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Development of soft dry electrodes: from materials to structure design

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Abstract

Bioelectric signals reflect our daily physiological activities, which can be recorded in the form of electroencephalography, electrocardiography, electromyography, *etc.* The traditional Ag/AgCl wet electrode is the gold standard for clinical monitoring of bioelectrical signals at present, while complicated preparation and gel evaporation limit its long-term application. Therefore, it is meaningful to research dry electrodes without conductive paste or additional adhesives. Unfortunately, the high interface impedance between electrodes and skin is a fatal defect of dry electrodes, which leads to excessive noise levels and poor signal quality. Consequently, more efforts are required to achieve conformal contact between dry electrodes and skin to reduce the contact impedance. From this perspective, we review the recent progress in capacitive electrodes, invasive microneedle electrodes, and common-contact dry electrodes. Material selection and structural design to obtain conformal contact are highlighted. Finally, we propose the future development direction of dry electrodes.

Keywords: Dry electrode, conformal contact, electrode-skin impedance, bioelectric signal

INTRODUCTION

Extensive and complex electrical signals in organisms are the reflection of their normal physiological activities. The common bioelectric signals collected in the clinic mainly include electrocardiography (ECG), which is closely related to cardiac diseases such as arrhythmia and myocardial infarction, electroencephalography (EEG), which is closely related to brain diseases such as Parkinson's disease and stroke, and electromyography (EMG), which is closely related to muscle atrophy[1]. Dry electrodes, after years of painstaking investigation, have emerged as crucial tools for monitoring bioelectric signals[2-4]. They have been widely applied in disease prevention and diagnosis[5,6], healthcare[7-9], and human-machine interfaces[10,11].

Using dry electrodes can avoid the disadvantages of the traditional Ag/AgCl wet electrodes. For instance, the usage of wet electrodes involves complex preparation work, including hair removal and grinding the horny layer[12]. Also, evaporation of the liquid phase of wet electrodes can lead to a severe loss of signal. However, unlike wet electrodes, dry electrodes are difficult to form conformal contact with the skin due to the lack of sufficient electrolytes. This leads to high interface impedance (more than several hundreds of kΩ[13,14]), in sharp contrast with wet electrodes (a few kΩ)[15]. Besides that, improper contact will also make dry electrodes slide inevitably on the skin, causing instability in monitoring electrophysiological signals. Therefore, achieving conformal contact between dry electrodes and skin is of vital importance.

Currently, dry electrodes can be divided into the following three types according to the contact between dry electrodes and skin [Figure 1]: capacitive electrodes (non-contacted dry electrodes), invasive microneedle electrodes, and common-contact dry electrodes[16,17]. Since interface contact affects impedance and plays a decisive role in the detection performance of electrodes, this review focuses on the latest research progress in the achievement of conformal contact for three types of dry electrodes. Three types of common-contact dry electrodes, namely self-adhesive dry electrodes, ultra-thin dry electrodes, and micro-structured dry electrodes, are elaborated in terms of material selection and structural design. Then, potential challenges and future development of dry electrodes towards practical application through bioelectric signals monitoring are discussed.

CAPACITIVE ELECTRODES

The capacitive electrode can collect signals without contacting the skin, which is similar to a capacitor and directly coupled with the skin, so it can avoid damaging the skin and measure electrophysiological signals in areas with more hair or through multiple layers of clothing[18]. However, considering the influence of coupling impedance and multi-layer insulation, conformal contact between dry electrodes and skin is not possible. Additionally, extremely high interface resistance and incomplete fixation are major obstacles for capacitive dry electrodes, leading to serious motion artifacts and noise[16]. Hence, the development of the capacitive electrode in conformal contact is actually limited, and it is not suitable for bioelectric signal monitoring, especially for dynamic monitoring.

INVASIVE MICRONEEDLE ELECTRODES

It is an important feature of the invasive microneedle electrode to use microneedles to pierce the horny layer and directly contact the growth layer of human skin in the process of monitoring electrical signals. Penetrating the horny layer formed by dead cells obtains superb conformal contact, avoiding unstable signal recording due to skin stretching or electrode movement. This method does not injure internal tissues and eliminates the high impedance of the horny layer[19,20]. Invasive microneedle electrodes can be roughly divided into dry electrodes with rigid microneedle arrays (silicon microneedle array and metal microneedle array) and dry electrodes with flexible microneedle arrays (polymer microneedle array)[17]. At present,

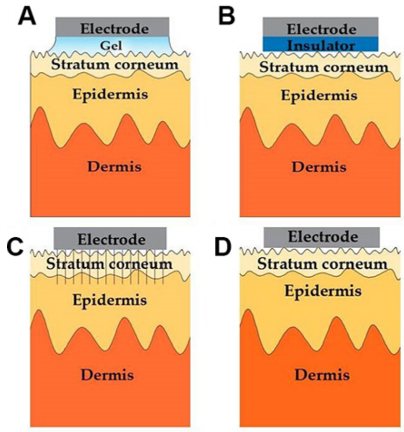

Figure 1. (A-D) Schematic diagram of contact between electrodes (the wet electrode, the capacitive electrode, the invasive microneedle electrode, and the common-contact dry electrode, respectively) and skin[16]. Reprinted with permission. Copyright 2021, MDPI.

silicon microneedles are fragile, so their development is limited. Although the metal microneedle has fine toughness, its rigid substrate is difficult to contact skin conformably. Polymer microneedles have good biocompatibility, but a few polymers with lower modulus are challenging to penetrate the horny layer. In addition, for human beings, the skin is the largest organ. Tough and soft skin has certain stretchability and elasticity[21]. As a result, owing to actualizing conformal contact between electrodes and skin, the best way is to develop a dry electrode that matches the Young's modulus of the skin. This can be achieved by using an invasive microneedle electrode with a soft substrate (lower modulus) and hard microneedle (higher modulus)[22,23]. The flexible substrate can comply with skin curvature to attain conformal contact, and the rigid microneedle can allow itself to pierce the horny layer smoothly at the same time.

