



Electro-induced interfacial enrichment polymerization enables flexible fiber zinc batteries with robust electrode-electrolyte interface and ultrahigh zinc utilization

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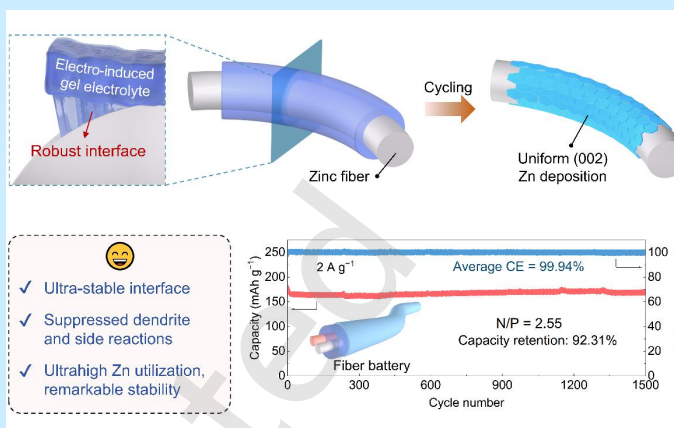
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A controllable electro-induced interfacial enrichment polymerization strategy is designed to construct highly robust fiber electrode-gel electrolyte interfaces, enabling flexible aqueous fiber Zn batteries with high electrochemical performance and stability.

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ABSTRACT

Flexible aqueous fiber zinc batteries are recognized to be promising for flexible and wearable electronics due to both high safety and energy-storage capability. However, their applications have long been hindered by poor fiber electrode-gel electrolyte interfaces and extremely low zinc utilization. Here we report a highly controllable one-step electro-induced interfacial enrichment polymerization strategy to simultaneously address these challenges. The in-situ hybrid polymer hydrogel electrolyte is directly constructed on the fiber electrode surface via synchronously electro-initiated interfacial free-radical polymerization and ionic crosslinking. The rational interface design not only establishes exceptionally robust fiber electrode-gel electrolyte interfaces via strong interfacial chemical bonding, but also guides preferential deposition of Zn (002) plane and suppresses side reactions and dendrite growth. Consequently, even under an ultrahigh zinc utilization of 85.4%, the in-situ fiber Zn//Zn cells exhibit an extended cycling life exceeding 300 hours, significantly outperforming previously reported fiber Zn//Zn cells. Furthermore, at a low N/P ratio of 2.55, the highly flexible aqueous Zn//polyaniline fiber batteries demonstrate a high capacity retention of 92.31% after 1500 cycles with an average coulombic efficiency of 99.94%. This work presents a general and effective strategy to construct robust electrode-gel electrolyte interfaces for high-performance flexible and wearable energy storage devices.

KEYWORDS

Flexible zinc batteries, Interfacial enrichment polymerization, Electrode-electrolyte interface, in-situ hydrogel electrolyte, High zinc utilization

1 Introduction

Flexible batteries that combine intrinsic safety and high flexibility are widely regarded as promising candidates for the next-generation power sources in smart wearable systems [1-3]. Replacing conventional flammable and toxic organic electrolytes with safe and non-toxic aqueous electrolytes is therefore a key pathway toward the above practical applications [4, 5]. In particular, aqueous fiber zinc batteries emerge as a compelling option owing to their inherent safety, high theoretical capacity, environmental benignity, and superior omnidirectional flexibility [6, 7]. However, they have been severely hindered by persistent challenges, including uncontrolled

zinc dendrite growth [8, 9], parasitic side reactions [10, 11], and limited zinc utilization [12]. To address these critical limitations, extensive research efforts have been devoted to rational zinc anode design [13-15], separator modification [16, 17], electrolyte additive engineering [18], and polymer gel electrolyte design [19, 20]. Among them, the use of gel electrolytes proves particularly effective in mitigating zinc anode-related issues. Furthermore, compared to their liquid counterparts, polymer gel electrolytes endow fiber zinc batteries with superior leakage resistance and mechanical stability [21], thereby holding great promise for next-generation flexible

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and wearable energy storage systems.

Different from conventional planar electrodes, the high-curvature geometry of fiber electrodes renders them far more difficult to establish mechanically robust and stable interfaces with gel electrolytes [22], particularly under the complex mechanical deformations during operations. Unfortunately, the poor fiber electrode-gel electrolyte interface in such fiber batteries often results in severe interfacial separation during prolonged cycling or repeated bending [23, 24], thereby leading to increased interfacial impedance, voltage polarization, and even battery failure [25, 26]. More importantly, the unstable interface accelerates localized Zn dendrite nucleation and parasitic side reactions [27, 28], which in turn cause rapid degradation of electrochemical performance including

shortened cycling lifespan and reduced coulombic efficiency [29], especially under high zinc utilization (i.e., depth of discharge, DOD). Unfortunately, most reported fiber zinc batteries were operated at extremely low Zn utilization (usually <15%) to maintain cycling stability [30, 31], which inevitably sacrifices their practical energy densities and severely hinders their large-scale applications. Therefore, it is imperative to develop robust and strongly bonded fiber electrode-gel electrolyte interfaces that can simultaneously suppress zinc dendrite growth and undesirable side reactions to achieve long-term stability even under high Zn utilization, thereby enabling highly stable, intrinsically safe, and high-performance fiber zinc batteries.

