Skin-Like Electronics for Perception and Interaction: Materials, Structural Designs, and Applications

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Recent advances in the material and structural design of skin-like electronics have enhanced the interactions between the virtual and physical worlds and between people and objects. Herein, the existing flexible sensing materials and microstructures, including flexible stimuli-responsive materials and response- and stretchability-enhanced microstructures, are reviewed. Five typical skin-like electronics with sensitivities analogous to the human senses (olfactory, visual, auditory, tactile, and gustatory senses) and their corresponding applications (gas, light, chemical composition, sound, and mechanical signal monitoring) are introduced. Finally, it is concluded with some perspectives on challenges and opportunities for future research in flexible hybrid electronics.

1. Introduction

The realization of perceptions and interactions is the physical basis of the communication between the virtual world and the physical world of humans and objects. Traditional hard electronics cannot be conformably integrated on the human body or soft robots, which limits the interface between the virtual and physical worlds. The substantial progress of skin-like electronics[1–5] has been achieved to enrich this interface,[6,7] increasing the commercial potential of human–machine interactions,[8–12] biomimetic robots,[13–15] implantable medical instruments,[16] and digital healthcare.[17–21]

2. Flexible Sensing Materials and Microstructures

2.1. Flexible Stimuli-Responsive Materials

Based on different mechanisms to achieve flexibility, skin-like electronics are divided into organic and inorganic flexible electronics. The organic flexible electronics[22–24] directly use flexible organic materials, such as organic semiconductors and conductive polymers, to realize sensing units and functional circuits. For the inorganic flexible electronics,[25,26] all functional components are dispersed and reside on rigid islands, which are linked by stretchable/deformable interconnects. Then, such systems with islands and interconnects integrated on or encapsulated into substrates achieve flexibility and stretchability. However, both organic and inorganic flexible electronics have disadvantages. For example, organic flexible electronics are constrained by the development of materials while flexible stimuli-responsive materials have not yet reached a high sensing sensitivity. As for inorganic flexible electronics, they need a complex preparation process due to their complex structures, which lead to low reliability.

In this study, we review the existing skin-like electronics used for the achievement of perception and interaction, including their materials, designs, and applications. In Section 2.1, we present six kinds of flexible stimuli-responsive materials (photo-, thermo-, humidity-, chemo-, mechano-, and magneto-responsive materials). In Section 2.2 and 2.3, we outline the development direction of the microstructure designs used to enhance the response sensitivity (for crack, porous, wave, and fluff microstructures) and stretchability (wrinkle, buckle, serpentine, network, helical, Kirigami, and Origami microstructures) of stimuli-responsive materials. Furthermore, we introduce five typical skin-like electronics with sensitivities analogous to the human senses (olfactory, visual, auditory, tactile, and gustatory) based on flexible stimuli-responsive materials and microstructures in Section 3 and their unique applications in Section 4. In Section 5, we summarize the challenges and prospects for further study on flexible stimuli-responsive materials and microstructures. Figure 1 shows the outline of this review.

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transmit through various stimuli-responsive materials for measurement and transmission. Inspired from the unprecedentedly rapid progress of flexible electronic imperative, extensive research on stimulus-responsive materials are springing up, which have become the basis for broadening the application and fulfilling further demands of the stimulus-responsive materials. Flexible stimuli-responsive materials can respond to stimuli from the external environment (light, temperature, humidity, chemical reagents, mechanical forces, and magnetic fields) due to the changes in their microstructures and states by modifying their chemical and physical properties.

2.1.1. Photo-Responsive Materials

Light is commonly used as a stimulus in adaptive materials because it is nondestructive and allows for rapid response and easy signal reading\[^{30-34}\](either through deformation or generation of electrical signals). However, when using light as a stimulus, materials have to undergo reversible photochemical reactions. Troyano et al.\[^{30}\] reported that a 2D-to-3D shape transformation of an autonomous soft mechanical device based on a metal–organic framework could be reversed by light-induced desorption. The reported strategy enables the application of flexible photo-responsive autonomous devices in robotics and auto-systems (top and bottom panels for light off and on, respectively, in Figure 2a). Han et al.\[^{33}\] reported an artificial muscle fabricated by a bilayer structure of poly (methyl methacrylate) and graphene oxide compounded with gold nanorods. Upon light irradiation, the patterned 2D film could be deformed into 3D structures within only 0.05–0.6 s. This advanced complex material provides an important reference for developing light-responsive sensors and systems.

Figure 1. Outline of the review.
2.1.2. Thermo-Responsive Materials

Thermal stimuli are the most accessible and widely used in responsive materials.\(^{35,41-47}\) Luo and Mather\(^{35}\) developed triple shape-memory polymers with much more controllable and complex shape-morphing ability. It is worth mentioning that the temporary shape shown in the middle panel of Figure 2b definitely altered the recovery pathway from the temporary shape in the top panel of Figure 2b to the permanent shape in the bottom panel of Figure 2b, in a discontinuous fashion. Intermediate shapes can only be found along the same recovery pathway. Reeder et al.\(^{41}\) reported a memory polymer with a novel shape as a substrate for organic thin field-effect transistors for implantable electronics that can withstand the natural motion of the surrounding soft materials.

