IJMMM International Journal of Minerals, Metallurgy and Materials 矿物冶金与材料学报（英文版）

Mechano-electrochemical perspectives on flexible lithium-ion batteries

Na Li, Shuangquan Yang, Haosen Chen, Shuqiang Jiao, and Weili Song

Cite this article as:
Na Li, Shuangquan Yang, Haosen Chen, Shuqiang Jiao, and Weili Song, Mechano-electrochemical perspectives on flexible lithium-ion batteries, *Int. J. Miner. Metall. Mater.*, 29(2022), No. 5, pp. 1019-1036. https://doi.org/10.1007/s12613-022-2486-4

View the article online at SpringerLink or IJMMM Webpage.

International Journal of Minerals, Metallurgy and Materials
Volume 29, Number 5, May 2022, Page 1019
https://doi.org/10.1007/s12613-022-2486-4

Invited Review

Mechano-electrochemical perspectives on flexible lithium-ion batteries

Na Li[1,2], *Shuangquan Yang*[1,2], *Haosen Chen*[1,2], *Shuqiang Jiao*[3], *and Weili Song*[1,2]✉

1) Institute of Advanced Structure Technology, Beijing Institute of Technology, Beijing 100081, China
2) Beijing Key Laboratory of Lightweight Multi-functional Composite Materials and Structures, Beijing Institute of Technology, Beijing 100081, China
3) State Key Laboratory of Advanced Metallurgy, University of Science and Technology Beijing, Beijing 100083, China
(Received: 20 January 2022; revised: 29 March 2022; accepted: 30 March 2022)

Abstract: With the advent of flexible/wearable electronic devices, flexible lithium-ion batteries (LIBs) have attracted significant attention as optimal power source candidates. Flexible LIBs with good flexibility, mechanical stability, and high energy density are still an enormous challenge. In recent years, many complex and diverse design methods for flexible LIBs have been reported. The design and evaluation of ideal flexible LIBs must take into consideration both mechanical and electrochemical factors. In this review, the recent progress and challenges of flexible LIBs are reviewed from a mechano-electrochemical perspective. The recent progress in flexible LIB design is addressed concerning flexible material and configuration design. The mechanical and electrochemical evaluations of flexible LIBs are also summarized. Furthermore, mechano-electrochemical perspectives for the future direction of flexible LIBs are also discussed. Finally, the relationship between mechanical loading and the electrode process is analyzed from a mechano-electrochemical perspective. The evaluation of flexible LIBs should be based on mechano-electrochemical processes. Reviews and perspectives are of great significance to the design and practicality of flexible LIBs, which is contributed to bridging the gap between laboratory exploration and practical applications.

Keywords: flexible lithium-ion batteries; flexible materials; structural design; mechanical and electrochemical coupling

1. Introduction

With the increasing interest in flexible/wearable electronic and medical devices, flexible energy-storage devices have attracted attention as power sources and have played an indispensable role in the development of these flexible technologies [1–6]. Lithium-ion batteries (LIBs) are the optimal candidates owing to their stable cycle life, high energy density, and low cost [7–8]. Conventional LIBs consist of rigid electrodes, electrolytes, separators, and packaging materials. Conventional LIBs structures mainly include prismatic, coin, and cylindrical shapes with rigid packages [9]. With conventional LIBs, their use with flexible/wearable devices is difficult as the active material can be removed from the current collector under a mechanical load. This phenomenon leads to an increase in internal resistance, particle isolation, and capacity decay. Compared with conventional LIBs, flexible LIBs possess excellent mechanical properties including stretchability, foldability, bendability, and twistability [10–11]. While flexible LIBs have decades of development history, they still present the greatest challenge for practical application in flexible/wearable devices [12].

The key challenges of flexible LIBs are overcoming the rigidity of traditional materials and battery structures and improving their safety under mechanical deformation. Matching between the materials and battery structure is an important factor that affects the performance of flexible LIBs [13]. To overcome these above greatest problems, enormous efforts have been expended in the development of flexible materials and structural designs. Various flexible materials have been used to replace traditional current collectors and rigid electrodes [14–17]. Different flexible carbon-based materials, such as carbon textiles [18–19], graphene film [20–22], or carbon nanotubes [23–26] have been reported to replace rigid materials due to excellent conductivity and flexibility. Other functional materials have also been developed for flexible LIBs, including flexible polymer electrolytes [27–32] and polymer packaging materials [33–34]. In addition, the flexibility of the battery has been improved by suppressing the stress concentration in electrodes during mechanical deformation through structural design [35]. A variety of novel flexible battery configurations have been widely studied, such as thin film [36], cable-type [37–38], wave-like [39], origami [40–41] and node-type [42]. These flexible battery configurations can maintain excellent electrochemical performance under mechanical deformation. Therefore, significant progress has been made in the development of flexible materials and structural designs. Nevertheless, the energy density cannot meet the demands of practical applications [13]. The electrochemical performance, mechanical properties, and flexibility are important factors to consider when designing flexible batteries. However, there is no unified

✉ Corresponding author: Weili Song E-mail: weilis@bit.edu.cn

standard for comprehensively the performance of flexible LIBs. Hence, there are still many major challenges in the design, manufacture, and evaluation of flexible batteries.

To date, there have been many excellent reviews on flexible LIBs, providing the development history and summarizing recent progress. Previous reviews have mainly focused on flexible materials [43] and designconfigurations [35], and have paid little attention to the methods used for the evaluation [44] of flexible batteries. In this review, the development and challenges of flexible LIBs were discussed from a mechano-electrochemical perspective. The recent progress in flexible LIBs design is addressed concerning flexible material and configuration design. We reviewed the evaluation methods with respect to mechano-electrochemical processes. Fig. 1 summarizes the flexible materials, configurations, and evaluation methods. Upon the current progress, major challenges and perspectives on the future development of flexible LIBs are highlighted from a mechano-electrochemical view. This review is expected to provide novel macro-scale and micro-scale perspectives for overcoming challenges, which can be used to guide the optimal design of flexible LIBs and for understanding the essential mechanisms.

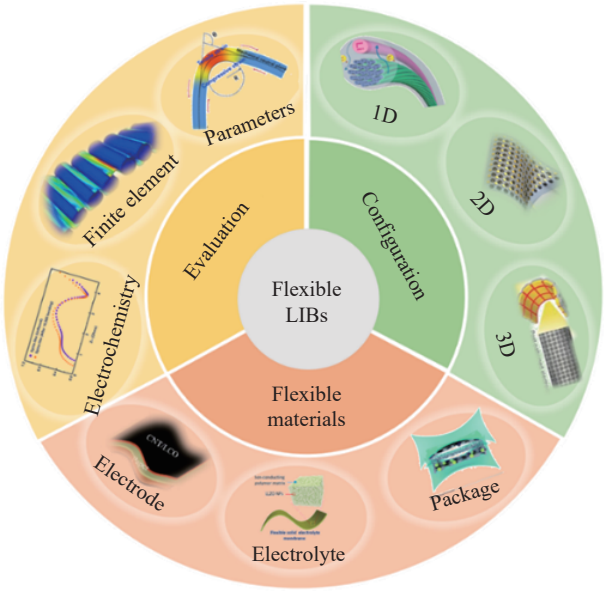


Fig. 1. Summary of flexible materials, configurations, and evaluation methods of flexible LIBs.

2. Flexible materials design

Conventional LIBs are prone to stress concentration and safety problems during mechanical deformation due to the presence of rigid materials. Thus, the rigid materials used in conventional LIBs are not suitable for flexible LIBs. Flexible materials are indispensable for flexible batteries. Designing suitable flexible materials is a major challenge for the development of flexible batteries with excellent performance. In recent years, great progress has been made in flexible materials, which can ensure that the batteries can adapt to mechanical deformations. Most researchers have focused on flexible electrode materials, electrolytes, and packaging materials.

2.1. Flexible electrodes

Traditional electrodes are prepared by coating a slurry of the active material, conductive carbon, and binder on metal current collectors (Cu or Al) [45]. The active materials were shaded from the current collectors during mechanical deformations owing to the weak binding force, which leads to the battery performance degradation [9,46]. Flexible LIBs should maintain excellent mechanical and electrochemical performance under various mechanical deformations. Flexible electrodes are the key factors affecting battery performance. A wide variety of flexible electrode materials has emerged, including self-supporting electrodes [47], flexible current collectors [48], and flexible electrode configurations [41].

2.1.1. Flexible carbon-based materials

Carbon-based materials such as carbon nanotubes [23,49], carbon cloth [19,50], and graphene [21–22], have been widely used as flexible electrodes and current collectors due to their excellent electrical conductivity and flexibility. Carbon nanotube (CNT) films are the most promising current collectors and have been widely applied in flexible LIBs. Hu *et al*. [51] have reported flexible paper flexible LIBs in which free-standing CNT thin films and flexible paper were used as current collectors and separators, respectively, as shown in Fig. 2(a). $LiCoO_2$(LCO) and $Li_4Ti_5O_{12}$(LTO) materials were coated onto CNT thin film to form flexible electrodes. Then, the flexible LCO/CNT and LTO/CNT were bonded to both sides of the paper peeling process with a rod to form a flexible paper battery with a thickness of only 300 μm. The cross-sectional SEM images of the paper battery are shown in Fig. 2(b) [51]. The self-discharging performance was approximately 0.04%, and the voltage decreased by 2% after cutting off the powder, as shown in Fig. 2(c) [51]. The flexible paper-based LIBs exhibited a high energy density of 108 $mW{\cdot}h{\cdot}g^{-1}$ and excellent mechanical flexibility due to the strong adhesion and mechanical durability of the flexible electrodes.

In order to improve the electrode conductivity and capacity of flexible LIBs, graphene films have also been used in flexible batteries as optimal candidates, due to their high surface area, stability, flexibility, and excellent conductivity. The graphene has higher specific surface area than CNT. Shi *et al*. [52] have designed flexible LIBs composed of integrated graphene-based electrodes. The flexible free-standing electrodes were prepared via vacuum filtration. As shown in Fig. 2(d) and (e), the graphene/LTO electrode (G–LTO) has a more stable interface than conventional electrodes(Al–LTO). The graphene-based electrode exhibited excellent flexibility (Fig. 2(f)). The potential difference increased with increasing current density; however the conventional electrodes showed a higher overpotential. As shown in Fig. 2(g)–(i), the flexible LIBs exhibited lower polarization and excellent rate performance compared to the conventional electrodes, which was attributed to the stable interface between the active materials and graphene. In conclusion, the G–LTO electrodes

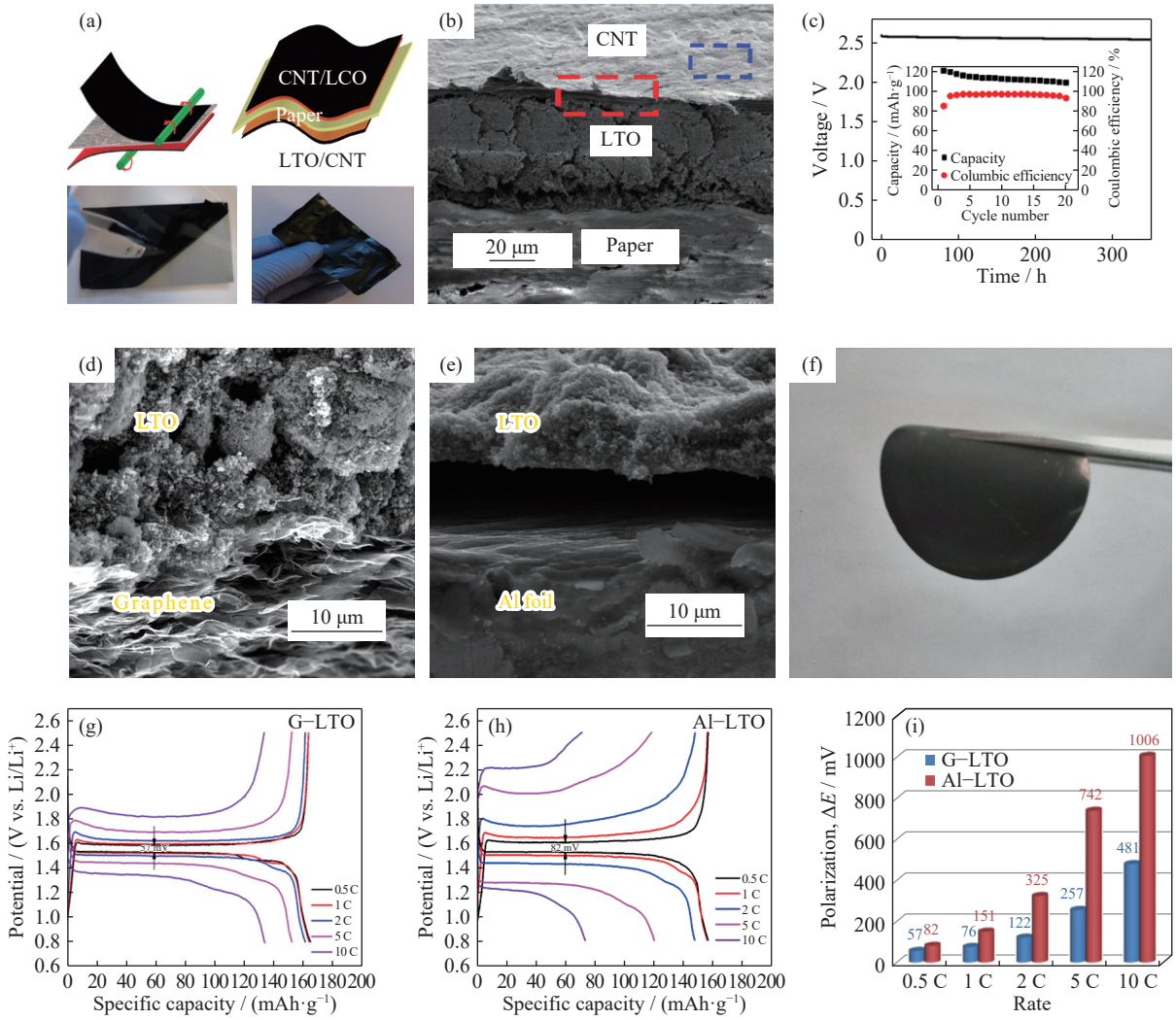


Fig. 2. Flexible carbon-based materials: (a) schematic of the paper LIB with free-standing film electrode; (b) SEM image of the cross-section of the paper LIB; (c) self-discharge curves of the full paper LIB; SEM images of the cross-section of (d) G–LTO and (e) Al–LTO; (f) photograph of a G–LFP; rate performance of (g) G–LTO and (h) Al–LTO; (i) comparison of the polarization of different electrodes at different current densities. (a–c) Reprinted with permission from L. Hu, H. Wu, F.L. Mantia, *et al.*, Thin, flexible secondary Li-ion paper batteries, *ACS Nano*, 4(2010), No. 10, p. 5843 [51]. Copyright 2010 American Chemical Society; (d–i) [52] © IOP Publishing. Reproduced with permission. All rights reserved.

exhibited stable electrode processes due to their faster ion transport and high electrolyte wettability. In conclusion, flexible carbon-based electrodes are thin, showed good flexibility and electrochemical properties. CNT and graphene films were used as a flexible substrates, and active materials coated on the substrate through doctor-blading or vacuum filtrations processes. The flexible electrode has poor adhesion, so the interface stability needs to be improved. However, compared with the traditional lithium-ion battery, the capacity of the flexible battery obtained by the above method is relatively low, which is difficult to meet the practical applications.

