

Flexible Batteries: From Mechanics to Devices

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ABSTRACT: With the rapid development of wearable electronics, it is desirable to design and develop flexible power supplies, especially rechargeable lithium ion batteries, with high performance and superior flexibility and durability for integration into electronics. Structures and materials are two key factors in achieving the flexibility of batteries. Therefore, it becomes important to understand the fundamental mechanics in order to better guide us in the design of flexible batteries to meet the requirements of wearable electronics. In this work, we review recent research progress on batteries for wearable electronics based on structures and materials, covering the fundamental mechanics underlying the structural design mechanism and intrinsically deformable materials as building blocks for flexible batteries. In addition, we review and discuss emerging new materials and structures that could potentially revolutionize the design concepts of flexible batteries for wearable electronics. Therefore, these flexible and wearable materials and structures are not limited to batteries. We can envision that the mechanics and design concepts can also be extended to other electronics for wearable applications.



Interest in wearable electronics has surged in the past several years. With the rapid development of wearable electronics, it is desirable to design and develop wearable power supplies for integration into the electronics. Rechargeable lithium ion batteries, as a relatively mature electrochemical energy storage technology, have been widely adopted by portable electronics as a power supply because of their high power, high energy density, and lack of memory effect. However, conventional lithium ion batteries are rigid and fragile and thus cannot meet the requirements of wearable electronics. Therefore, it is important to fabricate wearable batteries with superior flexibility and durability for wearable electronics.

Structures and materials are two key factors in achieving the flexibility of batteries. Stiff and brittle materials that are often used in conventional batteries can be made deformable via suitable structural design. Alternatively, batteries can also be made flexible at the component level by using intrinsically deformable materials. In this work, we review recent research progress on flexible batteries for wearable electronics, which can be categorized into two design approaches: structural design and material design. For the structural design approach, we first review the fundamental mechanics underlying the structural design mechanism to make stiff and brittle building blocks of batteries compliant and deformable and then review recent developments of flexible batteries based on such a structural design mechanism, including stretchable batteries and foldable batteries. For the material design approach, we review recent progress in developing intrinsically deformable building blocks for flexible batteries, including carbon nanotube (CNT),

graphene, and carbon nanofiber (CNF). Finally, we review emerging new materials and structures that could potentially revolutionize the design concepts of flexible batteries for wearable electronics, including paper and textile batteries, ultrathick flexible batteries, and one-dimensional (1D) fiber batteries. In the final section, conclusions and outlook for future development of flexible batteries are provided.

Fundamental Mechanics Underlying the Structural Design Mechanisms. In its early years, the inception of flexible electronics relied on structural design to achieve the deformability of intrinsically stiff and brittle electronic materials so that the resulting electronic devices can sustain repeated bending, twisting, and stretching. Typical structural design mechanisms include neutral plane design, serpentine patterning, and island architecture. In this section, we review the fundamental mechanics underlying these three structural design mechanisms.

When a film–substrate bilayer or a multilayer is bent against a cylindrical surface, shown in Figure 1a, the side facing outward is under tension and the other side facing inward is under compression. Therefore, there exists a plane in between where strain vanishes. Such a plane is referred to as the “neutral plane” when the structure is subject to bending. The strain in the film–substrate bilayer is linearly proportional to the

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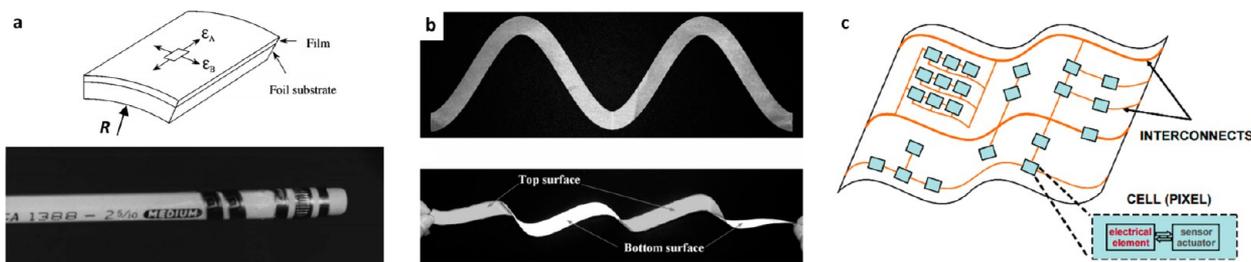


Figure 1. Three elementary structural design strategies to make electronics device deformable. (a) Achieving bendability by placing brittle functional layer on neutral plane. Top panel: Thin film coated to a substrate and bent against a cylindrical surface. Bottom panel: Two transistors bonded to a 25 μm thick steel substrate and wrapped around a pencil. (Reprinted with permission from ref 1. Copyright 1999 American Institute of Physics.) (b) Attaining stretch capability by patterning structure into serpentine design. A paper ribbon cut into serpentine shape (top panel). When pulled at two ends, the serpentine shape accommodates the elongation by deflecting out of the plane. (Reprinted with permission from ref 4. Copyright 2005 Materials Research Society.) (c) Island architecture. Rigid subcircuit islands which are mechanically isolated but electrically linked are distributed systematically on a compliant elastomer substrate. (Reprinted with permission from ref 11. Copyright 2004 Elsevier.)

distance from the neutral plane, and consequently, the maximum tensile strain in the film occurs at the top surface¹

$$\epsilon_{\text{top}} = \left(\frac{t_f + t_s}{2R} \right) \frac{(1 + 2\eta + \chi\eta^2)}{(1 + \eta)(1 + \chi\eta)} \quad (1)$$

where t is the thickness of film or substrate (as denoted by the subscripts, f for film and s for substrate), and R is the bending radius; $\eta = t_f = t_s$; and χ represents the ratio of elastic moduli, Y_f/Y_s . If the substrate is made of material much more compliant than the film, $Y_f/Y_s \gg 1$, then the neutral plane shifts toward the stiff film side and as a result reduces the strain in the top surface. Therefore, so as to diminish strain, in the design of a multilayer-structured bendable electronics device, for example, thin-film-on-polymer, it is always favorable to place the brittle thin film along or at least near the neutral plane of the multilayer. For example, in Figure 1b two transistors are bonded to a 25 μm thick steel substrate and subsequently wrapped around a pencil, and the transistors maintain their functionalities. Another effective way of achieving such a goal is to sandwich the brittle thin film between compliant encapsulating layer. When the material stiffness and thickness are carefully selected, the film can be located close to the neutral plane. As for a stacked structure composed of multiple layers, provided the mechanical property (plane-strain Young's modulus, \bar{E}) and thickness t of each layer, the position of the neutral plane can be predicted by the following equation:²

$$b = \frac{\sum_{i=1}^N \bar{E}_i t_i [(\sum_{j=1}^i t_j) - t_i/2]}{\sum_{i=1}^N \bar{E}_i t_i} \quad (2)$$

where b is the distance from neutral plane to the top surface, \bar{E}_i and t_i denote the plane-strain Young's modulus and thickness of the i th layer, respectively, with $i = 1$ corresponding to the top layer. The foregoing formula predicts the position of a single neutral plane. However, it is applicable only if the adjacent layers possess comparable stiffness. When stiffness alternation along the thickness direction is introduced, for example, bonding the stiffer layers with highly compliant ones, the sole neutral plane splits into several planes.³ The split of neutral planes greatly expands the dimension in which brittle functional electronic thin films can be used in flexible devices.

While neutral plane design is applicable to electronic devices that are mainly subject to bending deformation, such a design concept cannot mitigate the strain in flexible devices subject to

in-plane stretch. Most widely used inorganic electronic materials are stiff and fracture at small tensile strains (a few percent). To this end, it has been demonstrated that a thin film of stiff and brittle material can be made compliant and deformable by suitable patterning in the plane of the thin film. To illustrate this concept, a piece of paper is cut into a serpentine-shaped ribbon and pulled apart at two ends, as shown in Figure 1b. The serpentine paper ribbon, initially planar, twists and bends out of the plane to accommodate the elongation. Consequently, the resulting strain in the paper ribbon is small. By patterning a planar structure into a serpentine shape, its stretch capability can be elevated to up to tens of percent with otherwise much less deformable materials. Factors that influence the stretch capability of device structures patterned into a serpentine include the stiffness of the underlying substrate, as well as the serpentine geometry. A stiffer substrate poses more mechanical constraint to the out-of-plane deflection and twisting of the serpentine when it is stretched, therefore leading to a higher resulting strain in the device structure. Accordingly, a compliant substrate is preferred to achieve better device stretch capability.⁴ The dependence of device stretch capability on serpentine geometry has also been systematically studied and can serve as the guideline of structural design to achieve a certain result.^{5–7} The serpentine patterning concept has inspired a variety of structural designs to achieve large stretch capability of flexible and stretchable devices; these approaches are reviewed in the following sections.

