

# The Functionalization of Miniature Energy-Storage Devices

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In addition to fabricating miniaturized but high-performance energy-storage devices, integrating them with additional intelligent functions is also of great significance to accelerate the development of portable and wearable electronics. The integration of functional materials into energy-storage devices can endow them with impressive properties besides energy storage, allowing many unprecedented applications to be possible. Here, the recent progress and methodology of constructing functionalized miniature energy-storage devices that can change color, memorize shapes, and self-heal are summarized. The strategies in the integration of different functions and the performances of these intelligent energy-storage devices are then highlighted. The remaining challenges and future directions are provided in conclusion.

## 1. Introduction

The ever-growing demand for sustainable energy has pushed forward the development of efficient energy-storage devices.<sup>[1–3]</sup> In particular, with the increasing attention in portable and wearable electronics, tremendous efforts have been devoted to make miniature but powerful energy-storage devices.<sup>[4–8]</sup> Hence, high-performance energy-storage devices, including batteries and supercapacitors, are designed to be flexible, lightweight, and miniature, such as thin film,<sup>[9]</sup> micrometer fiber,<sup>[10]</sup> and ring-like structures.<sup>[11]</sup> The aforementioned work has been summarized in many previous reviews, remarking on the achievements from fundamental research to technical innovation, and thus will not be included here.

Apart from the continuing efforts to improve the energy-storage performance and fabricate miniaturized configurations for better compatibility, considerable interests have also been attracted to endow energy devices with desired functions. Besides the basic function of storing electrical energy, the integration of additional smart functions into miniature energy-storage devices can make them more suitable under various circumstances, allowing many unprecedented applications to be possible. An ideal miniature energy-storage device should not only possess high electrochemical performance with good durability, but also be endowed with impressive properties for specific purposes, such as the capability to self-heal after mechanical damage or self-regulate

its own temperature for better safety management. In this regard, transparent, compressible or stretchable energy-storage devices have been extensively studied as a class of electronic devices with promising properties.<sup>[12–15]</sup> However, it is noted that these properties are far from intelligent functions, so the scope of recent research on energy devices has been further extended to other more intriguing and challenging smart functions. Here, we focus on functionalized energy-storage devices, including smart functional materials introduced into conventional energy devices and novel structures aimed at one or more smart

functions. In recent years, significant breakthroughs have been made in the fields of electrochromic, shape-memory, and self-healing energy devices, which are responsive to external stimuli of voltage, mechanical deformation, and configurational integrity, respectively.<sup>[16–18]</sup> Here, the recent progress in these three subtopics, including smart functional materials and working mechanism of each function, are introduced, with an emphasis on the strategies of device integration and resultant performance. Finally, several ideas regarding the current challenges and further improvement of multifunctional energy-storage devices are proposed.

## 2. Electrochromic Function

Being intellectualized for miniature devices has become mainstream among various kinds of functionalization, the key point of which lies on the intelligent interaction between users and devices.<sup>[19,20]</sup> Fabricating intelligent devices that can feed back real-time working conditions via different visualizations is an effective strategy. On the basis of this idea, efforts have been devoted to developing various electrochromic devices such as smart windows, which can modulate their color and transparency by electrochromism and have been applied to various fields including the aircraft industry, the construction industry, and automobiles.<sup>[21,22]</sup> These smart devices are based on electrochromism, in which an apparent coloration process can be observed by the naked eye accompanying charge insertion/extraction or redox reaction occurring in an active electrochromic materials (**Figure 1a**).<sup>[23,24]</sup> Typically, electrochromic devices are produced by incorporating electrochromic materials onto transparent current collectors.<sup>[25]</sup>

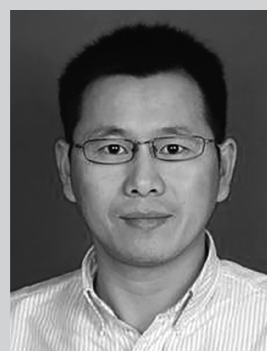
A large proportion of energy-storage devices (i.e., supercapacitors and batteries) with Faradic reactions at the electrodes are interlinked with electrochromic devices considering their similar active material types and related device structure and