2.1.2. Flexible electrode structure design

Although the flexible film electrodes have excellent flexibility, stress concentration would still occur in the electrodes during continuous mechanical deformation. To solve this problem, novel electrode configurations and three-dimensional electrodes have been designed. Bao *et al.* [53] have reported patterned electrodes with serpentine-shaped networks. The patterned electrodes were prepared by 3D-printed technology, as shown in Fig. 3(a), and then the free-standing serpentine-shaped electrodes were acquired through vacuum drying and demoulding. The stretchability of the serpentine-shaped patterned electrode network was demonstrated by finite element simulation. For the flexible LIBs, $Li_4Ti_5O_{12}$ (LTO) and $LiFePO_4$ (LFP) were chosen as the positive electrode and negative electrode respectively. The electrolyte is 1 M $LiPF_6$ in EC-DEC with 1:1 by volume. The capacity retentions of LFP and LTO electrodes could still reach 92% and 88% after stretching 100 times, as shown in Fig. 3(b). The full batteries showed stable electrochemical performance, powering a light-emitting diode (LED) under different mechanical deformations. The excellent extendibility and mechanical durability were attributed to suitable configuration design. This serpentine-shaped electrode would provide a new strategy for the design and manufacture of stretchable LIBs [53]. Poor contact occurs inside the free-standing serpentine-shaped electrode, and the free-standing electrode cracked during extreme deformation. Hence, elevating inter-

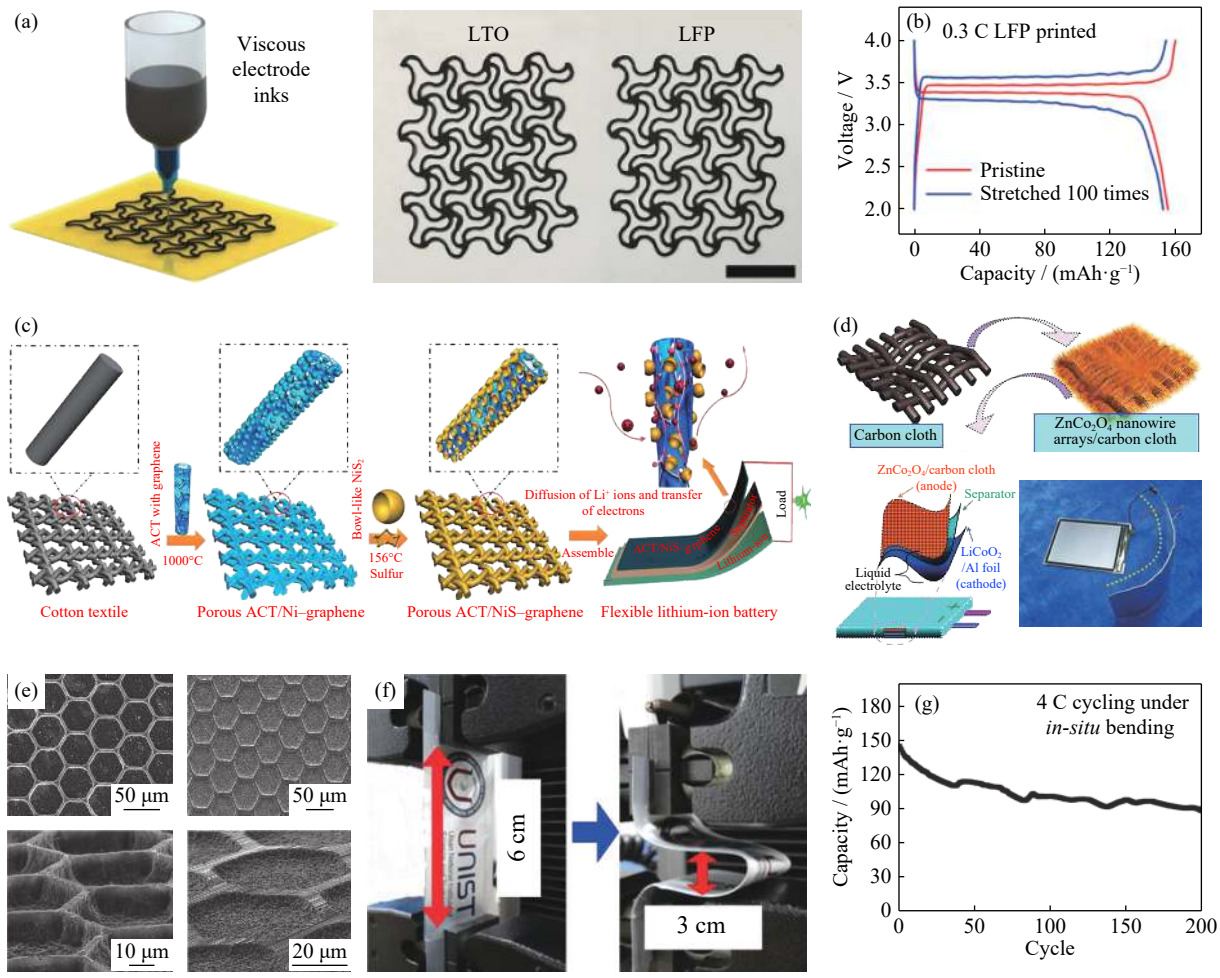


Fig. 3. Flexible electrode structure design: (a) 3D-printed patterned stretchable electrodes; (b) cycle performance of LFP cathode at 0.3 C under different states: pristine and stretched 100 times; (c) schematic of the fabrication process of flexible lithium-ion batteries with active cotton-textile; (d) schematic of the fabrication of flexible lithium-ion battery with the hierarchical 3D $ZnCo_2O_4$ nanowire arrays/carbon cloth as negative; (e) SEM images of patterned current collectors: Al and Cu foil; (f) photographs of a flexible battery under mechanical deformation; (g) discharge capacity of the flexible battery at the current density of 4 C under in-situ bending: a bending speed of 30 mm·min^{-1} with an applied force of 11.0 N. (a–b) Reprinted from *Energy Storage Mater.*, 33, Y.H. Bao, Y. Liu, Y.D. Kuang, *et al.*, 3D-printed highly deformable electrodes for flexible lithium ion batteries, 55-61, Copyright 2020, with permission from Elsevier [53]; (c) reprinted with permission from Z. Gao, N.N. Song, Y.Y. Zhang, *et al.*, Cotton-textile-enabled, flexible lithium-ion batteries with enhanced capacity and extended lifespan, *Nano Lett.*, 15(2015), No. 12, p. 8194 [18]. Copyright 2015 American Chemical Society; (d) reprinted with permission from B. Liu, J. Zhang, X.F. Wang, *et al.*, Hierarchical three-dimensional $ZnCo_2O_4$ nanowire arrays/carbon cloth anodes for a novel class of high-performance flexible lithium-ion batteries, *Nano Lett.*, 12(2012), No. 6, p. 3005 [19]. Copyright 2012 American Chemical Society; (e–g) reprinted with permission from M.H. Park, M. Noh, S. Lee, *et al.*, Flexible high-energy Li-ion batteries with fast-charging capability, *Nano Lett.*, 14(2014), No. 7, p. 4083 [54]. Copyright 2014 American Chemical Society.

face contact is very important. Chemical contact is more stable than physical contact. In-situ grown on flexible substrate is an effective method.

Three-dimensional flexible current collectors have been developed to improve the stability of electrodes. Carbon cloth and conductive carbon textiles have been the most frequently reported owing to their excellent mechanical strength, commercial feasibility, and flexibility. Many electrode materials can be directly grown on the three-dimensional carbon-based substrates, such as nanowires, nanosheets, and nanotubes. Such three-dimensional free-standing electrodes fabricated using an in situ growth method is an effective method to prepare flexible electrodes for LIBs. Gao *et al.* [18] have reported the use of an active cotton textile (ACT) as the current collector. The ACT/NiS_2-graphene electrode was prepared by two-step heat treatment, as shown in Fig. 3(c). The flexible LIBs assembled using the ACT/NiS_2-graphene electrode showed excellent flexibility and electrochemical performance during bending, which was attributed to the multiscale porous hierarchical structure of ACT. During mechanical deformation, no cracks were generated in the active particles because the hollow structure of the ACT fibers buffered the stress concentration. Liu *et al.* [19] have fabricated hierarchical 3D $ZnCo_2O_4$ nanowire arrays/carbon cloth anodes for flexible LIBs. Fig. 3(d) shows the schematics of the electrodes and flexible LIBs. After bending 120 times, the flexible LIBs exhibited similar discharge capacity and platforms. In conclusion, 3D carbon-based current collectors have been

demonstrated to provide a good opportunity for the commercialization of flexible batteries owing to their lightweight, high mechanical strength, and strong adhesion with the active materials. In particular, the strong adhesion with the active materials can maintain the structural stability of the electrode and improve its conductivity. The porous structure increases the wettability of the electrolyte and the active surface area, which is beneficial for accelerating Li^+ transportation and the stability of electrode reactions [19].

The above-mentioned flexible electrodes are different from conventional electrodes, such as energy density and conductivity. The design of flexible batteries based on traditional metal collectors is an inevitable trend. Active materials are removed from the metal current collector during mechanical deformation owing to their lower adhesion. To solve this problem, many researchers have studied patterned metal foil. Park *et al*. [54] have reported the use of comb-patterned Cu and Al foils as current collectors, which can improve adhesion to the electrode (Fig. 3(e)). The width of the omb-patterned Cu and Al foil was 50 μm, which was optimal for elevating the mechanical strength of the current collector and adhesion to active materials. The flexible full batteries using comb-patterned Cu and Al foils were bent by a testing machine (Fig. 3(f)) at a bending speed of 30 $mm \cdot min^{-1}$ under an applied force of 11.0 N. The flexible full battery showed the discharge capacity of 90 $mAh \cdot g^{-1}$ after 200 cycles under in-situ bending at a current density of 4 C (Fig. 3(g)) [54]. The comb-patterned Cu and Al foil, a combination of microsacle and nanoscale patterns, significantly increases the mechanical strength of the current collector and the adhesion of active materials to the current collector. Compared with the electrodes that active materials shedding from the current collector, the comb-patterned current collector exhibited better rate performance. This work has developed flexible batteries with fast charging capability by using high-power cathode and anode materials and honeycomb-patterned current collectors. Hence, a patterned current collector is an effective method for fabricating flexible LIBs.

In summary, free-standing carbon-based electrodes and flexible electrode structure designs are the future development directions for flexible electrodes. They exhibit strong adhesion to active materials, which can enhance Li^+ transport. The good conductivity and porous structure can provide pathways for ion and electron transport, which is beneficial for the stability of the electrochemical kinetic process during mechanical deformation. However, during extreme deformations, the flexible electrodes maybe occur wrinkle, fractures, and delamination. The manufacturing processes of the above-mentioned flexible electrodes are complex and the cost is expensive. It is enhancing major challenge to enhance the energy density of flexible LIBs.

2.2. Flexible polymer electrolyte

The electrolyte is an indispensable part of LIBs, and plays a role in transporting Li-ions. The electrolyte should have high ionic conductivity and insulation resistivity. The electrolytes of conventional LIBs are liquid, mostly composed of ester and ether electrolytes that are combustible. LIBs are prone to liquid leakage under continuous mechanical loads, causing safety hazards. Furthermore, side reactions including dendrites growth, electrolyte decomposition, and gas evolution may occur on the surface of the electrode or electrolyte. Lithium dendrites can pierce the separator, resulting in thermal runaway and explosions. In recent years, the further development of inorganic solid electrolytes and polymer electrolytes has significantly enhanced the safety of LIBs. Inorganic solid electrolytes (ISEs) have high ionic conductivity, high rigidness, and high interface resistance, which are not suitable for flexible LIBs [9]. Solid polymer electrolytes are widely used in flexible batteries because of their good mechanical flexibility and excellent interface stability.

Lee and his colleagues have prepared a new type of ultrathin and deformable polymer electrolyte (N-PCPE) composed of a plastic crystalline polymer electrolyte and porous non-woven polyethylene terephthalate (PET) fabric [28]. N-PCPE possesses excellent mechanical strength owing to its three-dimensional network structure. The N-PCPE electrolyte could be deformed arbitrarily, as shown in Fig. 4(a). The LTO negative electrode, N-PCPE, and LCO positive electrode were assembled into a battery with an all-plastic film. The resulting pouch battery-powered LED in both bent and no-bend states (Fig. 4(b)–(d)). As shown in Fig. 4(c), the voltage remained stable during the different bending deformation processes. The pouch battery assembled with N-PCPE exhibited stable electrochemical performance even in a severely wrinkled state, and there was no internal short circuit between the electrodes. N-PCPE exhibits both high ionic conductivity and mechanical flexibility, and this electrolyte can provide important support for the development of flexible batteries, especially for safety. In addition, flame-retardant and stable polymer electrolyte membranes have been fabricated by the Kyu's research group. The polymer electrolyte was prepared using cross-linkable polyurethane as the precursor; the preparation process is shown in Fig. 4(e) [55]. The polymer electrolyte membrane (PEM) was transparent and could be arbitrarily bent and twisted (Fig. 4(f)). The PEM exhibited an ionic conductivity of 8×10^{-4} $S \cdot cm^{-1}$ at room temperature. The LFP and graphite were used as the positive and negative electrodes. The full battery was composed of LFP, PEM, and graphite, and could powder a LED during the bending process (Fig. 4(g)). The PEM electrolyte exhibited good electrochemical performance in the full battery, with 80% capacity retention after 250 cycles.