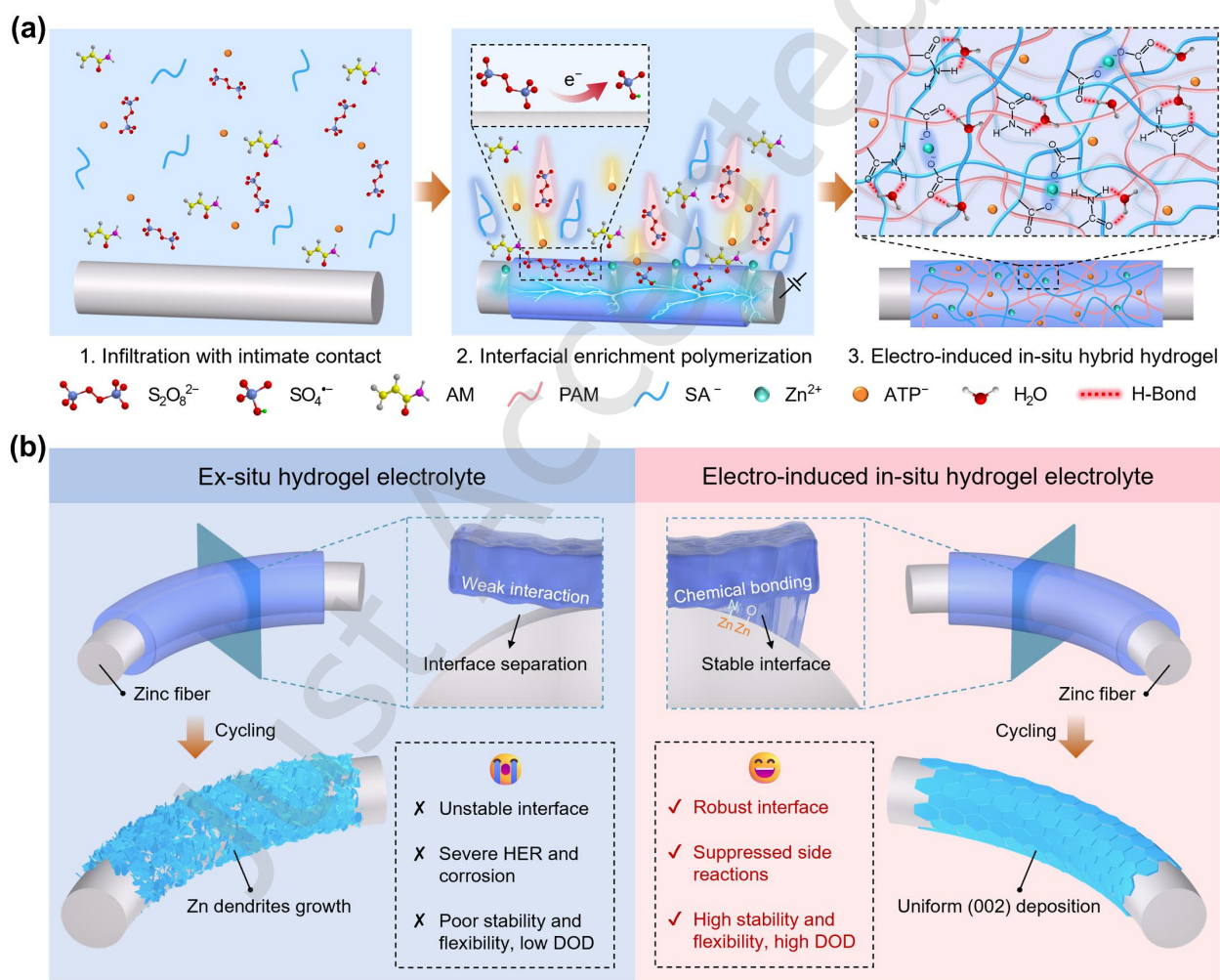


Figure 1 Polymerization mechanism and advantages of the electro-induced in-situ hybrid polymer hydrogel electrolyte. (a) Schematic illustration of the electro-induced interfacial enrichment polymerization mechanism for constructing robust in-situ hybrid hydrogel electrolyte directly on the fiber electrode. (b) Comparison of the fiber electrode-gel electrolyte interfaces between conventional ex-situ and electro-induced in-situ hybrid hydrogel electrolytes.

Herein, a one-step electro-induced interfacial enrichment polymerization strategy is proposed to achieve intimate and robust interfaces between high-curvature fiber electrode and hydrogel electrolyte (Fig. 1(a)). Under electro-induced localized enrichment effect, the in-situ hybrid polymer hydrogel electrolyte was

directly synthesized on the fiber electrode surface by concurrently initiated interfacial free-radical polymerization of acrylamide (AM) monomers and ionic crosslinking of sodium alginate (SA) triggered by electric current. Particularly, the interfacial stability between fiber electrode and hydrogel electrolyte is substantially

enhanced owing to the unique interfacial enrichment effect before in-situ polymerization and the formation of strong Zn–N and Zn–O bonding (Fig. 1(b)). Moreover, the dense in-situ hybrid polymer network is enriched with functional groups, which can accelerate Zn^{2+} desolvation, guide the preferential deposition of Zn (002) crystal plane, and reduce the activity of interfacial free H_2O molecules, thereby effectively suppressing dendrite growth and undesirable side reactions. Compared with conventional preparation methods, our electro-induced interfacial enrichment polymerization offers markedly improved controllability and has been demonstrated to be crucial for achieving outstanding interfacial stability, electrochemical performance and flexibility for aqueous fiber Zn batteries. Consequently, both Zn//Zn and Zn//polyaniline (PANI) fiber cells exhibit superb electrochemical performance even under high Zn utilization. This work establishes a viable avenue for developing next-generation flexible batteries with robust electrode-gel electrolyte interfaces and enhanced electrochemical performance.

2 Results and discussion

A simple and highly controllable electro-induced interfacial enrichment polymerization was developed to construct robust and tightly bonded interfaces between flexible fiber electrode and hybrid hydrogel electrolyte. Initially, the precursor solution with low viscosity was allowed to thoroughly infiltrate the surface of fiber electrode to achieve intimate contact. When an appropriate electric current was applied, the electric field induced a pronounced localized enrichment of charged species on the surface of fiber electrode. Simultaneously, the interfacial free-radical polymerization coupled with ionic crosslinking was triggered to fabricate a robust in-situ hybrid hydrogel directly on the fiber electrode (Fig. S1 in the Electronic Supplementary Material (ESM)). The reaction mechanism can be described as follows: the zinc electrode released Zn^{2+} ions under appropriate current. Meanwhile, the anions of $\text{S}_2\text{O}_8^{2-}$, $(\text{C}_6\text{H}_7\text{O}_6^-)_n$ of SA^- and attapulgite (ATP^-) migrate toward the fiber electrode surface under the electric field. With the generated Zn^{2+} , the SA hydrogel network is constructed on fiber electrode surface through the complexation between Zn^{2+} and $-\text{COO}^-$ via interfacial ionic coordination and crosslinking. Simultaneously, the gathered $\text{S}_2\text{O}_8^{2-}$ on fiber Zn surface were reduced to reactive $\text{SO}_4^{\cdot-}$ radicals, which synchronously initiate the interfacial free-radical polymerization of acrylamide (AM) monomers, thereby forming an interconnected PAM-SA-ATP hybrid polymer hydrogel network that is firmly anchored on the fiber electrode surface.

The polymerization reactions were initiated at tightly coupled interface between the fiber electrode and infiltrated liquid precursor, ensuring intimate interfacial contact and chemical interaction. The resulting in-situ hybrid hydrogel electrolyte exhibited a strong interfacial

interaction with the fiber electrode (Fig. 2(a)), which is crucial for constructing stable fiber electrode-gel electrolyte interface. More importantly, compared with thermal-induced in-situ hydrogel electrolyte, our electro-induced enrichment polymerization strategy constructed a markedly denser and gradient hybrid polymer network within the interfacial region adjacent to the fiber electrode (Fig. 2(b)), which is due to the migration and interfacial enrichment of anions (e.g., ATP^- and SA^-) during the polymerization process (Fig. S2 in the ESM). In contrast, the regular thermal-induced in-situ hydrogel electrolyte was uniform and showed no enrichment phenomenon on the fiber electrode (Fig. 2(c)). Additionally, as shown in the time-dependent interfacial composition analysis, the continuous blue shift of the C–O stretching vibration was ascribed to the progressive enrichment of SA^- during the electro-induced polymerization (Fig. 2(d)), which further demonstrated the interfacial enrichment behavior [32]. The denser hydrogel network of electro-induced in-situ hybrid hydrogel electrolyte on the electrode surface was also verified by the much lower electric double-layer capacitance (Fig. S3 in the ESM). Consequently, the electro-induced in-situ hydrogel electrolyte demonstrated substantially higher peeling force and shear strength than its thermal-induced in-situ counterpart (Fig. S4 in the ESM), which is derived from the stronger interfacial interactions offered by the compact layer.

Benefited from the high controllability of interfacial polymerization reaction, the loading amount of the electro-induced in-situ hybrid hydrogel electrolyte on fiber electrode could be readily tuned over a wide range (Fig. S5 in the ESM). The diameter of fiber electrode coated with hybrid hydrogel electrolyte gradually increased from 300 to 1534 μm as the applying time of electric current increased from 0 to 150 s (Fig. S6 in the ESM). The successful preparation of hybrid hydrogel electrolyte was demonstrated by the characteristic peaks of C=O (1656.8 cm^{-1}), N–H (1597.9 cm^{-1}) and C–O (1033.7 cm^{-1}) from fourier-transform infrared (FTIR) spectroscopy (Fig. S7 in the ESM) [33]. The morphology of the in-situ and ex-situ electrolytes was further characterized using scanning electron microscopy (SEM). The freeze-dried in-situ hybrid hydrogel revealed a continuous and interconnected porous architecture (Fig. S8 in the ESM), which facilitates the electrolyte accommodation, enhances the ion transport, and alleviates mechanical stress.

