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# Bioinspired Self-healing Soft Electronics

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Inspired by nature, various self-healing materials that 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 can 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. This review presents a detailed discussion on bioinspired self-healing soft electronics. 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. This cutting-edge and promising field is believed to stimulate more excellent cross-discipline works in material science, flexible electronics, and novel sensors, accelerating the development of applications in human motion monitoring, environmental sensing, information transmission, etc.

of electronic devices such as computers and mobile phones is moving toward 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 Internet of Things technologies.<sup>[1]</sup> For instance, Matsuhisa et al. reported a stretchable polymer diode and implemented an on-skin strain sensor and wireless display system,<sup>[2]</sup> Lu et al. fabricated a flexible neural probe using AgNW-coated fiber,<sup>[3]</sup> and Liu et al. constructed a soft TENG for energy harvesting based on a hybrid hydrogel-elastomer.<sup>[4]</sup> Compared with traditional electronic devices, soft electronics can be seamlessly integrated with the human body or shape, which significantly expands their applications in healthcare monitoring,<sup>[5]</sup> electronic skins,<sup>[6]</sup> human-machine interface,<sup>[7]</sup> and other fields.<sup>[8,9]</sup> 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

## 1. Introduction


Electronic devices have dramatically revolutionized way people's daily life since their inception. Recently, the development

can be damaged and fractured when cut by sharp objects or subjected to 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

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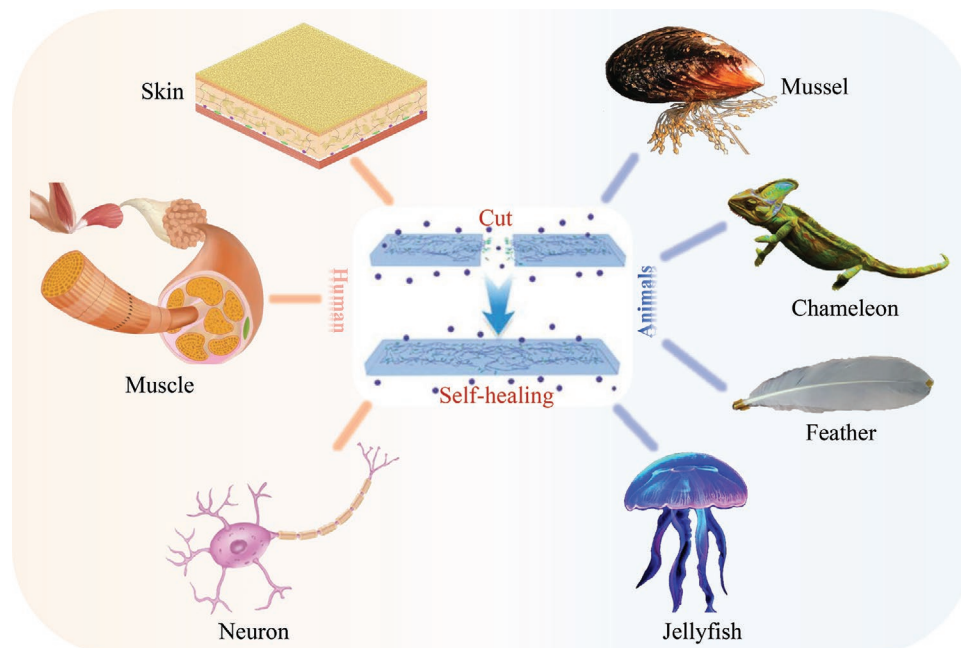
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this thorny problem. During the long evolutionary process in nature, animals and plants have acquired self-healing abilities. For example, the leaves of plants and the skin of animals can repair themselves after injury, which allows them to survive in harsh environments. Inspired by nature, self-healing materials capable of recovering their physical properties after damage can be adopted to improve the service life of soft electronics.<sup>[10,11]</sup> This unique functional material was proposed in the 1980s and drew tremendous attention.<sup>[12]</sup> The self-healing process is achieved through dynamic bond exchanges or supramolecular interactions in the polymer chain segments or by releasing encapsulated healing reagents. Studies on self-healing materials have long focused on mechanisms, fabrications and characterizations.<sup>[13]</sup> Recently, the self-healing property has been integrated into soft electronics, blazing a promising trail in the field of self-healing soft electronics.<sup>[14–17]</sup> These self-healing soft electronics, including OFETs, TENGs, capacitors, and electronic skins, have been developed for various scenarios such as strain sensing,<sup>[18–20]</sup> human motion monitoring,<sup>[21–24]</sup> energy harvesting,<sup>[25–29]</sup> and wound healing.<sup>[30,31]</sup>

Besides the self-healing properties, various biological self-healing materials in nature can also intrinsically integrate other properties depending on their living environments and functions (**Figure 1**). Taking the human body as an example, the human skin, as the largest sensory organ, is stretchable, resilient, and can perceive external stimuli.<sup>[33,38,39]</sup> Human muscle comprises arranged collagen fibers and a hierarchy of bundles, which has high mechanical strength to perform various movements.<sup>[19,40]</sup> 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.<sup>[34,41]</sup> 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,<sup>[35,42–46]</sup> chameleons can change color depending on the environment to protect themselves,<sup>[36]</sup> feathers are lightweight and can repair themselves due to their cascade slide-lock system,<sup>[47]</sup> while the beautiful jellyfish has green fluorescent protein, and the research on its light-emitting principle has won the Nobel Prize in Chemistry.<sup>[48]</sup> All these fascinating properties have inspired researchers to develop unique self-healing materials. This review summarizes fabrication strategies and mechanisms, such as encapsulating healing agents and introducing supramolecular interactions for functional self-healing soft materials. The working principles of self-healing soft electronic devices are presented, 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 from films to 3D structures, and potential applications of self-healing soft electronics are summarized and prospected. This review aims to provide a powerful reference for developing bioinspired self-healing soft electronics that can adapt to different application scenarios. As a comprehensive research field, self-healing soft electronics will



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



inspire more interdisciplinary research on material synthesis, processing and shaping, device fabrication, and novel sensors.

