

Recent Advances in Materials, Designs and Applications of Skin Electronics

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ABSTRACT As electronic devices get smaller, more portable, and smarter, a new approach of realizing electronics in thin, soft, and even stretchable style that could be worn and attached to the skin, which is called skin electronics, has emerged and attracted much attention. To achieve well compliance, extend the maximum stretchability, promote the comfortability of wearing, and make the most use of the skin electronics, researchers are making efforts in different aspects. In this article, we summarized the recent advances in categories of materials science, design strategies and novel applications. Examples of skin electronics using various functional materials including piezoelectric, thermoelectric, etc., and soft conductive materials including PEDOT: PSS-based conductive polymer, carbon nanomaterials, metal-based materials and hydrogels were given. Different mechanics design strategies for enhancing mechanical performance and comfortability design strategies for better wearing experience were introduced. Lastly, practical applications of skin electronics in fields of smart healthcare and human-machine interface were discussed. Research focused on these aspects all boosted the development of skin electronics in different dimensions, with which combined together may help skin electronics take a leap into truly ubiquitous use in our daily life.

INDEX TERMS Engineering in medicine and biology/biomedical engineering/biomedical electronics, industrial electronics/assembly systems/flexible electronics, sensors/wearable sensors.

I. INTRODUCTION

Electronics has been evolving over a century since J.A. Fleming invented the first vacuum diode in 1897, as the sizes of components and boards got smaller and smaller, integrated circuits helped people shrink massive computers into portable, mobile, and even wearable devices. However, as traditional electronics are based on silicon and rigid printed circuit boards (PCB), they apparently don't perfectly suit wearable scenarios, where movements, deformations, perspirations and breathing of the skin might be hindered due to the mismatching between Young's moduli, flexibility and permeability of rigid electronics and human body. In this context, the concept of soft, thin and stretchable skin electronics is brought up for

filling these gaps between rigid electronics and soft human body, which gives more opportunities for electronics in various fields and may become a next evolutionary platform for future electronics.

Since the first stretchable single crystal silicon wavy ribbon is demonstrated in 2006 [1], people have explored numerous new materials, methods, structures, advanced fabrication process, and innovative applications for the development of soft, flexible and stretchable electronics. Apart from those for implanted use, skin electronics is typically applied on the skin outside of the body, functions for sensing physiological signals, monitoring physical activities, harvesting energy from daily life, and delivering medical therapies, stimulations, or perceptible information to the human body. The goal of

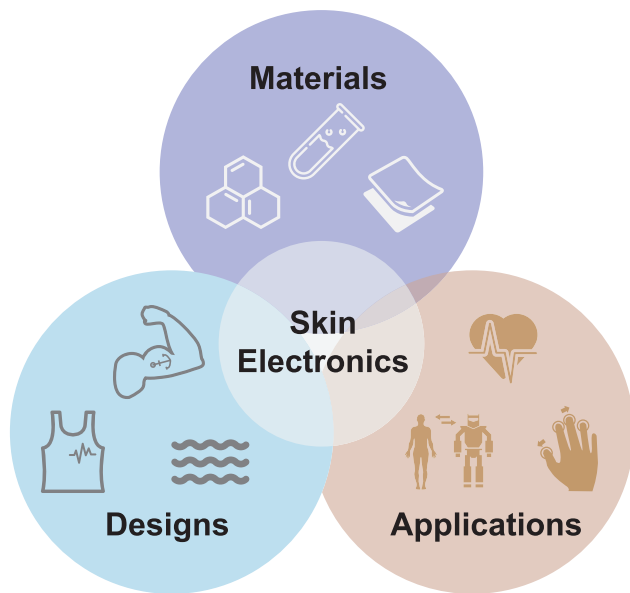


FIGURE 1. Schematics of skin electronics in the aspects of materials, designs, and applications.

realizing high-fidelity signal or information interacting with the skin means the device should be able to intimately contact with the skin without peeling off, and maintains stable electrical or mechanical performance even when it undergoes stretching, bending or twisting caused by natural movements and deformations of the skin. That means the materials for composing skin electronics should either be intrinsically or engineered to be flexible and stretchable. Meanwhile, the adhesiveness, bio-compatibility (that won't cause allergic response), moisture permeability, wireless independence and unobtrusiveness are also desired for developing ideal skin electronics.

To develop high performance skin electronics devices and systems that meet the needs mentioned above, researchers made efforts in different aspects of it, from basic chemistry and materials parts of the device, the mechanics and electrical engineering parts, integration and miniaturization, to exploring the potential applications in various fields. In this review, we summarized recent high impact and representative research advances in skin electronics and categorized them in three main topics, materials, designs and applications, each for one chapter (Fig. 1). For the materials part, we firstly introduce most commonly used options in substrate selection, followed by examples in functional materials with specific physical or chemical properties for different purposes, and more importantly, the typical soft conductive materials which are essential for constructing electrical connections and circuits, to make them flexible and stretchable enough for stably working on soft and deforming skin. The designs chapter discussed several mechanics design strategies that help turn unstretchable materials (e.g., metal) into stretchable formats, and different routes for fabricating the skin electronics with higher comfortability for wearing, in aspects

of thin and conformable enough to be imperceptible, and permeable, breathable in styles of textiles and clothes. Last but not least, we listed a series of advanced research work on skin electronics in different applications, which can be roughly categorized into two sub-groups according to their purposes, which are healthcare-oriented and human-machine interface (HMI) related skin electronics.

II. MATERIALS FOR SKIN ELECTRONICS

A. SUBSTRATE AND ENCAPSULATION

For soft and stretchable electronics, related substrate materials largely determine their functional performance, as almost all functional electronic devices and circuit connections must be carried by various substrates during the manufacturing process or as final products. Required substrate materials for flexible electronics manufacturing are usually soft and bendable films, such as common polyethylene terephthalate (PET), polyimide (PI), etc. However, for skin electronics, requirements for the intrinsic properties of substrate materials are more stringent, such as the biocompatibility between substrate materials and the skin (non-toxic to the skin, and low likelihood of causing allergic to the skin), similarity or matching of Young's modulus of skin and substrate materials, the conformality between substrate materials and the skin, and the ductility of substrate materials, etc. Often reported soft substrate materials for skin electronics are silicone-based elastomers (Ecoflex, PDMS) [2], [3] and rubbers (SBS, SEBS) [4], [5], and more, hydrogels, as three-dimensional network polymer materials filled with water, possessing good flexibility and biocompatibility, therefore are increasingly used as interface materials between skin and soft electronics. The intrinsic superior compliance of Ecoflex substrates makes them often used in skin electronics with large curvature deformation and huge stretch [6], [7], [8]. Unlike the preparation process involving chemical cross-linking of silicone elastomers, polystyrene-block-polybutadiene-block-polystyrene (SBS) and polystyrene-block-poly(ethylene butylene)-block-polystyrene (SEBS) are typical block copolymers, and can be prepared into elastomers by physical cross-linking method (e.g. solvent evaporation), which allows them to match more substrate manufacturing crafts [4], [5]. Hydrogels, which closely resemble human tissue due to their abundant water content, are considered very promising as tissue-electronic interfaces [9], [10]. Besides, some water-soluble substrate materials, such as PVA, are widely used in skin electronics' fabrication/transfer process [11].

