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Emerging epidermal electrodes towards digital health and on-skin digitalization

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Abstract

Epidermal electrodes can be directly attached to the human skin for high-fidelity electrophysiological monitoring owing to their preponderance in thinness, lightweight, conformability, biocompatibility, self-adhesiveness, mechanical flexibility, gas-permeability, *etc*. These devices have attracted immense attention due to their emerging applications in personalized health care, human/brain-machine interfaces, and soft robotics. This Perspective focuses on the most recent significant progress in this area, especially materials, properties, and applications. Challenges and prospects are summarized to underscore the unexploited areas and future directions toward digital health and on-skin digitalization.

Keywords: Epidermal electrodes, ultrathinness, ultra-conformability, electrophysiology, digital health, on-skin digitalization

INTRODUCTION

Epidermal electrodes have seen tremendous developments in the last two decades, both in materials and structures and prominent applications, such as health monitoring, diagnosis and therapy, human/brain-machine interfaces (HMIs/BMIs), prosthetics, robotics, and augmented reality (AR) and virtual reality (VR) communications[1-5]. In particular, for health monitoring, epidermal electrodes have attracted intensive attention for non-invasive electrophysiological recording, such as electromyogram (EMG) (amplitude between 50 and 5,000 μV, frequency between 5 and 500 Hz), electrocardiogram (ECG) (amplitude between 50 and 5,000 μV, frequency between 0.5 and 100 Hz), electrooculogram (EOG) (amplitude between 10 and 3,500 μV, frequency between 0.1 and 30 Hz), and electroencephalogram (EEG) (amplitude less than 100 μV, frequency between 0.5 and 100 Hz)[6-9]. Owing to their ultrathinness, lightweight, high conductivity, low skin-contact impedance, and skin-like mechanical properties, epidermal electrodes can pick up delicate ionic conduction signals on the epidermis induced by brain, muscle, eyeball, and heart activities[10,11]. Different types of electrophysiological signals provide valuable insights into the functioning of various tissues and organs. Together with the capability of electrostimulation, epidermal electrodes play a crucial role in sleep monitoring, wound healing, fatigue alerts, neurofeedback training, muscle and neurological disorder theranostic, HMIs, BMIs, *etc*.[12,13]. Besides digital health, epidermal electrodes are emerging devices to the realization of on-skin digitalization that aims to create a seamless interface between humans and devices and enable remote health monitoring and human-cyber interactions[7].

MATERIALS

Some pioneering work has been done in epidermal electrodes by introducing structural engineering on metal and polymeric films[14,15]. Structure engineering is an effective strategy to endow rigid electronic devices that are conformable and stretchable for skin applicability. Another significant strategy is to design and utilize intrinsically stretchable materials[16,17]. To achieve high conductivity for epidermal electrodes, a variety of electrical materials have been employed, such as conducting polymers, ionic liquids, liquid metals, low-dimensional nanomaterials (e.g., carbon/metallic-based nanomaterials and MXenes), and hydrogels [Figure 1A-E][18-22]. Figure 1A demonstrates the utilization of ionic liquid, bis(trifluoromethane) sulfonimide lithium salt (LiTFSI), integrated with poly(ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) and ethylene glycol for the fabrication of dermal electrodes[18]. In Figure 1B, micromeshes made of gallium liquid metals are created onto elastomer sponges for EMG electrodes[19]. A solvent-free supramolecular ion-conductive elastomer has been developed as an ionic tattoo for various electrophysiological monitoring applications [Figure 1C][20]. Figure 1D shows a sweat-stable EMG electrode made of MXene[21]. Besides MXene, other prevailing low-dimensional nanomaterials for epidermal electrodes include silver nanowires[23], gold nanowires[24], graphene[25], and carbon nanotubes[26]. Figure 1E presents bioadhesive hydrogel materials for rapid, robust, conformal, and electrically conductive integration between bioelectronic devices and various wet dynamic tissues[22]. It is worth noting that among them, conducting polymers[27,28], liquid metal, low-dimensional nanomaterials[29], and their hybrid[30] have been widely exploited for both dry and wet epidermal electrodes[31-34]. Wet electrodes refer to the use of conductive hydrogels or paste materials, such as the commercialized Ag/AgCl, which have good biocompatibility, skin adhesion, mechanical softness, and low skin impedance. Therefore, wet electrodes generally can provide high-fidelity and low-noise signals. On-skin dry electrodes are fabricated by carbon and metal-based materials and composites. Unlike wet electrodes, dry electrodes rely on external forces or van der Waal forces to achieve high skin contact, normally with high skin impedance and high-noise signals. However, their electrical and electromechanical properties can be tailored by various materials and structures[35,36]. Due to their versatile tunability on electrical and mechanical properties, hydrogels have emerged as advantageous materials for the development of epidermal electrodes, both as conductive materials and polymeric substrates[37,38]. Other representative polymeric substrate materials include PDMS[39,40] and polymeric electrospun nanofibers[41-43].