## 2. Self-Healing Mechanisms

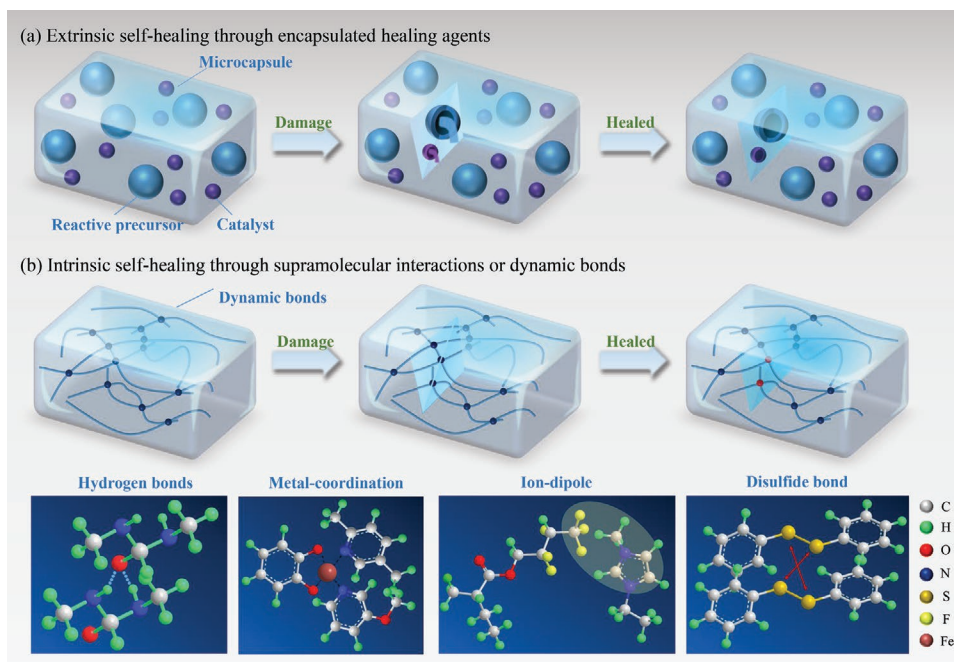
Over the past 20 years, flourishing research works have been focused on self-healing materials that can recover their physical properties upon being subjected to external damage.<sup>[10,49]</sup> Numerous synthetic polymeric materials have been developed and applied in various fields, such as protective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

Studies on self-healing mechanisms are the basis for the design of numerous materials. **Figure 2** depicts two main self-healing mechanisms: extrinsic and intrinsic. In extrinsic approaches (Figure 2a), microcapsules containing healing agents such as reactive precursors and catalysts are encapsulated into the materials.<sup>[53]</sup> 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 incorporating 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.<sup>[54]</sup> 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,<sup>[25,55,56]</sup> metal-coordination bonds,<sup>[57]</sup> ion-dipole interactions,<sup>[58]</sup> etc. 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.<sup>[59]</sup> Metal-coordination bond is a strong noncovalent interaction in which a central metallic atom is surrounded by bound molecules or ions.<sup>[60]</sup> 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.<sup>[32]</sup> 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.<sup>[61]</sup> Notably, the aforementioned chemical approaches can be arbitrarily combined to construct self-healing materials with fascinating characteristics.

## 3. Self-Healing Materials in Soft Electronics

Numerous self-healing materials have been prepared and studied based on the above mechanisms. 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 that 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. This section discusses the materials, representative working mechanisms, and applications of each component.



**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.

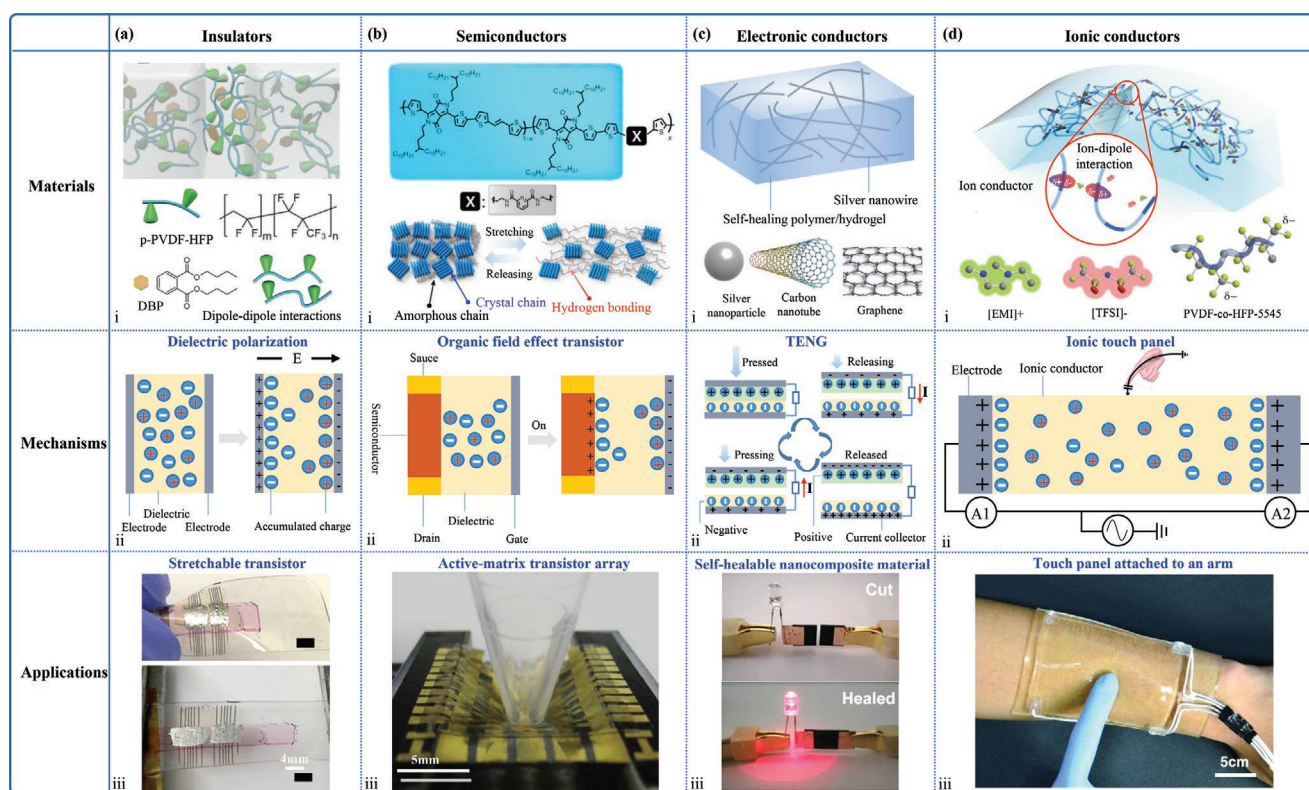
### 3.1. Insulators

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.<sup>[62]</sup> 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.<sup>[49]</sup> 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 the dielectric being 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.<sup>[63]</sup> 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 rather than the integrated OFET device.

