the hydrogel. The hydrogel can adhere to various surfaces, such as cherry, glass, mouse liver, and metal. Nanozyme catalysis endows the hydrogel with self-setting properties, which will facilitate their injection at the injury sites. The hydrogel can also be used as adhesive bioelectronics for EMG signal detection. Mussel-inspired self-healing soft electronics with adhesiveness are in a booming stage of development, and their mechanism, structure, and applications are expected to be further investigated.

5.2. Chameleon-inspired self-healing soft electronics

Chameleons can adjust their body color with the changing environments to hide themselves and capture preys because of their unique structural color, which is produced by micro- and nanostructures.^[100] When being stimulated, chameleons stretch their skin, causing the distance between the guanine nanocrystals under the skin to widen, further causing a change in reflectivity that changes the color they display.^[101] As shown in Figure 8a, inspired by the structural color of the chameleon and the adhesive protein of the mussel, Wang et al. fabricated a self-healing film by doping conductive CNTs-PDA fillers into PU inverse opal matrix.^[36] First, silica nanoparticles were self-assembled into ordered hexagonal arrays, then PU solution was introduced into the arrays, and the mixed film solidified after solvent evaporation. Finally, after etching the silica nanoparticles, PU inverse opal matrix was obtained. In this system, PU film provides stretchability and self-healing ability, while CNTs deliver conductivity. When the hybrid film is stretched, the resistance of the film increases, and at the same time, the reflection spectrum is blue-shifted due to the reduction of the distance between the diffraction planes. As a demonstration, the hydrogel was adhered to the human finger (Figure 8b), wrist (Figure 8c) and elbow (Figure 8d), respectively, for human motion detection. Taking the finger as an example, the film's structural color changes from orange-red to green with increasing bending angle of the finger with good repeatability, while the increase in resistance corresponds to the change in wavelength. Compared with traditional soft electronic sensors, this dual-signal sensing extends the expression of the signal. In addition, benefiting from the self-healing ability, films with different structural colors can be stitched together to obtain complex patterned films.

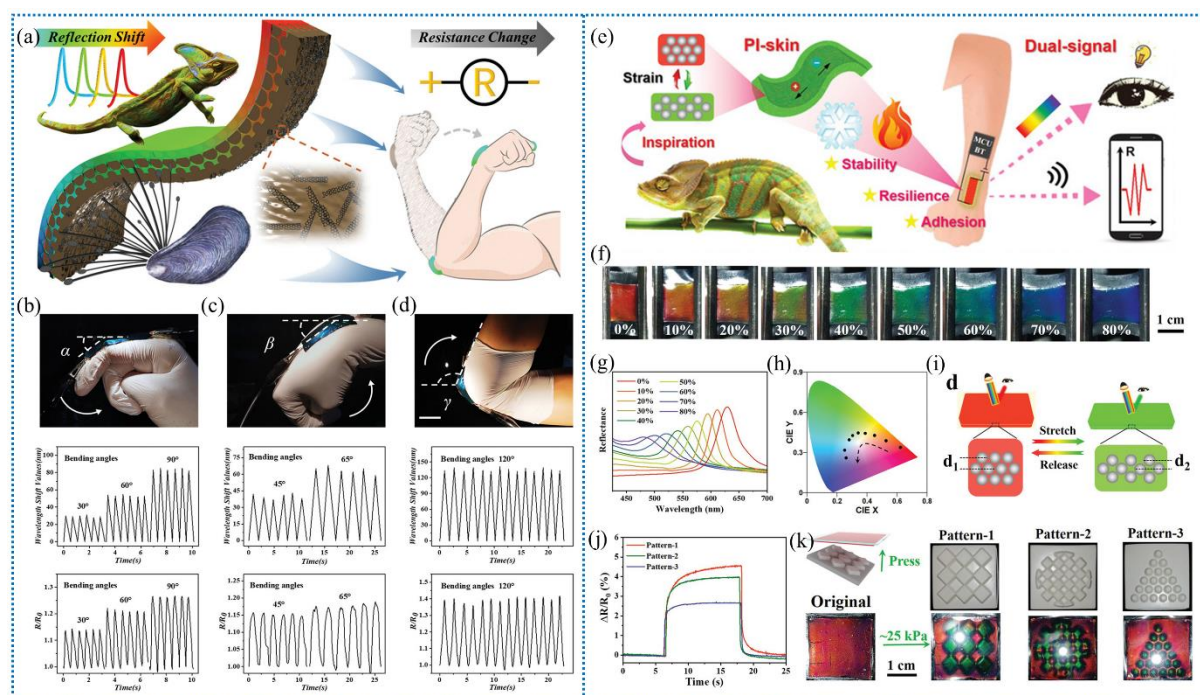


Figure 8. Chameleon-inspired self-healing soft electronics. a) Schematic diagram of the self-healing structural color film inspired by chameleon. b-d) The structural color films adhered to human finger (b), wrist (c), and elbow (d), respectively, are applied to human motion monitoring by recording wavelength and resistance variations in real time. a-d) Reproduced with permission.^[36] Copyright 2020, Wiley-VCH GmbH. e) Schematic diagram of chameleon-inspired photonic-ionic skin (PI-skin). f) PI-skin color changes under different strains. g) Reflectance spectra of PI-skin under different strains. h) The corresponding CIE chromaticity diagram. i) Diagram of the photonic structure change during the stretching process, the lattice spacing in the vertical direction decreases. j) Resistance change when PI-skin is pressed with different patterns. k) Photographs of the PI-skin when pressed with different patterns. The color of the pressed area changes from red to green. e-k) Reproduced with permission.^[20] Copyright 2022, Wiley-VCH GmbH.

