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Polymer-based flexible bioelectronics

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ABSTRACT

Due to the mechanical mismatch between conventional rigid electronic devices and soft tissues at nature, a lot of interests have been attracted to develop flexible bioelectronics that work well both in vitro and in vivo. To this end, polymers that can be used for both key components and substrates are indispensable to achieve high performances such as high sensitivity and long-term stability for sensing applications. Here we will summarize the recent advances on the synthesis of a variety of polymers, the design of typical architectures and the integration of different functions for the flexible bioelectronic devices. The remaining challenges and promising directions are highlighted to provide inspirations for the future study on the emerging flexible bioelectronics at end.

1. Introduction

With the rapid development on cooperation studies among materials science [1,2], electronic engineering, artificial intelligence, and biomedical science, there emerges a new and multidisciplinary direction which is named bioelectronics [3–9]. In most cases, it represents the design and application of electronic devices such as sensors to solve the problems of biomedical fields, including both in vitro and in vivo. Obviously, the conventional rigid electronic devices typically based on metal and silicon electrodes cannot effectively meet the requirements of soft tissues mainly in mechanical property such as stiffness. If a rigid device is attached to the skin or implanted into the body, the surrounding tissues may get hurt and the resulting scars will damage or even disable the devices [10]. Out of question, it is well expected that bioelectronic devices should mechanically behave like the soft tissue after being adhered to or implanted. They should be also biocompatible and stable upon contact with biofluids to avoid the coating failure, delamination or corrosion [11]. In the case of their applications in vivo, they would not be peeled off or extracted from the body to avoid second injury after use. In other words, they would better degrade in body or be assimilated/metabolized by the body [12]. In addition, an ideal electronic device should be implanted with minimal traumas [13] (e.g., by injection through the use of a needle) and may accommodate body movements (e.g., it is able to bear stretching, compressing, folding or other deformations) [14]. Furthermore, it is better to easily integrate the other functions such as powering and data transferring in the bioelectronic devices with small sizes [3,15,16], which are vital to the applications both in vitro and in vivo.

To fully satisfy the biomedical applications, increasing interests have been thus attracted to develop the next-generation flexible electronic devices with a variety of functions in recent years. They can be soft, deformable, stable, biocompatible and even degradable to open up a new avenue in the advance of detecting signals or diseases, understanding mechanisms of biological activities, treating diseases and communicating [17]. The effective applications are mainly derived from the good and dynamically matching interfaces [6,16]. For the use of a flexible device in vitro, it can closely attach on curved skin surface with a stable interface and thus works efficiently under moving [18–20]. In the case of the flexible device in vivo, the stable and dynamically matching interfaces between the flexible electronic device and tissues are also found to play a critical role in high biocompatibility [21–23]; In contrast, the stiff implants induce astrocytic scars and microglia populations (Fig. 1).

The effective interfaces are closely related to the use of soft materials for key components or/and substrates, the design of structures and the realization of functions in flexible bioelectronic devices [17]. Polymers are first highlighted due to the fact that they have been typically explored as the promising soft materials, in addition to the basis of the structure and property design. The representative structures and properties are then summarized for the available flexible electronic devices. The remaining challenges and promising directions are finally presented to give an inspiration to the future study in flexible bioelectronics.

