small methods
www.small-methods.com

Tissue-Matchable and Implantable Batteries Toward Biomedical Applications

Bing Yan, Yang Zhao, and Huisheng Peng**

Implantable electronic devices can realize real-time and reliable health monitoring, diagnosis, and treatment of human body, which are expected to overcome important bottlenecks in the biomedical field. However, the commonly used energy supply devices for them are implantable batteries based on conventional rigid device design with toxic components, which both mechanically and biologically mismatch soft biological tissues. Therefore, the development of highly soft, safe, and implantable tissue-matchable flexible batteries is of great significance and urgency for implantable bioelectronics. In this work, the recent advances of tissue-matchable and implantable flexible batteries are overviewed, focusing on the design strategies of electrodes/batteries and their biomedical applications. The mechanical flexibility, biocompatibility, and electrochemical performance in vitro and in vivo of these flexible electrodes/batteries are then discussed. Finally, perspectives are provided on the current challenges and possible directions of this field in the future.

1. Introduction

Implantable electronic devices that can monitor,[1–3] diagnose,[4–6] and treat human diseases[7–11] are of great importance for the revolution of biomedical and healthcare fields.[12–16] Implantable bioelectronics have been developed rapidly in the past decades.[17–20] For example, implantable biosensors can realize recordings of internal biophysical and biochemical information in vivo for personalized healthcare.[21–25] In order to ensure stable operations of these implantable electronic devices, sufficient power must be continuously supplied to them. Therefore, implantable power sources that can work stably in vivo are particularly crucial, mainly including energy storage devices (such as batteries[26–34] and supercapacitors[35–39]) and energy harvesting devices (such as biofuel cells,[40] triboelectric,[41–44] piezoelectric[45–47] and thermoelectric devices[48–50]). Among them, batteries are important and promising power sources for implanted electronic devices due to their high energy densities and stable voltage outputs.[49,51]

Tissues in living organisms are mechanically soft, dynamic, curvilinear, and sensitive.[52,53] Unfortunately, the clinically implanted batteries currently used are usually conventional rigid and bulky batteries with Young's moduli of 10^7 to 10^8 kPa,[54,55] which mechanically mismatch soft tissues such as skin and organs (Young's moduli of $<10^2$ kPa).[56,57] The mechanical mismatch between implanted batteries and tissues may lead to unstable battery-tissue interfaces, which caused many potential problems such as foreign body responses and tissue damage.[58] In addition, to avoid damaging vital organs and tissues, the implanted areas of tissue-unmatched batteries are largely limited to a few definite regions such as subcutis.[29,31] The limited implantation region has greatly restricted the development and application range of these implantable batteries in our bodies. Therefore, it is crucial to develop highly flexible and implantable batteries with tissue-matchable softness and compatibility.

In particular, 1D fiber-shaped and 2D thin-film flexible batteries that share tissue-matchable softness are promising candidates for implantable bioelectronic devices.[30,59] In fact, tissues/organs in different parts of the body have quite different softness and biological fluid environments.[56] Therefore, an ideal tissue-matchable flexible battery should simultaneously own high mechanical flexibility and electrochemical stability to effectively match the softness and bio-environments of various tissues and organs. Furthermore, both high biocompatibility and safety are required for implantable flexible batteries. However, most of flexible batteries employ conventional organic electrolytes that are intrinsically toxic and flammable,[60–63] which posed potential safety hazards when implanted in the human body.[54,64] Therefore, to better meet the development and application requirements of fast-growing implantable bioelectronics, it is also critical to design safe and soft batteries

B. Yan, Y. Zhao
Institute of Flexible Electronics and Research and Development Institute of Northwestern Polytechnical University in Shenzhen
Northwestern Polytechnical University
Xi'an 710072, China
E-mail: iamyzhao@nwpu.edu.cn
Y. Zhao
State Key Laboratory of Organic Electronics and Information Displays and Institute of Advanced Materials (IAM)
Nanjing University of Posts and Telecommunications
Nanjing 210023, China
H. Peng
State Key Laboratory of Molecular Engineering of Polymers
Department of Macromolecular Science and Laboratory of Advanced Materials
Fudan University
Shanghai 200438, China
E-mail: penghs@fudan.edu.cn

The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/smtd.202300501

DOI: 10.1002/smtd.202300501

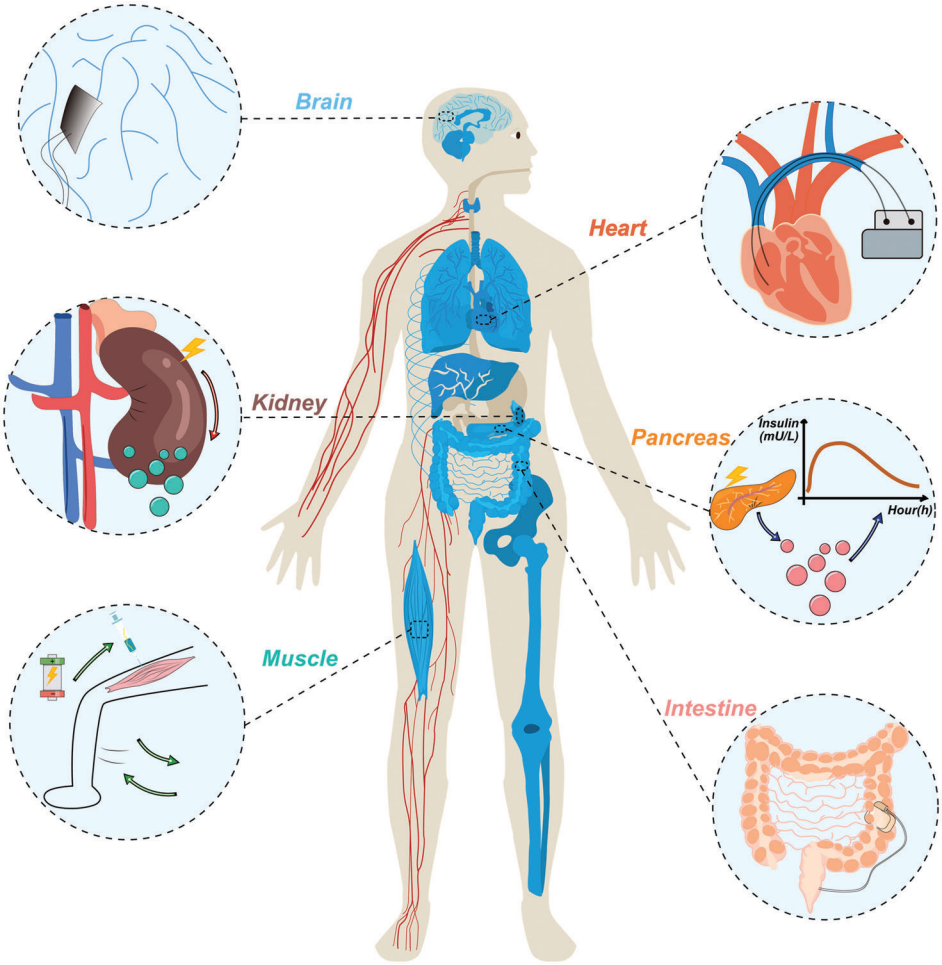

Figure 1. Schematic diagram of the potential implantation regions and application scenarios of tissue-matchable and implantable flexible batteries. Brain: the flexible and implantable pressure sensors powered by tissue-matchable batteries can monitor intracranial pressure for the accurate diagnosis of traumatic brain injury.[6,65] Heart: the pacemakers powered by implanted batteries can treat heart diseases such as chronic arrhythmia.[66] Kidney: the low-intensity electrical stimulation can promote the secretion of catecholamine molecules with anti-inflammatory effects.[67] Pancreas: the insulin release can be regulated by controlling the membrane potential of pancreatic β-cells through implanted bioelectronic devices.[68] Muscle: the precise electrical stimulation of neuromuscular can assist paralyzed patients with functional movements.[69] Intestine: the sacral electrical nerve stimulation by implanted batteries can treat fecal incontinence and constipation.[70] In addition, the biomedical applications of implantable batteries also include modulating cell behavior[31,71] and controlling drug release,[72] etc.

to effectively supply power in different regions of the body (**Figure 1**).

In this review, we aim to summarize the recent advances in the field of tissue-matchable and implantable flexible batteries. First, design strategies and preparations of different kinds of tissue-matchable flexible electrodes are discussed. Second, the structure design, fabrication, flexibility, biocompatibility, and electrochemical performance in vitro and in vivo of various implantable batteries are overviewed and highlighted. The application demonstrations of these batteries in implantable and biomedical fields are also described. Finally, we present the existing challenges and future perspectives in this fast-growing field.

2. Design of Tissue-Matchable Electrodes

Among different implantable power supply devices, batteries are considered the primary power source for driving implantable electronic devices due to their relatively high energy densities and stable voltage outputs.[73–75] Compared with traditional organic batteries, aqueous batteries employ non-toxic and non-flammable aqueous electrolytes with much higher safety,[76–79] which are important candidates for implantable flexible batteries.[80,81] Aqueous flexible batteries were mainly based on lithium-ion batteries,[82,83] sodium-ion batteries,[84,85] zinc batteries,[86,87] magnesium batteries,[88,89] and aluminum-ion batteries.[90,91] According to the device structure, flexible batteries can be mainly divided into 2D thin-film[92–94] and 1D fiber-shaped[95–98] batteries, which have attracted a lot of research attention in recent years. An ideal implantable battery should have a similar or identical softness to that of biological tissues (Young's moduli of $<10^2$ kPa). The key to achieving tissue-matchable and implantable flexible batteries lies in the design of tissue-matched flexible electrodes, which should have ultra-high flexibility (low intrinsic Young's moduli) and safety (high biocompatibility)