Island architecture is an approach to attaining stretch capability by patterning rigid island array onto a compliant substrate,^{8–11} as sketched in Figure 1c.¹¹ The rigid islands, on which functional circuit components (transistors, resistors, capacitors, etc.) are fabricated, are electrically connected through stretchable metallic traces, such as microstructured gold film or serpentine interconnect, which are in turn supported by a compliant substrate. When the whole structure is stretched, most of the deformation is carried by the compliant substrate but the rigid islands experience negligible strain. However, the huge strain localization near the island–substrate juncture can lead to cracking of the metal interconnect traces, a key challenge to the island architecture concept. To avoid the interconnect cracking, it is suggested to use a much more compliant substrate and to keep the size of the device islands small (e.g., less than hundreds of micro-

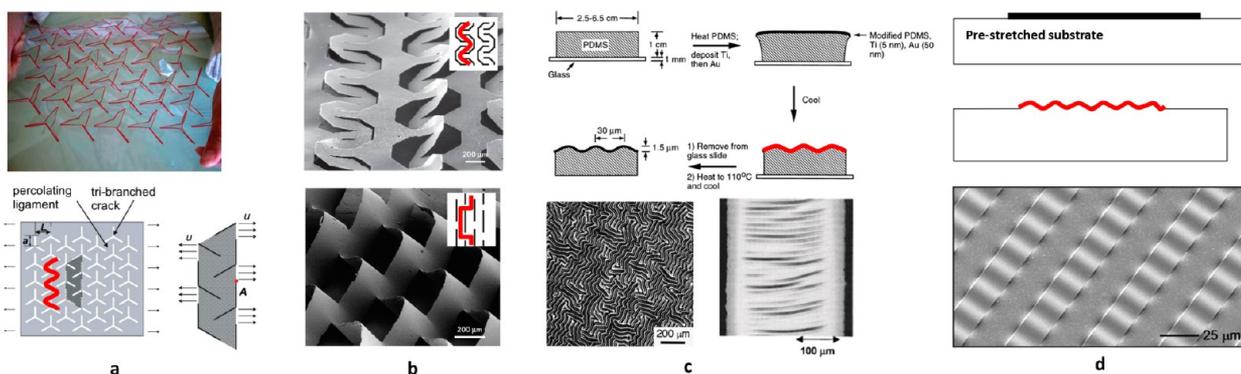


Figure 2. Examples of various approaches to introducing serpentine-like patterns to otherwise undeformable structures, by patterning line defects on a plane (a, b) and by forming superficial waves (c, d). (a) Mechanism of reversible stretch capability of thin microcracked Au film on elastomeric substrate. Top panel: Array of tribranched seams cut on a transparency makes the sheet stretchable. Under stretching, the applied displacement is conformed by the out-of-plane motion of the flaps at the cutting edges. Bottom panel: Schematic of tribranched microcracks generated in deposition processes of Au on polydimethylsiloxane (PDMS) substrate. These tribranched microcracks carve serpentine-shaped ligaments (marked by red line in bottom panel) out of the Au film. (Reprinted with permission from ref 12. Copyright 2006 American Institute of Physics.) (b) Line defect patterns created by oxygen-plasma etching on graphene oxide–poly(vinyl alcohol) (GO-PVA) nanocomposite thin films. When stretch is applied, the ligaments extend the structure through out-of-plane buckling. Inset: cut patterns and serpentine ligaments marked in red. (Reprinted with permission from ref 13. Copyright 2015 Nature Publishing Group.) (c) Top panel: fabrication sequence of surface wave by deposition of metal on elastomer. Marked by red line is the cross-sectional view of the surface wave geometry. (Reprinted with permission from ref 14. Copyright 1998 Nature Publishing Group.) Bottom panel: randomly patterned wavy structure and a stripe of wavy gold film evaporated through a shadow mask. (Reprinted with permission from ref 18. Copyright 2003 American Institute of Physics.) (d) Sketch of depositing thin film on prestretched substrate (top panel) (Reprinted with permission from ref 11. Copyright 2004 Elsevier.) and SEM image of wavy Si ribbons on PDMS substrate (bottom panel). (Reprinted with permission from ref 20. Copyright 2006 AAAS.) The Si ribbons are bonded to a prestretched PDMS substrate, and the sequential release of the stretch after bonding causes compression in the Si ribbons. The ribbons respond to the compressive stress with a superficial periodic wavy structure.

meters) and the interisland spacing large to mitigate the strain localization.

Deformable Structural Designs. In this section, we review an array of structural designs that enable much enhanced deformability of intrinsically much less deformable materials, based on the serpentine patterning mechanism.

It is found that a thin gold film deposited on polydimethylsiloxane (PDMS) can be made with built-in branched microcracks ($<1 \mu\text{m}$ in branch length).^{11,12} Such microcracked gold films are capable of retaining both electrical conductivity and mechanical reversibility to up to 32% applied strain for more than 100 load–unload cycles, in sharp comparison with the low rupture strain (1–2%) for a continuous thin gold film. It is revealed that such a large and repeatable stretch capability of the thin gold film results from the fact that the branched microcracks can effectively pattern the thin gold film into a network of serpentes. To illustrate such a mechanism, Figure 2a shows a transparency film with periodic tribranched cuts (to mimic the branched microcracks) being stretched. The bottom panel of Figure 2a reveals that the percolating ligaments as demarcated by the tribranched cracks form a wavy serpentine. Therefore, subject to tension, these ligaments deflect and twist out of the plane to accommodate the elongation, while the resulting tensile strain in the ligaments is small. As a result, the transparency film with the cuts (i.e., the microcracked thin gold film) can sustain repeated elongations without mechanical failure.

Recent advances in nanofabrication enable patterning microscale line defects on nanocomposite thin films in a more controllable manner to make the resulting structure deformable.¹³ Figure 2b shows two examples of line defect patterned by oxygen-plasma etching on graphene oxide–poly(vinyl alcohol) (GO-PVA) nanocomposite thin films ($\sim 5 \mu\text{m}$ in thickness). The ligaments carved out by these patterns

are highlighted by red lines in the insets. When an elongation is applied, these wave-shaped ligaments are able to deflect and twist out of the plane, aligning to the direction of the applied load, extending the length of the structure; as a result, the strain level in the structure is greatly reduced. Experiment and finite element method simulation identify three stages in the tensile behavior of sheets patterned with serpentine-shaped ligaments: (1) When the applied strain is less than a few percent, which is the initial elastic regime, the thin film follows the behavior of a pristine sheet without patterned line defects. (2) Upon further stretching, the load surpasses the critical buckling stress of the ligaments and out-of-plane deflection starts to occur. (3) At elevated stretch, the strain exceeds the capability of conforming stretch by rigid body rotation; the structure collapses with ligaments oriented in alignment with the load direction, and further stretching will cause fracture.

The above deformable structures are planar in the undeformed state and contain serpentine patterns in their plane. There also exist other deformable structures with out-of-plane waviness in the undeformed state. Such serpentine waves can flatten when the structure is stretched in the plane. As a result, the tensile strain experienced by the structure is small; thus, the structure becomes deformable. For example, during the deposition process of gold on an elastomeric substrate, the substrate is heated and consequently expands, as shown in the top panel of Figure 2c.¹⁴ Because of thermal mismatch between the substrate and the gold film upon cooling to ambient temperature, the substrate contracts more than the gold film does. Thus, the thin gold film ($\sim 100 \text{ nm}$ thick) is under compression and therefore buckles out of the plane to form a wavy pattern.^{14–18} It has been shown that the stretch capability of such a wavy gold thin film can serve as a deformable conductor with a stretch capability up to 123%.¹⁸ An alternate technique to develop well-aligned wave pattern is to attach a

thin stiff film to a prestretched substrate. Upon releasing the prestretch, the substrate tends to contract and recover its original length. As a result, it poses compression to the thin stiff film; thus, the film buckles out-of-plane into a wavy pattern.^{11,18–21} The critical buckling strain and the postbuckling morphology of the wavy pattern can be predicted by analytical models provided in refs 22 and 23. The desired superficial wave structure can be tuned by the prestretch level.

Many stretchable electronic structures which invoke much similarity to the serpentine structure might also be highly enlightening to the development of wearable batteries. Recent experiments demonstrate that a thin metal film with hollow-out can be coated onto a PDMS substrate.^{24,25} Panels a and b of Figure 3 show a schematic of a side view and a SEM image of

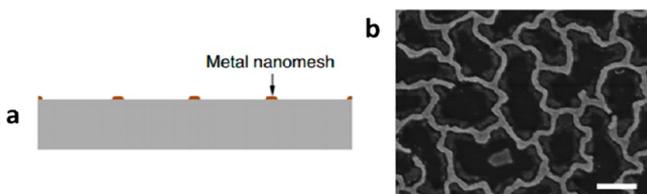


Figure 3. (a) Schematic of metal nanomesh developed on Si mother wafer by photolithography through In_2O_3 grain boundary and (b) SEM image of the as-fabricated metal network (bright image; scale bar, 500 nm). (Reprinted with permission from ref 25. Copyright 2014 Nature Publishing Group.)

the top view, respectively, of gold nanomesh fabricated by grain boundary photolithography. The gold nanomesh sustains

exceptionally high stretch and remains conductive at up to 160% tensile strain. The high deformability is attributed to two factors: (1) The ligaments of Au nanomesh bend and twist out of the plane to accommodate the stretch. (2) the PDMS support stabilizes the fracture in the ligaments. Such a structure can be made useful for augmenting the deformability of thin-film LIB.