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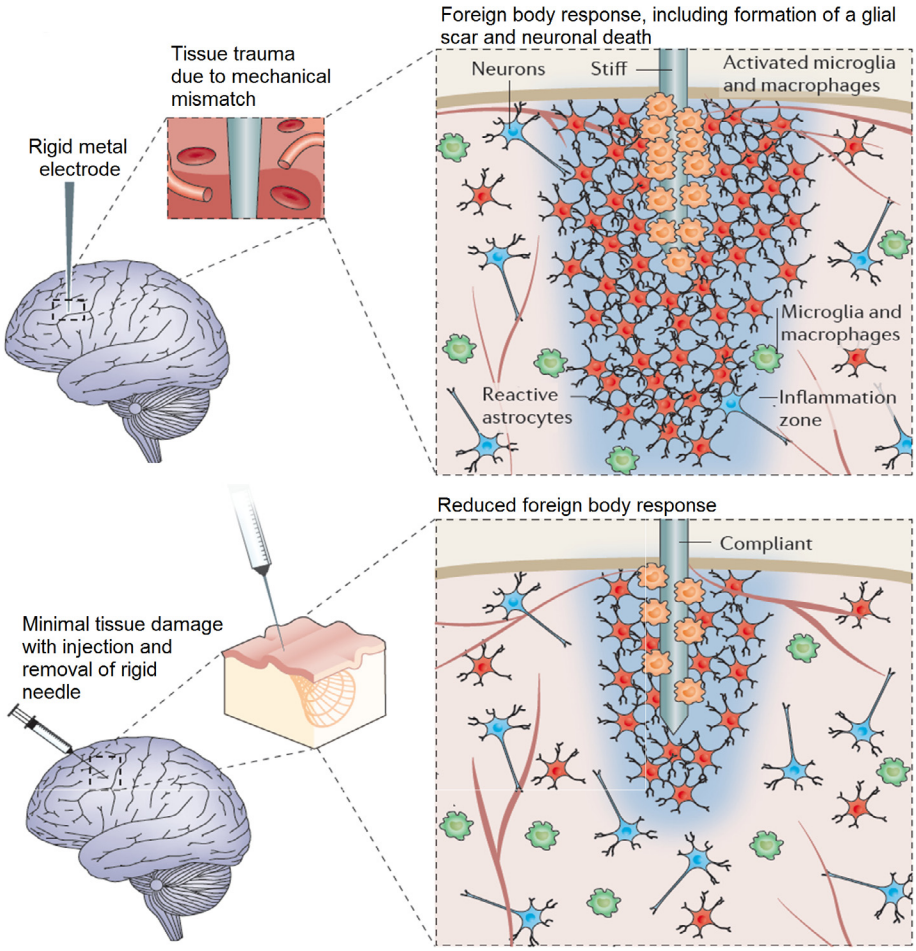

Fig. 1. (Color online) Interface demands for implantable bioelectronics. A schematic comparison on the foreign body response of stiff (top) and compliant (bottom) bioelectronic devices. Reprinted with permission from Ref. [4], Copyright 2017 Nature Publishing Group.

2. Polymer materials

The diversity and synthetic tunability of polymer materials offer them good electrical conductivity for electrodes, optical transparency for substrates and biodegradability for sacrificial layers in bioelectronic devices [24] (Fig. 2a). Polyaniline (PANI), poly(3,4-ethylenedioxythiophene) (PEDOT) and polypyrrole (PPy) are widely used as electrodes [1,25,26]. Generally, if organic compounds including high boiling solvents like dimethyl sulfoxide and methyl pyrrolidone, ionic liquids and surfactants are added, their electrical conductivities may increase to be comparable to indium tin oxide (ITO), a typical inorganic electrode material. Silk, cellulose, collagen, gelatine and other natural polymers with both high biocompatibility and biodegradation are good candidates for sacrificial layers that can degrade and dissolve after implanting, thus minimizing the invasion for better recording or stimulation [27–29]. Meanwhile, synthetic polymers such as polylactic acid, polyethylene glycol and their copolymers are also competitive alternatives as sacrificial layers due to their tuneable degradation rates and good biocompatibility [7]. As moisture and dielectric barriers, polymers like polydimethylsiloxane (PDMS), poly(*p*-xylylene) (mostly used as a trade name of Parylene C), and polyimide (PI) are widely explored as insulation substrates to protect electric components from impairing under direct contacts with biofluids [5,27,30]. These polymers are transparent, flexible and biocompatible without immune responses. Note that polymers are advantageous for large-scale and low-cost preparations through solution processes such as spin coating [31].