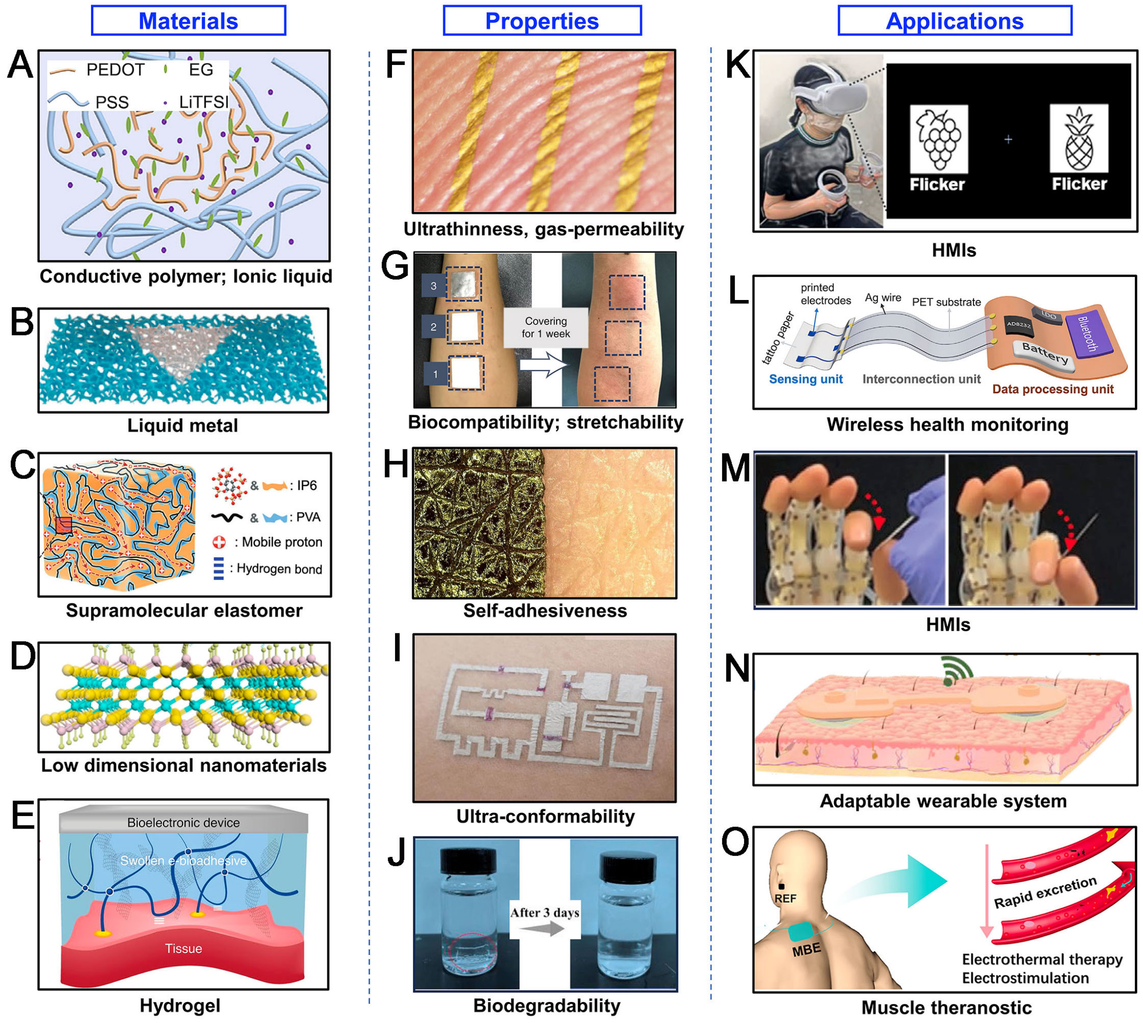

Figure 1. Recent representative examples of advanced epidermal electrodes (2017-2023). Material examples: (A) conductive polymer and ionic liquids[18]; (B) liquid metals[19]; (C) supramolecular elastomer[20]; (D) low dimensional nanomaterials[21]; and (E) hydrogels[22]. Property examples: (F) ultrathinness and gas-permeability[50]; (G) biocompatibility and stretchability[53]; (H) self-adhesiveness[44]; (I) ultra-conformability[13]; and (J) biodegradability[62]. Application examples: (K) HMIs[55]; (L) wireless health monitoring[18]; (M) HMIs[70]; (N) adaptable wearable system[71]; and (O) muscle theranostic[21]. Figure 1A adapted with permission from ref.[18]. Copyright 2023 Elsevier; Figure 1B adapted with permission from ref.[19]. Copyright 2022 American Chemical Society; Figure 1C adapted with permission from ref.[20]. Copyright 2023 John Wiley and Sons; Figure 1D adapted with permission from ref.[21]. Copyright 2022 American Chemical Society; Figure 1E adapted with permission from ref.[22]. Copyright 2022 Springer Nature; Figure 1F adapted with permission from ref.[50]. Copyright 2017 Springer Nature; Figure 1G adapted with permission from ref.[53]. Copyright 2021 Springer Nature; Figure 1H adapted with permission from ref.[44]. Copyright 2021 National Academy of Sciences; Figure 1I adapted with permission from ref.[13]. Copyright 2020 Springer Nature; Figure 1J adapted with permission from ref.[62]. Copyright 2023 John Wiley and Sons; Figure 1K adapted with permission from ref.[55]. Copyright 2022 American Association for the Advancement of Science; Figure 1L adapted with permission from ref.[18]. Copyright 2023 Elsevier; Figure 1M adapted with permission from ref.[70]. Copyright 2020 John Wiley and Sons; Figure 1N adapted with permission from ref.[71]. Copyright 2023 American Chemical Society; Figure 1O adapted with permission from ref.[21]. Copyright 2022 American Chemical Society. EG: Ethylene glycol; HMIs: human-machine interfaces; LiTFSI: bis(trifluoromethane) sulfonimide lithium salt; PEDOT: poly(ethylenedioxythiophene); PSS: poly(styrenesulfonate).

PROPERTIES

Depending on the target applications, different materials and structures are chosen to obtain the desired properties. Regardless of the type of application, the basic requirements of epidermal electrodes comprise biocompatibility, stretchability, sufficient thinness, and mechanical durability. Notably, the epidermal electrode with a compliant and comfortable interface guarantees high-quality bioelectrical signals where a low skin impedance can be attained. According to the flexural rigidity equation, flexural