Li *et al.* reported a dry electrode based on the polyimide microneedle array (PI-MNA) [Figure 2A]. Ti was used as an adhesive layer, and Au was used as a conductive layer, both of which were deposited onto the surface of PI-MNA by magnetron sputtering. The flexible substrate (thickness of about 50 μm) and solid microneedles (diameter less than 10 μm) of the electrode met the development of emerging invasive microneedle electrodes. Furthermore, the normalized electrode-skin contact impedance of PI-MNA electrodes had not only hit all-time lows (approximately 1/250 of that of standard electrodes), but the cost of $0.35 per electrode ensured industrial preparation of large quantities[24]. When ambient temperature or body temperature rises, sweat inevitably is produced on human skin, resulting in the accumulation of liquid at the interface of conformal contact. This not only affects signal output but also triggers skin allergies, so it is essential to ensure the permeability of dry electrodes. Hou *et al.* fabricated a dry electrode with microneedles at the top and Miura-ori structured substrate by polydimethylsiloxane (PDMS) molding process using a high-precision machining direct molding mold [Figure 2B]. The Miura-ori, which originated from origami, was a typical flat-folded mosaic structure. During the smooth unfolding of the origami, the parallelogram, as a component unit, remained flat and unbent. The Miura-ori structure at the bottom of the electrode not only had air circulation channels to help sweat escape but also had two-directional in-plane bendability (bending radius of 1 cm) to accommodate the curvature of skin. Compared to wet electrodes, this electrode showed an improvement in EMG signal quality (signal-to-noise ratio is 15.24 dB; conversely, the wet electrode has a signal-to-noise ratio of 7.79 dB). However, the preparation process of invasive microneedle electrodes with a Miura-ori structure was more complex. The complex structure made the homogeneity of metal plating poor[25]. Nowadays, invasive microneedle electrodes mostly adopt vertical microneedles.

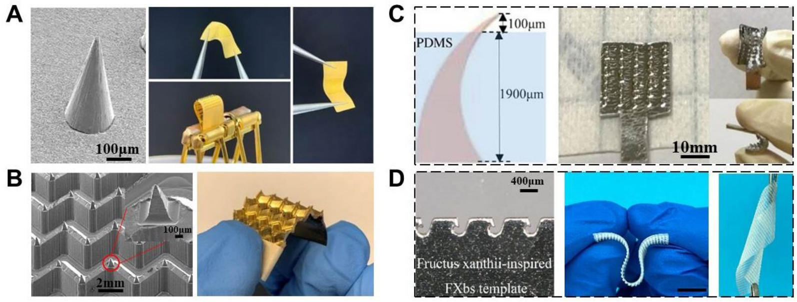

Figure 2. Invasive microneedle electrodes. (A) SEM image of the polyimide microneedle array electrode and its sample diagram[24]. Reprinted with permission. Copyright 2022, Springer Nature. (B) SEM image of the Miura-ori structured electrode (illustration inside is an enlarged SEM image of a single microneedle) and its sample diagram[25]. Reprinted with permission. Copyright 2021, Springer Nature. (C) Size and shape drawing of the hook electrode and its sample diagram[26]. Reprinted with permission. Copyright 2020, Elsevier. (D) SEM image of the barbed electrode and its physical picture[27]. Reprinted with permission. Copyright 2022, American Chemical Society. SEM: Scanning Electron Microscope.

Interestingly, inspired by the thorn of golden margined century plant leaf, Li *et al.* successfully prepared the dry electrode with hook-type microneedles [Figure 2C]. The microneedle structure was fabricated by laser milling means and then embedded inside the flexible PDMS substrate. The hook-type microneedle had a smaller penetration force (0.939 N) but a larger pullout force (0.149 N), so it was laborious to break and detach[26]. Besides that, Niu *et al*. proposed a new stacking template technique for the first time to rapidly prepare Ag/AgCl thermoplastic polyurethane (TPU) electrodes [Figure 2D]. They interleaved and fixed the bent pins, which eventually became a multi-stacked stencil. TPU was poured into the templates for curing, and silver conductive layers were deposited by chemical silver plating. The electrode had a structure similar to the inverted thorn of Xanthium and an excellent dynamic friction coefficient (about 38.8% higher than that of a flat plate structure). Accordingly, this electrode showed a high signal-to-noise ratio in dynamic signal acquisition (32.27, 30.13, and 27.77 dB when the arm was swung at 30, 60, and 90; conversely, the signal-to-noise ratio of the wet electrode was 31.89, 26.73, and 21.92 dB)[27]. In fact, the hooked curved electrode does provide better conformal contact and lower contact impedance compared to the upright microneedle electrode. However, the curved needle that tightly binds to the skin is difficult to pull out after testing and is prone to cause greater skin damage, which limits its practical application.

For invasive microneedle electrodes, softness is an important prerequisite for their further development in the field of conformal contact. Flexible microneedle electrodes can adapt to skin profiles to obtain lower signal-to-noise ratios in dynamic measurements. Nevertheless, as the manufacturing engineering of invasive microneedle electrodes becomes increasingly mature, researchers gradually discover that microneedles of any material all have a sharp structure, bringing discomfort and infection. This is also the challenge that limits the practical application of invasive microneedle electrodes.

COMMON-CONTACT DRY ELECTRODES

The common-contact dry electrode means that it can directly contact skin but does not pierce the horny layer. Compared with the invasive microneedle electrode, it cannot completely adapt to the rough skin surface, thus resulting in high and unstable interface impedance[17]. In order to gain conformal contact, scientists improved traditional dry electrodes and invented the following types of the common-contact dry electrode, including self-adhesive dry electrodes, ultra-thin dry electrodes, and micro-structured dry electrodes.

Self-adhesive dry electrodes

To achieve conformal contact between dry electrodes and skin, it is an effective approach to endow dry electrodes with self-adhesive characteristics. Poly (3, 4-ethylenedioxythiophene): poly (styrene sulfonate) (PEDOT:PSS) is a polymer material with the advantages of electrical conductivity, transparency, and environmental stability, which is often used in the field of flexible electronics. From Ouyang's work, a stretchable and self-adhesive dry electrode (PWS) was reported via solution processing of the biocompatible mixture of PEDOT:PSS, waterborne polyurethane (WPU), and D-sorbitol [Figure 3A]. Intrinsically conductive PEDOT:PSS did not have good elasticity and adhesion. O-H, N-H, and C=O groups derived °C from WPU and D-sorbitol were able to form strong physical adsorption (hydrogen bonding) with keratin and lipids on the skin. Naturally, the PWS electrode exhibited lower contact impedance (82 $K\Omega cm^2$ at 10 Hz) than the Ag/AgCl electrode (148 $K\Omega cm^2$ at 10 Hz), which could accurately record high-quality ECG signals (peak-to-peak voltage of 1.84 mV for QRS complex)[28]. Although the self-adhesive dry electrode is capable of detecting high-quality signals in motion, its adhesion is greatly deteriorated by sweat. Consequently, future research should focus on dry electrodes with high self-adhesive properties for wet skin. Cao *et al.* proposed a self-adhesive and highly sweat-resistant dry electrode (PPT) by drop-casting aqueous solution. The electrode was the blends of PEDOT:PSS, polyvinyl alcohol (PVA), tannic acid (TA), and ethylene glycol (EG) [Figure 3B]. Fundamentally, the increase in conductivity (122 Scm^{-1}) attributed to EG was able to induce secondary doping of PEDOT:PSS. As a soft polymer, PVA could disperse energy and improve the breaking elongation (54%) of the electrode. The hydroxyl group and hydrophobic benzene ring of TA could strongly increase the adhesion of the PPT electrode in dry and clammy environments (0.32 and 0.28 Ncm^{-1}, respectively)[29]. Similarly, Park *et al.* chose PEDOT:PSS as the conductive material to make a degradable and environment-friendly dry electrode (PDC) by blending the component solutions [Figure 3C]. It was worth mentioning that they added dopamine and hydroxyethyl cellulose (HEC) into the PDC film. The amine group of dopamine formed a hydrogen bond and electrostatic interaction with the hydroxyl group on HEC, giving the PDC electrode excellent tensile properties (only 8% of resistance change under 110 of fully flat-bended cycles). More importantly, HEC could be dissolved in water, so this electrode was able to be degraded under 6 °C distilled water immersion or washing[30]. In fact, the degradable property enables the post-processing of electrodes and increases the sustainability of the product, which has profound application value.