B. FUNCTIONAL MATERIALS

With further research on skin electronics, varieties of functional skin electronics have been designed and developed to satisfy people's individual needs, such as resistance-based sensors for strain and tactile detection, electronic skins with energy harvesting devices, or electro-chromic/luminescent functions, etc. To meet the functional requirements mentioned

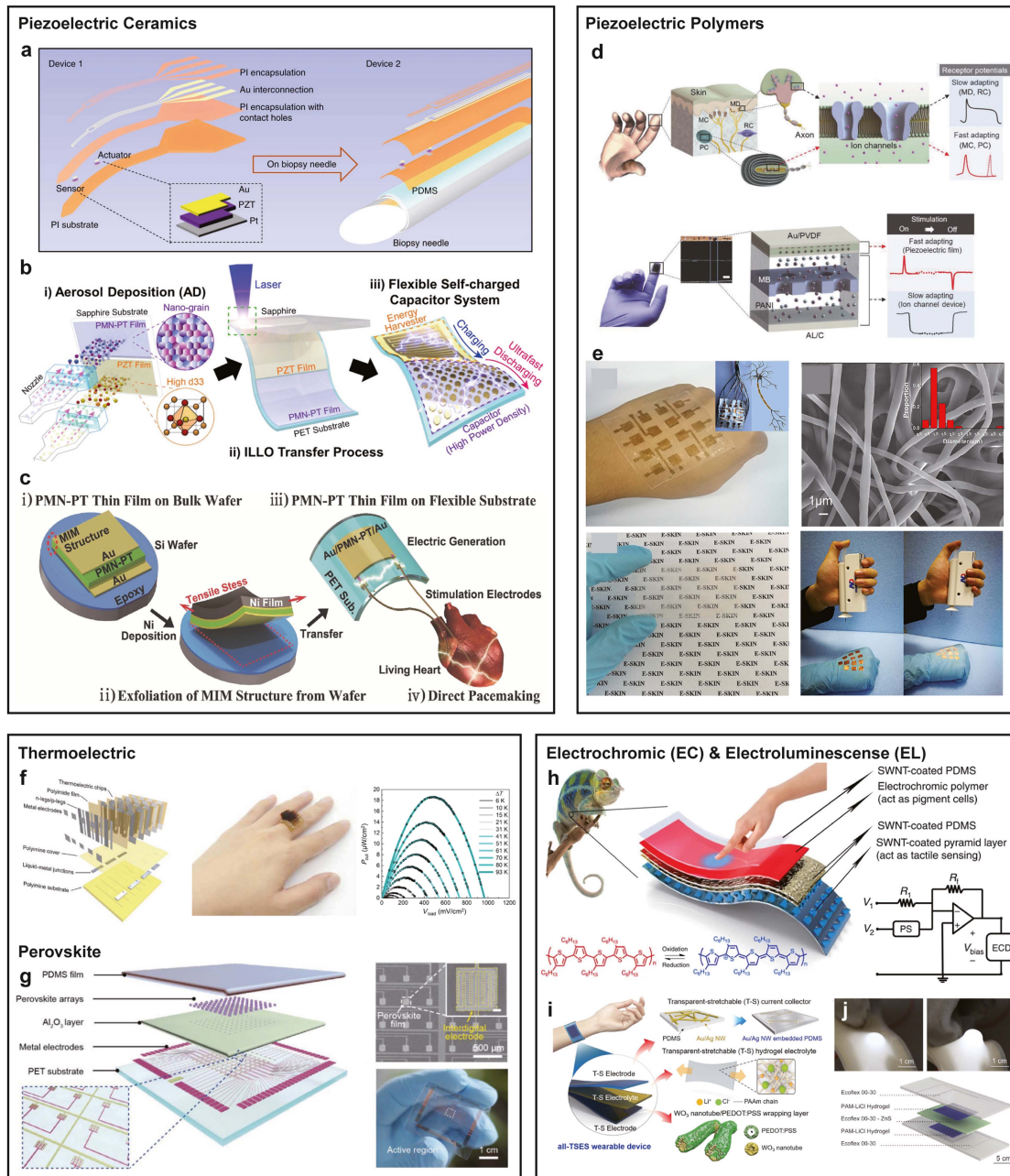


FIGURE 2. Functional materials for skin electronics. (a) PZT-based piezoelectric actuator and sensor for tissue modulus detection [12]; (b) Self-powered system with integrated PZT-based energy generator and PMN-PT-based capacitor [13]; (c) PMN-PT-based piezoelectric film for direct electrical stimulation of cardiac pacing [14]; (d) PVDF-based self-powered mechanoreceptor for e-skin with stimulus recognition [15]; (e) Piezoelectric fiber films by electrospinning PVDF organic solution for e-skin [16]; (f) Wearable thermoelectric generator through Lego-like reconfigurable design based on Bi and Sb chalcogenides thermoelectric legs [19]; (g) Flexible perovskite arrays for photodetection [20]; (h) Electrochromic polymer-based e-skin for touch sensing [22]; (i) electrochromic e-skin based on inorganic electrochromic material (WO_3) [23]; (j) Stretchable electroluminescent e-skin based on ZnO phosphors [2].

above and principles for wearables, the selection and design of functional materials are essential to manufacturing skin electronics. To start with, piezoelectric ceramic materials are often utilized in transducing between mechanical pressure and electrical signals, and thus can be adopted for pressure sensors. Yu et al. [12] fabricated piezoelectric ceramic lead zirconate titanate ($Pb(Zr_{0.52}Ti_{0.48})O_3$, PTZ) films with

a thickness of 500 nm, and utilized the piezoelectric effect and inverse piezoelectric effect of PZT to detect tissue modulus (Fig. 2(a)). Peddigari et al. [13] designed an all-in-one skin electronic system integrating energy harvesting and storage, in which flexible PZT film was used as the energy harvesting material due to its excellent piezoelectric properties, and $Pb(Mg_{1/3}Nb_{2/3})O_3$ - $PbTiO_3$ (PMN-PT) was

used as the capacitor material (Fig. 2(b)). Similarly, work by Hwang et al. [14] employed flexible PMN-PT thin film as piezoelectric layer for developing a self-powered cardiac pacemaker (Fig. 2(c)). In addition to the piezoelectric ceramics mentioned above, piezoelectric polymers are also common functional materials for flexible piezoelectric skin electronics due to their better compliance and capacity to be fabricated into desired structures through common processes. Chun et al. [15] reported a flexible self-powered mechanoreceptor sensor using polyvinylidene fluoride (PVDF) film as piezoelectric layer to achieve simulating the perception of complex pressure stimuli by human skin (Fig. 2(d)). Wang et al. [16] dissolved PVDF in an organic solvent, then prepared fiber films with both piezoelectric and pyroelectric effects by electrospinning the mixture solution (Fig. 2(e)).

Flexible electronic devices based on thermoelectric energy conversion are another promising alternative, since, skin electronics are in direct or close contact with the human body, which provides the entire system with continuous and stable heat. There are mainly two aspects to the design of thermoelectric energy conversion devices in skin electronics: 1) the original fabrication of functional thermoelectric energy conversion components (including fundamental material design and process development, etc.); 2) the optimization of structure and mechanics based on commercially available thermoelectric energy conversion devices. Cao et al. [17] fabricated BiTe-SbTe-based flexible thermocouples using screen printing process, and investigated the effect of different binders on the thermoelectric performance. Based on commercially available thermoelectric legs, Kim et al. [18] conducted structural design and optimization research on thermoelectric energy conversion devices to achieve flexibility and functionality of the overall device through experiments and simulations. Ren et al. [19] developed a wearable and self-healing, reconfigurable thermoelectric generator (Fig. 2(f)). This device is based on thin film Bi and Sb chalcogenides deposited on PI films as the n/p-type legs, and utilized liquid metal's self-healing capability to realize Lego-like reconfiguring, which helped it to be stretchable while owning a high open circuit voltage of $1\text{V}/\text{cm}^2$ and a maximum power output of $19\mu\text{W}/\text{cm}^2$ at temperature difference of 95K. Functional perovskite-based devices have exciting applications in skin electronics, especially photodetectors, work by Wu et al. [20] demonstrated fine flexible perovskite arrays for light sensing and real-time light trajectory capture (Fig. 2(g)). And Lei et al. [21] achieved fabrication of ultrathin perovskite film ($\sim 600\text{ nm}$ to $\sim 100\ \mu\text{m}$) with precise control by lithography-assisted epitaxial growth method for single-crystal photovoltaic devices.

For a long time, controllable and flexible optoelectronic display devices have been a popular research area for expanding skin electronic functions, which can provide the most direct means for human-machine interaction and human-environment interaction. There are two main novel methods for realizing wearable and flexible optoelectronic display: electrochromic (EC) and electroluminescence (EL), both of

which can change human visual perception under the effect of electrical energy. Chou et al. [22] fabricated stretchable electrochromic thin layers by spin-coating electrochromic polymers, and achieved tactile sensing through color changes by pressing-induced changes in applied voltage (Fig. 2(h)). Similarly, inorganic material WO_3 is also often employed to fabricate electrochromic devices, for instance, Yun et al. [23] fabricated stretchable electrochromic supercapacitors basing WO_3 nanotubes coated with PEDOT: PSS (Fig. 2(i)). Electroluminescent materials offer exciting prospects for skin electronics with machine-vision interaction and flexible display. Work by Larson et al. [2] reported an electroluminescent elastomer that can withstand huge deformation while electroluminescence by embedding electroluminescent materials (ZnS phosphor) into the elastomer and forming a multi-layer structure (Fig. 2(j)). Similarly, Zhao et al. [24] synthesized ZnS: Cu, Al phosphor particles with higher luminous efficiency as electroluminescent materials for self-powered touch display.