![Figure 2. Flexible stimuli-responsive materials. a) Flexible photo-responsive materials. Reproduced with permission.\(^{30}\) Copyright 2019, Wiley-VCH. b) Flexible thermo-responsive materials. Reproduced with permission.\(^{35}\) Copyright 2010, Wiley-VCH. c) Flexible humidity-responsive materials. Reproduced with permission.\(^{36}\) Copyright 2018, Wiley-VCH. d) Flexible chemo-responsive materials. Reproduced with permission.\(^{37}\) Copyright 2018, American Chemical Society. e) Flexible mechanical-responsive materials. Reproduced with permission.\(^{38}\) Copyright 2019, Royal Society of Chemistry. f) Flexible magneto-responsive materials. Reproduced with permission.\(^{39}\) Copyright 2019, Wiley-VCH. g) Flexible multiple-responsive materials. Reproduced with permission.\(^{40}\) Copyright 2016, Wiley-VCH.](image-url)
tissue. Under external heating, the shape memory polymer substrate can reshape into the appropriate 3D shape for implanting into the tissue. The shape memory polymer substrate can not only address the challenging biomedical problems but also broaden the application of flexible thermo-responsive materials. Flexible thermo-responsive materials can be used as simple and flexible sensors but with slow flow responsiveness.

2.1.3. Humidity-Responsive Materials

Flexible humidity-responsive materials can undergo deformations and electrical and color changes upon variations in humidity with the merit of simplicity and feasibility, but at the cost of responsive speed.\textsuperscript{13,14,19-21} Shen et al.\textsuperscript{13} reported a humidity-responsive material that can produce electrical signals upon moisture variations, as shown in Figure 2c. As shown in the bottom panel of Figure 2c, the magnitude of the generated voltage has a quasi-ideal linear relationship with the relative humidity level. These results reveal the possibility of developing self-powered flexible humidity sensors without an external power supply.

2.1.4. Chemo-Responsive Materials

Flexible chemo-responsive materials are useful for monitoring human health by analyzing a person’s secrets. Flexible chemo-responsive materials work through state changes or morphology transformations induced by chemical reagents induced molecules. They have high specificity and selectivity but slow responsiveness.\textsuperscript{17,50-52} Lee et al.\textsuperscript{17} developed a bio-inspired phage nanostructure-based color-sensor array by constructing structural color matrices that are composed of liquid-crystalline bundled nanofibers from self-assembled phages. As shown in Figure 2d, the ability to detect the rapid color changes upon exposure to various target chemicals (benzene, toluene, xylene, and aniline) is implemented by designed phages to express cross-responsive receptors on their major coat protein.

| Converted signals | Response time | Advantage | Disadvantage | Application |
|-------------------|---------------|-----------|--------------|-------------|
| Photo-responsive | Deformation,\textsuperscript{10-13} Electrical signal\textsuperscript{34} | $\approx$0.1–1 s, $\approx$1 s–100 ms | Nondestructive, Rapid responsiveness | Limitation supply of light and photo-chromatic energy |
| Thermo-responsive | Deformation,\textsuperscript{35,41-45} Electrical signal\textsuperscript{34} Optical signal\textsuperscript{47} | $\approx$1 s–1 min, $\approx$<100 ms, $\approx$<100 ns | Simple, Rapid responsiveness | Slow responsiveness, Temperature |
| Humidity-responsive | Electrical signal\textsuperscript{16,48} Deformation\textsuperscript{49} | $\approx$1 s–1 min, $\approx$10 s | Simple, Feasible | Slow responsiveness, Skin moisture, Air humidity |
| Chemo-responsive | Optical signal\textsuperscript{17} Electrical signal\textsuperscript{35,51} Deformation\textsuperscript{52} | $\approx$100 ms, $\approx$10 s, $\approx$1 min–10 h | Simple, Selectivity | Slow responsiveness, Body fluid components, Toxic Chemicals |
| Mechanical-responsive | Electrical signal\textsuperscript{39,53} Light\textsuperscript{61} Optical signal\textsuperscript{58,67} | $\approx$1 min–100 ms, $\approx$1 ms, $\approx$<1 s | Rapid responsiveness | Requirements for high durability, Limitation source for magnetic materials, Low transparency |
| Magnet-responsive | Deformation\textsuperscript{19} Electrical signal\textsuperscript{35,60} | $\approx$<1 s | Nondestructive, Rapid responsiveness | Magnetic field |

2.1.5. Mechanical-Responsive Materials

Flexible mechanical-responsive materials with versatile responsiveness and high-end precision detect various regimes of pressure or strain at different positions on the human body during physical activities. Mechanical-responsive materials with rapid responsiveness have been reported.\textsuperscript{38,53-58} Han et al.\textsuperscript{38} innovatively developed a versatile 3D carbon nanofiber networks (CNFNs) by a simple and cost-efficient strategy with superior pressure-sensitivity through electro-spinning and thermal treatment (top panel for sample, and bottom panel for its relative resistance change ratios versus pressure, Figure 2e). These CNFNs showed a pressure sensitivity of up to 1.41 kPa$^{-1}$, which is the largest among similar 3D porous materials. Moreover, they exhibit excellent flexibility, stable resilience, and super compressibility (>95%).

2.1.6. Magneto-Responsive Materials

Flexible magneto-responsive materials offer a noninvasive monitoring approach via the magneto-mechanical stimulation of materials, compensating for the low resilience of the materials to light, heat, pressure, humidity, and chemical stimuli.\textsuperscript{39,59,60} Ahmed et al.\textsuperscript{39} has engineered a soft magnetic shape-memory composite obtained by dispersing liquid droplets of magneto-rheological fluid into a polydimethylsiloxane (PDMS) matrix. This composite showed anti-thermal behavior, fast shape-memory, and almost 30-fold stiffening. As shown in Figure 2f, the shear strain evolution upon magnetic stimulation experiences four stages. The red curve shows the material recovery process in the absence of a magnetic field, whereas the blue curve shows that the material retains the deformation in the presence of a magnetic field. However, this material is particularly susceptible to light, heat, pressure, humidity, and chemicals, which limits its application in biomedical and wearable devices.