Therefore, The safety and flexibility of flexible LIBs are improved by using solid polymer electrolytes. However, compared with liquid electrolytes, the ionic conductivity and interface stability still needs to be improved. Currently, in situ polymerization methods and additives have been widely used to evaluate interface stability and reduce interface resistance. Hence, the development of novel solid-state polymer electrolytes with high conductivity is very important for flexible LIBs. In addition, enhancing the stability of the interface between solid polymer electrolyte and electrode should be paid attention to flexible lithium-ion battery.

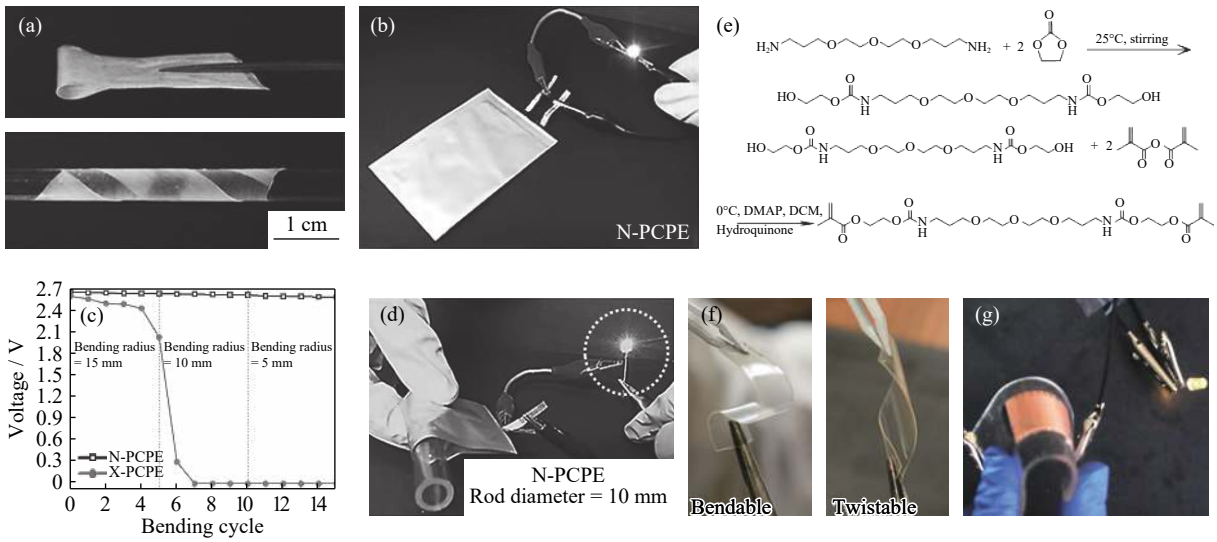


Fig. 4. Flexible polymer electrolytes: (a) photograph of an N-PCPE electrolyte with the different bending states; photographs of pouch LIBs with the N-PCPE electrolyte in (b) the pristine and (c) bent states; (d) voltage of the pouch LIBs during the bending test; (e) synthesis of polyethylene glycol-bis-carbamate (PEGBC) and polyethylene glycol-bis-carbamate dimethacrylate (PEGBC-DMA); (f) PEM electrolyte under the bent and twisted states; (g) photograph of flexible solid-state LIB powering LED. (a–d) K.H. Choi, S.J. Cho, S.H. Kim, *et al*., *Adv. Funct. Mater*., 24, 44 (2014) [28]. Copyright 2014 Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission; (e–g) reprinted from *Solid State Ionics*, 320, G.P. Fu, M.D. Soucek, and T. Kyu, Fully flexible lithium ion battery based on a flame retardant, solid-state polymer electrolyte membrane, 310-315, Copyright 2018, with permission from Elsevier [55].

2.3. Flexible packaging materials

LIBs are packaged in sealed containers to prevent the electrolyte from coming into contact with the external atmosphere. The packaging materials generally comprise either an aluminum-plastic film, steel shell, or aluminum shell, which are inert and do not participate in any reactions. However, these packaging materials have high stiffness, which results in uneven stress in the electrode and the electrolyte. Currently, most of the reported flexible LIBs are mainly based on pouch batteries, in which laminated or wound cells are encapsulated by an aluminum-plastic film. The aluminum-plastic film is mainly divided into three layers: nylon, metal Al foil, and polypropylene film. The aluminum-plastic film has low flexibility, which depends on the thickness of the nylon and metal layers. Flexible packaging materials that prevent electrolyte leakage are very important for flexible LIBs.

To address the inflexibility of aluminum-plastic films, most polymer materials have been applied to packaging flexible LIBs, such as polydimethylsiloxane (PDMS) [56] and polyimide films (PI) [57]. Gao and his colleagues have proposed a new type of flexible and self-healing all-fiber-based solid-state lithium-ion battery shaped like a spring. The SnO_2 quantum dots @ reduced graphene oxide (RGO) and $LiCoO_2$ were used as negative and positive electrodes, respectively. The electrolyte used poly(vinylidenefluoride-co-hexa-fluoropropylene, PVDF-co-HFP) soaked in $LiClO_4$ with EDC/EC. Self-healing carboxylated polyurethane (PU) was used as the packaging material for self-healing the spring-like LIBs as shown in Fig. 5(a) [34]. The self-healing spring-like LIBs exhibited good electrochemical performance. The capacity of the flexible self-healing LIBs was 82.6 $mAh \cdot g^{-1}$ under a series of mechanical deformations. When the current density was 0.1 $A \cdot g^{-1}$, it maintained a capacity of 50.1 $mAh \cdot g^{-1}$ after the fifth healing. Polydimethylsiloxane (PDMS) exhibits excellent mechanical strength and flexibility. Most of the currently reported thin-film batteries are encapsulated with PDMS to maintain good deformability. The flexibility of thin-film batteries may mainly originate from the PDMS packaging materials, as the flexibility of the electrode or electrolyte materials has not been well proven. Koo *et al*. [36] have reported bendable inorganic thin-film battery based on PDMS that was solid-state (Fig. 5(b)). The solid-state bendable battery exhibited a capacity of 106 $\mu Ah \cdot cm^{-2}$.

PDMS was also known as a silicone high-elastomer with good tensile properties that can withstand more than 100% stretch. Hence, it is widely used in stretchable flexible LIBs. Xu *et al*. [56] have designed a stretchable flexible battery with a typical structure and PDMS, which was used to isolate the electrodes and electrolytes from the air to protect them. As shown in Fig. 5(c), the stretchable flexible battery exhibited stretchable deformation ability. The excellent stretchability was attributed to the self-similar serpentine interconnects and low-modulus silicone elastomers. The stretchable batteries can achieve reversibility of up to 300% while maintaining a capacity density of 1.1 $mAh \cdot cm^{-2}$.

Compared with aluminum-plastic films, the flexibility of polymer materials is more suitable for the packaging and practical application of thin-film batteries. Flexible packaging is a novel approach for next-generation flexible electronic devices. However, polymer packaging materials have some deficiencies. For example, waterproofing ability is not as good as that of aluminum-plastic films. In the future, improved flexibility of aluminum-plastic films is a great challenge for the development of flexible LIBs.

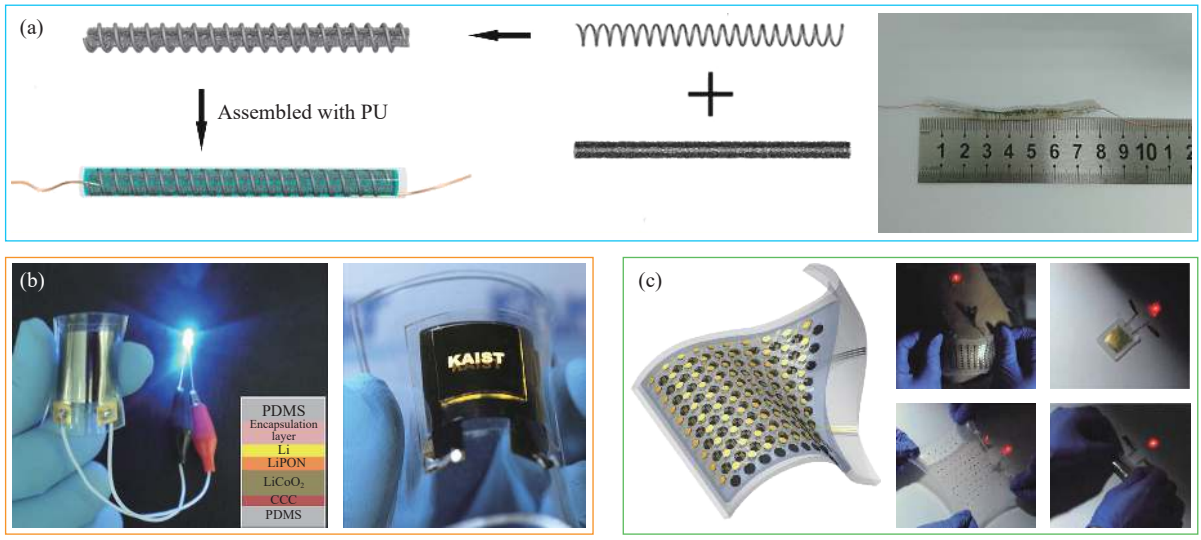


Fig. 5. Flexible polymer packaging materials: (a) self-healing carboxylated polyurethane (PU); (b) photographs of a bendable inorganic thin-film battery with PDMS packaging material; (c) stretchable batteries with PDMS. (a) Reprinted from *Nano Energy*, 51, J.Y. Rao, N.S. Liu, Z. Zhang, *et al.*, All-fiber-based quasi-solid-state lithium-ion battery towards wearable electronic devices with outstanding flexibility and self-healing ability, 425-433, Copyright 2018, with permission from Elsevier [34]; (b) reprinted with permission from M. Koo, K.I. Park, S.H. Lee, *et al.*, Bendable inorganic thin-film battery for fully flexible electronic systems, *Nano Lett.*, 12(2012), No. 9, p. 4810 [36]. Copyright 2012 American Chemical Society; (c) reprinted by permission from Springer Nature: *Nat. Commun.*, Stretchable batteries with self-similar serpentine interconnects and integrated wireless recharging systems, S. Xu, Y.H. Zhang, J. Cho, *et al.*, Copyright 2013 [56].

3. Mechanical structure of flexible batteries

In recent years, great efforts have been made to realize flexibility by changing the battery design. During mechanical deformation, it was observed that the active materials were removed from the current collector, increasing the contact resistance [9,58–60]. Therefore, it is necessary to design flexible LIBs based on the traditional industrial electrode structure and manufacturing methods that maintain relatively high energy density and electrochemical performance. A series of novel structural designs were applied to flexible LIBs to enhance their mechanical flexibility and electrochemical performance stability. When the flexible LIBs were deformed, the stress concentration mainly occurred in specific regions. In this section, the recent developments of flexible LIBs with regard to structural design are discussed, including electrode configurations and battery configurations.

3.1. Patterned configuration

The development of the electrode configuration is reviewed. The electrode configurations mainly focus on designing novel electrode structures and optimizing traditional electrode structures to enhance adhesion and mechanical stability, such as fiber electrodes, kirigami-based planar electrodes, stretchable electrodes, and 3D electrodes. Park *et al.* [42] have designed post-patterned electrodes based on a traditional electrode, which were then wound into node-type flexible LIBs (Fig. 6(a)). The post-patterned electrodes were fabricated using an additional traditional electrode prepared by slurry coating and rolling processes. The traditional electrode exhibited delaminated active materials and cracks in both positive and negative electrodes after 200 cycles (Fig. 6(b)). Nevertheless, the post-patterned electrodes retained a complete surface with no damage. Furthermore, the node-type flexible LIBs with post-patterned electrodes exhibited better electrochemical performance and flexibility. The node-type flexible LIBs could power LED under different bending states (Fig. 6(c)). After bending 3000 times, up to 80% capacity was retained (Fig. 6(d)), and the node-type flexible LIBs showed no discernible capacity loss compared with unstressed LIBs.

However, post-patterned electrodes may cause an electrode contact mismatch during battery assembly, and the preparation processes are complicated, increasing the time and cost [61]. Therefore, this research group has designed a 3D interlocking electrode structure with opposite patterning. The 3D interlocking electrode structure was fabricated by patterned rolling the entire cell, as shown in Fig. 6(e) [61]. Compared to the above patterning process, the 3D interlocking structure electrodes exhibited no structural deformation under mechanical deformation, mainly because of the good matching between the electrodes. The 3D interlocked LIBs exhibited better electrochemical performance, with 80% capacity retention after 100 cycles after flexing 4000 times (Fig. 6(f)). While the normal LIBs retained only 50% of their capacity. The 3D interlocked LIBs were used to power LED under mechanical deformation, as shown in Fig. 6(g) and (h). The patterned electrode structure demonstrates that the mechanical and electrochemical stability of flexible LIBs can be enhanced by designing commercial electrode configurations. These studies provide a new opportunity for the development of high-energy flexible LIBs based on manufacturing processes of the traditional LIBs. However, during preparation processes of patterned electrodes may cause irre-