Sun et al. fabricated a biomimetic photonic-ionic skin (PI-skin) by sandwiching PS@SiO₂ microspheres in an ionic liquid located polyzwitterionic PSBMA network.^[20] SEM image shows that PS@SiO₂ microspheres possess an ordered array structure with the polymer filled in the gaps of the microspheres, locked by covalent cross-linking and non-covalent bonds to avoid separation of the microspheres from the polymer upon stretching. Similarly, the PI-skin produces dual visual and electrical signals when touched (Figure 8e). Figure 8f displays the change in color of the PI-skin from red to blue when stretched from the original state to the strain of 80%, indicating sufficient sensitivity. The corresponding reflection spectra and CIE chromaticity diagrams during the stretching process are summarized in Figure 8g-h. Figure 8i depicts the mechanism of this process, where the lattice spacing in the vertical direction decreases when stretching, leading to a blue shift of the maximum reflection peak. In addition, the PI-skin can be used as a distributed pressure sensor. Three templates with different patterns are fabricated and pressed on the PI-skin. As shown in Figure 8j, the difference in the pressurized area is indicated by the electrical signal, while the pressure distribution cannot be seen. However, the shape, location, and distribution of pressure can be shown simply and clearly through visual color changes (Figure 8k). This provides a promising solution for the emerging field of human-machine interaction sensing.

5.3. Other animals-inspired self-healing soft electronics

In addition to the aforementioned mussels and chameleons, there are many other bioinspired self-healing soft materials. Figure 9a shows the composition of the jellyfish, combining hydrogen bonding-assembled hierarchical fluorescent proteins and biomembrane structures.^[37] This endows jellyfish with survivabilities such as finding food and mating partners in the dark.

Inspired by this, Liu et al. synthesized a fluorescent and self-healing hydrogel via aqueous self-assembly and polymerization of micelles. The micelles exhibit hydrophobic cores that impart toughness to the hydrogel and enhance fluorescence emission, the polymer incorporated in the micelles can ionize and respond to electric fields. The electric field-induced bending behavior of the hydrogel was investigated, and Figure 9b shows a schematic representation of the experiment, where sulfonate ions are immobilized in the hydrogel and the counter Na^+ ions move freely to the cathode, causing osmotic pressure difference to bend the hydrogel. Figure 9c shows the photographs of the hydrogel during bending, where the bending angle increases with time, reaching almost 90° after 5 min, and changing the electrode direction induces the opposite bending. Due to the bending behavior of hydrogel, it can be used as a "hydrogel cantilever" for lifting cargo (Figure 9d). The authors also demonstrated the self-healing property of the hydrogel through rheological tests.

Moreover, the perovskite-polymer matrix has recently been adopted to mimic the luminescent properties of jellyfish to construct a self-healing elastomer for harsh aquatic environments.^[102] Despite their excellent light emission properties, metal halide perovskites suffer from poor stability. The researchers exploited the positively charged nature of perovskites to form ion-dipole interactions with all-dipole fluorine elastomer, which both protected the perovskites and provided aquatic self-healing capabilities. The luminescent elastomer possesses large-scale manufacturing capability and can be fabricated into different forms, such as film, fiber, and textile.