C. CONDUCTIVE MATERIALS

For the material system of skin electronics, in addition to functional materials for specific functions and substrate/encapsulation materials for overall protection, conductive materials are essential to achieve the interconnection of soft functional components and the complete function of the whole circuits. To match specific application scenarios, the design and selection of conductive materials in skin electronics is very critical to maintaining overall flexibility and functionality, and common conductive materials for interconnection of electronic components can be roughly divided into four categories according to conductive properties: 1) conductive polymer [25], [26]; 2) carbon-based conductive nanomaterials (graphene [27], [28], carbon nanotube [29] et al.); 3) metal-based conductive materials (gold [11], silver [25], [27] etc.); 4) hydrogel-based conductive materials [30]. A type of conductive material may also be combined with other conductive components or modified in molecular structures for achieving better compliance and functionality. For example, PEDOT: PSS is the most typical and widely studied conductive polymer in skin electronics. As shown in Fig. 3(a), Ershad et al. [25] mixed silver flakes into PEDOT: PSS solution to obtain a conductive ink for mask printing directly on the skin surface, and the mixture ink also can be used to write conductive connections on skin with a ball-point pen. A topological supramolecular structure based on PEDOT: PSS was designed by Jiang et al. [26] to achieve high conductivity and better stretchability in small-scale fabrication (Fig. 3(b)). Carbon-based nanomaterials are regarded as promising conductive interconnections in skin electronics due to their excellent processability and compatibility with various functional/substrate materials. Graphene patterns with low sheet resistance can be easily fabricated by transient laser heating PI films [31], for instance, patterns fabricated by carbon dioxide laser-induced graphene (LIG) are employed for

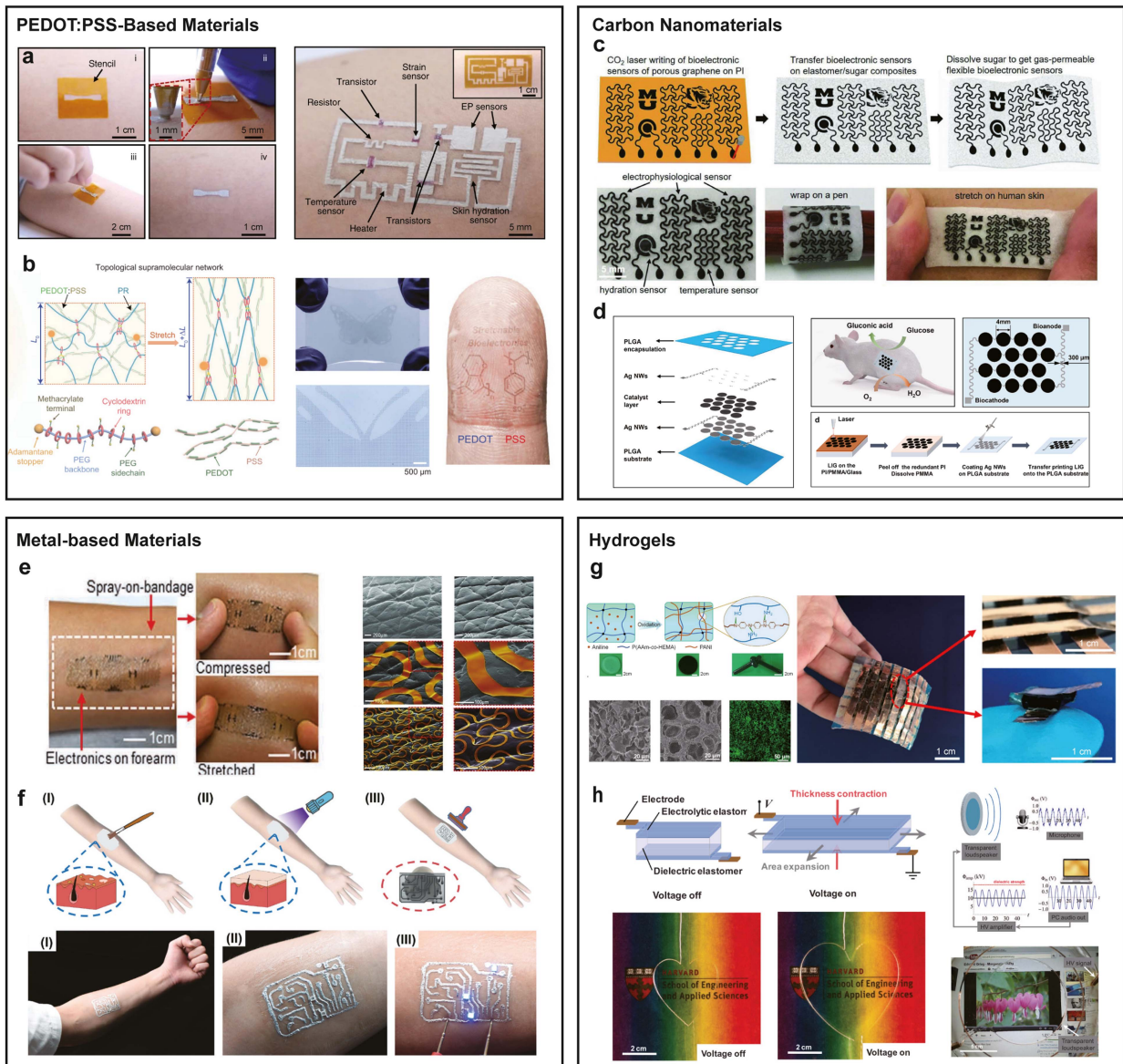


FIGURE 3. Conductive materials for skin electronics. (a) Hybrid conductive ink composed of Ag flakes and PEDOT: PSS for in-situ printing [25]; (b) PEDOT: PSS-based topological supramolecular structure with better stretchability and high conductivity at tiny scales [26]; (c) LIG-based gas-permeable electrophysiological sensor [28]; (d) Biofuel cells with LIG as catalyst carrier [27]; (e) Serpentine Au foils designed for conformal skin electronics [32]; (f) Liquid-metal-based e-skin by in-situ stamp printing [34]; (g) Conductive hydrogel based on PANI polymer network [36]; (j) Ionic conducting hydrogels with NaCl as electrolyte [41].

electrophysiological signals detection [28] (Fig. 3(c)) and biofuel cells [27]. Sun et al. et [28] utilized the precise patterning of LIG to fabricate complex functional circuits, while the high porosity of graphene provides a new option for comfortable and breathable skin electronics. Huang et al. proposed a transient, ultrathin and implantable biofuel cell by LIG and Au nanoparticles [27] (Fig. 3(d)), which harvests energy from the oxidation of glucose and obtains high open circuit potential of 0.77V and maximum power density of 483.1 μW/cm². Kim et al. [29] developed a reconfigurable, low-modulus conductive clay based on carbon nanotubes for electronic skin with thermoelectric energy conversion.

The advantage that metal-based conductive materials have over other conductive materials is their high electrical conductivity. However, bulk metals are mostly rigid and unbendable, which seems to be incompatible with soft and stretchable skin electronics. Therefore, necessary structural design and process optimization are required for intrinsically rigid metal connections in skin electronics (detailed description in next section); for instance, gas-permeable Au nanomeshes [11] and lightweight serpentine Au foils [32] (Fig. 3(e)) are fabricated to form conformal contact with skin. Another class of metal-based materials that are very promising for e-skin fabrication are gallium-based liquid metals, which, like the

well-known mercury (Hg), are fluid at room temperature and have metallic electrical conductivity. Notably, unlike metallic mercury, gallium-based liquid metals have low vapor pressures and negligible toxicity [33], which enables them to be used as conductive connections in skin electronics. Wu et al. [34] stamp-printed liquid-metal-based conductive nanoclay to fabricate e-skin with excellent electrical conductivity in situ on the arm (Fig. 3(f)). Not only bulk liquid metals can be used to print e-skins in situ, but liquid metal particles similarly. Lee et al. [35] achieved high-precision mask printing of electronic tattoos directly on the skin based on liquid metal particle coated with Pt-modified CNTs. Hydrogel-based conductive materials have become a popular option as connecting components in skin electronics due to their biocompatibility, pluripotency, and functionality easy to extend and modify. The design ideas of hydrogel-based conductive materials can be roughly divided into three categories according to the components that play a conductive role, namely polymer conduction [36], penetrating network conduction [37] and ionic conduction [38], [39]. Work by Wang et al. [36] proposed an interpenetrating network hydrogel in which poly(acrylamide-co-hydroxyethyl methyl acrylate) (P(AAm-co-HEMA)) network endowed the hydrogel system with toughness and interpenetrating polyaniline (PANI) polymer network assumed the conductive function (Fig. 3(g)). Ohm et al. [37] mixed silver flakes into PAAm-alginate hydrogels, and dehydrated them so that the silver flakes formed penetrating networks inside hydrogels to fabricate stretchable and highly conductive hydrogels. Similarly, Tringides et al. [40] mixed CNTs and graphene into alginate to form conductive penetrating networks to enhance the electrical conductivity of alginate hydrogels for electrode array. Since water in the hydrogel occupies the absolute dominant component, water-soluble ions can dissolve in the hydrogel and serve as main conductive component; both sodium chloride [38] and potassium [39] chloride are common ion-conducting components in conductive hydrogels. To obtain much better stretchability and transparency than metal conductors, Keplinger et al. [41] et al. fabricated totally transparent ionically conductive hydrogels with sodium chloride as electrolyte for stretchable actuators that generate strain under high voltage, and can even be used as a transparent loudspeaker (Fig. 3(h)).