Importantly, a robust interfacial interaction between the fiber electrode and electro-induced in-situ hydrogel electrolyte was well preserved even after repeated bending deformations (Fig. 2(e)). In contrast, the ex-situ electrolyte exhibited pronounced interfacial gaps with the flexible electrode (Figs. 2(f) and S9 in the ESM). The interfacial interaction between the flexible Zn electrode and different hydrogel electrolytes was quantitatively evaluated through peeling-force and shear-strength

measurements (Fig. 2(g)). Specifically, the in-situ hybrid hydrogel electrolyte demonstrated an exceptional peeling force of up to 1081 N m^{-1} , nearly 34 times higher than that of ex-situ hydrogel electrolyte (32 N m^{-1}). The in-situ hybrid hydrogel electrolyte also delivered a shear strength of up to 124.4 kPa , surpassing the ex-situ electrolyte (12.3 kPa). The strong peeling force and high shear strength ensured a robust interface between fiber electrode and hydrogel electrolyte, which is essential for flexible batteries subjected to various mechanical deformations [34, 35]. The key interfacial chemical interaction between the in-situ hybrid hydrogel electrolyte and Zn electrode was

revealed by X-ray photoelectron spectroscopy (XPS) analysis, elucidating the underlying mechanisms responsible for the enhanced interfacial interactions and stability. High-resolution XPS spectra displayed characteristic peaks corresponding to amide and carboxyl groups, while distinct Zn–N and Zn–O bonding signals were clearly identified at 399.8 eV in N 1s and 533.0 eV in O 1s regions (Figs. 2(h–k)) [36], respectively. The interfacial bonding ensured strong interactions between the fiber Zn electrode and in-situ hybrid hydrogel electrolyte, enabling a robust electrode-electrolyte interface.

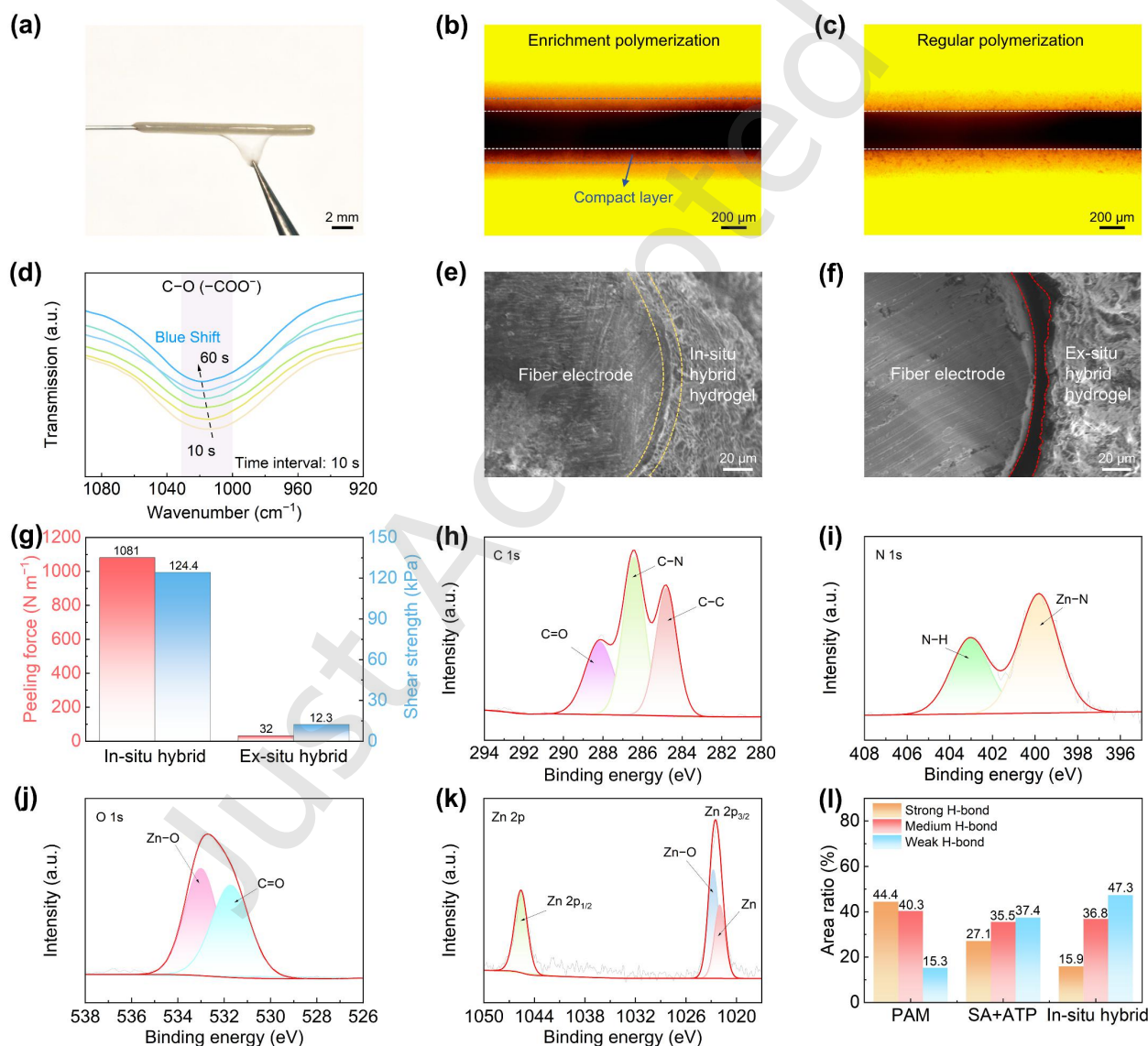


Figure 2 Characterization and interfacial interactions between fiber electrode and different hydrogel electrolytes. (a) Optical photograph of the electro-induced in-situ hybrid hydrogel electrolyte under pulling, exhibiting strong interfacial interactions with the fiber electrode. (b, c) Optical microscope images of the fiber electrodes covered with in-situ hydrogel electrolytes prepared by electro-induced interfacial enrichment polymerization and regular thermal-induced polymerization, respectively. (d) FTIR spectra of the electro-induced in-situ hydrogel electrolytes at the electrode interface after polymerization for different durations. (e, f) Cross-sectional SEM images of the fiber electrode-gel electrolyte interfaces for electro-induced in-situ and ex-situ hybrid hydrogel electrolytes after bending deformations, respectively. (g) Peeling force and shear strength of the in-situ and ex-situ hybrid hydrogel electrolytes on Zn electrodes. (h–k) High-resolution XPS spectra of C 1s (h), N 1s (i), O

1s (j), and Zn 2p (k) for the Zn electrode with in-situ hybrid hydrogel electrolyte. (l) Proportions of strong, medium, and weak hydrogen-bond types in different hydrogel electrolytes obtained from FTIR spectra.

The in-situ hybrid hydrogel electrolyte exhibited a much higher ionic conductivity (18 mS cm^{-1}) than other hydrogel electrolytes (Fig. S10 in the ESM), which was attributed to the interactions between Zn^{2+} and -COO^- that alter the solvation structure of Zn^{2+} . Both the hydrophilic groups such as amide bonds in PAM and the ion-guidance function of ATP can promote the transport of hydrated Zn^{2+} . Particularly, the in-situ hybrid hydrogel electrolyte exhibited a high Zn^{2+} transference number ($t_{\text{Zn}^{2+}}$) of 0.70 (Fig. S11 in the ESM), which was beneficial to reduce the overpotential and enhance the electrochemical performance of the battery. The water environment of different hydrogel electrolytes was investigated by the FTIR spectroscopy (Fig. S12 in the ESM), which provided direct evidence of electrolyte environment reconstruction (Fig. 2(l)). Notably, the in-situ hybrid hydrogel electrolyte exhibited a progressively decreased ratio of strong H-bond (15.9%) compared with PAM (44.4%) and SA+ATP (27.1%) hydrogel electrolytes, indicating the reduced water activity to suppress hydrogen evolution and Zn corrosion side reactions. In addition, the in-situ hybrid hydrogel electrolyte showed a broadened electrochemical stability window from the linear sweep voltammetry (LSV) measurements (Fig. S13 in the ESM), which is attributable to the suppressed activity of free H_2O molecules [37].