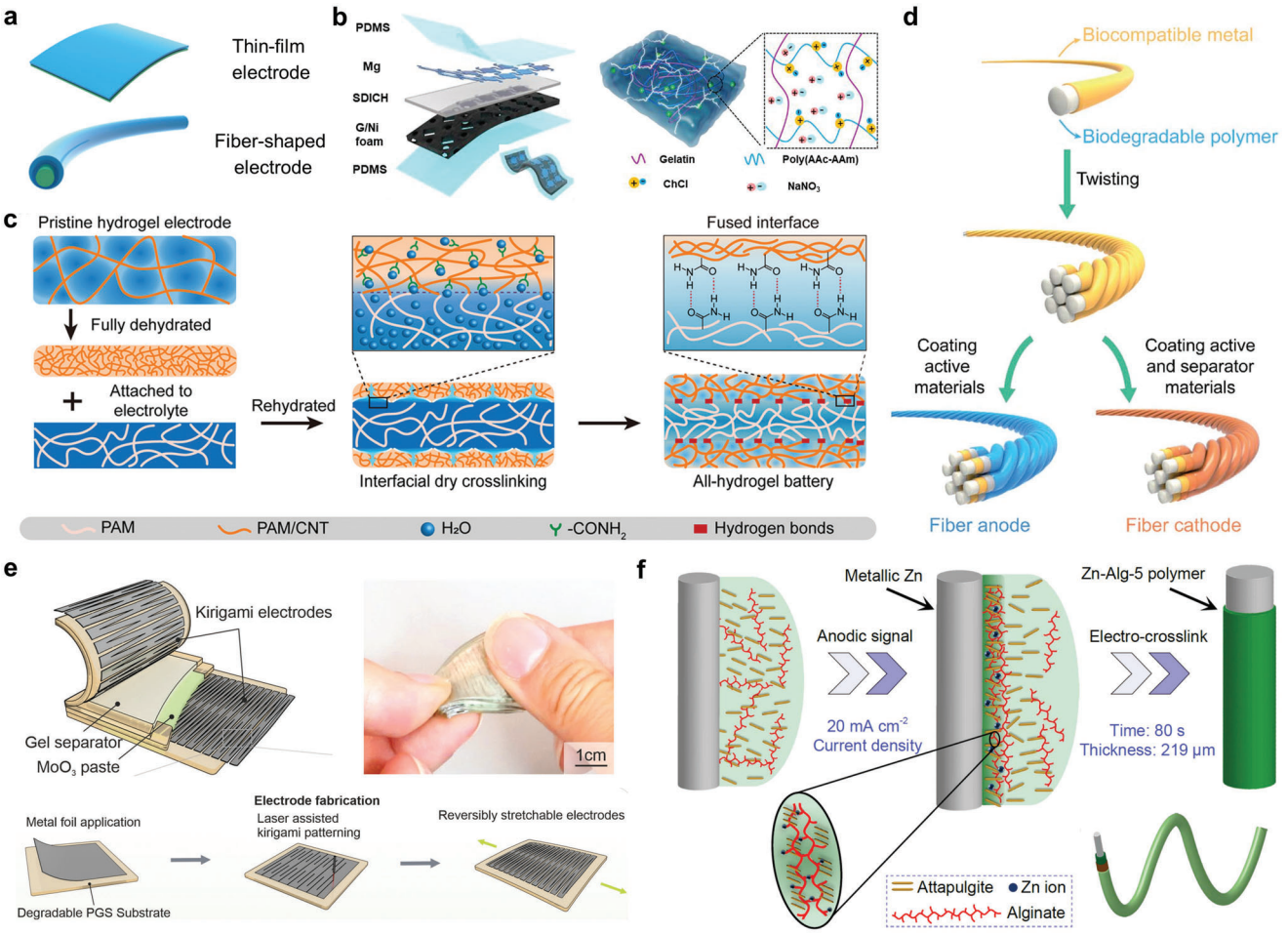

Figure 2. Design and preparation of different types of tissue-matchable electrodes. a) Schematic diagram of typical thin-film and fiber electrodes. Reproduced with permission.[51] Copyright 2022, Wiley-VCH. b) Schematic illustration of a flexible and stretchable thin-film magnesium-air battery with Mg foil and graphene/nickel foam as electrodes. Reproduced with permission.[99] Copyright 2022, Wiley-VCH. c) Preparation processes of all-hydrogel battery based on both hydrogel electrodes and electrolyte. Reproduced with permission.[59] Copyright 2022, Wiley-VCH. d) Schematic diagram of preparation processes of flexible and biodegradable fiber electrodes. Reproduced with permission.[33] Copyright 2021, Royal Society of Chemistry. e) Schematic illustration of preparation process of the pattern stretchable and biodegradable kirigami-patterned electrodes. Reproduced with permission.[105] Copyright 2022, Wiley-VCH. f) Schematic diagram of in situ synthesis of polymer electrolyte on the surface of fiber Zn anode. Reproduced with permission.[100] Copyright 2023, Oxford University Press.

to form stable electrode-tissue interfaces with soft biological tissues.

To realize tissue-matched flexible electrodes, there are generally two main design strategies including structural design and material design. As two typical flexible electrodes, thin-film and fiber-shaped electrodes have their own advantages (**Figure 2**a). Thin-film electrodes can sustain bending deformations and maintain relatively stable electrochemical performances. In addition to ordinary bending deformations, fiber electrodes can withstand complex deformations such as arbitrary twisting and knotting, showing better flexibility and compatibility.[101,102] Particularly, the fiber shape and high softness allow them to form stable electrode-tissue interfaces with vital organs such as heart[30] and brain.[103] In addition, the optimized design of the electrode materials (current collector materials and electrode active materials) is also one of the key factors in designing tissue-matched electrodes. Carbon nanotube fibers and biodegradable polymer films/fibers are highly flexible and biocompatible, allowing a good match with target tissues. Hydrogel materials have a natural advantage in adhering to the surface of the target tissue/organ, which are also good candidates for tissue-matched electrodes/electrolytes.[104]

The key to designing tissue-matched electrodes is to prepare desiring current collector materials and safe electrode active materials. The optional current collector materials and electrode active materials for tissue-matched electrodes may vary, depending on the type of batteries. Magnesium-air batteries display advantages of high theoretical energy density, low cost, and high safety,[106,107] which are promising candidates for wearable and implantable power sources. For instance, a thin-film magnesium-air battery simultaneously with high flexibility, stretchability, and biocompatibility is presented here as a demonstration (Figure 2b).[109] A metallic Mg foil with a thickness of 0.05 mm and graphene/nickel foam was used as the flexible

anode and cathode for the magnesium-air battery, respectively. The flexible electrodes showed a large specific surface area and superior electrical conductivity, which can provide high electrochemical performance for the full battery. A dual-ion-conducting hydrogel was used as the electrolyte to ensure both high flexibility and strength of the magnesium-air battery. Moreover, the hydrogel electrolyte showed high freedom in the design of the polymer networks to regulate migration and transport of ions.

The use of metallic materials usually reduces flexibility of electrodes, thereby reducing their mechanical compatibility with biological tissues. Hydrogel materials show both high flexibility and compatibility with tissues,[108,109] which are usually used as biomedical materials and electrolytes for flexible devices.[110,111] Interestingly, hydrogels are also promising candidates for tissue-matchable electrodes. Ultrasoft all-hydrogel battery had been recently created from both hydrogel electrodes and electrolytes (Figure 2c).[59] Polyacrylamide/carbon nanotube loaded with active materials acted as cathode and anode. The original hydrogel electrode was completely dehydrated to obtain a dehydrated electrode, which was then attached to the hydrogel electrolyte to form a superior fusion interface under the combined effect of osmotic pressure and hydrophilic amide groups. After reaching rehydration equilibrium, the rehydrated hydrogel electrode can be softened again. The obtained hydrogel electrodes exhibited high electrical conductivities and Young's moduli of 70 to 100 kPa, which are well-matched biological tissues (Young's moduli of $<10^2$ kPa).

As alternatives to metal wires, carbon nanotube fibers and polymer fibers are two commonly used flexible substrate materials for tissue-matchable electrodes.[33,112,113] Carbon nanotube fibers display remarkable electrical conductivity and flexibility, but it is challenging to make them into biodegradable electrodes. In contrast, polymer fibers are highly flexible and biocompatible. Their electrical conductivities are typically low and need modifications for use as electrodes. After modifications, flexible polymer fiber electrodes had been designed to biodegrade in vivo (Figure 2d).[33] The biodegradable polymer fiber was first prepared by isotropic twisting of a biodegradable polymer yarn, which was sputtered with a thin layer of gold with a thickness of 80 nm. The biodegradable polymer fibers were then loaded with different active materials to obtain fiber cathode and anode. Without encapsulation, the fiber electrodes were injected directly into the target tissues by a micro syringe, which can effectively avoid the pain and risk of infection for the patients during surgical implantation or device replacement.

Besides biodegradability, stretchability is also an important property for tissue-matchable electrodes, which enables the electrodes with better softness. Interestingly, the stretchable and biodegradable metal electrodes were fabricated by a laser-assisted kirigami-patterning method (Figure 2e), which could achieve over a thousand strain cycles with minimal resistance increase.[105] A biodegradable and elastic polymer of poly(glycerol sebacate) (PGS) was used as the substrate, which was synthesized by a polycondensation reaction. The magnesium and molybdenum metal foils were pressed and adhered well onto the PGS gel. Through laser-assisted patterning, the metal foils were then cut into a designed kirigami pattern to obtain stretchable and biodegradable electrodes.

Among different aqueous battery systems, aqueous zinc batteries are also considered to be promising candidates for future implantable batteries due to their high theoretical specific capacity (820 $mAh \cdot g^{-1}$), high safety, and good stability of metallic zinc.[114,115,116] However, the biosafety design for aqueous zinc batteries in biomedical applications is still a huge challenge. To this end, a biocompatible aqueous zinc battery was made from flexible electrodes and Zn-alginate polymer electrolyte with intrinsic safety (Figure 2f).[100] A fiber Zn electrode and a metal steel foil loaded with MnO_2 active materials were used as anode and cathode, respectively. After applying a certain level of voltage, free Zn^{2+} in the electrolyte was cross-linked with the biomass salt to in situ form polymer electrolyte on the surface of Zn electrode.

3. Tissue-Matchable and Implantable Batteries: Structure, Fabrication and Performance

An ideal tissue-matchable and implantable battery should have tissue-like soft properties, high safety, and high electrochemical stability in complex bio-environments. The key to achieving such batteries lies in the rational design of appropriate battery structure and component materials. Current flexible batteries are mainly divided into 1D fiber-shaped and 2D thin-film batteries according to their shape. Tissue-matchable batteries are usually produced from flexible electrodes, electrolytes, and encapsulation materials with high flexibility, safety, and compatibility. The structure and components of various tissue-matchable and implantable flexible batteries including cathode, anode, separator, electrolyte, binder, and encapsulation materials are shown in **Table 1**. The fabrication and electrochemical performance of different types of tissue-matchable and implantable batteries are carefully discussed in this section.

3.1. Fabrication

Sodium ions are main positive ions in body fluids with inherent safety and biocompatibility. Therefore, aqueous sodium-ion batteries that employ sodium-rich aqueous electrolytes show high safety, so they represent promising candidates for implantable energy storage devices. A fiber-shaped aqueous sodium-ion battery was discovered with high flexibility and safety.[73] The fiber battery was based on a carbon nanotube/$Na_{0.44}MnO_2$ fiber cathode, carbon nanotube/$NaTi_2(PO_4)_3$ fiber anode, and an aqueous Na_2SO_4 electrolyte. Interestingly, when the normal saline (0.9 wt.% NaCl) and cell culture medium (Dulbecco's modified Eagle's medium) were used as electrolytes, the fiber-shaped aqueous sodium-ion batteries still showed effective charge–discharge performance. The high safety and flexibility of the aqueous fiber sodium-ion batteries made them promising for implantable and medical applications.

Carbon nanotube fibers have good mechanical properties, high electrical conductivity, and superior biocompatibility,[119,120] which can be used as current collector materials for implantable fiber electrodes. However, oxygen reduction side reactions usually occur on the interface of anode and oxygen-containing electrolytes, causing rapidly decayed charge/discharge performance and cycle stability of aqueous batteries.[73] Recently, an implantable aqueous sodium-ion fiber battery was designed with desiring properties (**Figure 3**a).[30] The aqueous fiber battery was

Table 1. The structure and component materials of various tissue-matchable and implantable flexible batteries.

Battery type	Structure	Cathode	Anode	Electrolyte	Separator	Binder	Encapsulation	Reference
Li-ion battery	Microchip	$LiCoO_2$	/	LIPON	/	/	Silicon dioxide	[117]
Li-ion battery	Film	$LiMn_2O_4$	$LiTi_2(PO_4)_3$	PAM/LiCl hydrogel	/	PVDF	Parafilm/ hydrogel	[59]
Na-ion battery	Fiber	CNT/$Na_{0.44}MnO_2$	CNT/MoO_3/polypyrrole	Body fluids	Chitosan	/	/	[30]
Na-ion battery	Fiber	Gold/MnO_2	Gold/Polydopamine/ polypyrrole	Body fluids	Chitosan	/	/	[33]
Na-ion battery	Film	$Na_2VTi(PO_4)_3$	$Na_2VTi(PO_4)_3$	Silk fibroin hydrogel electrolytes	/	/	Silk fibroin hydrogel	[26]
Zn-ion battery	Film	α-MnO_2/rGO	Zn	Cellulose aerogel-gelatin electrolyte	/	PTFE	Silk protein	[28]
Zn battery	Film	Polyimide	Zn	Body fluids	Glass fiber	PTFE	/	[118]