III. ADVANCED DESIGN STRATEGIES

Human skin is mechanically flexible and may deform seriously during movements and actions, thus proper mechanics design strategies are needed for endowing skin electronics a certain degree of flexibility and stretchability so as to ensure conformability of the skin interface, stable electrical performance and signal fidelity during usage in daily life. Meanwhile, as skin electronics are directly attached to or integrated on human skin, wearing comfortability is very important, and lots of efforts have also been made by researchers on the comfortability designs of skin electronics.

In this section, we summarized six representative types of mechanics design strategies, which are serpentine structure, fractal pattern, honeycomb structure, trampoline-inspired

structure, wavy pattern, and pre-stretching based 2D/3D structure. Then we summarized three types of comfortability design to enhance the user's experience of skin electronics, which conclude tattoo-based, textile and garment-based, and fiber-based skin electronics.

A. MECHANICS DESIGNS

Serpentine structure, as a kind of structure commonly used in flexible electronics, has been proven to significantly improve the elasticity and stretchability of intrinsically rigid metal interconnections in stretchable electronic devices [42], [43]. Song et al. [44] introduced stacked multilayer network materials as a synergistic platform with serpentine interconnects to integrate and encapsulate electronic devices (Fig. 4(a)). The proposed strategy demonstrates a miniaturized electronic system (11 mm × 10 mm), with the elastic stretchability of ~20% and large area coverage (~110%), which has great potential in compass displays, somatosensory mouse and physiological signal monitoring. Serpentine-based skin electronics is also very essential for infant health clinical care, Chung et al. [45] developed an epidermal electronic system (EES), which can be used for electrocardiogram (ECG) and PPG monitoring (Fig. 4(b)). The electronic layer contains a set of thin, narrow serpentine metal traces that interconnect multiple, chip-scale integrated circuit components. With this structure, the device can bend to smaller radii (6.4 mm and 5mm) than required (>~140 mm and >~50 mm for the chest and foot) without damage. When uniaxial stretching the ECG EES up to 16% and PPG EES up to 13%, the electronics and antenna structure remain below the limit of plastic deformation (~0.3%). And even when device is stretched to 20% strain, the inductance, Q factor and resonant frequency of the antenna vary very little (< 5%).

Fractal pattern refers to the self-similarity of a geometric pattern, complete or approximate resemblance to a part of itself [46]. Especially in mechanics design of skin electronics, such as von Koch curve, Peano curve, Hilbert curve, Moore curve, Vicsek fractal, Greek cross, etc., which can be engineered to accommodate enhanced elastic strains along selected dimensions and support biaxial, radial, and other deformation modes. With "All Vertical" Peano pattern design, silicon nanomembrane can reach a largest deformation with 100% strain and high areal coverage [47] (Fig. 4(c)). Tian et al. [48] fabricated a three-set electrode array, each consists 17 fractal mesh electrodes in Greek-cross patterns, and this fractal mesh electrode suggests an elastic stretchability of 18% (assuming a yield strain of 0.3% for the gold), which is sufficient for straining during natural motion of skin.

Originating from the natural honeycomb of bees, honeycomb structure, which consists of uniformly-distributed hexagonal cells has been extensively studied in soft electronics, because this porous structure has a large surface area to volume ratio and excellent mechanical properties. Recently, Chen et al. [49] combined an organic semiconductor film with a honeycomb structure to develop a high-performance organic electrochemical (OECT) transistor (Fig. 4(d)),

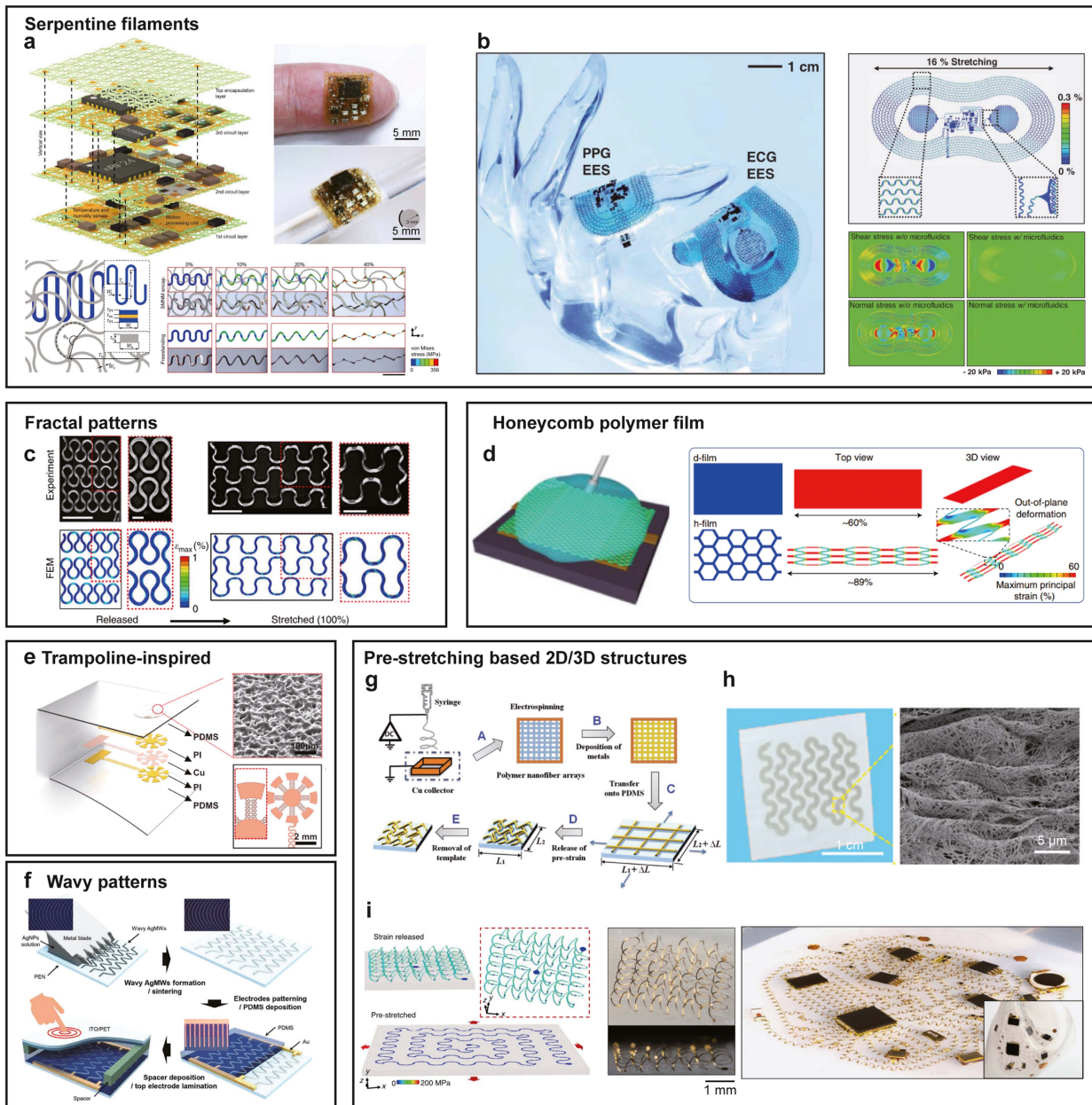


FIGURE 4. Mechanics designs in skin electronics. (a) Stacked multilayer network materials as a synergistic platform with serpentine interconnects. [44] (b) An epidermal electronic system (EES), which can be used for electrocardiogram (ECG) and PPG monitoring. [45] (c) Stretchable silicon nanomembrane using fractal patterns in Peano-curve. [47] (d) Honeycomb-inspired structure in polymer film. [49] (e) Trampoline-inspired structure. [50] (f) Wavy pattern-based structure. [51] (g-i) Pre-stretching based 2D and 3D structure. [52]–[54].

which has a charge transport stability over 30~140% tensile strain, only limited by metal contact fatigue. This honeycomb-semiconductor maintains high ion uptake and stable electrochemical and mechanical properties over 1500 redox cycles with 104 stretching cycles with 30% strain. With stable ECG signal recording cycles and synapse responses under varying strains, it offers a great opportunity for advanced skin electronics.