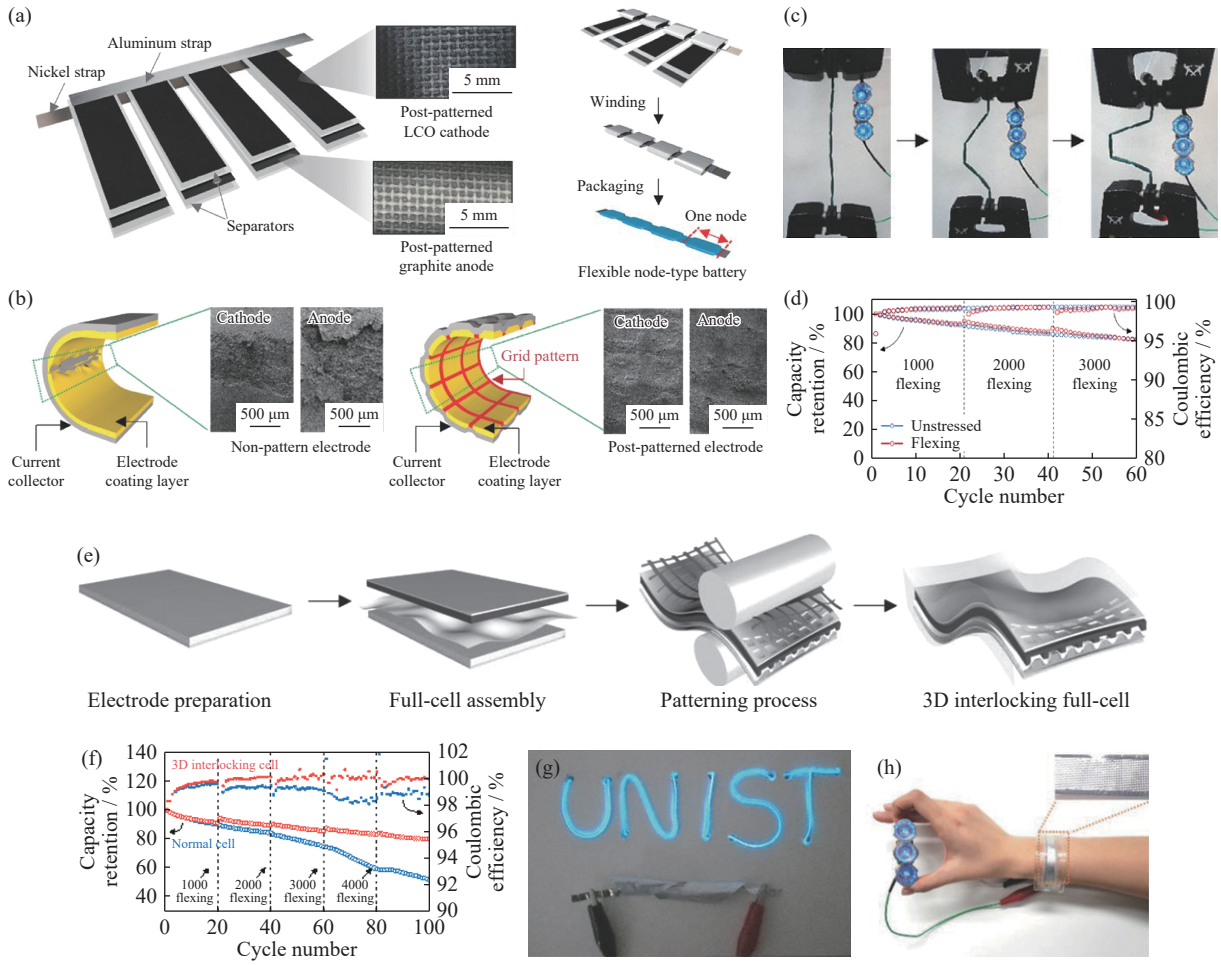
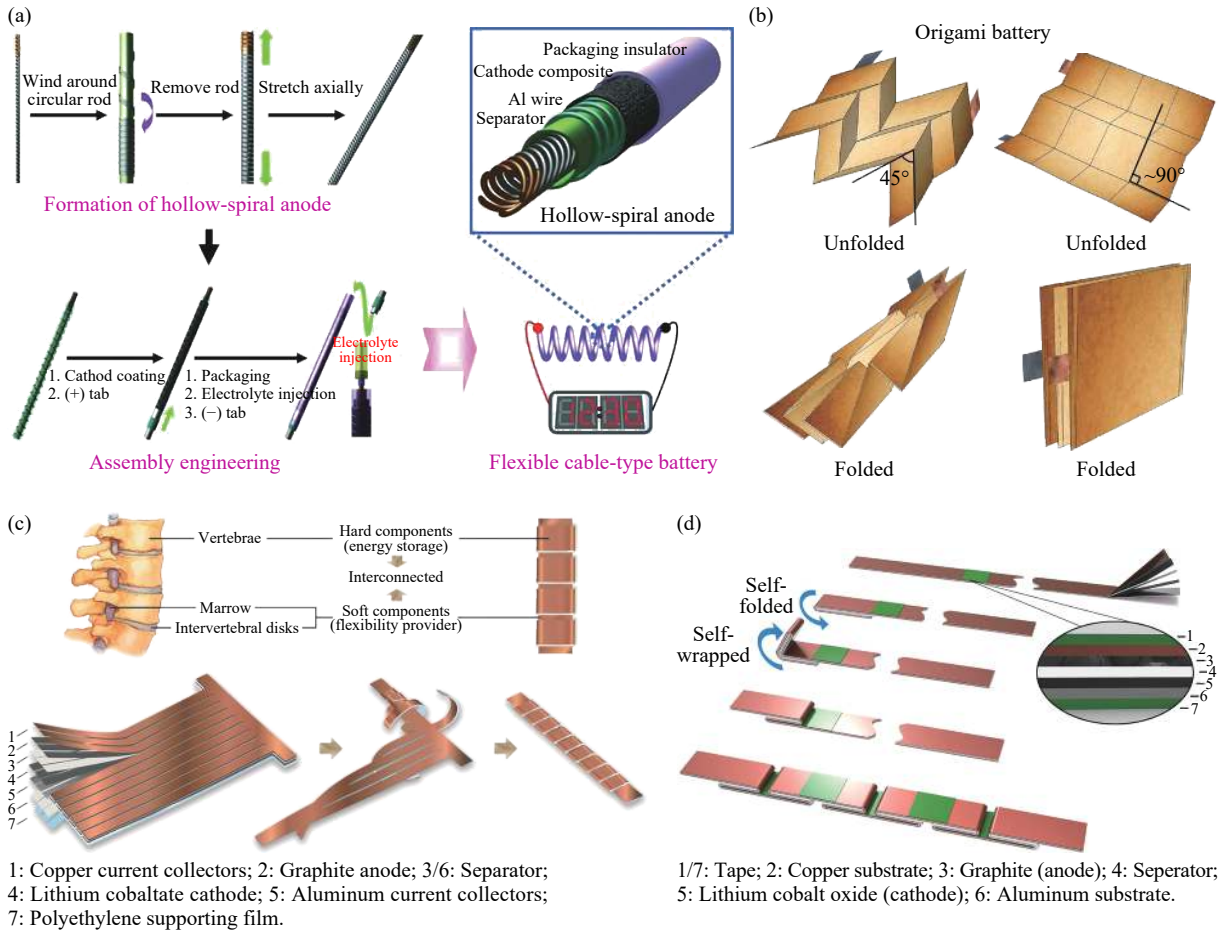


Fig. 6. Patterned configuration: (a) schematic of the node-type LIBs with postpatterned $LiCoO_2$ and graphite electrodes; (b) damage sites of nonpatterned and postpatterned electrodes after 200 cycles; (c) photographs of the node-type LIBs under flexing test; (d) cycle performance of the node-type LIBs with flexing and in the unstressed state; (e) schematic of the assembly process for flexible 3D interlocking LIBs; (f) cycle performance of flexible 3D interlocking LIBs; (g–h) application scenario display of flexible 3D interlocking LIBs. (a–d) M. Park, H. Cha, Y. Lee, J. Hong, S.Y. Kim, and J. Cho, *Adv. Mater.*, 29, 1605773 (2017) [42]. Copyright 2017 Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission; (e–h) H. Cha, Y. Lee, J. Kim, M. Park, and J. Cho, *Adv. Energy Mater.*, 8, 1801917 (2018) [61]. Copyright 2018 Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission.

versible damage to the original electrodes, which would make the electrchemical performance of battery decreased owing to side reactions.

3.2. Full battery configuration

Recent developments in battery configurations for flexible LIBs have been widely reported. The battery configurations focus on multidimensional structures. The novel flexible LIBs configurations mainly include fabric-like [62], cable-type [37], fabric-like [63], wave-like [29,39,64], and origami structures [65]. The fabric-type lithium-ion battery has been considered to have great potential in flexible wearable devices [66]. Peng's research group has been committed to developing fiber-like batteries in recent years, with the goal of large-scale industrialization of fiber batteries. To manufacture long fiber electrodes and high-performance flexible fiber batteries has become a key problem [67–69]. Kwon *et al*. [37] have designed a cable-type battery with hollow multihelix electrodes, in which the balance of battery capacity was achieved by adjusting the number of negative electrodes and the thickness of the positive electrodes (Fig. 7(a)). The cable-type battery can be applied in any field owing to linear shape and omni-bearing flexibility, providing a reliable strategy for the realization of wearable devices. The cable-type battery belongs to 1D linear structure design, which may damage the fiber electrode with deformations. The electrode damage can be alleviated by reserving the stress concentration positions based on Origami technology. Origami/kirigami technology can be applied to the optimal design of traditional LIBs for planar structures. Recently, Song *et al*. [65] have reported origami LIBs with good folding, bending, and twisting properties, as shown in Fig. 7(b). The origami LIBs were fabricated by Miura folding to form "mountain" and "valley" creases, which played a connecting role. Hence, the origami LIBs can be compressed in one or two directions to realize various angular deformations. Owing to the limitations of the origami structure in the stretching direction, this team also designed kirigami-based flexible LIBs to expand stretchability by combining cutting and folding. The cable-type and origami flexible LIBs exhibited good



1: Copper current collectors; 2: Graphite anode; 3/6: Separator; 4: Lithium cobaltate cathode; 5: Aluminum current collectors; 7: Polyethylene supporting film.

1/7: Tape; 2: Copper substrate; 3: Graphite (anode); 4: Seperator; 5: Lithium cobalt oxide (cathode); 6: Aluminum substrate.

Fig. 7. Battery configurations: (a) cable-type battery; (b) origami LIBs; (c) spine-like structure; (d) zigzag-like design. (a) Y.H. Kwon, S.W. Woo, H.R. Jung, *et al*., *Adv. Mater*., 24, 5192 (2012) [37]. Copyright 2012 Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission; (b) reprinted by permission from Springer Nature: *Nat. Commun*., Origami lithium-ion batteries, Z.M. Song, T. Ma, R. Tang, *et al*., Copyright 2014 [65]; (c) G.Y. Qian, B. Zhu, X.B. Liao, *et al*., *Adv. Mater*., 30, 1704947 (2018) [70]. Copyright 2018 Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission; (d) X.B. Liao, C.M. Shi, T.Y. Wang, *et al*., *Adv. Energy Mater*., 9, 1802998 (2019) [71]. Copyright 2019 Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission.

electrochemical performance, while their energy density was relatively low.

Although great progresses have been made in flexible LIBs, it remains challenging to simultaneously obtain high flexibility and high energy density. In order to elevate the high energy density, inspired by the spine structure, Qian *et al*. [70] have designed a spine-like flexible, as shown in Fig. 7(c). The spine-like configuration realized the decoupling of energy storage regions and the flexible regions that endured deformation. The spine-like flexible LIBs have used $LiCoO_2$ and graphite as the positive and negative electrodes, respectively. The energy density of spine-like flexible LIBs exceeds 85% of that of prismatic batteries with the same volume. The energy density of the flexible full LIBs can reach 242 $W{\cdot}h{\cdot}L^{-1}$ because 90% of the area is used to store energy. The spine-like batteries maintain excellent performance under dynamic and different mechanical deformations owing to the decoupling design. Inspired by origami folding, this team also reported a zigzag-like battery with high foldability and high energy density, as shown in Fig. 7(d) [71]. As the length ratio of the foldable flexure hinge between the rigid electrode parts can approach zero, the energy density of zigzag-like batteries could reach 275 $W{\cdot}h{\cdot}L^{-1}$, which is 96.4% of that of traditional stacking batteries. The zigzag-like battery showed stable cycle performance during 45000 cycles of continuous dynamic loading with 130 degrees.

The high energy density flexible LIBs, such as spine-like and zigzag-like, can bend only in a single direction. Flexible LIBs may require arbitrary deformations for practical applications. Our research group reported novel bidirectional flexible snake-origami LIBs inspired by chemical molecular structures, as shown in Fig. 8(a) [72]. The bidirectional flexible snake-origami batteries are composed of rigid and soft regions. The rigid regions were used to provide energy, and soft regions were designed as deformation units. The flexible snake-origami batteries exhibited a stable electrochemical performance during different deformations, as shown in Fig. 8(b). As shown in Fig. 8(c) and (d), compared to the previous and commercial flexible LIBs, the flexible snake-origami batteries showed a high energy density of 357 $W{\cdot}h{\cdot}L^{-1}$ and unique bi-directional deformation owing to the decoupling design. We have also established a principle between geometric parameters, energy density, and flexibility, which is used to demonstrate the validity of utilizing rigid-soft

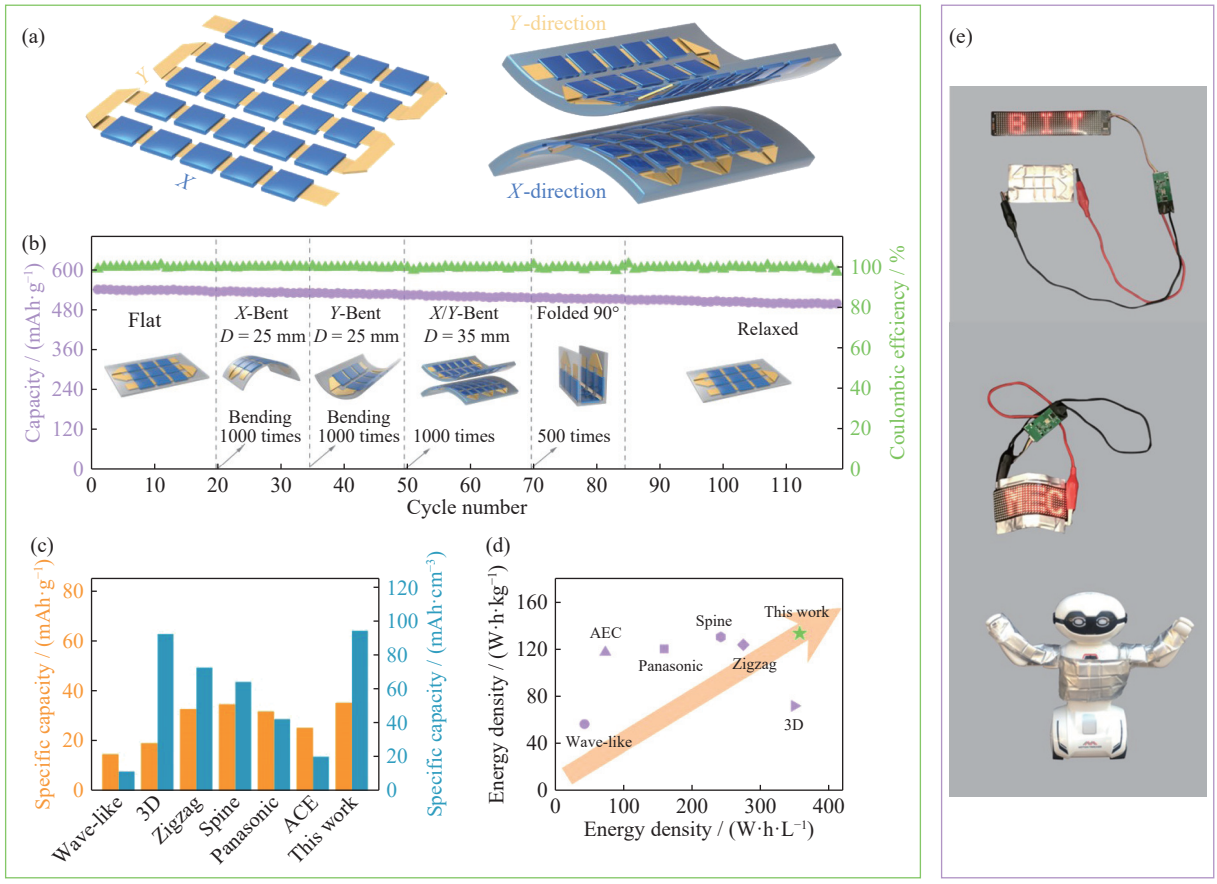


Fig. 8. Bi-directional snake-origami batteries: (a) schematic illustration; (b) electrochemical performance under different mechanical deformations; (c–d) performance comparison with previously reported battery; (e) snake-origami batteries powered flexible LED display and robot. Reprinted from Ref. [72].

coupled structures to endure various deformations [72]. The flexible snake-origami batteries were used to provide energy for flexible LED screens and smart robots to demonstrate possible application scenarios.

