A Self-Healing Flexible Quasi-Solid Zinc-Ion Battery Using All-In-One Electrodes

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Self-healing and flexibility are significant for many emerging applications of secondary batteries, which have attracted broad attention. Herein, a self-healing flexible quasi-solid Zn-ion battery composing of flexible all-in-one cathode (VS$_2$ nanosheets growing on carbon cloth) and anode (electro-chemically deposited Zn nanowires), and a self-healing hydrogel electrolyte, is presented. The free-standing all-in-one electrodes enable a high capacity and robust structure during flexible transformation of the battery, and the hydrogel electrolyte possesses a good self-healing performance. The presented battery remains as a high retention potential even after healing from being cut into six pieces. When bending at 60°, 90°, and 180°, the battery capacities remain 124, 125, and 114 mAh g$^{-1}$, respectively, cycling at a current density of 50 mA g$^{-1}$. Moreover, after cutting and healing twice, the battery still delivers a stable capacity, indicating a potential use of self-healing and wearable electronics.

Introduction

Self-healing technology is developed rapidly in recent years, and has been applied in several fields including biological medicines, sensors, surface sciences, and intelligent functional materials. It realizes an automatic self-repair function without manual operation, thereby improving the stability and long cycle life, as well as greatly reducing the maintenance cost. Recently, self-healing has also become a hotspot for emerging energy storage systems. Many self-healing batteries have been reported, such as the self-healing Li-ion, Na-ion, and Mg-ion batteries. For example, Zhao et al. reported a Li-ion battery by using aligned carbon nanotube sheets loaded with LiMn$_2$O$_4$ and LiTi$_2$(PO$_4$)$_3$ nanoparticles on a self-healing polymer as electrode, while lithium sulfate/carboxymethylcellulose (Li$_2$SO$_4$/CMC) gel as both electrolyte and separator. The self-healing performance was ascribed to the reconnection of hydrogen bonds and the van der Waals force on the surface of the breaking place. The battery delivered a capacity that decreased from 28.2 to 17.2 mAh g$^{-1}$ at the rate of 50 mA g$^{-1}$ after fifth cutting/self-healing cycle. Zhong et al. reported a quasi-solid-state Na-ion battery by using polyacrylamide (PAM) hydrogel as the electrolyte, which provided N and H atoms to form hydrogen bonds. Besides, self-healing Mg-ion battery was also successfully constructed. Among many self-healing batteries, there are very few studies on the aqueous Zn-ion battery with a self-healing function. Zn-ion batteries possess some attractive features such as high capacity, high abundance of Zn and low cost, high conductivity, low redox potential (−0.762 V vs standard hydrogen electrode), good safety, which enable them to be applied broadly such as in portable and wearable electronics, and large-scale energy storage.

Many previous self-healing investigations focus on the synthesis of self-healing electrolytes. For instance, Wang et al. prepared a PAM hydrogel and immersed it in a solution containing ZnSO$_4$ and MnSO$_4$ to get a hydrogel electrolyte, which was used in a self-healing Zn-ion battery. After cutting/healing, the tensile stress of the hydrogel could maintain 66% and the Young’s modulus was slightly raised on the basis of the hydrogen bonds between carboxyl groups. The δ-MnO$_2$ cathode-based battery exhibited a capacity from 122 to 108 mAh g$^{-1}$ after 100 cycles at a rate of 10 C. Moreover, Huang et al. prepared a poly(vinyl alcohol)[PVA]/Zn(CF$_3$SO$_3$)$_2$ composite as the self-healing electrolyte. The large amount of hydroxide (−OH) in PVA chains was able to form hydrogen bonds. After cutting/self-healing four times, the battery delivered a capacity of 81.4 mAh g$^{-1}$. Currently, it remains a big challenge to achieve a high self-healing performance with compatible electrolyte and electrode systems.
Moreover, flexible batteries are of great significance to expand the applications of energy storage systems, which can be used widely in flexible electronics including wearable devices and robots. Some flexible substrates were employed as the current collectors. For example, Mo et al. deposited Ge- and N-doped graphene on porous Ni foam to form an anode for Li-ion batteries. Some other electrodes based on the substrates of carbon cloth (CC)/carbon fibers/stainless-steel mesh/polypropylene membrane also displayed a flexible performance and maintained a good electrochemical performance after bending, twisting, or folding. Recently, constructing an emerging self-healing flexible battery becomes a great interest. Once the battery is broken, it is able to repair spontaneously and keep a good flexibility simultaneously. Recently, a Zn-ion battery reported by Lu et al. was able to recover after deformation on the basis of the reversible ionic bonds and hydrogen bonds between two polymers of gelatin and sodium alginate. By bending the battery into diameters of 10 and 30 mm, their V_2O_5/carbon nanotube batteries remained 86% and 89% of the initial capacity, respectively. Li et al. reported a Zn–MnO_2 battery in which the electrolyte was carboxyl-modified PVA. The self-healing mechanism of the hydrogel was ascribed to the COO–Fe bonds. Ma et al. reported an all-solid-state Zn-ion battery in which poly(vinylidene fluoride–hexafluoropropylene) (PVDF–HFP) and poly(ethylene oxide) (PEO) co-polymer provided hydrogen bonds to enhance the flexibility. As seen from these achievements, it is promising to construct a high-performance self-healing flexible Zn-ion battery by using hydrogel electrolyte and all-in-one 3D electrodes, which have been rarely studied.