Trampoline-inspired structure design is a strategy for improving the electrical performance of skin electronics under stretching and out-of-plane deformations. Combined with the

microstructure modification using sandpapers (Fig. 4(e)), the triboelectric nanogenerator (TENG) exhibits great sensitivity of 0.367 mV Pa-1 and accurate unchanged signal outputs even under 35% strain, which shows great potential for self-powered sensor and human-machine interfaces [50].

Wavy pattern is widely used in layout designs for stretchable skin electronics. Kang et al. [51] designed a vibrotactile actuator based on wavy Ag microwire array, which highly promotes vibration intensity and cycling performance because of great areal coverage and improved mechanical stability (Fig. 4(f)). As a result, this vibrotactile actuator can be

adopted on a commercial wearable smart watch directly and exhibits stable performance over 1000 actuating cycles.

When 2D and 3D buckling structures are needed, pre-stretching turns up to be a commonly used method for preparing these special structures. Huang et al. [52] presented an extremely stretchable flexible transparent electrode by transferring metal nanofibers to a pre-stretched PDMS substrate and then release to induce an in-plane sinusoidal wavy structure, which achieved the superior mechanical stretchability with tensile strains up to 300% in Au nanotrrough networks (Fig. 4(g)). The Au nanotrrough networks can be stretched up to 120% repeatedly for 100000 cycles, demonstrating enormous potential as a stretchable flexible transparent electrode in skin electronics. Ma et al. [4] fabricated a liquid-metal fiber mat through pre-stretching method, with this step, the permeability of the mat was activated, and the liquid metal formed a laterally mesh-like and vertically wrinkled structure, which has an ultrahigh stretchability (adapt to omnidirectional stretching over 1800% strain) and electrical stability. With pre-stretching method, 3D structure can also be realized easily. Xu et al. [53] reported a 3D wavy structure through spray printing Ag NWs on pre-stretched multiscale porous polystyreneblock-poly(ethylene-ran-butylene)-block-polystyrene (SEBS), which shows a negligible change of electrical resistance ($\sim 1\%$) at a bending radius of 1 mm and increases $\sim 17\%$ under 100% uniaxial strain (Fig. 4(h)). Jang et al. [54] developed a 3D architecture by releasing the pre-strain to cause the 2D precursor to transform geometrically, through a coordinated collection of in-plane and out-of-plane translational and rotational motions (Fig. 4(i)). With the help of a soft encapsulation strategy that applies an encapsulation material as a liquid precursor at a state of partial release of the substrate prestrain, then crosslinking it into a solid form and completing the release. The elastic stretchability is enhanced ~ 2.2 and 2.8 times for uniaxial and radial stretching compared to the usual case of encapsulation, which can significantly solve the engineering constraints and performance limitations of 2D materials.

B. COMFORTABILITY DESIGNS

As for skin electronics with excellent mechanical properties, when applied to the human body, it is very important to consider its comfortability because it usually directly attaches to the skin. Therefore, there is plenty of research concentrated on comfortability designs to make it thin, flexible, lightweight, breathable, and biocompatible.

Tattoo-like skin electronic has attracted many concentrations due to its properties of ultra-thin, light-weighted enough to be imperceptible and could be seamlessly attached to the skin. Wong et al. [55] reported a tattoo-like triboelectric nanogenerator (TL-TENG) with thickness of tens of micrometers, which can interface with the skin without additional adhesive layers (Fig. 5(a)). The TL-TENG demonstrates the stable performance of energy harvesting with the open-circuit voltage reach to ≈ 180 V and short-circuit current ≈ 2.2 μ A

under constant tapping (≈ 16 kPa). It can also be customized into various tattoo patterns, which indicates great potential in wearable nanogenerators and human-machine interface. Norton et al. [56] developed a fractal mesh geometry of soft foldable electrode, which can be mounted on the complex surface topology of the auricle and the mastoid directly and chronically. It can provide high-fidelity and long-term capture of electroencephalogram (EEG), and can be equipped for two weeks throughout daily activities, such as vigorous exercise, swimming, sleeping, and bathing (Fig. 5(b)).

Integrating skin electronics into clothes is also an ingenious design to make wearable devices portable and breathable. Jiang et al. [57] integrated a nanofiber composite (LPPS-NFC) based hybrid TENG and PENG energy harvester (TPENG) to the top and a PENG to the bottom side of a shoe pad for collecting the energy from walking and running, which has a promising for self-powered textile sensors and wearable electronics (Fig. 5(c)). Textile-based materials can realize great breathability for skin electronics, Liang et al. [58] developed a topology-optimized droplet energy harvesting fabric to obtain stable and efficient output through the falling raindrops (Fig. 5(d)). Based on this self-powered device, a wireless wearable prototype with great flexibility and breathability is successfully demonstrated to monitor the droplet properties, such as temperature, pH, and salinity. Fan et al. [59] fabricated a triboelectric all-textile sensor with high sensing performance (pressure sensitivity of 7.84 mV Pa $^{-1}$, response time of 20 ms, and stability of over 105 cycles). When stitched into a wristband, fingerstall, sock, and chest strap, it can measure the pulse at the neck, wrist, fingertip, and ankle (Fig. 5(e)). In addition, it has a good machine washability of over 40 washes, which is greatly promising for future healthcare monitoring.

Fiber materials are always used to make wearable electronics due to their stretchability and porosity. Zhuang et al. [60] reported a liquid metal (LM)-superlyophilic and stretchable fibrous thin-film scaffold fabricated by electrospinning of SBS, which shows high conductivity of 155900 S cm $^{-1}$ and a marginal resistance change of only 2.5-fold at 2500% strain (Fig. 5(f)). This fiber can be patterned with printing technologies for wearable device design. Xiao et al. [61] developed a recharged solid-state Zn/MnO $_2$ fiber battery with over 500 hours cycle performance and maintaining 98.0% capacity after more than 1000 charging/recharging cycles, which can power sensors to realize healthcare monitoring, including heart rate, temperature, humidity, and altitude monitoring (Fig. 5(g)).

IV. APPLICATIONS

A. HEALTHCARE & E-MEDICINE

The wearable and flexible electronic devices are crucial for accurate, continuous and long-termed healthcare and e-medicine, which can cozily cover the human epidermis acting like a portable and intelligent health examination center



FIGURE 5. Comfortability designs in skin electronics. (a) Tattoo-like ultrathin triboelectric nanogenerators (TL-TENG) [55]. (b) Ultrathin electroencephalography (EEG) electrodes as brain-computer interface [56]. (c) Nanofiber composite based energy harvester of shoepad for collecting energy from walk and running [57]. (d) Textile-based skin electronics as energy harvesting through raindrops [58]. (e) Textile-based triboelectric sensor with good washability [59]. (f) Liquid metal - superlyophilic and stretchable fiber based material for wearable device design [60]. (g) Fiber-based Zn/MnO₂ battery with high cycling stability [61].

without the limitation of time and space. Skin is an important source to transmit and generate physiological signals.

1) ELECTROPHYSIOLOGY SIGNAL

Electrophysiology signals are the measurement of voltage changes or electric current or modification covering a wide range of scales, from single ion channel proteins to vast organs like the brain. On-skin electrophysiology monitoring techniques mainly involve electrocardiology (ECG), which gives comprehensive information about heart illness from the ventricles and atria for the study of arrhythmia, atrial fibrillation and other cardiac disorders; Electromyography (EMG), which analyzes and collect the electrical signals produced by the skeletal muscles for the analysis of abnormalities, motor unit recruitment and biomechanics of human movements;

Electroencephalography (EEG), which records electrogram of the electrical activity on the scalp representing the macroscopic activity of the brain's surface layer beneath. Electrooculography (EOG) is a technique for measuring the standing potential of the corneo-retinal between the front and the back of the human eye.

