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Review article

Bio-inspired flexible electronics for smart E-skin☆

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A B S T R A C T

"Learning from nature" provides endless inspiration for scientists to invent new materials and devices. Here, we review state-of-the-art technologies in flexible electronics, with a focus on bio-inspired smart skins. This review focuses on the development of E-skin for sensing a variety of parameters such as mechanical loads, temperature, light, and biochemical cues, with a trend of increased integration of multiple functions. It highlights the most recent advances in flexible electronics inspired by animals such as chameleons, squids, and octopi whose bodies have remarkable camouflage, mimicry, or self-healing attributes. Implantable devices, being overlapped with smart E-skin in a broad sense, are included in this review. This review outlines the remaining challenges in flexible electronics and the prospects for future development for biomedical applications.

Statement of significance

This article reviews the state-of-the-art technologies of bio-inspired smart electronic skin (E-skin) developed in a "learning-mimicking-creating" (LMC) cycle. We emphasize the most recent innovations in the development of E-skin for sensing physical changes and biochemical cues, and for integrating multiple sensing modalities. We discuss the achievements in implantable materials, wireless communication, and device design pertaining to implantable flexible electronics. This review will provide prospective insights integrating material, electronics, and mechanical engineering viewpoints to foster new ideas for next-generation smart E-skin.

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1. Introduction

Smart electronic skins have been promoted by advances in materials, mechanics, electronics, and information technologies. For instance, with the evolution of stretchable polymeric materials, a diverse set of stretchable sensors for biomimetics has been demonstrated, such as pressure/strain sensors, thermal sensors, optical sensors, and biochemical sensors. The incorporation of repeatable bond-forming in response to specific stimuli in polymer gels has endowed flexible electronics with self-healing features. New types of organic semiconductors expand the frontiers of smart E-skin to the integration of artificial nerves, both synaptic and afferent. Smart electronic skins offer attractive opportunities in various fields such as robotics, prosthetics, and health-monitoring technologies.

Natural skins have always been a source of great inspiration for the development of smart electronic skins. Human skin, which is highly integrated with numerous mechanoreceptors, plays a critical role in the perception of the physical world. Scientific questions, such as how to reconstruct a natural sensing ability, motivate the evolution of smart flexible electronics. Researchers have actively practiced a "learning-mimicking-creating" (LMC) cycle inspired by natural human skin. Various flexible sensors have been developed to detect haptic stimuli by mimicking the tactile sensation of human skin. The development of flexible sensors started with a focus on physical variable detection, such as pressure or temperature sensing. Subsequently, the functionalities of the devices were expanded to monitor biochemical markers, such as lactic acid or glucose. Smart flexible electronic skins have been advancing in the LMC cycle, with inspiration from animal skins. The unique skin features of chameleon, mussels, and octopus have encouraged researchers to render electronic skins with the ability

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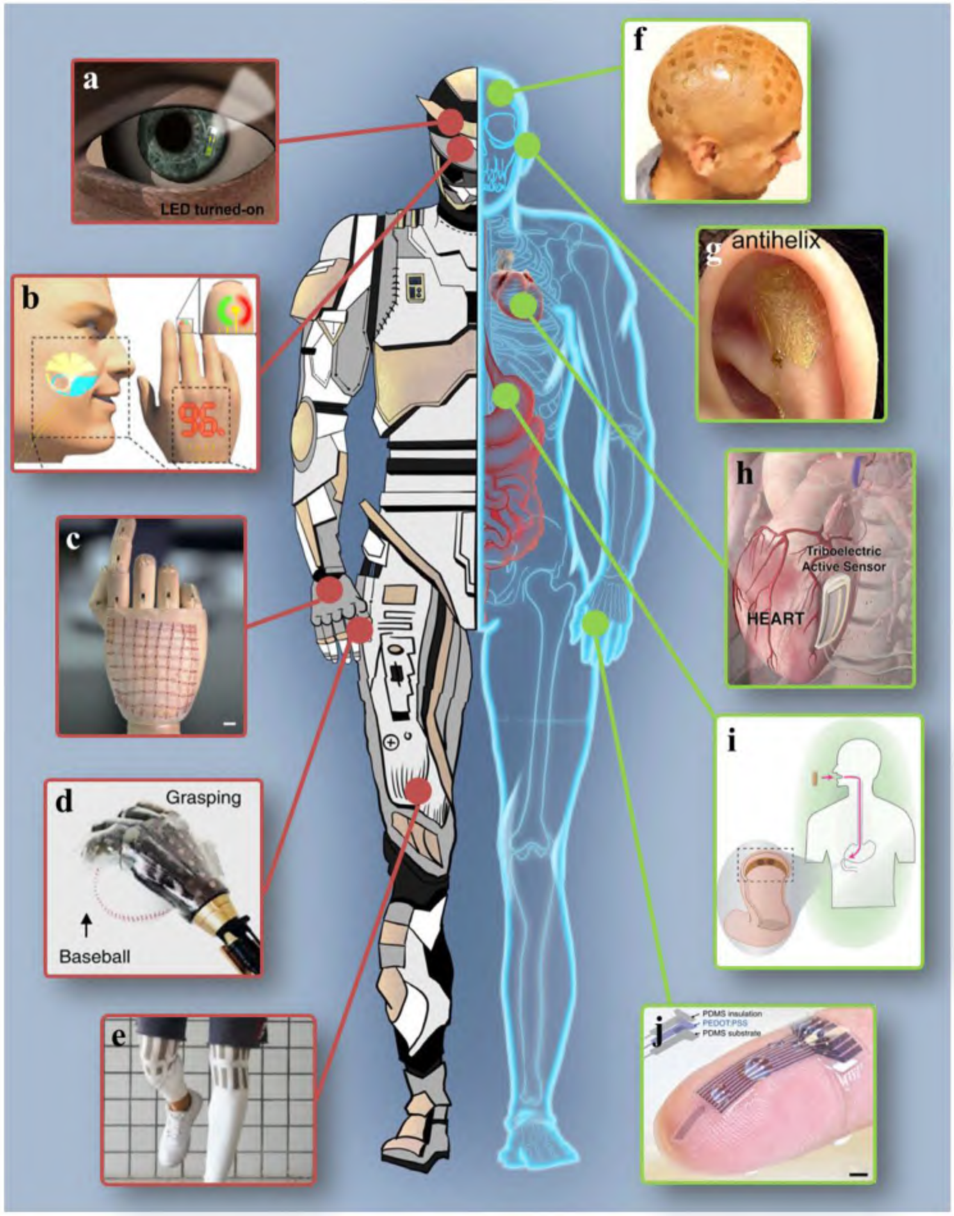

Fig. 1. Examples of bio-inspired smart electronic skin (E-skin) developed in the "learning-mimicking-creating" (LMC) cycle. (a) Smart contact lens containing functional devices (rectifier, LED, and glucose sensor). Reproduced with permission from [156]. (b) Flexible optoelectronic skins. Reproduced with permission from [56]. (c) A conformable multifunctional sensor matrix on a hand. Reproduced with permission from [69]. (d) A prosthetic hand integrated with stretchable sensors and actuators catching a baseball. Reproduced with permission from [68]. (e) A knitted flexible sensor on legs for recognition of walking motion. Reproduced with permission from [157]. (f) Epidermal EEG electrodes. Reproduced with permission from [158]. (g) Antihelix-mounted artificial skin for EEG recording. Reproduced with permission from [86]. (h) Implantable triboelectric active sensor for heart rate monitoring. Reproduced with permission from [138]. (i) Edible current sources. Reproduced with permission from [132]. (j) A soft neural probe. Reproduced with permission from [159].

to change colors or shapes in response to certain external stimuli. Additionally, the LMC cycle is derived not only from the skin outside the body, but also from the organ envelopes inside the body. Many flexible electronic devices, such as implantable sensors, power source/energy harvesters, artificial neurons, and synaptics, have been proposed with attractive functionality and biocompatibility. Augmentative features are continuously added to enhance their performance. Some smart flexible electronic devices have the ability to sense multiple stimuli, record sensation information, or even generate feedback in terms of mechanical or nociceptive modality to the skin.

In this review, we highlight state-of-the-art technologies in the field of bio-inspired smart flexible electronics (Fig. 1). They are summarized according to the device development streamline, from relatively simple designs/isolated functions to increasingly integrated formats for sensing not only physical variables (pressure, strain, temperature, light, etc.), but also biochemical cues in skin secretions or near the skin tissues. Flexible electronics mimicking chameleon or octopus skins are included in our discussion to promote inspiration for future development. This review integrates the general development of implantable flexible electronics as "smart skins" for tissue/organ envelopes. There are several comprehensive reviews on the development of bio-inspired smart skins [1–5]. However, this is a rapidly growing field of research. Here, we attempt to highlight the latest advances in smart E-skin, including those featured with wireless communications and implantable devices, followed by several remaining critical challenges that the community must address. It will provide useful prospective views inspiring the development of next-generation smart E-skin, especially new materials engineering and integration with artificial neural networks.