Here, in order to develop safe, portable, and wearable secondary battery systems, we present a novel self-healing and flexible Zn-ion battery composing of PVA/Zn(CH_3COO)_2/Mn(CH_3COO)_2 (named as PVA–Zn/Mn) hydrogel as electrolyte, VS_2 nanosheets growing on CC as cathode, and Zn-nanowire-deposited CC as anode, as illustrated in Figure 1a. The PVA–Zn/Mn hydrogel electrolyte possesses a large amount of –OH group on the PVA chains, which can easily form hydrogen bonds to achieve a self-healing function. Dense VS_2 nanosheets were in situ grown on CC through a hydrothermal method, forming a free-standing cathode, while Zn anode was prepared by the electrochemical deposition of Zn nanowires on CC. The presented Zn-ion battery displays both flexible and self-healing properties. Furthermore, even though the battery was cut into six pieces, it can be healed quickly with a slight decay of the overall battery potential.

Results and Discussion

VS_2 nanosheets were hydrothermally grown on the conductive and flexible CC current collector, forming a free-standing...
all-in-one VS$_2$/CC cathode. The scanning electron microscopy (SEM) images of the VS$_2$/CC cathode are shown in Figure 1b,c. The surface of the carbon fibers is covered by dense VS$_2$ nanosheets, displaying a loading of about 15.1 mg cm$^{-2}$. In Figure S1a (Supporting Information), the thickness of each nanosheet is about 180 nm. The transmission electron microscopy (TEM) images are displayed in Figure S1b,c (Supporting Information) and the high-resolution TEM (HRTEM) image of a piece of VS$_2$ (Figure 1d) shows a lattice spacing of 0.25 nm, corresponding to the (102) plane of the hexagonal phase of VS$_2$. The selected-area electron diffraction (SAED) pattern (Figure S1d, Supporting Information) displays a series of concentric rings which are indexed to the (001), (101), (102), and (110) planes of VS$_2$, indicating a polycrystalline structure. The diffraction peaks in the X-ray diffraction (XRD) pattern (Figure S1e, Supporting Information) are indexed to the hexagonal VS$_2$, indicating a polycrystalline structure. The diffrac-tion peaks in the X-ray diffraction (XRD) pattern (Figure S1e, Supporting Information) are indexed to the hexagonal VS$_2$, indicating a polycrystalline structure. The diffraction peaks in the X-ray diffraction (XRD) pattern (Figure S1e, Supporting Information) are indexed to the hexagonal VS$_2$, indicating a polycrystalline structure. The diffraction peaks in the X-ray diffraction (XRD) pattern (Figure S1e, Supporting Information) are indexed to the hexagonal VS$_2$, indicating a polycrystalline structure.