Metal and silicon can be in fact made into nanomaterials to realize flexibility on the device level, though they cannot change their intrinsic rigid property, e.g., high elastic modulus of ~100 GPa for silicon. In contrast, polymers show much lower and tuneable elastic moduli ranged from 100 kPa to 10 GPa, which may be designed to match biological tissues (typically 1 kPa to 1 GPa for soft tissues and 10 GPa for hard tissues) to great degree [6,32,33] (Fig. 2b). Compared with silicon, the use of inherently soft polymer materials to directly contact biological tissues may greatly reduce negative reactions for bioelectronic devices owing to the better interfaces between the devices and tissues.

To prevent the failure of bioelectronic devices, it is also necessary to seal them from exposure to warm, aqueous and saline environments associated with the foreign-body response mostly by the use of polymers. Polymers like PDMS and poly(*p*-xylylene) that are commonly explored for clinical applications have been widely investigated as effective sealing layers. If cracks still occur, we may introduce self-healing polymers in advance to bioelectronic devices and restore them with high stability [20,34,35] (Fig. 2c).

3. Structure designs

For human skins, organs and brain skulls with curved surfaces, it is challenging for typical thin-film bioelectronic devices to attach to them seamlessly, which for instance results in acute stimulation or inaccurate detection. Consequently, hydrogels and adhesive polymers may be used to improve compliant contacts by increasing the adhesion force between devices and tissues [3,23].