Apart from PEDOT:PSS[31-33], elastomers that inherently possess viscous properties are often used as substrate materials for self-adhesive dry electrodes. However, poor conductivity is a fatal defect of elastomers. Therefore, enhancing the electrical conductivity of elastomers is the key to their wide application in self-adhesive dry electrodes. Generally speaking, the common method to elevate the conductivity of elastomers is to add conductive fillers, such as metal nanoparticles, metal nanowires, and carbon nanotubes. Nevertheless, the traditional conductive fillers will not maintain a conductive network under strain, resulting in a rapid decline in conductivity. In recent years, liquid metals (LMs) with excellent conductivity and ductility have gradually come into people's view. Moreover, LMs with tensile properties matching elastomers enable them to overcome the drawback of conventional fillers[34-36]. Pei *et al.* applied LMs as fillers to endow viscoelastic elastomer with high tensile conductivity [Figure 3D]. EGaIn droplets were uniformly dispersed into the uncured elastomer precursor composed of a catechol group and thiooctyl monomer. On account of the existence of the LM, hydrophobic alkyl chain, dynamic disulfide bond, catechol group, and carboxyl group in the system, this electrode had high conductivity (1.3×104 Sm^{-1}), outstanding reversible wet bonding strength (670 kPa), and fine self-healing function under high temperature/light (90% healing efficiency after 4 hours of healing at 60 °C)[37]. Practically, the self-healing property gives elastomers the ability for long-term monitoring, which further expands the application of dry electrodes.

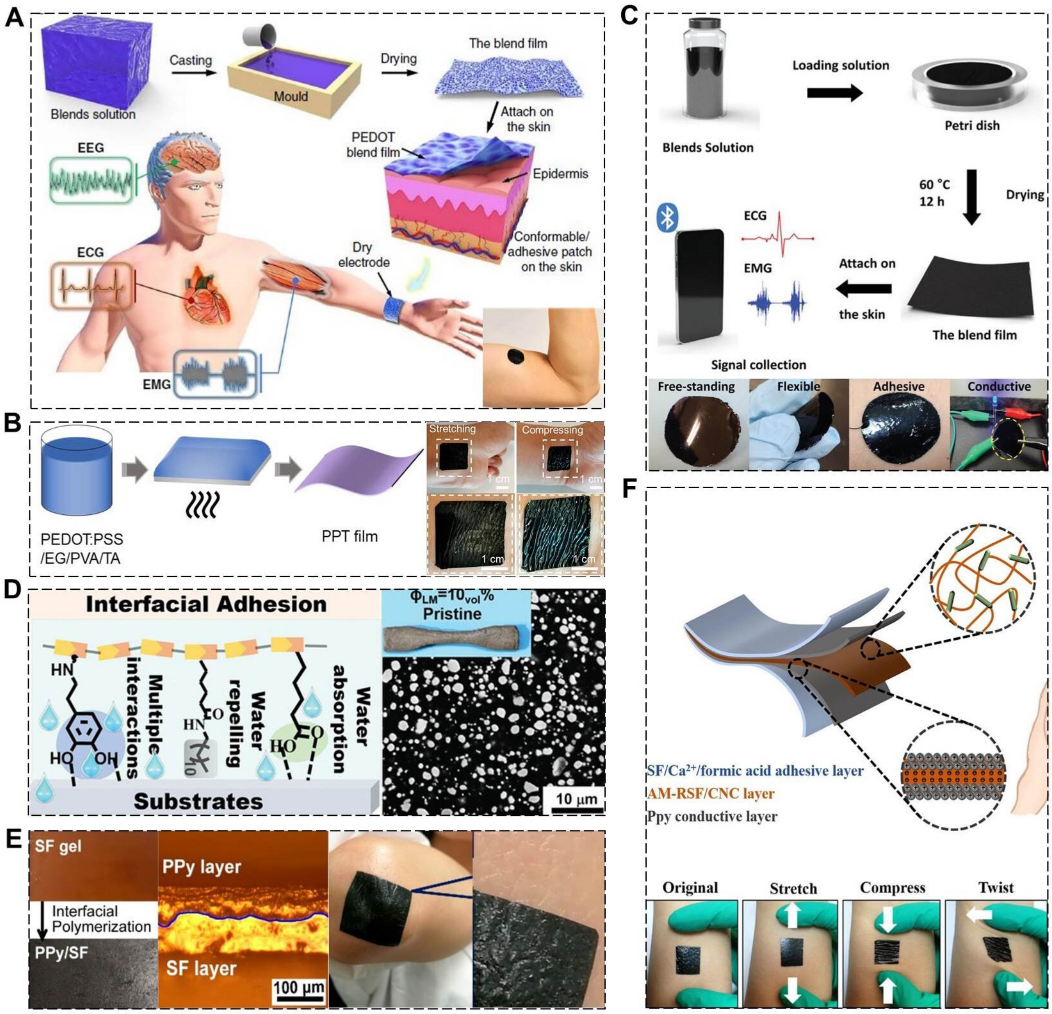

Figure 3. Self-adhesive dry electrodes. (A) Preparation flow chart of the electrode based on PEDOT:PSS (the illustration in the lower right corner is its physical picture)[28]. Reprinted with permission. Copyright 2020, Springer Nature. (B) Preparation diagram of the highly sweat-resistant dry electrode and photos of its stretch/compression at the wrist[29]. Reprinted with permission. Copyright 2022, American Chemical Society. (C) Formation diagram of the environment-friendly dry electrode and its photos showing various characteristics[30]. Reprinted with permission. Copyright 2022, Wiley-VCH. (D) Explanatory diagram of the interface adhesion mechanism of the electrodes based on LM-doped elastomers and its SEM image (the illustration in the upper right corner is a sample picture)[37]. Reprinted with permission. Copyright 2022, Wiley-VCH. (E) Pictures of PPy and SF before and after interfacial polymerization and a sample picture of the electrode based on this interface interlocking structure[40]. Reprinted with permission. Copyright 2020, American Chemical Society. (F) Structure diagram of the electrode with a three-layer structure and its contact pictures with the skin under different conditions[41]. Reprinted with permission. Copyright 2022, Elsevier. LM: Liquid metal; PEDOT:PSS: poly (3, 4-ethylenedioxythiophene): poly (styrene sulfonate); PPy: polypyrrole; SF: silk fibroins.