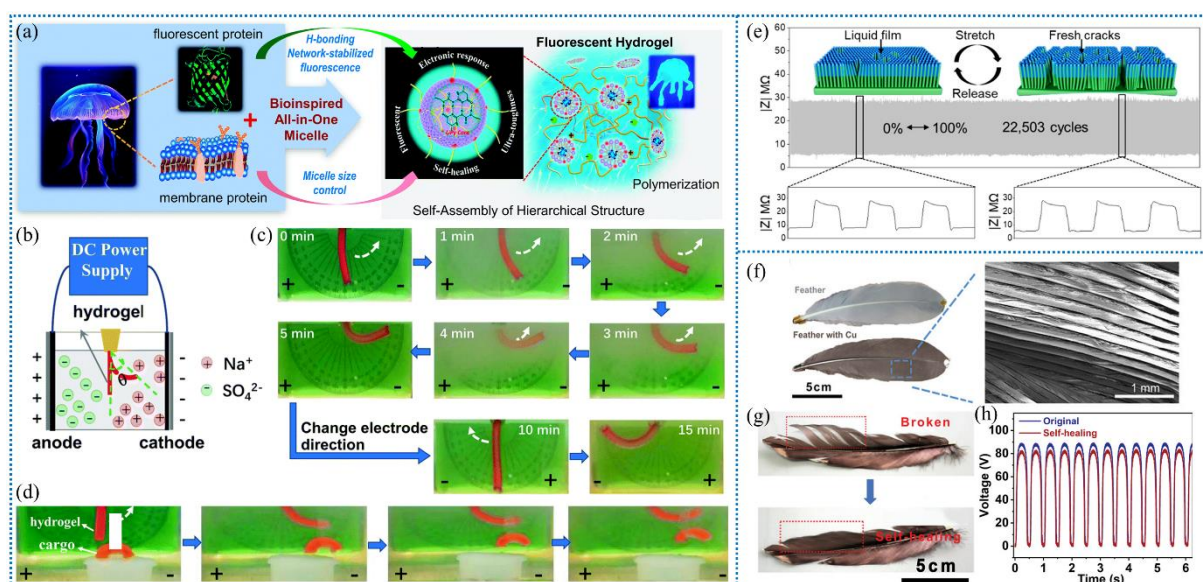


Figure 9. Other animals-inspired self-healing soft electronics. a) Left: Schematic representation of fluorescent and membrane proteins in jellyfish. Right: Fluorescent hydrogel with a hierarchical structure inspired by jellyfish. b) Schematic diagram of the electric field actuation

setup for testing the bending behavior of the fluorescent hydrogel. The SO_4^{2-} ions are immobilized in the hydrogel and the counter Na^+ ions move to the cathode, causing osmotic pressure difference to bend the hydrogel. c) The fluorescent hydrogel bends when an electric field is applied. The bending angle increases with time, and changing the electrode direction causes the hydrogel to bend in the opposite direction. d) The hydrogel can be used as a “hydrogel cantilever” to lift the cargo. a-d) Reproduced with permission.^[37] Copyright 2021, Royal Society of Chemistry. e) Schematic illustration of the resistive strain sensor based on self-healing wear-free liquid films inspired by tear films on animal corneas. The cracks formed between the microvilli during stretching can self-heal after release, and the resistance variation remains stable even after 22500 stretch-release cycles. Reproduced with permission.^[103] Copyright 2019, American Chemical Society. f) Photography and SEM image copper-deposited feather. g) Self-healing behavior of the feather-based single-electrode triboelectric nanogenerator (F-STENG). h) The output voltages of the F-STENG before damage and after self-healing are comparable. f-h) Reproduced with permission.^[27] Copyright 2021, Wiley-VCH GmbH.

Miao et al. fabricated a self-healing liquid film by mimicking the structure of tear films on animal corneas.^[103] As shown in Figure 9e, an ethanol solution of ionic liquid was dropped on the PDMS microvilli, and as the ethanol evaporated, a liquid-gas interface appeared and a meniscus was formed between adjacent microvilli. The conductivity of the liquid film can be adjusted by the ionic liquid concentration. Cracks are formed between the microvilli during stretching, and self-repair occurs after release due to capillary forces. Compared to other self-healing soft electronics, the liquid film exhibits superior durability, maintaining a steady resistance variation even after 22500 stretch-release cycles due to wet friction. The liquid film is then used as a strain sensor for human motion monitoring, where the resistance increases with the bending of the finger or wrist.

In addition to biomimetics, primitive biomaterials themselves have recently been used as self-healing electronics.^[27] As shown in Figure 9f, Zhu et al. deposited copper particles on a feather with a long and narrow shape and used it as a single-electrode mode TENG. When in contact with external objects, triboelectric charges will accumulate on the feather and generate a loop current. Due to the hook structure on the microscopic size, feathers can heal themselves to their original shape when damaged (Figure 9g). Figure 9h compares the voltages generated by TENG based on the original feather and the self-healing feather, indicating the self-healing ability of the electronic device. This method uses feathers that fall off the animals to construct self-

healing TENG without any post-chemical treatment process, providing a new idea for the construction of self-healing electronics.

6. Conclusions and outlook

In this review, bioinspired self-healing soft electronics are discussed in detail from mechanisms and materials to properties. There are two main self-healing mechanisms, namely extrinsic self-healing via encapsulation of healing agents and intrinsic self-healing through supramolecular chemistry or dynamic bonds. Based on the two self-healing mechanisms, polymer materials and their composite systems with conductive nanoparticles or ionic liquids have been designed and implemented for self-healing soft electronics, including insulators, semiconductors, electronic conductors, and ionic conductors. Natural biological systems have other fantastic properties in addition to self-healing, which inspire many unique self-healing soft electronics (Figure 10a). Electronic skins and ionic skins mimic the stretchability of human skins to prevent cracking, and they are commonly used as strain sensors to monitor human motions.^[104] Inspired by the mechanical toughness that human muscles possess to perform movements, muscle-inspired soft electronics are achieved by mimicking the structure of muscle fibers or the D-A interaction of the skeletal muscle protein titin.^[77] Neurons communicate with each other through spatial axons, allowing signals to be transmitted throughout the body, and thus inspired polymers and devices have excellent mechanical properties and information transfer capabilities, respectively. Mussels can attach their soft body to wet rocks to survive in the fierce waves or tides, which inspires self-healing soft electronics with adhesive properties.^[89] The soft electronic device mimicking the structural color of a chameleon has a dual-signal sensing capability when stretched, with simultaneous changes in resistance and color.^[36] Typical works in these areas are comprehensively discussed including material synthesis, device fabrication, and applications.