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reaction kinetics, which demonstrate promising integration potential of these two kinds of devices.<sup>[26]</sup> More specifically, a miniature electrochromic energy-storage device can be regarded as a rechargeable battery with electrochromic materials incorporated in the transparent electrodes with a highly compact, miniature structure. The significance of integrating the electrochromic function into miniature energy-storage systems at the level of the materials mainly lies in the following aspects. Firstly, the real-time display of charging–discharging status can be realized, which may protect the device from overcharging and avoid unexpected powering off. Considering the limited dimension of miniature energy-storage devices, this tactic is more easily implemented compared with installing an energy-consuming indicator light-emitting diode (LED) into the minimized space. Secondly, overheating can be avoided for the internal structure of energy storage by effectively tuning the transparency of the device surface to resist excessive light absorption. Besides, a tunable transparency of the device can also ensure the privacy of its users. In short, the integration of electrochromic devices and energy-storage systems at a miniature size can hugely improve the usage experience and safety toward real-world applications.

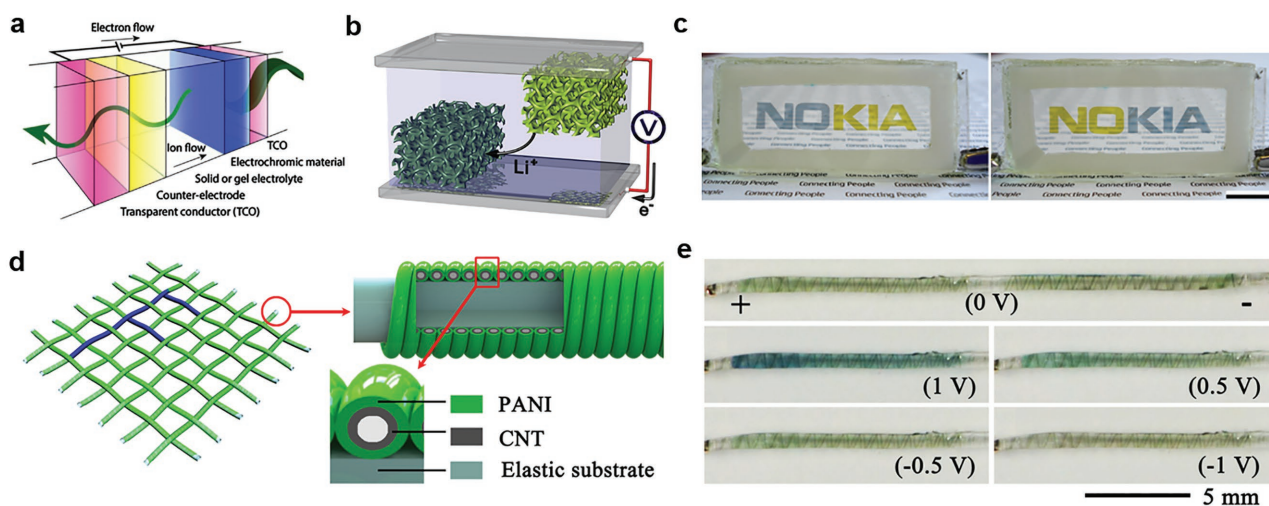
Metal oxides endowed with electrochromism are promising materials from which to construct miniature electrochromic energy-storage devices. For instance, as widely adopted candidates, tungsten oxides ( $\text{WO}_3$ ) have been incorporated on the surface of transparent conducting electrodes via the thermal-evaporation method to form thin-layered composite electrodes composed of  $\text{WO}_3$ . The resulting devices demonstrated effective color variations from transparent to deep blue and demonstrated desirable specific capacity as energy-storage devices.<sup>[27]</sup> Additionally, transparent conducting glass can be replaced by a flexible poly(dimethylsiloxane) (PDMS) substrate embedded with a conducting Ag-nanowire network, which enables great potential toward advanced applications such as e-skins and e-papers.<sup>[28]</sup> Niobium oxides have also proved to be proper



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candidates for similar electrochromism.<sup>[29]</sup> The aforementioned electrochromic oxides demonstrated their coloration process during ion insertion. There are also electrochromic metal oxides for color variations under ion extraction, among which nickel oxide has been widely selected to fabricate functional energy-storage devices due to its desirable cyclic reversibility as well as low cost.<sup>[30]</sup> Toward the construction of supercapacitors, manganese oxide is a competitive candidate given its high theoretical specific capacity, facile preparation process, and extensive sources.<sup>[31]</sup> Besides, suitable nanostructures can be constructed to jointly enhance the electrochromic performance, as well as the power and energy density. A gyroid-nanostructured vanadium pentoxide ( $\text{V}_2\text{O}_5$ ) network was synthesized and it was demonstrated to gain more desirable electrochromic (Figure 1b,c) and electrochemical performances when applied in the fabrication of supercapacitors.<sup>[32]</sup>