Dry electrodes are applied to the delicate skin of the human body. Once electrodes are toxic or irritating, they can lead to skin damage and cause discomfort to the user. Therefore, biocompatibility is an important criterion for measuring dry electrodes. Some natural adhesive materials, such as silk fibroins (SF), have attracted people's attention because of their exceptional biocompatibility and superior conformal adhesion[38,39]. Yang *et al.* obtained the structural interlock of Polypyrrole (PPy) and SF by means of interfacial polymerization and fabricated a self-adhesive dry electrode with water-activated conformal properties [Figure 3E]. At higher relative humidity (94.6%), SF with Young's modulus (2.1 MPa) close to that of skin could boost interfacial adhesion (40 J/m^2). Meanwhile, the chain structure of PPy and SF promoted the tensile stability of this electrode (R/R_0 at 100 cycles of stretch was approximately 1). These features ensured that this electrode was capable of collecting stable and reliable ECG signals in the environment of sweating and exercise (sensitivity of 0.5; conversely, the sensitivity of the wet electrode was 0.31)[40]. Additionally, a self-adhesive dry electrode (PPy@AM-SF/CNC) with low impedance was prepared by Meng *et al.* and was able to easily detach from the skin [Figure 3F]. The group used acid-modified silk/ cellulose nanocrystal film as the substrate, coated PPy as the conductive layer, and finally spread calcium-modified SF as the adhesive layer. Among these three layers, the SF layer was modified with Ca to facilitate the adhesion of the PPy@AM-SF/CNC electrode (12.85 N/m on pigskin) because Ca ions could chelate with metals and trap water. Notably, when the electrode was immersed in water, the cohesive force decreased due to the obvious increase of water content (peel force dropped sharply from 13.5 N/m to 9.22 N/m in 10 s), so this effectively achieved a non-destructive separation from skin[41].

Adhesion of self-adhesive dry electrodes is profitable for their conformal contact, thereby reducing skin contact impedance and noise. However, the adhesion of electrodes on wet skin is not ideal, so the development of self-adhesive dry electrodes that can be applied to both wet and dry skin is a future endeavor. From the perspective of materials, PEDOT:PSS and elastomers are common substrate materials for the fabrication of stretchable self-adhesive electrodes. Although their intrinsic properties are limited, this is compensated by the doping of certain substances (TA, LMs, *etc.*). Additionally, with the increasing research on the practical application of dry electrodes, people are paying more attention to the biocompatibility of electrodes. Some biocompatible viscous substances (SF) can help us to obtain self-adhesive dry electrodes with little skin damage. However, poor stretchability and low conductivity of viscous substances limit them to further development. Hence, the modification of natural viscous substances is an important trend for self-adhesive dry electrodes.

Ultra-thin dry electrodes

It is believed that thick electrodes are hard to adhere tightly to the skin[42]. This leads to a decrease in the contact area at the electrode-skin interface, affecting the quality of signals. Specifically speaking, for the same material, thick electrodes are usually more difficult to bend than ultra-thin electrodes. Therefore, it cannot establish a wonderfully conformal contact with skin, which is more prone to generate noise and artifacts. On the contrary, the ultra-thin electrode with better conformability is capable of filling the wrinkles and depressions of the skin, thus maintaining a larger contact area and obtaining better signal quality[43]. In brief, reducing the thickness of electrodes is an uncomplicated and valid tactic of conformal contact. However, the imbalance between the mechanical and electrical properties of ultra-thin electrodes is a common conundrum in research. From this, Tang *et al.* utilized solution processing of PEDOT:PSS, polysorbate, and glycerol to fabricate a delamination-resistant imperceptible dry electrode [Figure 4A]. The polar group in glycerol could lead to the phase separation of PEDOT and PSS; that is to say, the partial aggregation of PEDOT formed a conductive network, and PSS became a dispersed soft matrix. As a functional bridging unit of PEDOT and PSS, the biocompatible polysorbate could dissipate the tensile force through its hydrophilic/hydrophobic interaction, which endowed the electrode with high fracture strain (elongation at break was higher than 90%, and the corresponding resistance increased only 1.7 times). More importantly, the thickness of several microns enhanced the compliance of the electrode to the skin, obtaining a good signal-to-noise ratio for EMG (35.23 dB)[44]. Jiang *et al.* evaporated the gold film onto the PDMS film and proposed a PDMS-gold conductor [Figure 4B] with a thickness of only 1.3 microns. During the preparation process, the thick PDMS film expanded, and the thin gold film emerged with controlled micro-cracks. This micro-cracked structure maintained steady conductivity of the electrode during stretching (resistance only increases by 1.7% at 0% strain after 5000 cycles at 100% strain). Furthermore, the electrode could record ECG signals with a high signal-to-noise ratio (37.5 dB)[45].