Advances of the film–substrate architecture also seek to improve the support conditions provided by the substrate.²⁶ A thin gold film is deposited on polymer foam which has microscopic holes (Figure 4a). As a resultant, the film is brought into partial contact with the substrate in such a way that the film covers the surface cell as a drumhead placed over a drum. When subject to stretching, the cracks and folds are well confined within areas above the surface cells, leaving other regions bonded to the walls crack-free (Figure 4c), in contrast with the better understood homogeneous cracking pattern created over an elongated thin gold film coated onto a plane polymer substrate, as shown in Figure 4b. Figure 4d is an optical image of a strained film–foam system. The cells on the face are visible. Tribranched lines represent the walls enclosing the hollow and further indicate the honeycomb-shaped unit cell of the structure. Such a hexagon unit cell can be divided into four distinctive regions, as illustrated by the inset of Figure 4e. The average strains within the gold film over these four regions all increase with the increasing applied stretch. The minimum value always occurs in the gold film bonded to zone A, while the maximum value occurs in the regions over the cell hollows (zones C and D). When the system is stretched, the two neighboring zone-A wall ligaments rotate toward each other,

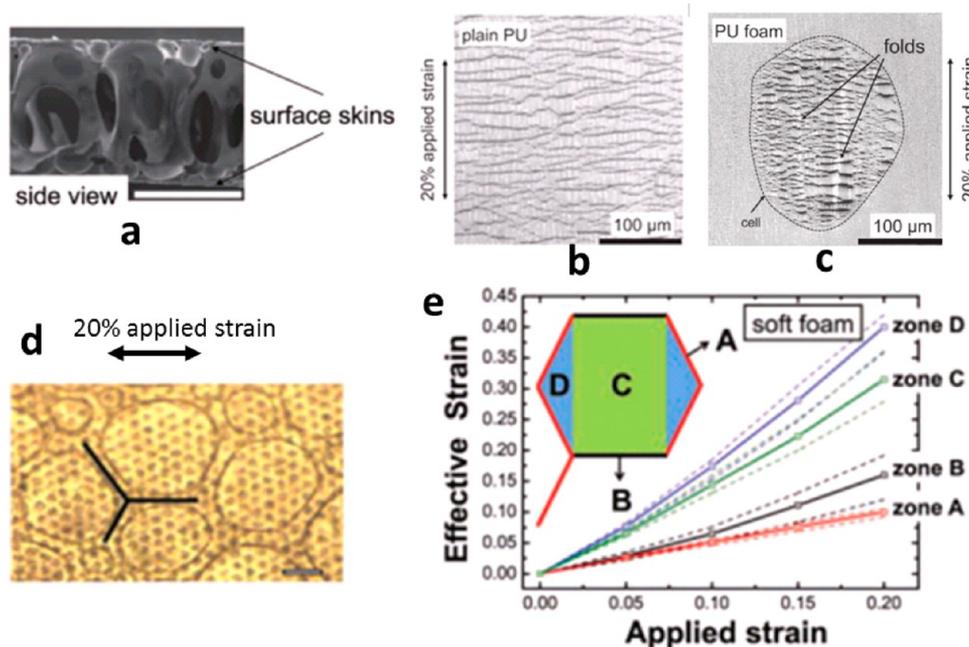


Figure 4. (a) SEM image of cross-sectional view of polyurethane (PU) foam. The foam features a microcellular structure. On both top and bottom surfaces, the cells are covered by a thin PU surface skin layer ($<100\ \mu\text{m}$ thick) which is formed spontaneously in the fabrication process (scale bar, 0.5 mm). (b, c) The homogeneous cracking pattern formed on a film-on-planar-substrate structure is characterized by uniform crack spacing (b), whereas a stretched film-on-foamy-substrate system shows morphology with cracks and folds trapped within the region above the surface cells (c) ($\epsilon_{\text{appl}} = 20\%$; scale bars, $100\ \mu\text{m}$). (d) Optical image under 20% applied strain. Tribranched lines represent a minimal unit of the surrounding walls which enclose the surface cell. Stretch direction is marked with the arrows (scale bar, $100\ \mu\text{m}$). (e) Average strains in the film over four distinctive regions (inset): A, cell walls oriented at an angle of 60° to the direction of the applied stretch; B, wall ligaments along the stretch direction; C, central rectangular region over the cell; and D, the remaining regions over the cell. (Reprinted with permission from ref 26. Copyright 2013 Wiley.)

largely in rigid body motion which accommodates the applied strain, resulting in an elongated hexagon and effectively reducing the film strain within these regions. The motif of releasing strain recurs again in this design.

Flexible Batteries: Structural Design. In this section we review the design of stretchable and wearable batteries based on or inspired by the deformable structural designs as reviewed above.

A recent development of flexible lithium ion battery features in an array of electrode–collector pads electrically connected by serpentine shaped interconnects (Figure 5a,b).²⁷ The metal

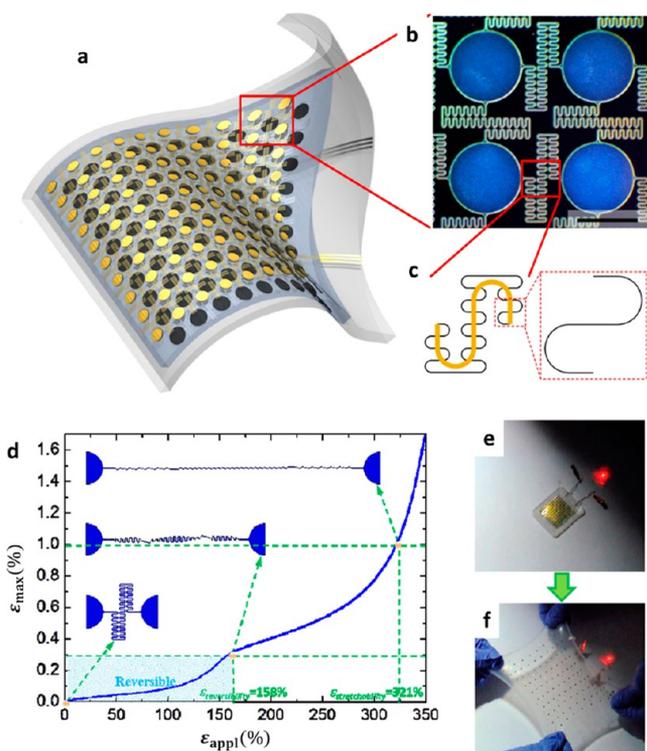


Figure 5. (a) Schematic of flexible LIB assembly under stretch and bending. (b) Optical image of copper electrode current collectors and serpentine interconnects fabricated using photolithography (scale bar, 2 mm). (c) Illustration of “self-similar” serpentine interconnect which has two levels: first level, black line; second level, yellow line. (d) Maximum strain, ϵ_{\max} , in a stretched and buckling out-of-plane serpentine interconnect as a function of applied strain, ϵ_{appl} . The “self-similar” serpentine design substantially reduces the strain to more than 2 orders of magnitude. Insets show three states during stretch: intact pads and interconnects (bottom), the serpentine interconnect stretched to its reversible limit (middle), and stretchable limit (top). (e, f) The stretchable LIB powers an LED at its original state (e) and at a biaxially stretched state (f). (Reprinted with permission from ref 27. Copyright 2013 Nature Publishing Group.)

collector pads and interconnects are coated between two polyimide layers (PI) to form a sandwiched PI(1.2 μm)/metal(0.6 μm)/PI(1.2 μm) structure so that the metal layers are placed at the neutral plane of the multilayer. The anode side and cathode side are separated by deformable gel electrolyte, and the entire architecture is encapsulated in very compliant silicone (Young’s modulus, 60 kPa). Compared to the conventional serpentine structure which has only one level of meandering wire, the design here has an advanced “self-similar” structure composed of two levels, as illustrated in Figure 5c: the

smaller size serpentine (black, first level) forms a larger serpentine (yellow, second level). The hierarchical geometry of serpentine design leads to an ordered unravelling, i.e., when the interconnect is subject to stretch, the first level serpentine expands until it fully collapses along the stretching direction, which is followed by the expansion of the second level serpentine upon further lengthening, as shown in the inset of Figure 5d. The mechanical advantage of this “self-similar” serpentine is that it yields a second opportunity to release the strain by the expansion of the second level, as indicated by the turning point of the ϵ_{\max} – ϵ_{appl} curve around $\epsilon_{\text{appl}} = 160\%$. The reversibility and stretch capability of the two-leveled serpentine are determined by equaling the ϵ_{\max} to the elastic limit and ultimate tensile strain the interconnect metal, in this case $\epsilon_{\text{reversibility}} = 158\%$ and $\epsilon_{\text{stretchability}} = 321\%$. In Figure 5e,f, a red light-emitting diode (LED) is powered by a stretchable battery; upon biaxial stretching of the battery, no visible dimming in the light can be observed. This shows the battery is extraordinarily deformable. This is a classic example of how various building blocks of design elements (neutral plane, serpentine interconnect, and island concept) can be put together to secure the deformability of a battery.