Table 1 presents a list of converted signals, response time, advantages, disadvantages, and potential application of recently developed stimuli-responsive materials.
materials can be incorporated into various sensors and attached or laminated to different positions of the human body to record the corresponding electrophysiological activity and physiological responses and provide information on the health condition.

Multi-mode and multi-integrated flexible stimuli-responsive devices, as well as human–machine interfaces and the flexible multiple-responsive materials\(^\text{[60,62,63]}\) are highly demanded for monitoring personal health conditions. Ho et al.\(^\text{[40]}\) successfully fabricated a sensor array, in which each unit has humidity sensitivity (left panel of Figure 2g), temperature sensitivity (middle panel of Figure 2g), and pressure sensitivity (right panel of Figure 2g), hence realizing a humidity, temperature, and pressure triple sensing function. Importantly, each sensor unit is independent of the others.

Because traditional stimuli-responsive materials are not flexible, sensors made by these materials cannot be conformally integrated with the human body, which reduces human comfort and makes it difficult to capture the human body’s weak physiological parameters. The flexible stimuli-responsive materials enable traditional bio-sensors to be flexible and stretchable without degrading sensitivity, and therefore have great application prospects in the preparation of bio-electronics.

2.2. Response-Enhanced Microstructures

The stimuli-responsive material of a sensor needs to be chosen depending on the application. However, even when using the same composition and chemical bonds, the sensing materials may have different properties. The structural design with specific processing technology corresponds to a unique function.\(^\text{[64]}\) Therefore, the structural design is an effective approach for skin-like electronics to enhance the response of materials.\(^\text{[65]}\)

2.2.1. Crack Structure

Spiders can efficiently detect signals around them through crack-shaped slit organs.\(^\text{[66]}\) The slits in their organ induce a large strain under small external stimuli, which results in ultrasensitive displacement detection.\(^\text{[67]}\) Inspired by the spider’s slit organ, the crack-based sensors have been fabricated and exhibited high gauge factors.\(^\text{[68–70]}\) Figure 3a shows an ultrasensitive crack-based strain sensor fabricated by Kang et al.\(^\text{[68]}\) The platinum layer was deposited on a flexible substrate with a thickness of 20 nm, and then the crack was generated by bending.\(^\text{[68]}\) Sensors based on nanoscale crack junctions have shown ultrahigh sensitivity with a gauge factor of over 2000 in the 0–2% strain range. Compared to sensors with no cracks, the resistance change in the specimens with cracks was 450 times higher at a 0.5% strain (Figure 3a, inset). The ultra-high sensitivity was attributed to the disconnection–reconnection process undergone by the zig-like nanoscale crack junctions under strain. Furthermore, the tunneling effect of the carrier transport has been shown to lead to very high sensitivity.\(^\text{[64]}\)

2.2.2. Porous Structure

The change in the resistance of strain/stress sensors depends on the deformability of the material used, which is related to its Young’s modulus. Porous structures can be used to decrease the Young’s modulus, hence increasing the sensitivity of the materials.\(^\text{[9,74–78]}\) For example, reverse micelles have been used to introduce pores into conventional pressure-sensitive rubbers (left panel of Figure 3b).\(^\text{[9]}\) The morphology, pore characteristics, and system modulus of pressure-sensitive rubbers can be readily controlled, and the change in the resistance of the porous pressure-sensitive rubbers depends on the applied tensile strain. Moreover, the modulus of the porous pressure-sensitive rubbers is low due to the pores. The porous pressure-sensitive rubber sensors have good sensitivity over a wide range of induced strains and exhibit a resistance two times higher than that of sensors without pores (right panel of Figure 3b). In addition, conductive polymers containing hollow spheres,\(^\text{[74]}\) porous carbon nanotube sponges,\(^\text{[75]}\) metal–polymer hybrid nanocable sponges,\(^\text{[76]}\) porous carbon nanotube–elastomer hybrid nanocomposite,\(^\text{[77]}\) and gold–film–polyurethane sponges\(^\text{[78]}\) have shown enhanced sensitivity to mechanical deformations.

2.2.3. Wave Structure

Compared with unstructured materials, microstructured flexible materials show higher sensitivity and faster response time.\(^\text{[40]}\) The left panel of Figure 3c\(^\text{[71]}\) shows a capacitive sensor with wrinkles; the capacitance of this sensor changes with the distance of each ridge (closer) or valley (further) from the sensor. Thus, the pressure can be measured by the voltage output signal of the sensor. For pressures below 1.0 kPa, the sensitivities of the non-wrinkled and wrinkled sensors are 4.8 × 10⁻⁶ and 0.013 kPa⁻¹, respectively (right panel of Figure 3c). Moreover, the contact surface area of the highly wrinkled surface changes with the external pressure condition, which results in the change in the resistance between the electrodes. Therefore, the wrinkled structure leads to high-pressure sensitivity due to the high surface roughness mismatch.\(^\text{[79]}\) In addition, the fingerprints still contain ridge or valley structures similar to the wrinkle, which also results in high sensitivity within a wide pressure range.\(^\text{[80,81]}\) Furthermore, the wrinkle structure of elastic materials shortens the relaxation time of the material deformation and improves the response-ability of the material to an external pressure load. In other words, the response time and release time of the wrinkle pressure sensor are significantly improved compared with that of the nonwrinkle pressure sensor.\(^\text{[82]}\)