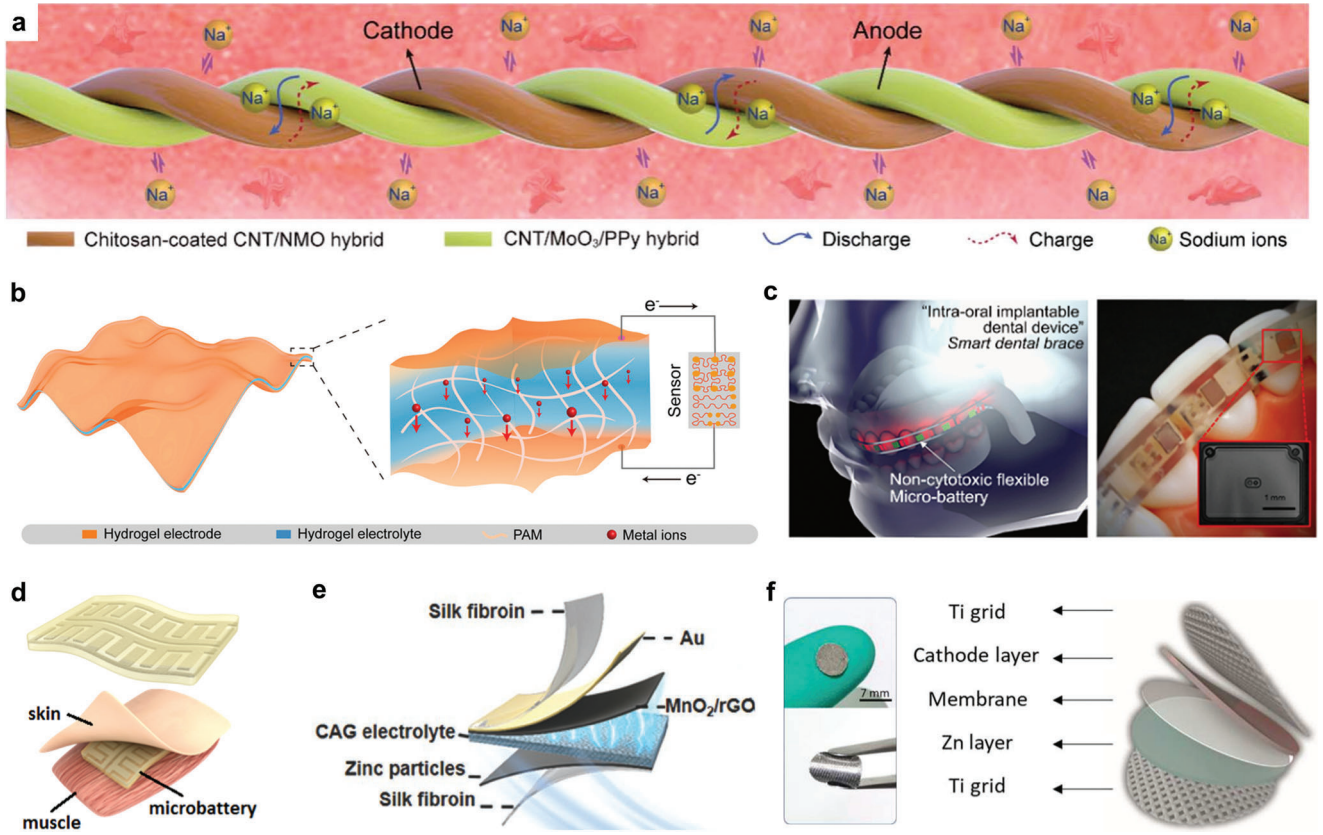

Figure 3. Fabrication of various tissue-matchable and implantable flexible batteries. a) Schematic illustration of working mechanism of flexible and implantable aqueous sodium-ion fiber battery. Reproduced with permission.[30] Copyright 2021, Royal Society of Chemistry. b) Schematic diagram of mechanism of soft all-hydrogel battery. Reproduced with permission.[59] Copyright 2022, Wiley-VCH. c) Schematic diagram and photograph of flexible and implantable thin-film lithium-ion micro-batteries for intra-oral phototherapeutic system. Reproduced with permission.[117] Copyright 2017, Springer Nature. d) Schematic illustration of flexible and implantable thin-film sodium-ion microbattery. Reproduced with permission.[26] Copyright 2018, American Chemical Society. e) Schematic sketch of biodegradable and implantable thin-film zinc-ion battery. Reproduced with permission.[28] Copyright 2022, Wiley-VCH. f) Schematic of flexible and implantable self-charging zinc battery. Reproduced with permission.[118] Copyright 2023, American Association for the Advancement of Science.

fabricated with carbon nanotube fibers loaded with $Na_{0.44}MnO_2$ (NMO) and molybdenum trioxide/polypyridine (MoO_3/PPy) as the cathode and anode, respectively. A layer of chitosan with good biocompatibility was coated on the surface of carbon nanotube/NMO fiber electrode as a separator, which could prevent short circuits during operation of fiber battery. MoO_3 and PPy coated on carbon nanotube fibers were proved to effectively inhibit oxygen reduction side reactions on the anode. The continuous insertion/extraction of sodium ions between fiber electrodes and body fluid ensured stable charging and discharging of the fiber battery in body fluids.

Young's moduli of conventional rigid batteries vary typically between 10^7 and 10^8 kPa, which are intrinsically incompatible with soft tissues (<10^2 kPa). In order to better integrate implantable batteries with biological tissues in a variety of dynamic environments, reducing their intrinsic Young's moduli becomes an effective solution. The ideal state for an implantable flexible battery is to have a similar or identical softness to that of biological tissue. Particularly, an implantable all-hydrogel battery had been successfully constructed with comparable softness to tissue by using an interfacial dry cross-linking strategy (Figure 3b).[59] The hydrogel electrolyte was sandwiched between two dehydrated hydrogel electrodes to fabricate the all-hydrogel battery with both high electrical conductivity and interfacial charge transfer efficiency. After reduction of its intrinsic Young's moduli, the all-hydrogel battery is well-matched soft tissues to ensure the battery works properly in the organism.

Miniaturization is another effective strategy to achieve flexible and implantable batteries. For instance, flexible, biocompatible, and implantable thin-film lithium-ion micro-battery was further explored for intra-oral phototherapeutic systems (Figure 3c).[117] The thin-film battery employed lithium cobalt oxide as a cathode, glass-like solid-state lithium phosphorous oxynitride as electrolyte, and titanium as an anode current collector. Based on the flexible micro-batteries, a pragmatic and highly effective integration strategy was developed for an implantable orthodontic system. Interestingly, a flexible and implantable thin-film symmetric sodium-ion microbattery was also developed based on the heteronanomat bifunctional electrodes and biocompatible electrolyte (Figure 3d),[26] showing good reliability and performance in vivo.

Traditional implantable batteries need to be removed from the body through a second surgery after completing their missions, which can lead to additional safety risks. Particularly, implanted batteries that degrade at a controlled rate can effectively reduce potential risks such as chronic inflammation, patient pain, and healthcare cost. For example, a biodegradable and implantable thin-film zinc-ion battery was developed based on cellulose aerogel-gelatin solid electrolyte (Figure 3e).[28] The complete degradation of the zinc-ion battery occurred within 30 days in buffered proteinase K solution. Similarly, a biodegradable thin-film sodium-ion battery was reported by using biodegradable materials as battery components, which can decompose in nature into non-toxic compounds or elements via hydrolysis and/or fungal degradation.[121] Recently, a flexible and implantable self-charging zinc battery was developed based on a biocompatible polyimide electrode and zinc electrode (Figure 3f).[118] Particularly, the battery could modulate the hypoxia level in tumors by consuming oxygen during discharge/charge cycles for antitumor therapy. In addition to the electrode materials included in Table 1, other metals (such as magnesium) and conductive polymers are also potential candidates for tissue-matchable and implantable flexible batteries.[99]

3.2. Electrochemical Performance In Vitro

Electrochemical performance is key for all implantable batteries. The all-hydrogel lithium-ion battery demonstrated a discharge voltage plateau of 1.5 V and a discharge specific capacity of 82 $mAh{\cdot}g^{-1}$ at the current density of 0.5 $A{\cdot}g^{-1}$ (**Figure 4**a).[59] The lithium-ion full battery also demonstrated good cycling performance with 65% capacity retention after 400 charge–discharge cycles at 1 $A{\cdot}g^{-1}$. When the hydrogel electrodes were loaded with $NH_4V_4O_{10}$ and zinc nanosheets as active materials, the resulting all-hydrogel zinc-ion battery was also able to work normally. The zinc-ion full battery showed a discharge-specific capacity of 370 $mAh{\cdot}g^{-1}$ at a current density of 0.5 $A{\cdot}g^{-1}$, and 73% of capacity was maintained after 200 cycles at 2 $A{\cdot}g^{-1}$. During bending, stretching, twisting, and shearing each for 5000 cycles, the capacity of the all-hydrogel battery remained stable without obvious loss of capacity (Figure 4b), demonstrating superior adaptability and stability under complex and variable conditions.

Fiber-shaped batteries also exhibit superior electrochemical performance in vitro. For example, the biocompatible fiber-shaped aqueous zinc batteries (Figure 4c) based on safe Zn-alginate polymer electrolyte showed a specific capacity of 206 $mAh{\cdot}g^{-1}$ at current density of 1 $A{\cdot}g^{-1}$, with capacity retention of 95% after 100 charge–discharge cycles (Figure 4d).[100] The aqueous zinc battery also showed high capacity retention under different bending states (Figure 4e). The bent fiber zinc batteries were able to light up a small light-emitting device stably (Figure 4f). The implantable aqueous sodium-ion fiber battery achieved a discharge-specific capacity of 47.52 $mAh{\cdot}g^{-1}$ in an electrolyte of physiological saline at 1 $A{\cdot}g^{-1}$ (Figure 4g).[30] The aqueous sodium-ion full battery also showed good rate capability and high cycling performance (Figure 4h,i). The biodegradable and unencapsulated fiber-shaped battery demonstrated a decent specific capacity of 24.4 $mAh{\cdot}g^{-1}$ at 1 $A{\cdot}g^{-1}$ (Figure 4j).[33] The capacity continuously decreased for two weeks due to the degradation of the battery (Figure 4k), and the degradation rate and stable operation interval could be regulated through rational design of biodegradable polymers. After 1000 cycles of bending at a bending angle of 180°, 89.1% of the capacity was maintained for the fiber battery (Figure 4l).

3.3. Electrochemical Performance In Vivo

Due to the unique 1D shape and high flexibility, fiber-shaped batteries can be implanted into the body via a miniature syringe injection process (**Figure 5**a). The ability to maintain stable electrochemical performance in various implantation regions with different biochemical environments is of paramount importance for implantable batteries. Particularly, the unencapsulated aqueous fiber-shaped sodium-ion battery can be injected into the subcutaneous, cardiac, and brain regions of mice (Figure 5b), and they showed close contact with tissues.[30] The implanted sodium-ion fiber-shaped batteries presented discharge-specific capacities of