The ancient Japanese arts of origami (namely, paper-folding) and kirigami (paper-folding and -cutting) have also lent inspiration of imparting high linear and areal deformability into structure design.^{13,28} The potential of using these schemes in developing deformable lithium ion batteries has been recently demonstrated.^{29,30} The origami and kirigami lithium ion batteries have the conventional stacked structure of planar battery, developed by coating current collector with active electrode slurry, separating the cathode side from anode side with separator, and assembling in packaging material. The total thickness of the as-fabricated planar battery is $\sim 380 \mu\text{m}$. Then by folding the planar battery along a grid of preplanned creases the plane has been divided into parallelograms, as the schematic illustrates in Figure 6a. When the two parallel sides are pulled together, the battery is folded via the predefined creases and collapses into the corresponding contracted state, strain occurs only at the creases and vertices, however the parallelograms, which undergo rigid body motion about the creases, maintain strain-free. Figure 6b depicts the expanded and the folded state of origami lithium ion battery, and the output voltage exhibits consistency during folding and unfolding. Kirigami includes both folding and cutting into topological design. Figure 6c demonstrates the sequence of the developing a kirigami lithium ion battery. Dragged at two ends, the parallelograms rotate about the hinges to accommodate the stretch. A kirigami battery continuously powers an LED during the deformation without observable dimming of the light. Note that even though by implementing the strategy of origami and kirigami stress vanishes almost on the entire battery, the region at creases and hinges experiences intense stress concentration which originates from the loading cycles of folding/unfolding the battery. The cyclic nature of the loading may wear out the battery material by fatigue mechanisms. Therefore, the success of origami/kirigami batteries in withstanding the loading cycles critically depends on endurance of the material and the quality of interlayer bonding (for example, in origami battery case, CNT coated paper conductor has been employed as tough current collector to sustain fatigue). Promoted by the revolutionary advancement in materials, origami- and kirigami-based techniques provide us with access to an extensive collection of designs which seek to accommodate large

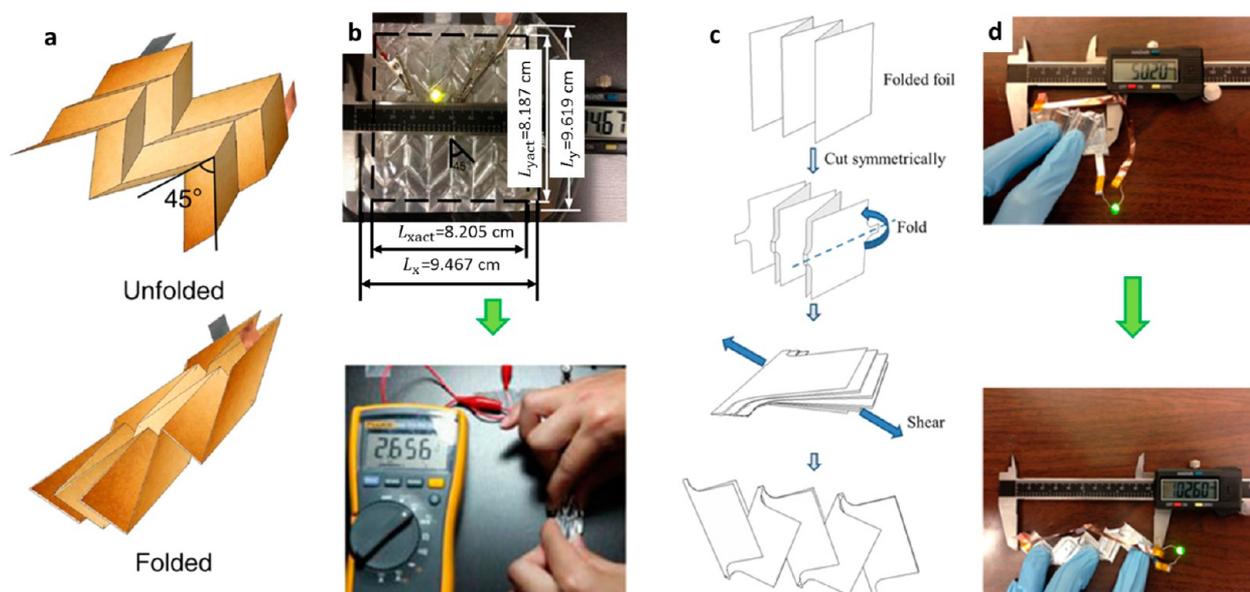


Figure 6. Schematic (a) and experimental image (b) showing an origami lithium ion battery in its unfolded and folded state. (Reprinted with permission from ref 29. Copyright 2014 Nature Publishing Group.) (c) Sequence of folding and cutting a planar battery into design. (d) A kirigami battery continuously powers an LED during the deformation. (Reprinted with permission from ref 30. Copyright 2015 Nature Publishing Group.)

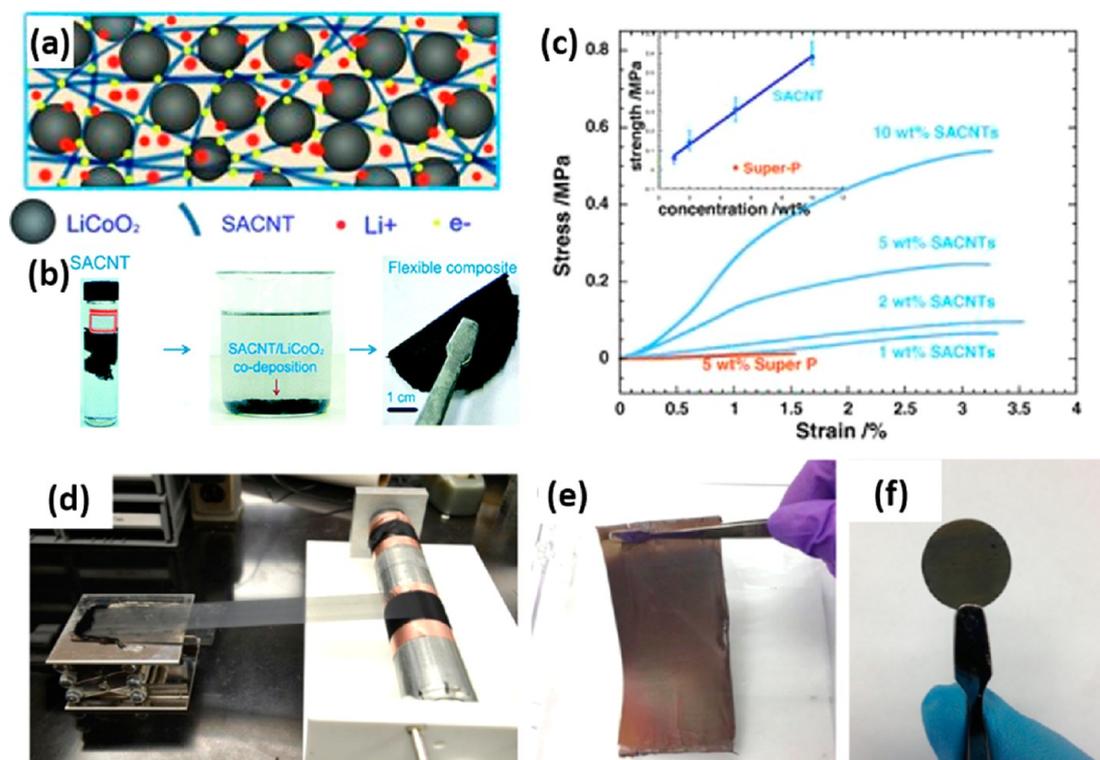


Figure 7. (a) Schematic of the free-standing $\text{LiCoO}_2/\text{CNT}$ cathode. (b) Fabrication process of the electrode by using ultrasonication and codeposition method. (c) Mechanical properties of the electrode. Inset is the relation between electrode strength and CNT content. (d) A wide, continuous aligned CNT sheet was drawn from CNT forests and rolled on a cylinder. (e) Free-standing and binder-free Si-CNT film. (f) Punched electrode for coin cell test. (Reprinted with permission from refs 31 and 44. Copyright 2012 and 2013, respectively, Wiley.)

displacement through decomposition into localized deformation and rigid body motion.

Wearable Batteries: Material Design. In addition to achieving flexibility by those delicate structure designs, selecting materials with intrinsically good flexibility is the other important prerequisite to enable good flexibility. Here, we reviewed two

types of building block materials: one is electrically conductive materials, focusing on flexible carbon materials including carbon nanotube (CNT), graphene, and carbon nanofiber (CNF); the other is ionically conductive materials, focusing on solid-state electrolytes, as important battery components for use in flexible battery design.

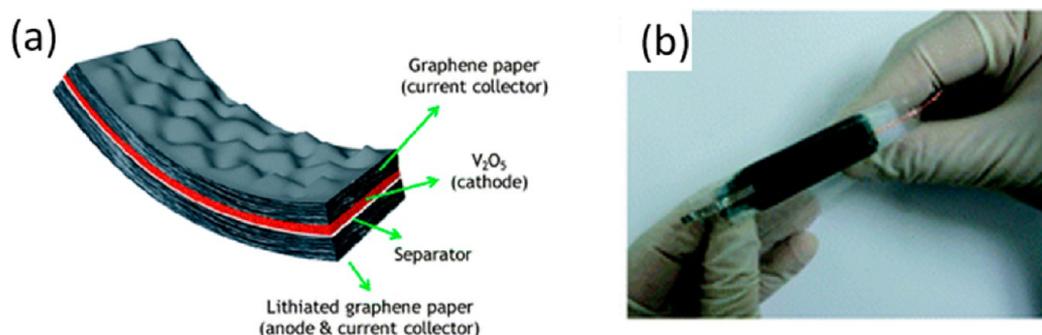


Figure 8. (a) Schematic of a graphene-based flexible lithium ion battery using V_2O_5 /graphene as cathode and pure graphene paper as anode and (b) bending of the graphene-based battery. (Reprinted with permission from ref 56. Copyright 2011 Royal Society of Chemistry.)