The Zn deposition behavior of fiber Zn//Zn symmetric cells (Fig. S14 in the ESM) employing the in-situ and ex-situ hydrogel electrolytes exhibited distinct differences, as revealed by the morphological characterization of Zn anodes. The pristine Zn electrode displayed a heterogeneous surface with inherent defects (Fig. S15a in the ESM). In contrast, the surface of fiber Zn electrode became quite smooth after the applying of electric current during hydrogel synthesis process (Fig. S15b in the ESM), which was favorable for the uniform deposition of Zn metal. After cycling, the fiber Zn electrode employing electro-induced in-situ hybrid hydrogel electrolyte exhibited a much smoother surface compared with those using other electrolytes (Fig. S16 in the ESM). In comparison, numerous randomly distributed Zn dendrites were observed on the surface of Zn anode with ex-situ electrolytes (Fig. 3(a)). Notably, the Zn plating layer formed in electro-induced in-situ hybrid hydrogel electrolyte was composed of compact and densely packed flakes after the same cycling (Fig. 3(b)). This observation confirms that the electro-induced interfacial polymerized electrolyte establishes a highly uniform ion transport environment and a robust electrode-electrolyte interface, thereby enabling homogeneous Zn nucleation and suppressing dendrite growth.

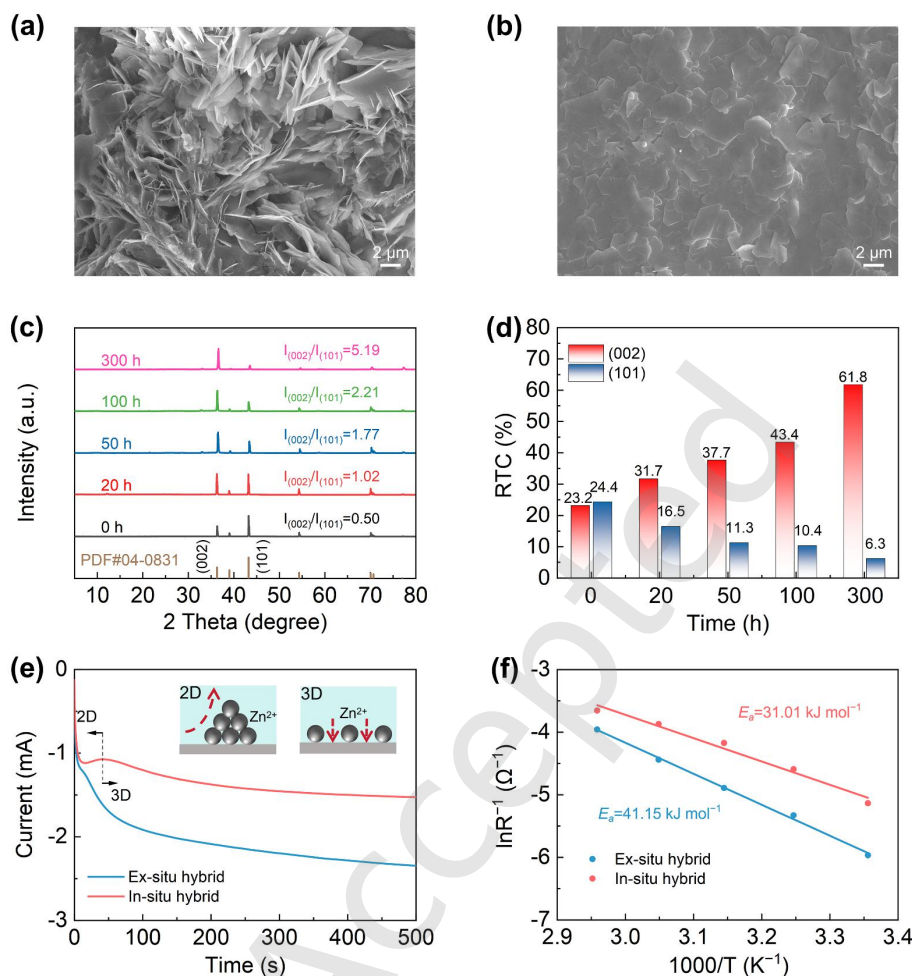


Figure 3 Characterization of the fiber Zn anodes using different hybrid hydrogel electrolytes. (a, b) SEM images of fiber Zn anodes after cycling in ex-situ and electro-induced in-situ hybrid hydrogel electrolytes, respectively. (c) XRD patterns of Zn anodes cycled for various durations at 1 mA cm^{-2} and 1 mAh cm^{-2} using the in-situ hybrid hydrogel electrolyte. (d) Calculated RTC values of different crystal planes after cycling for various durations with in-situ hybrid hydrogel electrolyte. (e) CA curves of fiber Zn//Zn symmetric cells with ex-situ and in-situ hybrid electrolytes at an overpotential of -150 mV. (f) Calculated desolvation energy barriers of Zn//Zn cells with different hybrid electrolytes.

The evolution of zinc deposition configuration during cycling was characterized by X-ray diffraction (XRD), revealing a distinct crystallographic transition process (Fig. 3(c)). The diffraction peaks observed at 36.3° and 43.2° were assigned to the (002) and (101) planes, respectively. During the initial 20 h of cycling, the (101) plane dominated the zinc anode, exhibiting a stronger diffraction intensity than other crystal planes. However, prolonged cycling induced a gradual transition in the preferred orientation, and the (002) plane progressively intensified and eventually became dominant. The evolution of crystallographic orientation during cycling was further quantified using relative texture coefficient (RTC) [38], calculated by comparing the XRD peak intensities of the (*hkl*) crystal planes to those of a standard reference sample (Table S1). The quantitative RTC analysis revealed a pronounced crystallographic evolution during the cycling of Zn//Zn cells with electro-induced in-situ hybrid hydrogel electrolyte (Fig. 3(d)). The RTC of Zn (002) plane exhibited a steady

increase from 23.2% to 61.8%, while the (101) plane's RTC correspondingly decreased from 24.4% to 6.3%. Therefore, the in-situ hybrid hydrogel electrolyte obtained by electro-induced interfacial enrichment polymerization enabled the (002)-textured Zn deposition with suppressed dendrite formation, which is attributed to the intimate interfacial contact between the in-situ hydrogel electrolyte and the fiber electrode, as well as the presence of zincophilic functional groups.

The nucleation behavior of Zn²⁺ in different electrolytes was evaluated by chronoamperometry (CA) measurements (Fig. 3(e)). The ex-situ hydrogel exhibited a rapid current surge within 500 s, indicating a predominant two-dimensional (2D) diffusion process. The 2D diffusion facilitated irregular nucleation and consequently led to uncontrolled growth of zinc dendrites. In contrast, the Zn//Zn symmetric cells employing electro-induced in-situ hydrogel electrolyte exhibited a faster initial 2D diffusion process before reaching a steady-state current, attributed to more electrochemically

active sites, followed by a three-dimensional (3D) diffusion process that favored uniform Zn deposition. To elucidate the reaction kinetics of Zn^{2+} , the interfacial activation energy (E_a) was calculated to evaluate the influence of different electrolytes on the Zn^{2+} desolvation process (Fig. S17 in the ESM). The fiber Zn//Zn cells with in-situ hybrid hydrogel electrolyte exhibited a lower energy barrier compared with those using ex-situ electrolyte (Fig. 3(f)), which effectively enhanced the reaction kinetics. In addition, the electro-induced interfacial enrichment polymerization strategy enabled the hydrogel electrolyte with improved nucleation behavior and a substantially lower desolvation energy barrier compared with its thermal-induced counterpart, thereby effectively inhibiting the dendrite growth (Fig. S18 in the ESM).

The electrochemical performance of the fiber Zn anode was systematically evaluated in various hydrogel electrolytes. Long-term galvanostatic cycling of fiber Zn//Zn cells was conducted at a current density of 1 mA cm^{-2} and an areal capacity of 1 mAh cm^{-2} . The fiber Zn//Zn cells using PAM, SA+ATP and ex-situ hybrid electrolytes exhibited a sudden and irreversible voltage drop or surge within 670 h (Fig. 4(a)). In contrast, the symmetric Zn cells employing electro-induced in-situ hybrid hydrogel electrolyte maintained highly stable cycling for over 2500 h. The enlarged voltage profiles indicated that the Zn//Zn cells using in-situ hybrid hydrogel electrolyte exhibited the smallest voltage hysteresis among various hydrogel electrolytes (Fig. S19 in the ESM). Additionally, the rate performance of the symmetric Zn cells was evaluated at a series of current densities ranging from 1 to 50 mA cm^{-2} with a fixed capacity of 1 mAh cm^{-2} (Fig. 4(b)). The symmetric Zn cells using in-situ hybrid electrolyte operated stably even at a high current density up to 50 mA cm^{-2} and demonstrated a lower voltage hysteresis compared with alternative hydrogel electrolytes.