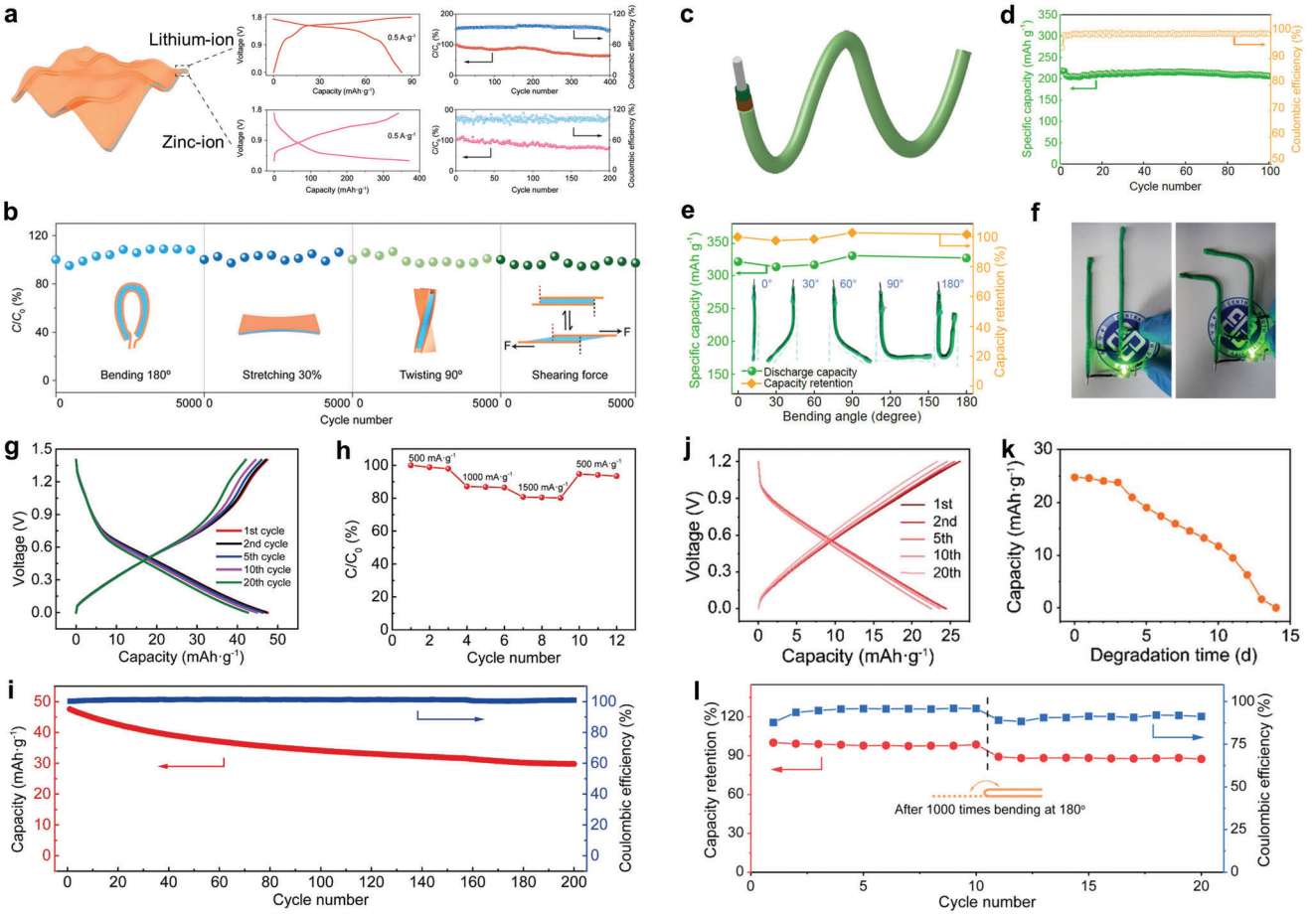

Figure 4. Electrochemical performances of tissue-matchable batteries in vitro. a) Galvanostatic charge–discharge curves and cycling performance of all-hydrogel lithium-ion and zinc-ion batteries. b) Capacity retention of all-hydrogel battery under various bending, stretching, twisting, and shearing cycles. Reproduced with permission.[59] Copyright 2022, Wiley-VCH. c,d) Schematic diagram and cycling performance of biocompatible fiber-shaped aqueous zinc battery. e) Capacity retention of aqueous zinc battery under different bending angles. f) Photographs of fiber-shaped aqueous zinc batteries before and after bending at 90°. Reproduced with permission.[100] Copyright 2023, Oxford University Press. g,h) Galvanostatic charge–discharge curves and rate performance of implantable aqueous fiber-shaped sodium-ion batteries in the electrolyte of physiological saline. i) Cycling performance of aqueous fiber-shaped sodium-ion battery at current density of 1 $A \cdot g^{-1}$. Reproduced with permission.[30] Copyright 2021, Royal Society of Chemistry. j) Galvanostatic charge–discharge curves of biodegradable fiber-shaped battery at 1 $A \cdot g^{-1}$. k) Capacity retention of biodegradable fiber-shaped battery on increasing degradation time at 1 $A \cdot g^{-1}$. l) Capacity retention rate of biodegradable fiber-shaped battery before and after 1000 bending cycles at 180°. Reproduced with permission.[33] Copyright 2021, Royal Society of Chemistry.

43.05, 39.36, and 33.53 $mAh \cdot g^{-1}$ in above three tissues at 1 $A \cdot g^{-1}$, demonstrating superior charge–discharge performances in different regions in vivo.

Due to the potential inflammatory risk, psychological stress, and pain of patients caused by the second surgery for battery retrieval, implanted batteries may be designed to entirely biodegrade after completing the power supply mission in vivo. In particular, a biodegradable fiber-shaped battery could be injected into the body of a mouse with minor wound of only ≈300 μm in size, and part of fiber electrodes could be extracted outside the body for easy charging when necessary (Figure 5c).[33] The implanted biodegradable fiber-shaped battery showed normal open circuit voltage without a short circuit (Figure 5d) and exhibited a discharge-specific capacity of 25.6 $mAh \cdot g^{-1}$ at current density of 1 $A \cdot g^{-1}$ in vivo (Figure 5e). The discharge-specific capacity of implanted fiber-shaped battery was tested to be 28.7 and 20.6 mAh g^{-1} at current densities of 0.5 and 1.5 $A \cdot g^{-1}$ (Figure 5f), respectively. The biodegradable fiber battery achieved a capacity retention rate of 69.1% after 200 charge–discharge cycles at 1 $A \cdot g^{-1}$ (Figure 5g). The fiber battery completely disappeared after being implanted for ten weeks. The fabrication approach, electrochemical performance, and advantages/disadvantages of various tissue-matchable and implantable flexible batteries are summarized in **Table 2**.

4. Applications in Biomedical Fields

In the near future, more and more bioelectronic devices will be implanted in people's bodies, and implanted batteries will be one of the main energy supply devices for them. Tissue-matchable and implantable flexible batteries are expected to realize various functions including modulating cell behavior, controlling drug release, applying electrical stimulation, and treating diseases, etc. However, high biocompatibility and safety of these batteries are

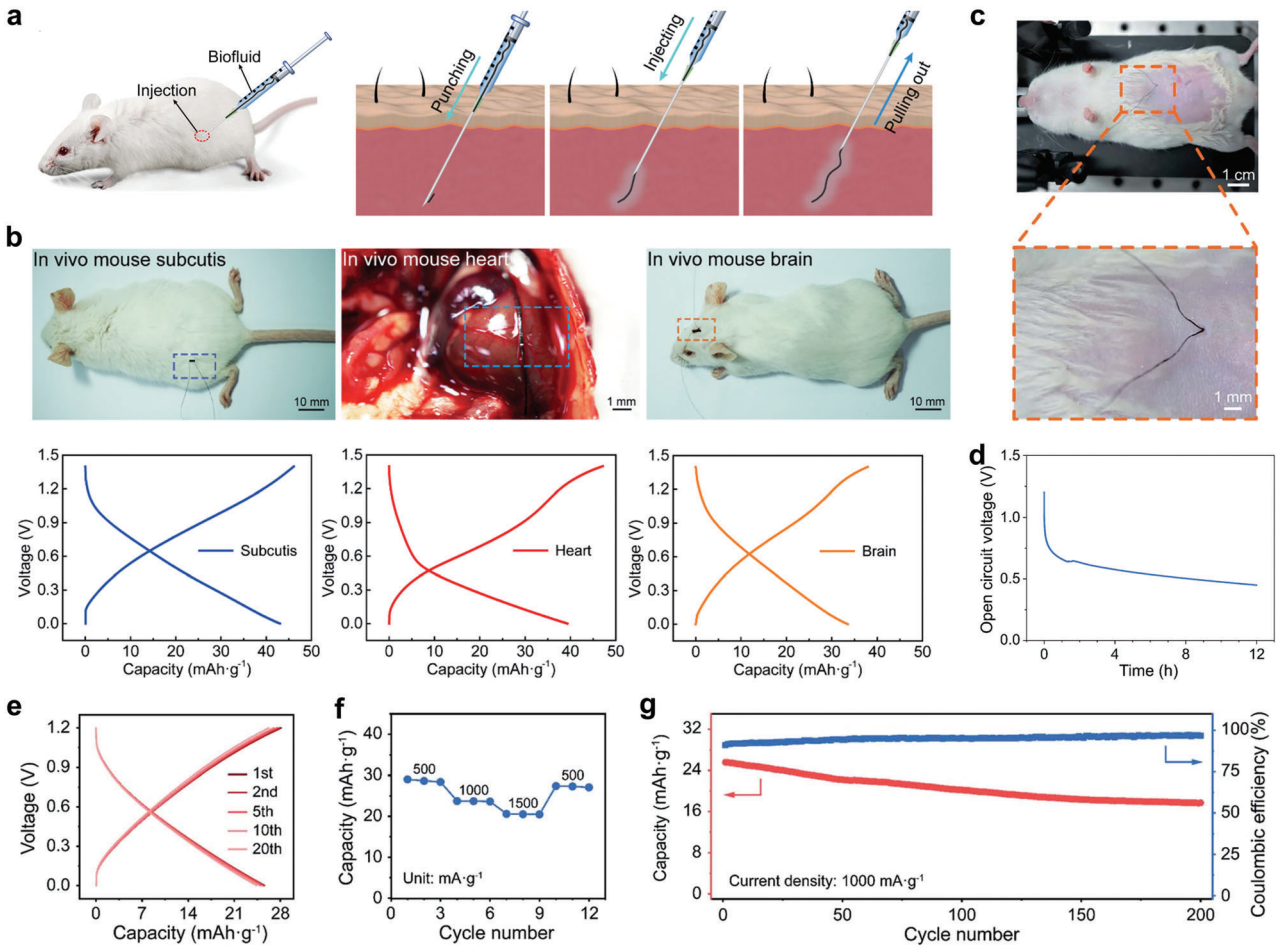

Figure 5. Electrochemical performance of tissue-matchable and implantable flexible batteries in vivo. a) Schematic illustration of the syringe injection process of fiber-shaped battery. b) Photographs and corresponding galvanostatic charge–discharge curves of aqueous fiber-shaped sodium-ion batteries in subcutis, heart, and brain of mice. Reproduced with permission.[30] Copyright 2021, Royal Society of Chemistry. c) Photographs of an injected biodegradable fiber-shaped battery in the body of a mouse. d) Open circuit voltage of the implanted biodegradable fiber-shaped battery. e–g) Galvanostatic charge–discharge curves, rate capability, and cycling performance of biodegradable fiber-shaped battery in vivo. Reproduced with permission.[33] Copyright 2021, Royal Society of Chemistry.

prerequisites for their further practical biomedical applications, which need to be carefully evaluated. Therefore, this section summarizes the biocompatibility studies and biomedical applications of tissue-matchable and implantable flexible batteries.

4.1. Biocompatibility

Biocompatibility of implantable batteries determines whether serious rejection reactions will occur after they are implanted into the body, which is undoubtedly important for their biomedical applications. Therefore, biocompatibility studies of batteries are essential and critical before further clinical medical applications can be carried out.

All-hydrogel lithium-ion battery was ultrasoft and could form a close and steady contact interface with the heart (**Figure 6**a).[59] The potential risk posed by implantable batteries to vital organs is an important aspect to evaluate their biocompatibility, which will largely influence further clinical medical applications. One month after the implantation of the all-hydrogel lithium-ion battery into the mice, there was no noticeable pathological change in the hematoxylin and eosin (H&E) stained tissue sections of the major organs (such as heart, liver, spleen, lung, and kidney) compared to the blank control group (Figure 6b). Besides, various indicators of the organ-specific disease and blood levels of different enzymes for the implanted group were found to be within the confidence intervals of normal values of those of control group (Figure 6c). These results demonstrated high biocompatibility of all-hydrogel batteries, indicating their possibility to power implantable bioelectronic devices.

The non-encapsulated aqueous fiber-shaped sodium-ion batteries showed superior biocompatibility and non-toxicity (Figure 6d).[30] After the implantation of fiber batteries for 30 days, no significant difference was found in the H&E stained tissue sections of the subcutaneous tissues compared to control group without a fiber-shaped battery (Figure 6e), demonstrating the high integration and compatibility between the fiber battery and tissues. Furthermore, the immune response fluorescence

Table 2. The fabrication approach and performance of various tissue-matchable and implantable flexible batteries.

Battery type	Structure	Fabrication approach	Electrochemical performance	Advantage	Disadvantage	Reference
Li-ion battery	Microchip	XeF_2 etching	146 μAh·cm^{-2} at 130 μA·cm^{-2} in vitro	○ High operating voltage	○ Complex fabrication approach	[117]
Li-ion battery	Film	Cross-linking reaction occurred with stirring	82 mAh·g^{-1} at 0.5 A·g^{-1} in vitro	○ Highly soft ○ High specific capacity	○ Water retention performance	[59]
Na-ion battery	Fiber	Electrodeposition	Power density of 78.9 mW·cm^{-3} in vivo	○ High flexibility ○ Mini-invasive syringe injection method	○ Low specific capacity and operating voltage	[30]
Na-ion battery	Fiber	Electrodeposition	25.6 mAh·g^{-1} at 1 A·g^{-1} in vivo	○ Biodegradability ○ Mini-invasively injection method	○ Performance degradation during use	[33]
Na-ion battery	Film	Electrospinning and electrospraying	53.3 mAh·g^{-1} at 1C in vitro	○ High stability	○ Limited flexibility	[26]
Zn-ion battery	Film	Hydrothermal method	211.5 mAh·g^{-1} at 61.6 mA·g^{-1} in vitro	○ High specific capacity ○ Biodegradability	○ Limited flexibility	[28]
Zn battery	Film	Liquid phase synthesis	≈120 mAh·g^{-1} at 1 A·g^{-1} in vitro	○ Regulate the microenvironment of tumor	○ High cost for large-scale production	[118]

staining signals also showed no significant difference between the subcutaneous tissues with/without implanted fiber-shaped battery after 30 days, proving the high biocompatibility of the implanted fiber battery. After implantation of the fiber batteries for 30 days, no carbon nanotube fragment was found in H&E-stained tissue sections of major organs of mice (Figure 6f), proving the non-toxicity, reliability, and structure stability of the implanted fiber-shaped batteries.