rigidity can be calculated as $D = \frac{Et^3}{12(1-\nu^2)}$, where E, t, and ν represent Young's modulus, thickness, and Poisson's ratio, respectively, of a thin film[44]. Therefore, reducing thickness is the most effective approach to decrease flexural rigidity, thus leading to higher skin compliance[45,46]. Towards this end, lots of electronic tattoo electrodes have been developed based on conducting polymers and two-dimensional nanomaterials[10,37,47,48]. Besides reducing thickness to obtain high skin compliance, dry electrodes should also be mechanically stretchable and durable to secure continuous attachment on the human skin[49].

With the introduction of electrospun nanomeshes, skin electronics have evolved from a thin-film form factor to a gas-permeable, biocompatible ultrathinness form factor[50-52] [Figure 1F]. Ma *et al.* reported biocompatible and permeable ECG electrodes using a liquid-metal fiber mat with a stretchability of over 1,800% strain [Figure 1G][53]. A self-adhesive electrode has been developed by reducing thickness to 165 nm employing Au-coated PDMS nanofilm [Figure 1H][44]. Another efficacious strategy to improve adhesiveness is to directly paint/draw inks/gels on the human skin [Figure 1I][13,54]. A recent example is a paintable epidermal electrode from thermal-controlled phase change gelatin-based hydrogels, which overcomes the limited conformability on hairy areas such as the scalp[55]. Taking advantage of the adhesive properties of hydrogels, many researchers have been working on simultaneously improving their gas-permeability for long-term skin applicability. There are two typical approaches: (1) ultrathin enough (a few μm-thick) to be permeable[20,56] and (2) macroscopic porous structure to be permeable[57,58].

Most existing wearable electronics are not decomposable and can lead to serious electronic waste (e-waste) and burden to Mother Earth[59]. To this end, biodegradable materials have been utilized to develop transient epidermal electrodes with zero waste footprint[60,61]. Lately, Ye *et al.* developed a fully biodegradable and biocompatible ionotronic skin that was made by carboxylated chitosan (CCS) and sulfobetaine methacrylate (SBMA) polymerized in glycerol and water followed by cross-linking with hydrogen bonds and electrostatic attraction[62]. As shown in Figure 1J, the developed ionic epidermal electrodes can accurately record action potentials and fully degrade in only three days without any residue. Other properties, such as washability[63], waterproof[64], self-healing[65], and antibacterial characteristics[66], have also been implemented for specific application scenarios.