The combination of self-healing capabilities and other bio-inspired properties in soft electronics is attractive. However, despite the fascinating properties of these soft electronics, their applications were largely focused on strain sensing, wound healing, and TENG, without fully utilizing the properties learned from biological systems. Thus, the development of self-healing electronics for specific application scenarios is promising. For example, muscle-inspired self-healing soft electronics can be applied to soft actuators,^[113,114] mussel-inspired self-healing soft electronics can be applied to underwater communication,^[115] and chameleon-inspired self-healing soft electronics can be applied to optical camouflage.^[32] Moreover, in addition to human motion detection^[109] and electric generator,^[110] self-healing soft electronics are expected to be combined with artificial intelligence (AI) analysis systems for environmental sensing (Figure 10c).^[111] AI is also promising in motivating the evolution of perception to cognition, for example, it can distinguish materials through a single touch.^[112] Moreover, most of the reported research focuses on the self-healing of materials rather than fabricated devices, which is obviously insufficient for practical applications. Self-healing of soft electronic devices requires self-healing of all components and good binding ability and compatibility between different parts. Finally, the currently reported self-healing materials are mainly films and fibers fabricated by spin coating or molding, restricting their large-scale production. Thus, it is essential to expand the fabrication approaches for self-healing materials, such as extrusion molding for films,^[105] thermal-drawing technique for fibers,^[106] and even weaving technique for textiles (Figure 10b).^[116] This will facilitate the introduction of self-healing soft electronics into flexible wearable electronics, which is considered to be the next generation of information technology.^[117] Moreover, the 3D printing technique has been adopted for fabricating self-healing soft electronics with precise and designable device structures.^[32, 108, 118, 119] It is worth mentioning that both the above-mentioned film extrusion process and thermal fiber drawing technique require materials to be thermoplastic, and 3D printing also has material selectivity depending on the printing technology. Therefore, satisfying the mechanical properties of materials without sacrificing self-healing ability through rational molecular design is the key to the application of self-healing materials in various manufacturing methods. Dedicated and systematic research on bioinspired self-healing soft electronics is expected to motivate work in life sciences, organic synthesis, materials processing, device manufacturing, environmental monitoring and AI to improve the service life of electrical devices and even provide a new generation of electronic devices.

Acknowledgements

M. Q. and R. Y. contributed equally to this work. This work was supported by the Research Initiation Project of Zhejiang Lab funding (No. 2022MG0PI01), National Natural Science Foundation of China (52202191). The authors would like to acknowledge Dr. Xuyang Wang for editing the pictures.

Received: ((will be filled in by the editorial staff))

Revised: ((will be filled in by the editorial staff))

Published online: ((will be filled in by the editorial staff))

References

- [1] C. Wang, C. Wang, Z. Huang, S. Xu, *Adv. Mater.* **2020**, *30*, 1801368.
- [2] N. Matsuhisa, S. Niu, S. J. K. O' Neill, J. Kang, Y. Ochiai, T. Katsumata, H. C. Wu, M. Ashizawa, G. J. N. Wang, D. Zhong, X. Wang, X. Gong, R. Ning, H. Gong, I. You, Y. Zheng, Z. Zhang, J. B. H. Tok, X. Chen, Z. Bao, *Nature* **2021**, *600*, 246.
- [3] C. Lu, S. Park, T. J. Richner, A. Derry, I. Brown, C. Hou, S. Rao, J. Kang, C. T. Moritz, Y. Fink, P. Anikeeva, *Sci. Adv.* **2017**, *3*, e1600955.
- [4] T. Liu, M. Liu, S. Dou, J. Sun, Z. Cong, C. Jiang, C. Du, X. Pu, W. Hu, Z. Wang, *ACS Nano* **2018**, *12*, 2818.
- [5] R. C. Webb, A. P. Bonifas, A. Behnaz, Y. Zhang, K. J. Yu, H. Cheng, M. Shi, Z. Bian, Z. Liu, Y. S. Kim, W. H. Yeo, J. S. Park, J. Song, Y. Li, Y. Huang, A. M. Gorbach, J. A. Rogers, *Nat. Mater.* **2013**, *12*, 938.
- [6] B. C. K. Tee, A. Chortos, A. Berndt, A. K. Nguyen, A. Tom, A. McGuire, Z. C. Lin, K. Tien, W. G. Bae, H. Wang, P. Mei, H. H. Chou, B. Cui, K. Deisseroth, T. N. Ng, Z. Bao, *Science* **2015**, *350*, 313.
- [7] S. Lim, D. Son, J. Kim, Y. B. Lee, J. K. Song, S. Choi, D. J. Lee, J. H. Kim, M. Lee, T. Hyeon, D. H. Kim, *Adv. Funct. Mater.* **2014**, *25*, 375.
- [8] S. I. Rich, R. J. Wood, C. Majidi, *Nat. Electron.* **2018**, *1*, 102.
- [9] M. Zarek, M. Layani, I. Cooperstein, E. Sachyani, D. Cohn, S. Magdassi, *Adv. Mater.* **2016**, *28*, 4449.
- [10] M. D. Hager, P. Greil, C. Leyens, S. Zwaag, U. S. Schubert, *Adv. Mater.* **2010**, *22*, 5424.
- [11] Y. J. Tan, G. J. Susanto, H. P. A. Ali, B. C. K. Tee, *Adv. Mater.* **2021**, *33*, 2002800.
- [12] K. Jud, H. H. Kausch, J. G. Williams, *J. Mater. Sci.* **1981**, *16*, 204.