In summary, the structural design has proven to be an effective strategy for flexible batteries with both high energy density and excellent mechanical stability. During mechanical deformation, the stress concentration mainly emerges in specific areas of the flexible LIBs. However, the overall performance of the battery is slightly affected by continuous deformations owing to unstable kinetic reactions and the uneven distribution of electrolytes. When designing a flexible battery, it is necessary to carefully consider the balance between energy density, geometric configuration, and flexibility, which is used to achieve balanced performance for practical application. Therefore, it is still a great challenge to realize the actual flexibility and no damage, as the flexible LIBs must maintain stable electrochemical reactions under various mechanical loads. In the future, the coupling design of flexible material and battery structure is the development direction of the flexible battery.

4. Mechano-electrochemical coupling evaluation

Flexible LIBs have made great progress in terms of flexible materials and structural design in recent years, providing important technical support for the development of wearable devices. However, effective evaluation of the flexibility, durability, and electrochemical performance of flexible LIBs has become a key challenge. Recently reported evaluation methods and parameters are diverse and lack unified standards, which makes it difficult to accurately and uniformly evaluate flexible LIBs. The evaluation of flexible LIBs should be based on mechano-electrochemical processes. The common deformations mainly included bending, stretching, folding, and twisting. The evaluation methods mainly focus on finite element simulation, geometric parameters, and electrochemical analysis.

4.1. Mechanical stability

Numerical simulations, such as finite element simulations, are widely used to analyze the stress distribution of flexible batteries under mechanical loadings [73–75]. As shown in Fig. 9(a) and (b), the stress distribution of the joint-like flexible battery was studied using finite element simulation under different deformation states [76]. The finite element simulation results have demonstrated that the stress was mainly concentrated on the connecting part between the two cylinders. The stress distribution of electrode materials with different shapes can also be analyzed using a finite element simulation. Chen and his colleagues have analyzed different

electrode configurations, and found that a wavy electrode structure was more suitable for the flexible batteries. The simulation results have demonstrated that the wrinkled structure can release strain by changing its shape with an out-of-plane bend, as shown in Fig. 9(c), even though the electrode materials are stiff [77]. Song *et al*. [65] designed an origami battery that can withstand high-strength deformation, which can create a compact, deformable 3D structure by folding two-dimensional planar structures along predetermined creases. In Fig. 9(d), the finite element simulation results of the origami structure under different deformations indicate that the strain was very small at a strain level of 0.001%, which demonstrated the stability of the battery structure [65]. Qian *et al*. [70] have analyzed the effective bending stiffness of spine-like batteries using an analytic method. They defined a relative bending efficiency parameter, k/k_0, that describes flexiblity. As shown in Fig. 9(e), k/k_0 is a function of the gap length, denoted by x. The finite element analysis of the stress distribution of the spine-like, prismatic, and stacked batteries in a bending state was conducted. The results demonstrated that the spine-like structure had high flexibility, as shown in Fig. 9(f), with the stress mainly concentrated in the gap. For the structural design of flexible batteries, finite element simulation is a very effective method for determining the rationality and stress distribution of structures. Nevertheless, the model of battery structure in finite element simulation is quite different from the real geometric configuration, ignoring the influence of the electrochemical environment.

4.2. Mechanical and electrochemical coupling process

The flexible LIBs can bring about opposite tensile/compressive stresses on the electrodes under mechanical loading. The geometric parameters commonly used to evaluate the flexibility of flexible limbs are R, θ, and L. R is used to de-

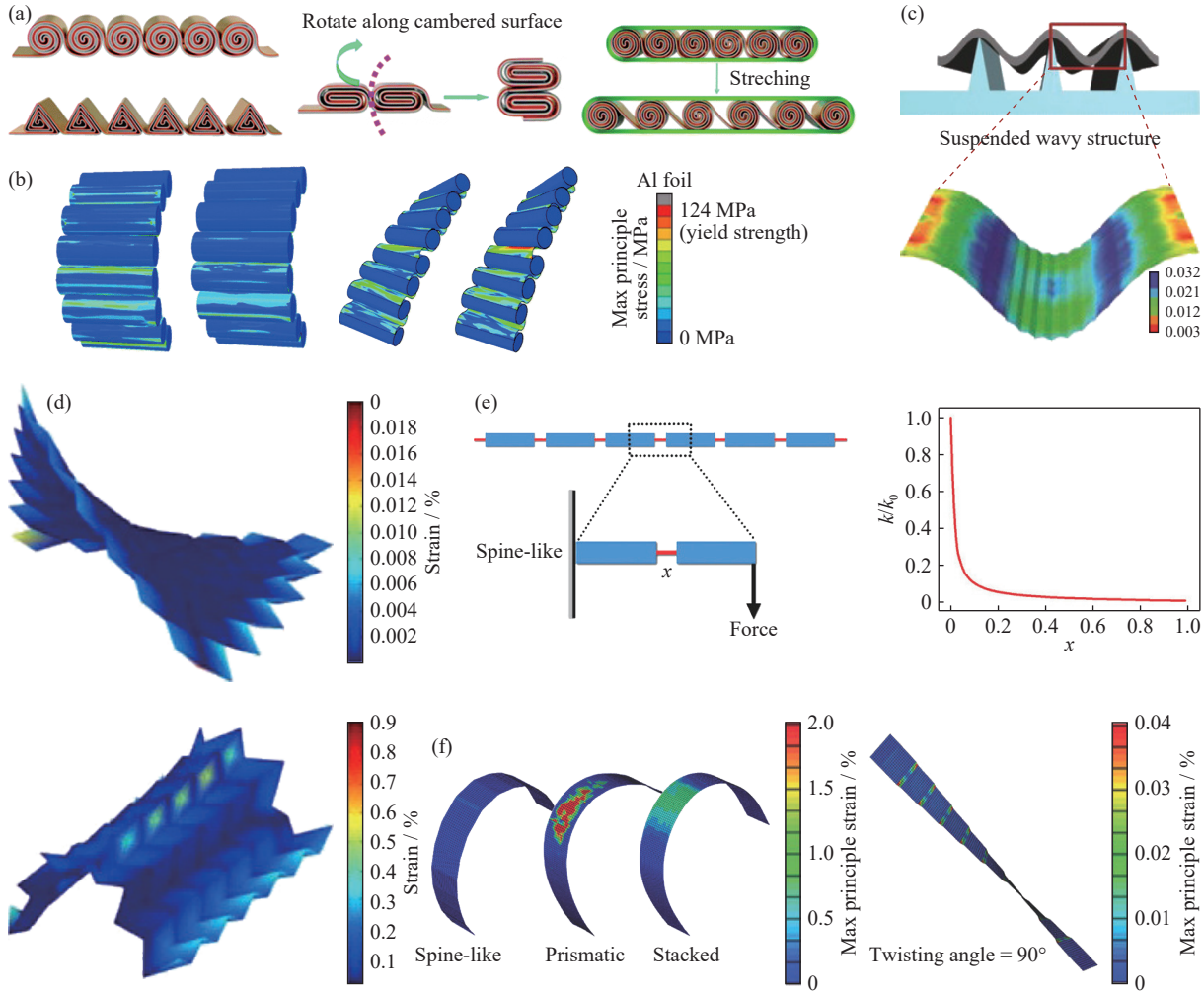


Fig. 9. Finite element simulation: (a) human-joint-inspired structural design; (b) finite element simulation results of a bendable and twistable flexible battery; (c) strain distribution of wavy-like battery; (d) strain distribution of Origami LIBs; (e–f) finite element simulation of spine-like battery. (a–b) reprinted with permission from A. Chen, X. Guo, S. Yang, *et al*., Human joint-inspired structural design for a bendable/foldable/stretchable/twistable battery: Achieving multiple deformabilities, *Energy Environ. Sci*., 14(2021), No. 6, p. 3599 [76]. Copyright 2021 Royal Society of Chemical; (c) D.P. Qi, Z.Y. Liu, Y. Liu, *et al*., *Adv. Mater*., 27, 5559 (2015) [77]. Copyright 2015 Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission; (d) reprinted by permission from Springer Nature: *Nat. Commun*., Origami lithium-ion batteries, Z.M. Song, T. Ma, R. Tang, *et al*., Copyright 2014 [65]; (e–f) G.Y. Qian, B. Zhu, X.B. Liao, *et al*., *Adv. Mater*., 30, 1704947 (2018) [70]. Copyright 2018 Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission.

scribe the bending radius, and θ is the bending angle. The distance between the two ends of the cell after bending is L. While the three parameters are suitable for macro-scale stress analysis, they are not suitable for accurately describing the stress distribution of internal multi-layer structures. If parameter b is used to describe the thickness of a neutral plate, then a multilayer battery structure can be regarded as a composite plate structure with the friction force between the plates determined using the analytical method. The frictional force is related to the thickness of the neutral plane and the force caused by bending. However, the change in the friction coefficient due to the uneven distribution of the electrolytes is not considered. Thus, the analysis of the accuracy of the internal stress distribution requires further study. The internal environment of a flexible battery is a multifield coupling environment under mechanical loading. The changes in the internal structure and environment are dynamic processes, which complicate the accurate description of the internal stress distribution.

Currently, most researchers have focused on the stability of the electrochemical performance during different mechanical deformations described by the above geometric parameters. In Fig. 10(a) and (b), Kown *et al*. [37] have investigated the voltage stability with various bending deformations every 20 min, with cable-type batteries exhibiting good voltage stability. In addition, Li *et al*. [78] have also reported three parameters (L, θ, and R) to explain the relationship between the electrochemical performance and the bending state, where L is also described as the device length, as shown in Fig. 10(c). The capacity retention was used to represent the stability of

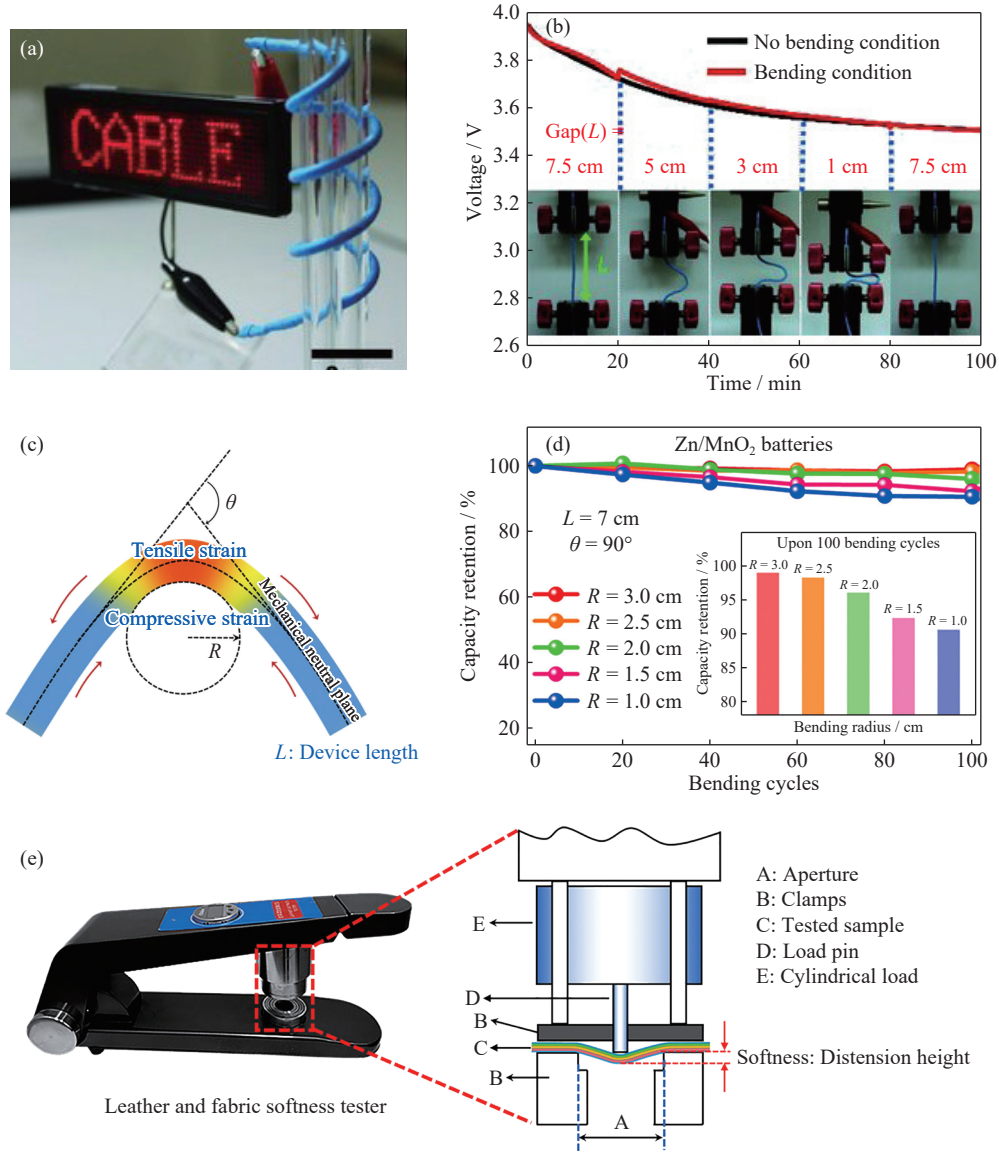


Fig. 10. (a) Photographs of cable-type battery under twisting deformation; (b) discharge curves with different bending states (L denotes the distance between the two ends, which describes the bending state); (c) schematics of the bending state with three key parameters; (d) capacity retention of flexible batteries; (e) schematic illustration of softness. (a–b) Y.H. Kwon, S.W. Woo, H.R. Jung, *et al*., *Adv. Mater*., 24, 5192 (2012) [37]. Copyright 2012 Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission; (c–e) reprinted from *Joule*, 3, H.F. Li, Z.J. Tang, Z.X. Liu, *et al*., Evaluating flexibility and wearability of flexible energy storage devices, 613-619, Copyright 2019, with permission from Elsevier [78].

the electrochemical performance under changing geometric parameters. The capacity retention decreased with a decrease in bending radius, as shown in Fig. 10(d), indicating that capacity retention is closely related to the geometric parameters. In Fig. 10(e), softness is defined as the distension height and was used to represent the deformation ability. In conclusion, the researchers have proposed evaluation parameters and corresponding test methods for flexible batteries and electrode materials. Most experimental characterization methods are based on measuring the electrochemical performance under different deformation processes and evaluating the deformability with corresponding geometric parameters.