The cycling performance of fiber Zn//Zn symmetric cells with various hydrogel electrolytes was further evaluated at elevated current densities and areal capacities. At a much higher current density of 30 mA cm^{-2} , the symmetric Zn cells employing in-situ hybrid hydrogel electrolyte exhibited highly stable Zn plating/stripping for over 200 h (Fig. S20 in the ESM), whereas those using ex-situ and other electrolytes suffered from rapid failure and markedly shortened cycle life. In addition, even at a

high capacity of 10 mAh cm^{-2} (10 mA cm^{-2}), the fiber Zn//Zn cells with in-situ hybrid hydrogel electrolyte delivered stable cycling over 612 h (Figs. 4(c) and S21 in the ESM), markedly outperforming those using PAM (4 h), SA+ATP (76 h), and the ex-situ (164 h) hydrogel electrolytes. Particularly, even under an ultrahigh areal capacity of 37.5 mAh cm^{-2} (85.4% Zn utilization), the electro-induced in-situ fiber Zn//Zn cells maintained a stable cycling for over 300 h (Fig. 4(d)), significantly surpassing the counterparts with other electrolytes (less than 50 h). Furthermore, compared with thermal-induced in-situ symmetric Zn cell, the fiber Zn//Zn cell using electro-induced hydrogel electrolyte demonstrated a markedly prolonged cycling lifespan under 85.4% DOD (Fig. S22 in the ESM), highlighting the critical role of interfacial enrichment polymerization.

Compared with previously reported fiber Zn//Zn cells, our electro-induced in-situ Zn cells exhibit outstanding advantages in both Zn utilization and cycling performance (Fig. 4(e) and Table S2 in the ESM) [5-7, 27, 39-45]. The superior electrochemical performance arises from several synergistic factors. First, in contrast to conventional hydrogel electrolytes, the unique electro-induced interfacial enrichment polymerization constructs a much more condensed hybrid polymer network on the fiber electrode surface, thereby strengthening interfacial interaction and yielding a mechanically and electrochemically robust fiber electrode-gel electrolyte interface. Second, the concurrently initiated interfacial free-radical polymerization of AM and ionic crosslinking of SA triggered by electric current establish strong interfacial interactions between the fiber electrode and hydrogel electrolyte via chemical bonding, which effectively restrict the physical space available for dendrite propagation and thus suppress zinc dendrite formation. Third, the incorporation of zinc-affinitive functional groups (e.g., amide bonds, carboxyl groups) within the dense hybrid networks accelerates Zn^{2+} desolvation and guides uniform Zn^{2+} nucleation and deposition, enabling the preferential growth of two-dimensional Zn (002) texture. Finally, the interfacial hydrogen-bonding interactions between specific functional groups of polymers and H_2O molecules effectively reduce the activity of interfacial free water [46, 47], thereby inhibiting hydrogen evolution and corrosion reactions at the fiber electrode-gel electrolyte interface and ultimately prolonging the cycling lifespan of fiber cells.

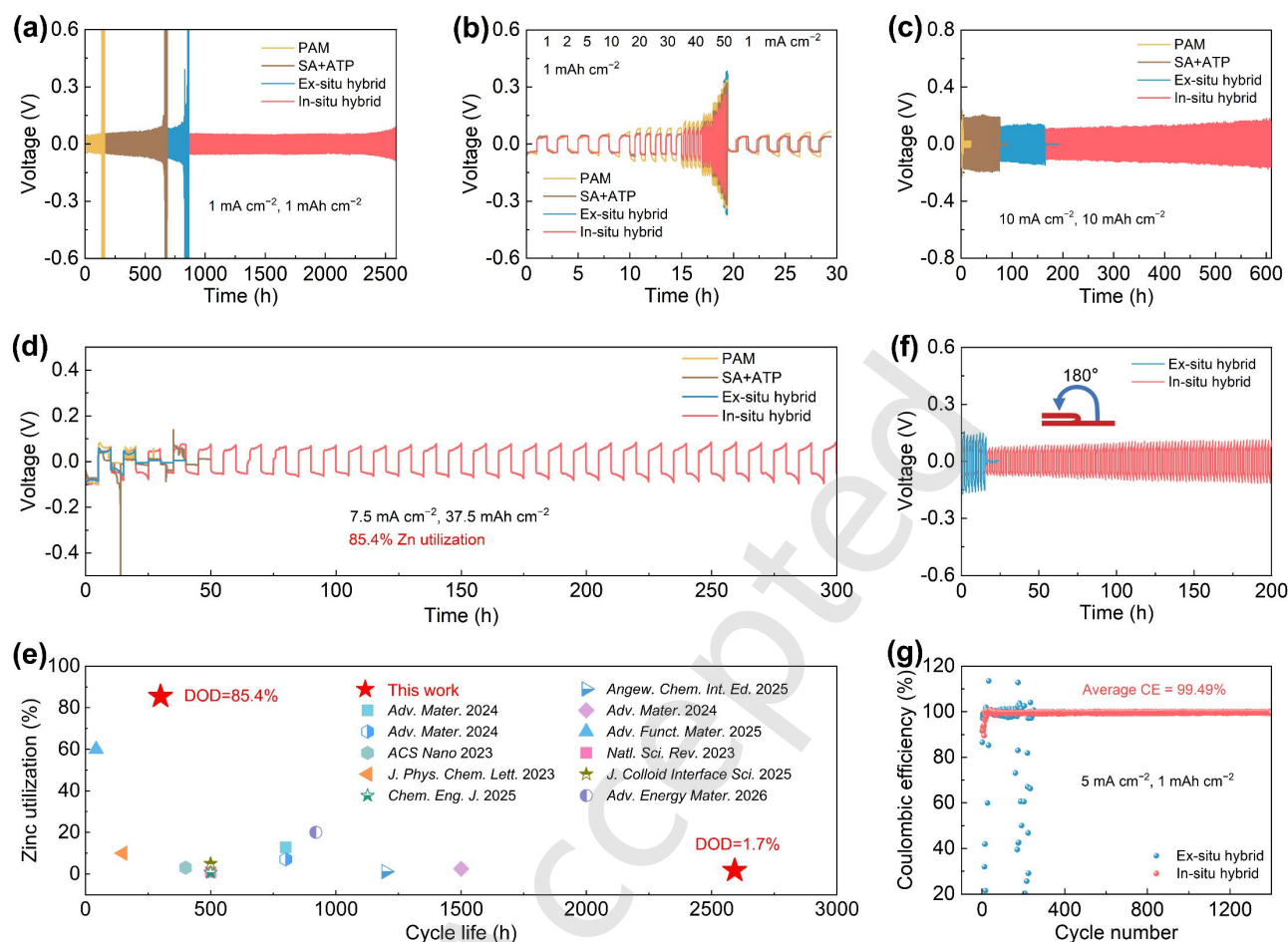


Figure 4. Long-term cycling stability of fiber Zn anodes with different hydrogel electrolytes. (a) Cycling performance of fiber Zn//Zn symmetric cells with various hydrogel electrolytes at 1 mA cm⁻² and 1 mAh cm⁻². (b) Rate performance of Zn//Zn cells with different electrolytes. (c) Cycling performance of fiber Zn//Zn cells at 10 mA cm⁻² and 10 mAh cm⁻². (d) Cycling performance of various fiber Zn//Zn cells under ultrahigh Zn utilization of 85.4%. (e) Comparisons of Zn utilization and cycle life of this work and those of previously reported fiber Zn//Zn cells. (f) Cycling performance of fiber Zn//Zn cells with different hydrogel electrolytes under a bending angle of 180° at 10 mA cm⁻² and 10 mAh cm⁻². (g) Coulombic efficiency of fiber Zn//Cu asymmetric cells with ex-situ and electro-induced in-situ hybrid hydrogel electrolytes at 5 mA cm⁻² and 1 mAh cm⁻².