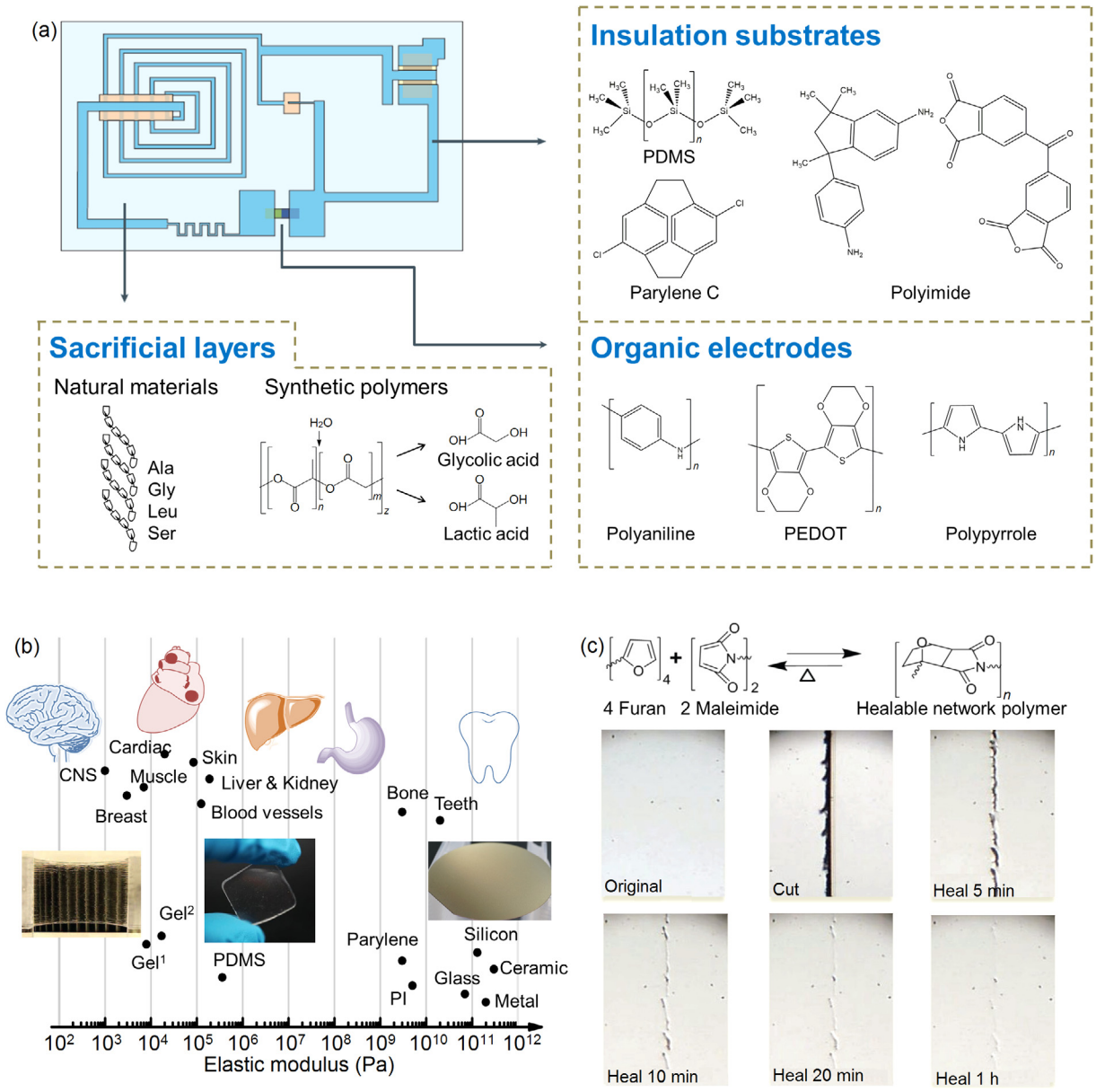

Fig. 2. (Color online) Polymers for soft interface and functionalization. (a) Different components with different polymers of a flexible bioelectronic device. (b) Logarithmic plot on the comparison of elastic moduli for various biological tissues and materials. (c) The realization of self-healing function by the use of self-healing polymer. Reprinted with permission from Refs. [4] and [34], Copyright 2017 Nature Publishing Group and 2013 Wiley-VCH Verlag.

A representative strategy is to modify the structure of the bioelectronic devices by coating soft polymers on rigid electrodes [21,30,36]. For instance, biodegradable poly (lactic-*co*-glycolic acid) was used as the sacrificial layer or substrate, followed by coating electrodes, interconnects and interlayer dielectrics to produce a transient hydration sensor [7]. It might dissolve completely to prevent the second injury caused by peeling off (Fig. 3a). Similarly, we may implant the flexible bioelectronic device by a temporary hard shuttle and then remove it after insertion so that only the soft part remains in vivo. For instance, cellular-scale optoelectronic devices were produced from a thin polymer layer of epoxy and bottom bioresorbable adhesive layer of biopolymer like silk fibroin, which made it possible to remove the rigid microneedle after implanting [4] (Fig. 3b, i). The bioelectronic devices might even be stretchable to adapt to the curved surfaces through the use of elastomeric substrates [37] (Fig. 3b, ii). For the above structure designs, the polymer substrate layer may reduce the mismatch in stiffness between devices and tissues.

Stretchability is key to the bioelectronic devices [14,19,31], e.g., a strain of more than 80% on the knuckle, over 50% on the knee joint and no less than 10% volume change in cardiac tissue [6]. Besides the use of elastomeric substrates, another general and promising strategy is to make them into specific architectures that are stretchable. For the first typical method, Peano curve was designed for the bioelectronic devices that could be stretched although the building blocks themselves were rigid at nature [38] (Fig. 3c). The tensile strains for the wires attached to skin replica with conformable contact exceeded 30% in both horizonal and vertical directions (Fig. 3d, i). For the second typical method, biaxially stretchable devices based on single crystalline silicon were realized by designing a wavy architecture [39] (Fig. 3d, ii). For the third method, a multielectrode array was fabricated on