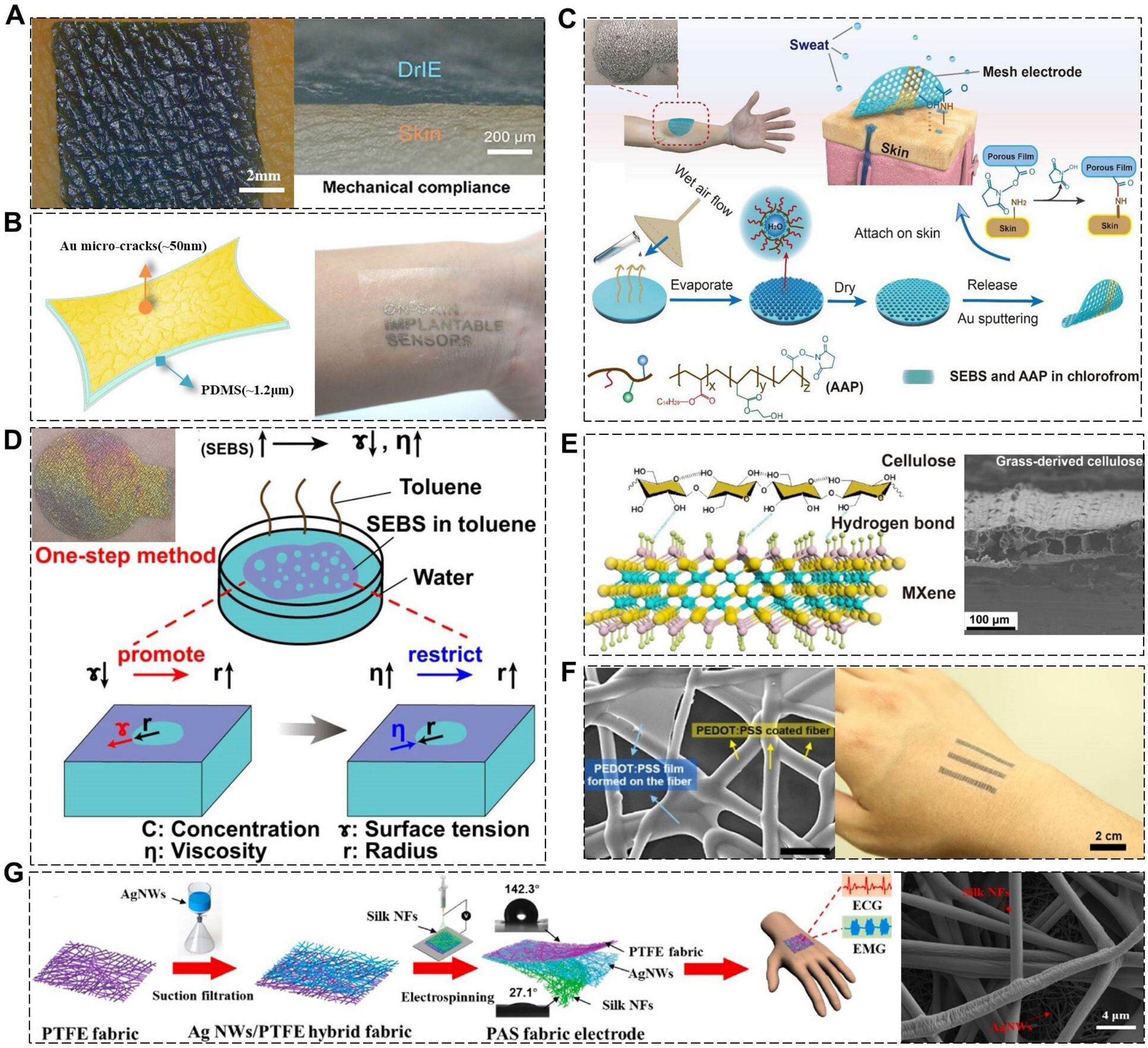

Figure 4. Ultra-thin dry electrodes. (A) The physical picture of the delamination-resistant imperceptible dry electrode and its cross-section optical microscope image after contacting pig skin[44]. Reprinted with permission. Copyright 2021, American Chemical Society. (B) Structure diagram of PDMS-gold conductor and its sample photo[45]. Reprinted with permission. Copyright 2022, Springer Nature. (C) Schematic diagram of the single-layer porous membrane electrode (the illustration in the upper right corner is its real photo)[48]. Reprinted with permission. Copyright 2022, American Chemical Society. (D) Description of the principle of the porous ultra-thin electrode (the illustration in the upper right corner is its physical picture)[49]. Reprinted with permission. Copyright 2022, Springer Nature. (E) Schematic diagram of hydrogen bond in the ultrathin MXene-based electrode and SEM diagram based on the pore structure of grass-derived cellulose[51]. Reprinted with permission. Copyright 2022, American Chemical Society. (F) Physical image and SEM image of the breathable electrode based on the electrospinning method[54]. Reprinted with permission. Copyright 2021, American Chemical Society. (G) Process flow diagram of the ultra-thin (10 μm) fabric electrode and its SEM diagram[55]. Reprinted with permission. Copyright 2022, Elsevier. PDMS: polydimethylsiloxane.

Because the ultra-thin electrode has excellent bonding ability with skin, bacteria are easy to breed, and signal quality is poor in sweaty and humid environments. More attention should be paid to the permeability of ultra-thin electrodes. Porosity is a means to elevate air permeability[46,47]. Tian *et al.* reported a single-layer porous membrane electrode [Figure 4C] using the breath figure method (BFM). N_2 was blown onto the surface of the chloroform solution of styrene-ethylene-butylene-styrene (SEBS)/adhesive and amphiphilic polymer (AAP). The chloroform rapidly evaporated, resulting in a porous film structure. Then, gold was magnetically sputtered on the surface of the porous film. The pore size and wall thickness were less than 10 μm, and it was well matched with the pores of human skin, making it comparable to the ultrahigh permeability of naked skin (Approximately 25.3 $gm^{-2}d^{-1}$). Moreover, this electrode was clearly able to observe the ECG waveform and obtain a heart rate of approximately 62 bpm[48]. Xie *et al.* fabricated a porous ultra-thin (200 nm) electrode [Figure 4D] using the Marangoni effect and magnetron sputtering gold. Compared to water, the surface tension of SEBS solution was lower, so an ultra-thin layer of SEBS solution would appear on the water surface. The gradual evaporation of toluene increased the concentration of SEBS, resulting in an increase in surface tension and a decrease in the viscosity of SEBS. When the surface tension of the SEBS solution decreased, material transport would occur, inducing the formation of membrane pores. At the same time, the increase in the viscosity of the SEBS solution restricted the flow and the increase in pores. When the toluene was completely evaporated, an ultra-thin porous film (25.3 $gm^{-2}h^{-1}$) was accomplished. Finally, sputtering gold endowed this electrode with conductivity. The signal-to-noise ratio of this electrode during sweating was close to that of the wet electrode (about 13 dB)[49]. Additionally, Yu *et al.* reported a highly breathable and ultrathin MXene-based electrode (MBE) [Figure 4E]. Transition metal carbides or nitrides, known as MXene[50], are two-dimensional materials with superb conductivity and biocompatibility. Inspired by the natural porous structure of plants, they chemically etched Pennisetum to prepare a flexible porous cellulose skeleton and then impregnated the MXene sheet. Principally, the functional groups of -O, -OH, and -F in MXene could form hydrogen bonds with -COOH and -OH in cellulose. This hydrogen-bonded MXene nanosheet made MBE breathable and excellent conductive (10 Ω/sq). Surprisingly, MBE had wonderful joule heating ability, which could be used as a combination of electrical stimulation and electrotherapy functions in muscle therapy[51].

The nanofiber network structure is another way to increase air permeability. As is well-known, electrospinning is popularly used to prepare porous nanofiber network substrate[52,53]. Jeong *et al.* proposed a breathable electrode [Figure 4F] that sprayed PEDOT:PSS on TPU elastomer nanofiber produced by electrospinning. This structure with nano-network was the source of outstanding breathability (12.0 $gm^{-2}h^{-1}$), expanding the application prospect of ultra-thin electrodes[54]. Yan *et al.* fabricated an ultrathin (10 μm) fabric electrode (PAS) [Figure 4G]. They covered Ag NWs (as a conductive layer) on the surface of hydrophobic polytetrafluoroethylene (PTFE) non-woven fabric (as an external self-cleaning layer) by suction filtration operation. Then, viscous SFs (Silk NFs) (as a skin contact layer) were electrospun on the conductive layer. This PAS electrode had brilliant permeability (2553 $gm^{-2}d^{-1}$), surface hydrophobicity (contact angle 142.3°), and outstanding conductivity (3.58 Ω/sq). These brilliant characteristics made the electrode have low noise and high signal-to-noise ratios (22.59 dB). However, since Ag NWs were bonded to PTFE using physical suction filtration, the cyclic stretching conductivity of this electrode was a weak point for high-quality monitoring of long-term motion[55].