### 3.2. Semiconductors

Semiconductors play an important role in the fabrication of OFETs. Nevertheless, fabricating fully self-healing semiconductors remains challenging due to the thickness-limited replicating 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.<sup>[64]</sup> Heating and solvent annealing can promote the movement of polymer chains, thereby repairing the nanocracks formed by fatigue. The research group further fabricated a semiconducting polymer that can self-repair without external forces through



**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.<sup>[62]</sup> Copyright 2018, Wiley-VCH GmbH. Reproduced with permission.<sup>[63]</sup> 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.<sup>[64]</sup> Copyright 2016, Springer Nature. Reproduced with permission.<sup>[65]</sup> 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.<sup>[73]</sup> Copyright 2020, Wiley-VCH GmbH. d) Ionic conductors composed of ionic hydrogel are used as a touch panel, and the working mechanism of the device is presented. Reproduced with permission.<sup>[58]</sup> Copyright 2019, Springer Nature. Reproduced with permission.<sup>[76]</sup> Copyright 2016, American Association for the Advancement of Science.

metal-coordination interactions.<sup>[65]</sup> The cut and recovery test of the self-healing semiconductor was 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, 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).<sup>[66–70]</sup> 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 the potential difference till the 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.<sup>[71,72]</sup> Figure 3c iii shows a photograph of a nanocomposite conductor with its conductivity after self-healing confirmed by lighting an LED.<sup>[73]</sup> A noteworthy issue is that incorporating nanofillers may sacrifice the mechanical strength and self-healing ability of polymer or hydrogel. Still, an insufficient filling may result in poor conductivity, so it is essential 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.<sup>[74,75]</sup> Although ionic conductors typically have higher resistance than electronic conductors, they are still widely studied because 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.<sup>[58]</sup> Kim et al. reported an ionic touch panel constructed by an ionic conductor (Figure 3d iii), which is soft, transparent, and stretchable.<sup>[76]</sup> 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. 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 applied 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.

## 4. Human-Inspired Self-Healing Soft Electronics

In addition to self-healing properties, various biological self-healing materials in nature 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.<sup>[84,85]</sup> Humans are one of the nature's most wondrous creations, with skin that protects the entire body and senses the external environment, bones that support the body, muscles that perform the movement, and nerves that connect the whole body to transmit signals. In addition, the human body has a strong ability to heal itself and can regenerate after injuries.<sup>[86]</sup> 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 merits, such as stretchability, elasticity, self-healing, and superior sensory capabilities.<sup>[87–92]</sup> **Figure 4d** depicts the hierarchical structure of human skin, in which the waterproof epidermis provides protection, the dermis contains elastic tissue and fibers that give it resilience and toughness, and the hypodermis deforms to attenuate external pressure.<sup>[83]</sup>

Inspired by biological skins, tremendous efforts have been invested in developing artificial skins, including electronic and ionic skins. Electronic skins can mimic human skin sensations and introduce self-healing features to enhance durability and longevity. Tee et al. reported the first room-temperature self-healing electronic skin prepared by doping  $\mu\text{Ni}$  particles in a random branched polymer network connected by hydrogen bonds (Figure 4a).<sup>[56]</sup> The electrical conductivity of the organic-inorganic composite can be tuned by the content of  $\mu\text{Ni}$  particles. Due to the enhancement of mechanical strength by  $\mu\text{Ni}$  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. Then after 5 min of self-healing at room temperature, the LED lights 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  $\mu\text{Ni}$  particles decreases, reducing the composite's resistance, which ultimately leads to an increase in the brightness of the LED connected in series.

Ionic skins have also been widely studied and applied in sensing fields.<sup>[21,82,93–95]</sup> Recently, Wang et al. fabricated a fatigue-free ionic skin by embedding a polyurethane (PU)



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

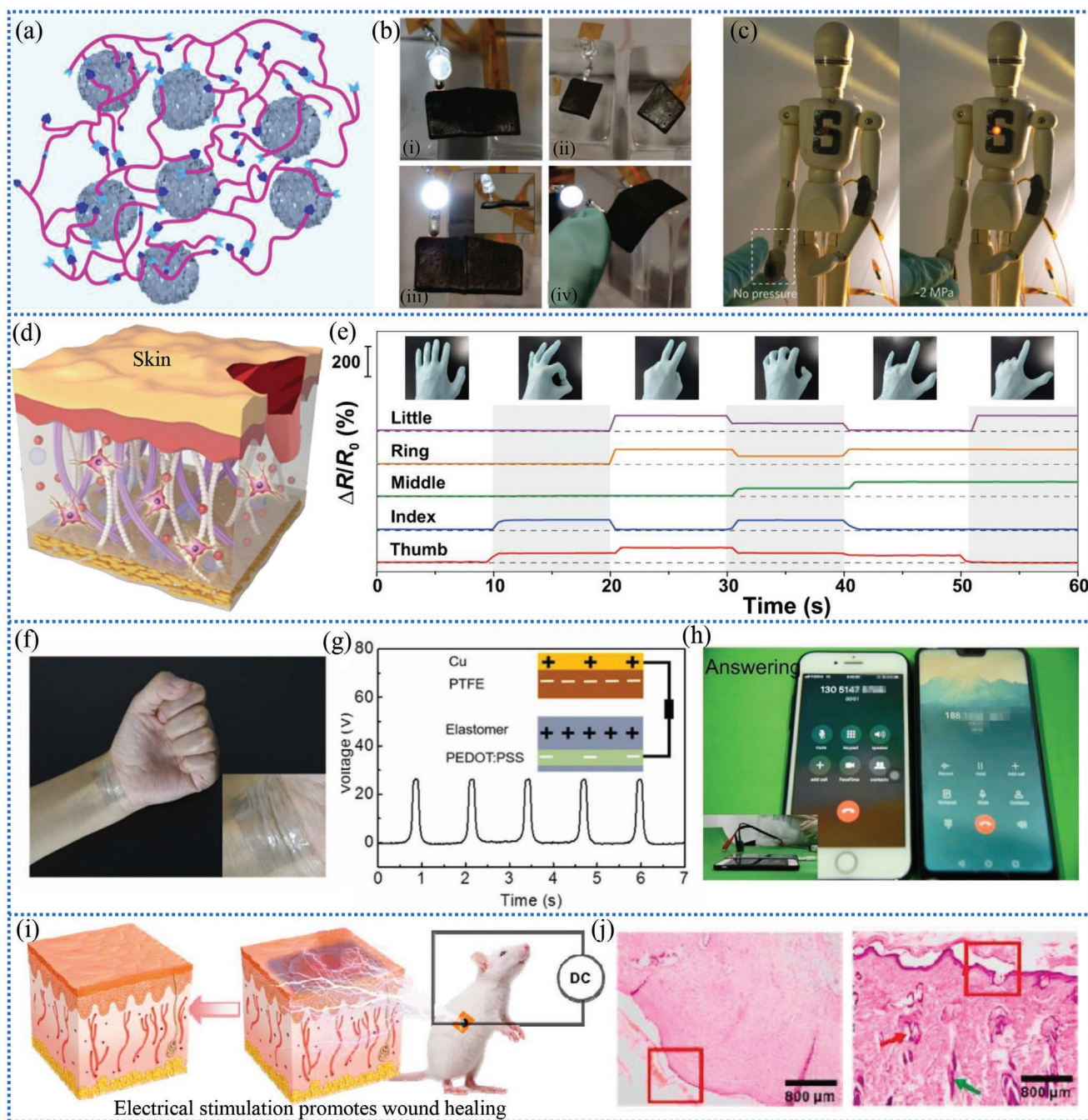
Function	Composition	Self-healing mechanism	Application	ref.
Insulator	PHMA/PEI blend polymer	Hydrogen bond	OFET and chemical sensor	[77]
Insulator	poly(urea-urethane) (PUU)	Disulfide bond	Transistor	[78]
Insulator	FeCl <sub>2</sub> -PDMS	Metal-coordination	OFET	[63]
Insulator	p-PVDF-HFP-DBP complex	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	[79]
Semiconductor	DPP/PDCA copolymer	Hydrogen bond	OFET	[64]
Semiconductor	DPP-TVt-PDCA/PDMS-PDCA-Fe elastomer	Metal-coordination	OFET	[65]
Semiconductor	PDPP <sub>urethane</sub> Copolymer	Hydrogen bond	OFET	[80]
Electronic conductor	μNi particles doped complex	Hydrogen bond	Pressure sensor	[56]
Electronic conductor	PU-DHP/Fe2	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	[81]
Electronic conductor	CNT embedded PDMS-MPU <sub>0.4</sub> -IU <sub>0.6</sub>	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 <sup>+</sup> and (B(OH) <sub>4</sub> ) <sup>-</sup> ions in PVA	Hydrogen bond	Ionic skin TENG	[71]
Ionic conductor	PVDF-co-HFP-5545/EMIOTf materials	Ion-dipole interaction	Soft actuator	[74]
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	[82]
Ionic conductor	PU scaffold in ionic liquid	Hydrogen bond and disulfide bond	Ionic skin	[83]