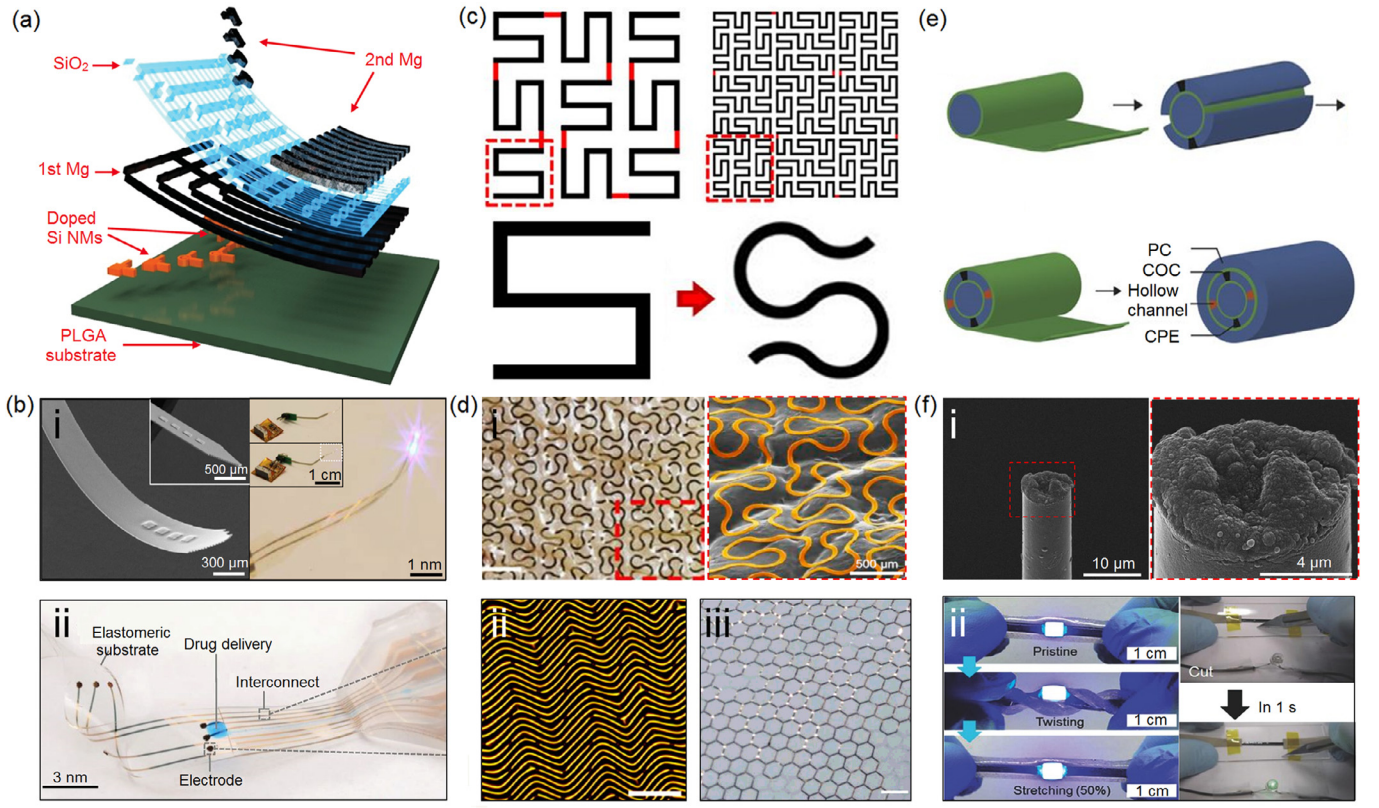

Fig. 3. (Color online) Structure design of flexible bioelectronic devices for dynamically mechanical stability. (a), (b) Structure and demonstration of typical planar compliant bioelectronic devices, respectively. (c), (d) Structure and demonstration of stretchable bioelectronic devices, respectively. (e), (f) Structure and demonstration of cable-like bioelectronic devices, respectively. Reprinted with permission from Refs. [6,2,37–40,46,7] and [25], Copyright Wiley-VCH Verlag, Science Publishing Group, Nature Publishing Group and American Chemical Society.

honeycomb grid-typed Parylene C substrate to become stretchable with good non-thrombogenicity and stability for living rat hearts [40] (Fig. 3d, iii). The stretchable devices may better accommodate deformations particularly in three dimensions for higher dynamic stability.