**Figure 1.** a) Schematic illustration of the typical structure of electrochromic devices. Reproduced with permission.<sup>[24]</sup> Copyright 2014, The Royal Society of Chemistry. b) Schematic design of the gyroid  $\text{V}_2\text{O}_5$  electrochromic supercapacitor. c) Photographs of the supercapacitor device in (b) displaying color change upon charge and discharge. b,c) Reproduced with permission.<sup>[32]</sup> Copyright 2012, American Chemical Society. d) Schematic of the structure of electrochromic fiber-shaped supercapacitor. e) Chromatic transitions during the charge–discharge process of the supercapacitor in (d). d,e) Reproduced with permission.<sup>[39]</sup> Copyright 2014, Wiley-VCH.

For the application of smart energy-storage systems, conducting polymers have also been considerably developed. As a consequence of their intrinsically soft molecular chains,<sup>[33]</sup> the resulting miniature electrochromic energy-storage devices are of high flexibility and even stretchability compared to the easy occurrence of surface cracks in metal oxides, significantly expanding the matching potential for specific usage scenarios such as wearable microelectronics. For instance, polyaniline was demonstrated to be blue with the pernigraniline form, green in a slightly oxidized state, and yellow with leucoemeraldine, and the varied colors were determined according to the applied voltage.<sup>[34]</sup> Energy-storage smart windows with polyaniline films were endowed with both stable electrochemical and electrochromic performances.<sup>[35]</sup> It is worth noting that dopant-modified polypyrrole has also been proved to be an ideal candidate for fabricating miniature electrochromic supercapacitors due to its high theoretical capacity and mechanical properties.<sup>[36]</sup>

Apart from directly fabricating electrodes composed of active electrochromic materials, the strategy of constructing miniature electrochromic energy-storage systems based on carbon nanotubes (CNTs) and graphene has gained ever-increasing popularity in recent years. Conventional electrochromic devices typically consist of an electrode of electrochromic materials, a matching counterpart, two transparent current collectors, supporting materials, and an electrolyte.<sup>[28]</sup> This multilayered structure results in corresponding instabilities among the various interfaces and difficulties in the fabrication process, which will severely contradict the trend of making electronic devices more miniature. In comparison, by introducing aligned multiwall carbon-nanotube (MWCNT) films with high transparency and conductivity, the fabrication process could be greatly simplified with enhanced performances. Aligned MWCNT films were paved onto thin pre-stretched PDMS layers to form flexible and electrically conducting substrates, followed by facile electropolymerization of the monomer to prepare composite electrochromic electrodes. Two as-fabricated functional electrodes were then piled up with a gel electrolyte layer stacked between them to construct a miniature electrochromic supercapacitor.<sup>[37]</sup> Besides the simplified fabrication process and the enhanced flexibility, the composite structure of conducting polymers and aligned MWCNTs was demonstrated to generate higher cyclic reversibility and more tunable color variation of electrochromic performance, which could be regarded as a synergistic effect of the incorporated materials.<sup>[38]</sup> Furthermore, attributed to the optical and mechanical properties of aligned MWCNT films, similar fabrication tactics of incorporating a functional guest could also be adopted to construct miniature fiber-shaped electrochromic supercapacitors, demonstrating great potential to be woven into wearable power textiles (Figure 1d,e).<sup>[39]</sup> Similar to MWCNTs, graphene is also promising in applications of both energy-storage systems and optoelectronic devices. Due to the unique gate-tunable optical properties of multilayer graphene (MLG), the transparency of the electrode composed of MLG was found to be controlled via reversible intercalation of charges into the single layer, resulting from electrostatic doping without incorporating conventional electrochromic guest materials.<sup>[40]</sup> A continuous production of this