2. E-skin for sensing physical variables

The skin of a human or animal can perceive a wide range of sensations, including pressure, stress, temperature, and light. In this section, we discuss the skin-inspired sensors for sensing these physical stimuli based on the transducing principle, improving sensing performance, and integrating functionalities for multiple input detections.

2.1. Flexible electronics for pressure sensing

Flexible pressure sensors constructed on soft materials usually convert mechanical stimuli to electrical signals, such as resistance, capacitance, and electrical potential. Researchers have explored approaches to improve the sensing performance of various types of devices. Below, we highlight key developments in flexible pressure sensors based on resistive, capacitive, and piezoelectric sensing mechanisms.

As one of the most frequently employed approaches, resistive sensing devices are usually constructed on soft polymers that combine a number of electrically conductive sensing elements, such as metal, semiconductor thin films, and nanocomposites. External mechanical stimuli are transduced into the geometrical changes of sensing elements, inducing variations in either bulk resistivity or in contact resistance between the two conductive modules. In the bulk resistivity change mode, the most explored methods include creating cracks [6], detachments, and defects [7] in the conductive sensing film under mechanical pressure. Contact resistance is another tunable parameter based on mechanical forces/pressures [8–10]. Zhu et al. devised a resistive pressure sensor by assembling a microstructured polydimethylsiloxane (PDMS) film coated with reduced graphene oxide (rGO) and a flexible electrode layer. The pyramid microstructures deformed under external pressure, resulting in an increase in the contact area between the rGO-coated pyramids and the electrode substrate (Fig. 2a) [11]. Such surface microstructures cause highly sensitive deformation at low pressure levels, whereas they decrease the sensitivity at high pressure levels, leading to nonlinearity over a large pressure range. Introducing multi-layered, hierarchical structures (Fig. 2b) [12], or porous foams [13] has been demonstrated to improve the sensitivity and linearity of the contact deformation over a wide pressure range. These valid approaches have broadened the applicability of these devices to prosthetic E-skin.

A capacitive flexible pressure sensor is mainly constructed as a parallel-plate capacitor with a dielectric polymer sandwiched between two flexible electrode layers. Sensors transduce external forces/pressures into mechanical deformations/deflections of the dielectrics or electrodes, which results in changes in the dielectric constant, overlapping area, or separation distance [14–17]. In general, the relationship between pressure and mechanical deformation is dominated by the modulus of the dielectric layer; therefore, a well-designed material with a reduced modulus would dramatically improve the sensitivity. Strategies for introducing surface microstructures into dielectrics have been widely investigated. Bao et al. constructed dielectric PDMS layers with micropatterns as a sensing dielectric, allowing for a reduction in the modulus and viscoelasticity of the elastomer (Fig. 2c) [17–19]. Liu et al. developed a porous structure in a soft Ecoflex elastomer with four parallel-plate capacitors to resolve both normal and tangential mechanical loads [16]. Additionally, the interfacial capacitance at the electronic-ionic interface leads to another trend in capacitive sensing. Nie et al. introduced ionic fillers into a hydrogel matrix as a dielectric in a flexible sensor with an ultrahigh interfacial capacitance, achieving a limit of detection as low as 1.8 Pa with an E-skin device [15].

To mimic the ability of human skin to sense high-frequency vibrations, piezoelectric pressure sensors provide feasible technical solutions for highly sensitive dynamic force sensations. The sensors generally utilize piezoelectric materials that can produce piezopotential by strain-induced ionic polarization under variations in applied pressure. Remarkably, piezoelectric pressure sensors do not require external battery components and provide an excellent response to dynamic forces. Polymeric materials of polyvinylidene fluoride (PVDF) and its copolymers, and inorganic materials such as zinc oxide, lead zirconate titanate, and barium titanate are good candidates for piezoelectric pressure sensing solutions (ZnO, InN and GaN mixed coaxial piezoelectric fibers) [20–24]. Zhu et al. developed InN and GaN mixed coaxial piezoelectric fibers to reach up to 10.89 mV kPa^{-1} in the pressure range of 80–230 kPa (Fig. 2d) [23]. Liu et al. developed a 2D piezotronic transistor array by well ordering a ZnO nanoplatelet array, achieving a high sensitivity of 78.23 meV MPa^{-1} and a high resolution of 12700 dpi [25]. Alternatively, triboelectric-based sensor combining triboelectrification and electrostatic induction is another research area of intense focus in active pressure detection [26,27]. Triboelectric-based flexible devices are not only suitable for dynamic pressure sensing, but also pave the way for the development of self-powered flexible electronics. In a recent report, Wang et al. exhibited an ultralight single-electrode triboelectric yarn for harvesting biomechanical energy and monitoring tiny signals from humans or insects (Fig. 2e) [28].

2.2. Stretchable electronics for strain sensing

Stretchable strain sensors mainly measure changes in resistance due to the disconnection or tunneling effect between adjacent conductive fillers in stretchable polymeric materials [29,30]. In contrast to conventional strain gauge sensors, stretchable strain sensors are suitable for large strain-associated detections, especially those associated with human body motions such as stretching, bending, and torsion [31–33]. Boland et al. achieved an excellent conductive stretchable composite by infusing graphene into natural rubber, displaying a 10^4-fold increase in resistance at strains exceeding 800% (Fig. 3a) [34]. In this method, the increase of filler contents in polymers can improve the conductivity of the composite materials. It is of importance to make conductive particles uniformly dispersed in the polymers in order to reduce location-to-location variations in conductivity [35]. As a widely-use method, a conductive composite can be prepared by blending the suspension of conductive fillers and pre-cured elastomer, followed by evaporation of the blending solvents. Alternative methods have been adapted by directly embedding a stretchable conductive network in an elastic matrix [36]. Zhang et al. successfully developed an elastic strain sensor using carbonized plain weave cotton fabric and encapsulated it in an elastic matrix [37]. In addition, to customize the sensitivity and stretchability of a strain sensor, Chen et al. proposed a mechanocombinatorial strategy by utilizing a mechanically heterogeneous substrate. The authors demonstrated that strain, governed by both mechanical and structural parameters, could be redistributed over a stretchable film with heterogeneous patterns (Fig. 3b) [38]. The development of anisotropic conductive materials achieved high gauge factors while maintaining resiliency to various loading conditions such as bending, twisting, and pressing. [39]. Miniaturization in the thickness of an electronic tattoo enabled a highly stretchable, conformal, and sticky strain sensor, with the crease amplification effect which can boost the output signal by three times [40].