The observation toward the cut sections indicate a similar morphology as the one before cutting (Figure 1e,f). It is found that two pieces adhere tightly. The tensile tests were conducted under a series of loading weights from 100 to 500 g, as displayed in Figure 2c. For example, at a loading of 500 g, the healed hydrogel keeps well. The self-healing mechanism is ascribed to the formation of abundant hydrogen bonds between the hydroxyls in the PVA chains. Since that, the self-healing performance would be affected mainly by the density of functional groups and the thickness of the film. In order to improve the self-healing performance, improving the functional groups within the hydrogel electrolyte and optimizing the thickness of hydrogel film would be valuable.

To evaluate the self-healing properties of the Zn-ion battery composing of the hydrogel electrolyte, VS$_2$/CC cathode, and Zn/CC anode, a fully charged battery with the potential of 0.901 V was cut into different pieces. In the first, second, and third demonstrations, the battery was vertically cut, while the fourth time, the battery was horizontally cut. In Figure 2d, it is found that potentials of the battery remain 0.893, 0.891, 0.848, and 0.801 V after each cut/self-healing process. The potential retention is as high as 88.9% after each time of self-healing. In addition, two batteries were connected in series to lighten a light-emitting diode (LED), as displayed in Figure 2e. The LED was lighted even after four times of the cut/healing process, indicating a promising potential for practical self-healing applications.

Figure 3a,b presents the cycling performance of the self-healing Zn-ion battery at a current density of 200 mA g$^{-1}$. The plateaus at 0.7 and 0.6 V in discharge and the ones at 0.7 and 0.75 V during charge well match the redox peaks in cyclic voltammetry (CV) curves (Figure 3c), respectively. The discharge capacities at the 1st, 2nd, 3rd, 10th, and 40th cycles are 175, 161, 161, 157, and 123 mAh g$^{-1}$, respectively. The initial Coulombic efficiency is 68%, which is attributed to the formation of solid-electrolyte interface (SEI) film, while it becomes as high as 99.7% after 40 cycles. The capacity fade is ascribed to the passivation of Zn anode[23] and the formation of Zn(OH)$_2^{2-}$ and ZnO, which further prevent the reactions with Zn. [24] The SEM images of the postcycled cathode verify a robust structure, as shown in Figure S6 (Supporting Information). In addition, Figure S7a,b
Figure 2. a) Photos of the PVA-Zn/Mn hydrogel electrolyte: i) before and ii) after being stretched. b) Self-healing demonstration: i) two pieces of hydrogels with different colors; ii) two half pieces; iii, iv) the two pieces contact for iii) 0 min and iv) 30 min. c) Weight-bearing tests of the healed hydrogel with a series of loading weights of i) 100, ii) 200, iii) 300, iv) 400, and v) 500 g. d, e) Photographs of the self-healing flexible Zn-ion battery after first, second, third, and fourth times of cutting and self-healing: d) battery potentials and e) LEDs lightened by the battery after each time of the self-healing process.
Figure 3. Electrochemical performance of the Zn-ion batteries. a) Galvanostatic charge–discharge curves and b) cycling performance at 200 mA g\(^{-1}\). c) CV curves at 0.1 mV s\(^{-1}\). d) Rate performance. e) Ex situ XRD patterns of the VS\(_2\)/CC cathode when charging–discharging at various potentials. f) Corresponding charge–discharge profile illustrates each potential position. g) CV curves at a series of rates from 0.1 to 1.0 mV s\(^{-1}\). h) The fitted lines of log(i) versus log(v) plots for different peaks. i) Ratios of the capacitive to diffusion-controlled contributions. j) Nyquist plots of the battery within one charge–discharge cycle. The inset shows the equivalent circuit used for fitting.
The reaction in charge and discharge processes is presented in Figure 3c, resulting in that the XRD pattern at 1.0 V can be indexed to the (001), (100), (101), (102), and (110) planes of VS₂. It is noted that there is a slightly negative shift, which is ascribed to the gradual Zn²⁺ insertion in the interlayer spacing of VS₂, forming a VS₂/ZnVS₂. At 0.4 V, some new peaks at 17.3°, 27.5°, 28.3°, 42.3°, 44.4°, and 51.9° appear as marked by black squares, which are attributed to the transformation of VS₂, with continuous insertion of Zn²⁺ ions. The V(IV) is reduced depending on the insertion of Zn²⁺ ions, which leads to the phase transformation to Zn₂(VS₂). In the subsequent charging process, these peaks become weak and finally disappear at 1.0 V. Most of the V₃S₄ returns to VS₂, as displayed in Figure S8 (Supporting Information). Figure 3e shows the CV curves at the first ten cycles in the potential range of 1.0–0.4 V versus Zn/Zn²⁺ at a scanning rate of 0.1 mV s⁻¹. Two reduction peaks at 0.58 and 0.68 V correspond to the Zn²⁺ insertion, while the anodic peaks at 0.72 and 0.8 V are assigned to the Zn²⁺ extraction. Figure 3d displays the rate performance at different current densities. The capacities are 162, 152, 125, 87, and 75 mA h g⁻¹ at 50, 100, 200, 500, and 1000 mA g⁻¹, respectively. The capacity recovers to 146 mA h g⁻¹ when the rate is back to 50 mA g⁻¹. The capacity decay between the first and the second time cycling at 50 mA g⁻¹ is 9.8%.