In order to record physiological electric signals from different channels as many as possible and achieve long-termed and continuous MRI-compatible monitoring, Tian et al. [48] propose the manufacturing method of a large-area electronic interfaces and its applications on prosthetic control and cognitive monitoring (Fig. 6(a)). As for the detection of EMG signal, large-area, body-scale epidermal systems can offer comprehensive EMG recording capabilities across various muscle groups, which enables more precise control of

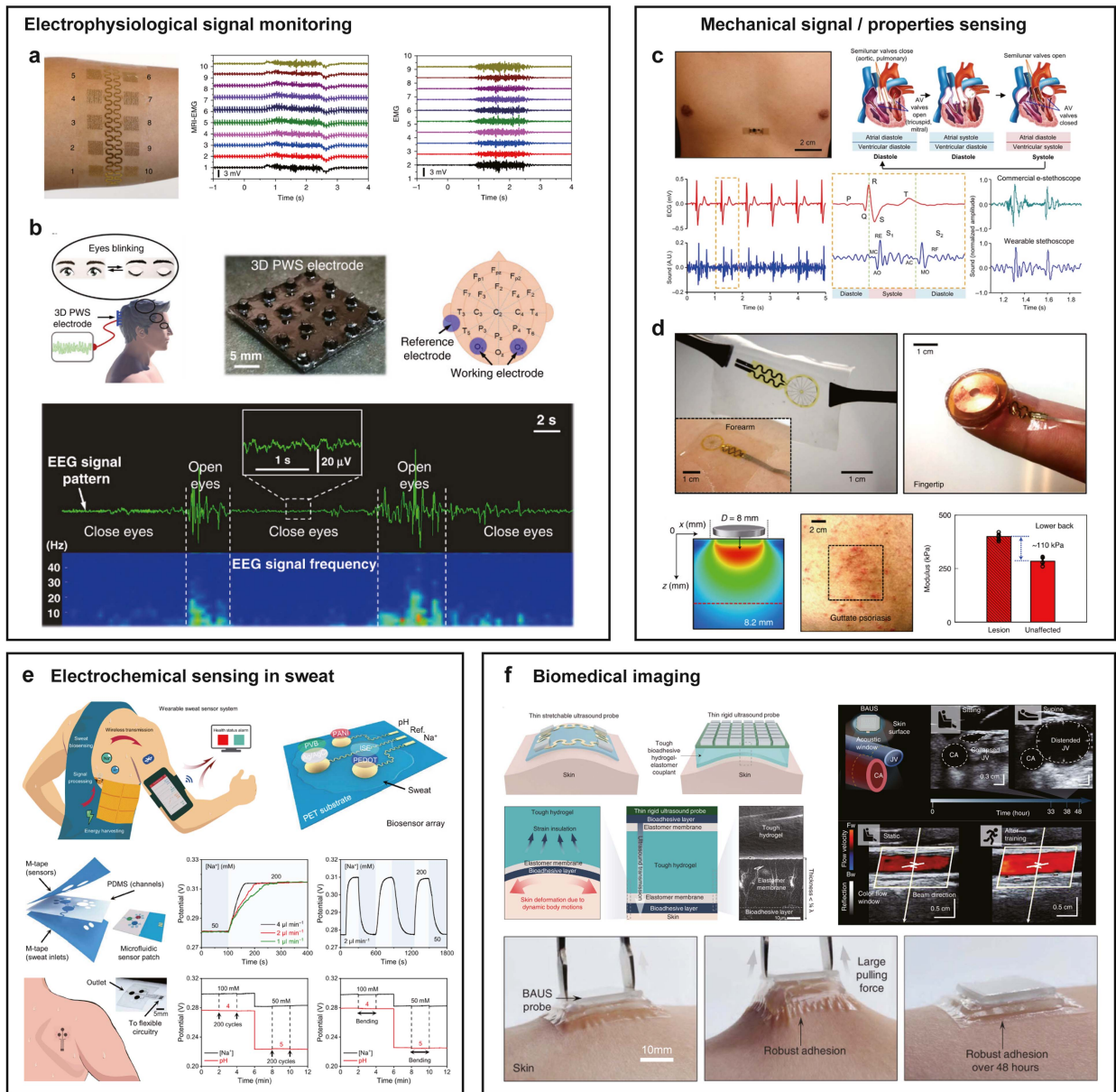


FIGURE 6. Applications of skin electronics in healthcare. (a) MRI-compatible epidermal electronic interface for multichannel EMG measurements [48]. (b) A self-adhesive, stretchable and intrinsically conductive polymer dry electrode for EEG sensing, where alpha wave were detected when eyes closed [62]. (c) Mechano-acoustic sensing patch for detecting physiological process and body motions [70]. (d) Miniaturized electromechanical devices for the monitoring of psoriasis lesions via the biomechanics sensing of deep tissue [74]. (e) A wearable wireless, battery free sweat sensor for monitoring Na^+ and pH, which is self-powered by body motion-driven TENG [76]. (f) A bioadhesive ultrasound device for 48 hours continuous imaging of living diverse organs [83].

multifunction prostheses. Fig. 6(a) shows the epidermal EMG array mounted on the right arm of the subject located in an MRI scanner, by which EMG signals of ten channels can be fully recorded respectively from one trial and after magnetic resonance artefact correction.

EEG acquisition usually requires multiple electrodes placing on the scalp to monitor epilepsy, sleeping state, detecting evoked potentials for brain-computer interfaces (BCI) and so on. However, commercial conductive gels-based EEG electrode becomes invalid easily as the water loses

quickly. Although the dry contact electrode is more durable and conductive, its application is also subject to limited stretchable and adhesive capacities. In this way, Zhang et al. [62] proposed a stretchable, self-adhesive and fully-organic dry electrode with unique fabrication processing of biocompatible blends of poly(ethylenedioxythiophene):poly(styrenesulfonate), waterborne polyurethane (WPU) and D-sorbitol. As Fig. 6(b) shown, multiple EEG electrodes are arranged to cover the scalp of the subject to collect the EEG signal during

eye-blinking, where alpha wave could be detected during eyes closed and a broader frequency range detected during eyes opened. Zulqarnain et al. proposed wireless and wearable ECG sensing device made of amorphous Indium Gallium Zinc oxide thin film transistors via NFC RF communications [63]. Kim et al. presented a reusable, multichannel, surface EMG sensor array that covers multiple muscles over sufficiently large areas, with compliant designs that provide varying degrees of stiffness for repetitive uses [64]. Ameri et al. proposed an undetectable EOG sensor system based on transparent, breathable, ultrathin, ultrasoft, and non-invasive graphene electronic tattoos (GET). The GET EOG can capture high-precision EOG with an angular resolution of 4° of eye movement, and eye movement can be correctly interpreted [65].

2) MECHANICAL SIGNALS

Mechanical signals generally refer to passive and active epidermal mechanical vibration signals [66]. Passive mechanical signals involve physiological mechanical signals produced by human tissues, such as cardiac rhythm, diastolic of skeletal muscles, gastrointestinal tract and so on. Electronic stethoscopes and accelerometers are the two effective and frequently-used methods to detect passive mechanical signals. The electronic stethoscopes can translate the vibration vibrations of their diaphragm into electric signals by piezoelectric [67] or triboelectric [68] materials. Unlike the ultra-thin and weak structure of electronic stethoscopes [69], the accelerometers have strong and durable designs. During vibration periods, the accelerometers can convert the inertia-based deformation of piezoelectric material into an electric signal. For example, Liu et al. [70] introduced a mechano-acoustic-electrophysiological sensing platform incorporating high-bandwidth triaxial accelerometers for both cardiovascular disease treating and human-machine interfaces (Fig. 6(c)). This platform records the mechano-acoustic responses to value the operation of heart like structural defects in heart valves, rather than appearing in the ECG traces directly. Simultaneous ECG and heart sound measurements matched up well where four auscultation sites (aortic, pulmonary, tricuspid, mitral) for heart murmurs can be distinguished at the magnified plot. What's more, mechanical sensors attached to the skin can capture the mechanical signal associated with body motion and other physiological processes. Lee et al. introduced a soft, wireless mechano-acoustic sensing device located at the suprasternal notch [71]. The different rotation angles of the head against the torso for body-orientation calibration test at the application of distinguishing four sleep stages can be sensed. This specific demonstration in various actual scenarios demonstrates the ability to respiration, vocal utterance, gait recognition and other healthcare applications.