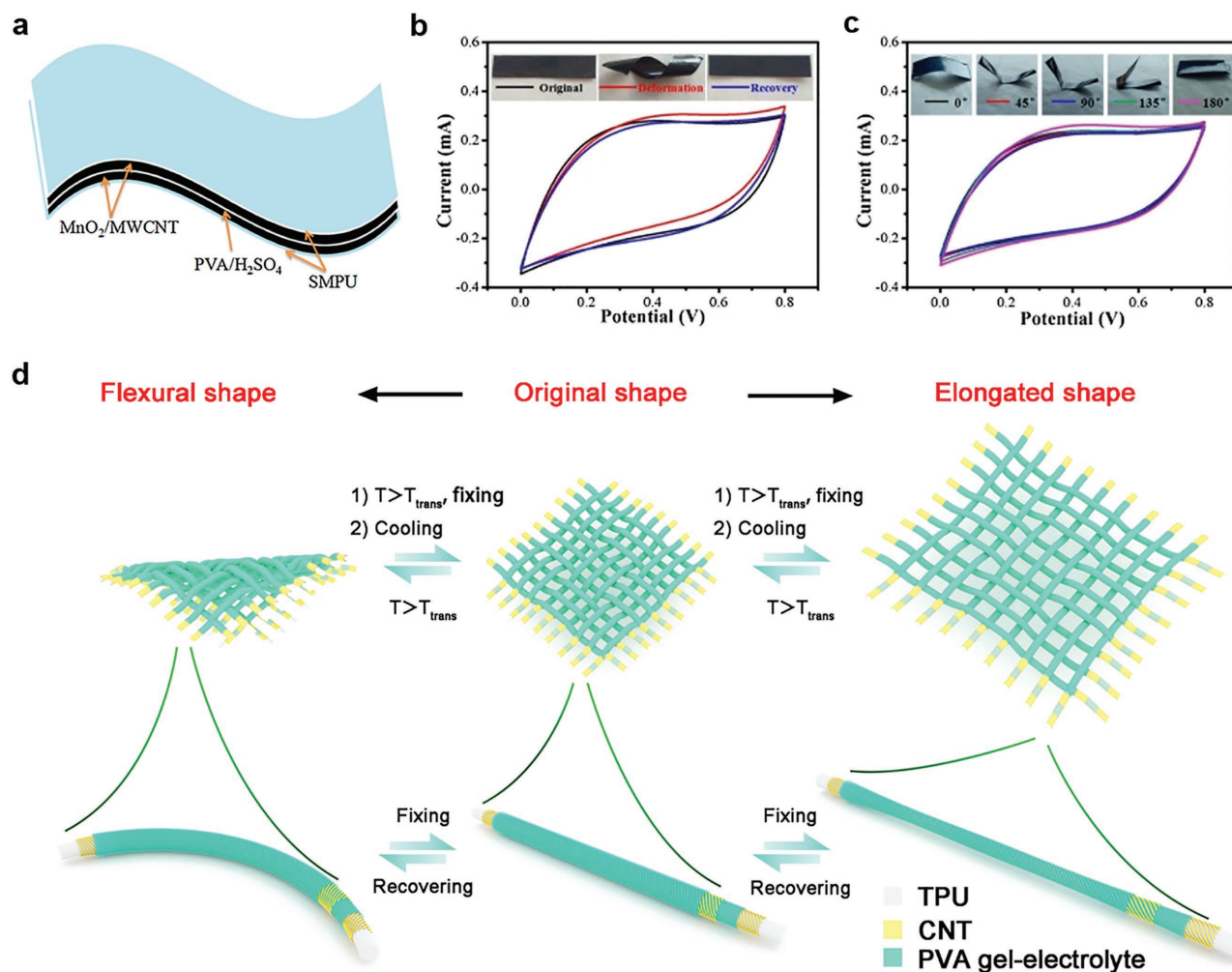
kind of sandwiched miniature supercapacitors may be realized through a roll-to-roll fabrication.

### 3. Shape-Memory Function

During practical application, miniature energy-storage devices may suffer irreversible deformations, which would result in the accumulation of local stress, structural fracture, and then their malfunction.<sup>[41]</sup> Introducing a shape-memory function into energy devices can enable them to recover back to their original shape or size in response to external stimuli, eliminating the local stress and then extending the lifetime of devices. Moreover, shape-memory devices can be controlled or tailored into designed shapes so as to meet the needs of various working conditions, and recover the original state whenever it is needed.<sup>[42–44]</sup> As a key component for shape-memory devices, shape-memory materials (SMMs) can be divided into two main categories: shape-memory alloys and shape-memory polymers.<sup>[45]</sup> By employing SMMs as the substrate or core of an electrode material, the as-prepared devices are endowed with shape-memory function as well as energy storage, providing a pathway for designing and fabricating smart integrated devices.<sup>[46]</sup>