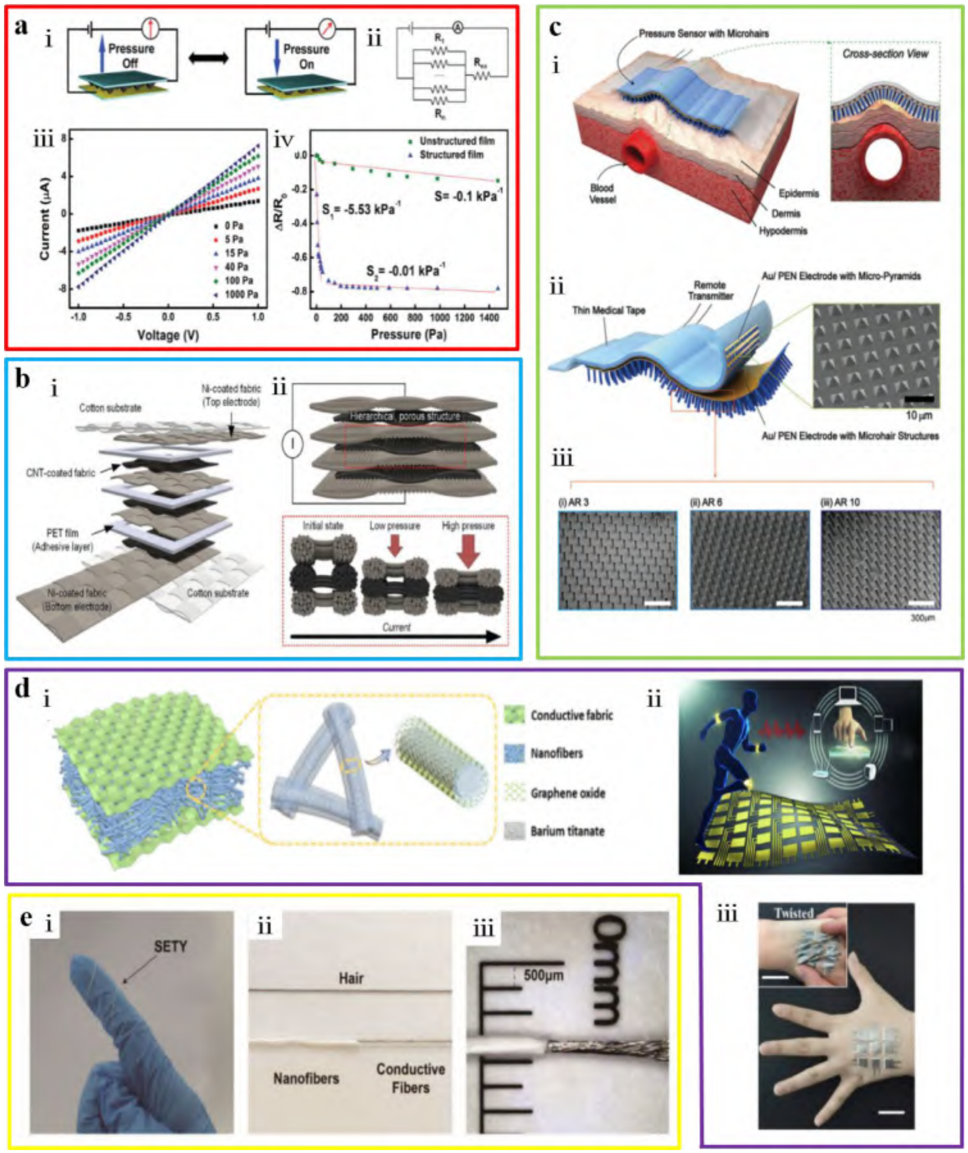

Fig. 2. (a) Schematic illustration of the working mechanism of the sensor in response to pressure; The equivalent circuit diagram; I–V curves of the pressure sensor with applied pressure; Pressure-response curves of the unstructured and microstructured rGO/PDMS films. Reproduced with permission from [11]. (b) Schematic illustration of the sensor structure, which alternately stacked Ni and CNT-fabrics, PET film, and cotton substrates; Sensing mechanism of the tactile sensor. Reproduced with permission from [12]. (c) A highly skin conforming and pulse-detectable pressure sensor using microhairs structures; Reproduced with permission from [19]. (d) Structure design of the highly shape adaptive electronic skin based on piezoelectric core-shell nanofibers; Optical photograph of the electronic skin conformably attached on the back of hand; Illustration of the proof-of-concept of fabricated electronic skin for joint motion monitoring and tactile sensing. Reproduced with permission from [23]. (e) The visual fineness contrast of the single-electrode triboelectric yarn with finger, hair and scale. Reproduced with permission from [28].

2.3. Flexible electronics for thermal sensing

In addition to sensing mechanical stimuli, accurate measurement of temperature is critical in the development of E-skin. Thermocouples are classical instruments for temperature sensing [41–43]. Nevertheless, it is difficult to apply thermocouples to flexible and conformable temperature sensing owing to the inflexible construction materials and complicated fabrication processes. Alternatively, thermistors render a wide range of working temperatures with good sensitivity, flexibility, and simple fabrication processes. Thermistors rely on the temperature-dependent resistance changes of building blocks, such as metals, carbon-based materials, polymers, or ionic hydrogels [44,45]. According to the resistive response to the temperature, there are two types of thermistors: positive temperature coefficient (PTC) resistors and negative temperature coefficient (NTC) resistors. Katerinopoulou et al. demonstrated a printable NTC ceramic-based temperature sensor that yields a high thermal coefficient with a 4.0% change in resistance at 25°C (Fig. 3c) [46]. PTC-based thermal sensors intrinsically suffer from low sensitivity and poor durability. Lee et al. addressed this challenge and greatly promoted the sensitivity of a polymer thermistor by adding silver fractal dendrites (AgFDs) to a polyacrylate (PA) matrix. The AgFDs–PA composite film exhibited a superior PTC effect (104 $\Omega°C^{-1}$) in the human body temperature range (34–37°C) (Fig. 3d) [47].

2.4. Flexible electronics for light sensing

Flexible electronics for optical sensing have been intensively investigated with new ideas inspired by animal eyes. Remarkably, organic photodetectors (OPDs) have received extensive attention in the fields of flexible health monitoring, image detection, and

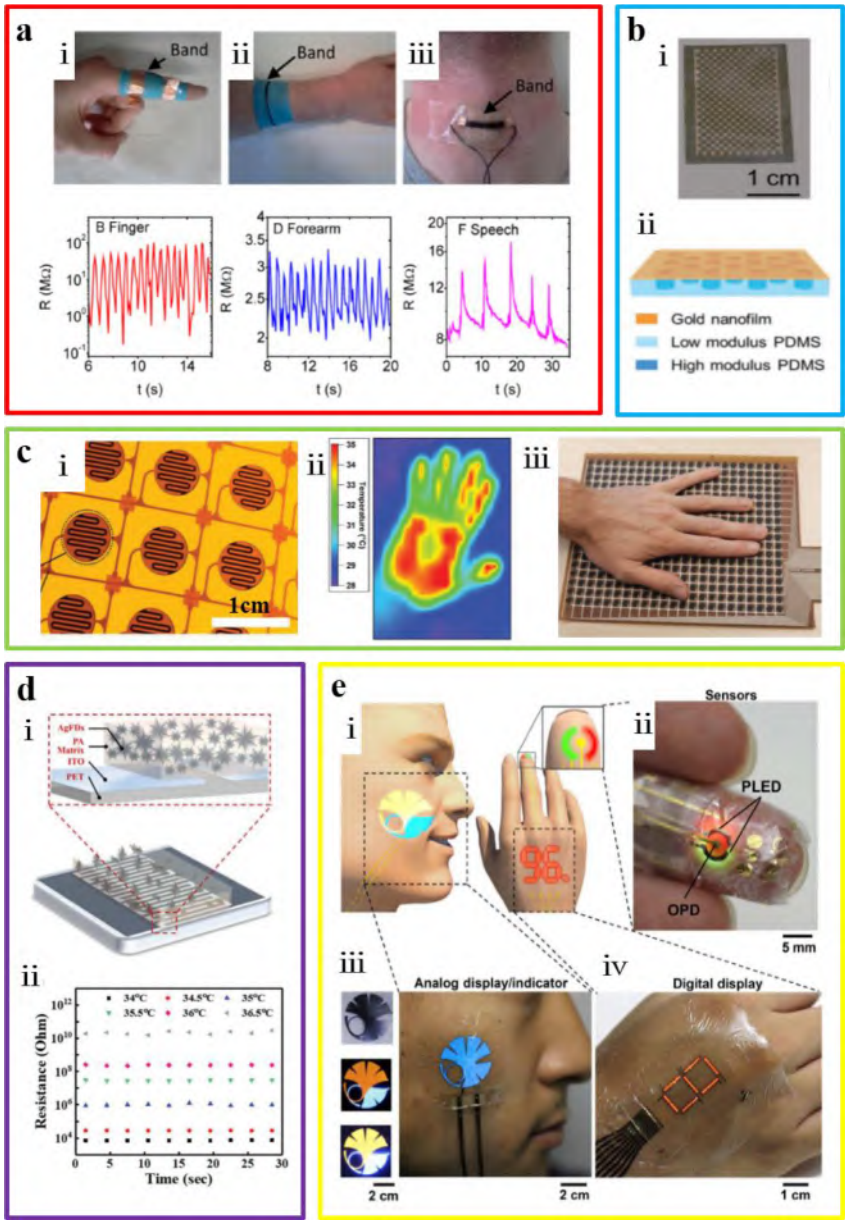

Fig. 3. (a) Applications of conductive rubber bands to detect body motions. Reproduced with permission from [34]. (b) Optical image and schematic of the heterogeneous substrate-based strain sensor. Reproduced with permission from [38]. (c) Optical images of screen-printable temperature sensors with calibrated readout of the temperature of a hand placed on a sensor sheet. Reproduced with permission from [46]. (d) Schematic illustration of a structure of a AgFDs–PA-based flexible thermistor; Changes in output resistance of the thermistor as a function of temperature; output resistance of the thermistor from 34°C to 36.5°C. Reproduced with permission from [47]. (e) Smart e-skin system comprising health-monitoring sensors, displays, and ultraflexible polymer light-emitting diodes. Reproduced with permission from [56].

artificial eyes [48–51]. Photodiodes and phototransistors are two of the most commonly used OPDs. Photodiodes are constructed with organic photosensing layers sandwiched between the anode and cathode electrodes. By absorbing photon energy under illumination, the active layer generates electron–hole pairs, which are separated and collected by the electrodes [52–54]. In the case of phototransistors, the active layer is typically integrated with a charge-transport layer. The electron–hole pairs generated after receiving the photon energy are dissociated from each other and form charge carriers with the assistance of a vertical electric field, resulting in the modulation of the electrical properties of the channel layer [55]. One groundbreaking optoelectronic skin was successfully fabricated on an ultrathin flexible system with a total thickness of 3a0~μm using polymer light-emitting diodes and organic photodetectors (Fig. 3e) [56]. The device not only functioned as a pulse oximeter, but also displayed visualized results. Similarly, Xu demonstrated an epidermal and flexible near-infrared (NIR) photoplethysmogram (PPG) sensor by integrating a high-sensitivity organic phototransistor with a high-efficiency inorganic light-emitting diode. The sensor achieved an NIR response as high as 3.5×10^5 AW^{-1} at a low operating voltage (<3 V) [57].

