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Nano energy for miniaturized systems

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ABSTRACT

Skin mountable electronic devices are in a high-speed development at the crossroads of materials science, electronics, and computer science. Sophisticated functions, such as sensing, actuating, and computing, are integrated into a soft electronic device that can be firmly mounted to any place of human body. These advanced electronic devices are capable of yielding abilities for us whenever they are needed and even expanding our abilities beyond their natural limitations. Despite the great promise of skin mounted electronic devices, they still lack satisfactory power supplies that are safe and continuous. This Perspective discusses the prospects of the development of energy storage devices for the next generation skin mountable electronic devices based on their unique requirements on flexibility and miniaturized size.

1. From microelectronic devices to cybernetic systems

Rapid development of electronic technologies has drastically reduced the dimension of electronic devices from meter scales (*e.g.*, personal computers), to millimeter scales (*e.g.*, portable devices), and, finally, to micro-/nanometer scales. These tiny electronic devices can be integrated into flexible substrate with the minimal compromise between functionality and flexibility, and help spawn the concept of wearable technologies. In terms of wearability, a mechanically comfortable interface with human body that minimizes the discomfort of worn or attached electronic devices is particularly important. Therefore, the resulting wearable devices should be inherently soft, which requires plastic substrates and soft electronic materials. As well as providing favorable interface with human body, wearables offer the vision of a cybernetic system powered by multimodal and multipoint sensing and stimulation, and rapidly evolve into the ultimate version, electronic skin, which will be directly attached to the motile surface of human skin (Fig. 1).

The progress from flexible electronic devices to electronic skin is achieved by the super-conformability of ultrathin-film devices with excellent flexibility and stretchability [1–3]. By further reducing the thickness, a new concept of imperceptibility is developed, which becomes one essential characteristic of electronic skin [4–7]. Another crucial feature is the ability to replicate the complex sensing ability of human skin to external environment through numerous receptors [5,8]. To do so, a soft platform comprising flexible sensor arrays and digital circuits such as shift registers, memories, wireless circuits, and amplifiers, has been developed (Fig. 2a) [4]. Apart from imperceptibility and multipoint sensing, limited area that can be used for attaching electronic skin raises the challenge of high integration density of electronic components, thus requiring 3D architectures. Whereas producing 3D systems by stacking 2D systems through planar microfabrication is intuitive, this strategy is relatively incompetent in reducing the footprint area because this target relies on demanding jobs of miniaturization of every electronic component and advances in electric circuit design [9–12]. Alternatively, shapeable materials allow for self-driven spatial rearrangement of 2D devices into 3D devices [13,14]. The manufacture of electronic components and design of electric circuit is based on the efficient planar microfabrication, while a self-assembly process reshapes the 2D system into a 3D object, which simultaneously achieves a complex system and satisfies the requirement of minimal footprint area. Fig. 2b shows a prototype of a Swiss-roll electronic device consisting of inductors, transformers, and resonators [15]. The footprint area of the 2D precursor system is approximately 100 mm^2 and drastically reduces to approximately 2 mm^2 after transforming into the Swiss-roll. In addition to the

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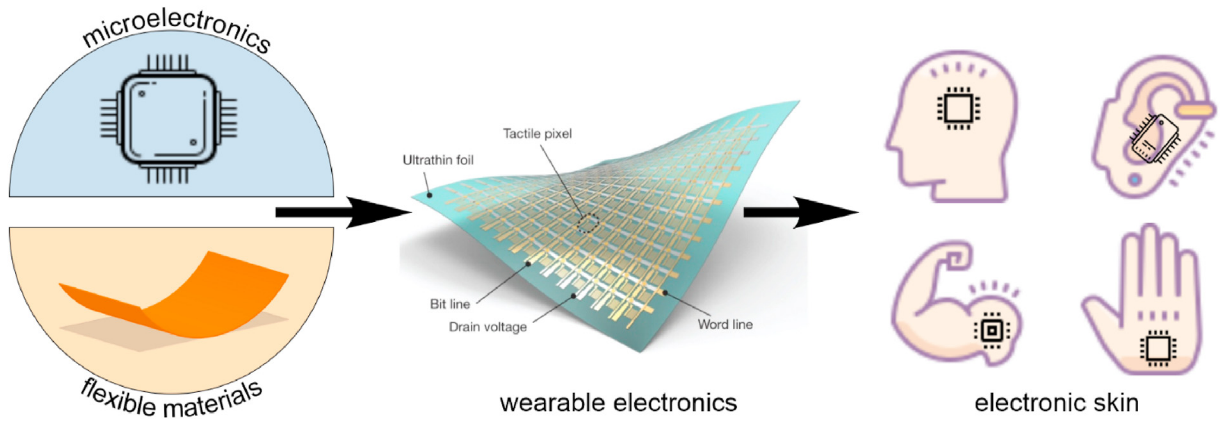

Fig. 1. The union of microelectronics and flexible materials enables the fast development of flexible electronics (Reproduced with permission [4]. Copyright 2013, Springer Nature) and provides a vision of cybernetic devices.