scaffold containing disulfide bonds into an ionic matrix containing hydrogen bonds.<sup>[83]</sup> The self-healing PU was electrospun and collected to obtain PU nanomesh. 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, 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 conductivity and mechanical strength of the ionic skin can recover. 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.<sup>[71,96]</sup> Liu et al. synthesized a self-healing elastomer by mixing a hard phase containing disulfide bonds with a soft matrix containing hydrogen bonds.<sup>[28]</sup> Different samples with thicknesses ranging from 3 to 100 μm

were fabricated by spin-coating. 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 as ultrathin and transparent. Using PTFE as triboelectrically negative material, the prepared TENG generates an alternating voltage during the contact-separation process (Figure 4g). The output voltages of TENG before cutting and after self-healing for 5 min are nearly identical, indicating the fast self-healing ability at the device level. Finally, a screen protector with TENG was 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).<sup>[31,97,98]</sup> Zheng et al. prepared a gelatin-based hydrogel containing PEDOT:PSS and MWCNTs to enhance the conductivity.<sup>[31]</sup> 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



**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.<sup>[56]</sup> 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.<sup>[83]</sup> 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 under the terms of CC BY license.<sup>[28]</sup> Copyright 2022, The Authors, published by MDPI. i) Schematic illustration of skin-inspired bio-electronic hydrogel used to promote wound healing after electrical stimulation. j) Hematoxylin and eosin 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.<sup>[31]</sup> Copyright 2021, Elsevier.

records the epidermal changes in the wound injury area. Thus, the hydrogel was used for wound healing promotion. As shown in Figure 4j, 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

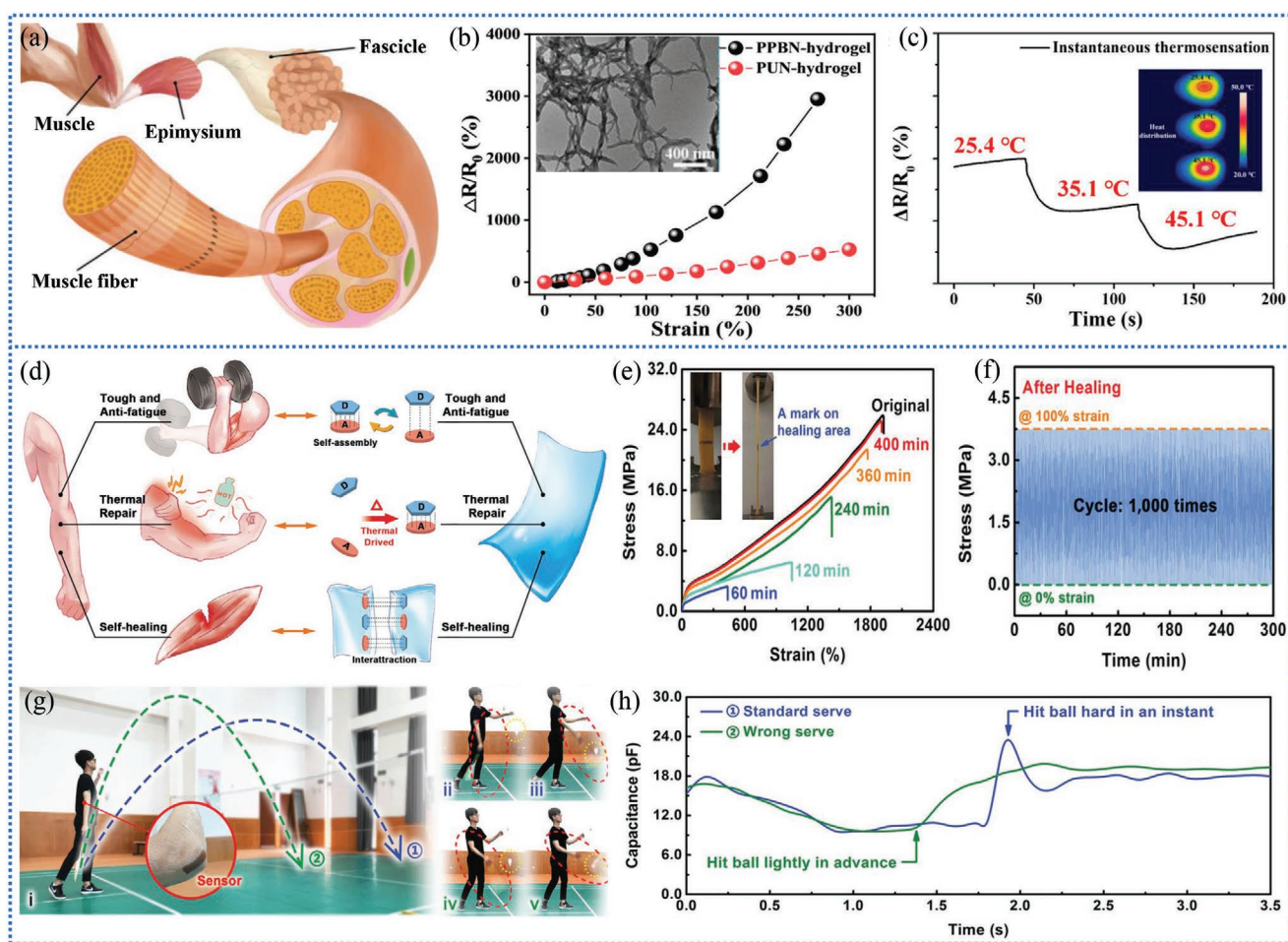


promoted the wound healing. This enables the hydrogel to be not only a strain sensor but also a scaffold for wound healing.