Flexible materials are important components to determine the flexibility of LIBs. Carbon materials, especially for carbon nanotubes (CNT), graphene, and carbon nanofibers (CNF), have been largely used in flexible batteries as conducting elements and flexible matrix. In addition, the high surface area, porous structure, good mechanical property, and high chemical stability of carbon materials will facilitate the electrochemical performance of flexible batteries.

CNT can be made into flexible membranes and bendable yarns by wet-processing method, such as vacuum filtration, or dry-processing method, like directly drawing. The lightweight, flexible, and highly conductive CNT networks can be combined with active materials for use as cathode and anode electrodes.

In the cathode, CNT network can replace conventional aluminum (Al) current collector, so that high voltage tolerance, lightweight, and high contact area between active materials and current collectors can be achieved. In addition, the porous CNT network can allow better liquid electrolyte adsorption. Cathode materials, including $Na_xV_2O_5$, V_2O_5 , $LiMn_2O_4$, and $LiCoO_2$, have been used to make composite with CNT to construct flexible cathode electrodes.^{31–34} A representative work to prepare flexible cathode is using ultrasonication and codeposition method. Figure 7a and 7b shows the simple method to produce flexible electrode with commercial $LiCoO_2$ embedded in CNT network.³¹ The active $LiCoO_2$ ink was dispersed onto the superaligned carbon nanotubes (SACNT). After sonication, the composite were entangled into a robust and continuous three-dimensional (3D) structure to obtain dense composite. This cathode structure is highly flexible and strong, which can be applied to further battery fabrication processes without cracking. The stress–strain curves in Figure 7c show that the strength and Young’s modulus of the conventional battery composite were only 3% of the strength and 10% of the Young’s modulus of the developed flexible electrode. The inset shows that the strength of the flexible electrode increased linearly with the amount of CNT.

For the anode design, CNT can be used as the anode material directly in the form of infiltrated membrane and as self-assembled membrane by direct chemical vapor deposition growth. CNT has the lithium storage capability of 100–600 mAh/g.^{35–39} In order to achieve better electrochemical performance, higher-capacity anode materials such as Si, Ge, Sn, and transition-metal oxides can be introduced.^{40,41} For these anode materials, the CNT network not only provides good electrical substrate but also creates a porous matrix to buffer the volume expansion of the anode materials during lithiation, minimizing the internal stress of electrode and addressing the delamination challenge of electrode coating and

current collector. The other is using a dry-processable CNT film by drawing aligned CNT arrays to fabricate a flexible electrode.⁴² Such dry-processable CNT film has a superaligned structure that can be made into current collector for loading active materials, such as SnO_2 .⁴³ When active material was coated directly onto CNT, such as Fe_3O_4 by magnetron sputtering or Si via chemical vapor deposition (CVD), the core–shell structure exhibited a high capacity, good rate capability, and high capacity retention.^{44,45} Figure 7d shows a continuous aligned CNT film drawn from the spinnable CNT arrays and rolled on a cylinder. After Si deposition by using silane gas in CVD furnace, the free-standing Si–CNT with a core–shell structure was obtained and showed good flexibility, as shown in Figure 7e.⁴⁴ This film can be punched into a disk for lithium ion battery testing (Figure 7f).

Because of the one-dimensional structure, CNT is a good elementary building block for making a flexible substrate. Its large surface area and high electric conductivity can provide a short electron-transfer pathway to active materials, and the free space in the CNT assembly can have better liquid electrolyte penetration and enable fast Li ion transport. Although extensive studies have demonstrated CNT’s great potential use in Li ion batteries, some concerns and challenges of CNT remain to be addressed. The first is related to the Coulombic efficiency. The high surface area and structural defects will lead to heavy formation of solid–electrolyte interface (SEI) and lower the Coulombic efficiency, especially the initial Coulombic efficiency. The second is about the CNT assembly and the CNT-based composites. CNT assembly tends to agglomerate, and CNT generally has a weak adhesion with active materials. The SEI formation on CNT may block electron transfer at the interface, leading to capacity decay. The third is the challenge of reducing cost and being adopted into the state-of-the-art battery manufacturing. If the aforementioned concerns and challenges can be addressed, CNT can boost the development of flexible Li ion batteries toward high performance and high flexibility.

Graphene as a two-dimensional “wonder” material has been explored for electrochemical energy storage for years because of the superior properties in terms of high surface area, good electrical conductivity, good chemical stability, and good mechanical flexibility. The role of graphene for electrochemical energy storage becomes much more important with continuously developing methods to produce graphene at low cost at large scale.⁴⁶ Because of properties similar to those of CNT, graphene materials not only can be used as conducting additives for making conventional flexible electrode but also can be prepared into graphene papers by using various

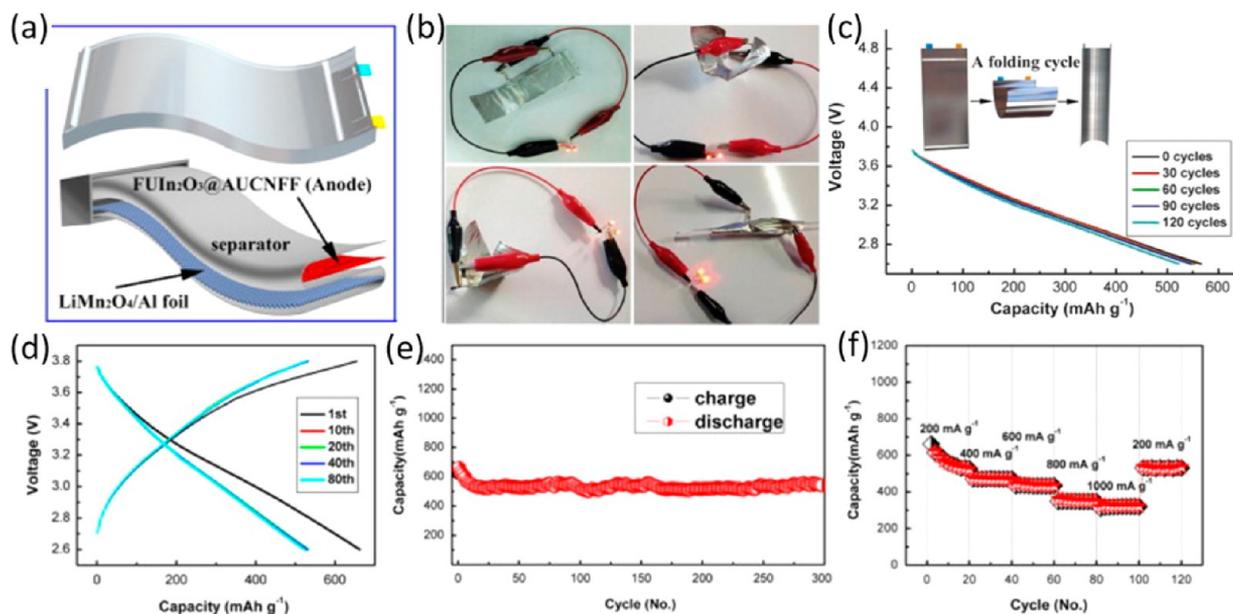


Figure 9. (a) Schematic of CNF-based flexible battery, (b) LED lit up by the flexible battery under different bending and twisting states, (c) discharge curves of battery at different cycles during repeated bending test, (d) charge and discharge curves of battery, (e) cycling stability, and (f) rate performance. (Reprinted with permission from ref 63. Copyright 2015 Elsevier.)

fabrication methods, such as vacuum infiltration of graphene inks and mechanical pressing of graphene aerogels, for use as anode electrode directly, or use as conducting substrate such as current collectors and 3D matrix to load with high-capacity cathode (V_2O_5 , $LiFePO_4$, FeF_3) and anode active materials (Si , SnO_2 , Fe_3O_4 , Co_3O_4 , MnO_2 , TiO_2).^{47–55}

Through the integration of cathode and anode, the flexible graphene-based electrode can be made into a flexible full cell. Figure 8a shows a schematic of a graphene-based flexible lithium ion battery using V_2O_5 /graphene as cathode and pure graphene paper as anode.⁵⁶ The graphene anode was prelithiated before assembling into the full cell. As shown in the schematic, the wrinkle and ripple surface of graphene can increase the contact area of active materials with the current collector. The full cell can be operated at regular charge–discharge capability under full mechanical flexibility, such as bending and twisting (Figure 8b). Similarly, graphene foams (GF) synthesized by using templated growth graphene as a free-standing, lightweight 3D structure containing a $LiFePO_4$ cathode and a $Li_4Ti_5O_{12}$ anode were fabricated.⁵⁷ No metal current collector, carbon black additive, or binder was used in this design. The bendable full cell was prepared by a free-standing cathode and anode with a thickness of ~ 100 μm to laminate onto both sides of a commercial separator and then sealed with a poly(dimethylsiloxane) (PDMA) using conventional ether-based liquid electrolyte. The full cell had a good flexibility and bendability; there was no structural failure after repeated bending of a < 5 mm radius, and this cell can power a light-emitting diode (LED) in a bending state. These results demonstrate that graphene is a promising carbon material for constructing highly conductive and highly flexible electrodes toward full cell integration.