2.2.4. Cilia Structure

Many animals perceive ambient signals through hair cells, e.g., spiders, fish, and mammals through the cilia, lateral line, and inner ears’ hair cells, respectively.\(^\text{[69,83,84]}\) The sensory hair in the hair cell has a large aspect ratio, so it can be easily deflected under pressure, resulting in the interaction between the cilia and the environment.\(^\text{[85]}\) After the load release, the cilia (hair) will return to the initial position. (Figure 3d).\(^\text{[72]}\) A cilium is similar to a cantilever beam, which acts as a mechanical signal amplifier. When the cilium is under bending deformation, the stress decreases gradually with the distance from the cilium root. Therefore, biomimetic cilia sensors should be designed with a nonuniform stress distribution.\(^\text{[83]}\) In particular, out-of-plane
structures similar to cilia can significantly enhance the capability of the system to perceive the environment. In addition, various out-of-plane microstructures (e.g., pyramids, cubes, lines, hemispheres, and columns) are fabricated using patterned mold transfer or photolithography techniques which greatly improve the responsiveness of sensors.

2.2.5. Structural Design

In addition to enhancing the sensing ability of sensors, the structural design can induce other new capabilities. Figure 3e shows a mechanochromic device that can change color in response to the opening and closing of cracks. The mechanochromism is achieved by coating green and orange fluorescent molecules on a PDMS layer. The overall fluorescent color is determined by the linear combination of green and orange fluorescence, which equals to the area ratio of opening cracks and non-opening-crack surface. With the increase in the strain, the color of the material changes from green to yellow to orange. Once the strain is released, the cracks return to their initial state and can be completely closed, which results in light-shielding, preventing the observation of the luminescence of the material. The mechanochromism can also be controlled by nanocylinders. Cylindrical arrays can be fabricated on the surface of flexible materials using silicon template transfer printing. With the increase in the strain, the resonance peak of the nanocylinders is red-shifted. Furthermore, thin-film wrinkling or unfolding have also been shown to lead to variable transparency.

2.3. Stretchability-Enhanced Microstructures

Representative topological patterns generated by micro buckling, latticing, spinning, cutting, and folding, are typically used to support reversible stretchability for various stimuli-responsive materials while maintaining their original functionalities. Stretchability-enhanced microstructures, such as wrinkles, buckles, serpentesines, networks, helical designs, Kirigami, and Origami, are frequently used for the fabrication of skin-like electronics.

Figure 3. Response-enhanced microstructures. a) Crack-enhanced microstructures. Reproduced with permission. Copyright 2014, Nature Publishing Group. b) Porous-enhanced microstructures. Reproduced with permission. Copyright 2014, Wiley-VCH. c) Wave-enhanced microstructures. Reproduced with permission. Copyright 2016, IEEE Press. d) Cilia-enhanced microstructures. Reproduced with permission. Copyright 2018, Royal Society of Chemistry. e) Structure-induced color alteration microstructures. Reproduced with permission. Copyright 2016, Nature Publishing Group.
2.3.1. Wrinkle Structure

For a film/fiber with high modulus bonded onto a compliant pre-strain substrate, the release of the pre-strain leads to the wrinkling of the stiff film into wave-like shapes.[90–96] This system has a compliant response (low elastic modulus) because the stiffness of the wrinkled film/fiber is negligible. Typically out-of-plane and in-plane wrinkles are shown in Figure 4a. Zang et al.[96] developed extremely stretchable wrinkling forms of graphene papers by combining graphene with a pre-strained compliant substrate. The left-top panel of Figure 4a shows the initially flat graphene paper bonded on the pre-strained substrate, and the left-bottom panel of Figure 4a shows the out-of-plane wrinkled form after releasing the pre-strain. Ryu et al.[95] formed in-plane wrinkling silicon nanowires on a pre-strained substrate. The right-top and -bottom panels of Figure 4a shows the morphology of the silicon nanowire before and after releasing the pre-strain, respectively.

2.3.2. Buckle Structure

Buckle is a basic physical phenomenon resulting from instability. A combination of the rigid functional films with compliant substrates in the form of buckles enables the films to minimize the spatial constraint.[97,98,106–108] Methods based on stress-induced bending initiate a buckling process with deterministic control over buckling geometries. Ko et al.[98] discussed the effects of the adhesion energy and degree of pre-strain on the buckling modes of Si circuit meshes integrated on PDMS membranes and found that different adhesion behaviors resulted in contour shapes, including single and multiple buckling. The left-top[97] and right-top[98] panels of Figure 4b are pre-fabricated circuit meshes bonded on a pre-strained substrate. The left-bottom[97] and right-bottom[98] panels of Figure 4b show the single and multiple buckling of the meshes after releasing the pre-strain, respectively.

2.3.3. Serpentine Structure

Both wrinkle and buckle structures require the release of a pre-stretch to achieve stretchability. Serpentine[25,99,109–111] is a planar structure that can be stretchable itself without the need for pre-strain release. Gonzalez et al.[99] studied different serpentine shapes (i.e., “U”, half-circle, elliptical, and “horsehoe” shapes), and found that the “horseshoe” design induced a high stretchability (i.e., minimum stress concentration). The left-top and -bottom panels of Figure 4c show “horseshoe” metal interconnects embedded into a substrate before and after 25% elongation. Here, the buckle of the serpentine due to elongation is in-plane. Out-of-plane buckling occurs if the serpentine is thin enough, which further improves the stretchability of the serpentine. Kim et al.[125] compared serpentine with in-plane and out-of-plane buckling under large elongation and found that fewer cracks were produced with out-of-plane buckling. The right-top and -bottom panels of Figure 4c show out-of-plane buckling of the serpentine before and after elongation, respectively.