Figure 3e displays the ex-situ XRD patterns of the all-in-one VS₂/CC cathode at the selected potentials indicated in Figure 3f. During the discharge from 0.7 to 0.6, 0.5, and to 0.4 V, the peaks located at 15.4°, 32.1°, 35.7°, 45.2°, and 57.1° correspond to the (001), (100), (101), (102), and (110) planes of VS₂. It is noted that there is a slightly negative shift, which is ascribed to the gradual Zn²⁺ insertion in the interlayer spacing of VS₂, forming a Zn₂(VS₂). At 0.4 V, some new peaks at 17.3°, 27.5°, 28.3°, 42.3°, 44.4°, and 51.9° appear as marked by black squares, which are attributed to the transformation of VS₂, with continuous insertion of Zn²⁺ ions. The V(IV) is reduced depending on the insertion of Zn²⁺ ions, which leads to the phase transformation to Zn₂(VS₂). In the subsequent charging process, these peaks become weak and finally disappear at 1.0 V. Most of the V₃S₄ returns to VS₂, as displayed in Figure 3f.

The CV curves at various scan rates from 0.1 to 1.0 mV s⁻¹ are displayed in Figure 3g. The shape of the CV profiles keeps close along with a slightly shifting of each peak, which is ascribed to the polarization under the increasing rates. The capacitive and diffusion-controlled behaviors are investigated by equation \( i = a v^b \), where \( a \) and \( b \) are the adjustable parameters. It is transformed to equation \( \log(i) = \log(a) + b \log(v) \), where \( \log(b) \) is determined by the slope of \( \log(i) \) and \( \log(v) \). When \( b = 0.5 \), it is related to a diffusion-controlled process, whereas \( b = 1 \) reflects a capacitive behavior.[26] In Figure 3h, the \( b \) values of peaks 1, 2, 3, and 4 are 0.438, 0.310, 0.466, and 0.529, respectively, indicating both capacitive and diffusion-controlled behaviors of the battery. Moreover, the contribution ratios at various sweep rates can be calculated by \( k_1 i_1 \) and \( k_2 i_1^{1/2} \) according to the equation \( i(V) = k_1 i_1 + k_2 i_1^{1/2} \) and \( i(V) i_1^{1/2} = k_1 i_1^{1/2} + k_2 i_1 \), where \( k_1 \) and \( k_2 \) are constants. The \( k_1 \) and \( k_2 \) can be calculated by plotting the fitting lines of \( i(V) i_1^{1/2} \) and \( k_2 i_1 \). Since that, the capacitive contribution \( (k_1) \) and diffusion-controlled contribution \( (k_2 i_1^{1/2}) \) at various scan rates are obtained, as shown in Figure 3i. The capacitive contribution increases with the increase of scanning rates. The electrochemical impedance spectra (EIS) at a series of potentials within one cycle are shown in Figure 3j. The fresh cell was discharged from 0.9 to 0.4 V, and then charged back to 1.0 V. It exhibits an increasing trend of charge-transfer resistance when discharging to 0.4 V. The fitted resistances are shown in Table S1 (Supporting Information). The oblique line disappears at the discharge potential of 0.6 V. It is ascribed to the gradually formation of sulfur, which reduces the conductivity. The subsequent charging processes show a decreasing trend of charge-transfer resistance, which is attributed to the decrease of sulfur component.