In general, graphene can be used as an active material or an inactive component as conducting additive, and in the past years, a large amount work on graphene and its composites has been focused on energy storage applications. However, it is elusive whether graphene is a “wonder” material that can potentially boost the development of energy storage, especially

flexible Li ion batteries with high energy density. It is important to increase the packing densities both gravimetrically and volumetrically to achieve batteries with high energy density. It is essential to avoid the restack of graphene to maximize the fast electron and ion transport with active materials at the interface. Increasing the Coulombic efficiency is the key for the practical consideration of batteries. How to build up a mechanically stable 3D graphene matrix that can sustain repeatable bending is the main challenge for making flexible batteries. If these concerns and challenges can be addressed, we still optimistically see a bright future for graphene in flexible energy storage applications.

Beyond CNT and graphene, carbon nanofiber (CNF) has attracted significant interest for electrochemical energy storage, especially flexible energy storage applications, because of the low-cost, fast production rate, and versatile structural design. Spinning is the main technique for producing nanofibers, followed by carbonization at high temperature in a protective gas environment. Because spinning is part of the textile industry, the large-scale production of CNF becomes possible, for example, by using electrospinning and centrifugal spinning techniques. Details can be found in reviews in the literature.^{58,59}

To synthesize cathode and anode materials, active material salt precursors/nanoparticles need to dissolve/disperse into polymer solutions to get a uniform composite solution, followed by spinning (electrospinning, centrifugal spinning) and thermal treatment (stabilization and carbonization processes) to get functional CNF composite as electrode materials. Extensive studies have been conducted based on CNF by tailoring the structure to address the fundamental challenge of active materials such as Si and Sn and use the intrinsic flexibility of CNF to fabricate free-standing electrodes and demonstrate flexible and bendable batteries.^{60–62} Recently, a flexible lithium ion full cell was reported by using ultrafine In_2O_3 nanocrystals@aligned ultralong CNF film as anode and $LiMn_2O_4/Al$ as cathode; the schematic is shown in Figure 9a.⁶³ The cathode and anode were wrapped between commercial separators and encapsulated in a flexible plastic shell case.

Figure 9b shows that two LEDs were lit up by the flexible battery even when the battery was in a bendable condition. Figure 9c indicates that the battery performance remained the same when the cell was bent from different directions for long-term cycles. The full cell can deliver a capacity of ~ 500 mAh/g at a current density of 200 mA/g with a voltage of 2.6–3.8 V (Figure 9d). The full cell exhibited a stable cycling performance and good rate capacity at different current densities (Figure 9e,f).

To enable high power and high energy density, it is required that CNF should have higher electric conductivity, low sheet resistance, interconnection between nanofibers, and low area density. If CNF can meet these requirements, we will see a promising future for flexible CNF in the application of flexible batteries due to its good mechanical properties, versatile synthesis of CNF-based composite as cathode and anode, and fast and low cost in large-scale production for making superior flexible batteries.

In addition to the electrically conductive materials, ionically conductive materials as the other important building block materials are believed to play a vital role in securing cell safety. Conventional liquid electrolyte-based batteries contain organic liquid electrolyte to transport Li ions between cathode and anode. The flammable and fluidic organic liquid electrolytes have intrinsic safety concerns that may pose safety issues to batteries, especially flexible batteries. To avoid potential safety concerns, solid-state ionically conductive materials can be used to replace the organic liquid electrolyte in flexible batteries. In this section, we review two different types of solid-state electrolytes, gel polymer electrolytes and ceramic/polymer electrolytes, which can be potentially applied to flexible battery design.

Gel polymer electrolytes have the advantages of being electrolyte leakage-proof and having low flammability, low toxicity, and relatively high ionic conductivity. The gel polymer electrolytes generally still contain liquid organic electrolytes, but these liquid electrolytes are trapped in polymers. To enhance the mechanical flexibility and liquid electrolyte uptake, fillers such as nanoparticles are added into the polymer matrix. Although gel polymer electrolytes are categorized as solid-state electrolytes, strictly speaking, the trapped liquid electrolytes may still possess safety concerns similar to those of conventional batteries.

Ceramic/polymer electrolytes are an important type of solid-state electrolytes. Intrinsically, ceramic/polymer electrolytes can be considered as pure solid solids because there is no liquid electrolyte in the electrolyte system. A polymer matrix mixed with ceramic ion conductors and Li salt are the main components in the composite electrolytes.^{64–67} Poly(ethylene oxide) (PEO) is the most widely studied material for solid electrolytes.⁶⁸ Ceramic ion conductors can influence the crystallization kinetics of the PEO polymer chains and promote local amorphous regions, and the ceramic ion conductors can provide extended ionic transport pathways in the polymer matrix. Recently, studies have shown that continuous ceramic ion conductors within the polymer matrix can enhance the ionic conductivity of solid electrolytes, and a flexible solid-state electrolyte with 3D ion conductive networks was reported.^{65,67} Figure 10 shows the schematic of the flexible solid electrolyte composite, where ceramic ion conductive nanofibers function as the reinforcement and the Li ion conducting polymer as the matrix.⁶⁷ The 3D nanofiber networks form a continuous ion-conducting pathway in the polymer matrix. The reported ionic

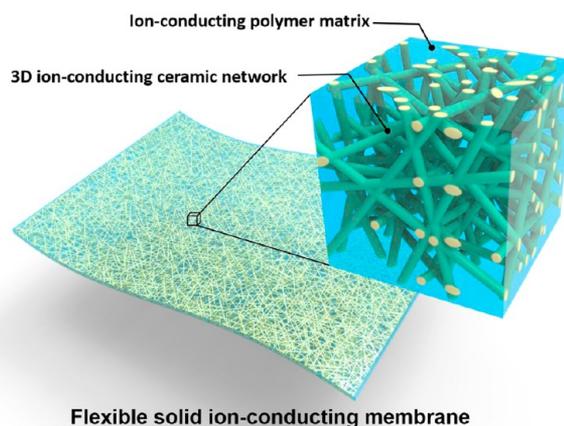


Figure 10. Schematic of flexible solid-state composite electrolyte. (Reprinted with permission from ref 67. Copyright 2016 National Academy of Sciences.)

conductivity was 2.5×10^{-4} S/cm at room temperature. This flexible solid-state electrolyte membrane can be potentially applied to flexible batteries.

Although ceramic/polymer electrolytes are good ion transport media, challenges remain in terms of low ionic conductivity and high interfacial resistance with electrodes.⁶⁹ Therefore, extensive studies are highly expected to address these challenges for flexible batteries and fundamental study of solid-state ionically conductive materials.

New Material and Structural Designs. To design a flexible battery, one straightforward idea is using a flexible substrate to replace the conventional rigid metal substrate and following industrial battery fabrication processes to manufacture flexible electrodes and batteries. Inspired from the papers and textiles, their ultraflexibility can be used to fabricate conducting flexible substrate for assembling flexible batteries. Details about paper and textile energy storage can be found in the literature.^{70,71} The first paper battery was developed by Hu et al.; a CNT slurry-coated paper loaded with LiMn_2O_4 and $\text{Li}_4\text{Ti}_5\text{O}_{12}$ active materials was fabricated for the demonstration of a flexible battery (Figure 11a).⁷² In their work, commercial paper was made into a highly conductive substrate with a sheet resistance at 1 ohm per square, and the sheet resistance remained unchanged when different bending radius was applied (Figure 11b). After being loaded with active materials, the battery showed a performance similar to that of conventional batteries.

The first textile battery was also reported in work by Hu et al., in which they reported cotton fabrics dyed by using CNT inks.⁷³ The CNT-coated fabrics with cathode and anode materials were then assembled into a battery. Figure 11c shows the schematic of CNT wrapping around cellulose fibers to form a 3D porous structure.⁷³ Because of the mechanical flexibility of CNT and high surface area of cellulose textile fibers, together with the large water absorption of the fibers in the dyeing process, surface contact areas between CNT and cellulose fibers will be maximized, leading to a highly conductive textile with a sheet resistance of ~ 4 Ω /square (Figure 11d,e). Figure 11f is the SEM image of a fabric sheet coated with CNT on the fabric fiber surface. The textile porous structure can facilitate the easy access of electrolyte and the adsorption of electrode locally. After CNT coating, the cotton fabric still preserved the textile pattern, indicating that the CNT coating has no influence on the structural change of textile fiber assembly.