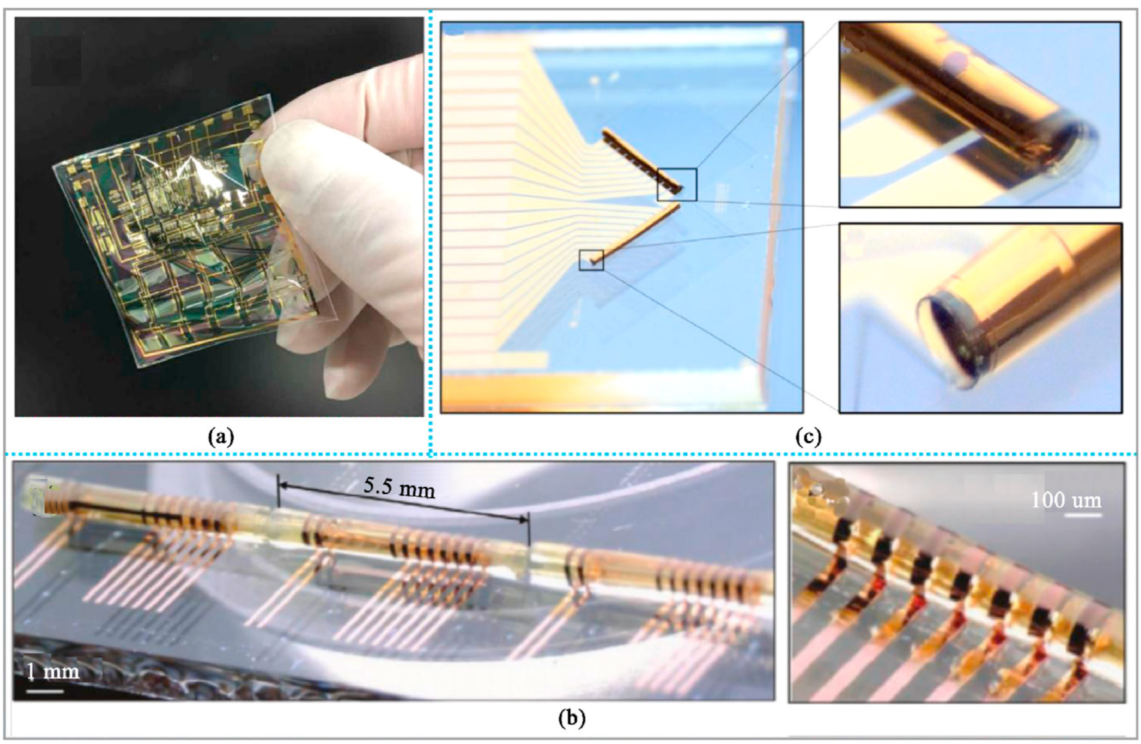

Fig. 2. Miniaturization of complex systems. (a) The imperceptible magnetic sensor matrix (Reproduced with permission [17]. Copyright 2020, AAAS). (b) The rolled-up compact magnetic inductors, transformers, and resonators (Reproduced with permission [15]. Copyright 2018, Wiley). (c) The rolled-up 3D magnetic field vector angular encoder (Reproduced with permission [16]. Copyright 2019, Wiley).

efficiency in saving the footprint area, the unique Swiss-roll structure allows for spatial sensing ability. Fig. 2c shows a 3D magnetic field vector angular encoder comprising a pair of 90° positioned Swiss-roll sensors, which can sense with high accuracy in all three dimensions [16]. Apparently, the advanced 3D micromachining technology opens up a new route towards highly compact and multifunctional microelectronics, and hence facilitates the development of electronic skin with a minimal footprint area, which puts a whole new perspective on cybernetic systems that can precisely monitor physiological signals, stimulate tissues, restore lost functions, and even impose new abilities in human.