Biocompatibility and safety of biodegradable batteries are crucial for their implantable applications. The biodegradable and implantable thin-film zinc-ion battery was implanted into the subcutaneous region of rats for 30 days (Figure 6g).[28] During the month after implantation, the rats showed no signs of disease or obvious debilitation. The fluorescence images of RAW 264.7 cells incubated with the four battery materials also showed no significant difference in cell viability (Figure 6h), proving the good biocompatibility of the biodegradable thin-film zinc-ion battery. Even at different states of charge, the biodegradable fiber-shaped battery demonstrated negligible toxicity and side effects as revealed by the H&E stained sections of subcutaneous tissues at implantation sites (Figure 6i),[33] suggesting good biocompatibility. Due to the employment of biocompatible battery components, the implantable self-charging battery also showed good biocompatibility (Figure 6j).[118]

4.2. Biomedical Applications

In the near future, implantable bioelectronic devices with different functions (such as biosensors) will be implanted in different positions of the human body. Therefore, the corresponding implantable batteries should meet the power supply requirements in various regions of the human body. Due to the non-encapsulated design, 1D architecture, high flexibility, and small size, aqueous fiber-shaped sodium-ion battery could be injected into three typical regions including subcutis, heart, and brain of the body through an injection process (**Figure 7**a),[30] which was particularly promising for various potential biomedical applications. Similarly, the biodegradable fiber-shaped battery was successfully implanted into the subcutis of a mouse (Figure 7b), which could power an implanted fiber-shaped sensor to effectively detect external pressure changes in the implanted area (Figure 7c).[33]

As an application demonstration, an aqueous fiber-shaped sodium-ion battery was implanted into the subcutis of a mouse to efficiently power an implanted sensor by a minimally invasive injection process (Figure 7d), which realized accurate monitoring of the mouse respiration under relaxed and frightened conditions (Figure 7e).[30] It is crucial to efficiently charge implanted batteries. The implanted tissue-matchable batteries can be charged through the extracted flexible electrodes outside the skin. However, this charging method is inconvenient and has potential safety risks. It is desired to develop new charging methods for them with no percutaneous leads and a low risk of infection. Particularly, wireless charging technology can transmit energy through inductive coupling between the transmitter coils outside the body and the receiver coils inside the body.[49] The efficiency of energy transfer is affected by the distance, resonance frequency, and alignment between transmitter and receiver coils, which varies in different environments.[122] Therefore, the effective integration between implantable flexible batteries and miniaturized flexible inductive coupling or radio-frequency energy harvesting devices enables efficient charging of implantable flexible batteries wirelessly.

The integration between all-hydrogel lithium-ion battery and hydrogel strain sensor was achieved by attaching them to the surface of the heart to monitor heartbeat signals (Figure 7f).[59] The obtained detection system was soft enough to form close and steady contact with the heart, enabling accurate monitoring of heartbeat signals without voltage fluctuation of the all-hydrogel battery (Figure 7g). Interestingly, the implantable self-charging zinc battery could continuously consume oxygen

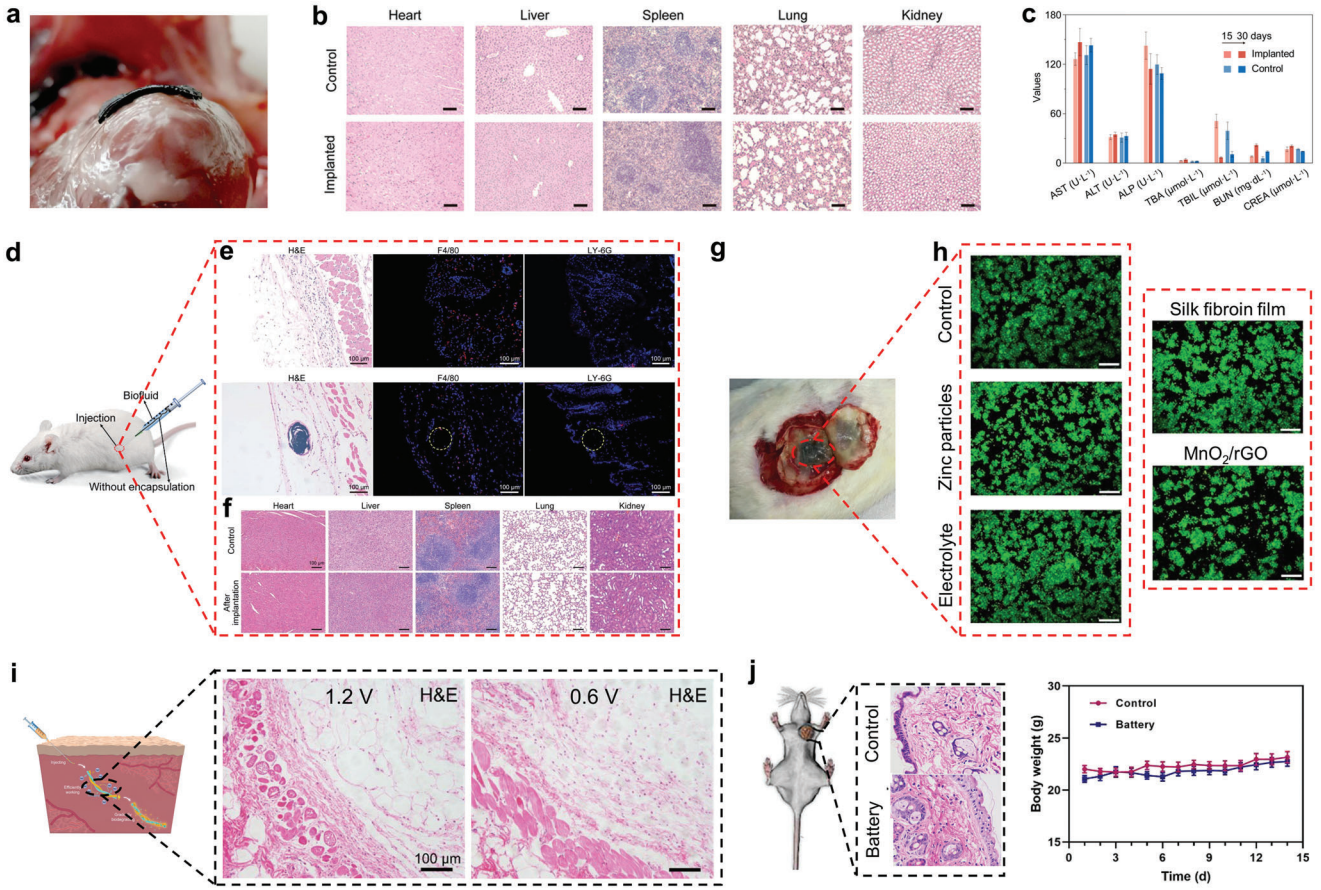

Figure 6. Biocompatibility of tissue-matchable and implantable flexible batteries. a) Photograph showing the interface between all-hydrogel battery and heart. b) Representative H&E staining images of major organs of control and all-hydrogel battery group after implanted for 30 days. c) Changes in the indicators of organ-specific diseases and blood levels of enzymes and electrolytes after implantation of all-hydrogel batteries. Reproduced with permission.[59] Copyright 2022, Wiley-VCH. d) Schematic illustration of injection of aqueous fiber-shaped sodium-ion battery into the body through a syringe. e,f) H&E stainings, F4/80 and LY-6G immunofluorescence stainings of subcutaneous tissues, and H&E-stained tissue sections of major organs of mice without and with aqueous fiber-shaped sodium-ion batteries after implanted for 30 days. Reproduced with permission.[30] Copyright 2021, Royal Society of Chemistry. g) Photograph of the implanted biodegradable thin-film zinc-ion battery in the subcutaneous region of a rat. h) Fluorescence images of RAW 264.7 cells incubated with the four materials of thin-film zinc-ion battery. Reproduced with permission.[28] Copyright 2022, Wiley-VCH. i) H&E stained sections of subcutaneous tissues at implantation sites of biodegradable fiber-shaped battery at different states of charge. Reproduced with permission.[33] Copyright 2021, Royal Society of Chemistry. j) H&E staining images of tumor tissues implanted with a self-charging battery and the corresponding average body weight change of mice. Reproduced with permission.[118] Copyright 2023, American Association for the Advancement of Science.

during the discharge/self-charge cycles to create a tumorous hypoxic environment for effective antitumor therapy (Figure 7h). The intratumoral hypoxic conditions were helpful for the hypoxia-activated prodrugs to kill tumor cells, and the produced reactive oxygen species during the battery reaction could prevent the formation of tumors, showing a notable tumorigenesis suppression effect.[118,11] Therefore, the biomedical applications of tissue-matchable and implantable batteries mainly include supplying power, modulating cell behavior, controlling drug release, applying electrical stimulation, and treating diseases.

5. Perspectives

Since the first pacemaker was successfully implanted in the human body in 1958, many attempts have been made in the field of implantable electronic devices, which have made a significant contribution to saving lives. As the key energy supply unit, implantable batteries are crucial for steady operation of implanted electronic devices in the body. Conventional rigid and bulky batteries mechanically mismatch soft tissues and employ toxic electrolytes, posing potential immune rejection, tissue damage, and safety risks. Therefore, tissue-matchable batteries with tissue-like soft properties, high safety, and high stability in complex bio-environments are ideal candidates for implantable batteries.

The recent advances in tissue-matchable and implantable flexible batteries have been systematically summarized in this review, including injectable aqueous fiber-shaped batteries, all-hydrogel thin-film batteries, thin-film sodium-ion microbattery, biodegradable thin-film zinc-ion battery, biodegradable aqueous fiber-shaped batteries, and self-charging zinc battery. These tissue-matchable batteries exhibited high flexibility, good biocompatibility, and superior electrochemical perfor-

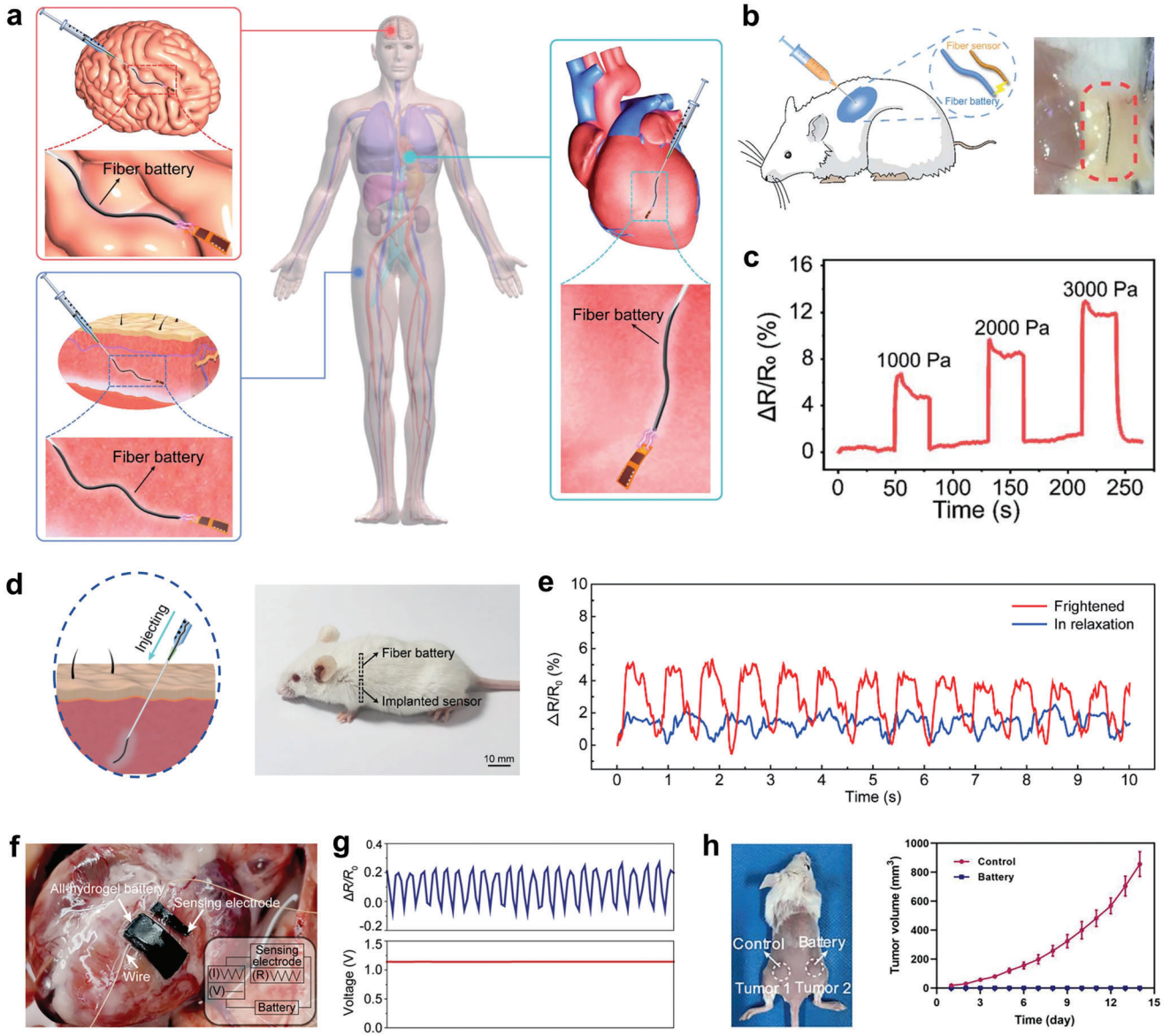

Figure 7. Biomedical applications of tissue-matchable and implantable flexible batteries. a) Schematic illustration of three typical injection regions (subcutis, heart, and brain) of injectable aqueous fiber-shaped batteries. Reproduced with permission.[30] Copyright 2021, Royal Society of Chemistry. b,c) Schematic diagram of a biodegradable fiber-shaped battery powering a fiber pressure sensor and the detected external pressure changes in the implanted area. Reproduced with permission.[33] Copyright 2021, Royal Society of Chemistry. d) Schematic illustration of injection and photograph of the mouse implanted with an aqueous fiber-shaped sodium-ion battery and sensor. e) Respiration monitoring signals recorded from the implanted sensor powered by injected fiber-shaped battery. Reproduced with permission.[30] Copyright 2021, Royal Society of Chemistry. f) Photograph showing the detection system with an all-hydrogel lithium-ion battery and sensor attached to the surface of the heart. g) Resistance variation curve of the sensor and the voltage variation curve of the all-hydrogel battery under heart beating. Reproduced with permission.[59] Copyright 2022, Wiley-VCH. h) Changes in the tumor volumes of mice in control group (saline) and battery group implanted with self-charging batteries. Reproduced with permission.[118] Copyright 2023, American Association for the Advancement of Science.