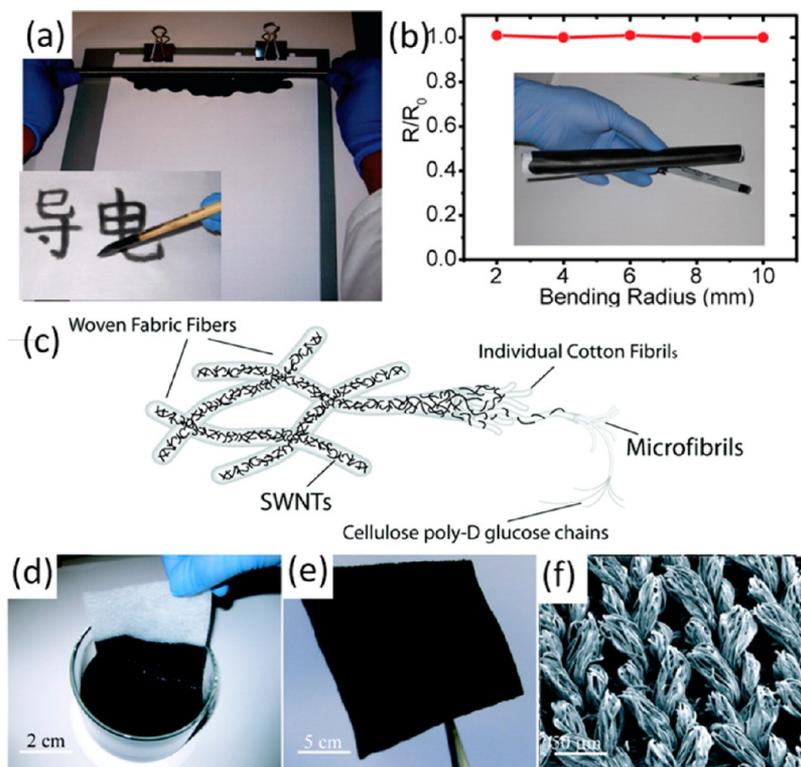


Figure 11. (a) Meyer rod coating of CNT or Ag NW ink on commercial copy paper. Inset is the direct writing of CNT ink on the paper with Chinese characters. *Daodian* means conductive. (b) Sheet resistance changes after bending conductive paper into different radius. (Reprinted with permission from ref 72. Copyright 2009 National Academy of Sciences.) (c) Schematic of SWNTs wrapping around cellulose fibers to form a 3D porous structure. (d) Conductive textiles are fabricated by dipping textile into an aqueous SWNT ink followed by drying in oven at 120 °C for 10 min. (e) A thin, 10 × 10 cm² textile conductor based on a fabric sheet with 100% cotton and sheet resistance of 4 Ω/sq. (f) SEM image of CNT coated textile fabric. The scale bar is 50 μm. (Reprinted from ref 73. Copyright 2010 American Chemical Society.)

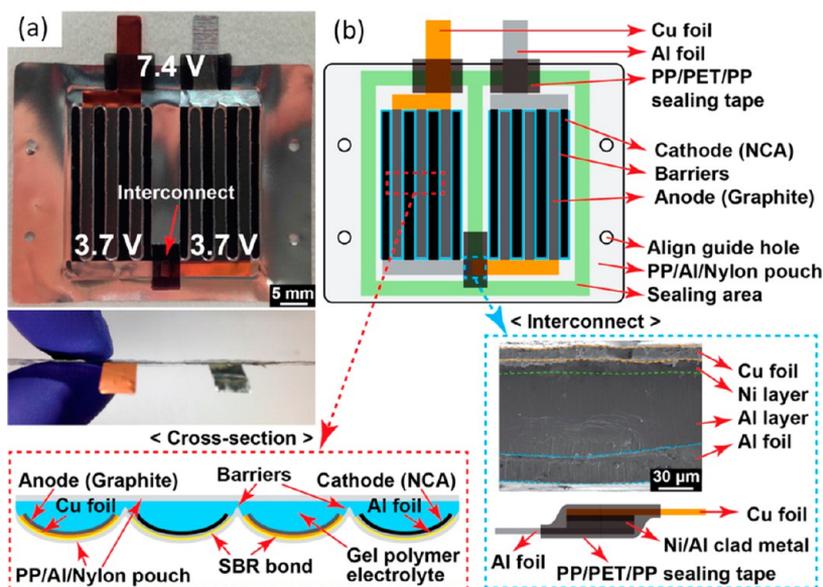


Figure 12. (a) Top-view and cross-sectional photographs of a coplanar flexible battery in which two 3.7 V cells are connected in series. (b) Schematic illustration of a coplanar flexible battery showing the cell configuration composed of interdigitated electrodes, interconnect, metal tails for external electric connection, and pouch. (Bottom left) A cross-sectional illustration of the interdigitated electrodes with pouch barriers between adjacent anodes and cathodes by the curvy geometry of each electrode. (Bottom right) Ni/Al clad metal intermediate layers to bridge Al and Cu interconnects. Al and Cu are not feasible to be directly welded. (Reprinted from ref 74. Copyright 2010 American Chemical Society.)

Paper- and textile-based batteries have demonstrated good flexibility, high active material mass loading, and good cycling

performance, which are ideal substrates to fabricate flexible and wearable batteries. One potential disadvantage is the high liquid

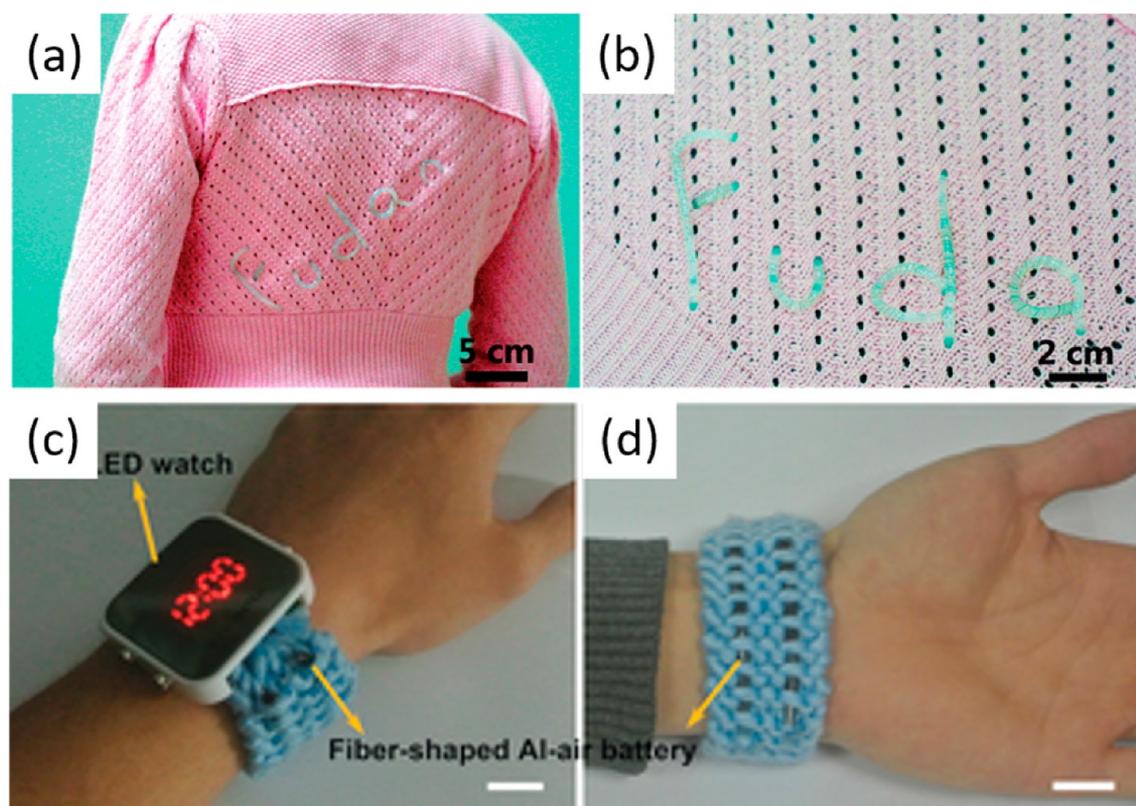


Figure 13. (a, b) Photographs of stretchable fiber-shaped batteries being woven into a knitted sweater. (Reprinted with permission from ref 75. Copyright 2014 Royal Society of Chemistry.) (c, d) Photographs of a commercial LED watch powered by two fiber-shaped Al-air batteries woven into a fabric and connected in series (scale bars, 2 cm). (Reprinted with permission from ref 75. Copyright 2016 Wiley.)

electrolyte uptake in the porous paper and textile structures. Although more electrolyte can enable better ion transport, the safety concern remains in the high risk of flammability and leakage. In addition, more electrolyte will increase the total weight of batteries and diminish its energy density. In the future, design ultralight paper and textiles structure and use aqueous electrolyte to make aqueous electrolyte-based flexible Li ion batteries are becoming promising.

For most of the flexible lithium ion batteries, a conventional stacking cell configuration is widely applied, in which cathode, separator, and anode are sandwiched together. For a thin battery application, stacking cell configuration can maintain good flexibility. However, it becomes a challenge to have a thick battery and meanwhile maintain good flexibility. For the thick battery, the flexibility of the stacking cell will be compromised and delamination may occur once the battery is bent with a small bending radius. Therefore, it is important to design a novel battery configuration that can meet high mass loading and small bending radius at the same time.