2.5. Sensors in integrated formats (multifunctional sensing)

Multifunctionalities are essential for the development of next-generation smart skins. Many studies have reported artificial skins with bimodal sensing achieved by utilizing the specific effects of materials, such as the piezoresistive, piezoelectric, or pyroelectric effects [58–61]. Bao et al. fabricated a field-effect transistor (FET) sensor by integrating pressure-sensitive gate dielectrics and

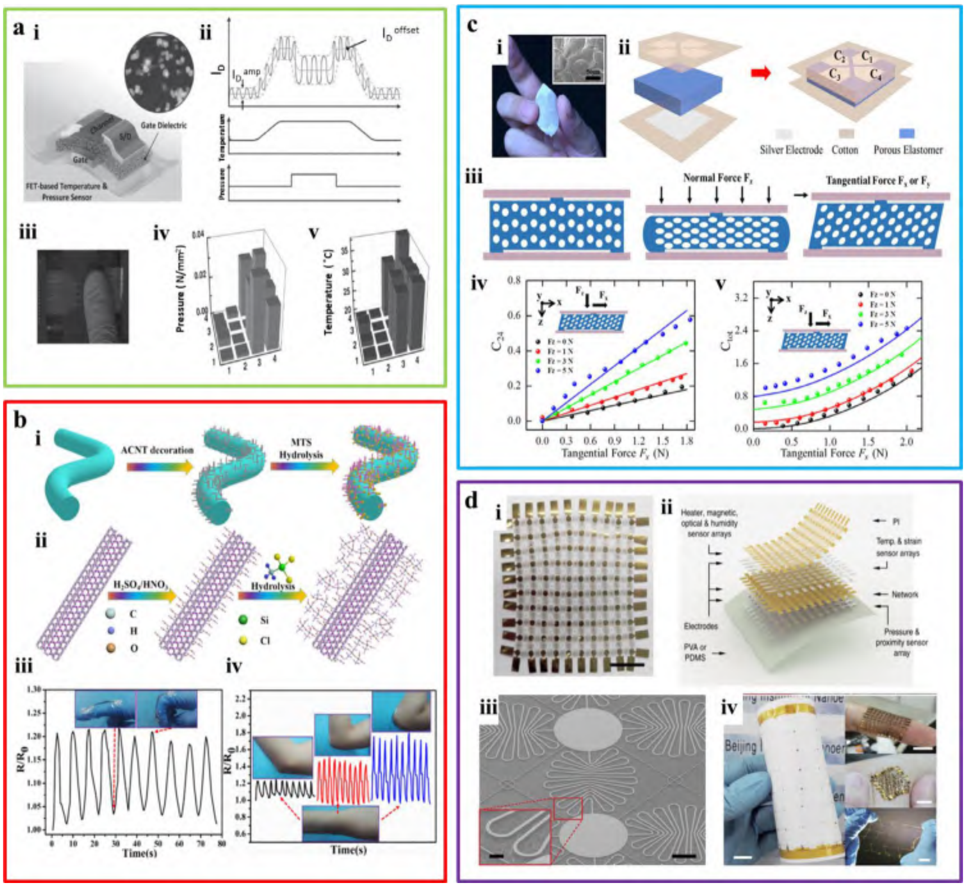

Fig. 4. (a) The structure of physically responsive field-effect transistor with the bottom-gated and top-contact structure, where the gate dielectric is comprised of P(VDF-TrFE) or nanocomposite of P(VDF-TrFE) and $BaTiO_3$ nanoparticles and the channel is organic semiconductor of pentacene; Changes in current signals of the FET with P(VDF-TrFE) upon applying pressure and temperature; Read-out pressure and temperature measurements of a sensor array with half of the sensor's devices are being depressed by a human thumb. Reproduced with permission from [62]. (b) Schematic illustration for the preparation of nanofiber composite and the methyltrichlorosilane hydrolysis on the methyltrichlorosilane surface and the application of detection the body motions. Reproduced with permission from [63]. (c) Photograph of a porous dielectric elastomer-based force (PDiF) sensor; Schematic illustration of the assembly and the structure of the PDiF sensor; Schematic deformations of the porous elastomer under a normal force or a tangential force in the cross-sectional view. The differential capacitance C_{24} and the total capacitance change C_{tot} of the sensor against tangential loads (F_x) under various normal forces (F_z). Reproduced with permission from [16]. (d) Optical image of the fabricated polyimide network along with the schematic layout of the integrated sensor array with eight functions. Reproduced with permission from [69].

a temperature-responsive semi-conductive channel layer based on piezoelectric and pyroelectric effects (Fig. 4a) [62]. Additionally, Gao et al. demonstrated a stretchable resistive sensor that is sensitive to both chemical toluene vapor and mechanical strain, with good selectivity and high sensitivity (Fig. 4b) [63]. Bai et al. presented a sensor composed of two parallel elastomeric lightguides that incorporated unique chromatic patterns. By combining frustrated total internal reflection and absorption, the sensor achieved multilocational distinguishment and multimodal mechanical deformation decoupling such as the modes of stretch, bend, and press. Their device allowed for reconfiguration of finger joint movements and external presses in real time [64]. Multifunctional detection can also be achieved by an adaptive electrode configuration in a single device [65–67]. Liu et al. reported a flexible capacitive sensor by configuring the electrode in four-parallel capacitors for the sensitive detection of spatial forces in arbitrary directions. The normal and tangential forces were detected by the average and differential capacitances, respectively (Fig. 4c) [16]. Moreover, the integration of more than two functions in a single device has been deployed in recent years, such as sensing of touch, temperature, humidity, strain, compliance, and chemical vapor. (Fig. 4d) [68–70]. In addition to the intrinsic nature of soft and stretchable devices, the characteristics of self-healing and self-power are attractive features of multifunctional devices [71–73].

3. E-skin for sensing biochemical cues or exhibiting extraordinary functions

In addition to physical variable detection, electronic skins can also provide augmentative functions. A sensor can be used as a chemical cue for skin/dermal skin secretion. Beyond human skin, the extraordinary features of the chameleon or octopus skins, such as mimicry or self-healing inspire integration of biomimetic functions to the devices. Below, we will focus on smart electrical skins with the abilities of biochemical sensing of glucose and lactic acid, bioelectrical signal detection, adaptive changes in color/shape inspired by the chameleon, and self-healing inspired by amphibians.

3.1. Flexible electronics for sensing metabolites

The liquid mixtures secreted by sweat glands on mammalian skin contain various biochemical molecules, such as glucose, lactic acid, and electrolyte salts. Sensing glucose in sweat is particularly important [74,75]. Lee et al. reported a patch-type glucose sensor integrating the modules for blood glucose detection and drug release, based on the chemical vapor synthesis of graphene and on doping with a gold network structure for electrochemical enhancement (Fig. 5a-i) [76]. The patch sensor can measure pH, temperature, humidity, and strain to carefully calibrate any effects