Among the various shape-memory alloys, nickel–titanium (NiTi) has been mostly explored, given their high biocompatibility and mechanical and electrical properties, which enable them to be ideal flexible substrates for fabricating energy devices.<sup>[47,48]</sup> For instance, shape-memory supercapacitors with body-temperature-inducible shape-memory ability were successfully prepared by utilizing graphene-coated NiTi alloy as the negative electrode and ultrathin MnO<sub>2</sub>/Ni film as the positive electrode.<sup>[49]</sup> The resulting supercapacitor exhibited distinctive shape-memory ability, and could recover to its original shape after being placed in contact with a human wrist. Similarly, wire-shaped NiTi-alloy-based shape-memory supercapacitors were designed and prepared by using NiTi wires as the current collector and substrate for active materials of manganese dioxide and polypyrrole.<sup>[50]</sup> When the temperature reached the activation point, this wire-shaped device would recover to its original undistorted state within a few seconds despite serious plastic deformation, with its electrochemical performance remaining nearly unchanged.

Although NiTi alloys provide a high driving force that can actuate more complex structures, their heavy weight may result in a low energy density of the whole device. In comparison to NiTi alloys, shape-memory polymers are much lighter with better flexibility and stretchability, which make them promising candidates for preparing shape-memory devices.<sup>[51–55]</sup> As shown in **Figure 2a**, flexible thin-film supercapacitors were prepared by electrodes of CNT/MnO<sub>2</sub> composite film coated onto shape-memory polyurethane (SMPU) substrates.<sup>[56]</sup> The resulting shape-memory supercapacitors were able to be changed into arbitrary shapes as required, and they maintained them with good shape fixity. Moreover, this device could recover to its original shape in a few seconds once the temperature exceeded the glass-transition temperature of the SMPU. The electrochemical performance of this shape-memory supercapacitor remained unchanged during the deformation and



**Figure 2.** a) Schematic illustration of shape-memory film supercapacitor. b) CV curves of the film supercapacitor with and without being twisted. c) CV curves of the supercapacitor at different bending angles. a-c) Reproduced with permission.<sup>[56]</sup> Copyright 2016, Elsevier B.V. d) Schematic design of fiber-shaped shape-memory supercapacitor and the resulting textile that is reversibly transformed into flexural or elongated states and recovered to the original shape. Reproduced with permission.<sup>[58]</sup> Copyright 2015, Wiley-VCH.

shape-recovery process (Figure 2b,c). When this shape-memory supercapacitor was tailored into various shapes, its capacitance was well proportional to the area.

Compared with the conventional planar structure, fiber-shaped energy devices possess unique and promising advantages, particularly the ability to be woven into lightweight, flexible energy textiles.<sup>[57]</sup> As shown in Figure 2d, a coaxial-fiber-shaped shape-memory supercapacitor was fabricated by winding aligned CNT sheets on a SMPU substrate.<sup>[58]</sup> The as-fabricated devices were able to be programmed into different shapes to meet the needs of various working conditions, or woven into textiles to fabricate smart clothes for wearable electronic devices. In spite of its flexibility and stretchability, this fiber-shaped device could be deformed into various shapes and sizes as designed, and recovered to its original state whenever required. The capacitance of this device was well maintained during different kinds of deformations, in the deformed states, and after recovery. Furthermore, electronic textiles woven by such fiber-shaped supercapacitors also exhibited efficient

shape-memory ability, and improved voltage or discharging time when they were connected in series or parallel, respectively. These properties are particularly attractive for the development of next-generation wearable electronics.

Despite the recent progress in shape-memory energy devices, there remain many problems that need to be solved for future improvements. Firstly, the abovementioned shape-memory energy supercapacitors are thermoresponsive, and the shape-recovery process can only be stimulated by heat. Instantaneous shape recovery under ambient conditions or shape-memory function induced by other forms of external stimulation (e.g., light, humidity, etc.) are desired.<sup>[59–61]</sup> Secondly, it is not suitable for large-scale preparation to use the above-mentioned methods to fabricate shape-memory energy devices. Recently, wet-spinning methods were developed to fabricate shape-memory fiber-shaped supercapacitors, which employed wet-spun shape-memory polymer fiber as a substrate, followed by continuous deposition of active materials and coating of gel electrolyte.<sup>[62]</sup> With the advantages of low

cost, facile fabrication, and easy scalability, such a wet-spinning process may be a promising approach for large-scale fabrication of shape-memory devices.<sup>[63]</sup>