#### 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.<sup>[19]</sup> 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  $-26\text{ }^{\circ}\text{C}$ . The PPBN-hydrogel can withstand fracture stress over 35 kPa and reach a fracture strain of nearly 1000%. After 6 h 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



**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.<sup>[19]</sup> Copyright 2019, American Chemical Society. d) Schematic diagram of the muscle-inspired self-healing elastomer. Donor–acceptor 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 min. 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.<sup>[79]</sup> Copyright 2021, Wiley-VCH GmbH.

is achieved by using 1D 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.

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).<sup>[79]</sup> The DA-PU possesses super-tough mechanical properties, which can withstand a fracture stress of  $\approx 26$  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 was fabricated using DA-PU as the dielectric layer and filling layer 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 badminton players correct and incorrect movements (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, resulting in the recorded capacitance curve reaching 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 significant 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 >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).<sup>[41]</sup> 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 min, 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.<sup>[34]</sup> 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  $\text{Li}^+$  content, this is due to the breakage of the oriented arrangement of hydrogen bonds caused by  $\text{Li}^+$ , 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. 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, 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.

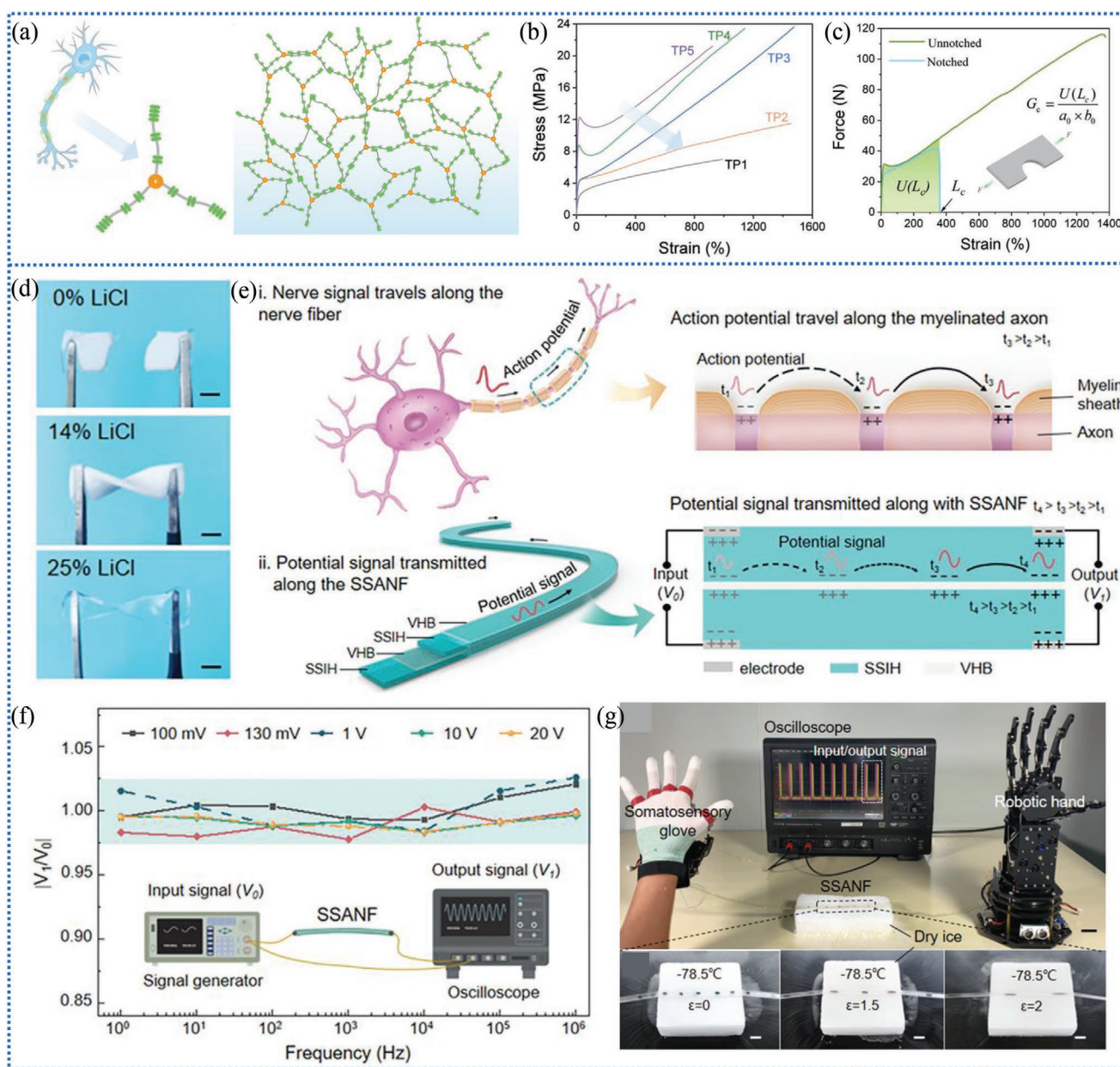
## 5. Animals-Inspired Self-Healing Soft Electronics

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).<sup>[99–102]</sup> Dopamine molecules contain catechol and amine groups, which are essential for mussels





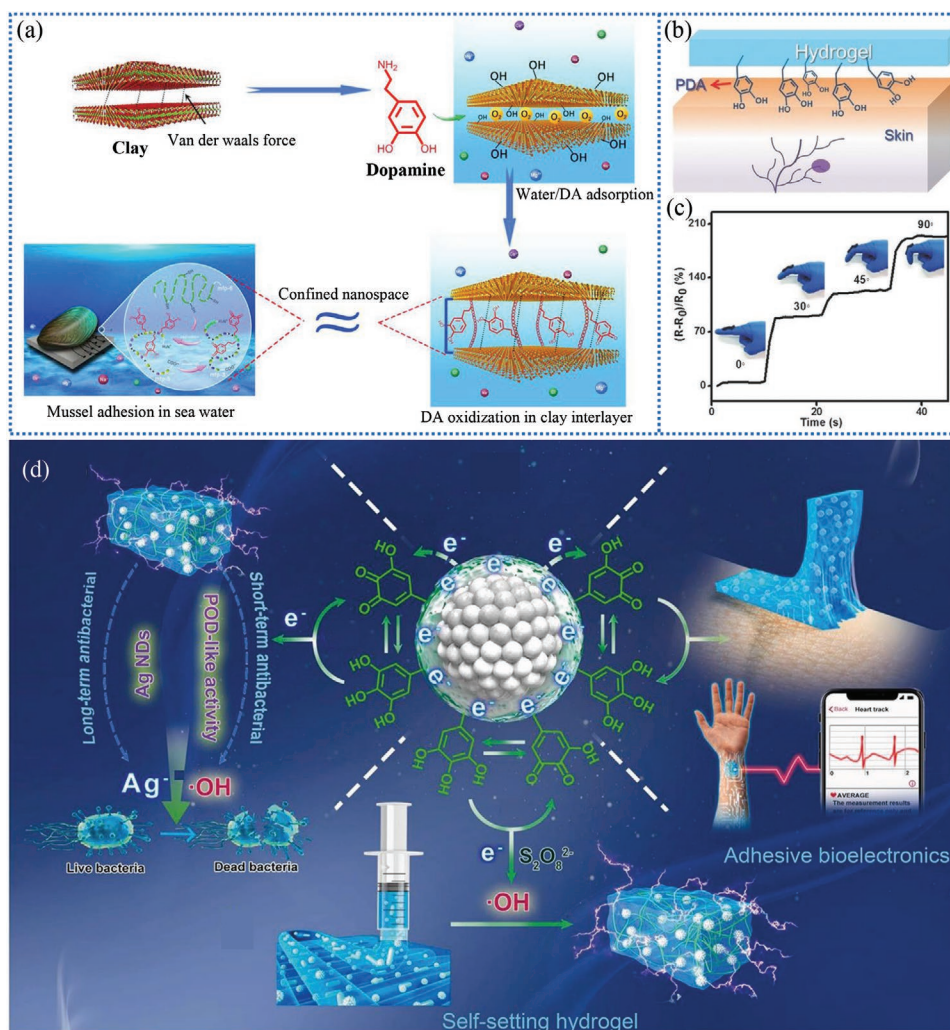
**Figure 6.** Neuron-inspired self-healing soft electronics. a) Schematic of the neuron-inspired self-healing telechelic polyurea (TP). b) Stress-strain curves of TPx, with TP3 exhibiting the best overall mechanical properties. c) Force–displacement curves for notched and unnotched TP3 samples, showing great crack tolerance. a–c) Reproduced with permission.<sup>[41]</sup> Copyright 2022, Wiley-VCH GmbH. d) Self-Healing ionic hydrogels with different LiCl contents. The transparency and flexibility of the hydrogel increase with Li<sup>+</sup> 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.<sup>[34]</sup> Copyright 2022, Wiley-VCH GmbH.