To more efficiently meet the application requirements for three-dimensional deformations, one-dimensional bioelectronic devices such as fiber-shaped ones have recently attracted increasing interests [41–44]. A fiber device may move back and forth along the axial direction and bear the twisting motion at the radial direction [45]. A variety of fiber-shaped bioelectronic devices based on polymers were fabricated by means of a thermal drawing process that further allows for the integration of multiple polymer materials [46,47]. Generally, those different polymer materials share both glass transition and melting temperatures, e.g., polycarbonate (PC), cyclic olefin copolymer (COC) and conductive polyethylene (CPE) were processed in a single fiber device, which was stable under bending (Fig. 3e). The fiber-shaped bioelectronic devices may be also produced by the typical coating methods. For instance, Parylene C could be deposited onto the surface of a carbon fiber with a core-shell structure to record single-neuron in early chronic experiments [8]. Due to an order of magnitude smaller than traditional recording electrodes, it showed higher compliant property for the central nervous system (Fig. 3f, i). The fiber-shaped bioelectronic devices can be further made by simply mixing conducting polymers, e.g., poly (3,4-ethylenedioxythiophene)-poly (styrenesulfonate), and highly deformable viscoelastic polymers together through a solution process [25]. They were self-healing, foldable and stretchable (Fig. 4f, ii). The fiber-shaped bioelectronic devices are thin typically with sizes of micrometers to millimeters, and they thus share promising and unique advantages to be implanted with minimally invasive operations in the biological tissues.

4. Functions

A spectrum of properties can be integrated into the bioelectronic devices for more functions. Here we highlight them in accordance with the above three typical structures to provide some clues along this boosting direction.

Self-powered electronic devices that can measure biometric signals on Parylene C substrates might be seamlessly attached onto human skins or other tissues [16,48]. The high flexibility was indicated by wrapping them over a spatula rod, and they showed accurate and continuous detection of physiological signals without the need of an external power supply or bulky connecting wires [3] (Fig. 4a). As an application demonstration, the self-powered electronic device was attached to a rat heart with the working model shown in Fig. 4b. Due to the close contact between the heart tissue and the device, very high signal-to-noise ratio was achieved (Fig. 4c).

To further enhance the dynamic conformability with the movement of tissues, a lot of efforts have been made to fabricate stretchable hydrogel-based elastic electronic devices with elastic moduli of kilopascals [23,49]. A typical elastic device here consisted of a highly conductive soft hydrogel as the conductor and an elastic photoresist as the insulating substrate [3] (Fig. 4d). As a result of the highly matchable modulus between the device and tissue, the interfacial impedance was significantly reduced. It showed approximately 30 times higher current-injection density than those of platinum electrodes (Fig. 4f) and stable electrical perfor-