However, some human tissues like skin are unable to vibrate. Active mechanical signals generally refer to calculating the biomechanics of deep tissues by loading external pressure via mechanical vibration actuator and detecting

deformation in tissue via piezoelectric and electromagnetic sensor, which are normally used to monitor and diagnose tissue lesions from corresponding skin diseases [72]. Degdeviren et al. presented an ultrathin and stretchable mechanical sensing device consisting of mechanical actuators and sensors made of nanoribbons of PZT, which can apply to measure the viscoelasticity in the near-surface areas of the epidermis. But most of these methods only concern about the superficial depth of epidermal skin tissues [73]. In this way, Song et al. put forward an miniaturized electromechanical devices used to accurately measure modulus of deep tissue (~ 8 mm) for locating lesions associated with psoriasis [74]. Fig. 6(d) shows the module measurement of guttate psoriasis based on the principle of quasi-static, tensile stress-strain responses via magnetic electromechanical modulus sensor. Furthermore, multiple sensors can be configured into arrays which indicates its potential application in wounds of arbitrary shapes.

3) SWEAT SENSORS

Sweat contains many important chemical components as physiological indicators such as sweat glucose, pH and other metabolites and nutrients. The flexible and wearable sweat sensor could help monitor relevant physiochemical parameters and the risk alarms for an accidental physiological failure during sweating after strenuous exercise. Bandonkar et al. proposed a wireless, battery-free and skin-interfaced electronic sweat sensor integrating with colorimetric reagents on the single-use microfluidic platform, which provided real-time sensing of chloride, lactate and glucose after strenuous exercise [75]. What's more, Song et al. proposed a technology of triboelectric nanogenerators (TENGs) based sweat sensor, which can detect dynamically monitor key sweat biomarkers with continuous power supply created from human motion (Fig. 6(e)) [76]. However, the variety of physiochemical parameters is limited to the restricted methods beyond ion-selective and enzymatic electrodes or direct oxidation of electroactive molecules. As the application scenarios of sweat collection are also limited by the volume of sweat, most sweat sensors are only applied after strenuous exercise. In this way, Wang et al. presented a new method of wearable electrochemical biosensor based on the unique in situ regeneration and calibration technologies, which enables the sensitive, selective and continuous monitoring of a wide range of trace-level biomarkers including all nine essential amino acids as well as vitamins, metabolites and lipids commonly found in human sweat [77]. Additionally, the continuous and sufficient sweat collection can be achieved by the new strategies of the carbochol iontophoresis-based sweat induction and microfluidic-based surrounding sweat sampling.

4) IMAGING

Biomedical imaging technologies such as magnetic resonance imaging (MRI), X-rays, computed tomography (CT) and electrical impedance tomography (EIT) are typically used to image tissue deep into body, which are for locating the lesions

and diagnosing multiple chronic diseases. But these imaging devices are normally heavy and expensive, which only provide a short period of tissue imaging. To overcome this disadvantage, the wearable sensing based on variable methods of optical [45], thermal [78], chemical [79], terahertz [80], radio frequency mechanisms [81] are proposed, which penetration depth or spatial resolution are unsatisfactory for human tissue imaging. As the ultrasound has significant penetration depth and proven biocompatibility in human tissues, ultrasound-based wearable and flexible imaging sensor deserve attention. Wang et al. put forward the stretchable ultrasonic phased arrays which can achieve continuous monitoring of deep-tissue hemodynamics [82]. The stretchable ultrasonic device eliminated the air gap between traditional ultrasonic probe and skin surface. However, the short continuous imaging duration and the imaging stability disturbed by the displacement between device and skin are still the technology barriers to the development of wearable ultrasonic imaging. Wang et al. described a bioadhesive ultrasound device based on a specific bioadhesive hydrogel-elastomer hybrid, which provides 48 hours of continuous imaging of diverse internal organs, including blood vessels, muscle, heart, gastrointestinal tract, diaphragm, and lung [83]. Fig. 6(f) shows the robust adhesion between the bioadhesive ultrasound device and the skin. This device can be used to observe the hemodynamic changes in the body, such as the diameter of the jugular vein in the subject increasing from the sitting or standing position to the supine position, and the blood flow rate dramatically increasing after 0.5 hours of physical exercise.

B. HUMAN-MACHINE INTERFACE

Human-Machine Interface (HMI) is a user interface or method that a user can interact with a machine through various bionic signal such as motions, touches and so on. Recent developments in electronics, materials, and mechanical designs have offered more possibilities for wearable HMI devices.

1) TACTILE FEEDBACK

Tactile feedback mainly refers to provide users with perceptible tactile information by directly stimulating the skin via mechanical, pneumatic, electrical or thermal methods. Various applications have been explored, which integrates with robot hand, touch screen and other feedback systems. Yu et al. proposed a wireless and battery-free haptic interface cooperating with virtual reality (VR) and augmented reality (AR), which used tactile feedback to simulate the virtual world in addition to visual and audial feedback, which makes VR experience more immersive [84]. Mechanical-based haptic interface induces tactile sensation on the skin via vibration from electrical motors, electromagnetic actuators or piezoelectric devices. Fig. 7(a) presents the applications of online social communication and robot hand control based on the skin-integrated haptic interface. What's more, Jung et al. put forward a new strategy for haptic interface with the promotion of light weight and array precision to improve the strength

and power efficiency of haptic engagement [85]. An amputee volunteer can adjust the force of robot hand to grasp an eggshell via 30 pressure sensors on the hand and haptic feedback interface on the upper arm. Additionally, Sun et al. [86] proposed augmented tactile-perception and haptic-feedback rings (ATH-Rings) for human-machine interface based on VR devices, which achieved finger bending sensing by TENG sensor and temperature detecting by flexible pyroelectric sensors with vibration and heating haptic feedback respectively from eccentric rotating mass vibrators and nichrome metal wires (Fig. 6(b)). ATH-Rings with comfortable, self-powered and low voltage-driving designs realized tactile feedback based on the advanced internet of things (IoT) portable platform.

2) TACTILE SENSING

Among variable kinds of HMI technologies, tactile sensing is one of the most fundamental mutual signal transmissions to achieve communication and control between human and machine. Tactile sensing normally refers to processing contact or physical pressure signals via variable strategies such as resistive [87], capacitive [88], piezoelectric [89], triboelectric [90] and optical [91] technologies. Zhu et al. proposed a skin-electrode mechanosensing structure which consists of two electrodes and the skin [92]. The skin acts as an ionic interface where electrons and ions are separated by tiny voltage without occurring chemical reactions. As the capacitance of skin is in direct proportion to the contact area of skin-electrode interface, the mechanosensing structure can detect the minor changes in the contact area and quantify in the form of capacitance value. Fig. 7(c) demonstrates the working principle of the skin-electrode mechanosensing structure. Textile-based skin-electrode mechanosensing structure enables cost-effective and convenient wearables for tactile sensing. As Fig. 7(c) shows, the smart glove indicates pressure distributions of grasping balloon and beaker separately.

Human skin perceives tactile stimuli through skin mechanical receptors including slow adaptive (SA) and fast adaptive (FA), where separately responds to the static pressure and the high-frequency dynamic pressure. However, the strategies of tactile sensing mostly realize one of slow adaptive and fast adaptive. Chonet al. raise an artificial neural tactile sensing system consisting of sensors that simultaneously mimic FA and SA mechanoreceptors and a neural stimulator that generates sensory neuron-like signals which mimicked the real neural signals activated from the afferent nerves [92]. Robot equipped with this artificial neural tactile sensing system can be used for surface texture recognition based on a deep learning technique that classify twenty fabrics with different textures.