to achieve adhesion.<sup>[18]</sup> Inspired by this, Han et al. oxidized dopamine to form polydopamine (PDA) in clay nanosheets and generated sufficient free catechol groups. Then acrylamide monomer was added for in situ polymerization. The formed hydrogel possesses good adhesion and superior toughness due to its mussel-like structure (Figure 7a).<sup>[103]</sup> The researchers have also used PDA to reduce graphene oxide (GO) following acrylamide monomer polymerization.<sup>[100]</sup> 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  $\pi$ - $\pi$  stacking interactions. The hydrogel can bond to human skin tightly due to the free catechol moiety of PDA (Figure 7b).<sup>[22]</sup> Incorporating functionalized single-wall





**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.<sup>[103]</sup> 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.<sup>[22]</sup> 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.<sup>[45]</sup> Copyright 2021, Royal Society of Chemistry.

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 s 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.<sup>[42]</sup>

Jia et al. synthesized a mussel-inspired ultra-small tannic acid chelated Ag (TA-Ag) nanozyme catalyzed hydrogel.<sup>[45]</sup> Figure 7d shows the structure of the nanozyme, in which the

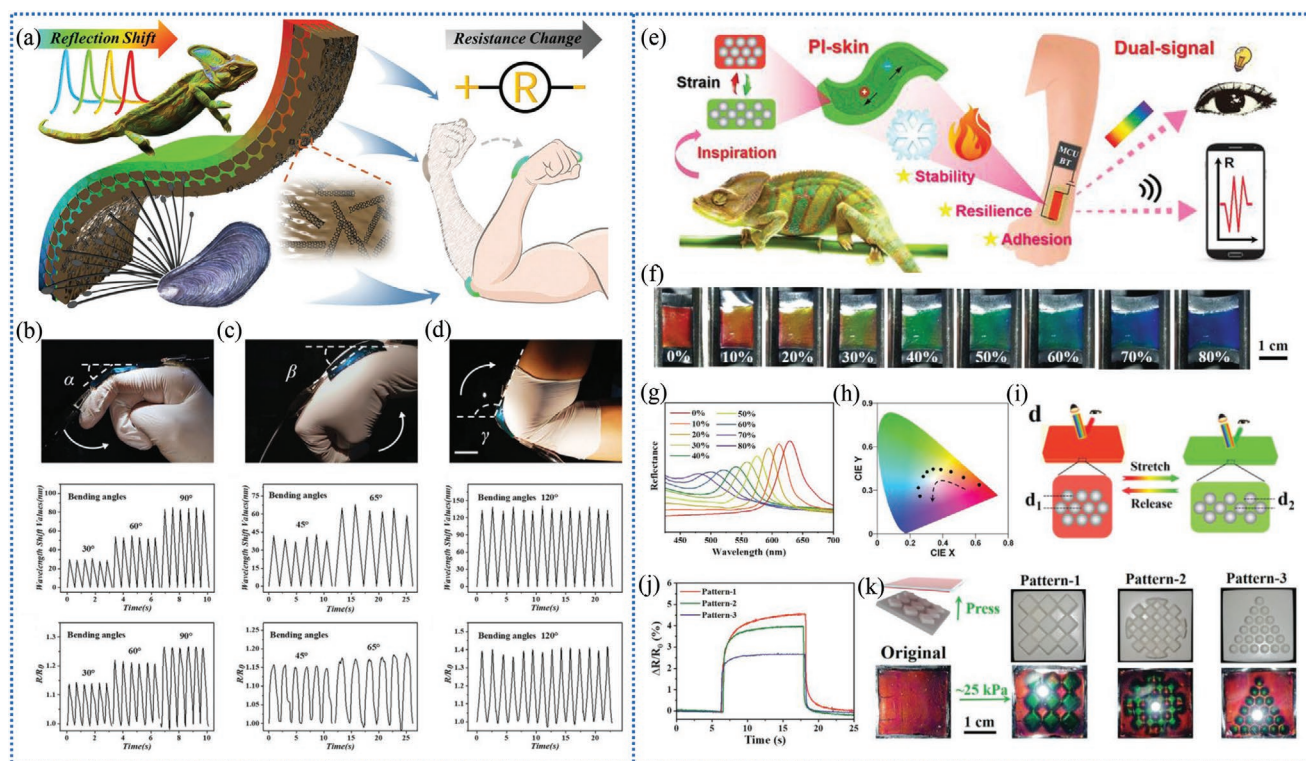
abundant phenolic hydroxyl groups ensure the adhesiveness of the hydrogel. The hydrogel can adhere to various surfaces, such as cherry, glass, mouse liver, and metal. Nanozyme catalysis endows the hydrogel with self-setting properties, facilitating their injection at the injury sites. The hydrogel can 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 and capture prey because of their unique structural color, which is produced by micro- and nanostructures.<sup>[104]</sup> When stimulated, chameleons stretch their skin, causing the distance between the guanine nanocrystals under the skin to

widen, further driving a change in reflectivity that changes the color they display.<sup>[105]</sup> As shown in **Figure 8a**, inspired by the chameleon's structural color and the mussel adhesive protein, Wang et al. fabricated a self-healing film by doping conductive CNTs-PDA fillers into PU inverse opal matrix.<sup>[36]</sup> 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. Meanwhile, 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.