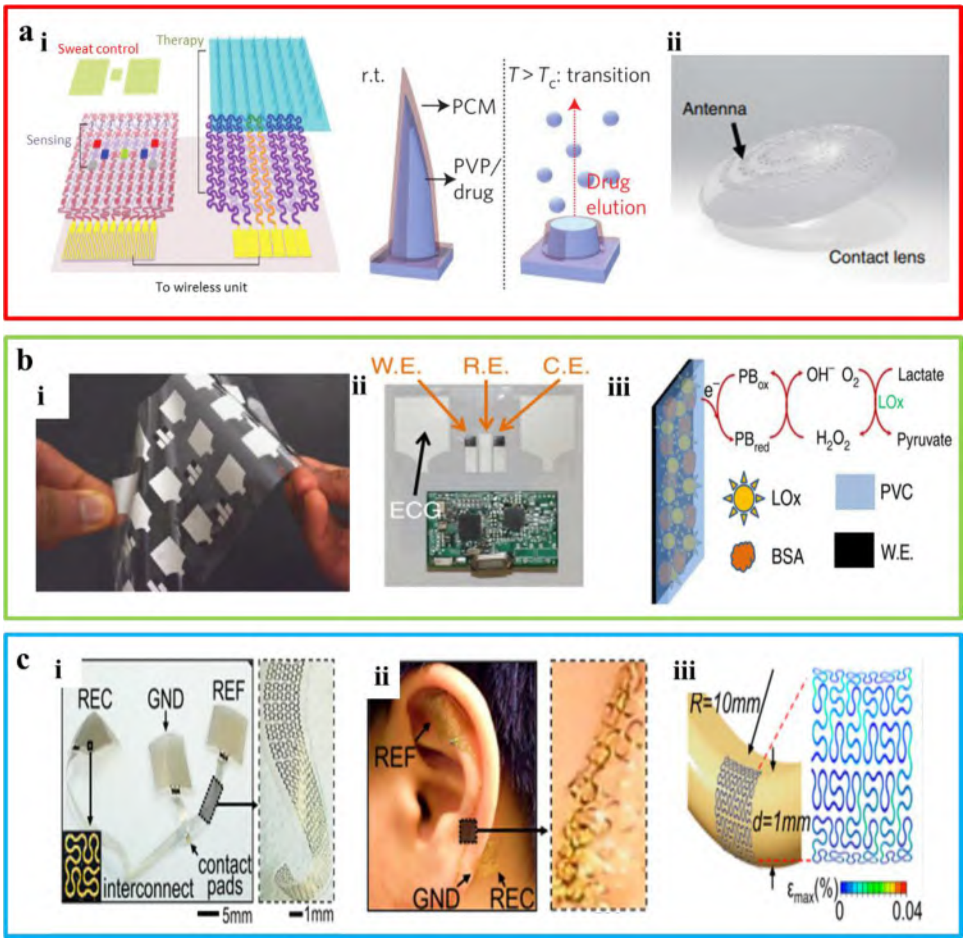

Fig. 5. (a) Schematic diagram of the integrated smart patch for blood glucose detection and treatment. Schematic diagram of a wireless blood glucose contact lens sensor based on tear detection. Reproduced with permission from [76,77]. (b) An array of printed flexible patches; Image of a patch along with the wireless electronics; Schematic showing the lactate biosensor along with the enzymatic and detection reactions. Reproduced with permission from [85]. (c) Epidermal electronics with fractal layouts, composed of three electrodes (REC, GND, and REF) and interconnect (left), with magnified view of the latter (right). Fractal device architectures and mechanical properties of EEG measurement systems; Device laminated on the auricle and mastoid (left) and the magnified interconnect (right); Simulation results for simultaneous bending along two orthogonal. Reproduced with permission from [86].

on glucose detection. In the scenario of sensing high glucose content in sweat, the second module triggers the release of the multi-channel drug via microneedles. They demonstrated accurate measurement of the blood glucose concentration in mouse models and “on-demand” release of drugs for treatment.

In addition, tear fluid is attractive owing to its enriched biological information composed of proteins, lactic acid, and glucose. Kim et al. reported a non-contact wireless blood glucose detection contact lens based on graphene and nano-silver materials prepared using the principle of LC oscillation circuits (Fig. 5a-ii) [77]. The spiral electrode was made of a hybrid structure of graphene and nano-silver, which allowed for high optical transmittance. Using polyethylene terephthalate and polydimethylsiloxane (PDMS) as flexible substrates, the sensor can endure bending with different radii of curvature. The glucose in the tear can be oxidized to gluconic acid by the enzyme of glucose oxidase deposited on the substrate, while generating hydrogen peroxide to take part in the subsequent redox reaction for electrochemical signals. The antenna coil captured tiny changes in the electrical current and transmitted them to the reader coil for the detection of glucose concentration.

Under conditions of insufficient oxygen, glucose produces lactic acid by an anaerobic reaction. Lactic acid is an important indicator of human health [78]. Payne et al. reported a flexible device for sensing lactic acid based on tetrathiafulvalene and carbon nanomaterials [79]. In addition, the pH changes induced by various physiological activities can be monitored using new types of E-skin, including a sweat sticker sensor using pH-sensitive phase-changing polymers (polyaniline, PANI) [80], or an ion field effect transistor (ISFET) flexible pH sensor [81].

3.2. Flexible electronics for ECG/EEG/EMG sensing

In addition to chemical species sensing, artificial skin devices have been used for the detection of bioelectrical signals, such as electrocardiography (ECG) [82], electroencephalography (EEG) [83] and electromyography (EMG) [84]. Because ECG signals usually have large electrical amplitudes, detection is easier than with other types of signals. A typical configuration for ECG measurement includes bipolar high-impedance electrodes with a layer of conductive hydrogel adhesives attached to surface locations on the human body, such as the intercostal space of the chest (Fig. 5b) [85]. In conventional EEG/EMG signal measurements, rigid electrodes are combined with electrolyte gels, where the uncomfortable contact and low signal-to-noise ratio (SNR) are the main constraints for wearable and long-term signal recording. Norton et al. proposed a foldable neural electrode platform with a thickness of 5 µm for long-term, high-fidelity EEG recording of signals by using the outer ear and adjacent regions as mounting locations (Fig. 5c) [86]. Han et al. demonstrated that electrodes could be configured as electrode-electrolyte gels or as single conductive gels

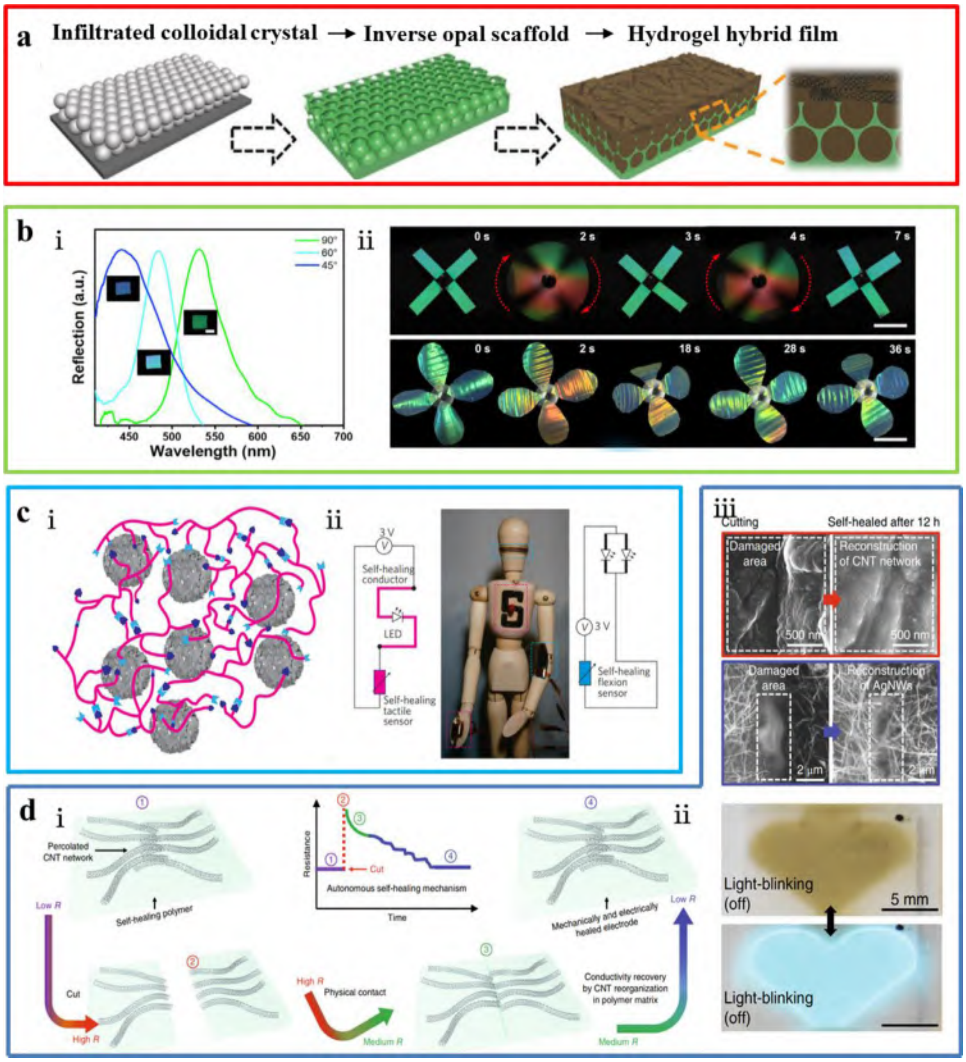

Fig. 6. (a) Illustration of fabrication of polymer-conductive hydrogel hybrid structural color films. Reproduced with permission from [89]. (b) The Bragg diffraction red-shifts by altering the glancing angle. Insets show the corresponding images of inverse opal PTMPTA films; The non-patterned structural colored pinwheel keeps rotating with a color change under intermittent nitrogen-acetone mixed flow (top images), and the patterned structural colored flower blooms and closes with a color change under exposure to acetone flow (bottom images). Reproduced with permission from [90]. (c) Proposed interaction of oligomer chains with μNi particles; Self-healing flexion and tactile sensor circuit schematic and mounting on a fully articulated wooden mannequin. Reproduced with permission from [110]. (d) Proposed recovery mechanism for CNTs embedded in self-healing polymer matrix; Self-healable heart-shaped pixel that blinks when a heartbeat is detected; SEM images of self-healable composite electrodes before and after self-healing. Reproduced with permission from [113].