mance in different regions, showing promising applications in biomedical and healthcare fields. Despite the rapid development and progress in this field, there are still many challenges that need to be addressed prior to clinical applications (**Figure 8**).

High safety is one of the most crucial properties of implantable flexible batteries. The key to achieving high safety lies in the design of biocompatible components that can form stable interfaces with soft biological tissues, including electrodes, electrolytes, and encapsulation materials. Although tissue-matchable batteries are available, some components of these batteries are still not biocompatible, which may hinder their long-term implantations with potential safety risks. To this end, more efforts should be made to develop safer electrodes, electrolytes, and encapsulation materials with intrinsic biocompatibility and high flexibility. The extraction of electrode/electrolyte materials from

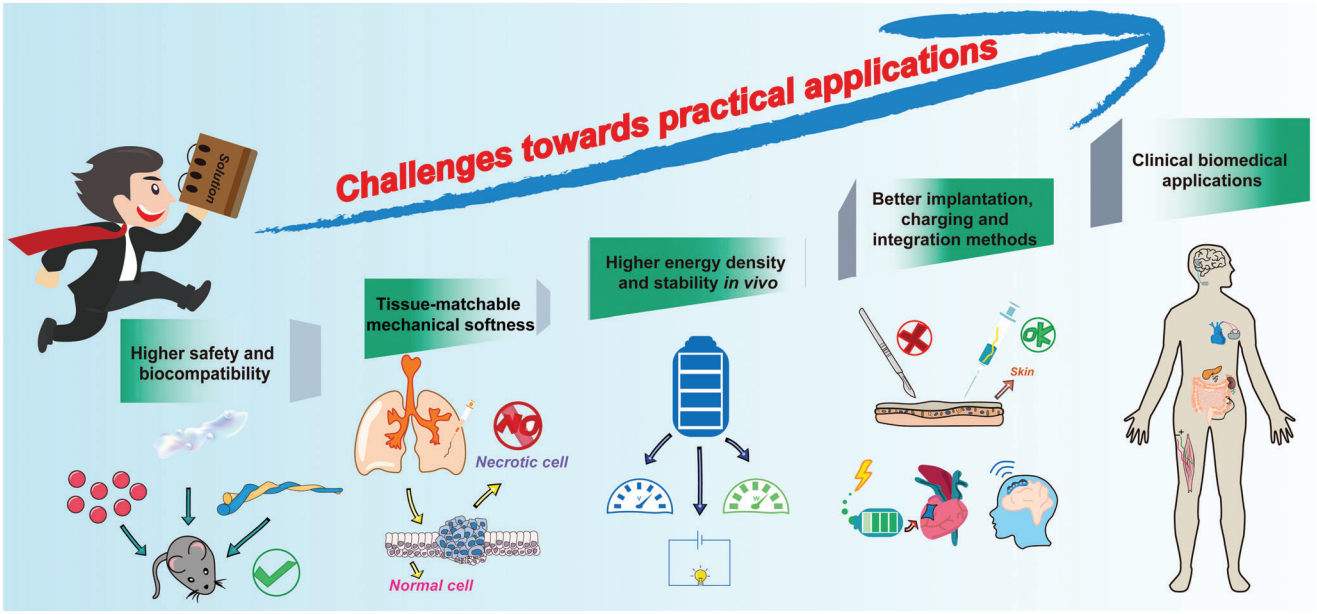

Figure 8. Challenges and roadmaps of tissue-matchable and implantable batteries toward practical biomedical applications.

natural biomass materials might be an effective and sustainable pathway.

Obtaining matched Young's moduli with soft and dynamic biological tissues is particularly important for the steady operation of tissue-matchable and implantable flexible batteries. Unstable battery-tissue interface caused by the mechanical mismatch will result in potential inflammatory response and tissue damage. Although several tissue-matchable flexible batteries have been developed with high softness close to biological tissues, there are still many implantable batteries that adopt the traditional rigid battery structure design with almost no flexibility. The large gap in mechanical softness between these implanted batteries and tissues may lead to a series of problems such as foreign body response and tissue damage after a long-term implantation. Therefore, it is crucial to develop highly flexible implantable batteries through a fully flexible design for all battery components. For example, designing new hydrogel-like electrode materials and electrolytes is an effective strategy for novel tissue-matchable flexible batteries.

In order to realize safe batteries for implantable bioelectronic devices, it is very important to design suitable systems. Aqueous batteries are a safer alternative to organic batteries. Interestingly, aqueous fiber-shaped sodium-ion batteries have been demonstrated to effectively work in body fluids without using additional electrolytes or encapsulation materials, which can greatly improve their flexibility. However, low energy density and poor stability of them have limited their applications. Their energy density is mainly restricted by the low specific capacity and low operating voltage. Currently, these implantable batteries can power some low-power devices, but may not be able to drive high-power devices such as pacemakers. This problem can be solved by developing new high-capacity electrode materials with high voltage and safety. Biodegradable batteries can be degraded after implantation to avoid surgical removal, whereas their continuously decreased electrochemical properties largely lower the stability of power supply. Thus, it is necessary to discover implantable aqueous batteries with much higher energy density and stability.

Injectable fiber-shaped batteries have provided a new implantation solution by injecting them into the body through a syringe. Compared with surgery implantation procedure, the syringe-assisted injection method is simpler and less invasive, offering decreased health risks, pain, and morbidity. However, most implantable batteries appear in thin films and can only be implanted by surgeries, which cause additional pain and health risk to patients. Therefore, more efforts should be made to develop mini-invasive implantation methods for implantable thin-film batteries. Moreover, it remains difficult to efficiently charge implanted batteries. Developing wireless charging solutions are urgently needed. Multifunctional integration methods and efficient connection methods of various implanted bioelectronic devices are also required for real clinical applications.

In summary, tissue-matchable and implantable flexible batteries are still in the early stages of development, and there is still a long way to go prior to their large-scale applications. The above challenges need close collaborations from researchers with a variety of backgrounds in Chemistry, Physics, Materials Science, and Biomedical Engineering. It is believed that these tissue-matchable and implantable batteries surely serve as the next-generation power systems for biomedical devices and may revolutionize healthcare fields.

Acknowledgements

This work was supported by the Science and Technology Commission of Shanghai Municipality (21511104900, 20JC1414902), the Natural Science Basic Research Program of Shaanxi (2022JQ-351), the Guangdong Basic and Applied Basic Research Foundation (2021A1515110593), the open research fund of State Key Laboratory of Organic Electronics and Information Displays, the National Postdoctoral Program for Innovative Talents (BX2021245), China Postdoctoral Science Foundation

(2021M702661), and the Fundamental Research Funds for the Central Universities (31020200QD042).

Conflict of Interest

The authors declare no conflict of interest.