To address this challenge, recently a new flexible battery strategy was developed: a coplanar cell structure in which cathodes and anodes are interdigitated.⁷⁴ The coplanar electrode design has the advantage of improved bending tolerance and increased mass loading. Panels a and b of Figure 12 show a photo image of the complete cell before sealing and the schematic showing the cell configuration, respectively.⁷⁴ The interelectrode barrier with the curvy electrode structure plays an important role in the electrochemical performance of the battery under bending conditions. Such an interdigitated flexible battery has an entire cell thickness of 0.5 mm and a stable battery performance over a 5000 repetition bending test

with a bending radius of 5 mm. In addition, the interdigitated pouch case can effectively prevent Li dendrite growth and short-circuit because of the barrier at the point between anode and cathode. Therefore, the combined approach of the coplanar geometry of electrode patterns and the novel interelectrode barriers will enable the robust bending capability and good electrochemical performance. In addition, important inspirations from this design are that the pattern of each single cell can be arbitrary designed and the series-/parallel-connection of the cell can be tailored according to the requirements of the electronic devices.

This battery design successfully addressed the strain and stress issues generated under deformation. However, the proposed interdigitated electrodes configuration may limit the electrochemical performance of the cell at high current rate because of the relatively long ion diffusion distance between cathode and anode. To achieve fast ion rate in batteries, it is desirable to design a stacked cell with electrode–separator–electrode configuration while adopting the proposed strategy to design high-performance flexible batteries.

Textile is the art of fibers. Inspired from textile structure, building fiber batteries can be the key elemental building block to realize complete flexibility of batteries toward ultimate wearable applications. Recently, a new family of fiber-shaped batteries has been extensively studied. Because of the one-dimensional structure, fiber-based batteries can effectively work under different deformations, and the fiber structure can be potentially adopted into current textile manufacturing processes to fabricate truly textile batteries. The woven or knitting pattern of fiber-based batteries can mimic real textile fabrics to allow moisture vapor and air to pass through. In addition, the flexible

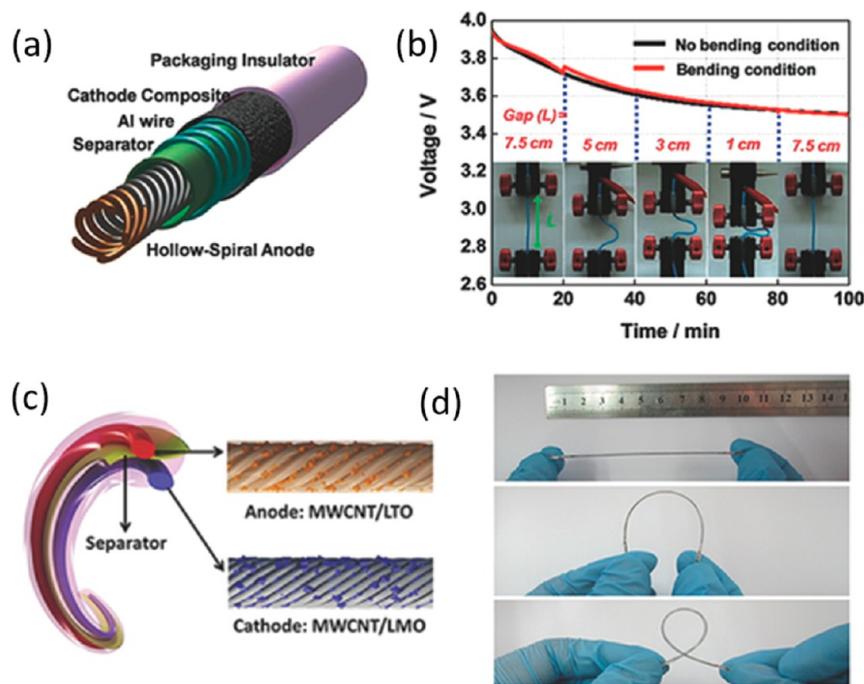


Figure 14. (a) Schematic illustration of the structure of a helically coaxial fiber LIB, (b) discharge characteristics of the cable-type LIB under bending, (c) schematic illustration of the parallel structure of a fiber LIB, and (d) images of the parallel fiber-shaped LIB being deformed. (Reprinted with permission from refs 78 and 79. Copyright 2012 and 2014, respectively, Wiley.)

fiber-based textile batteries can be integrated into garments to realize highly wearable applications, as demonstrated in Figure 13.^{75,76} The integrated batteries will not influence the appearance of the garment. These unique features allow one-dimensional fiber-based batteries a promising future for wearables compared to that of conventional planar batteries. Details of fiber-based batteries are reported in the review by Peng's group.⁷⁷

A fiber-shaped battery generally consists of two twisted fiber electrodes, including two typical structures: helically coaxial and parallel structures. Figure 14a shows a typical cable-type battery made with a helically coaxial structure.⁷⁸ Ni–Sn was used as hollow spiral anode, which was prepared by depositing Ni–Sn on a hollow-spiral Cu wire. Al wire coated with LiCoO₂ as cathode was wound around the anode with a polymer separator in between. This cable-type battery showed good flexibility and stable battery performance regarding to the increased bending angle (Figure 14b). However, because of the large diameter (in the millimeter range), strictly speaking, this cable-type battery cannot be called a fiber battery. Reducing the diameter of the cable-type battery is important to achieve the possibility of weaving or knitting. Therefore, the other type of fiber battery having the parallel structure shown in Figure 14c is more like a “fiber-based battery” that can demonstrate the weaving function for wearable batteries.⁷⁹ In this design, LiMn₂O₄/CNT is used as cathode and Li₄Ti₅O₁₂/CNT is used as anode. The CNT yarn was prepared by direct-drawing from the aligned CNT arrays, followed by twisting. The twisted CNT yarn can exhibit superior mechanical properties. To assemble the battery, a heat-shrinkable tube is used to encapsulate the twisted electrodes and separator. Figure 14d shows a photo image of bending an electrode into different shapes to demonstrate its good flexibility.⁷⁹

One-dimensional fiber-based batteries are still in the early stage, and their future development will mainly rely on

The gap between structural and material design is trending down in the sense that structural design can be performed at the material level to develop deformable materials. The coordination of structure and material design is anticipated to further enrich our toolkit for wearable battery invention.

materials. As the basic building block, aligned CNT fibers and bundles require higher electric conductivity to enable long and continuous electrodes with lower internal resistance. Besides Li ion batteries, use of other types of battery chemistry, such as lithium–sulfur batteries, lithium–oxygen, zinc–air, and aluminum–air, can significantly increase the energy density, and very recently cable-based lithium–sulfur batteries have been reported by the Hui group.^{76,80–82} In addition, minimizing inactive components in fiber batteries is important for the increase of battery energy density. With the increasing development of wearable electronics, we can envision that flexible fiber batteries will play an important role in future development of wearable energy storage devices.

Summary and Future Outlook. In this review, we discussed the mechanics in flexible structure design and reviewed recent work of flexible and wearable batteries based on two aspects: structural design and material design. For the structural design aspect, we impart the principles of technologies which enabled flexibility in otherwise undeformable structures and sort out the developments in flexible electronics which represent possible routes toward flexible batteries. We expect these principles in structural design could inspire novel structures that are

applicable for flexible batteries. Also, we note the tendency that the boundary between structural design and material design actually blurs. The gap between structural and material design is trending down in the sense that structural design can be performed at the material level to develop deformable materials. The coordination of structure and material design is anticipated to further enrich our toolkit for wearable battery invention.

New material and structural design, including paper and textile batteries, ultrathick batteries with unique curvy electrode structure, and one-dimensional fiber-based batteries, are also reviewed for the advanced flexible and wearable battery design.

Downsizing the diameter of the battery device will ultimately realize the integration of batteries into flexible substrates and other flexible electronics serving a truly wearable function.

Downsizing the diameter of the battery device will ultimately realize the integration of batteries into flexible substrates and other flexible electronics serving a truly wearable function. Such dimension reduction of the building blocks indeed enlarges the degree of freedom which can be attained in flexible batteries. Therefore, these flexible and wearable materials and structures are not limited to the batteries; we can envision that the mechanics and design concepts can be also extended to other electronics for wearable applications.

Good flexibility and high electrochemical performance are two key factors to evaluate wearable batteries. It is important to find balanced properties between flexibility and electrochemical performance, and it is necessary to evaluate the two properties on the full cell rather than a single-component level. In addition, most research communities contributed great effort on materials and less to structure and mechanics considerations in flexible battery design. On the basis of our understanding, we propose the following challenges and perspectives for the future development of flexible batteries:

- (1) Standard tests and evaluations for flexible batteries are urgently needed. Flexibility tests for batteries, including flexible electrodes, electrolytes, packaging, bending test, twisting test, stretching test, fatigue test, and safety test, are important to ensure the high quality of flexible batteries.
- (2) Cooperation between scientific research community and industry partners is important. Flexible battery design and development should meet with practical use and better follow current state-of-the-art battery manufacturing processes.
- (3) Great effort has been focused on materials study for single battery components, such as electrodes and electrolytes. More future work should focus on full cell integration and full cell evaluation. Through the engineering of full cell structures, flexible full cells are expected to achieve electrochemical performance comparable to that of conventional Li ion batteries.
- (4) It is preferable to have fundamental study on the understanding of structures and mechanics in flexible battery under bending and other complicated deformations.

- (5) Safety is the top priority for batteries, especially for wearables. Conventional organic liquid electrolytes have safety concerns due to their high flammability. To replace the organic electrolyte, recently developed of solid-state and aqueous electrolytes can be applied to flexible batteries.
- (6) To meet the high energy density requirement, it is important to adopt new battery chemistry, such as lithium–sulfur and lithium–air, in the development of flexible batteries.

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