Sun et al. fabricated a biomimetic photonic-ionic skin (PI-skin) by sandwiching PS@SiO<sub>2</sub> microspheres in an ionic liquid located polyzwitterionic PSBMA network.<sup>[20]</sup> SEM image shows that PS@SiO<sub>2</sub> microspheres possess an ordered array structure with the polymer filling in the gaps of the microspheres, locked by covalent cross-linking and noncovalent 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. In contrast, visual color changes can clearly show the shape, location, and pressure distribution (**Figure 8k**). This provides a promising solution for the emerging field of human-machine interaction sensing.



**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.<sup>[36]</sup> 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 changes 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.<sup>[20]</sup> Copyright 2022, Wiley-VCH GmbH.



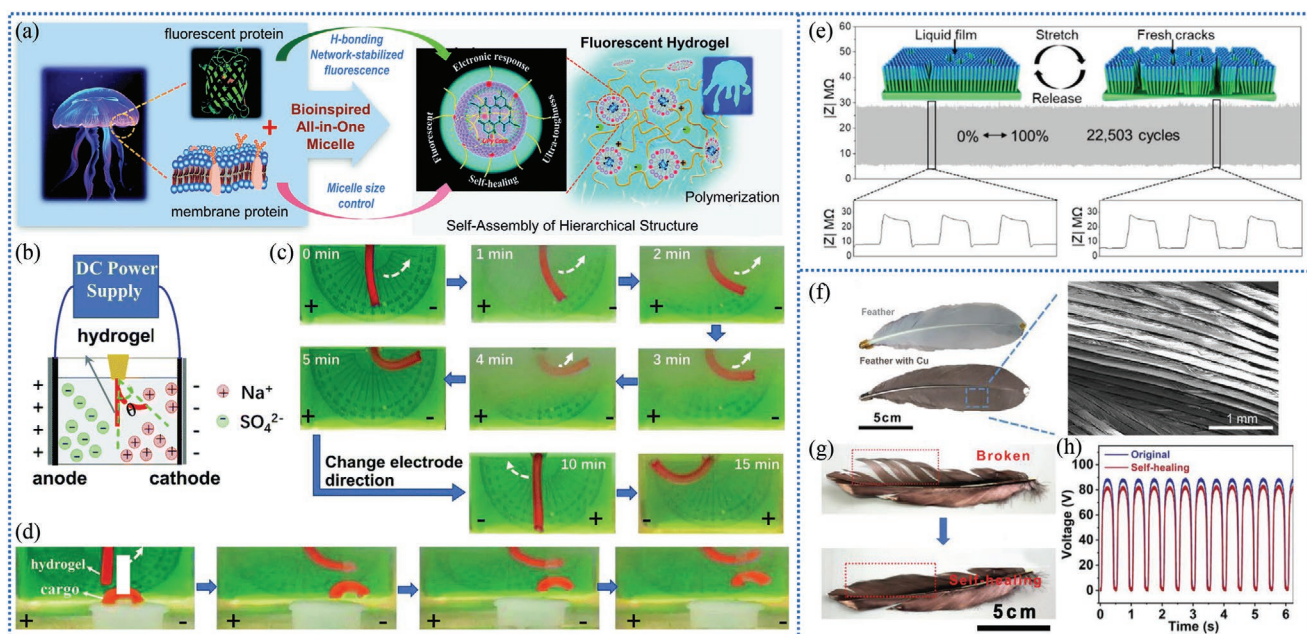
### 5.3. Other Animals-Inspired Self-Healing Soft Electronics

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.<sup>[37]</sup> 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, while 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  $\text{Na}^+$  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  $90^\circ$  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 researchers tested the stress–strain curves of the self-healing hydrogels after multiple electric pulses. The results showed

that the electrical stimulation only slightly affected the self-healing properties. The introduction of self-healing properties improves the fatigue resistance of the device. Such hydrogels with all-around properties are believed to provide new insights into smart hydrogels, such as applications in soft robotics and artificial muscles.

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.<sup>[106]</sup> 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.

Miao et al. fabricated a self-healing liquid film by mimicking the structure of tear films on animal corneas.<sup>[107]</sup> As shown in **Figure 9e**, an ethanol solution of ionic liquid was dropped on the PDMS microvilli. 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 were formed between the microvilli during stretching, and self-repair occurred after release due to capillary forces. Compared to



**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  $\text{SO}_4^{2-}$  ions are immobilized in the hydrogel and the counter  $\text{Na}^+$  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.<sup>[37]</sup> 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.<sup>[107]</sup> 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.<sup>[27]</sup> Copyright 2021, Wiley-VCH GmbH.



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 was then used as a strain sensor for human motion monitoring, where the resistance increased with the bending of the finger or wrist.

In addition to biomimetics, primitive biomaterials have recently been used as self-healing electronics.<sup>[27]</sup> 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 building self-healing electronics.