#### 4. Self-Healing Function

Miniature and wearable energy-storage devices may inevitably suffer from all kinds of local stresses under bending or other deformations in their long life cycles, which is a common problem for practical applications.<sup>[64]</sup> Within the devices, the electrode materials are susceptible to structural fracture under bending, while polymeric flexible substrates may possibly undergo structural damage caused by deformation over time or accidental cutting.<sup>[65]</sup> Such unavoidable failure would cause the breakdown of an electrochemical device, which will seriously limit the lifespan and reliability of the wearable electronics. Therefore, an ideal wearable device should not only present superior electrochemical performance but also be endowed with additional functions such as the ability to inhibit the structural fracture of the electrodes, or to restore its electrical property and configuration integrity after mechanical damage.<sup>[66]</sup>

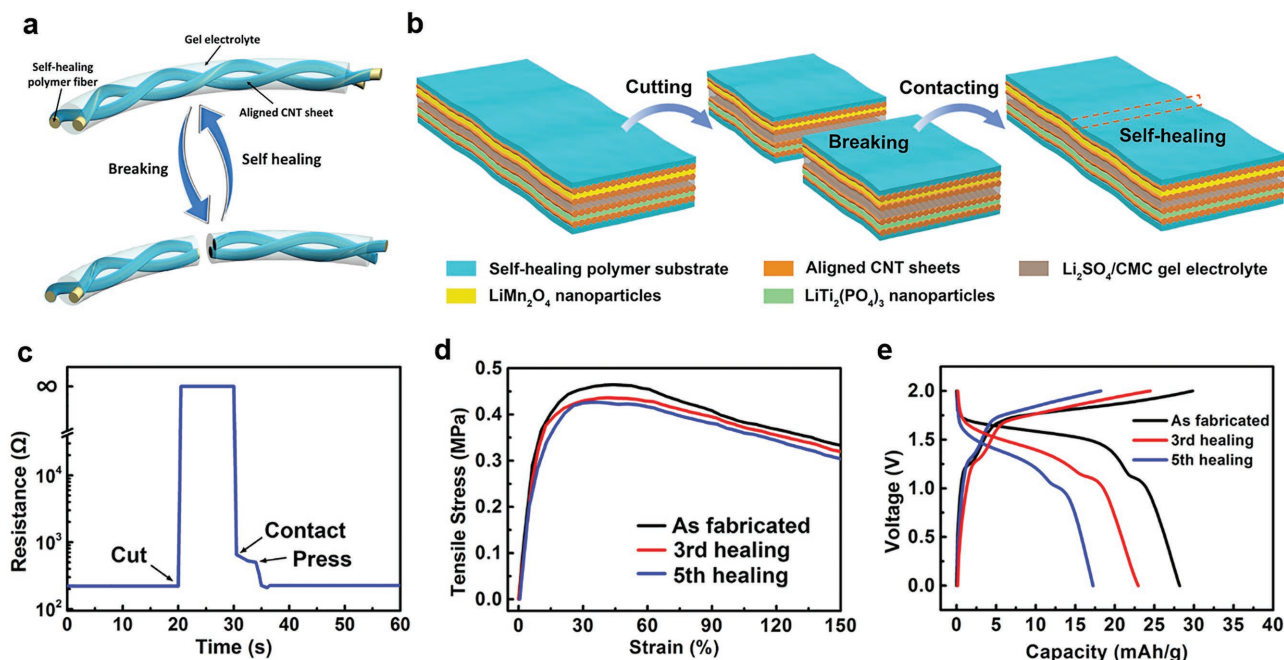
Self-healing materials that can repair the structural damage they have sustained have been developed over the past decade.<sup>[67–69]</sup> To date, a variety of self-healing materials have been developed, which can be divided into two kinds: intrinsic ones that can heal the breaking interface by using reversible covalent bonding or noncovalent interactions, and extrinsic ones that release healing agents from capsules or vascular networks after crack propagation.<sup>[70]</sup> Besides, self-healing materials have been successfully prepared with desirable physical (e.g., electrical, electronic, and thermal) and chemical (e.g., electrochemical and photochemical) properties.<sup>[71]</sup> In particular, for application in energy-storage devices, the restoration of their electrical properties after damage is of importance for self-healing electrochemical devices.<sup>[72]</sup> As a key component in self-healing energy devices, many self-healable electronic conductors with good self-healability and conductivity have been reported.<sup>[73–75]</sup> For example, conductive self-healable film was fabricated by spreading silver nanowires on self-healing polymer film, and the healing process can be repeatable at the same location for multiple cycles with 97% recovery of the surface conductivity in a short time.<sup>[76]</sup> These pioneering studies have greatly put forward the development of self-healing energy-storage devices. By integration of appropriate self-healing materials into miniaturized devices, the structural fractures of such electrode materials could be healed as well as the configuration integrity and electrical properties of devices being restored, which can perfectly solve the problem of portable and wearable electronics suffering from mechanical damage. In the past few years, self-healing polymers have been incorporated into various energy-storage devices, such as supercapacitors, lithium-ion batteries (LIBs) and solar cells, serving as substrates, coating shells, or electrolytes.<sup>[77–81]</sup> Self-healing polymers recover the mechanical integrity of the device while the healing of the conductivity is based on the recovery of the mechanical integrity.<sup>[82]</sup>