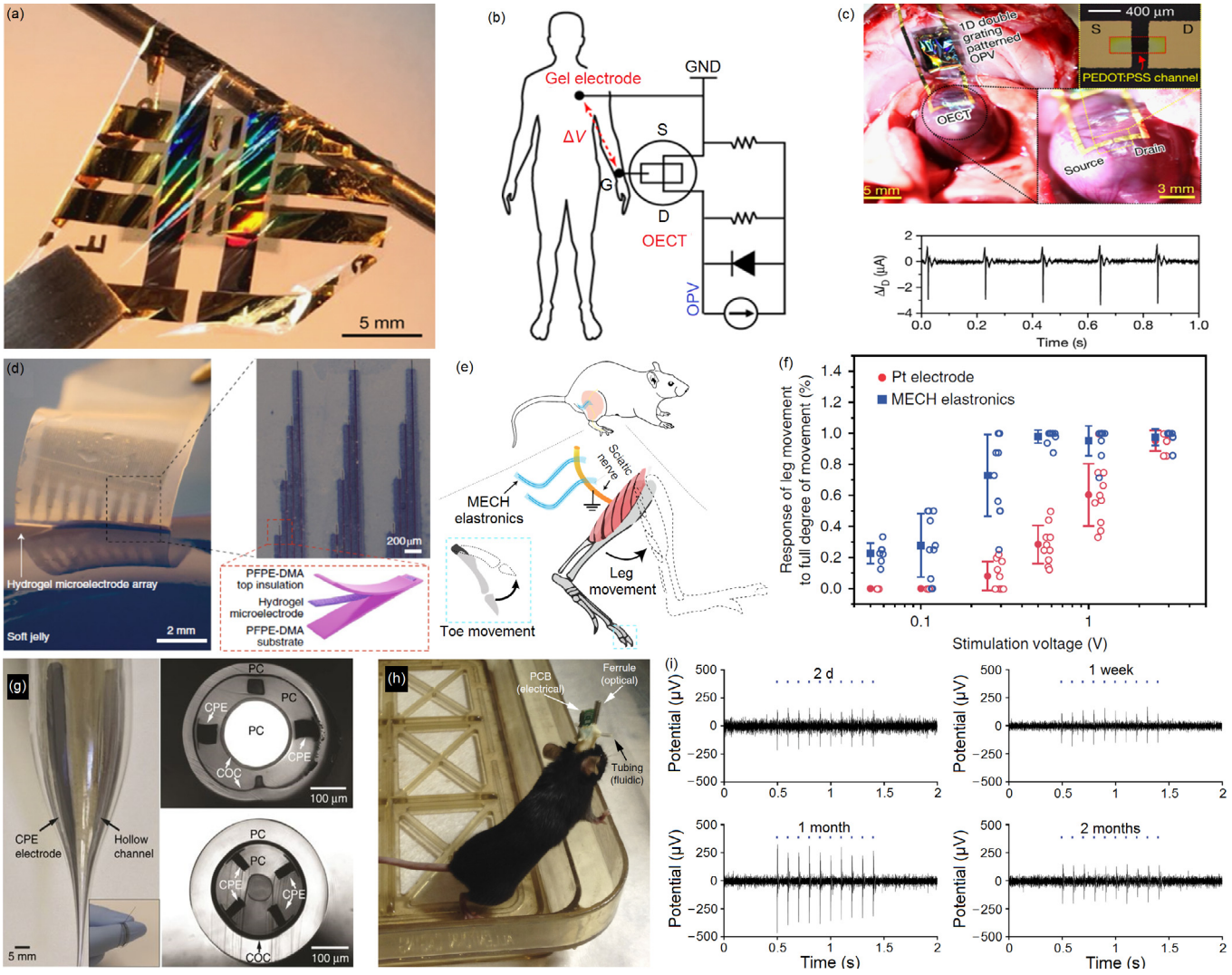

Fig. 4. (Color online) Performance characterization of the polymer-based bioelectronics. Representative illustration of compliant (a)–(c) and stretchable (d)–(f) bioelectronics. (a) Photograph of the flexible self-powered integrated electronic device wrapped over a spatula rod. (b) Scheme of the detecting mechanism. (c) The application to a rat heart with typical photograph (top) and output current signal (right). (d) Photographs and structure scheme of stretchable electrode array. (e) Scheme of the in vivo neural stimulation experiment. (f) The comparison on the percentage of leg movement with respect to the full degree of movement under different stimulation voltages between polymer and metal electrodes with the same exposed area ($n = 9$). (g), (h) Representative illustration of fiber bioelectronics. (g) Photographs of the preparation and cross-sectional structure of a fiber-shaped probe. (h) Photograph of a transgenic mouse with implanted probe. (i) Chronic electrophysiological recording after injection of CNQX during optogenetic stimulation (10 Hz) up to two months. Reprinted with permission from Refs. [1,3] and [46], Copyright Nature Publishing Group.

mances under deformations due to the high volumetric capacitance of the hydrogel materials. These elastic bioelectronic devices could be developed for localized low-voltage electrical stimulation of the sciatic nerve in free-moving animals (Fig. 4e).