The interfacial stability between fiber electrode and hydrogel electrolyte plays a pivotal role in achieving high-performance flexible batteries, particularly under complex deformations. As the bending angle increased from 0 to 90°, the cycling life of fiber Zn//Zn cells with ex-situ hybrid hydrogel electrolyte decreased significantly from 139 to 44 h at 10 mA cm⁻² and 10 mAh cm⁻² (Fig. S23 in the ESM), which can be attributed to the interfacial separation between the fiber electrode and ex-situ hydrogel electrolyte. In contrast, the electro-induced in-situ fiber Zn//Zn cells exhibited stable cycling performance under increasing bending angles. Notably, even at a high bending angle of 180°, the electro-induced in-situ symmetric Zn cells still maintained stable cycling over 200 h (Fig. 4(f)), far outperforming their ex-situ and thermal-induced in-situ counterparts (Fig. S24 in the ESM). These results clearly demonstrated the superior interfacial stability between the fiber electrodes and

electro-induced in-situ hybrid hydrogel electrolyte. In addition, as shown in the electrochemical impedance spectroscopy (EIS) curves, the interfacial impedance of the in-situ symmetric Zn cells was much lower than that of the cells employing other electrolytes after cycling (Fig. S25 in the ESM). Consequently, the electro-induced in-situ fiber symmetric Zn cells demonstrated faster ion transport kinetics, stronger interfacial interactions, and enhanced electrochemical stability compared with their ex-situ counterparts.

The improvement of zinc deposition behavior and cycling stability achieved by employing in-situ hybrid hydrogel electrolyte was further confirmed using Zn//Cu asymmetric cells. As shown in cyclic voltammetry (CV) curves, the use of in-situ hybrid hydrogel electrolyte markedly reduced the Zn²⁺ nucleation overpotential by 67 mV compared with the ex-situ electrolyte (Fig. S26 in the ESM), indicating a reduced nucleation energy barrier and

accelerated nucleation kinetics. Consistently, the deposition voltage profile of fiber Zn//Cu cells with in-situ hybrid hydrogel electrolyte also exhibited a much lower nucleation overpotential than those with ex-situ electrolyte (Fig. S27 in the ESM), further corroborating the enhanced reaction kinetics. At 5 mA cm^{-2} and 1 mAh cm^{-2} , the Zn//Cu cells with ex-situ hydrogel electrolyte exhibited sharp fluctuations in coulombic efficiency (CE) and a short cycling life of merely 248 cycles (Fig. 4(g)). In contrast, the fiber Zn//Cu cells employing electro-induced in-situ hydrogel electrolyte demonstrated highly stable cycling for 1400 cycles with an average CE of 99.49%, indicating significantly enhanced interfacial stability and reversibility because of the effective suppression of dendrite growth and parasitic side reactions.

The voltage profiles of the Zn//Cu cells with ex-situ hybrid hydrogel electrolyte exhibited a pronounced increase in overpotential during cycling, accompanied by poor voltage stability (Fig. S28a in the ESM). In contrast, the Zn//Cu cells employing electro-induced in-situ hybrid hydrogel electrolyte displayed remarkably stable and smooth voltage profiles throughout cycling (Fig. S28b in the ESM), indicating that in-situ electrolyte effectively suppressed dendrite formation and mitigated side reactions. Additionally, the corrosion behavior of metallic Zn in different hydrogel electrolytes was investigated using Tafel curves (Fig. S29 in the ESM). A lower corrosion current of 0.129 mA cm^{-2} was achieved for in-situ hybrid hydrogel electrolyte, which was markedly lower than that of PAM (0.249 mA cm^{-2}) and SA+ATP (0.277 A cm^{-2}) electrolytes, validating its superior corrosion inhibition effect on Zn anode.

To verify the practical feasibility of the electro-induced in-situ hybrid hydrogel electrolyte in fiber zinc batteries, carbon fiber (CF)/PANI was selected as the fiber cathode (Fig. S30 in the ESM) to construct aqueous Zn//PANI fiber full cells (Figs. 5(a) and S31 in the ESM). The linear resistances of CF and CF/PANI were 113.6 and $56.8 \Omega \text{ cm}^{-1}$, respectively (Fig. S32 in the ESM). The SEM images of CF/PANI clearly revealed that PANI was uniformly coated on the smooth surface of carbon fibers (Fig. S33 in the ESM), forming a continuous and homogeneous coating. Moreover, the CV curves of the Zn//PANI fiber cells with in-situ hybrid hydrogel electrolyte exhibited highly similar shapes at various scan rates (Fig. S34 in the ESM), suggesting good electrochemical reversibility. The oxidation and reduction peaks shifted toward higher and lower potentials, respectively, due to the enhanced polarization effect. The relationship between peak current (i) and scan rate (v) follows the equation $i = av^b$, where a and b are variable parameters. When the b value approaches 1, the electrochemical process is dominated by pseudocapacitive behavior, whereas a b value close to 0.5 indicates a diffusion-controlled process. The calculated b values were 0.73 and 0.64 (Fig. S35 in the ESM),

suggesting that the charge/discharge reactions in the fiber battery were dominated by pseudocapacitive behavior with fast redox kinetics (Fig. S36 in the ESM). The capacitive contribution of the fiber full cells gradually increased with increasing scan rates, further proving the primary capacitive-controlled behavior at high rates (Fig. S37 in the ESM).

Compared with ex-situ fiber cells, the Zn//PANI fiber cells with electro-induced in-situ hybrid hydrogel electrolyte exhibited a larger enclosed CV area, higher redox peak currents, and reduced peak separation (Fig. 5(b)), indicating an enhanced electrochemically active area, faster reaction kinetics, and improved electrochemical reversibility enabled by the intimate electrode/electrolyte interfacial contact. Moreover, the galvanostatic charge-discharge curves of the in-situ Zn//PANI fiber cells delivered a high reversible specific capacity of $233.86 \text{ mAh g}^{-1}$ at a current density of 0.2 A g^{-1} (Fig. 5(c)), exhibiting a high energy density of 210.6 Wh kg^{-1} . Furthermore, the reversible capacities of the in-situ fiber full cells were consistently higher than ex-situ counterparts across all current densities (Fig. 5(d)), demonstrating markedly enhanced rate capability. Even at a high current density of 10 A g^{-1} , the fiber full cells with in-situ hybrid electrolyte retained a considerable capacity of $135.32 \text{ mAh g}^{-1}$. Additionally, when the current density was reduced back to 0.2 A g^{-1} , the capacity of in-situ fiber cells recovered to $231.89 \text{ mAh g}^{-1}$ (Fig. S38 in the ESM), corresponding to a high capacity retention of 99.2%.

The aqueous Zn//PANI fiber cell with electro-induced in-situ hybrid polymer hydrogel electrolyte exhibited a lower interfacial impedance after 100 charge-discharge cycles (Figs. 5(e) and S39 in the ESM). After cycling, the Warburg factor (σ) of fiber full cells with ex-situ hydrogel electrolyte increased sharply from 851.4 to 1966.2 (Figs. 5(f) and S40 in the ESM), indicating the decrease of ion-diffusion efficiency. In contrast, the Warburg factor of fiber cells employing in-situ hybrid electrolyte increased only slightly from 351.9 to 360.3 after cycling. Therefore, the in-situ fiber cells exhibited faster ionic transport at the electrode-electrolyte interface compared with ex-situ counterparts. Furthermore, the self-discharge behavior of the Zn//PANI fiber cells with in-situ hybrid electrolyte was effectively mitigated (Fig. S41 in the ESM). After resting for 48 h, the in-situ fiber full cells still retained 75.5% of its initial capacity owing to the effective suppression of side reactions, which was notably higher than that of ex-situ fiber cells (58.5%). Furthermore, the use of in-situ hybrid hydrogel electrolyte significantly enhanced the cycling stability of the Zn//PANI fiber cells. Specifically, after 1800 charge-discharge cycles at 5 A g^{-1} , the in-situ fiber cells maintained a high reversible capacity of $150.11 \text{ mAh g}^{-1}$ with 94.28% capacity retention (Fig. S42 in the ESM), far exceeding that of cells using ex-situ hydrogel electrolytes.