A typical example is a self-healing supercapacitor, with its conductive electrode prepared from a spreading single-walled

carbon nanotube (SWCNT) film onto a self-healing polymer substrate filled with hierarchical TiO<sub>2</sub> nanoflowers.<sup>[83]</sup> After mechanical damage, lateral movement of the self-healing composite layers enabled the separated SWCNT layers to contact with each other, thus realizing the restoration of the configuration and conductivity of the devices. The self-healing supercapacitors showed good electrochemical performance, and 85.7% of their specific capacitance could be restored after the 5th cutting. Self-healing function is also of great importance for fiber-shaped energy devices that are susceptible to structural fracture during practical applications in wearable electronics.<sup>[84]</sup> Toward this end, highly conductive and self-healable wires were fabricated by winding aligned CNT sheets around self-healing polymer fibers.<sup>[85]</sup> The resulting core–sheath fibers showed a low electrical resistance of 140 Ω cm<sup>-1</sup>, which increased by 8.5% after breaking and self-healing process. It was found that an aligned CNT strand was formed along the cross section of broken ends after cutting, which acted as conductive bridge to reconnect two broken ends of CNTs upon contact, for the self-healing of the electrical properties. Besides, the mechanical strength of the resulting fibers was maintained respectively at 79.1% and 72.5% after one and five cycles of breaking/healing processes. The self-healing fiber with good recovery of electrical and mechanical properties holds great potential for wire-shaped electronic devices. As a demonstration, the self-healing core–sheath fibers were utilized to prepare wire-shaped supercapacitors (Figure 3a). With two self-healing fibers twisted as electrodes, the resulting wire-shaped supercapacitor exhibited a maximal specific capacitance of 11.1 F g<sup>-1</sup> and 82.6% of the capacitance was regained after five breaking/healing cycles. In order to further improve the electrochemical performance, polyaniline was introduced into the aligned CNT sheets as a second active phase to fabricate an asymmetric supercapacitor, whose specific capacitance of 140.0 F g<sup>-1</sup> was maintained at 92% after self-healing.

However, the above-mentioned self-healable supercapacitors have employed self-healing polymers as an extra layer, which will reduce the volumetric/mass capacitance since the self-healing polymers are electrochemically inactive. Given that the electrolyte is indispensable, a new strategy to prepare self-healable devices is to use novel hydrogel electrolytes that are both self-healable and ionically conductive.<sup>[86–91]</sup> For example, a novel electrolyte comprising poly(acrylic acid) dual-crosslinked by hydrogen bonding and vinyl hybrid silica nanoparticles was prepared, which possessed a high self-healability, stretchability, and tunable ionic conductivity.<sup>[92]</sup> This dual-crosslinked poly-electrolyte was able to heal wounds at room temperature and recover most of its original ionic conductivity after multiple breaking/healing cycles. Supercapacitors prepared with this self-healable electrolyte were able to retain the capacitance completely even after 20 cycles of breaking/healing cycles, outperforming all other reported self-healable devices.