3) GESTURE SENSING

With the development of artificial intelligence, gesture sensing become a hot research object. Wearable and flexible

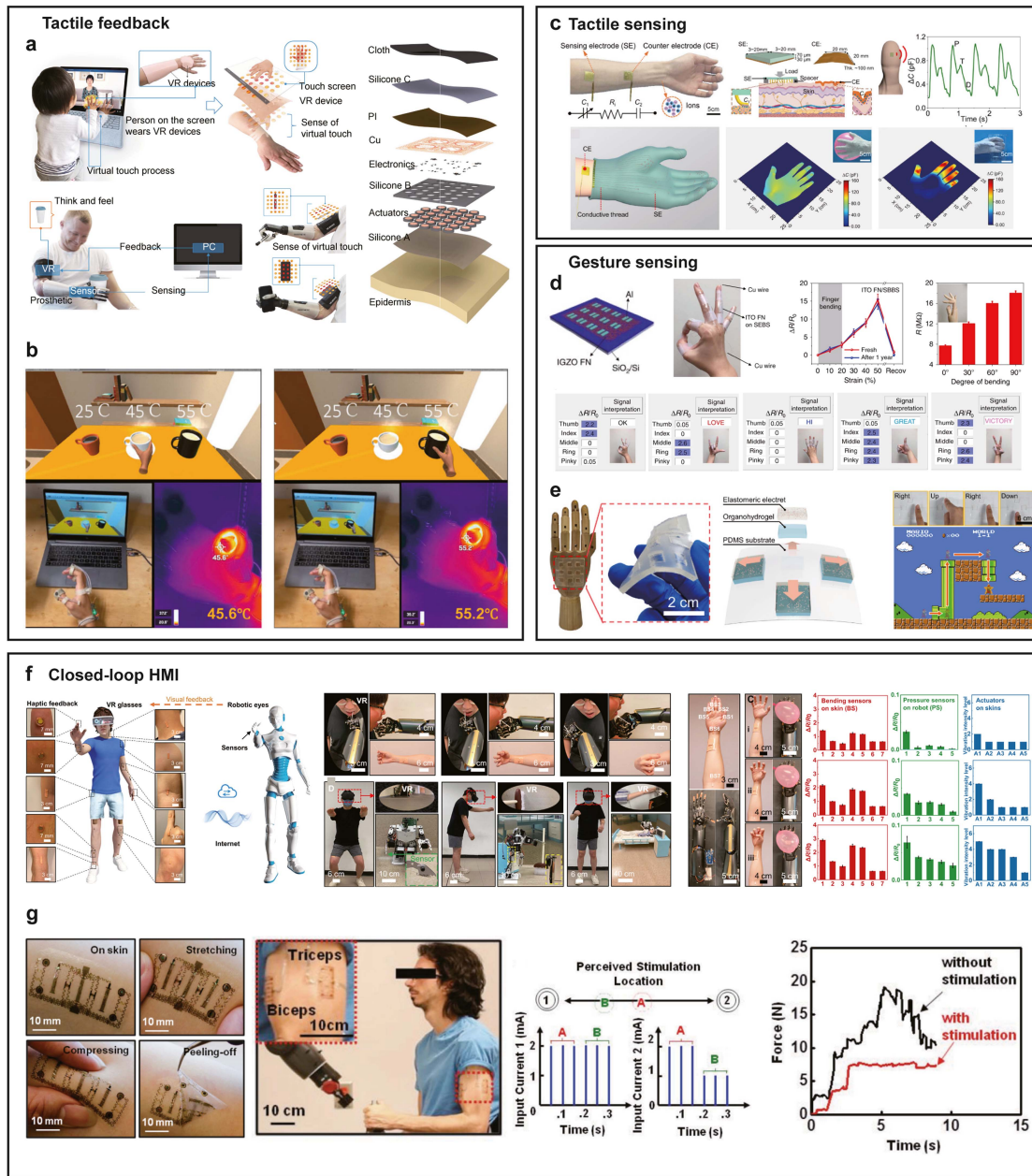


FIGURE 7. Applications of skin electronics in human-machine interface. (a) A skin-integrated wireless haptic interfaces establishing feedback for social communication and control of active prostheses [84]. (b) An augmented tactile feedback ring that enables thermo-haptic feedback in VR.[86]. (c) A skin-electrode iontronic interface for tactile sensing, which can be applied to the smart glove for pressure indication of balloon and beaker [92]. (d) A flexible and stretchable electronic device based on metal oxide nanofiber networks for gesture sensing [93]. (e) A bioinspired soft electroreceptor for untouched somatosensation, which achieves the gesture distinguishing for movement control of game characters [94]. (f) A wireless human-machine interface for closed-loop control between robot and human based on robotic VR [95]. (g) An epidermal electronic platform for EMG sensing and electrostimulation, which enables closed-loop control of the robotic arm to grip a bottle precisely [97].

electronics for precise gesture recognition are desirable for lightweight and mechanically stretchable functions. Wang et al. raised a preparation method of flexible and stretchable metal oxide nanofiber networks for multimodal and monolithically integrated wearable electronics [93] (Fig. 7(d)). Hydrothermally derived oxide nanowires have been explored via the blow-spinning method. The resistivity change of devices made from metal oxide nanofiber represents different

bending angles of finger fitted with the devices, which enables to translate hand gestures. Furthermore, Guo et al. [94] presented bio-inspired soft electroreceptors for artificial precontact somatosensation, which is capable of sensing approaching targets without contact by detecting the charges that they carry naturally. Fig. 7(e) shows the electroreceptor matrix and its application in playing Super Mario using touchless control pad with four function keys.

4) CLOSED-LOOP HMI

To realize a closer teaming between humans and robots, engagement, comprehension, and communication are required. Wearable devices make it possible to construct a closed-loop control, sensing and feedback HMI system, where information sensed by robotics could be felt by users. Liu et al. presented a closed-loop HMI system based on skin-integrated electronics that complies with the whole body to provide wireless motion capturing and haptic feedback via Bluetooth, Wireless Fidelity (Wi-Fi), and Internet [95]. Fig. 7(f) shows the closed-loop HMI which is applied to saliva sample collection by nasopharyngeal swabs and difficult sport tasks such as coughing, room cleaning, and patient care. What's more, Gu et al. proposed a soft neuroprosthetic hand providing simultaneous myoelectric control and tactile feedback [96]. It is driven by pneumatic actuation, controlled by EMG signals and provides electrotactile feedback on the residual limb. The soft neuroprosthetic hand has a better performance than neuroprosthetic hand in speed and dexterity. A person with a transradial amputation can use the soft neuroprosthetic hand to recreate closed-loop control and primitive touch in tests with blindfolds and acoustical isolation. Additionally, Xu et al. [97] presented an HMI system applying the tactile feedback of electric stimulation with multiple sensors of electromyography (EMG), temperature and strain, which is constructed from patterned metal traces and polymer dielectric materials on a thin layer of silicone elastomer. As Fig. 7(g) shown, the subject can control the gripping motion of robotic hand by EMG signal, where the gripping force can be adjusted from the electric stimulation based on the force transducer on the robotic hand.

V. CONCLUSION

The emerging of skin electronics makes it possible for people to wear all kinds of healthcare monitoring or therapy devices, and also smart communication or entertainment devices directly on the skin, lightens the burden of carrying bulky or tethering devices, and also makes it simpler to acquire signals, harvest energy and deliver stimulations to/from the skin. Undoubtedly it sheds some light on the style and platform of next generation of ubiquitous electronics equipment in the near future. However, the difficulties in efficiency, stability, reusability, mass manufacturing and cost may temporarily hinder it from successful commercialization and universal use in the modern society. To overcome these hurdles, researchers made great efforts to innovations of the functions, improvements of the performances, and maturation of fabrication process in developing better skin electronics.

A systematic developing process of a complete research work in skin electronics may start from exploiting novel functional materials, then finding a proper design strategy to adopt this novel material into a specific application scenario to achieve better performance, or it could also start from noticing the urgent need in a specific situation, and then try to find a suitable design strategy or materials options to construct a

device or system that fulfills this need. These are two completely different lines of thought of researchers, but they both helped the development of skin electronics. Since skin electronics involves multidiscipline efforts including materials science and engineering, mechanical engineering, electrical engineering, computational science, clinical medicine and even industrial or production designs, the seamless cooperation among researchers in different fields becomes especially essential.

Here we provided representative examples and organized them in dimensions of materials, designs and applications for the reference of developing novel skin electronics. However, due to the length limitation we cannot include all of them. There are many other materials types worth mentioning, such as semiconductors, dielectric elastomers, etc., and other designs such as antenna designs for wireless communications, other applications such as wearable display, ambient or motion energy harvesting, etc. We believed that although challenges still remain, with the rapid development of the engineering and fabrication techniques, and with the great need for wearable multifunctional personal equipment, skin electronics could become mature products in consumer markets just like mobile phones do, which will definitely change our lifestyle, makes it more convenient, instant and seamlessly connected for no matter point-of-care, entertainment or communication.

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