2.3.4. Network Structure

In biological tissues, curved and chained microstructures, such as collagen fibrils, consist of cross-linked fiber networks that have a superior stretchability.[100,101,112–118] Therefore, artificial materials with network design could be utilized to design stretchable electronics. Figure 4d shows the stretching behaviors of a deterministic network and cross-linked network. Yang et al.[100] integrated optimized network geometries (i.e., triangular, square, honeycomb, and kagome) into low-modulus elastomers as a structural reinforcement, with accurately designed stress/strain responses. The left and middle panels of Figure 4d show the unstrained and strained triangular networks, respectively. Yang et al.[101] investigated the extreme tear resistance of skin associated with cross-linked collagen fiber networks. The right-top and -bottom panels of Figure 4d show the arrangement of curved collagen fibers before and after stretching, respectively.

2.3.5. Helical Design

Many native tissues, such as the muscles and tendons of animals and the tendrils and cirri of plants, are extremely stretchable. The stretchability of these tissues can be attributed to their crimped fibrous structures having hierarchical helices of single fibers (at the nanoscale) to tissue bundles (at the mesoscale). The helical structure[102,103,119–121] exhibits exceptional stretchability and toughness in comparison with the primary yarn and can be stretched up to 15 times its initial length without rupture. Inspired by the hierarchical helical structure of muscles and tendons, Li et al.[102] developed a highly stretchable and tough nanofiber helix microtissue. This microtissue is shown in the left-top and -bottom panels of Figure 4e before and after stretching, respectively. The elastic helical filament proposed by Gerbode et al.[103] showed an initially compliant response (right-top panel of Figure 4e) and straightening of the helical filament after stretching (right-bottom panel of Figure 4e).

2.3.6. Kirigami and Origami

Kirigami and origami can be used to design stretchable 2D materials with various complex 3D structures by cutting and folding. There exist pierced strips in kirigami constructs, which can help sustain further deformations, including stretching, twisting, and shearing. In kirigami designs, the stretching mechanism is dominated by the out-of-plane bending mechanism of the parts around each slit.[104,122,123] In origami design, folds of each paper can be adjusted to form different structures. Also the 2D/3D origami type structures can be formed by reversible control using the strain-driven self-assembly. Morikawa et al.[104] developed an ultra-stretchable bioprobe film device with a kirigami design applicable to tissues and organs. The morphologies of this device with a slit pattern before and after stretching are shown in the left-top and -bottom panels of Figure 4f, respectively. Yan et al.[105] introduced a strategy to exploit mechanical buckling for the autonomous assembly of 3D structures into origami shapes in materials with different length scales. The right-top and -bottom panels of Figure 4f show a plastic soccer ball with a 3D origami and 2D structures, respectively.
Figure 4. Stretchability-enhanced microstructures. a) Wrinkle: a flat graphene paper on a pre-strained substrate before (left-top) and after (left-bottom) releasing the pre-strain (Reproduced with permission.\cite{96} Copyright 2014, Nature Publishing Group) and a silicon nanowire on a PDMS substrate initially without (right-top) and with (right-bottom) the use of pre-strain (Reproduced with permission.\cite{95} Copyright 2009, American Chemical Society). b) Buckle: global (left-top and -bottom) (Reproduced with permission.\cite{97} Copyright 2008, the National Academy of Sciences of the United States of America) and local (right-top and -bottom) (Reproduced with permission.\cite{98} Copyright 2009, Wiley-VCH) buckling modes of prefabricated circuit meshes on substrate before and after relaxing the pre-strain. c) Serpentine: in-plane (Reproduced with permission.\cite{99} Copyright 2008, Elsevier Ltd.) and out-of-plane (Reproduced with permission.\cite{25} Copyright 2010, Wiley-VCH) buckling serpentine before (left- and right-top) and after (left- and right-bottom) elongation. d) Network: triangular networks without (left) and with (middle) tensile strain (Reproduced with permission.\cite{100} Copyright 2015, Nature Publishing Group), and cross-linked collagen fiber networks before (right-top) and after (right-bottom) stretching (Reproduced with permission.\cite{101} Copyright 2015, Nature Publishing Group). e) Helical design: unstrained (left-top) and strained (left-bottom) hierarchical helix nanofiber yarn (Reproduced with permission.\cite{102} Copyright 2019, the National Academy of Sciences of the United States of America), and elastic helical filament before (right-top) and after (right-bottom) tension (Reproduced with permission.\cite{103} Copyright 2012, the American Association for the Advancement of Science). f) Kirigami and origami: kirigami paper before (left-top) and after (left-bottom) stretching (Reproduced with permission.\cite{104} Copyright 2018, Wiley-VCH), and a plastic soccer ball with 3D (right-top) origami structure due to an initially 2D (right-bottom) form with pre-strain (Reproduced with permission.\cite{105} Copyright 2016, Wiley-VCH).
3. Bio-Electronics

The five sensory systems (olfactory, visual, gustatory, auditory, and tactile) allow humans to gain information from the external world. Recent innovations in the types and structural design of materials have allowed the development of bio-inspired skin-like devices that can simulate the human sensory systems. Figure 5 lists the six flexible sensing materials and microstructures introduced in Section 2.1 and their application range in five sensory systems.\textsuperscript{125–134} The photo-responsive materials are used for visual (e.g., UV, color, and light intensity) perception. The thermo-responsive materials make the sensor have thermal tactile sensing function. The humidity-responsive materials are used to sniff the moisture content of gas. The chemo-responsive materials sense the chemical composition in the external environment, including gas and liquid compositions. The mechanical-responsive materials are used to fabricate the auditory (e.g., sound) and tactile (e.g., pressure, strain, and stress) devices. The magneto-responsive materials sense the distance and position of objects, which are broad visual and tactile.