Keywords

batteries, biomedical applications, flexibility, implantables

Received: April 14, 2023
Revised: June 30, 2023
Published online: July 19, 2023

[1] J. J. Jun, N. A. Steinmetz, J. H. Siegle, D. J. Denman, M. Bauza, B. Barbarits, A. K. Lee, C. A. Anastassiou, A. Andrei, C. Aydin, M. Barbic, T. J. Blanche, V. Bonin, J. Couto, B. Dutta, S. L. Gratiy, D. A. Gutnisky, M. Hausser, B. Karsh, P. Ledochowitsch, C. M. Lopez, C. Mitelut, S. Musa, M. Okun, M. Pachitariu, J. Putzeys, P. D. Rich, C. Rossant, W. L. Sun, K. Svoboda, et al., *Nature* **2017**, *551*, 232.
[2] C. M. Boutry, Y. Kaizawa, B. C. Schroeder, A. Chortos, A. Legrand, Z. Wang, J. Chang, P. Fox, Z. Bao, *Nat. Electron.* **2018**, *1*, 314.
[3] L. Wang, S. Xie, Z. Wang, F. Liu, Y. Yang, C. Tang, X. Wu, P. Liu, Y. Li, H. Saiyin, S. Zheng, X. Sun, F. Xu, H. Yu, H. Peng, *Nat. Biomed. Eng.* **2020**, *4*, 159.
[4] N. L. Opie, S. E. John, G. S. Rind, S. M. Ronayne, Y. T. Wong, G. Gerboni, P. E. Yoo, T. J. Lovell, T. C. Scordas, S. L. Wilson, A. Dornom, T. Vale, T. J. O'Brien, D. B. Grayden, C. N. May, T. J. Oxley, *Nat. Biomed. Eng.* **2018**, *2*, 907.
[5] D. Qi, Z. Liu, Y. Liu, Y. Jiang, W. R. Leow, M. Pal, S. Pan, H. Yang, Y. Wang, X. Zhang, J. Yu, B. Li, Z. Yu, W. Wang, X. Chen, *Adv. Mater.* **2017**, *29*, 1702800.
[6] P. Li, G. H. Lee, S. Y. Kim, S. Y. Kwon, H. R. Kim, S. Park, *ACS Nano* **2021**, *15*, 1960.
[7] K. Yamagishi, I. Kirino, I. Takahashi, H. Amano, S. Takeoka, Y. Morimoto, T. Fujie, *Nat. Biomed. Eng.* **2019**, *3*, 27.
[8] Y. S. Choi, R. T. Yin, A. Pfenniger, J. Koo, R. Avila, K. Benjamin Lee, S. W. Chen, G. Lee, G. Li, Y. Qiao, A. Murillo-Berlioz, A. Kiss, S. Han, S. M. Lee, C. Li, Z. Xie, Y. Y. Chen, A. Burrell, B. Geist, H. Jeong, J. Kim, H. J. Yoon, A. Banks, S. K. Kang, Z. J. Zhang, C. R. Haney, A. V. Sahakian, D. Johnson, T. Efimova, Y. Huang, et al., *Nat. Biotechnol.* **2021**, *39*, 1228.
[9] Y. Liu, J. Li, S. Song, J. Kang, Y. Tsao, S. Chen, V. Mottini, K. McConnell, W. Xu, Y. Q. Zheng, J. B. H. Tok, P. M. George, Z. Bao, *Nat. Biotechnol.* **2020**, *38*, 1031.
[10] Y. Song, J. Min, W. Gao, *ACS Nano* **2019**, *13*, 12280.
[11] Y. Zhao, C. Chen, Y. Qiu, T. Mei, L. Ye, H. Feng, Y. Zhang, L. Wang, Y. Guo, X. Sun, J. Wu, H. Peng, *Adv. Fiber Mater.* **2022**, *4*, 246.
[12] Q. Zheng, Q. Tang, Z. L. Wang, Z. Li, *Nat Rev Cardiol* **2021**, *18*, 7.
[13] C. Li, C. Guo, V. Fitzpatrick, A. Ibrahim, M. J. Zwierstra, P. Hanna, A. Lechtig, A. Nazarian, S. J. Lin, D. L. Kaplan, *Nat. Rev. Mater.* **2020**, *5*, 61.
[14] G. H. Lee, H. Moon, H. Kim, G. H. Lee, W. Kwon, S. Yoo, D. Myung, S. H. Yun, Z. Bao, S. K. Hahn, *Nat. Rev. Mater.* **2020**, *5*, 149.
[15] F. Fallegger, G. Schiavone, S. P. Lacour, *Adv. Mater.* **2020**, *32*, 1903904.
[16] J. H. Koo, J. K. Song, S. Yoo, S. H. Sunwoo, D. Son, D. H. Kim, *Adv. Mater. Technol.* **2020**, *5*, 2000407.
[17] Z. Zhao, G. D. Spyropoulos, C. Cea, J. N. Gelinas, D. Khodagholy, *Sci. Adv.* **2022**, *8*, eabm7851.
[18] S. Choi, S. I. Han, D. Jung, H. J. Hwang, C. Lim, S. Bae, O. K. Park, C. M. Tschabrunn, M. Lee, S. Y. Bae, J. W. Yu, J. H. Ryu, S. W. Lee, K. Park, P. M. Kang, W. B. Lee, R. Nezafat, T. Hyeon, D. H. Kim, *Nat. Nanotechnol.* **2018**, *13*, 1048.
[19] J. H. Lee, Y. S. Rim, W. K. Min, K. Park, H. T. Kim, G. Hwang, J. Song, H. J. Kim, *Adv. Funct. Mater.* **2021**, *31*, 2107074.
[20] T. M. Jang, J. H. Lee, H. Zhou, J. Joo, B. H. Lim, H. Cheng, S. H. Kim, I. S. Kang, K. S. Lee, E. Park, S. W. Hwang, *Sci. Adv.* **2020**, *6*, eabc9675.
[21] M. Mayer, A. J. Baeumner, *Chem. Rev.* **2019**, *119*, 7996.
[22] J. Zhou, Z. Ma, X. Hong, H. M. Wu, S.-Y. Ma, Y. Li, D. J. Chen, H. Y. Yu, X. J. Huang, *ACS Sens.* **2019**, *4*, 931.
[23] M. J. Malone-Povolny, E. P. Merricks, L. E. Wimsey, T. C. Nichols, M. H. Schoenfisch, *ACS Sens.* **2019**, *4*, 3257.
[24] K. Kaefer, K. Krüger, F. Schlapp, H. Uzun, S. Celiksoy, B. Flietel, A. Heimann, T. Schroeder, O. Kempski, C. Sonnichsen, *Nano Lett.* **2021**, *21*, 3325.
[25] J. Feng, C. Chen, X. Sun, H. Peng, *Acc Mater Res* **2021**, *2*, 138.
[26] G. Zhang, F. Geng, T. Zhao, F. Zhou, N. Zhang, S. Zhang, C. Deng, *ACS Appl. Mater. Interfaces* **2018**, *10*, 42268.
[27] H. Chen, Z. Bai, X. Dai, X. Zeng, Z. P. Cano, X. Xie, M. Zhao, M. Li, H. Wang, Z. Chen, L. Yang, J. Lu, *Angew. Chem., Int. Ed.* **2019**, *131*, 6735.
[28] J. Zhou, R. Zhang, R. Xu, Y. Li, W. Tian, M. Gao, M. Wang, D. Li, X. Liang, L. Xie, K. Liang, P. Chen, B. Kong, *Adv. Funct. Mater.* **2022**, *32*, 2111406.
[29] X. Huang, D. Wang, Z. Yuan, W. Xie, Y. Wu, R. Li, Y. Zhao, D. Luo, L. Cen, B. Chen, H. Wu, H. Xu, X. Sheng, M. Zhang, L. Zhao, L. Yin, *Small* **2018**, *14*, 1800994.
[30] Y. Zhao, T. Mei, L. Ye, Y. Li, L. Wang, Y. Zhang, P. Chen, X. Sun, C. Wang, H. Peng, *J. Mater. Chem. A* **2021**, *9*, 1463.
[31] X. Huang, H. Hou, B. Yu, J. Bai, Y. Guan, L. Wang, K. Chen, X. Wang, P. Sun, Y. Deng, S. Liu, X. Cai, Y. Wang, J. Peng, X. Sheng, W. Xiong, L. Yin, *ACS Nano* **2023**, *17*, 5727.
[32] X. Jia, C. Wang, V. Ranganathan, B. Napier, C. Yu, Y. Chao, M. Forsyth, F. G. Omenetto, D. R. MacFarlane, G. G. Wallace, *ACS Energy Lett.* **2017**, *2*, 831.
[33] T. Mei, C. Wang, M. Liao, J. Li, L. Wang, C. Tang, X. Sun, B. Wang, H. Peng, *J. Mater. Chem. A* **2021**, *9*, 10104.
[34] P. Simons, S. A. Schenk, M. A. Gysel, L. F. Olbrich, J. L. Rupp, *Adv. Mater.* **2022**, *34*, 2109075.
[35] S. He, Y. Hu, J. Wan, Q. Gao, Y. Wang, S. Xie, L. Qiu, C. Wang, G. Zheng, B. Wang, H. Peng, *Carbon* **2017**, *122*, 162.
[36] H. J. Sim, C. Choi, D. Y. Lee, H. Kim, J. H. Yun, J. M. Kim, T. M. Kang, R. Ovalle, R. H. Baughman, C. W. Kee, S. J. Kim, *Nano Energy* **2018**, *47*, 385.
[37] H. Li, C. Zhao, X. Wang, J. Meng, Y. Zou, S. Noreen, L. Zhao, Z. Liu, H. Ouyang, P. Tan, M. Yu, Y. Fan, Z. L. Wang, Z. Li, *Adv. Sci.* **2019**, *6*, 1801625.
[38] W. Tian, Y. Li, J. Zhou, T. Wang, R. Zhang, J. Cao, M. Luo, N. Li, N. Zhang, H. Gong, J. Zhang, L. Xie, B. Kong, *ACS Appl. Mater. Interfaces* **2021**, *13*, 8285.
[39] Z. Wang, S. Yao, S. Wang, Z. Liu, X. Wan, Q. Hu, Y. Zhao, C. Xiong, L. Li, *Chem. Eng. J.* **2023**, *463*, 142427.
[40] L. Wang, E. He, R. Gao, X. Wu, A. Zhou, J. Lu, T. Zhao, J. Li, Y. Yun, L. Li, T. Ye, Y. Jiao, J. Wang, H. Chen, D. Li, X. Ning, D. Wu, H. Peng, Y. Zhang, *Adv. Funct. Mater.* **2021**, *31*, 2107160.
[41] R. Hinchet, H. J. Yoon, H. Ryu, M. K. Kim, E. K. Choi, D. S. Kim, S. W. Kim, *Science* **2019**, *365*, 491.
[42] Z. Li, H. Feng, Q. Zheng, H. Li, C. Zhao, H. Ouyang, S. Noreen, M. Yu, F. Su, R. Liu, L. Li, Z. L. Wang, Z. Li, *Nano Energy* **2018**, *54*, 390.
[43] Z. Liu, H. Li, B. Shi, Y. Fan, Z. L. Wang, Z. Li, *Adv. Funct. Mater.* **2019**, *29*, 1808820.