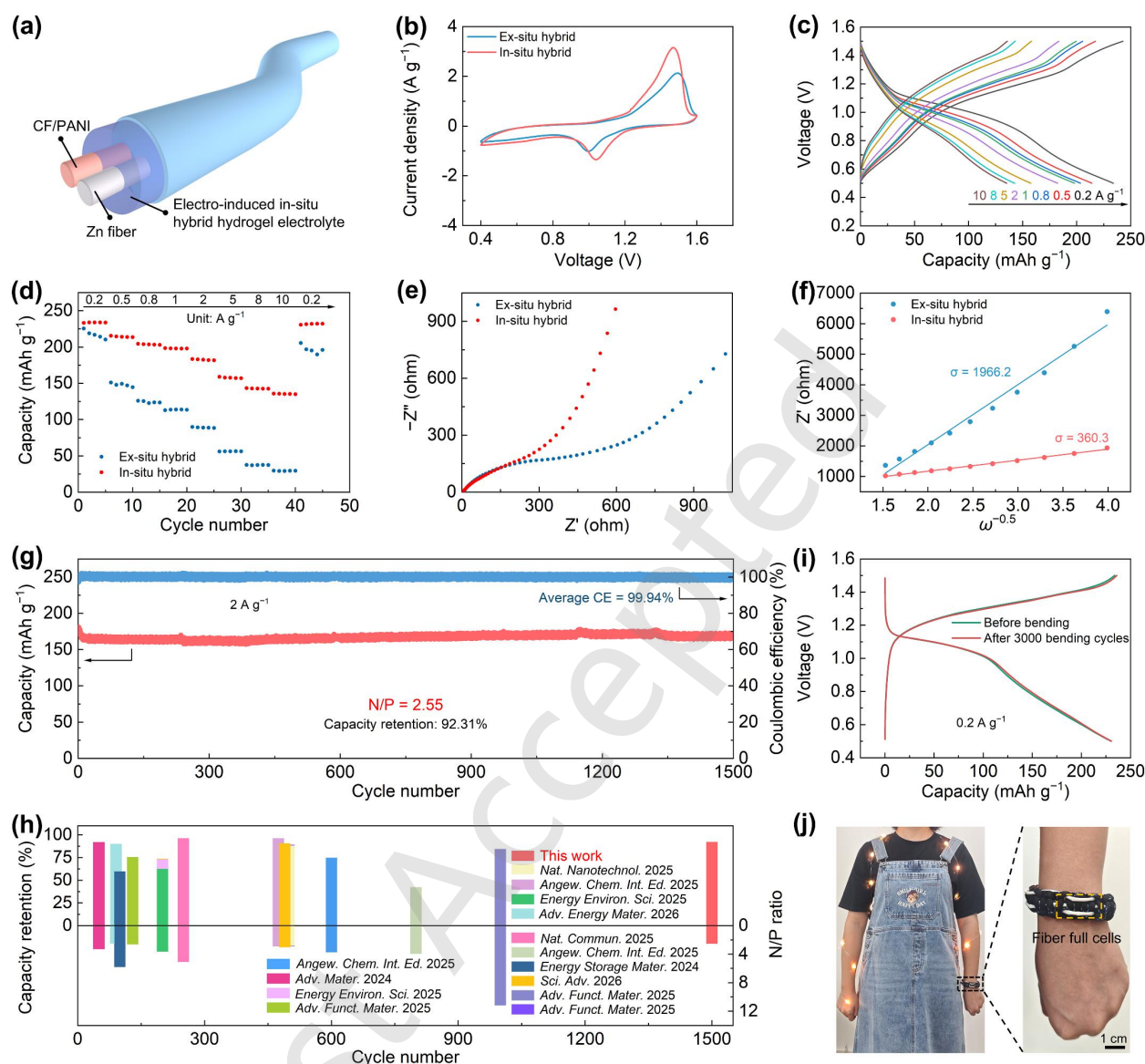


Figure 5. Electrochemical performance of flexible aqueous Zn//PANI fiber cells. (a) Schematic illustration of the Zn//PANI fiber cell employing electro-induced in-situ hybrid hydrogel electrolyte. (b) CV curves of the full cells with ex-situ and in-situ hydrogel electrolytes at a scan rate of 1 mV s⁻¹. (c) Galvanostatic charge-discharge curves at increasing current densities from 0.2 to 10 A g⁻¹. (d) Rate performance of the Zn//PANI fiber cells using different electrolytes. (e) EIS curves of the Zn//PANI fiber cells after 100 cycles. (f) Relationship curves of Z' and $\omega^{-0.5}$ in the diffusion-controlled frequency range after cycling. (g) Long-term cycling performance of the electro-induced in-situ fiber cells at a low N/P ratio of 2.55. (h) Comparisons of N/P ratio and cycling performance of this work and those of previously reported flexible Zn-based full cells. (i) Charge-discharge curves of the flexible fiber cells before and after 3000 bending cycles at a bending angle of 180°. (j) Photographs of the flexible Zn//PANI fiber cells powering a wearable LED strip.

Even at a low N/P ratio of 2.55, the electro-induced in-situ fiber batteries still maintained a high capacity retention of 92.31% after 1500 cycles, with an impressive average coulombic efficiency of 99.94% (Fig. S5g and S43 in the ESM). Compared with previously reported flexible Zn-based full cells [48–61], our in-situ Zn//PANI fiber cells exhibit remarkable advantages in both N/P ratio and long-term durability (Fig. 5(h) and Table S3 in the ESM). Moreover, the electro-induced in-situ fiber batteries demonstrated impressive specific capacity, cycling lifespan, capacity retention rate, and average coulombic efficiency

when compared with the state-of-the-art fiber batteries (Fig. S44 and Table S4 in the ESM). Beyond their exceptional electrochemical performance, the aqueous fiber full cells also demonstrated remarkable mechanical flexibility and structural stability. Even after 3000 bending cycles at a bending angle of 180°, the in-situ fiber full cells retained 99.9% of their initial capacity (Fig. 5(i)), exhibiting superior mechanical robustness. In contrast, the ex-situ fiber cells retained only 77.5% (Fig. S45 in the ESM), highlighting the superior interfacial stability of the in-situ system. Furthermore, the output voltage and

capacity of the device could be readily enhanced by connecting multiple batteries in series or in parallel (Fig. S46 in the ESM). Benefiting from their superior electrochemical performance, high flexibility, and intrinsic safety, the flexible aqueous Zn//PANI fiber batteries hold great promise for wearable energy storage application. As a practical demonstration, a wearable LED strip was continuously powered by the fiber Zn batteries woven into a knitted wristband (Fig. 5(j)), demonstrating their broad potential in next-generation smart and wearable electronics.

3 Conclusion

In summary, we have directly constructed a tightly bonded in-situ hybrid hydrogel electrolyte on high-curvature flexible fiber electrode through a highly controllable, one-step electro-induced interfacial enrichment polymerization. Particularly, the unique design not only effectively enhances the interfacial stability between fiber electrode and hydrogel electrolyte through the interfacial enrichment effect and the formation of strong interfacial chemical bonding, but also promotes the uniform zinc deposition and suppresses side reactions and dendrite growth by specific functional groups in the dense hybrid network. In comparison with regular polymerization approaches, the unique electro-induced interfacial enrichment polymerization has been demonstrated to substantially enhance the interfacial interactions and electrochemical performance of the flexible fiber batteries. Notably, even under an ultrahigh Zn utilization of 85.4%, the electro-induced in-situ hybrid hydrogel electrolyte enables fiber Zn//Zn cells to operate stably for over 300 h. Furthermore, the aqueous Zn//PANI fiber full cells demonstrate a high capacity retention of 92.31% after 1500 cycles at a low N/P ratio of 2.55. These aqueous batteries also demonstrate remarkable flexibility, safety, and wearability. This work offers new insights into the rational design of highly robust electrode-gel electrolyte interfaces for next-generation flexible and wearable electronic devices.