The skin-like devices are light-weighted, deformable, and function reconfigurable. These advantages enable them conformably attach with human tissues or organs, which not only decreases the uncomfortable feeling of devices wearing but also eliminates the mismatch impact of the devices and human body on the measurement accuracy of the physiological signal. Figure 6 shows examples of skin-like devices developed considering the five human sensory systems.\textsuperscript{125–134}

3.1. Olfactory Devices

The human nose contains almost 400 olfactory receptors with the capability of detecting the gaseous chemicals at a distance. Inspired by the role of the human noses play in identifying gases, skin-like flexible gas sensors have been successfully fabricated. These flexible gas sensors show good sensitivity and accuracy in the detection of various gases, including nitrogen dioxide (NO\textsubscript{2}), carbon dioxide (CO\textsubscript{2}), ammonia (NH\textsubscript{3}), hydrogen sulfide (H\textsubscript{2}S), and hydrogen (H\textsubscript{2}). Figure 6a shows a micro-supercapacitor integrated on a deformable substrate and used to drive the graphene-based gas sensor. The sensing mechanism of the NO\textsubscript{2} gas sensor is based on the change in electrical conductivity of p-type graphene after the absorption of NO\textsubscript{2}, which is a strong oxidizer and could withdraw electrons from graphene. The synthetic patterned-graphene gas sensor integrated with the micro-supercapacitors can detect NO\textsubscript{2} gas for more than 50 min even under a 50% uniaxial stretching; it is suitable for flexible and stretchable environments such as the human body.\textsuperscript{125} In addition to the detection of gases, noses can also respond to gases by, e.g., sneezing. Hou et al.\textsuperscript{126} developed a semicrystalline polymer actuator that can be driven by solvent vapor synthesized with crystallizable polyester segment poly (ε-caprolactone) and isophorone diisocyanate trimmer. Due to the reversible swelling–crystallization conversion of the crystallizable polyester segment, the synthesized polymer can expand and shrink in response to organic solvent vapor (Figure 6c). The contraction of the polymer-based actuator (1 mm thick) needs only ≈4 s in room temperature and can exhibit a fast self-oscillation when exposed to air.

3.2. Visual Devices

As one of the most sophisticated organ systems in animals, visual systems play a significant role in the survival of creatures as they allow animals to search, move safely, copulate, and identify information. These systems transform electromagnetic radiation into visual physiological signals and account for more than 80% of the external information sent into the brain. Traditional digital cameras are usually based on 2D planar substrates integrated with patterned photoelectric detectors. However, they fail to fully capture the information of the observed object, especially in 3D imaging.\textsuperscript{139} Currently, bio-inspired image-capturing optical systems characterized integrated with hemispherical micro-lens array-type compound sensors have been widely exploited for various imaging techniques, ranging from complementary metal-oxide semiconductor and charge coupled device imaging sensors to compact optical systems, along with the flexible and stretchable electronic techniques.\textsuperscript{140–142} Figure 6d shows a photograph of the deformable and hemispherical-shaped camera inspired by arthropod eyes.\textsuperscript{127} The surfaces of the fabricated arthropod-inspired cameras were densely distributed with 180 artificial imaging elements, which is approximately the number of eyes of dark beetles and fire ants. The devices were fabricated from 2D planar geometries to hemispherical shapes integrating the deformable arrays of thin silicon photodetectors and elastomeric compound optical elements. Figure 6e shows a photograph of the line-art illustrations of a “Horus eye” (an Egyptian hieroglyphic character) taken with the hemispherical-shaped compound eye cameras.\textsuperscript{127} Another developed hemispherical eye system constructed with origami silicon optoelectronics is shown in Figure 6f.\textsuperscript{128} The artificial compound eyes consisted of the focal plane arrays based on the single-crystalline silicon and featured with hemisphere shape. The convex isogonal polyhedral concepts make it feasible for various polygons to form into spherical formats. Each polygon block in the silicon-based devices could be used as an independent sensor pixel. The fabricated electronic eye prototypes represent simple and low-cost methods as well as flexible optimization parameters in terms of pixel density and design.\textsuperscript{128}
3.3. Gustatory Devices

Animal tongues use taste buds to distinguish sourness, saltiness, sweetness, bitterness, and other types of taste. Each taste bud contains 50–150 taste receptor cells coupled with protein to deliver the relative chemical information to the brains to detect the types of foods. The role tongues play in animal survival has attracted considerable interest to develop bio-inspired gustatory devices as flexible electrochemical devices,[143] which show high potential in the measurement and monitoring of bio-fluids.