The thickness of ultra-thin electrodes is usually at the nanometer or micrometer level, which makes them better conform to the curves of skin, less prone to falling off, and less likely to create gaps at the interface. Nevertheless, it is a challenging task to balance the mechanical and electrical properties of ultra-thin electrodes while controlling the low thickness. In addition, how to enhance the permeability of ultra-thin electrodes to avoid skin inflammation is also worthy of in-depth consideration. In fact, porous membranes and fiber network structures are two means to ensure the air permeability of electrodes.

Micro-structured dry electrodes

The micro-structured electrode refers to the design of a special microstructure on the surface of electrodes so that the electrode itself can pass through the hair but not penetrate the skin. The microstructure ensures a larger contact area, lower contact impedance, and better conformal contact[56]. At present, some microstructures have been prepared extensively by researchers, such as claw-like, spring-like, micropillarlike, and bionic microstructures.

Xing *et al.* reported a tentacle electrode whose shape was made by molding [Figure 5A]. They chose TPU, a polymeric elastomer, as the primary manufacturing material. The electrode was designed as a claw-like structure with a diameter of 14 mm and had a hemispherical tip featuring eight small fingers of 6 mm in length and 2 mm in diameter. This structure and elasticity could help fingers pass through hair and touch skin to record EEG signals with high signal-to-noise ratios (12.83 ± 2.85 dB; conversely, a signal-to-noise ratio of 14.1 ± 3.24 dB for the wet electrode)[57]. Liu *et al.* developed a novel dry electrode consisting of an upper printed circuit board (PCB), a lower PCB, a thread holder, and spring probes [Figure 5B]. The buffering effects provided by spring contact probes and the flexible cap enabled dry electrodes to attach well to the scalp without pain. Simultaneously, the EEG signal of this electrode was not significantly different from that of the wet electrode (relative error between 0.8179 ~ 0.9677), which could be easily and quickly used in clinical applications[58]. Using anodic oxidation (AAO) as a template, Niu *et al.* reported a micro-nanopillar array-structured dry electrode [Figure 5C]. They used in situ polymerization method to polymerize PANI nanopillars on the pore wall of AAO and spin-coated TPU/N,N-dimethylformamide (DMF) solution on AAO/PANI. After curing, the AAO was removed to obtain the PANI/TPU dry electrode. Compared with the planar wet electrode structure (2.257 MΩ at 0.1 Hz; 13.54 ± 7.86 dB), the electrode with the micro-nanopillar array structure could increase the contact area, thus showing a lower contact impedance (375.5 kΩ at 0.1 Hz) with a higher ECG signal-to-noise ratio (21.33 ± 5.48 dB) in a dynamic environment as well[59].

The bionic microstructure electrode fabricated by imitating the unique physical structure of organisms has several distinctive properties, such as adhesion[60] and the directional transport of water[61]. Inspired by grasshopper feet, Stauffer *et al.* reported a micro-structured dry electrode using the template method [Figure 5D]. This electrode was a combination of a soft porous electrode at the bottom (with an elastic modulus of ≈ 1 MPa similar to that of human skin), a macroscopic pillar electrode in the middle, and a micro-structured electrode at the top. This dry electrode could pass through thick hair and make conformal contact with skin. Furthermore, the skin contact impedance was comparable to that of clinical standard electrodes (approximately 50 kΩ cm^2 at 10 Hz)[62]. However, a challenge remains in terms of the adhesion of dry electrodes in human sweating environments. To solve this problem, Kim *et al.* reported a water-drainable dry electrode with a hexagonal micro-pattern inspired by the hexagonal micro-pattern of a tree frog toe pad and convex cup in an octopus sucker [Figure 5E]. They first prepared silicon templates using photolithography and reactive ion etching techniques. PDMS was fabricated into microstructures using simple micro-contact printing methods and then was sprayed with reduced graphene oxide (1.2 μm in thickness). The hexagonal micro-patterned channel facilitated sweat elimination, and convex cups on its upper surface improved skin adhesion in sweaty environments (2.36 Ncm^{-2} in dry conditions; 1.92 Ncm^{-2} in sweaty conditions) so that the electrode avoided detachment from the skin in both static and dynamic environments[63]. The surface of the forelegs of male diving beetles has a micro-cavity sucker structure and micro-wrinkles under the micro-cavity. The existence of micro-cups and micro-wrinkles makes the forelegs of beetles possess strong suction, liquid drainage, and high tensile strength. Inspired by this, Min *et al.* fabricated a flexible electrode with isotropic folded suction cups by compounding multi-walled carbon nanotubes (MWCNT) with PDMS via a processable selective transfer technique [Figure 5F]. Specifically, MWCNT suspensions were sprayed on plasma-treated and pre-strained PDMS. After tension release, the sprayed MWCNT wrinkled PDMS. This electrode had excellent adhesion (2.2 Ncm^{-2} on dry pig skin; 1.5 Ncm^{-2} on sweaty pig skin), superb electrical properties (approximately 87 Sm^{-1}; conductivity change of only 0.1 even at 30% tensile strain), and good moisture resistance (contact angle < 141.9°)[64]. Inspired by the conical beak of shorebirds, a sweat-proof Janus Au NWs/nitrocellulose (NC) electrode with a conical