[87]. The authors structured a conductive, self-adhesive, stretchable hydrogel by partially converting graphene oxide (GO) to conductive graphene through polydopamine reduction. Owing to its excellent conductivity and good adhesion to human skin, the performance of the hydrogel was highly competitive with that of commercially available electrodes.

3.3. Camouflaging and mimicry with flexible electronics

Recent studies have revealed the unexpected potential of the skin for sensing light. Skin cells can be transformed into photosensitive nerve cells [88]. The extraordinary functions of some creatures, such as mussels, chameleons, and octopus, have inspired researchers to develop new types of biomimetic flexible electronics. Zhao et al. combined carbon nanotube-embedded polydopamine (PDA) and elastic polyurethane (PU) to fabricate flexible electronics with a color-tuning ability (Fig. 6a) [89]. Their devices were designed to mimic the color shift mechanism of chameleon skin, featuring photonic responses from a lattice of small non close packed guanine nanocrystals on the surface. Using a silica-templated method, they hybridized the PU hydrogel film with the ordered inverse opal nanostructures as a scaffold of brilliant structure color. Any structural change of the scaffold by the force would produce a different pattern of light reflection according to the Bragg's Law. They demonstrated reversible color changes and potential motion-monitoring applications by responding to the stretching process with an interactive dual-signaling mechanism. In addition, Du et al. reported a design (Fig. 6b) integrating both color and shape changes driven by the solvent vapor swelling process [90]. The device was made of inverse opals with high porosity and periodicity and a vapor-responsive polymer stent (polytrimethylolpropane triacrylate, PTMPTA). The light reflection of the inverse opal/ PTMPTA films can be exaggerated by the fast vapor-absorbing/desorbing property of PTMPTA. Therefore, their devices were able to perform quick responses, including color changes or programmed shape transformations triggered by the external vapor. Alternatively, Bao et al. developed a chameleon-inspired stretchable sensor with interactive color-changing and stretching-sensing properties by combining a stretchable resistive pressure sensor and an organic electrochromic device [91]. A layer of pyramidal-microstructured PDMS coated with single-wall carbon nanotubes (SWNTs) was fabricated as the highly tunable resistive pressure sensing unit. Simultaneously they integrated another layer of poly(3-hexylthiophene-2,5,diyl) with the electrochromic property as a pigment cell for the demonstration of pressure-initiated color changes. Park et al. presented a chameleon-like multi-layered pressure sensor with dual signaling of pressure sensing and electroluminescent sensing [92]. Their device was constructured on a two-terminal capacitor with six constituent layers in the typical order of electroluminescent display. Light emission was modulated by the capacitance change of the insulator under the pressure, thus allowing for direct visualization of the static and dynamic information of the position, shape, and size of pressurizing object.

New flexible electronic devices have emerged, inspired by the skins of coleoid cephalopods such as squid, octopuses, and cuttlefish [93,94]. Xu et al. developed a bionic system capable of changing the reflection of infrared light in response to external stimuli, either broadband or narrowband [95]. An aluminum (Al) metal layer or alternating layers of titanium dioxide (TiO_2) and silicon dioxide (SiO_2) were deposited on photoconducting electrodes containing pre-coated acrylate membranes using electron-beam evaporation. The device was able to modulate the infrared light reflection profiles in response to external actuation, including mechanical actuation and electrical actuation. A similar strategy was employed to construct flexible devices based on thermoregulatory materials [96].

3.4. Haptic-feedback bionic electronics

Smart E-skin involves all aspects of tactile properties, ranging from receptors to neural coding to sensory feedback [97]. Importantly, sensory feedback is crucial to reflect the touch of sense in the interactive events in the real word in order to make immediate adjustments. There have been exciting progresses in the development of smart E-skin with the fusion of sensing and feedback functions. Mechanical simulators, including vibration motors, microfluidic/pneumatic or wired actuators, are widely used for the integration of haptic feedbacks [98–100]. Yu et al. reported battery-free electronic systems with haptic interfaces by incorporating arrays of millimeter-scale vibratory actuators in a soft and conformal elastomer. The system can be laminated onto skin and reprogrammed via a remote computer system, thus allowing for establishment of an epidermal virtual reality experience [98]. Zhu et al. proposed a haptic-feedback smart glove integrating triboelectric-based finger bending sensors, palm sliding sensor, and piezoelectric mechanical stimulators. It can detect the finger motions with multiple degrees of freedoms, or sense the normal and shear forces in eight directions. The smart glove amplified the sensation of the impact in real time via piezoelectric haptic stimulation [101]. Sensory feedback-based smart E-skin is compelling in the future development of bionic system, as a critical module to provide users with more interactive experience.

3.5. Self-healing flexible electronics

The biological tissues of many creatures, such as zebrafish, newt, octopus, and others, exhibit a remarkable property of autonomous healing from wounds to avoid further damage [102–104]. Inspired by their mechanisms, smart electronic materials have been proposed that incorporate novel chemistry to mimic self-healing performance [105–107]. Self-healing materials can be classified into two types based on the self-healing mechanism used: extrinsic or intrinsic. Extrinsic self-healing relies on the bond-forming or reactions of the pre-distributed reactive reagents in the mixture, while intrinsic self-healing is dominantly contributed by the reorganization of the material itself [108,109]. The molecular interactions for self-healing include covalent bonds, noncovalent bonds or intermolecular forces, such as hydrogen bond, ionic bond, dipole−dipole, and van der Waal's interactions. Reversible covalent interactions are generally relied on external stimuli (e.g. heat or pH changes) to trigger the reactions. For instance, Diels−Alder (DA) reaction for self-healing typically requires thermally reversible DA cycloaddition to highly crosslinked furan−maleimide polymers. In contrast, self-healing materials involving noncovalent reactions are not heavily dependent on external stimuli. For example, the self-healing process of supramolecular polymers is dominant by the reversible host−guest interactions without the external triggers. However, those self-healing materials independent on the external stimuli usually exhibit lower recovery rates than the others which request external triggers.

Tee1 et al. developed a mixture of micro-nickel (μNi) microparticles and a self-healing polymeric material as the major components in their self-healing devices (Fig. 6c) [110]. When the polymeric networking of the material was damaged by mechanical forces, the dynamical hydrogen bonds of the supramolecular structures could facilitate networking reconstruction at room temperature, thus enabling a self-healing process at the fracture surface. They demonstrated dual-sensing of external pressure stimuli and flexion, due to the re-arrangement of the μNi microparticles in the mixture induced by the external mechanical loads, with potential applications in sensing body movements such as hand shaking and arm bending. Carbon particles or graphene also serve as good candidates for construction of self-healable devices [111,112]. Conductive networks incorporating self-healing polymers can significantly enhance electromechanical recovery (Fig. 6d) [113]. For instance, patterned silver nanowires embedded in a self-healing polymer can undergo dynamic rearrangement, thus allowing for electrical conduction recovery after application of external mechanical loads. Ionic conductors have been used to build functional components in self-healing flexible electronics [114–116]. Cao et al. mixed a stretchable polymer with an ionic liquid to form crosslinks via ion-dipole interactions, which endowed the flexible sensor with an excellent self-healing property [117]. Their material, poly-(vinylidene fluoride-co-hexafluoropropylene) (PVDF-co-HFP), was featured with crossing-linking between the polar groups on the polymer and the ionic salt, providing the foundation of dynamic self-healing process. This strategy was extended to fabricate more flexible and self-healable sensors under various conditions [118].

4. Implantable flexible electronics – "smart skins"

In a general sense, "smart skins" would involve flexible electronics that operate inside the body, namely, implantable electronics. They would measure pressure, temperature, and electrophysiological signals and provide vital information for disease diagnosis and post-surgery monitoring. In this section, we discuss the constructive materials for implantable devices, followed by two categories of implants: active and passive devices. Details including the energy source and device architecture in device development are described.