A typical application of flexible electrochemical devices is the monitoring of intravascular blood glucose level in human bodies. Conventional commercial glucose monitoring relies on invasive lancet approaches (i.e., repeated finger pricks), which not only hinder patient compliance because of pain but also result in skin irritation and bacterial infections.[129] Therefore, Chen et al.[129] fabricated a skin-like biosensor (Figure 6g) for highly accurate monitoring of intravascular blood glucose in a noninvasively and in situ manner. The ultrathin skin-like biosensor in the biosensor system utilized a paper-like battery powered by electrochemical twin channels (ETCs) to drive the intravascular blood glucose out of the vessels. Then the glucose at the surface of human skin could be further analyzed by the biosensor. However, this noninvasive glucose monitoring system consists of two independent systems, and an integrated system combining the ETCs and the sensing electrodes should be developed.

Sweat is a representative bio-fluid that can be easily collected noninvasively and contains important biomarkers reflecting the status of human health, such as proteins, electrolytes, and small molecules. The quantitative analysis of chemicals in sweat can be an alternative to blood analyses.[144,145] Thereby, Koh et al.[130] designed a microfluidic device with good flexibility and stretchability. This device could not only collect the sweat produced from human skin via the microfluidic method but also...
wirelessly communicate the measurements via integrated electronics bonded to the surface of the human skin without mechanical and chemical irritation. The fabricated microfluidic device could be used to monitor and assess the concentration of glucose, lactate, pH, creatinine, and chloride ions in sweat with the colorimetric sensing approach (Figure 6h). Though body fluids (i.e., sweat, saliva, tears) are potential candidates for direct monitoring of the physical signs of the human body, their correlation with blood levels is confounding and great challenge due to the impact of water evaporation, seasonal fluid volume changes, as well as the difference of chemicals density (e.g., the density of glucose in those fluids is only 1–10% of that in the blood).

3.4. Auditory Devices

External sound signals can induce the vibration of animal eardrums at precise frequencies and amplitudes. The working mechanism of eardrums can be exploited to noninvasively and quickly diagnose, evaluate, and continuously monitor diseases in the long term using acoustic and ultrasonic detecting devices. For example, Wang et al. developed a flexible skin-mounted device for the long-term and real-time monitoring and evaluation of bowel sounds based on a 3D-printed elastomeric resonator integrated on a flexible skin-mounted device (Figure 6i). Ultrasonic detecting technology is an outstanding noninvasive approach for the monitoring of the central blood pressure waveform from the jugular vein and carotid artery. The corresponding ultrasonic devices could provide important clinical data for the prediction and diagnosis of all-cause and cardiovascular mortality. Wang et al. developed an ultrasonic device (wrapped on a non-developable surface; see Figure 6j) that conforms to the human skin and can precisely evaluate the blood pressure waveforms at human venous and arterial sites. This ultrasonic device had ultrathin thickness (240 μm) and considerable stretchability (up to 60% strain), as well as the capability of continuously monitoring the cardiovascular status.

3.5. Tactile Devices

Human skin contains widely distributed nerve cells that can sense external-world stimuli, such as stress, slight pressure, temperature, and vibrations. The advances in bio-inspired flexible electronics have promoted the development of various epidermal tactile devices with the capability of simulating human skin senses, such as skin-like pressure sensors, epidermal temperature sensor, and flexible strain sensor. Chen et al. developed a biocompatible and ultra-flexible strain sensor (bent on a glass rod; see Figure 6k) for the real-time and long-term measurement of pulses and body motion based on shear lag theory and the liquid transfer printing method. The sensor was fabricated on a semi-permeable porous film, which showed good air permeability, biocompatibility, and waterproofness. Figure 6l shows a photograph of this temperature sensor after a 24 h in vitro wearing test, and the inset shows the microscopical structure of the sensing part of the temperature device after peeling off from the human skin. Until now, the complexity of the human skin has made it challenging to develop epidermal devices, or electronic skins, with real tactile senses as the human skin.

Although rapid progress has been made in skin-like devices, there are still many challenges, such as poor biocompatibility, low integration, and so on. The ample room for further technological innovations for high sensitivity and durability, multifunctional integration, and advanced fabrication necessitates continuous efforts in multiple disciplines.

4. Bio-Physical Interaction

The five delicate sensory systems described earlier (olfactory, visual, gustatory, auditory, and tactile devices) use five different types of signals (gas, light, chemical composition, sound, and mechanical signals). This section introduces the application of these delicate sensory systems for physical conditions, including breathing composition, blood oxygen, blood pressure, blood glucose, bowel sounds, and pulse.

4.1. Gas Monitoring

Breathing is the biological activity with the fastest exchange of materials between the human body and the outside environment. Therefore, the gas detection during breathing is an effective method to detect the physiological health of humans. Existing breathing gas detection techniques mainly include breathing rate detection and breathing gas composition analysis. Gas monitoring is also important for the diagnosis of ovarian cancer, which is a common malignant tumor in female reproductive organs. Flexible sensors based on gold nanoparticles can be used to diagnose ovarian cancer noninvasively from the exhaled breath. The left panel of Figure 7a shows the different bending states of a gold nanoparticle sensor with high response to a specific volatile organic compound in the exhaled gas linked with ovarian cancer. The normalized strain response of the sensors under exposure to different volatile organic compounds is shown in the right panel of Figure 7a. Pang et al. used a porous graphene network to prepare a wearable humidity sensor, which could be integrated inside a medical breathing mask (left panel of Figure 7b). Figure 7b shows the respiration monitoring results with fast, normal, and deep breathing.