2. Power supplies for electronic skin

As is known, electronic devices, regardless of sizes and functions, are powered by electricity. For bulky electronic devices, batteries have been widely used due to their high energy density and stable long-term performance. However, these mainstream batteries are large and rigid, making them incompatible with soft and small sized electronic skin. To match the requirement of electronic skin, batteries should be miniaturized and engineered into an ultrathin form for adequate flexibility. Both miniaturization and thinning of batteries lead to the reduction of electrode materials, which directly results in the significant compromise in energy output of batteries. The resultant flexible and microscale batteries show intolerant mismatch between stored energy and power requirement of electronic skin [18]. As a result, battery-free design becomes prevailing in developing electronic skin [19–27], which has bypassed the disadvantages of energy storage devices [28,29] and has brought wide application of soft energy harvesters that generate electricity through mechanical, solar, and electromagnetic energy (Fig. 3).

Nanogenerators, converting ubiquitous mechanical energy into

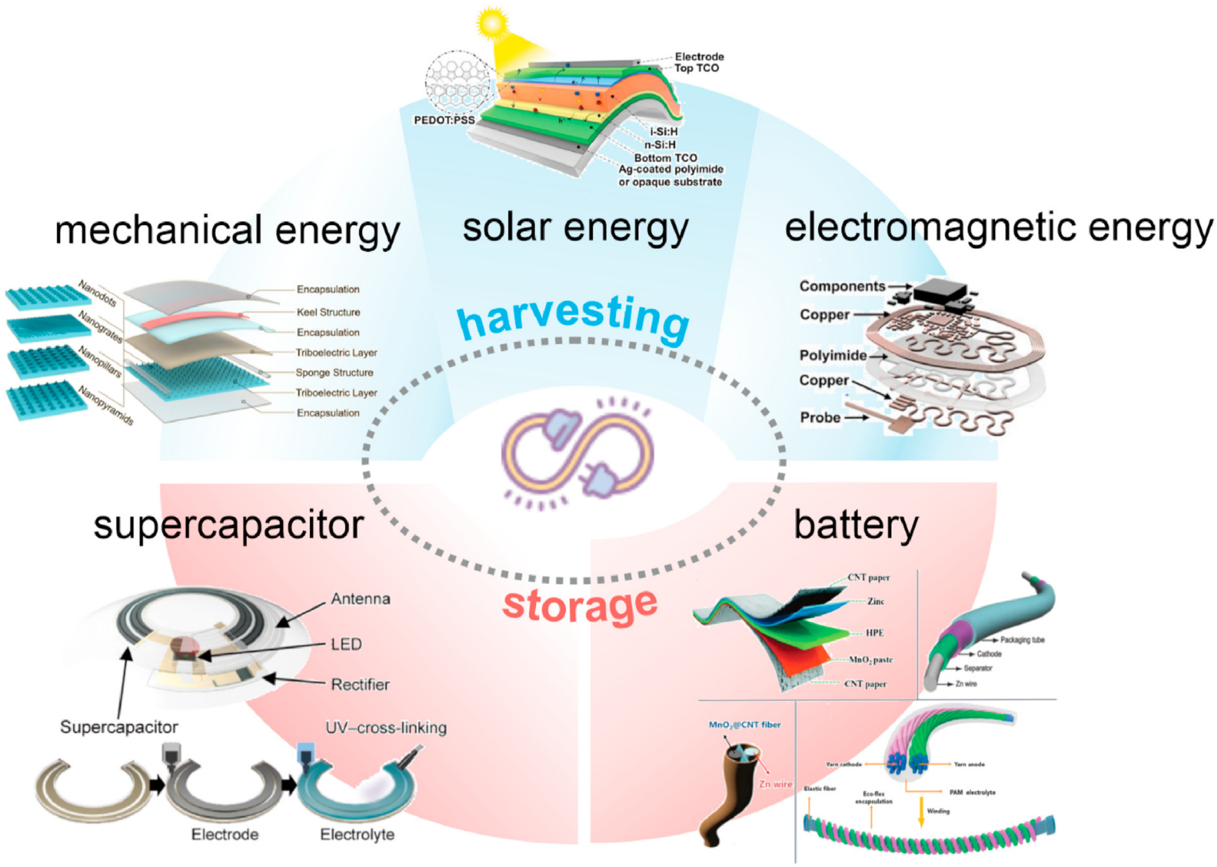

Fig. 3. Direct power supply by harvesting energy from mechanical energy (Reproduced with permission [20]. Copyright 2020, ACS), solar energy (Reproduced with permission [26]. Copyright 2018, Elsevier), and electromagnetic energy (Reproduced with permission [27]. Copyright 2019, NAS). Power supply from energy storage devices: supercapacitors (Reproduced with permission [28]. Copyright 2020, AAAS) and batteries (Reproduced with permission [29]. Copyright 2020, Wiley).