Polymer-based bioelectronic devices might be integrated with a variety of functions including optical, electrical and chemical interrogation of neural circuits [50]. Fig. 4g shows the function integration in a fiber-shaped device made by thermal drawing process and its application on moving mice [46]. Different polymers were designed into the fiber-shaped device with high performances. For instance, PC and COC exhibited low absorptions in the visible spectrum, and the differences between their refractive indexes permitted light confinement within PC, whereas CPE served as a recording electrode. The resulting waveguide, microfluidic channels and the electrodes can be adjusted to allow for simultaneous optical stimulation, drug delivery and neural recording in behaving mice with high resolution (Fig. 4h). It had further offered more than two months of optogenetic stimulation, drug perturbation, recordings and analysis of tissue response in vivo (Fig. 4i). The unique features of polymer materials and one-dimensional shape enabled stable brain-machine interfaces between the artificial electronic device and the biological tissue.

5. Outlook

Although a lot of polymer materials and structures have been investigated for the flexible bioelectronic devices, it remains difficult to make quantitative comparisons in flexibility, stretchability and biocompatibility because no general and efficient methods are available for evaluating the different sizes and shapes of devices to date. It is also rare to explore the application standards to assess whether and how they are good enough from a viewpoint of real applications, and to decide what the effective bioelectronic devices should be for a specific application.

For the current studies on bioelectronic devices, the same type of signals has been in fact detected at different sites in vivo [40]. It

is generally tried to find the same site of a rat or mouse based on the experiment accumulations, while it is not accurate. It is even more difficult to accurately repeat the same detecting site for different rats or mice. We need to develop efficient methods to fully repeat the detections with effective data in real applications.

The integration is always a mainstream direction in electronics [48], and it is of utmost importance for bioelectronic devices as they are often implanted in body. The additional functional parts should be reduced or even prevented for minimal invasion traumas. Although the integration of different functions has been attempted for several bioelectronic devices in recent years, great efforts are required to develop more efficient integration methods and introduce more functions. They should be also made as thin as possible to reduce the invasion trauma even after integration.

The applications of bioelectronic devices in brain science have attracted increasing interests just in the past years. It is even more difficult to design bioelectronic devices to satisfy the very soft brain tissue. Although the use of hydrogel or the design of injectable mesh had been proposed for the bioelectronic devices to better match the soft brain tissues, the future breakthrough needs the tight cooperation of synthesizing appropriate polymers and designing effective architectures.

It should be noted that the current bioelectronic devices are typically made at the lab scale. The development of continuous fabrication methods is out of question necessary to really solve the biomedical problems. The quantitative assessment system, repeated signal detection and stimulation control, low-cost materials and standard processing techniques are the necessary prerequisite to realize the commercialization of bioelectronics. Although great efforts have been devoted to these aspects, it deserves much more attentions from the viewpoint of both basic studies and practical applications. It is expected that these polymer-based flexible bioelectronics will be ultimately applied to the clinical and practical fields through unremitting efforts.

Conflict of interest

The authors declare that they have no conflict of interests.

Acknowledgments

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Author contributions

Xiaoying Wu and Huisheng Peng wrote the manuscript.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scib.2019.04.011.

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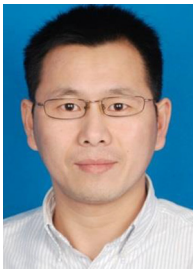

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