The change in the charge-discharge curves under deformation has also been explained by the relationship between mechanical loading and electrochemical reactions. As shown in Fig. 11(a) and (b), the zigzag-like flexible batteries were tested under different mechanical deformations [71]. The charge-discharge curves exhibited good stability in the bend, flexed, and folded states, as shown in Fig. 11(a). The charge-discharge curves exhibited regular changes with continuous folding/bending states, which demonstrated that the electrochemical reactions were affected by folding deformation. The voltage fluctuations were lower than 1 mV. Qian *et al.* [70] have proved the structural stability of spine-like batteries with dynamic flexing. The spine-like battery exhibited relatively stable electrochemical performance under dynamic flexion, as shown in Fig. 11(c). In contrast, the prismatic batteries exhibited severe voltage jitter with dynamic flexion, as shown in Fig. 11(d). The discharge voltage of the spine-like battery was stable even with continuous flexion and twisting, as shown in Fig. 11(e), with voltage fluctuation lower than 0.02 mV in Fig. 11(f). Therefore, the spine-like battery exhibited excellent flexibility and stable electrochemical performance.

From the charge-discharge curves of the flexible LIBs in Fig. 11 all showed voltage jitter under deformations. Hence, the polarization and other side reactions may occur in the battery. The kinetics of electrode processes is constantly changing with dynamic flexion. To summarise, the service process of flexible batteries is a mechano-electrochemical process. The voltage curve and electrochemical performance are only macroscopic representations of the battery, and they cannot represent internal electrode reaction changes. The above phenomena can only indicate that the internal electrochemical environment has changed. Yet the underlying reasons need to be analyzed and characterized by other experimental techniques.

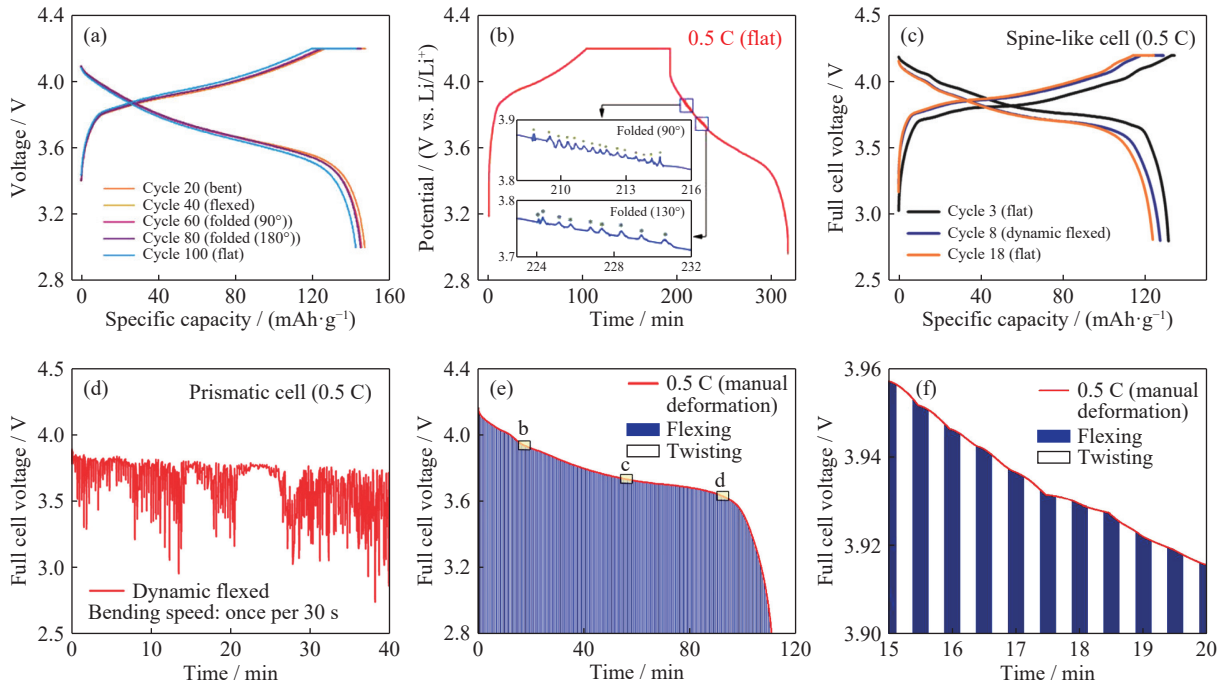


Fig. 11. Mechanical and electrochemical coupling process: (a–b) charge-discharge curves of zigzag-like flexible batteries with various mechanical deformations; (c) charge-discharge curves of spine-like flexible batteries under dynamic flex testing; (d) charge-discharge curves of prismatic batteries under dynamic flex testing; (e–f) discharge curve of the spine-like battery under continuously flexed and twisted states. (a–b) X.B. Liao, C.M. Shi, T.Y. Wang, *et al.*, *Adv. Energy Mater.*, 9, 1802998 (2019) [71]. Copyright 2019 Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission; (c–f) G.Y. Qian, B. Zhu, X.B. Liao, *et al.*, *Adv. Mater.*, 30, 1704947 (2018) [70]. Copyright 2018 Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission.

4.3. Kinetics of electrode processes

The electrode reactions are affected by the internal electrochemical environment, which changes with mechanical deformation. Hence, the electrode reactions depend on the mechano-electrochemical coupling process, which requires both excellent flexibility and stable electrochemical reaction processes. Establishing the relationship between mechanical flexibility and electrochemical performance to evaluate the

comprehensive performance of flexible batteries is a key problem.

When the battery is subjected to a certain external load, the internal complex electrochemical environment changes dynamically. Electrochemical impedance spectra (EIS) is commonly used to study the kinetics of electrode processes. Our group has used EIS to study the change in the kinetics of electrode processes with different mechanical deformations. Fig. 12(a) shows that there was a slight shift in the EIS spectra with every kind of mechanical deformation [72]. The distribution of relaxation time (DRT) method was used to further analyze the EIS results in Fig. 12(b). The resistance change may be caused by variations in the local electrolyte distribution or kinetics of electrode processes. Cho and his colleagues have reported 3D interlocked flexible LIBs with 3D patterned electrodes. The 3D interlocked flexible LIBs were flexed 5000 times, and the battery was disassembled for examination of the electrode. The disassembled electrodes were then reassembled into a half-cell. Electrochemical impedance tests were performed on the reassembled half-cells under a different state of charge (SOC). The images in Fig. 12(c) show that there were no significant changes in the dis-

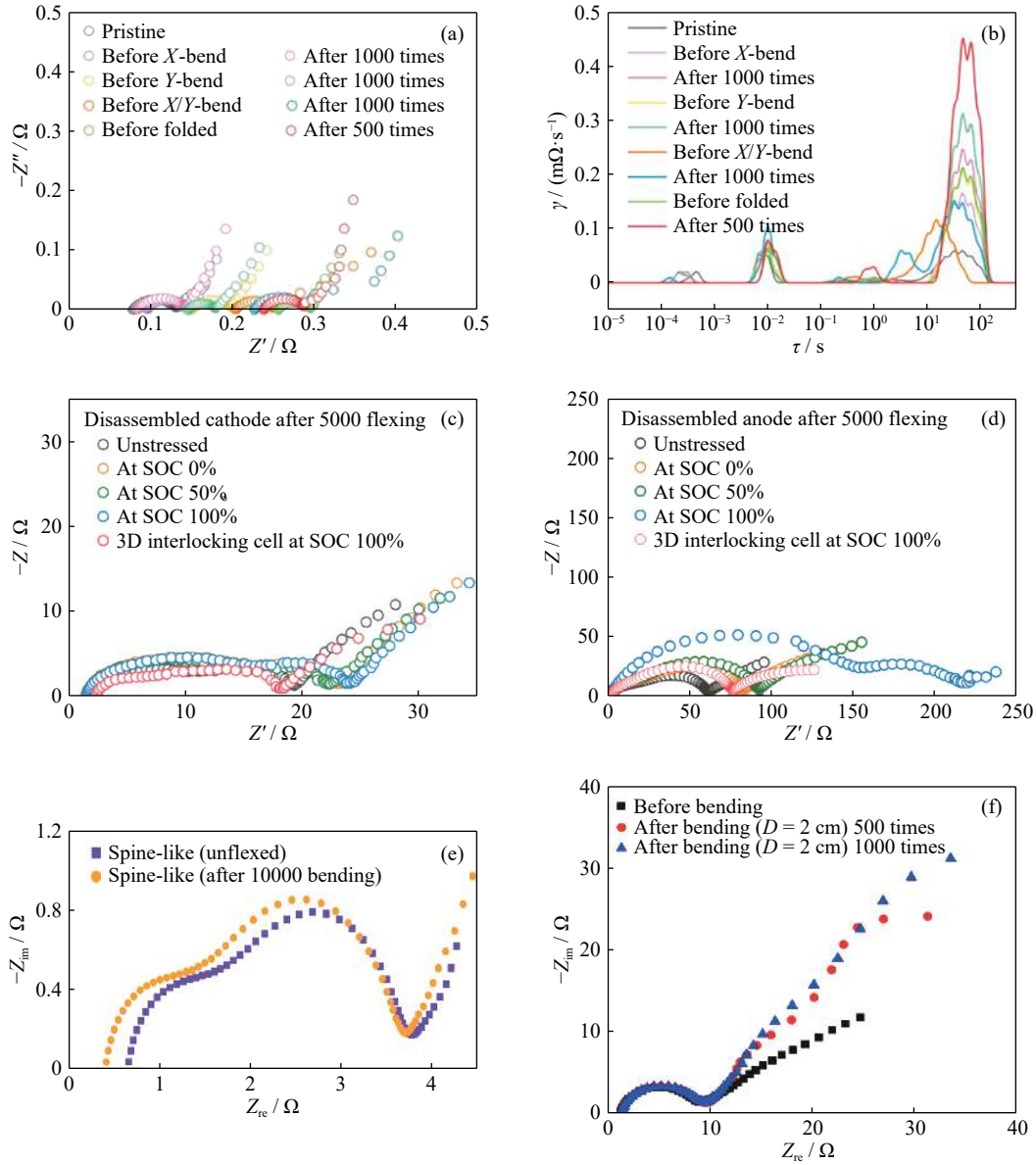


Fig. 12. Kinetics of the electrode processes: (a) electrochemistry impedance spectra and (b) distribution of relaxation time of bi-directional flexible LIBs with various mechanical deformations; electrochemistry impedance spectra of normal and 3D interlocking LIBs with the (c) reassembled cathode and (d) anode after flexing 5000 times; (e) electrochemistry impedance spectra of spine-like flexible LIBs for the unflexed state, and after bending 10000 times; (f) flectrochemistry impedance spectra of zigzag-like flexible batteries under different bending states. (a–b) Reprinted from Ref. [72]; (c–d) H. Cha, Y. Lee, J. Kim, *et al*., *Adv. Energy Mater*., 8, 1801917 (2018) [61]. Copyright 2018 Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission; (e) G.Y. Qian, B. Zhu, X.B. Liao, *et al*., *Adv. Mater*., 30, 1704947 (2018) [70]. Copyright 2018 Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission; (f) X.B. Liao, C.M. Shi, T.Y. Wang, *et al*., *Adv. Energy Mater*., 9, 1802998 (2019) [71]. Copyright 2019 Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission.

assembled positive electrode after flexing 5000 times, which demonstrates that the patterning process had little influence on the positive electrode [61]. It was found that the impedance changes of the disassembled graphite electrode were obvious with different SOC states, particularly the solid electrolyte interphase (SEI) resistance and charge transfer resistance, as shown in Fig. 12(d). When the flexible LIBs are subjected to several deformations, the adhesion between the components is weakened, leading to severe stratification or delamination of the active materials. Eventually, the SEI films reformed on the surface, and these active materials were removed from the electrode. As shown in Fig. 12(e)–(f), some researchers have studied the electrochemical impedance spectra of flexible batteries before and after bending, and the electrochemical impedance changes slightly [70–71]. This indicates that the internal electrochemical environment of the flexible battery changed with bending, and the corresponding kinetics of electrode processes were also affected. Electrochemical impedance can reflect the changes in the internal kinetic parameters to a certain extent. While most researchers have only studied the EIS of a single state, and the EIS with both various states and processes has been lacking.

Hence, it is necessary to establish the relationship between the charge-discharge curve, electrochemical impedance, and mechanical parameters, which are used to describe the mechano-electrochemical process. In conclusion, the internal environment of flexible batteries is affected by a complex multi-field coupling process. How to establish the relationship between macro deformation and internal dynamic change is the key challenge. We can develop novel experiments to explore the mechano-electrochemical mechanism, such as in-situ technology. Therefore, it is necessary to establish a multi-field coupling method to evaluate the flexibility and electrochemical performance of flexible batteries.