electricity, are based on the piezoelectric effect or the coupling effect of triboelectrification and electrostatic induction. When external force is applied to a piezoelectric/triboelectric nanogenerator, electric field will be established inside the nanogenerator, generating electron flow and hence powering electronic devices [8]. Owing to the principle of energy conversion, the output power of the nanogenerator relies on the frequency of external mechanical deformation. On the one hand, this unique feature allows for inherent sensing ability [20,21]. On the other hand, the dependence on external stimuli raises the concern of the stability of power supply, which will impairs the practicability of the nanogenerator as a reliable power supply for electronic skin. In addition to ubiquitous mechanical energy, solar energy is regarded as abundant and renewable power supplies for electronic skin. Flexible and stretchable photovoltaics that can be adapted to any device/substrate makes self-powered electronic skin come true [22]. For example, an organic solar cell has been integrated with ultraflexible electronics, which is lightweight and comfortable to human skin [23]. Similar to nanogenerators, solar cells are dependent on an external stimuli, incident illumination, and hence offer intermittent power from the view of long-term operation. Apart from direct energy conversion devices, wireless charging, a mature technology, has been used to power electronic devices. By making the coil flexible and reducing its size, the wireless charging modules become flexible, and hence compatible with electronic skin [3,24,25]. However, wireless charging has spatial restrictions, meaning that it, to some extent, impedes the human mobility, which runs counter to the aspiration, extending human abilities, of developing electronic skin for cybernetic systems.

3. Prospects of energy storage devices for electronic skin

Although soft energy harvesting devices have spawned the development of wireless electronic skin, the temporal power supply and spatial restrictions of these technologies limit continuous use of electronic skin. To address this critical issue, energy storage devices, nevertheless, are indispensable for electronic skin. Engineering challenges in materials and devices towards ultraflexible and/or microscale energy storage devices need to be overcome before we can fully exploit the benefits of electronic skin. For miniaturized electronic devices, the power consumption ranges from pW to μW depending on their integrated functions. Accordingly, a long-term operation after one charge process requires the miniaturized energy storage devices to provide energy at the level of μWh. In terms of overall service time, the batteries should be cycled for at least 100 cycles with the energy retention of more than 90%. As such batteries should be integrated in small systems, the battery size should be reduced to the sub-cm^2 scale. Moreover, a skin mounted system needs to be as safe as possible. As a result, the toxic or corrosive electrolyte should be well sealed to prevent any leakage. Alternatively, a safe aqueous electrolyte might circumvent such problem.

Microsupercapacitors have been integrated with a smart contact lens. A prototype microsupercapacitor using carbon materials with a large specific area and an ionic liquid-based electrolyte delivered the energy of ~5 μW h and successfully powered a LED [28]. However, minute-level operation time puts this pioneer energy storage device in flexible electronic devices at the disadvantage of inadequate energy output for continuous use. The fast decay of voltage also limits the output energy [30,31].