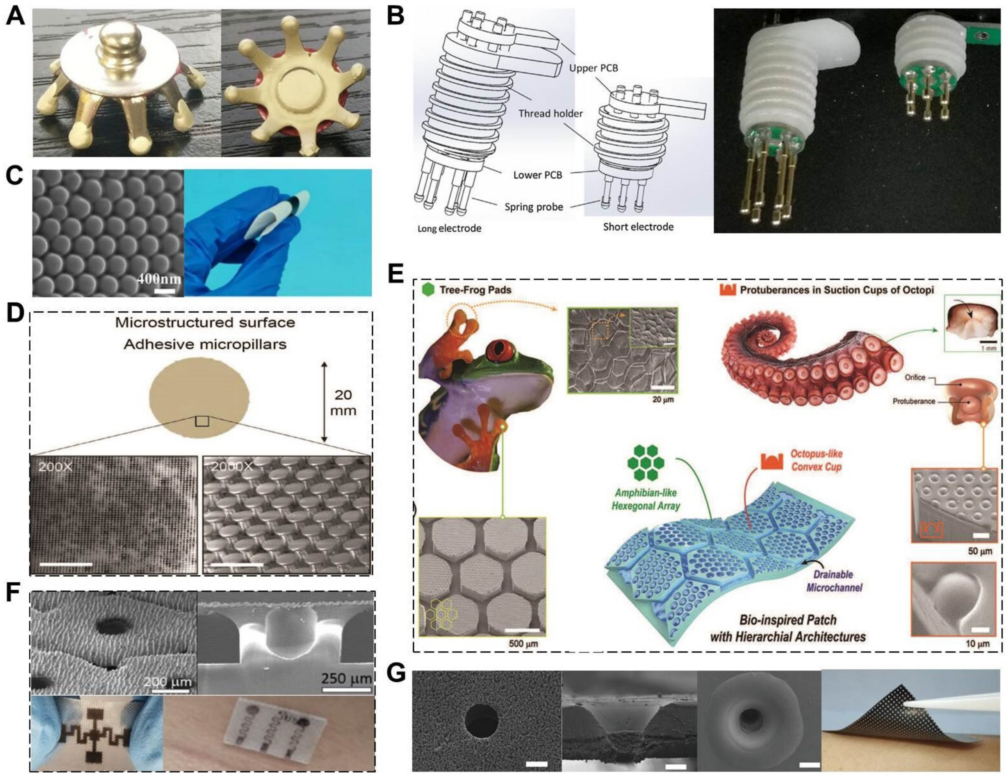

Figure 5. Surface microstructure electrodes. (A) Image of the caw-like dry electrode[57]. Reprinted with permission. Copyright 2018, Springer Nature. (B) Schematic diagram and a sample picture of the spring-like dry electrode[58]. Reprinted with permission. Copyright 2019, Elsevier. (C) SEM image of the micronanopillar electrode and its physical photo[59]. Reprinted with permission. Copyright 2022, American Chemical Society. (D) SEM image of the grasshopper-inspired bionic dry electrode with a scale of 200 μm (200 ×) and 20 μm (2000 ×)[62]. Reprinted with permission. Copyright 2018, Wiley-VCH. (E) Schematic diagram of the water-drainable dry electrode inspired by frog and octopus[63]. Reprinted with permission. Copyright 2019, Wiley-VCH. (F) SEM image of the microstructure electrode inspired by the forelegs of male diving beetles and its physical photo[64]. Reprinted with permission. Copyright 2021, Wiley-VCH. (G) SEM images from different angles of the Janus Au NWs/NC electrode inspired by the conical beak of shorebirds (the nitrocellulose side, the cross-section, and the Au NWs side from left to right, scale bars: 100 μm) and its physical picture[65]. Reprinted with permission. Copyright 2022, Wiley-VCH.

microporous structure was reported by Li *et al.* [Figure 5G]. Because the semi-cured polyurethane (PU) was highly adhesive, they prepared Au NWs on one side of the PU and lightly rolled the NC film onto the other side of the PU. The conical hole fabricated by the laser perforation method could spontaneously drive sweat from the electrode layer (the Au NWs side) to the sweat adsorption layer (the NC side), thus maintaining high interfacial adhesion (approximately 1.7 MPa) and low interfacial impedance (7 kΩ at 100 Hz; conversely, the impedance of bare Au NWs without microstructure is 10 kΩ) in the sweaty state[65].

With the development of micro and nanotechnology and the study of organisms in nature, people gradually focus on dry electrodes with surface microstructure. The microstructure generally has textural structure rules and certain periodicity. In addition, the microstructure can be arranged in different sizes and combinations to make electrodes exhibit different functional properties, such as adhesion, friction, and hydrophobicity. The current investigation of micro-structured dry electrodes involves multidisciplinary and has become a research hotspot. However, the development of micro-structured dry electrodes is still limited by processing technology. Thus, future research focus can be on the design and manufacturing technology of micro and nanostructures.

As shown in Table 1, among the three types of dry electrodes discussed, capacitive electrodes are able to avoid damage to the skin because they do not touch the skin. They can be applied to hairy areas and pass through multiple layers of clothing. However, they have the drawbacks of poor conformal contact with skin, the highest contact impedance, and lower quality in dynamic monitoring signals. Accordingly, capacitive electrodes are of little interest for research in the field of conformal contact. Invasive microneedle electrodes use microneedles to penetrate the horny layer, so they have better conformal contact with skin, minimal electrode-skin interface impedance, and superior signal-to-noise ratios. However, the discomfort and inflammation caused by the microneedle pose a threat to human health and limit the practical application of electrodes. Common-contact dry electrodes are characterized by direct contact with skin but do not penetrate the horny layer. This non-invasive nature determines relatively little skin damage and contact impedance, so the research prospects of common-contact contact dry electrodes are broad. However, complete conformal contact with skin is difficult, except with the help of certain properties (self-adhesive or ultra-thin) or special structures (microstructures). For self-adhesive dry electrodes, the humid environment limits their adhesion, which is not suitable for long-term monitoring of electrophysiological signals or detection under heavy sweating scenarios. Likewise, the weak breathability of ultra-thin dry electrodes easily causes skin inflammation and discomfort to users, making it difficult to detect signals on human skin for a long time. Nevertheless, there is a gap between the micro-structured dry electrode and the skin to remove gas and sweat generated by skin metabolism. Thus, inflammation is avoided, and long-term signal detection is facilitated. Actually, the signal-to-noise ratio of micro-structured dry electrodes is slightly lower than that of self-adhesive dry electrodes and ultra-thin dry electrodes, but reasonable design of microstructures can improve the contact situation to achieve the same level of signal-to-noise ratio. In addition, various organisms and phenomena in nature are our inspiration for designing microstructures. Bionic structures have more ingenious and interesting properties. Therefore, micro-structured dry electrodes have great research value and great prospects.

CONCLUSION AND OUTLOOK

Recent advances of dry electrodes are reviewed, focusing on their conformal interfacial contact. Capacitive electrodes have inferior interfacial contact and high impedance owing to the presence of a dielectric layer. Invasive microneedle electrodes have microneedles inserted into the horny layer of skin, providing excellent conformal contact but easily causing discomfort. Common-contact dry electrodes are preferred by scientists for their ability to realize conformal contact with the skin while being non-invasive. Moreover, three types of common-contact dry electrodes, namely self-adhesive dry electrodes, ultra-thin dry electrodes, and micro-structured dry electrodes, are mainly discussed. Simultaneously, their material selection and structural design to obtain conformal contact are elaborated. In addition, looking back at the development in recent years, it is not difficult to find trends and challenges of dry electrodes in the future.

During bioelectric signal measurement, the movement of human skin causes the electrode to slip on the skin and thus causes signal noise, which is the source of motion artifacts. Conformal contact with skin can reduce interface impedance and enhance signal quality. Therefore, dry electrodes in conformal contact with the skin are the best way to decrease motion artifacts. In general, stretchable, flexible, self-adhesive, ultra-thin, and micro-structured properties can be endowed to the dry electrode to achieve conformal contact with skin.