5. Conclusions and perspectives

5.1. Conclusions

In this review, we have discussed the progress of flexible LIBs concerning flexible materials, structural design, and evaluation methods. Firstly, flexible materials and structural design have been used to improve the flexibility of flexible batteries. The flexible electrodes and current collectors have been used to solve the active material delamination, and ensured the interface contact during the bending tests, such as free-standing electrodes. The electrolyte leakage during flexing tests, leading to unsafe events, is the key issue to achieving the stable performance of flexible LIBs. Therefore, flexible solid polymer electrolytes have been widely studied for flexible LIBs due to their high safety. The traditional packing materials are relatively rigid and have been replaced by PDMS and other polymer materials to promote flexibility. Secondly, in an effort to improve energy density and flexibility, structural designs of flexible LIBs have focused on electrode configurations and full-cell configurations. The main electrode configurations have focused on multi-dimension structures, including thin films, wavy-like, fabric-like, and island connections. However, combining the electrode configurations with flexible packing materials can improve the flexibility and stability of flexible LIBs. Novel configurations with the decoupling of energy storage and mechanical deformation have been designed, such as spine-like, zigzag-like, node-type, and snake-origami, allowing unprecedented electrochemical performance. In conclusion, the design rules of flexible LIBs should consider the balance between mechanical flexibility, energy density, and electrochemical performance. Up to date, the evaluations of flexible batteries have focused on numerical simulations and experimental characterizations that are used to analyze mechanical flexibility and electrochemical stability. The evaluation methods for the flexible batteries have not been unified, and it is necessary to develop a unified characterization method or evaluation standard. For example, the kinetics of electrode processes can be affected by mechanical loading, while the mechanism is not clear.

Although great progresses have been made in the flexible batteries in recent years, there are still some challenges that need to be overcome. In the future, the development of flexible batteries can focus on the following challenges: (1) novel structure design with arbitrary deformation direction; (2) development of unified evaluation methods and standards; (3) exploring of the mechanism about how mechanical deformations affect the electrode reactions; (4) large-scale and low-cost fabrication of flexible batteries. The development of flexible LIBs will promote the commercialization of wearable and flexible devices.

5.2. Perspectives

The electrode process of flexible LIBs can be divided into the following parts: liquid-phase mass transport in the electrolyte, charge transfer on the surface, and solid-phase diffusion in the particles [79]. The mechanical deformation has an impact on the electrode process, and the mechano-electrochemical mechanism is discussed based on the kinetics of electrode processes.

5.2.1. Mechano-electrochemical effects on the liquid-phase mass transport

It is noted that the mechanical forces directly impact the geometrical parameters of the flexible batteries, and mechano-electrochemical effects can vary in different electrode processes. In the liquid-phase mass transport, the mechanical influence is affected by the geometry of the internal structures, including the porous electrode, separators, and voids of the flexible batteries. The geometric parameters of the flexible LIB structure changed during the deformation process. When a certain force is applied to flexible LIBs, the battery deforms. The deformation of flexible LIBs can result in a change in the connection between the electrode and separator, and the spacing distribution between separator and electrode becomes inconsistent. For example, the porosity and tortuosity of the separators changed with deformation, and deformation of flexible LIBs may cause an uneven distri-

bution of electrolytes. In the flexible LIBs, the electrochemical processes of liquid-phase mass transport would be affected by the porosity and tortuosity of the separators, which results in voltage polarization and non-uniform ionic current between the electrodes.

5.2.2. Mechano-electrochemical effects on charge transfer on the surface

In the LIBs, the interface reactions induced by charge transfer can be divided into the following types: lithium dendrites, electrolyte decomposition, and solid electrolyte interphase (SEI). The interface reactions are related to charge transfer on various types of particle surfaces. Charge transfer depends on many factors, such as ion concentration, overpotential, and effective contact area. When a certain force is applied to flexible LIBs, some changes occur. In particular, there may be an uneven distribution of electrolytes and Li ions on the surface of the reaction particles, as well as overpotential changes on the local surface induced by the displacement of the electrode and separators. The separators and electrodes away from each other result in changes in the interface resistance in terms of geometric changes. The uneven distribution of electrolytes causes non-uniform ion distribution, resulting in non-uniformity of surface polarization. In addition, the peeling of the active material off the current collector has an impact on electron transfer, leading to the loss of reactivity. Therefore, the interface reactions are affected by the deformations of the flexible LIBs.

5.2.3. Mechano-electrochemical effects on the solid-phase mass transport

The solid-phase reaction is related to the ionic diffusion coefficient in the solid, which is affected by the mass transport, ionic diffusion at the porous electrode surface, and solid-phase diffusion [79]. The stress concentrates on the electrodes, resulting in surface morphology changes, cracks in the electrodes, phase transformations, etc. These challenges of electrodes affect the ion transport path and ionic diffusion coefficient. Furthermore, the uneven distribution of stress leads to the change in the intermolecular forces. Intermolecular forces have an impact on lattice mismatch and can result in possible phase formation/transformation.

Overall speaking, the mechanical deformations on flexible LIBs would have an impact on the kinetics of electrode processes. Without carefully considering the mechano-electrochemical effects, the mechanical deformations would induce unexpected side reactions, such as electrolyte decomposition, the evolution of lithium dendrites, and gas emission.

Acknowledgements

This study was supported by National Natural Science Foundation of China (No. 52074036), Technology Innovation Program of Beijing Institute of Technology (No. 2019CX01021), and BIT Teli Young Fellow.

Conflict of Interest

The authors declare no conflict of interests.

References

[1] C. Li, M.M. Islam, J. Moore, J. Sleppy, C. Morrison, K. Konstantinov, S.X. Dou, C. Renduchintala, and J. Thomas, Wearable energy-smart ribbons for synchronous energy harvest and storage, *Nat. Commun.*, 7(2016), art. No. 13319.

[2] A.E. Ostfeld, A.M. Gaikwad, Y. Khan, and A.C. Arias, High-performance flexible energy storage and harvesting system for wearable electronics, *Sci. Rep.*, 6(2016), art. No. 26122.

[3] D. Chen and Q.B. Pei, Electronic muscles and skins: A review of soft sensors and actuators, *Chem. Rev.*, 117(2017), No. 17, p. 11239.

[4] H. Nishide and K. Oyaizu, Toward flexible batteries, *Science*, 319(2008), No. 5864, p. 737.

[5] Y.F. Zhao and J.C. Guo, Development of flexible Li-ion batteries for flexible electronics, *InfoMat*, 2(2020), No. 5, p. 866.

[6] L.F. Wang, M.M. Geng, X.N. Ding, C. Fang, Y. Zhang, S.S. Shi, Y. Zheng, K. Yang, C. Zhan, and X.D. Wang, Research progress of the electrochemical impedance technique applied to the high-capacity lithium-ion battery, *Int. J. Miner. Metall. Mater.*, 28(2021), No. 4, p. 538.

[7] M. Li, J. Lu, Z.W. Chen, and K. Amine, 30 years of lithium-ion batteries, *Adv. Mater.*, 30(2018), No. 33, art. No. 1800561.

[8] T. Tao, S.G. Lu, and Y. Chen, A review of advanced flexible lithium-ion batteries, *Adv. Mater. Technol.*, 3(2018), No. 9, art. No. 1700375.

[9] Z.H. Fang, J. Wang, H.C. Wu, Q.Q. Li, S.S. Fan, and J.P. Wang, Progress and challenges of flexible lithium ion batteries, *J. Power Sources*, 454(2020), art. No. 227932.

[10] E. Foreman, W. Zakri, M.H. Sanatimoghaddam, A. Modjtahedi, S. Pathak, A.G. Kashkooli, N.G. Garafolo, and S. Farhad, A review of inactive materials and components of flexible lithium-ion batteries, *Adv. Sustainable Syst.*, 1(2017), No. 11, art. No. 1700061.

[11] G.M. Zhou, F. Li, and H.M. Cheng, Progress in flexible lithium batteries and future prospects, *Energy Environ. Sci.*, 7(2014), No. 4, p. 1307.

[12] Y.H. Hu and X.L. Sun, Flexible rechargeable lithium ion batteries: Advances and challenges in materials and process technologies, *J. Mater. Chem. A*, 2(2014), No. 28, p. 10712.

[13] J. Chang, Q.Y. Huang, Y. Gao, and Z.J. Zheng, Pathways of developing high-energy-density flexible lithium batteries, *Adv. Mater.*, 33(2021), No. 46, art. No. 2170363.

[14] C.Y. Wang and G.G. Wallace, Flexible electrodes and electrolytes for energy storage, *Electrochim. Acta*, 175(2015), p. 87.

[15] Y. Li, R.H. Wang, Z.N. Guo, Z. Xiao, H.D. Wang, X.L. Luo, and H. Zhang, Emerging two-dimensional noncarbon nanomaterials for flexible lithium-ion batteries: Opportunities and challenges, *J. Mater. Chem. A*, 7(2019), No. 44, p. 25227.

[16] B. Liu, J.G. Zhang, and G.Z. Shen, Pursuing two-dimensional nanomaterials for flexible lithium-ion batteries, *Nano Today*, 11(2016), No. 1, p. 82.

[17] O. Nyamaa, D.H. Seo, J.S. Lee, H.M. Jeong, S.C. Huh, J.H. Yang, E. Dolgor, and J.P. Noh, High electrochemical performance silicon thin-film free-standing electrodes based on buckypaper for flexible lithium-ion batteries, *Materials* (*Basel*), 14(2021), No. 8, art. No. 2053.

[18] Z. Gao, N.N. Song, Y.Y. Zhang, and X.D. Li, Cotton-textile-enabled, flexible lithium-ion batteries with enhanced capacity and extended lifespan, *Nano Lett.*, 15(2015), No. 12, p. 8194.

[19] B. Liu, J. Zhang, X.F. Wang, G. Chen, D. Chen, C.W. Zhou, and G.Z. Shen, Hierarchical three-dimensional $ZnCo_2O_4$ nanowire arrays/carbon cloth anodes for a novel class of high-performance flexible lithium-ion batteries, *Nano Lett.*, 12(2012), No. 6, p. 3005.

[20] J. Chen, L. Wen, R.P. Fang, D.W. Wang, H.M. Cheng, and F.

Li, Stress release in high-capacity flexible lithium-ion batteries through nested wrinkle texturing of graphene, *J. Energy Chem.*, 61(2021), p. 243.

[21] K. Rana, J. Singh, J.T. Lee, J.H. Park, and J.H. Ahn, Highly conductive freestanding graphene films as anode current collectors for flexible lithium-ion batteries, *ACS Appl. Mater. Interfaces*, 6(2014), No. 14, p. 11158.

[22] R.W. Mo, D. Rooney, K.N. Sun, and H.Y. Yang, 3D nitrogen-doped graphene foam with encapsulated germanium/nitrogen-doped graphene yolk-shell nanoarchitecture for high-performance flexible Li-ion battery, *Nat. Commun.*, 8(2017), art. No. 13949.

[23] X. Fang, C.F. Shen, M.Y. Ge, J.P. Rong, Y.H. Liu, A.Y. Zhang, F. Wei, and C.W. Zhou, High-power lithium ion batteries based on flexible and light-weight cathode of $LiNi_{0.5}Mn_{1.5}O_4$/carbon nanotube film, *Nano Energy*, 12(2015), p. 43.

[24] C.Z. Meng, C.H. Liu, and S.S. Fan, Flexible carbon nanotube/polyaniline paper-like films and their enhanced electrochemical properties, *Electrochem. Commun.*, 11(2009), No. 1, p. 186.

[25] X.L. Jia, C.Z. Yan, Z. Chen, R.R. Wang, Q. Zhang, L. Guo, F. Wei, and Y.F. Lu, Direct growth of flexible $LiMn_2O_4$/CNT lithium-ion cathodes, *Chem. Commun.*, 47(2011), No. 34, p. 9669.

[26] K. Amin, Q.H. Meng, A. Ahmad, M. Cheng, M. Zhang, L.J. Mao, K. Lu, and Z.X. Wei, A carbonyl compound-based flexible cathode with superior rate performance and cyclic stability for flexible lithium-ion batteries, *Adv. Mater.*, 30(2018), No. 4, art. No. 1703868.

[27] J.Y. Wan, J. Xie, X. Kong, Z. Liu, K. Liu, F.F. Shi, A. Pei, H. Chen, W. Chen, J. Chen, X.K. Zhang, L.Q. Zong, J.Y. Wang, L.Q. Chen, J. Qin, and Y. Cui, Ultrathin, flexible, solid polymer composite electrolyte enabled with aligned nanoporous host for lithium batteries, *Nat. Nanotechnol.*, 14(2019), No. 7, p. 705.

[28] K.H. Choi, S.J. Cho, S.H. Kim, Y.H. Kwon, J.Y. Kim, and S.Y. Lee, Thin, deformable, and safety-reinforced plastic crystal polymer electrolytes for high-performance flexible lithium-ion batteries, *Adv. Funct. Mater.*, 24(2014), No. 1, p. 44.

[29] W. Liu, J. Chen, Z. Chen, K. Liu, G.M. Zhou, Y.M. Sun, M.S. Song, Z.N. Bao, and Y. Cui, Stretchable lithium-ion batteries enabled by device-scaled wavy structure and elastic-sticky separator, *Adv. Energy Mater.*, 7(2017), No. 21, art. No. 1701076.

[30] X.D. Wang, Y. Lu, D.S. Geng, L. Li, D. Zhou, H.Y. Ye, Y.C. Zhu, and R.M. Wang, Planar fully stretchable lithium-ion batteries based on a lamellar conductive elastomer, *ACS Appl. Mater. Interfaces*, 12(2020), No. 48, p. 53774.

[31] S.H. Kim, K.H. Choi, S.J. Cho, E.H. Kil, and S.Y. Lee, Mechanically compliant and lithium dendrite growth-suppressing composite polymer electrolytes for flexible lithium-ion batteries, *J. Mater. Chem. A*, 1(2013), No. 16, p. 4949.

[32] C. Wang, R.J. Li, P. Chen, Y.S. Fu, X.Y. Ma, T. Shen, B.J. Zhou, K. Chen, J.J. Fu, X.F. Bao, W.W. Yan, and Y. Yang, Highly stretchable, non-flammable and notch-insensitive intrinsic self-healing solid-state polymer electrolyte for stable and safe flexible lithium batteries, *J. Mater. Chem. A*, 9(2021), No. 8, p. 4758.