The self-healing function is also highly desired for LIBs, which are one of main power sources for portable electronics due to their desirable energy density. Compared with self-healing supercapacitors, it is much more difficult to endow LIBs with self-healing ability since most of the materials used are vulnerable to ambient conditions (e.g., water, oxygen, etc.). Additionally, conventional organic electrolytes would



**Figure 3.** a) Schematic illustration of self-healing fiber-shaped supercapacitor. Reproduced with permission.<sup>[85]</sup> Copyright 2014, Wiley-VCH. b) Schematic illustration of self-healing aqueous lithium-ion battery. c) Electrical resistance of the self-healing electrode during a full cutting-healing cycle. d) Stress-strain curves of the self-healing electrode before and after the cutting-healing process. e) Galvanostatic charge-discharge curves of the aqueous lithium-ion battery after different numbers of self-healing cycles at a current density of  $0.5 \text{ A g}^{-1}$ . b-e) Reproduced with permission.<sup>[93]</sup> Copyright 2016, Wiley-VCH.

decompose rapidly in air once the batteries are damaged, which further increases the complexity to achieve the self-healing function of LIBs. Therefore, it is of both scientific and technological importance to design appropriate electrodes and battery structures that can self-heal after mechanical damage. Toward this end, a new family of all-solid-state and flexible aqueous LIBs with self-healing function were achieved.<sup>[93]</sup> The self-healing electrodes were fabricated by employing aligned CNT sheets loaded with LiMn<sub>2</sub>O<sub>4</sub> and LiTi<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub> nanoparticles on a self-healing polymer substrate (Figure 3b). Once the electrode was cut, the polymer substrates could be self-healed through hydrogen bonding, while aligned-CNT sheets attached onto the substrate could also be reconnected by van der Waals forces to recover the electrical conductivity of electrode. The self-healing electrodes demonstrated good electrical and mechanical self-healing performance, with a slight increase of electrical resistance by 2.3% and tensile strength maintained at 91.8% after five cutting-healing cycles (Figure 3c,d). The self-healing aqueous LIB was finally fabricated by pairing the self-healing cathode and anode with the aqueous lithium sulfate/sodium carboxymethylcellulose (Li<sub>2</sub>SO<sub>4</sub>/CMC) as both gel electrolyte and separator between them. Compared with the combustible and toxic organic electrolytes that will immediately decompose upon exposure to the air, the aqueous Li<sub>2</sub>SO<sub>4</sub> gel electrolyte is safe and nontoxic with a high stability in air. After being cut into two separate parts, the aqueous LIB could be healed to recover the normal functionality by simply bringing into contact the two parts for a few seconds. The specific capacitance of the aqueous LIBs could be restored to 61% after five breaking/healing cycles with good maintenance of the mechanical properties of

the whole device (Figure 3e). This self-healing aqueous LIB has paved the way for the development of self-healing energy-storage devices based on Li<sup>+</sup> insertion/extraction.

## 5. Conclusions and Outlook

To meet the growing requirements for portable and wearable energy-storage devices, tremendous efforts have been not only devoted to developing miniaturized but high-performance energy-storage devices, but also endow them with additional smart functions. As summarized here, the field of integrating electrochromic, shape-memory, and self-healing functions into miniature energy-storage devices has witnessed rapid development and remarkable achievements in the past few years. Smart functional materials, the strategies of device integration for each function, as well as the resulting performance in terms of their energy storage and additional smart functions are reviewed.

Although promising advances have been made for fabricating functional energy-storage devices, there is still a long way to go to further improve their performance for next-generation multifunctional electronics. Some important challenges that need to be overcome in future studies remain:

- i) Integrating appropriate functional materials to achieve optimized performance of energy storage and specific function is necessary. It is still challenging to find smart functional materials that not only are physically and chemically compatible with the devices, but also beneficial for improving

the electrochemical performance of devices. For the aforementioned functional energy devices, the basic energy-storage function and additional smart functions could not both be improved, or need to be compromised to reach a balance in some circumstances. Thus, developing novel functional materials, or optimizing the nanostructure/composition of existing functional materials is highly desired. Moreover, further optimization of the device integration is also needed for better utilization of these functional materials.

- ii) Introducing two or more additional functions into one energy-storage device is of great significance to satisfy the demands for various conditions. Compared with using several functional devices, developing multifunctional devices can effectively reduce the component amount with a smaller size. Nevertheless, this strategy would make the integrated energy devices more complex, which requires more efforts to optimize their structure and performance.
- iii) To date, most of the aforementioned functional energy devices are still limited to laboratory research, and cannot realize large-scale production. Developing and optimizing continuous methods to fabricate functional energy devices with low cost is important for practical applications.

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## Conflict of Interest

The authors declare no conflict of interest.

## Keywords

color change, energy storage, multifunction, self-healing, shape memory

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