Superb stretchability and flexibility are basic ideal features for dry electrodes. Specifically, electrodes better adapt to the deformation environment with the aid of high stretchability, while high flexibility helps electrodes better comply with skin. Intrinsically stretchable materials and structural design are two strategies to achieve the stretchability of electrodes. Intrinsically stretchable materials mainly include elastomers, gels,

Table 1. Advantages and disadvantages of capacitive electrodes, invasive microneedle electrodes, and common-contact dry electrodes

Type	Advantages	Disadvantages
Capacitive electrodes	No skin traumaSuitable for hairy and multi-layer clothing isolation	Non-conformal skin contact and high contact impedance
Invasive microneedle electrodes	Conformal contact and low contact resistance	Skin damage and user discomfort
Common-contact dry electrodes	Light skin damage	Incomplete conformal contact

and semiconductor polymers. Conductive materials, such as metals, are usually used as fillers to improve their conductivity. For some non-stretchable rigid materials, geometrical structures are designed to give them stretchability. For example, micro-cracks, serpentines, out-of-plane waves, open-mesh, and kirigami all exhibit outstanding anti-deformation capability[66]. Since dry electrodes work on soft skin, the electrodes must be flexible for conformal contact and wearing comfort. Considering the correlation between the Young's modulus and flexibility, the Young's modulus of electrodes is supposed to be reduced, which can smoothly conform to the skin and improve interface contact.

An effective means of maintaining conformal contact between electrodes and skin is interfacial adhesion. In the past, additional adhesives (conductive gels) were used to attain interfacial adhesion. For easy use of electrodes, scientists turn their focus to self-adhesive electrodes. However, self-adhesive organic dry electrodes always encounter the hassle of viscosity reduction in a humid environment. As a consequence, effective adhesion of self-adhesive dry electrodes on dry and wet skin is crucial for practical applications. Additionally, although strong interfacial adhesion facilitates conformal contact, electrodes, after use, are difficult to peel off from the skin and may even cause damage. Therefore, convenient detachment from the skin is also the way forward for dry electrodes.

The thickness of the electrode is closely related to the bending stiffness. Compared to thick electrodes, ultra-thin electrodes have less bending stiffness and stronger van der Waals forces at the interface, resulting in more conformal contact with skin. However, due to sweat and gas discharged from human skin, skin impedance will increase, and bacteria are more likely to breed, so we must leave no stone unturned to enhance the permeability of electrodes. Intrinsically breathable materials are poorly permeable, while the structural design can efficiently endow electrodes with excellent breathability. Examples include porous membranes prepared by blown bubbles, phase separation, and sacrificial sugar templates or fiber network membranes prepared by electrostatic spinning. In addition, ultrathin electrodes often face the problem of imbalance between mechanical and electrical conductivity, so a good balance between them is what researchers have been pursuing.

The ingenious microstructure allows electrodes to pass through hair and directly touch the skin, thus making better conformal contact and reducing contact resistance. When researchers design microstructures, inspiration should not be limited to daily life. The creatures and phenomena in nature may be helpful to us. After all, all living creatures change themselves to adapt to nature through cruel natural selection. Applying their physical characteristics and habits to electrodes can yield unexpected benefits.

Besides the above characteristics, the increasing demand for electrodes with long-term applications has made the study of self-healing and biocompatibility increasingly popular. Traditional conductive active materials (metal/conductive polymers) are prone to irreversible damage and fracture under external mechanical manipulation, so exploiting electrodes with rapid self-healing properties can effectively alleviate this problem, such as living assembled materials that take advantage of the healing properties of bacterial cell growth[67]. Biocompatibility is a constant theme in biomaterials research. Since dry electrodes are applied directly to the surface of human skin, biocompatibility is critical. Biocompatibility is a property of living body tissues that determines their reaction to a product. Better biocompatibility means that the product is less harmful to living organisms. During the design of dry electrodes, we need to consider whether the product triggers skin allergy or inflammation and whether it can remain low or even non-toxic in the long term. To improve the biocompatibility of dry electrodes, researchers should avoid using irritating and toxic substances as much as possible. Notably, the use of natural materials, such as silk protein SF and dopamine, can greatly enhance biocompatibility. Moreover, in response to the national double-carbon policy, the investigation of the degradable electrode, a kind of green product, should not be ignored.

Although significant progress has been made in the study of various aspects of performance just mentioned, the preparation of practical electrodes with excellent comprehensive performance is still a challenge. Consequently, it is imperative to achieve a perfect balance of all properties (stretchability, conductivity, flexibility, *etc*.).

It is worth mentioning that dry electrodes have not been actually on the market, and wet electrodes still occupy the main position. Nowadays, dry electrodes with conformal contact studied in the laboratory can reach the same level as wet electrodes in monitoring signals (signal-to-noise ratio). However, to truly become a viable commercial product, dry electrodes not only have excellent performance and mature process routes but also need to have low cost, good user experience, and long-term durability. In addition, the electrophysiological signals of the human body are unstable and complex. There is mutual crosstalk between various signals, such as mutual interference between myoelectricity and cardiac electricity. Therefore, how electrodes take specific signals while circumventing other signals needs to be further investigated by scientists. Unfortunately, we still have not found a perfect solution to coordinate all aspects of electrode performance. Can we realize large-scale commercial production in the future? This is an interesting topic for the practical application of dry electrodes. The idea that dry electrodes obtain mass commercial production merits careful consideration.

A product fulfills its specific practical value when it is applied. Designers must fully consider specific application scenarios so that they can create commercial products with maximum functionality to meet specific needs. Dry electrodes monitor electrophysiological signals to achieve health monitoring, making them highly promising in the biomedical field. For example, they are used in applications such as ECG scanning[68], respiratory monitoring[69], and pulse monitoring[70]. In the digital era, the rapid rise of artificial intelligence and the Internet of Things has led to a growing desire for human-computer interaction. The electrophysiological signals of the human body monitored by dry electrodes can be transformed into specific signals for human-computer interaction through a series of processing. Examples include hand gesture recognition[71], lip reading[72], and prosthetic control[73]. Smart wearable devices are the ideal future model for all types of electronics. Dry electrodes integrated into wearable devices capture information about the human body and do not interfere with the activities of users. Meanwhile, such smart wearable devices can transmit data with smartphones for easy monitoring[74-76]. Exercise is a normal part of life nowadays. Using dry electrodes for intelligent motion monitoring not only helps us to understand the state of the body during exercise but also provides scientific exercise guidance[77-79]. Significantly, a thorough understanding of the application scenarios of dry electrodes can lead researchers to carry out targeted designs. Furthermore, with the development of system integration and machine intelligence, the integration of dry electrodes into systems such as wearable devices or sensors[80] will be a very promising direction.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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