4.1. Constructive materials and delivery methods for implantable sensors

Constructive materials for implantable flexible electronics must be highly biocompatible. Typical in vivo phenomena, such as protein corona attached to the device surface, platelet accumulation, or oxidative stress leading to cell damage, must be carefully investigated for the appropriate choice of constructive ma-

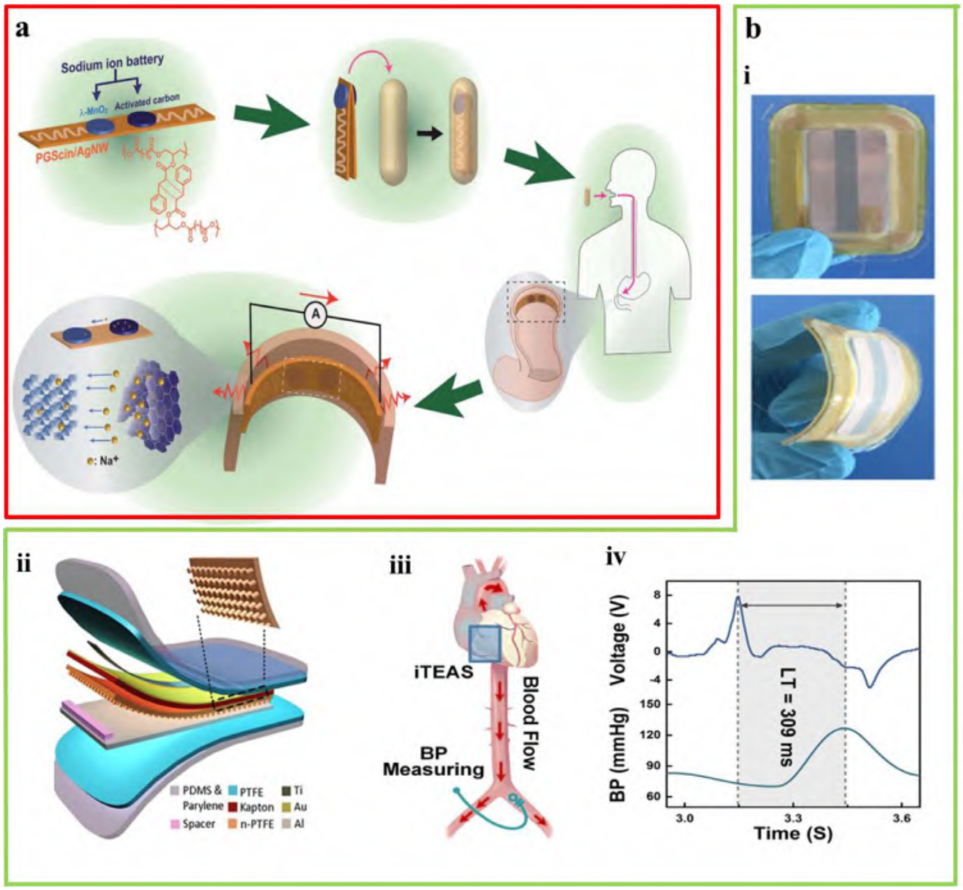

Fig. 7. (a) Envisioned deployment and operation of edible current sources. Reproduced with permission from [132]. (b) Photos of the implantable triboelectric sensor in original and bending states; Exploded-view illustration of the structure; Schematic diagram illustrating the mechanisms for monitoring the velocity of blood flow. Red arrows represent the direction of blood flow; Time interval (leading time, LT, shown in highlighted region) between the peaks of the two corresponding waveforms. Reproduced with permission from [138].

terials. Although many types of materials, including carbon nanotubes, graphene, liquid metals based on gallium/indium/bismuth elements, or conducting polymers, have been proposed to fabricate implantable electronics, their long-term effects in vivo are still largely unknown [119–121]. The biocompatibility of some materials may be improved by surface modification with well-studied substances, such as hyaluronic acid coated on polyetheretherketone (PEEK) devices [122]. The biodegradability of the materials also needs to be carefully balanced. In situ degradation of implantable devices can reduce the burden (or damage) of a second surgery to remove the device. The degradation rates are simultaneously influenced by the properties of the materials, ambient biofluids and trigger conditions [123]. For example, poly(lactic-co-glycolic) acid (PLGA) shows controllable biodegradable rates by tuning the lactic to glycolic mole percentage ratios in the process of copolymerization. The mass of PLGA (50/50, lactic/glycolic ratio) lost 20%, while PLGA (95/5) lost only 3% in 10 days in phosphate buffered saline (PBS) solution [124]. In addition, Rogers' group reported that temperature was effectively modulating the disintegration behaviors of the electronic devices. As the trigger temperature was reduced from 55 to 45 °C the failure of a wax – coated Mg resistors was prolonged by 8 min [125]. The stretchable sensors reported by Bao et al. integrated two biodegradable elastomers, poly(glycerol sebacate) (PGS) and poly(octamethylenemaleate (anhydride) citrate) (POMaC). They were tested in the demonstration of sensitive monitoring of pressure changes in orthopedic applications. The tensile modulus of the elastomer decreased at a rate of -11% per week according to their report [126].

Delivery methods are also critical for the application of implantable sensors. There are two main categories of implantation: orthotopic and remote. The former method attaches/adheres the sensors directly onto the target tissue site, which may require surgery. This method is difficult to apply to specific tissues, such as the inner wall of blood vessels or delicate organs. The latter method may address the limitations of the former in some cases. A typical example is the implantation of vascular stents [127]. The devices can be introduced from a relatively easier opening of a proximal vessel, and then guided along the vessel to reach distant target regions for final implantation.

4.2. Active implantable electronics

Active implantable devices typically require the effective management of energy sources (batteries or energy harvesters). As the device works in the body, the battery needs to be removed or dissolved in biofluids at its end of life. Bioresorbable batteries are popular choices for the development of transient implantable devices [128,129]. The design criteria for bioresorbable batteries include device dimensions, desorption rates, mechanical compliance, the attribute of complete transience, and the performance to meet device demands [130]. Examples include the utilization of water-activated battery technologies, in which the constituent materials physically disappear in a hydrographic environment [131]. Kim et al. introduced edible electronic power sources composed of flexible composite electrodes and a sodium ion electrochemical cell (Fig. 7a) [132]. Jia et al. reported a biodegradable battery