- [13] R. P. Wool, *Soft Matter* **2008**, *4*, 400.
- [14] J. Kang, J. B. H. Tok, Z. Bao, *Nat. Electron.* **2019**, *2*, 144.
- [15] K. Parida, J. Xiong, X. Zhou, P. S. Lee, *Nano Energy* **2019**, *59*, 237.
- [16] M. Khatib, O. Zohar, H. Haick, *Adv. Mater.* **2021**, *33*, 2004190.
- [17] Y. Gai, H. Li, Z. Li, *Small* **2021**, *17*, 2101383.
- [18] C. Shao, M. Wang, L. Meng, H. Chang, B. Wang, F. Xu, J. Yang, P. Wan, *Chem. Mater.* **2018**, *30*, 3110.
- [19] G. Ge, Y. Lu, X. Qu, W. Zhao, Y. Ren, W. Wang, Q. Wang, W. Huang, X. Dong, *ACS Nano* **2020**, *14*, 218.
- [20] Y. Sun, Y. Wang, Y. Liu, S. Wu, S. Zhang, W. Niu, *Adv. Funct. Mater.* **2022**, *32*, 2204467.
- [21] Z. Lei, Q. Wang, S. Sun, W. Zhu, P. Wu, *Adv. Mater.* **2017**, *29*, 1700321.
- [22] M. Liao, P. Wan, J. Wen, M. Gong, X. Wu, Y. Wang, R. Shi, L. Zhang, *Adv. Funct. Mater.* **2017**, *27*, 1703852.
- [23] L. Han, L. Yan, M. Wang, K. Wang, L. Fang, J. Zhou, J. Fang, F. Ren, X. Lu, *Chem. Mater.* **2018**, *30*, 5561.
- [24] J. Xue, Y. Zou, Y. Deng, Z. Li, *EcoMat* **2022**, *4*, e12209.
- [25] L. Shuai, Z. Guo, P. Zhang, J. Wan, X. Pu, Z. Wang, *Nano Energy* **2020**, *78*, 105389.
- [26] D. Yang, Y. Ni, H. Su, Y. Shi, Q. Liu, X. Chen, D. He, *Nano Energy* **2021**, *79*, 105394.
- [27] J. Zhu, Y. Cheng, S. Hao, Z. Wang, N. Wang, X. Cao, *Adv. Funct. Mater.* **2021**, *31*, 2100039.
- [28] R. Liu, Y. Lai, S. Li, F. Wu, J. Shao, D. Liu, X. Dong, J. Wang, Z. Wang, *Nano Energy* **2022**, *95*, 107056.
- [29] Y. Chen, X. Pu, M. Liu, S. Kuang, P. Zhang, Q. Hua, Z. Cong, W. Guo, W. Hu, Z. Wang, *ACS Nano* **2019**, *13*, 8936.
- [30] D. Yang, Y. Ni, X. Kong, S. Li, X. Chen, L. Zhang, Z. Wang, *ACS Nano* **2021**, *15*, 14653.
- [31] M. Zheng, X. Wang, O. Yue, M. Hou, H. Zhang, S. Beyer, A. M. Blocki, Q. Wang, G. Gong, X. Liu, J. Guo, *Biomaterials* **2021**, *276*, 121026.
- [32] Z. Yu, P. Wu, *Adv. Mater.* **2021**, *33*, 2008479.
- [33] K. Park, H. Yuk, M. Yang, J. Cho, H. Lee, J. Kim, *Sci. Robot.* **2022**, *7*, 7187.
- [34] C. Wang, Y. Liu, X. Qu, B. Shi, Q. Zheng, X. Lin, S. Chao, C. Wang, J. Zhou, Y. Sun, G. Mao, Z. Li, *Adv. Mater.* **2022**, *34*, 2105416.