4.2. Light Monitoring

Optical monitoring can be used to determine the morphology of the human body’s surface and physiological information in the deep tissue of the human body (e.g., blood oxygen, blood pressure, and blood glucose). Li et al. developed a flexible and stretchable optoelectronic device that can monitor blood oxygen (left panel of Figure 7c). The right panel of Figure 7c shows the percutaneous oxygen saturation (SpO2) and the pulse rate measured with this flexible and stretchable optoelectronic device.
Figure 7. Physiological signal monitoring application. a) Breathing gas monitoring for ovarian cancer diagnosis. Reproduced with permission.[152] Copyright 2015, American Chemical Society. b) Respiration monitoring for wearable humidity sensor. Reproduced with permission.[153] Copyright 2018, Elsevier. c) Blood oxygen monitoring. Reproduced with permission.[154] Copyright 2017, Wiley-VCH. d) Blood pressure monitoring. Reproduced with permission.[155] Copyright 2020, Oxford Academic. e) Blood glucose monitoring. Reproduced with permission.[156] Copyright 2017, American Association for the Advancement of Science. f) Sound monitoring for detecting intestinal diseases. Reproduced with permission.[157] Copyright 2019, Science China Press. g–i) Soft prosthetic finger with tactile sensor for (g) recording pulse waves, (h) kneading ping-pong ball, and (i) evaluating the hardness of balls. Reproduced with permission.[158] Copyright 2019, Wiley-VCH.
device. The SpO2 value calculated from the response of the device changed by only 0.1% while stretching the skin, indicating the high stability of the device. Li et al.[157] developed a wearable skin-like optoelectronic system (left panel of Figure 7d) that can be used to measure cuff-less continuous blood pressure. The right panel of Figure 7d compares the estimated systolic blood pressure measured by this optoelectronic system with that obtained by the invasive (intra-arterial) method, which were consistent.

4.3. Chemical Composition Monitoring

Water accounts for 60–70% of the human body weight. The bio-fluids (e.g., interstitial fluid (ISF), sweat,[159,160] tear fluid,[161] and saliva[162]) in the human body can be related to various diseases and metabolism. Chen et al.[159] developed an ultrathin (total thickness ≈3 μm) device for continuous blood glucose monitoring. The device was thin enough (total thickness ≈3 μm) to be biocompatible conformed to various complex surfaces (left panel of Figure 7e). A series of in vivo control tests, including a glucose tolerance test (a common screening method for diabetes), were performed to verify the effectiveness of the glucose biosensor. The correlation coefficient between the glucose biosensor test and blood test was 0.9997 (right panel of Figure 7e).

4.4. Sound Monitoring

Mechano-acoustic dynamics, represented by mechanical waves, have a direct or indirect relationship with natural physiological activity in the human body, such as vocal-cord vibration, skeletal muscle contraction, the closure of heart valves, and gastrointestinal tract movement.[147] Therefore, the real-time and quantitative monitoring of mechano-acoustic waves might be useful for clinical diagnoses and human healthcare. Bowel sounds, which can be aperiodic and random, are useful for the detection of intestinal diseases. Wang et al.[163] developed a flexible circuit board integrated with a 3D-printed elastomeric resonator (left panel of Figure 7f) that can continuously monitor intestinal sounds and wirelessly transmit the signals. The right panel of Figure 7f shows the same bowel sound signal measured with a traditional e-stethoscope and this flexible device, which verifies the reliability of the flexible device. They recorded the bowel sounds of healthy patients and patients suffering from mechanical intestinal obstruction and paralytic ileus, and found a clear difference in the number and amplitude of peaks in the unhealthy patients.

4.5. Mechanical Signal Monitoring

Physiological activities such as heartbeat, breathing, and joint movement are linked to the mechanical movement of human tissues. Flexible tactile devices can conformably attach to the surface of human tissues and measure these mechanical signals accurately. Liang et al.[156] presented a soft prosthetic finger integrated with flexible high-performance (good linearity, fast dynamic response, and high stability) force sensor arrays. Figure 7g shows the traditional Chinese medical pulse by using this soft prosthetic finger with the sensor array in the radial artery. Figure 7h demonstrates the flexibility of the sensor array. In this demonstration, a ping-pong ball is kneaded between two soft fingers, and the pressure distribution on the fingers is recorded with the sensor array. Subsequently, a soft prosthetic finger integrated with the sensor array measured the pressure distribution of balls made of Ecoflex-10 (Smooth-On Inc., USA), Ecoflex-30 (Smooth-On Inc., USA), PDMS (10.1, Sylgard 184, USA), and dragon skin 30 (Smooth-On Inc., USA). This experiment showed that balls with different hardness lead to different resistance change responses (right panel of Figure 7i).

5. Concluding Remarks

The advances in the material and structural design of skin-like electronics have allowed the achievement of the communication between people and objects, showing potential applications for soft robots and biomedical fields. Figure 8 shows the application prospects of skin-like electronics for humans (e.g., health care, sports, entertainment, virtual reality, and brain-computer interface) and objects (e.g., robots, Internet of Things, and equipment health). However, there are still many challenges in the development of skin-like electronics. First, the sensitivity and stability of skin-like electronics still need to be improved. Second, the durability of skin-like electronics has to be improved because they undergo large deformations with human tissues and are thus prone to fatigue failure. Therefore, it is necessary to develop materials and structures with high fatigue resistance. Third, the cost has to be lowered and mass production has to be achieved. Although skin-like electronics have been widely used in the laboratory, ordinary consumers have not benefited from skin-like electronics. Therefore, new materials and processes have to be developed to reduce the production cost of skin-like electronics.

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