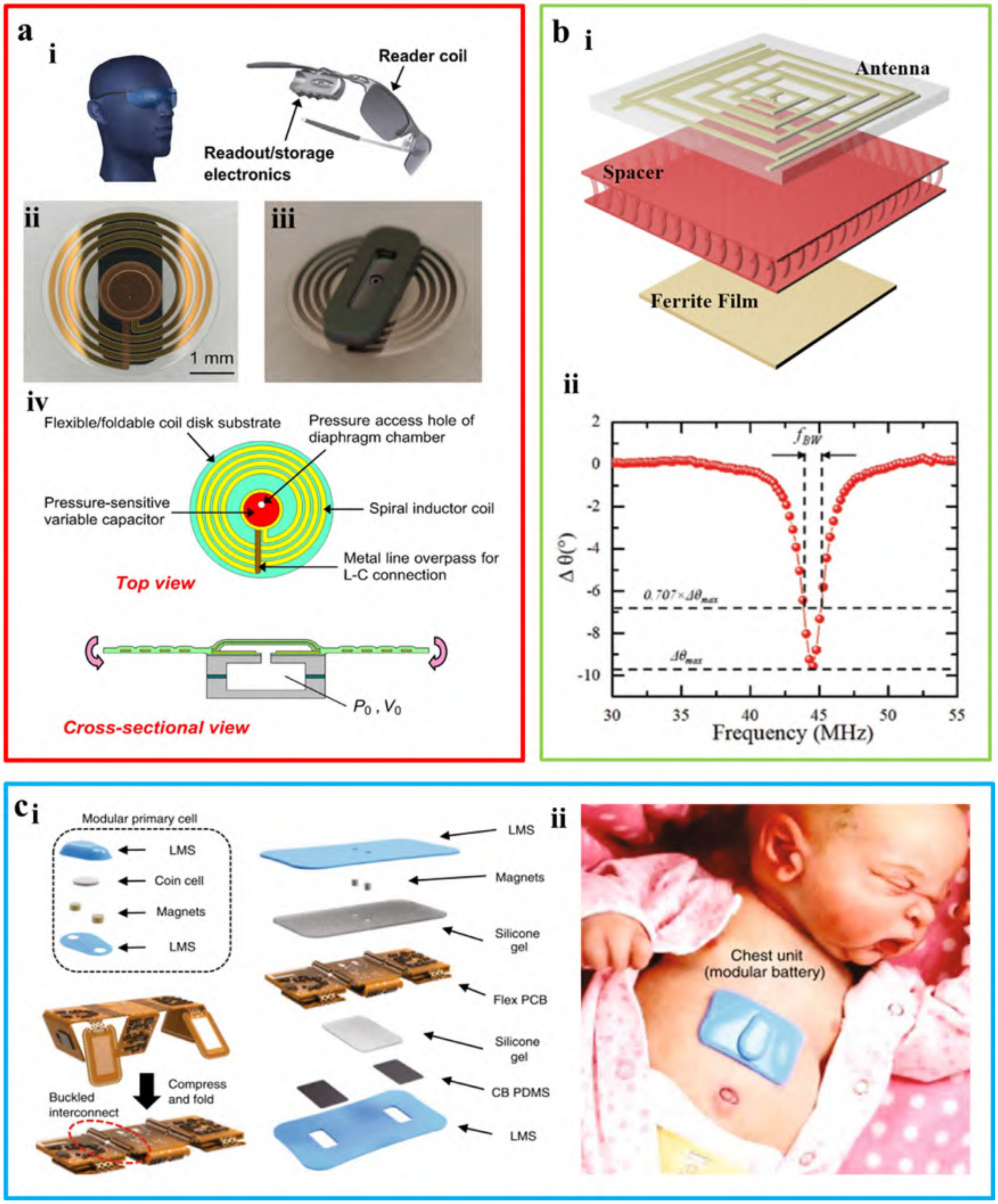

Fig. 8. (a) Proposed IOP monitoring method in practice; Full-scale photographs of the microfabricated flexible-coiled wireless pressure sensor with its (left) top and (right) bottom views before being packaged; Pressure sensor design schematics with flexible/foldable coil disk substrate. Reproduced with permission from [144]. (b) Schematic illustration of the 3D structure of a flexible wireless pressure sensor, consisting of a fabric spacer sandwiched between an antenna (top) and a piece of ferrite film (bottom); The phases changes of the read coil as a function of frequency when coupled to the pressure sensor. Reproduced with permission from [145]. (c) Schematic diagram and exploded-view illustration of a device with a modular primary battery; LMS, low-modulus silicone; PCB, printed circuit board; Photograph of the chest unit with a modular battery on a realistic model of a neonate. Reproduced with permission from [148].

by developing a polymer electrolyte (silk fibroin–choline nitrate) in a thin-film magnesium battery. The battery achieved a specific capacity of 0.06 mAh cm^{-2} and was completely degraded after 45 days in buffered protease solution [131]. In addition to bioresorbable batteries, energy harvesters are another appealing power supply strategy for medical implant devices [133–136]. These energy sources can scavenge the biomechanical energy generated by motions of internal organs, such as breath and cardiac motions. Zheng et al. reported a biodegradable triboelectric nanogenerator (BD-TENG) with a multilayer structure composed of a biodegradable encapsulation structure, friction layers, and electrode layers. The BD-TENG powered two inter-digit electrodes on which nerve cell growth was orientated, demonstrating the crucial impact on neural repair [137]. Implanted energy harvesters are promising energy supplies because they not only serve as energy sources but also as monitors, saving more space inside the body. Ma et al. reported a one-stop implantable triboelectric sensor to record heartbeat and breathing, showing a robust self-generated capability and exempting the necessity of an onboard battery (Fig. 7b) [138].

4.3. Passive implantable electronics

Passive implantable electronics do not have energy sources on their own, and they receive energy through external sources, such as radio frequency. The location and size of the implants inside the body, the data transmission distance, and the chemical environment of the surrounding tissue lead to the device architecture and communication mode [139]. One common architecture choice is an inductor-capacitor (LC) resonator, and the energy and data can be transmitted through electromagnetic coupling between the device inductor and an external coil [140–142]. The passive LC resonator has a unique resonator frequency, which is governed by the inductance and capacitance values. Any stimuli altering the inductance or capacitance will change the resonant frequency, which can be detected through the coupling coil. There are a few studies based on this working principle for invasively measuring intraocular pressure (IOP) sensors (Fig. 8a) [143, 144]. The capacitance was subject to the variation in the IOP, which could be reflected by the resonant frequency. Alternatively, Nie et al. proposed an LC-based pressure sensor array in which the pressure altered the inductance instead of the capacitance. By using a fabric spacer between the LC passive antennas and the ferrite film units, the resonant frequencies changed along the distance between the ferrite films and the LC antennas (Fig. 8b) [145]. In addition, the passive operating mode facilitates device miniaturization owing to its simple architecture. Bao et al. reported a passive, flexible, millimeter-scale sensor, scaled down to 1 × 1 × 0.1 cubic millimeters [128]. The LC tank was created by stacking a deformable dielectric layer between the two inductive spirals in a sandwich structure. However, the detection range of these passive wireless devices is typically on the order of several millimeters, making this technology less suitable for applications in tissues and organs deep within the body. Another architecture involves a radio-frequency antenna for data transmission and energy supply created by integrating an IC chip into a passive device [146,147]. This strategy has only been proven for on-skin applications, considering that the IC chip is not biocompatible and biodegradable (Fig. 8c) [148]. Another strategy for realizing passive implants depends on the surface acoustic wave (SAW) resonator on piezoelectric materials. Murphy et al. demonstrated the possibility of wirelessly monitoring left ventricle pressure using an SAW resonator and an antenna [149].

5. Challenges and prospects

The demand for next-generation bio/skin-inspired flexible electronics significantly improves its industrial performance. Currently, owing to the intrinsic hysteresis of polymeric materials, the flexible pressure sensors could suffer from low response time and large hysteresis error in loading and unloading process. This drawback might be partly addressed by incorporating micro/nanostructures in the active polymeric materials, such as porous elastomers [150,151] and silicon nanowires [152], yet *de novo* design of new materials and device structures is required. As another example, the dielectric materials of most flexible capacitive pressure sensors suffer from a relatively low dielectric constant ε, despite various efforts such as incorporating organic dipole molecules, adding high-permittivity ceramic fillers, and introducing conductive nanofillers. At present, the miniaturization of smart flexible electronics is still technically challenging. The typical size of flexible pressure sensors is at least one order of magnitude larger than that of silicon-based pressure sensors used in microelectromechanical systems (MEMS) (as small as hundreds of micrometers), which limits large-scale integration and mass production. In the future, more efforts in the miniaturization of flexible electronics are required to introduce novel device designs and high-level integration for smart E-skin.

For the development of multifunctional devices, researchers have demonstrated the feasibility of integrating two or more sensing modalities into a single device. However, the quantitative and precise measurement of an arbitrary input stimulus remains a critical problem. Typically, the electrical responses of these multifunctional sensing devices to known individual input stimuli can be well calibrated, but it remains a great challenge to decouple the convolved effect induced by simultaneous multiple input stimuli. Researchers have attempted to integrate commercial chips with different functions on flexible substrates using stretchable interconnection. However, it would require complicated fabrication and leave unaddressed the mechanical mismatch between the flexible connecter and the rigid chips. To solve the complicated input-output relationship, there is still a great need to develop smart decoupling methods, for example, a modified linear regression approach to decouple contact forces in spatially arbitrary directions [16]. Advanced signal analysis technologies, such as machine learning or deep learning, in concert with careful calibration of multiple sensing modalities, can bring new insights to this research direction in the future.

The biological safety issues of materials and devices should be highlighted, especially for implantable application scenarios. Nanomaterials, such as graphene, carbon tubes, and various inorganic/organic nanoparticles, have been demonstrated to be quite inert to the human body in the short term. However, the long-term effects of these materials *in vivo* or their detailed biodegradability actions are still largely unknown, which demands systematic investigation. Previous research indicated that the physicochemical parameters of high-aspect-ratio fibrous nanomaterials are critical in affecting lung diseases [153]. Long, rigid multiwall carbon nanotubes could possibly be carcinogenic for humans. In the future, the development of materials with significantly improved biosafety or controllable degradation rates is an attractive research area. In the special case of wireless data/power communication involving implantable devices, greatly increasing the communication distance is technically challenging owing to the limitation on the antenna size. For this application, innovative antenna designs based on biocompatible materials would become indispensable. To validate the bio-safety of implantable devices *in vivo*, standardized protocols should be established with care for full exploration.

Self-healing materials for the construction of flexible electronic devices are progressing rapidly. However, most of the materials in the literature can heal damage only on a very limited scale. New solutions are needed to screen robust materials based on novel self-healing mechanisms. In many cases, external stimuli such as heating, light exposure, and pH changes are required to trigger the self-healing process. Development of self-healing materials focusing on individually specific scenarios could bring new ideas. For example, diving equipment is soaked with a high concentration of salts in seawater; thus, self-healing flexible electronics for diving can be designed to take advantage of those ionic triggers. In contrast, next-generation E-Skin may require the development of new self-healing materials without the assistance of external stimulating factors.

There are several additional research directions that may highlight the future development of bio-inspired smart flexible electronics: 1) Development of materials with tunable mechanical properties depending on the external load rates, as inspired by the natural skin tissue structure of tendons and ligaments [2]. 2) The interfacing of smart E-skin with natural nerves to construct hybrid bioelectronics for neurorobotics and neuroprosthesis [154,155]. 3) Development of artificial motor systems integrated into E-skin [98]. Research efforts of addressing the critical issues discussed here promise new chances for next-generation smart E-skin.

Declaration of Competing Interest

There is no competing interest to declare.

Acknowledgments

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