The self-healing flexible battery before and after bending to 60°, 90°, and 180° was also cycled at a current density of 50 mA g⁻¹, as illustrated in Figure 4a. The corresponding performance is shown in Figure 4b,c. The battery without bending and bending at 60°, 90°, and 180° deliver capacities of 165, 180, 169, and 156 mA h g⁻¹, which remain 123, 110, 121, and 106 mA h g⁻¹ after 30 cycles, respectively. The decay with the increase of bending angle is attributed to the compressive and shear stresses on the battery system, which would impact the interfacial contact and properties.[28] It indicates the good flexibility of the fabricated battery in addition to the self-healing property, indicating its promising application in wearable electronics, self-healing intelligent devices, etc. In addition, the cutting and healing performances were also conducted during the cyclic process. In Figure 4d, the battery before cutting was initially cycled ten times. During the cutting, the electrodes and hydrogel were separated into two pieces. Then the two pieces were put together for contact, and the overall battery was packaged again by an adhesive polypropylene film. During the cutting process, the PVA chains would break and some free-moving PVA chain segments would expose on the fresh fracture surface. After the recontact (healing), the PVA chains would attract each other by hydrogen bonds, reforming a hydrogen bonding,[5] which makes the two pieces of hydrogel heal as a whole film again, as illustrated in Figure S10 (Supporting Information). In addition, the separated electrodes recontact by the adhesive effect of electrolyte. In our study, the self-healed battery was placed for 6 h before subsequent cycling tests. The corresponding cycling performance is displayed in Figure 4e. The fresh cell exhibits a capacity of 158 mA h g⁻¹ in the 10th cycle, and soon decays after the first cut-healing treatment. However, the capacity gradually recovers back to 142 mA h g⁻¹ in the 19th cycle, which is ascribed to the accomplished self-healing. The second time of cutting–healing was conducted at the 37th cycle. It shows almost no capacity at the next five cycles. Nevertheless, the capacity recovers back to 112 mA h g⁻¹, which is close to the first-round cutting–healing process. At last, the battery can be cycled normally after the cutting–healing treatments, indicating a potential for applications in many fields.

In summary, we present a self-healing flexible aqueous Zn-ion battery, which consists of a PVA–Zn/Mn electrolyte, all-in-one 3D VS₂/CC cathode, and Zn/CC anode. The constructed Zn-ion battery possesses both flexible and self-healing performances. After
Figure 4. a) Photos of the self-healing flexible Zn-ion battery after bending to different angles: i) no bending, ii) 60°, iii) 90°, and iv) 180°. b) Galvanostatic charge–discharge profiles of the battery under different bending states after cycling 30 times at 50 mA g\(^{-1}\). The inset shows the illustration of the bending angle. c–i) Corresponding discharge capacities of the batteries. d) Photo of the battery after cutting and healing and e) the corresponding cycling performance at 50 mA g\(^{-1}\).

cycling at a rate of 200 mA g\(^{-1}\) 40 times, the battery exhibits a stable capacity of 123 mAh g\(^{-1}\). After bending for 60°, 90°, and 180°, the battery capacities remain 110, 121, and 106 mAh g\(^{-1}\) after 30 cycles. Moreover, even though the battery was cut into six pieces, it was able to self-heal quickly with only a little decay of the overall potential of the battery, indicating a good self-healing stability. During charge–discharge, the battery was cut into two pieces; after self-healing, it delivered a recoverable capacity compared to the one before cutting. It is expected that the presented self-healing flexible Zn-ion battery could find important
applications for flexible electronics and self-healing supplies. Of course, the cycling performance at a high current density still needs further investigation. It could be enhanced by improving the Zn-ion concentration and ionic conductivity of the hydrogel electrolyte, which would have some interesting directions in future. Moreover, the all-in-one electrodes including both anode and cathode would be applicable for constructing a broad set of other emerging energy-storage systems.