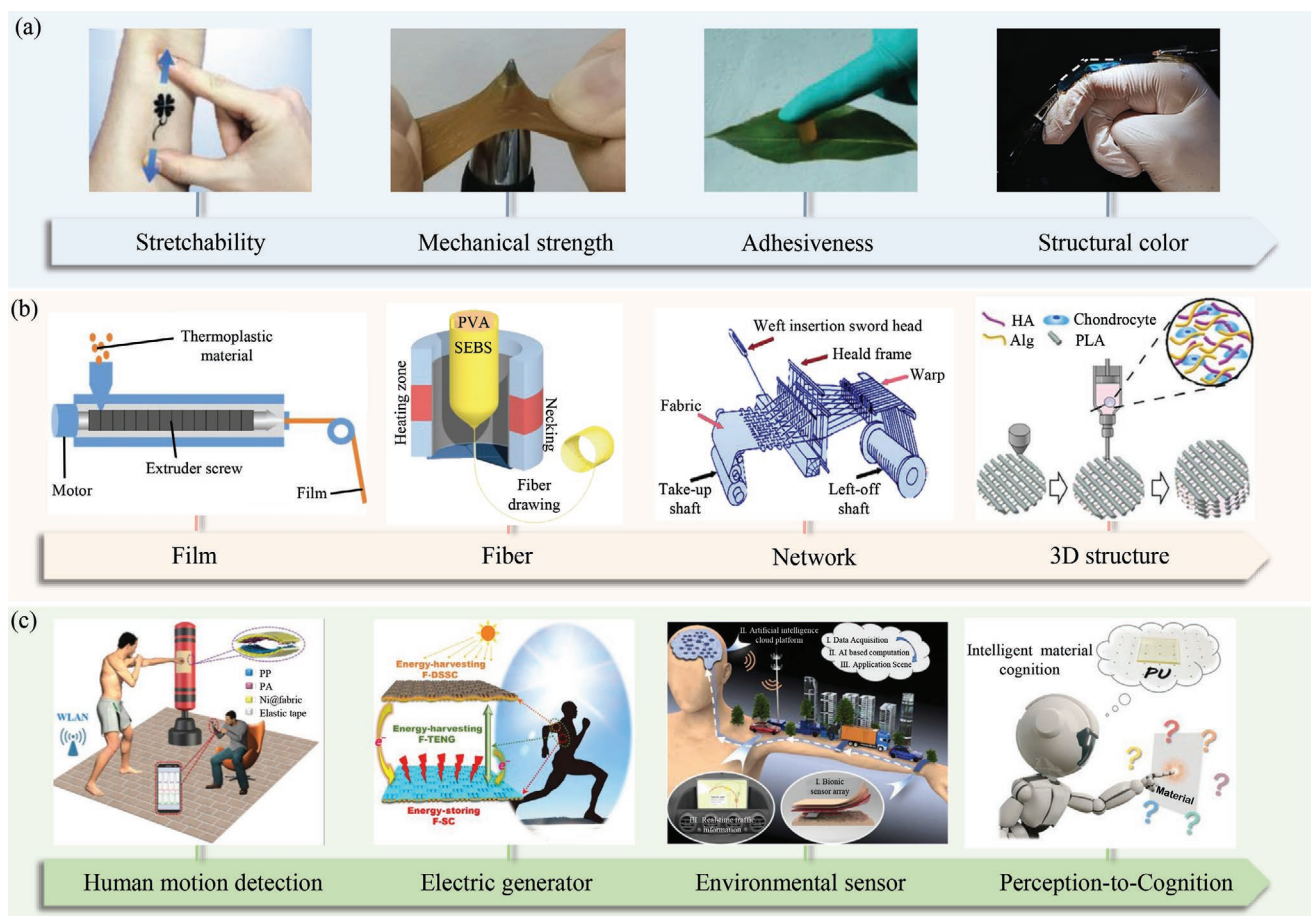
## 6. Conclusions and Outlook

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: 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 besides self-healing, inspiring many unique self-healing soft electronics (Figure 10a). Electronic and ionic skins mimic the stretchability of human skins to prevent cracking, and they are commonly used as strain sensors to monitor human motions.<sup>[108]</sup> 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.<sup>[79]</sup> Neurons communicate with each other through spatial axons, allowing signals to be transmitted throughout the body. Thus, polymers and devices with excellent mechanical properties and information transfer capabilities have been inspired. Mussels can attach their soft body to wet rocks to survive in the fierce waves or tides, which encourages self-healing soft electronics with adhesive properties.<sup>[93]</sup> 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.<sup>[36]</sup> Typical works in these areas are comprehensively discussed, including material synthesis, device fabrication, and applications.

The combination of self-healing capabilities and other bioinspired properties in soft electronics is attractive. However, despite these soft electronics' fascinating properties, their applications primarily 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, artificial muscles and biomimetic actuators have been designed inspired by the mechanical toughness of muscles, while self-healing properties enable the complete healing of realistic macroscopic injuries.<sup>[117,118]</sup> Mussels possess underwater adhesion and self-healing ability. Correspondingly, mussel-inspired soft electronics can be used as wearable devices to monitor human health during exercise and sweating.<sup>[119]</sup> The self-healing soft electronics inspired by the chameleon can be applied to optical camouflage, making the devices be hidden in the surrounding environment and having excellent application potential in military and underwater exploration.<sup>[32]</sup> Moreover, in addition to human motion detection<sup>[113]</sup> and electric generator,<sup>[114]</sup> self-healing soft electronics are expected to be combined with artificial intelligence (AI) analysis systems for environmental sensing (Figure 10c).<sup>[115]</sup> AI is also promising in motivating the evolution of perception to cognition, having been demonstrated to distinguish materials through a single touch.<sup>[116]</sup>

One concern in bioinspired self-healing materials is their mechanical performance. Self-healing properties rely on the exchange of dynamic bonds and reversible supramolecular interactions, which require good mobility of molecular chains. This conflicts with the chain rigidity that contributes to mechanical toughness. Rational molecular structure design can help address this issue, like combining strong and weak dynamic bonding or integrating hard segments into soft segments.<sup>[120–122]</sup> Self-healing of the entire electronic device after external damage is another challenge. Currently, most reported studies focus on self-healing a component in electronic devices. There are few reports on the self-healing of the whole device because it requires not only self-healing of all parts but also a good bonding interface between different components. Therefore, on the one hand, more self-healing functional materials, such as conductors and semiconductors, need to be designed and synthesized. On the other hand, adhesive self-healing materials are required to strengthen the interface between different components. It is also feasible to use the same polymer as the matrix of each layer of the device, which ensures good compatibility of the mechanical properties of the whole system during usage. 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,<sup>[109]</sup> thermal-drawing technique for fibers,<sup>[110]</sup> and even weaving technique for textiles (Figure 10b).<sup>[123]</sup> 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.<sup>[124]</sup> Moreover, the 3D printing technique has been adopted for fabricating self-healing soft electronics with precise and designable device structures.<sup>[32,108,125,126]</sup> It is worth mentioning that the before-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



**Figure 10.** The characteristics, possible preparation methods, and applications of bioinspired self-healing soft electronics. a) Stretchability, mechanical toughness, adhesiveness, and structural color of self-healing soft electronics inspired by human skin, muscles, mussels, and chameleons, respectively. Reproduced with permission.<sup>[108]</sup> Copyright 2019, Wiley-VCH GmbH. Reproduced with permission.<sup>[79]</sup> Copyright 2021, Wiley-VCH GmbH. Reproduced with permission.<sup>[93]</sup> Copyright 2020, Royal Society of Chemistry. Reproduced with permission.<sup>[36]</sup> Copyright 2020, Wiley-VCH GmbH. b) Possible approaches for large-scale manufacture of self-healing soft electronic materials: extrusion molding for films, thermal drawing technique for fibers, weaving for fabrics, and 3D printing for 3D structures. Reproduced with permission.<sup>[109]</sup> Copyright 2022, Springer Nature. Reproduced with permission.<sup>[110]</sup> Copyright 2021, Springer Nature. Reproduced under the terms of CC BY license.<sup>[111]</sup> Copyright 2021, The Authors, published by Public Library of Science. Reproduced with permission.<sup>[112]</sup> Copyright 2020, Elsevier. c) Tremendous applications of bioinspired self-healing soft electronics: human motion detection, electric generator, environmental sensor, and intelligent material cognition. Reproduced with permission.<sup>[113]</sup> Copyright 2021, Wiley-VCH GmbH. Reproduced with permission.<sup>[114]</sup> Copyright 2016, American Association for the Advancement of Science. Reproduced with permission.<sup>[115]</sup> Copyright 2022, Elsevier. Reproduced with permission.<sup>[116]</sup> Copyright 2022, Wiley-VCH GmbH.

the mechanical properties of materials without sacrificing self-healing ability through rational molecular design is the key to applying 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. It is hopeful of improving the service life of electrical devices and even providing a new generation of electronic devices.

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## Conflict of Interest

The authors declare no conflict of interest.

## Keywords

bioinspired electronics, hydrogel, polymers, self-healing, soft electronics

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