APPLICATIONS

It should be noted that a significant application of epidermal electrodes is continuous and long-term electrophysical monitoring due to its critical role in early disease prevention, screening, diagnosis, and treatment[28,67]. Generally speaking, the capability of continuous, long-term monitoring requires a combination of various properties, such as low skin impedance, high conformability, gas-permeability, robust skin-electrode interface, and mechanical durability. Owing to the advancement of ever-fast materials, a plethora of such epidermal electrodes have been realized for long-term ECG and EEG acquirement[44,55,68,69]. Furtherly, the collected high-fidelity electrophysiological signals can be adopted for BMIs[55], wireless health monitoring[18], HMIs[70], adaptable wearable systems[71], prosthetics[72], and muscle theranostics[21] [Figure 1K-O]. As high-fidelity EMG and EEG acquirement is significant for non-invasive high-precision HMIs/BMIs[12,72], it is highly demanding to develop high-performance epidermal electrodes. Additionally, to enable epidermal electrodes with unsacrificed functionality under extreme conditions, such as aqueous environments and polar regions, adaptable epidermal electrodes have attracted intensive attention over the last decade[73-75]. For instance, Wan *et al.* reported an all-in-one flexible system capable of working under intense motion, heavy sweating, and varied surface morphology, conducting *in situ* injection and photonic curing of a biocompatible and biodegradable light-curable conductive ink[71].

A closed-loop platform consisting of monitoring and therapy takes personalized healthcare to the next level. In interesting research, Song *et al*. developed an all-in-one, bioderived, air-permeable, and sweat-stable MXene electrode that can simultaneously record EMG signals and achieve electrostimulation and electrothermal therapy for muscle theranostics[21]. Specifically, the as-prepared MXene electrodes exhibit high breathability, are ultralightweight (~0.25 mg/cm^3), and have a low and stable electrode-skin interfacial impedance in various environments, enabling the long-term reliable monitoring of electrophysiology.

SUMMARY

In this work, we highlight recent key developments of epidermal electrodes. Materials, properties, and applications have been discussed individually. Remarkable progress has been made in this area due to the enormous efforts devoted by researchers worldwide. It is believed that epidermal electrodes have contributed a significant part to digital health and on-skin digitalization. However, there are some remaining issues waiting to be addressed before these devices can be seamlessly integrated into our daily lives.

Concurrent realization of combined promising properties, such as low skin impedance, robust electronic bonding, high skin compliance, mechanical durability, and gas-permeability[49]. It requires the development of advanced materials and fabrication techniques and an in-depth understanding of the soft-rigid interface interactions during constant dynamic skin motions. For example, it is important to develop unconventional gas-permeable materials to overcome the intrinsic tradeoff between mechanical durability and thinness geometry. To solve this issue, lots of efforts have been devoted to developing fiber-based or fiber-reinforced ultrathin, gas-permeable electronics[51]. Another approach in materials development is to design bulky gas-permeable gels that have high bioadhesive properties. In this case, delicate cross-linking networks should be considered to maintain excellent long-term stability, such as anti-drying and mechanical robustness.

System intelligence. Skin sensor-artificial intelligence (AI) networks are paramount to the development of both digital health and on-skin digitalization. The AI algorithms enable epidermal electrodes not only to detect the health status for health management in real-time[76] but also to enhance the interactions between humans and machines[77]. A recent work by Ouyang *et al*. demonstrated a system-on-a-chip with Bluetooth Low Energy for data transmission and a compressed deep-learning module for autonomous operation[78]. The system achieved applications in studies of sleep-wake regulation and for the programmable closed-loop pharmacological suppression of epileptic seizures in mice via feedback from EEG recording. Besides the incorporation of data management technologies, other issues, such as processing capacity, long-term stability, and data security, should also be taken into consideration.

Multichannel and multifaceted operation. Multichannel bioelectrical sensing is vital to achieve comprehensive electrophysiology information for high-precision diagnosis and treatment and precision control for HMIs/BMIs[7,11,79-81]. For example, Tian *et al*. reported a large area bioelectronic interface for electrophysiological recordings that enable coverage of the full scalp and the full circumference of the forearm[79]. The large-area sensing arrays enabled multifunctional control of a transhumeral prosthesis by patients who have undergone targeted muscle-reinnervation surgery, in long-term EEG, and in simultaneous EEG and structural and functional magnetic resonance imaging. Additionally, the fusion of multifaceted functionalities, such as biophysical and biochemical monitoring and self-powering, is appealing to realize a full-fledged epidermal electrode system.

To mitigate the aforementioned concerns, an intimate collaboration between researchers from interdisciplinary backgrounds is a must, not only between engineers and clinicians but also between engineers, biologists, and informaticians[82]. The combined efforts can promote setting the criteria of electrodes and sensing performance and the transformation from laboratory prototypes to commercial products.

DECLARATIONS

Authors' contributions

The author contributed solely to the article.

Availability of data and materials

All data needed to evaluate the conclusions in the paper are presented in the paper. Additional data related to this paper may be requested from the authors.

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Conflicts of interest

The author 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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