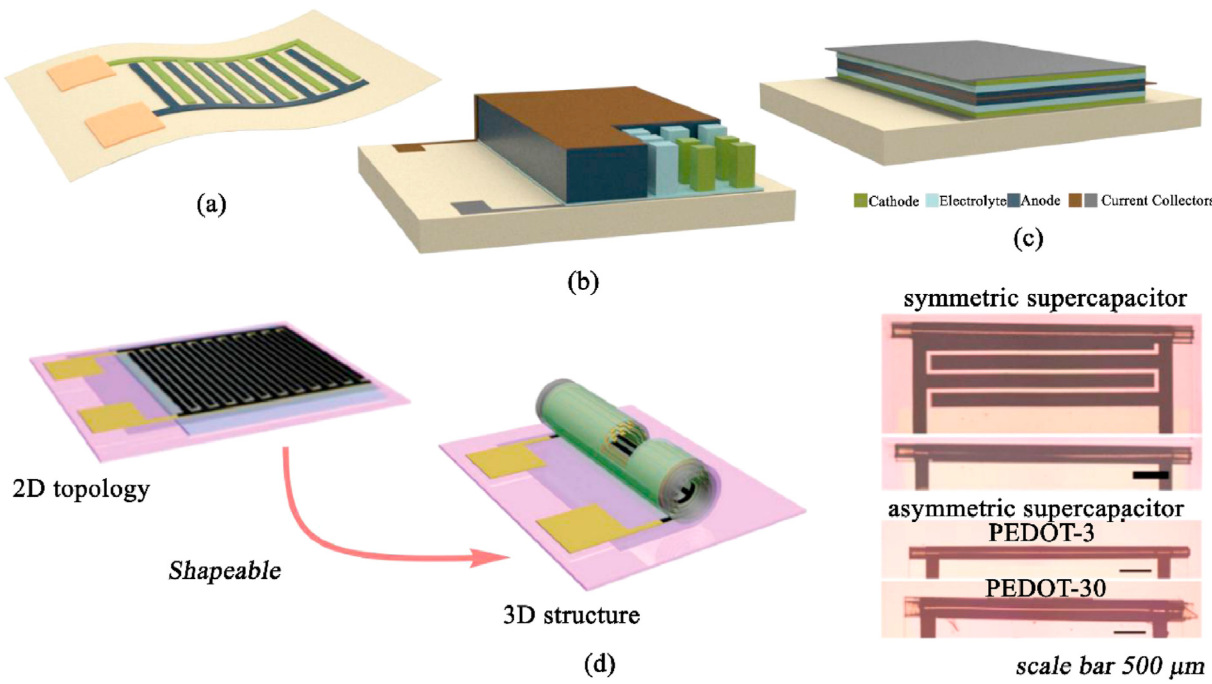

Fig. 4. Typical configurations of microbatteries: (a) interdigitated; (b) interpenetrating; and (c) stacked structure. (d) Reshape thin film devices into 3D Swiss-roll devices (Reproduced with permission [49,50]. Copyright 2019, ACS; Copyright 2019, Wiley).

The baseline of energy output can be elevated by replacing supercapacitors with batteries. However, safety concerns arise when we aim to make batteries skin-mountable. Any toxic substances and explosion risk should be eliminated for a device with intimate contact with human body. As such, the dominant battery chemistry based on lithium ions is not the ideal choice because any leakage of electrolyte and short-circuit in a battery will cause catastrophic results due to toxic and flammable organic solvents [32,33]. To solve this problem, solid-state or polymer-based electrolyte should be adapted to the miniaturized and flexible electronic systems. However, application of the solid-state electrolyte is plagued by its low ionic conductivity and brittleness [34,35]. Flexible polymer-based electrolyte, therefore, hold the promise to improve the safety and energy density of integrated lithium or lithium ion batteries [36,37].

By contrast, aqueous batteries using nontoxic and inflammable electrolyte rise to the challenge of high safety [38]. Despite the high safety, compared with organic electrolytes, aqueous electrolytes are less stable because water can easily evaporate at the room temperature, freeze at subzero temperatures, split when the voltage exceeds its thermodynamically stable window of 1.23 V. Efforts aimed at developing suitable aqueous batteries should be devoted to answer these challenges. For a start, evaporation and freezing of water can be suppressed by using hydrogel electrolytes. Organic frameworks and dissolved ions effectively interact with water molecules, which limit their activity, enhance the stability of electrolyte, and enlarge the working temperature range of aqueous batteries [39]. Furthermore, hydrogel electrolytes usually show excellent flexibility and stretchability and perfectly match the requirement of electronic skin [40,41]. Regarding methods to expand stable voltage window of water, manipulation of solvation is crucial, which also influences the stability of aqueous electrolytes under different temperatures [42]. Therefore, to adapt aqueous batteries for electronic skin, advances in hydrogel electrolyte with flexibility and the optimization of solvation structure need dovetail with each other.