[44] Z. Che, S. O'Donovan, X. Xiao, X. Wan, G. Chen, X. Zhao, Y. Zhou, J. Yin, J. Chen, *Small* **2023**, 2207600.
[45] P. Chen, P. Wu, X. Wan, Q. Wang, C. Xu, M. Yang, J. Feng, B. Hu, Z. Luo, *Nano Energy* **2021**, *86*, 106123.
[46] H. Zhou, Y. Zhang, Y. Qiu, H. Wu, W. Qin, Y. Liao, Q. Yu, H. Cheng, *Biosens. Bioelectron.* **2020**, *168*, 112569.
[47] Z. Xu, C. Jin, A. Cabe, D. Escobedo, A. Gruslova, S. Jenney, A. B. Closson, L. Dong, Z. Chen, M. D. Feldman, J. X. Zhang, *Adv. Healthcare Mater.* **2021**, *10*, 2002100.
[48] W. Gao, Y. Wang, F. Lai, *Smart Medicine* **2022**, *1*, e20220016.
[49] X. Huang, L. Wang, H. Wang, B. Zhang, X. Wang, R. Y. Stening, X. Sheng, L. Yin, *Small* **2020**, *16*, 1902827.
[50] T. Ghomian, S. Mehraeen, *Energy* **2019**, *178*, 33.
[51] P. Li, M. Liao, J. Li, L. Ye, X. Cheng, B. Wang, H. Peng, *Small Struct.* **2022**, *3*, 2200058.
[52] Q. Yang, T. Wei, R. T. Yin, M. Wu, Y. Xu, J. Koo, Y. S. Choi, Z. Xie, S. W. Chen, I. Kandela, S. Yao, Y. Deng, R. Avila, T. L. Liu, W. Bai, Y. Yang, M. Han, Q. Zhang, C. R. Haney, K. B. Lee, K. Aras, T. Wang, M. H. Seo, H. Luan, S. M. Lee, A. Brikha, N. Ghoreishi-Haack, L. Tran, I. Stepien, F. Aird, et al., *Nat. Mater.* **2021**, *20*, 1559.
[53] R. Feiner, T. Dvir, *Nat. Rev. Mater.* **2017**, *3*, 17076.
[54] M. Mimee, P. Nadeau, A. Hayward, S. Carim, S. Flanagan, L. Jerger, J. Collins, S. McDonnell, R. Swartwout, R. J. Citorik, V. Bulovic, R. Langer, G. Traverso, A. P. Chandrakasan, T. K. Lu, *Science* **2018**, *360*, 915.
[55] D. J. Young, P. Cong, M. A. Suster, M. Damaser, *Lab Chip* **2015**, *15*, 4338.
[56] C. F. Guimarães, L. Gasperini, A. P. Marques, R. L. Reis, *Nat. Rev. Mater.* **2020**, *5*, 351.
[57] Y. Ohm, C. Pan, M. J. Ford, X. Huang, J. Liao, C. Majidi, *Nat. Electron.* **2021**, *4*, 185.
[58] S. Park, S. W. Heo, W. Lee, D. Inoue, Z. Jiang, K. Yu, H. Jinno, D. Hashizume, M. Sekino, T. Yokota, K. Fukuda, K. Tajima, T. Someya, *Nature* **2018**, *561*, 516.
[59] T. Ye, J. Wang, Y. Jiao, L. Li, E. He, L. Wang, Y. Li, Y. Yun, D. Li, J. Lu, H. Chen, Q. Li, F. Li, R. Gao, H. Peng, Y. Zhang, *Adv. Mater.* **2022**, *34*, 2105120.
[60] Z. Fang, J. Wang, H. Wu, Q. Li, S. Fan, J. Wang, *J. Power Sources* **2020**, *454*, 227932.
[61] S. Ling, X. Li, T. Zhou, R. Yuan, S. Sun, H. He, C. Zhang, *Adv. Mater.* **2023**, *35*, 2211201.
[62] Q. Zhai, F. Xiang, F. Cheng, Y. Sun, X. Yang, W. Lu, L. Dai, *Energy Storage Mater.* **2020**, *33*, 116.
[63] F. Xiang, F. Cheng, Y. Sun, X. Yang, W. Lu, R. Amal, L. Dai, *Nano Res.* **2021**, *16*, 4821.
[64] C. H. Lai, D. S. Ashby, N. H. Bashian, J. Schoiber, T. C. Liu, G. S. Lee, S. Y. Chen, P. W. Wu, B. C. Melot, B. S. Dunn, *Adv. Energy Mater.* **2019**, *9*, 1900226.
[65] S. K. Kang, R. K. J. Murphy, S. W. Hwang, S. M. Lee, D. V. Harburg, N. A. Krueger, J. Shin, P. Gamble, H. Cheng, S. Yu, Z. Liu, J. G. McCall, M. Stephen, H. Ying, J. Kim, G. Park, R. C. Webb, C. H. Lee, S. Chung, D. S. Wie, A. D. Gujar, B. Vemulapalli, A. H. Kim, K. M. Lee, J. Cheng, Y. Huang, S. H. Lee, P. V. Braun, W. Z. Ray, J. A. Rogers, *Nature* **2016**, *530*, 71.
[66] J. Deng, X. Sun, H. Peng, *EcoMat* **2023**, *5*, 12343.
[67] R. Torres-Rosas, G. Yehia, G. Peña, P. Mishra, M. del Rocio Thompson-Bonilla, M. A. Moreno-Eutimio, L. A. Arriaga-Pizano, A. Isibasi, L. Ulloa, *Nat. Med.* **2014**, *20*, 291.
[68] K. Krawczyk, S. Xue, P. Buchmann, G. Charpin-El-Hamri, P. Saxena, M. D. Hussherr, J. Shao, H. Ye, M. Xie, M. Fussenegger, *Science* **2020**, *368*, 993.
[69] C. Marquez-Chin, M. R. Popovic, *Biomed. Eng. Online* **2020**, *19*, 34.
[70] E. V. Carrington, J. Evers, U. Grossi, P. G. Dinning, S. M. Scott, P. R. O'Connell, J. F. X. Jones, C. H. Knowles, *Neurogastroenterol Motil* **2014**, *26*, 1222.
[71] C. Yu, X. Liu, J. Zhang, Y. Chao, X. Jia, C. Wang, G. G. Wallace, *Small Methods* **2022**, *6*, 2200344.
[72] Y. Zhou, X. Jia, D. Pang, S. Jiang, M. Zhu, G. Lu, Y. Tian, C. Wang, D. Chao, G. Wallace, *Nat. Commun.* **2023**, *14*, 297.
[73] J. S. Chae, S. K. Park, K. C. Roh, H. S. Park, *Energy Storage Mater.* **2020**, *24*, 113.
[74] S. Stauss, I. Honma, *Bull. Chem. Soc. Jpn.* **2018**, *91*, 492.
[75] H. Sheng, X. Zhang, J. Liang, M. Shao, E. Xie, C. Yu, W. Lan, *Adv. Healthcare Mater.* **2021**, *10*, 2100199.
[76] D. Chao, W. Zhou, F. Xie, C. Ye, H. Li, M. Jaroniec, S. Z. Qiao, *Sci. Adv.* **2020**, *6*, eaba4098.
[77] Y. Liang, Y. Yao, *Nat. Rev. Mater.* **2023**, *8*, 109.
[78] Z. Wang, Y. Li, J. Wang, R. Ji, H. Yuan, Y. Wang, H. Wang, *Carbon Energy* **2022**, *4*, 411.
[79] F. Zhang, W. Zhang, D. Wexler, Z. Guo, *Adv. Mater.* **2022**, *34*, 2107965.
[80] S. Lei, Z. Liu, C. Liu, J. Li, B. Lu, S. Liang, J. Zhou, *Energy Environ. Sci.* **2022**, *15*, 4911.
[81] Z. Guo, Y. Zhao, Y. Ding, X. Dong, L. Chen, J. Cao, C. Wang, Y. Xia, H. Peng, Y. Wang, *Chem* **2017**, *3*, 348.
[82] A. von Wald Cresce, K. Xu, *Carbon Energy* **2021**, *3*, 721.
[83] D. Bin, Y. Wen, Y. Wang, Y. Xia, *J Energy Chem* **2018**, *27*, 1521.
[84] D. Bin, F. Wang, A. G. Tamirat, L. Suo, Y. Wang, C. Wang, Y. Xia, *Adv. Energy Mater.* **2018**, *8*, 1703008.
[85] T. Jin, X. Ji, P. F. Wang, K. Zhu, J. Zhang, L. Cao, L. Chen, C. Cui, T. Deng, S. Liu, N. Piao, Y. Liu, C. Shen, K. Xie, L. Jiao, C. Wang, *Angew. Chem., Int. Ed.* **2021**, *60*, 11943.
[86] P. Ruan, S. Liang, B. Lu, H. J. Fan, J. Zhou, *Angew. Chem., Int. Ed.* **2022**, *134*, 202200598.
[87] W. Du, E. H. Ang, Y. Yang, Y. Zhang, M. Ye, C. C. Li, *Energy Environ. Sci.* **2020**, *13*, 3330.
[88] M. Deng, L. Wang, B. Vaghefinazari, W. Xu, C. Feiler, S. V. Lamaka, D. Höche, M. L. Zheludkevich, D. Snihirova, *Energy Storage Mater.* **2021**, *43*, 238.
[89] F. Wang, X. Fan, T. Gao, W. Sun, Z. Ma, C. Yang, F. Han, K. Xu, C. Wang, *ACS Cent. Sci.* **2017**, *3*, 1121.
[90] M. C. Lin, M. Gong, B. Lu, Y. Wu, D.-Y. Wang, M. Guan, M. Angell, C. Chen, J. Yang, B. J. Hwang, H. Dai, *Nature* **2015**, *520*, 324.
[91] D. Yuan, J. Zhao, W. Manalastas Jr, S. Kumar, M. Srinivasan, *Nano Mater. Sci.* **2020**, *2*, 248.
[92] L. Hu, H. Wu, F. L.a Mantia, Y. Yang, Y. Cui, *ACS Nano* **2010**, *4*, 5843.
[93] J. Chang, J. Shang, Y. Sun, L. K. Ono, D. Wang, Z. Ma, Q. Huang, D. Chen, G. Liu, Y. Cui, Y. Qi, Z. Zheng, *Nat. Commun.* **2018**, *9*, 4480.
[94] X. Wang, S. Zheng, F. Zhou, J. Qin, X. Shi, S. Wang, C. Sun, X. Bao, Z. S. Wu, *Natl Sci Rev* **2020**, *7*, 64.
[95] H. Sun, Y. Zhang, J. Zhang, X. Sun, H. Peng, *Nat. Rev. Mater.* **2017**, *2*, 17023.
[96] F. Mo, G. Liang, Z. Huang, H. Li, D. Wang, C. Zhi, *Adv. Mater.* **2020**, *32*, 1902151.
[97] H. Wang, Y. Lu, Z. Nie, H. Liu, B. Dai, X. Shi, B. Yan, T. Zhao, Z. Zhang, J. Zhu, Y. Zhao, *Small* **2023**, *19*, 2206338.
[98] Y. Lu, H. Zhang, H. Liu, Z. Nie, F. Xu, Y. Zhao, J. Zhu, W. Huang, *Nano Lett.* **2021**, *21*, 9651.
[99] X. Huang, Y. Liu, W. Park, Z. Zhao, J. Li, C. K. Lim, T. H. Wong, C. K. Yiu, Y. Gao, J. Zhou, H. Li, L. Zhao, J. Li, B. Zhang, Y. Huang, R. Shi, D. Li, J. Mo, J. Wang, C. Zhang, Y. Li, Z. Wang, X. Yu, *InfoMat* **2022**, *5*, 12388.
[100] X. Xie, J. Li, Z. Xing, B. Lu, S. Liang, J. Zhou, *Natl Sci Rev* **2023**, *10*, nwac281.
[101] Y. H. Zhu, X. Y. Yang, T. Liu, X. B. Zhang, *Adv. Mater.* **2020**, *32*, 1901961.

[102] J. He, C. Lu, H. Jiang, F. Han, X. Shi, J. Wu, L. Wang, T. Chen, J. Wang, Y. Zhang, H. Yang, G. Zhang, X. Sun, B. Wang, P. Chen, Y. Wang, Y. Xia, H. Peng, *Nature* **2021**, *597*, 57.
[103] Y. Guo, C. Chen, J. Feng, L. Wang, J. Wang, C. Tang, X. Sun, H. Peng, *Small Methods* **2022**, *6*, 2200142.
[104] C. Yu, C. Wang, X. Liu, X. Jia, S. Naficy, K. Shu, M. Forsyth, G. G. Wallace, *Adv. Mater.* **2016**, *28*, 9349.
[105] M. Karami-Mosammam, D. Danninger, D. Schiller, M. Kaltenbrunner, *Adv. Mater.* **2022**, *34*, 2204457.
[106] L. Li, H. Chen, E. He, L. Wang, T. Ye, J. Lu, Y. Jiao, J. Wang, R. Gao, H. Peng, Y. Zhang, *Angew. Chem., Int. Ed.* **2021**, *133*, 15445.
[107] C. S. Li, Y. Sun, F. Gebert, S. L. Chou, *Adv. Energy Mater.* **2017**, *7*, 1700869.
[108] T. Zhu, Y. Ni, G. M. Biesold, Y. Cheng, M. Ge, H. Li, J. Huang, Z. Lin, Y. Lai, *Chem. Soc. Rev.* **2023**, *52*, 473.
[109] C. Xie, X. Wang, H. He, Y. Ding, X. Lu, *Adv. Funct. Mater.* **2020**, *30*, 1909954.
[110] A. C. Daly, L. Riley, T. Segura, J. A. Burdick, *Nat. Rev. Mater.* **2020**, *5*, 20.
[111] Z. Wang, H. Li, Z. Tang, Z. Liu, Z. Ruan, L. Ma, Q. Yang, D. Wang, C. Zhi, *Adv. Funct. Mater.* **2018**, *28*, 1804560.
[112] L. Lu, X. Fu, Y. Liew, Y. Zhang, S. Zhao, Z. Xu, J. Zhao, D. Li, Q. Li, G. B. Stanley, X. Duan, *Nano Lett.* **2019**, *19*, 1577.
[113] T. Zheng, P. Pour Shahid Saeed Abadi, J. Seo, B. H. Cha, B. Miccoli, Y. C. Li, K. Park, S. Park, S. J. Choi, R. Bayaniahangar, D. Zhang, S. H. Lee, C. K. Lee, A. Khademhosseini, S. R. Shin, *ACS Appl. Mater. Interfaces* **2019**, *11*, 20615.
[114] G. Fang, J. Zhou, A. Pan, S. Liang, *ACS Energy Lett.* **2018**, *3*, 2480.
[115] M. Wu, G. Zhang, H. Yang, X. Liu, M. Dubois, M. A. Gauthier, S. Sun, *InfoMat* **2022**, *4*, 12265.
[116] Z. Shao, S. Cheng, Y. Zhang, H. Guo, X. Cui, Z. Sun, Y. Liu, Y. Wu, P. Cui, J. Fu, Q. Su, E. Xie, *ACS Appl. Mater. Interfaces* **2021**, *13*, 34349.
[117] A. T. Kutbee, R. R. Bahabry, K. O. Alamoudi, M. T. Ghoneim, M. D. Cordero, A. S. Almuslem, A. Gumus, E. M. Diallo, J. M. Nassar, A. M. Hussain, N. M. Khashab, M. M. Hussain, *npj Flexible Electron.* **2017**, *1*, 7.
[118] J. Huang, P. Yu, M. Liao, X. Dong, J. Xu, J. Ming, D. Bin, Y. Wang, F. Zhang, Y. Xia, *Sci. Adv.* **2023**, *9*, eadf3992.
[119] S. Zhu, J. Sheng, Y. Chen, J. Ni, Y. Li, *Natl Sci Rev* **2021**, *8*, nwaa261.
[120] L. Li, L. Wang, T. Ye, H. Peng, Y. Zhang, *Small* **2021**, *17*, 2005015.
[121] M. H. Lee, J. Lee, S. K. Jung, D. Kang, M. S. Park, G. D. Cha, K. W. Cho, J. H. Song, S. Moon, Y. S. Yun, S. J. Kim, Y. W. Lim, D. H. Kim, K. Kang, *Adv. Mater.* **2021**, *33*, 2004902.
[122] G. Shin, A. M. Gomez, R. Al-Hasani, Y. R. Jeong, J. Kim, Z. Xie, A. Banks, S. M. Lee, S. Y. Han, C. J. Yoo, J.-L. Lee, S. H. Lee, J. Kurniawan, J. Tureb, Z. Guo, J. Yoon, S.-I. Park, S. Y. Bang, Y. Nam, M. C. Walicki, V. K. Samineni, A. D. Mickle, K. Lee, S. Y. Heo, J. G. McCall, T. Pan, L. Wang, X. Feng, T.-I. Kim, J. K. Kim, et al., *Neuron* **2017**, *93*, 509.

Bing Yan received his B.E. in metallurgical engineering from Lanzhou University of Technology in 2022. He is currently a M.S. student at Northwestern Polytechnical University, China. His work focuses on the fabrication of flexible and high-performance aqueous batteries.

Yang Zhao is currently an associate professor at Northwestern Polytechnical University. He received his Ph.D. degree in macromolecular chemistry and physics at Fudan University in 2020. His research interests are focused on the development of flexible and wearable energy storage devices.

Huisheng Peng is currently a professor at the Department of Macromolecular Science and Laboratory of Advanced Materials at Fudan University. He received his B.E. in polymer materials at Donghua University in China in 1999, his M.S. in macromolecular chemistry and physics at Fudan University in China in 2003, and his Ph.D. in chemical engineering at Tulane University in the USA in 2006. He then worked at Los Alamos National Laboratory before joining Fudan University in 2008. He focuses on the new direction of fiber electronics.