- [35] J. H. Waite, *J. Exp. Biol.* **2017**, *220*, 517.
- [36] Y. Wang, Y. Yu, J. Guo, Z. Zhang, X. Zhang, Y. Zhao, *Adv. Funct. Mater.* **2020**, *30*, 2000151.
- [37] T. Liu, F. Wang, Q. Wu, T. Chen, P. Sun, *Mater. Horizons* **2021**, *8*, 3096.
- [38] J. Y. Sun, C. Keplinger, G. M. Whitesides, Z. Suo, *Adv. Mater.* **2014**, *26*, 7608.
- [39] X. Xun, Z. Zhang, X. Zhao, B. Zhao, F. Gao, Z. Kang, Q. Liao, Y. Zhang, *ACS Nano* **2020**, *14*, 9066.
- [40] W. Kong, C. Wang, C. Jia, Y. Kuang, G. Pastel, C. Chen, G. Chen, S. He, H. Huang, J. Zhang, S. Wang, L. Hu, *Adv. Mater.* **2018**, *30*, 1801934.
- [41] H. Chen, Z. Sun, H. Lin, C. He, D. Mao, *Adv. Funct. Mater.* **2022**, *32*, 2204263.
- [42] Z. Wang, Y. Liu, D. Zhang, C. Gao, Y. Wu, *Compos. B. Eng.* **2021**, *224*, 109213.
- [43] A. H. Hofman, I. A. van Hees, J. Yang, M. Kamperman, *Adv. Mater.* **2018**, *30*, 1704640.
- [44] Q. Yu, Z. Zheng, X. Dong, R. Cao, S. Zhang, X. Wu, X. Zhang, *Soft Matter* **2021**, *17*, 8786.
- [45] Z. Jia, X. Lv, Y. Hou, K. Wang, F. Ren, D. Xu, Q. Wang, K. Fan, C. Xie, X. Lu, *Bioact. Mater.* **2021**, *6*, 2676.
- [46] N. R. Barros, Y. Chen, V. Hosseini, W. Wang, R. Nasiri, M. Mahmoodi, E. P. Yalcintas, R. Haghniaz, M. M. Mecwan, S. Karamikamkar, W. Dai, S. A. Sarabi, N. Falcone, P. Young, Y. Zhu, W. Sun, S. Zhang, J. Lee, K. Lee, S. Ahadian, M. R. Dokmeci, A. Khademhosseini, H. J. Kim, *Biomater. Sci.* **2021**, *9*, 6653.
- [47] M. Zhu, F. Zhang, X. Chen, *Small Struct.* **2020**, *1*, 2000045.
- [48] D. C. Baulcombe, S. Chapman, S. S. Cruz, *Plant J.* **1995**, *7*, 1045.
- [49] Y. Yang, M. W. Urban, *Chem. Soc. Rev.* **2013**, *42*, 7446.
- [50] F. Zhang, P. Ju, M. Pan, D. Zhang, Y. Huang, G. Li, X. Li, *Corros. Sci.* **2018**, *144*, 74.
- [51] X. Zhang, N. Sheng, L. Wang, Y. Tan, C. Liu, Y. Xia, Z. Nie, K. Sui, *Mater. Horizons* **2019**, *6*, 326.
- [52] L. Saunders, P. X. Ma, *Macromol. Biosci.* **2018**, *19*, 1800313.
- [53] Y. Cheng, X. Xiao, K. Pan, H. Pang, *Chem. Eng. J.* **2020**, *380*, 122565.
- [54] J. M. Lehn, *Science* **1993**, *260*, 1762.
- [55] L. B. Huang, X. Dai, Z. Sun, M. C. Wong, S. Y. Pang, J. Han, Q. Zheng, C. H. Zhao, J. Kong, J. Hao, *Nano Energy* **2021**, *82*, 105724.
- [56] B. C. K. Tee, C. Wang, R. Allen, Z. Bao, *Nat. Nanotechnol.* **2012**, *7*, 825.

- [57] Z. Wu, J. Chen, D. W. Boukhvalov, Z. Luo, L. Zhu, Y. Shi, *Nano Energy* **2021**, *85*, 105990.
- [58] Y. Cao, Y. J. Tan, S. Li, W. W. Lee, H. Guo, Y. Cai, C. Wang, B. C. K. Tee, *Nat. Electron.* **2019**, *2*, 75.
- [59] C. L. Perrin, J. B. Nielson, *Annu. Rev. Phys. Chem.* **1997**, *48*, 511.
- [60] P Heidarian, AZ Kouzani, A Kaynak, Bahador Bahrami, Mariana Paulino, Bijan Nasri-Nasrabadi, Russell J. Varley, *Macromol Rapid Commun.* 2020, *41*, 2000439.
- [61] Y. Lai, X. Kuang, P. Zhu, M. Huang, X. Dong, D. Wang, *Adv. Mater.* **2018**, *30*, 1802556.
- [62] Y. Cao, H. Wu, S. I. Allec, B. M. Wong, D. S. Nguyen, C. Wang, *Adv. Mater.* **2018**, *30*, 1804602.
- [63] Y. L. Rao, A. Chortos, R. Pfattner, F. Lissel, Y. C. Chiu, V. Feig, J. Xu, T. Kurosawa, X. Gu, C. Wang, M. He, J. W. Chung, Z. Bao, *J. Am. Chem. Soc.* **2016**, *138*, 6020.
- [64] J. Y. Oh, S. Rondeau-Gagné, Y. C. Chiu, A. Chortos, F. Lissel, G. N. Wang, B. C. Schroeder, T. Kurosawa, J. Lopez, T. Katsumata, J. Xu, C. Zhu, X. Gu, W. G. Bae, Y. Kim, L. Jin, J. W. Chung, J. B. H. Tok, Z. Bao, *Nature* **2016**, *539*, 411.
- [65] J.Y. Oh, D. Son, T. Katsumata, Y. Lee, Y. Kim, J. Lopez, H.-C. Wu, J. Kang, J. Park, X. Gu, J. Mun, N. G. J. Wang, Y. Yin, W. Cai, Y. Yun, J. Tok, Z. Bao, *Sci. Adv.* **2019**, *5*, 3097.
- [66] C. Gong, J. Liang, W. Hu, X. Niu, S. Ma, H.T. Hahn, Q. Pei, *Adv. Mater.* **2013**, *25*, 4186.
- [67] D. Son, J. Kang, O. Vardoulis, Y. Kim, N. Matsuhisa, J.Y. Oh, J.W. To, J. Mun, T. Katsumata, Y. Liu, A. F. McGuire, M. Krason, F. M. Lopez, J. Ham, U. Kraft, Y. Lee, Y. Yun, J. B. H. Tok, Z. Bao, *Nat. Nanotechnol.* **2018**, *13*, 1057.
- [68] D. Son, Z. Bao, *ACS Nano* **2018**, *12*, 11731.
- [69] K. Parida, V. Kumar, W. Jiangxin, V. Bhavanasi, R. Bendi, P.S. Lee, *Adv. Mater.* **2017**, *29*, 1702181.
- [70] H. Niu, X. Du, S. Zhao, Z. Yuan, X. Zhang, R. Cao, Y. Yin, C. Zhang, T. Zhou, C. Li, *RSC Adv.* **2018**, *8*, 30661.
- [71] X. Dai, L.B. Huang, Y. Du, J. Han, Q. Zheng, J. Kong, J. Hao, *Adv. Funct. Mater.* **2020**, *30*, 1910723.
- [72] Y. Cao, T.G. Morrissey, E. Acome, S.I. Allec, B.M. Wong, C. Keplinger, C. Wang, *Adv. Mater.* **2017**, *29*, 1605099.