Energy storage abilities largely depend on electrode materials, including their battery chemistry and mass loading. Battery chemistry defines the baseline of energy storage performance while mass loading of electrode materials set upper limit. In terms of batteries for electronic skin, electrodes should be thinned and miniaturized, leading to a limited mass loading of electrode materials. Besides, a thin separator or electrolyte layer increase the risk of short-cut over cycling. To tackle this, the separator or electrolyte layer should provide a high resistance to penetration. Choosing a proper battery chemistry is a central task to improve energy storage ability and to provide high safety. Aqueous batteries, such as alkaline batteries, have already been a strong competitor for lithium ion batteries in our daily lives. Therefore, the successful miniaturization of such microbatteries will be a promising solution. Recently, the primary alkaline battery is transformed into a rechargeable battery by using a monolithic zinc sponge anode [43]. In addition to the new electrode design, the reversibility of aqueous batteries has been dramatically improved by changing the electrolyte to acidic ones [44]. For instance, the Zn–MnO_2 battery can stably run for thousands of cycles. However, such batteries are limited by the capacity (300 mA h/g) and limited voltage (<2 V), which can be addressed by using electrolytic batteries [45]. Electrolysis of MnO_2, for example, maximizes the theoretical capacity of Zn-MnO_2 battery to 600 mA h/g with a high output voltage of approximately 2 V, leading to an ultrahigh energy density of 1.2 W h/g for the cathode material [46]. Moreover, MnO_2 can be deposited on the cathode side during the charging process, partially circumventing the limitation in loading of raw electrode materials [47,48]. As a result, the energy output of such battery can be finely tuned by controlling the charge depth, which enables on-demand power supply for electronic skin.

But transformation of a bulky battery to a skin-mountable battery will also have a bearing on the development of batteries for electronic skin. From a device viewpoint, both flexible batteries and soft miniaturized batteries are feasible. Flexible batteries should be ultrathin to minimize the discomfort when being attached to human skin. Engineering cell constituents, including current collectors, anodes, cathodes, separators, and electrolytes, into an ultrathin system is technologically difficult and introduces high risk of short-circuit. Therefore, the interdigitated design (Fig. 4a) has been developed to circumvent this issue. However, to offer a high energy density, a battery consisting of interdigitated electrodes occupies a large footprint area. Likewise, flexible batteries normally occupy large areas and have limitation in applicable positions on human body, which disobeys the design rule of cybernetic systems where electronic skin is placed in multiple positions and requires discrete energy supplies. In spite of high energy output, a 3D interpenetrated configuration (Fig. 4b) can hardly be thin and soft because a rigid porous template is necessary for the fabrication of interpenetrated electrodes [51]. The tangible progress towards batteries for electronic skin without any adaptability limitation, therefore, is expected to be made by advances in soft and miniaturized batteries. Alternatively, a thin film battery holds the promise of accomplishing soft and miniaturized batteries. One of the biggest challenges facing miniaturized thin film battery is how to enlarge electrode area with the restraint of a minimal footprint area. Apparently, an optimal battery for electronic skin requires a large electrode area and a miniaturized footprint area, which rises as the main obstacle for battery design. To overcome this obstacle, one promising strategy is to stacking multiple electrode and electrolyte layers on top of another and to keep a light weight (Fig. 4c). The challenges of this design rely on the precise laminating process, which otherwise will result in the failure of the whole battery. Besides, the flexibility of the battery after layering down multiple films becomes a tough challenge. Another strategy is to transform thin film to a soft 3D object, which recalls the self-driven rearrangement of a 2D device into a Swiss-roll like device through shapeable materials. Prototypes of Swiss-roll devices have been developed and shown competent energy density (Fig. 4) [49,50]. Soft polymers and ultrathin electrodes in such Swiss-roll energy storage devices are beneficial for flexible and miniaturized batteries. The vision of a flexible, microscale, and high-performance battery serving as a promising energy supply for continuous use of electronic skin relies on the combination of every strength in electrolyte stability, battery chemistry, and novel micromachining technology.

4. Conclusion

Electronic skin, providing continuous health monitoring, point-of-care diagnostics, and sense of touch for robotics, allow for various tasks current not possible, such as cybernetic system that interacts with human body to overcome the limitations of human biology. Despite the exciting prospect of electronic skin, the development of power supplies towards continuous use are lagging. In this regard, this Perspective sets out nano/microscale batteries as an effective way to enable continuous use of electronic skin through synergistic advancements in both battery chemistry and device architecture. We began by giving an outlook of cybernetic system, which is based on electronic skin attached to different human tissues. The complex system requires stable performance in all courses of operation, pointing out the disadvantage of mainstream power supplies used in current electronic skin, such as spatial and temporal restrictions. To overcome these disadvantages, of great importance are soft and miniaturized batteries with high energy output. Last but not least, we discussed prospects for high energy output with the limitation of dimension and requirement of flexibility, providing ideas to develop always-on electronic skin.

Declaration of competing interest

None.

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