**Experimental Section**

**Preparation of the Electrodes:** At first, a free-standing VS2 nanosheets/CC cathode was prepared through a solvothermal approach. All chemicals were analytical reagent grade purchased from Aladdin and used without purification. Typically, 3.6 mmol of NH$_4$VO$_3$ and 27 mmol of thiaacetamide were dissolved in 30 ml of deionized water, followed by the addition of 3.6 ml of NH$_3·$H$_2$O (37 wt%). After continuously stirring for 1 h, a piece of CC was immersed in the solution under ultrasonication, then the mixture was poured into a polytetrafluoroethylene (PTFE)-lined stainless-steel autoclave and sealed. The autoclave was placed in an oven at 180 °C for 8 h. After that, the CC growing with VS2 nanosheets was collected and washed by water and ethanol then dried at 60 °C under vacuum. The VS2 loading of the cathode was calculated by accurately weighing the mass of CC before and after VS2 growth.

**Preparation of the Electrolyte** was prepared by dissolving 13 g of PVA in 50 ml of deionized water then slowly magnetic stirring at 85 °C for 1 h, the sticky solution was ultrasonicated, and the solution was poured to a Petri dish and placed at room temperature overnight, forming a PVA–Zn/Mn hydrogel electrolyte. For comparison, pure PVA hydrogel was prepared by dissolving 13 g of PVA in 50 ml of deionized water then slowly stirred at 85 °C for 1 h.

**Characterization:** The morphology, structure, and composition were characterized by using SEM (Hitachi S-8100, operated at 5 kV), TEM, Hitachi HT7700), HRTEM (Tecnai G220STWIN/FEI), and XRD (Bruker D8 Advance) with the Cu Kα radiation at a wavelength of 1.5418 Å. XPS (ES-CALAB 250) was used to analyze the composition. The hydrogels were treated by liquid nitrogen before freeze-drying for 48 h to obtain the porous structure, then FTIR spectroscopy (IR-21IR-21) and Raman spectrometry (Renishaw, inVia) were used to study the groups of the PVA–Zn/Mn and pure PVA hydrogels.

**Battery Construction and Electrochemical Tests:** To study the flexibility and cutting/self-healing performances, the quasi-solid Zn-ion batteries were assembled by using the as-prepared PVA–Zn/Mn hydrogel electrolyte, VS2/CC cathode, and Zn/CC anode. The battery was packaged by adhesive polypropylene film. All procedures were carried out in an air surrounding. The bending angles were 60°, 90°, and 180°. The potentialsofthe batteries were measured by using a digital LCD display multimeter (Elecro, EM33D) before and after cutting/healing process. The photographs of a light emitting diode (LED) were taken after cutting and healing. In addition, for comparison, a conventional 2032-type coin cell was also assembled by using the prepared electrodes and a glass fiber separator. The electrolyte was prepared by dissolving 21.95 g of Zn(CH$_2$COO)$_2$·2H$_2$O and 2.45 g of Mn(CH$_2$COO)$_2$·4H$_2$O in 100 ml of water. The galvanostatic charge–discharge was conducted on a battery tester (Neware, CT-4008). The capacities were measured at current densities of 50 and 200 mA g$^{-1}$, respectively. The rate performance was tested at 50, 100, 200, 500, and 1000 mA g$^{-1}$ then back to 50 mA g$^{-1}$. Both the CV and EIS were measured on an electrochemical workstation (Chenhua, CHI-660E).

**Statistical Analysis:** The illustration models were constructed on a Maya software (Autodesk, San Rafael, CA, USA). The images were processed on a Fireworks software (Adobe, San Jose, CA, USA). The XRD, XPS, HRTEM, Raman, FTIR, and EIS spectra were analyzed on Jade software (Materials Data, Livermore, CA, USA), Peak Fit software (Systat software, Inc, San Jose, CA, USA), Digital Micrography software (Gatan Inc, Pleasanton, CA, USA), Labspec software (Horiba, Kyoto, JP), OMNIC (Thermo Fisher Scientific Inc, Waltham, MA, USA), and ZView (Scribner Associates, Inc, Charlotteville, VA, USA), respectively. Results were analyzed on an OriginPro software (Origin Lab, Northampton, MA, USA).

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

Research data are not shared.

**Keywords**

all-in-one, flexible, hydrogel electrolyte, self-healing, Zn-ion battery

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