- [73] Y. Dobashi, D. Yao, Y. Petel, T.N. Nguyen, M.S. Sarwar, Y. Thabet, C.L. Ng, E. Scabeni Glitz, G.T.M. Nguyen, C. Plesse, *Science* **2022**, 376, 502.
- [74] C. C. Kim, H. H. Lee, K.H. Oh, J. Y. Sun, *Science* **2016**, 353, 682.
- [75] W. Huang, K. Besar, Y. Zhang, S. Yang, G. Wiedman, Y. Liu, W. Guo, J. Song, K. Hemker, K. Hristova, *Adv. Funct. Mater.* **2015**, 25, 3745.
- [76] M. Khatib, T.P. Huynh, Y. Deng, Y.D. Horev, W. Saliba, W. Wu, H. Haick, *Small* **2019**, 15, 1803939.
- [77] W.B. Ying, G. Wang, Z. Kong, C.K. Yao, Y. Wang, H. Hu, F. Li, C. Chen, Y. Tian, J. Zhang, *Adv. Funct. Mater.* **2021**, 31, 2009869.
- [78] M.Y. Lee, S. Dharmapurikar, S.J. Lee, Y. Cho, C. Yang, J.H. Oh, *Chem. Mater.* **2020**, 32, 1914.
- [79] M. Khatib, O. Zohar, W. Saliba, H. Haick, *Adv. Mater.* **2020**, 32, 2000246.
- [80] T. Li, Y. Wang, S. Li, X. Liu, J. Sun, *Adv. Mater.* **2020**, 32, 20002706.
- [81] J. Wang, B. Wu, P. Wei, S. Sun, P. Wu, *Nat. Commun.* **2022**, 13, 1.
- [82] P. Fratzl, *J. R. Soc. Interface* **2007**, 4, 637.
- [83] N.K. Katiyar, G. Goel, S. Hawi, S. Goel, *NPG Asia Mater.* **2021**, 13, 1.
- [84] K. H. Chu, H. H. Tung, D. L. Clinciu, H. I. Hsu, Y. C. Wu, C. I. Hsu, S. W. Lin, S. J. Pan, *Gerontol. Geriatr. Med.* **2022**, 8, 23337214221077788.
- [85] S. Wang, J.Y. Oh, J. Xu, H. Tran, Z. Bao, *Acc. Chem. Res.* **2018**, 51, 1033.
- [86] A. Chortos, Z. Bao, *Mater. Today* **2014**, 17, 321.
- [87] S. Li, Y. Zhang, Y. Wang, K. Xia, Z. Yin, H. Wang, M. Zhang, X. Liang, H. Lu, M. Zhu, *InfoMat* **2020**, 2, 184.
- [88] Z. Ma, D. Kong, L. Pan, Z. Bao, *J. Semicond.* **2020**, 41, 041601.
- [89] J. Yin, S. Pan, L. Wu, L. Tan, D. Chen, S. Huang, Y. Zhang, P. He, *J. Mater. Chem. C* **2020**, 8, 17349.
- [90] Z. Zhang, L. Wang, H. Yu, F. Zhang, L. Tang, Y. Feng, W. Feng, *ACS Appl. Mater. Interfaces* **2020**, 12, 15657.
- [91] H. Chen, X. Ren, G. Gao, *ACS Appl. Mater. Interfaces* **2019**, 11, 28336.
- [92] J. Sun, X. Pu, M. Liu, A. Yu, C. Du, *ACS Nano* **2018**, 12, 6147.
- [93] Y. Qian, Y. Zheng, J. Jin, X. Wu, K. Xu, M. Dai, Q. Niu, H. Zheng, X. He, J. Shen, *Adv. Mater.* **2022**, 34, 2200521.
- [94] X. Liu, S. Tian, S. Xu, W. Lu, C. Zhong, Y. Long, Y. Ma, K. Yang, L. Zhang, J. Yang, *Biosens. Bioelectron.* **2022**, 214, 114528.
- [95] H. Lee, S.M. Dellatore, W.M. Miller, P.B. Messersmith, *Science* **2007**, 318, 426.