[33] H. Yim, S.H. Yu, S.H. Baek, Y.E. Sung, and J.W. Choi, Directly integrated all-solid-state flexible lithium batteries on polymer substrate, *J. Power Sources*, 455(2020), art. No. 227978.

[34] J.Y. Rao, N.S. Liu, Z. Zhang, J. Su, L.Y. Li, L. Xiong, and Y.H. Gao, All-fiber-based quasi-solid-state lithium-ion battery towards wearable electronic devices with outstanding flexibility and self-healing ability, *Nano Energy*, 51(2018), p. 425.

[35] G.Y. Qian, X.B. Liao, Y.X. Zhu, F. Pan, X. Chen, and Y. Yang, Designing flexible lithium-ion batteries by structural engineering, *ACS Energy Lett.*, 4(2019), No. 3, p. 690.

[36] M. Koo, K.I. Park, S.H. Lee, M. Suh, D.Y. Jeon, J.W. Choi, K. Kang, and K.J. Lee, Bendable inorganic thin-film battery for fully flexible electronic systems, *Nano Lett.*, 12(2012), No. 9, p. 4810.

[37] Y.H. Kwon, S.W. Woo, H.R. Jung, H.K. Yu, K. Kim, B.H. Oh, S. Ahn, S.Y. Lee, S.W. Song, J. Cho, H.C. Shin, and J.Y. Kim, Cable-type flexible lithium ion battery based on hollow multi-helix electrodes, *Adv. Mater.*, 24(2012), No. 38, p. 5192.

[38] S.Y. Lee, K.H. Choi, W.S. Choi, Y.H. Kwon, H.R. Jung, H.C. Shin, and J.Y. Kim, Progress in flexible energy storage and conversion systems, with a focus on cable-type lithium-ion batteries, *Energy Environ. Sci.*, 6(2013), No. 8, p. 2414.

[39] Y.N. Xu, K. Wang, J.W. Han, C. Liu, Y.B. An, Q.H. Meng, C. Li, X. Zhang, X.Z. Sun, Y.S. Zhang, L.J. Mao, Z.X. Wei, and Y.W. Ma, Scalable production of wearable solid-state Li-ion capacitors from N-doped hierarchical carbon, *Adv. Mater.*, 32(2020), No. 45, art. No. 2005531.

[40] Z.M. Song, X. Wang, C. Lv, Y.H. An, M.B. Liang, T. Ma, D. He, Y.J. Zheng, S.Q. Huang, H.Y. Yu, and H.Q. Jiang, Kirigami-based stretchable lithium-ion batteries, *Sci. Rep.*, 5(2015), art. No. 10988.

[41] Y.H. Bao, G.Q. Hong, Y. Chen, J. Chen, H.S. Chen, W.L. Song, and D.N. Fang, Customized kirigami electrodes for flexible and deformable lithium-ion batteries, *ACS Appl. Mater. Interfaces*, 12(2020), No. 1, p. 780.

[42] M. Park, H. Cha, Y. Lee, J. Hong, S.Y. Kim, and J. Cho, Post-patterned electrodes for flexible node-type lithium-ion batteries, *Adv. Mater.*, 29(2017), No. 11, art. No. 1605773.

[43] F.W. Xiang, F. Cheng, Y.J. Sun, X.P. Yang, W. Lu, R. Amal, and L.M. Dai, Recent advances in flexible batteries: From materials to applications, *Nano Res.*, 2021. DOI: 10.1007/s12274-021-3820-2.

[44] L.J. Mao, Q.H. Meng, A. Ahmad, and Z.X. Wei, Mechanical analyses and structural design requirements for flexible energy storage devices, *Adv. Energy Mater.*, 7(2017), No. 23, art. No. 1700535.

[45] D. Chen, Z. Lou, K. Jiang, and G.Z. Shen, Device configurations and future prospects of flexible/stretchable lithium-ion batteries, *Adv. Funct. Mater.*, 28(2018), No. 51, art. No. 1805596.

[46] H. Jeon, I. Cho, H. Jo, K. Kim, M.H. Ryou, and Y.M. Lee, Highly rough copper current collector: Improving adhesion property between a silicon electrode and current collector for flexible lithium-ion batteries, *RSC Adv.*, 7(2017), No. 57, p. 35681.

[47] Z.A. Zhang, Q. Li, K. Zhang, W. Chen, Y.Q. Lai, and J. Li, Titanium-dioxide-grafted carbon paper with immobilized sulfur as a flexible free-standing cathode for superior lithium–sulfur batteries, *J. Power Sources*, 290(2015), p. 159.

[48] S.W. Kim and K.Y. Cho, Current collectors for flexible lithium ion batteries: A review of materials, *J. Electrochem. Sci. Technol*, 6(2015), No. 1, p. 1.

[49] Y.F. Zhang, F.Z. Li, K. Yang, X. Liu, Y.G. Chen, Z.Q. Lao, K.C. Mai, and Z.S. Zhang, Polymer molecular engineering enables rapid electron/ion transport in ultra-thick electrode for high-energy-density flexible lithium-ion battery, *Adv. Funct. Mater.*, 31(2021), No. 19, art. No. 2100434.

[50] H.M. Shi, G.L. Wen, Y. Nie, G.H. Zhang, and H.G. Duan, Flexible 3D carbon cloth as a high-performing electrode for energy storage and conversion, *Nanoscale*, 12(2020), No. 9, p. 5261.

[51] L. Hu, H. Wu, F.L. Mantia, Y. Yang, and Y. Cui, Thin, flexible secondary Li-ion paper batteries, *ACS Nano*, 4(2010), No. 10, p. 5843.

[52] Y. Shi, L. Wen, G.M. Zhou, J. Chen, S.F. Pei, K. Huang, H.M. Cheng, and F. Li, Graphene-based integrated electrodes for flexible lithium ion batteries, *2D Mater.*, 2(2015), No. 2, art. No. 024004.

[53] Y.H. Bao, Y. Liu, Y.D. Kuang, D.N. Fang, and T. Li, 3D-prin-

ted highly deformable electrodes for flexible lithium ion batteries, *Energy Storage Mater.*, 33(2020), p. 55.
[54] M.H. Park, M. Noh, S. Lee, M. Ko, S. Chae, S. Sim, S. Choi, H. Kim, H. Nam, S. Park, and J. Cho, Flexible high-energy Li-ion batteries with fast-charging capability, *Nano Lett.*, 14(2014), No. 7, p. 4083.
[55] G.P. Fu, M.D. Soucek, and T. Kyu, Fully flexible lithium ion battery based on a flame retardant, solid-state polymer electrolyte membrane, *Solid State Ionics*, 320(2018), p. 310.
[56] S. Xu, Y.H. Zhang, J. Cho, J. Lee, X. Huang, L. Jia, J.A. Fan, Y.W. Su, J. Su, H.G. Zhang, H.Y. Cheng, B.W. Lu, C.J. Yu, C. Chuang, T.I. Kim, T. Song, K. Shigeta, S. Kang, C. Dagdeviren, I. Petrov, P.V. Braun, Y.G. Huang, U. Paik, and J.A. Rogers, Stretchable batteries with self-similar serpentine interconnects and integrated wireless recharging systems, *Nat. Commun.*, 4(2013), art. No. 1543.
[57] H. An, J. Mike, K.A. Smith, L. Swank, Y.H. Lin, S.L. Pesek, R. Verduzco, and J.L. Lutkenhaus, Highly flexible self-assembled V_2O_5 cathodes enabled by conducting diblock copolymers, *Sci. Rep.*, 5(2015), art. No. 14166.
[58] D.H. Kim, J.H. Ahn, W.M. Choi, H.S. Kim, T.H. Kim, J.Z. Song, Y.Y. Huang, Z.J. Liu, C. Lu, and J.A. Rogers, Stretchable and foldable silicon integrated circuits, *Science*, 320(2008), No. 5875, p. 507.
[59] H.L. Luo, J.E. Zhu, E. Sahraei, and Y. Xia, Adhesion strength of the cathode in lithium-ion batteries under combined tension/shear loadings, *RSC Adv.*, 8(2018), No. 8, p. 3996.
[60] A.J. Blake, R.R. Kohlmeyer, L.F. Drummy, J.S. Gutiérrez-Kolar, J. Carpena-Núñez, B. Maruyama, R. Shahbazian-Yassar, H. Huang, and M.F. Durstock, Creasable batteries: Understanding failure modes through dynamic electrochemical mechanical testing, *ACS Appl. Mater. Interfaces*, 8(2016), No. 8, p. 5196.
[61] H. Cha, Y. Lee, J. Kim, M. Park, and J. Cho, Flexible 3D interlocking lithium-ion batteries, *Adv. Energy Mater.*, 8(2018), No. 30, art. No. 1801917.
[62] J.Q. He, C.H. Lu, H.B. Jiang, F. Han, X. Shi, J.X. Wu, L.Y. Wang, T.Q. Chen, J.J. Wang, Y. Zhang, H. Yang, G.Q. Zhang, X.M. Sun, B.J. Wang, P.N. Chen, Y.G. Wang, Y.Y. Xia, and H.S. Peng, Scalable production of high-performing woven lithium-ion fibre batteries, *Nature*, 597(2021), No. 7874, p. 57.
[63] C.M. Shi, T.Y. Wang, X.B. Liao, B.Y. Qie, P.F. Yang, M.J. Chen, X. Wang, A. Srinivasan, Q. Cheng, Q. Ye, A.C. Li, X. Chen, and Y. Yang, Accordion-like stretchable Li-ion batteries with high energy density, *Energy Storage Mater.*, 17(2019), p. 136.
[64] W. Weng, Q. Sun, Y. Zhang, S.S. He, Q.Q. Wu, J. Deng, X. Fang, G.Z. Guan, J. Ren, and H.S. Peng, A gum-like lithium-ion battery based on a novel arched structure, *Adv. Mater.*, 27(2015), No. 8, p. 1363.
[65] Z.M. Song, T. Ma, R. Tang, Q. Cheng, X. Wang, D. Krishnaraju, R. Panat, C.K. Chan, H.Y. Yu, and H.Q. Jiang, Origami lithium-ion batteries, *Nat. Commun.*, 5(2014), art. No. 3140.
[66] F.N. Mo, G.J. Liang, Z.D. Huang, H.F. Li, D.H. Wang, and C.Y. Zhi, An overview of fiber-shaped batteries with a focus on multifunctionality, scalability, and technical difficulties, *Adv. Mater.*, 32(2020), No. 5, art. No. 1902151.
[67] Y.S. Chen, K.H. Chang, C.C. Hu, and T.T. Cheng, Performance comparisons and resistance modeling for multi-segment electrode designs of power-oriented lithium-ion batteries, *Electrochim. Acta*, 55(2010), No. 22, p. 6433.
[68] Y. Zhang, Y.H. Wang, L. Wang, C.M. Lo, Y. Zhao, Y.D. Jiao, G.F. Zheng, and H.S. Peng, A fiber-shaped aqueous lithium ion battery with high power density, *J. Mater. Chem. A*, 4(2016), No. 23, p. 9002.
[69] J. Ren, Y. Zhang, W.Y. Bai, X.L. Chen, Z.T. Zhang, X. Fang, W. Weng, Y.G. Wang, and H.S. Peng, Elastic and wearable wire-shaped lithium-ion battery with high electrochemical performance, *Angew. Chem. Int. Ed.*, 53(2014), No. 30, p. 7864.
[70] G.Y. Qian, B. Zhu, X.B. Liao, H.W. Zhai, A. Srinivasan, N.J. Fritz, Q. Cheng, M.Q. Ning, B.Y. Qie, Y. Li, S.L. Yuan, J. Zhu, X. Chen, and Y. Yang, Bioinspired, spine-like, flexible, rechargeable lithium-ion batteries with high energy density, *Adv. Mater.*, 30(2018), No. 12, art. No. 1704947.
[71] X.B. Liao, C.M. Shi, T.Y. Wang, B.Y. Qie, Y.L. Chen, P.F. Yang, Q. Cheng, H.W. Zhai, M.J. Chen, X. Wang, X. Chen, and Y. Yang, High-energy-density foldable battery enabled by zigzag-like design, *Adv. Energy Mater.*, 9(2019), No. 4, art. No. 1802998.
[72] N. Li, H.S. Chen, S.Q. Yang, H. Yang, S.Q. Jiao, and W.L. Song, Bidirectional planar flexible snake-origami batteries, *Adv. Sci.*, 8(2021), No. 20, art. No. 2101372.
[73] C.J. Xu, L. Weng, L. Ji, and J.Q. Zhou, An analytical model for the fracture behavior of the flexible lithium-ion batteries under bending deformation, *Eur. J. Mech. A/Solids*, 73(2019), p. 47.
[74] L.B. Jiang, J.J. Zhao, and Y.W. Gao, Mechanical analysis of a flexible cable battery using the finite element model, *AIP Adv.*, 9(2019), No. 1, art. No. 015013.
[75] C.J. Xu, L. Weng, B.B. Chen, L. Ji, J.Q. Zhou, R. Cai, and S.L. Lu, Modeling of the ratcheting behavior in flexible electrodes during cyclic deformation, *J. Power Sources*, 446(2020), art. No. 227353.
[76] A. Chen, X. Guo, S. Yang, G.J. Liang, Q. Li, Z. Chen, Z.D. Huang, Q. Yang, C.P. Han, and C.Y. Zhi, Human joint-inspired structural design for a bendable/foldable/stretchable/twistable battery: Achieving multiple deformabilities, *Energy Environ. Sci.*, 14(2021), No. 6, p. 3599.
[77] D.P. Qi, Z.Y. Liu, Y. Liu, W.R. Leow, B.W. Zhu, H. Yang, J.C. Yu, W. Wang, H. Wang, S.Y. Yin, and X.D. Chen, Suspended wavy graphene microribbons for highly stretchable microsupercapacitors, *Adv. Mater.*, 27(2015), No. 37, p. 5559.
[78] H.F. Li, Z.J. Tang, Z.X. Liu, and C.Y. Zhi, Evaluating flexibility and wearability of flexible energy storage devices, *Joule*, 3(2019), No. 3, p. 613.
[79] J.G. Tu, W.L. Song, H.P. Lei, Z.J. Yu, L.L. Chen, M.Y. Wang, and S.Q. Jiao, Nonaqueous rechargeable aluminum batteries: Progresses, challenges, and perspectives, *Chem. Rev.*, 121(2021), No. 8, p. 4903.