- [96] L. Han, X. Lu, M. Wang, D. Gan, W. Deng, K. Wang, L. Fang, K. Liu, C.W. Chan, Y. Tang, *Small* **2017**, *13*, 1601916.
- [97] X. Jing, H. Y. Mi, X. F. Peng, L. S. Turng, *Carbon* **2018**, *136*, 63.
- [98] X. Jing, H. Y. Mi, Y. J. Lin, E. Enriquez, X. F. Peng, L. S. Turng, *ACS Appl. Mater. Interfaces* **2018**, *10*, 20897.
- [99] L. Han, X. Lu, K. Liu, K. Wang, L. Fang, L.-T. Weng, H. Zhang, Y. Tang, F. Ren, C. Zhao, *ACS Nano* **2017**, *11*, 2561.
- [100] Y. Wang, H. Cui, Q. Zhao, X. Du, *Matter* **2019**, *1*, 626.
- [101] J. Teyssier, S.V. Saenko, D. Van Der Marel, M.C. Milinkovitch, *Nat. Commun.* **2015**, *6*, 1.
- [102] Y. Liu, T. Chen, Z. Jin, M. Li, D. Zhang, L. Duan, Z. Zhao, C. Wang, *Nat. Commun.* **2022**, *13*, 1.
- [103] W. Miao, D. Wang, Z. Liu, J. Tang, Z. Zhu, C. Wang, H. Liu, L. Wen, S. Zheng, Y. Tian, *ACS Nano* **2019**, *13*, 3225.
- [104] Q. Wang, S. Ling, X. Liang, H. Wang, H. Lu, Y. Zhang, *Adv. Funct. Mater.* **2019**, *29*, 1808695.
- [105] Q. Wang, W. Chen, W. Zhu, D.J. McClements, X. Liu, F. Liu, *NPJ Sci. Food* **2022**, *6*, 1.
- [106] M. Chen, Z. Wang, Q. Zhang, Z. Wang, W. Liu, M. Chen, L. Wei, *Nat. Commun.* **2021**, *12*, 1.
- [107] Y. Xiao, L. Shi, W. Zhou, F. Wan, W. Liu, *PloS one* **2021**, *16*, e0257629.
- [108] C. Antich, J. de Vicente, G. Jiménez, C. Chocarro, E. Carrillo, E. Montañez, P. Gálvez-Martín, J.A. Marchal, *Acta Biomater.* **2020**, *106*, 114.
- [109] J. Luo, W. Gao, Z.L. Wang, *Adv. Mater.* **2021**, *33*, 2004178.
- [110] Z. Wen, M.-H. Yeh, H. Guo, J. Wang, Y. Zi, W. Xu, J. Deng, L. Zhu, X. Wang, C. Hu, *Sci. Adv.* **2016**, *2*, e1600097.
- [111] Q. Zheng, Y. Hou, H. Yang, P. Tan, H. Shi, Z. Xu, Z. Ye, N. Chen, X. Qu, X. Han, *Nano Energy* **2022**, *98*, 107245.
- [112] H. Niu, H. Li, S. Gao, Y. Li, X. Wei, Y. Chen, W. Yue, W. Zhou, G. Shen, *Adv. Mater.* **2022**, *34*, 2202622.
- [113] S. Terryn, J. Brancart, D. Lefeber, G. Van Assche, B. Vanderborght, *Sci. Robot.* **2017**, *2*, eaa4268.
- [114] C. Lang, E.C. Lloyd, K.E. Matuszewski, Y. Xu, V. Ganesan, R. Huang, M. Kumar, R.J. Hickey, *Nat. Nanotechnol.* **2022**, *17*, 752.

- [115] C. Zhang, M. Wang, C. Jiang, P. Zhu, B. Sun, Q. Gao, C. Gao, R. Liu, *Nano Energy* **2022**, *95*, 106991.
- [116] X. Shi, Y. Zuo, P. Zhai, J. Shen, Y. Yang, Z. Gao, M. Liao, J. Wu, J. Wang, X. Xu, *Nature* **2021**, *591*, 240.
- [117] Y. Shen, Z. Wang, Z. Wang, J. Wang, X. Yang, X. Zheng, H. Chen, K. Li, L. Wei, T. Zhang, *InfoMat* **2022**, *4*, e12318.
- [118] M.A. Darabi, A. Khosrozadeh, R. Mbeleck, Y. Liu, Q. Chang, J. Jiang, J. Cai, Q. Wang, G. Luo, M. Xing, *Adv. Mater.* **2017**, *29*, 1700533.
- [119] A. Ph, A. Sg, B. Hy, A. Mp, A. Ak, C. Rv, A. Azk, *Carbohydr. Polym.* **2022**, *291*, 119545.

Self-healing soft electronic devices possess superior durability and can recover their physical properties after external damage. Biological systems provide unique properties such as stretchability, toughness, and adhesiveness in addition to self-healing capabilities. Self-healing soft electronics inspired by human skin, muscles, neurons, and animals such as mussels, chameleons, and jellyfish are summarized here in detail, including materials, properties, and applications.

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Bioinspired self-healing soft electronics

ToC figure