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Declaration of conflicting interests

The authors declare no conflicting interests regarding the content of this article.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and/or the Supplementary

Materials. Additional data related to this paper may be requested from the corresponding author upon request.

Electronic Supplementary Material: Supplementary material (including experiment details and additional characterizations) is available in the online version of this article at <https://doi.org/10.26599/NRE.2026.9120279>.

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Xudong Pei received his Bachelor's degree in 2022 and Master's degree in 2026 from Hubei University. His research interests focus on flexible and wearable aqueous zinc batteries.



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Electronic Supplementary Material

Electro-induced interfacial enrichment polymerization enables flexible fiber zinc batteries with robust electrode-electrolyte interface and ultrahigh zinc utilization

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1. Experimental Section:

1.1 Materials

Sodium alginate (SA, AR), acrylamide (AM, 99.0%), potassium persulfate (KPS, 99.99%), N,N-methylenebisacrylamide (MBA, 99.0%), $\text{ZnSO}_4 \cdot \text{H}_2\text{O}$ (99.9%), and ammonium persulfate (APS, AR) were purchased from Aladdin. Aniline (99.5%), HCl (AR) were purchased from Sinopharm. Attapulgit (ATP) was purchased from Shanghai Yuanye Bio-Technology. All the commercial reagents were used without further purification.

1.2 Material Preparation and Device Assembly

1.2.1 Preparation of Electro-Induced In-Situ Hybrid Hydrogel Electrolyte

0.65 g of SA and 1.32 g of ATP were completely dissolved in 20 mL of deionized water and stirred thoroughly at 70 °C. Subsequently, 0.165 g of AM, 0.825 mg of MBA, and 16 mg of KPS were added and completely dissolved at room temperature to form a precursor solution. Prior to use, the Zn wires were polished with sandpaper and subsequently rinsed with deionized water and ethanol. Using the electro-induced interfacial enrichment polymerization method, the in-situ hybrid hydrogel electrolyte was directly fabricated on the fiber Zn electrode (diameter of 0.3 mm) immersed in the above precursor solution with a two-electrode configuration, where the Zn electrode and stainless steel wire were used as the working electrode and counter electrode, respectively. After applying a constant electric current of 5 mA for a controlled duration (e.g., 10 to 150 s) using the electrochemical workstation (CHI 660E), the in-situ hybrid hydrogel was successfully grown on the fiber electrode. Finally, the fiber electrode covered with hybrid hydrogel was soaked into 2 M ZnSO_4 aqueous solution for 10 minutes to obtain the electro-induced in-situ hybrid hydrogel electrolyte. The suitable thickness of the hydrogel electrolyte on the fiber electrode was approximately 100-300 μm .

1.2.2 Preparation of Ex-Situ and Thermal-Induced In-Situ Hybrid Hydrogel Electrolytes

The in-situ hybrid hydrogel electrolyte was prepared on the surface of a fiber electrode by above electro-induced interfacial enrichment polymerization, which was then removed from the surface of the fiber electrode and immersed in 2 M ZnSO₄ aqueous solution for 10 minutes to obtain the ex-situ hybrid hydrogel electrolyte. When preparing the thermal-induced in-situ hydrogel electrolyte, the precursor solution for electro-induced polymerization was injected into a mold with a fixed fiber electrode, which was heated at 70 °C for 1 h and then immersed in 2 M ZnSO₄ aqueous solution for 10 minutes to obtain the thermal-induced in-situ hybrid hydrogel electrolyte.

1.2.3 Fabrication of Electro-Induced In-Situ Fiber Zn//Zn Symmetric Cells

Two parallel zinc wires were employed as the working electrode for electro-induced synthesis of in-situ hybrid hydrogel using the above-mentioned preparation method, followed by immersing in 2 M ZnSO₄ aqueous solution for 10 minutes. Finally, two fiber Zn electrodes covered with in-situ hybrid hydrogel electrolyte were assembled and sealed in a heat shrinkable tube to fabricate the in-situ fiber Zn//Zn symmetric cells. The thickness of in-situ hydrogel electrolyte layer in the fiber Zn//Zn symmetric cells was approximately 150 μm.

1.2.4 Fabrication of Ex-Situ and Thermal-Induced In-Situ Fiber Zn//Zn Symmetric Cells

Two parallel zinc wires covered with above ex-situ hydrogel electrolyte were assembled and sealed in a heat shrinkable tube to fabricate the ex-situ fiber Zn//Zn symmetric cells. When preparing the thermal-induced in-situ fiber Zn//Zn cells, the above precursor solution was injected into a mold with two fixed zinc wires, which was heated at 70 °C for 1 h and then immersed in 2 M ZnSO₄ aqueous solution for 10 minutes. Finally, the fiber Zn electrodes covered with thermal-induced in-situ hydrogel electrolyte were sealed in a heat shrinkable tube to fabricate the thermal-induced in-situ fiber Zn//Zn cells.

1.2.5 Assembly of Fiber Zn//Cu Asymmetric Cells

A zinc wire and a copper wire were placed in parallel to prepare the electro-induced in-situ hybrid hydrogel electrolyte, which were then assembled and sealed in a heat shrinkable

tube to fabricate the in-situ fiber Zn//Cu asymmetric cell. The ex-situ fiber Zn//Cu asymmetric cell was fabricated with a similar preparation method using ex-situ hydrogel electrolyte.

1.2.6 Synthesis of the Carbon Fiber/PANI Cathode

0.73 mL of aniline monomer was added into 30 mL of 1 M HCl aqueous solution at 0 °C. Carbon fibers were subsequently immersed in the homogeneous solution. After immersing for 1 h, 10 mL of 1 M HCl solution containing 0.456 g of APS was carefully added to the above solution. After reaction for 1 h, the samples were alternately immersed in deionized water and anhydrous ethanol, and then drying at 60 °C overnight in a vacuum oven to obtain the carbon fiber/PANI cathode. The PANI loading on the fiber electrode was determined by mass difference using a high-precision balance.

1.2.7 Fabrication of Aqueous Zn//PANI Fiber Full Cells

The carbon fiber/Zn electrode was prepared via a two-electrode electrodeposition process conducted under galvanostatic conditions, using carbon fiber as the working electrode, zinc wire as the counter electrode, and 2 M ZnSO₄ as the electrolyte. Two parallel carbon fiber/PANI and carbon fiber/Zn fiber electrodes were used as the working electrode to prepare the in-situ hybrid hydrogel electrolyte using the above mentioned electro-induced in-situ synthesis method. Subsequently, two fiber electrodes covered with in-situ hybrid hydrogel were immersed in 2 M ZnSO₄ aqueous solution. Finally, the two fiber electrodes covered with in-situ hybrid hydrogel electrolyte were assembled and sealed by a heat shrinkable tube to obtain the aqueous Zn//PANI fiber full cells. The N/P ratio was calculated by the capacity ratio between the negative and positive fiber electrodes.

2. Electrochemical Measurements and Calculation

The galvanostatic charge-discharge and rate performance tests of the fiber Zn//Zn symmetric cells, fiber Zn//Cu asymmetric cells, and fiber full cells were conducted by the NEWARE battery test system. The electrochemical measurements were performed at an

ambient temperature of 24-26 °C. The cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS), chronoamperometry (CA), Tafel and linear sweep voltammetry (LSV) tests were performed by the electrochemical workstation (CHI 660E). Tafel curves were carried out in a two-electrode setup with a scan rate of 10 mV s⁻¹.

The ionic conductivity (σ) of different hydrogel electrolytes was obtained by the EIS results of the symmetric cells. The electrolyte resistance (R_b) was derived from the EIS curves and used to calculate the ionic conductivity by the following equation:

$$\sigma = \frac{l}{R_b S} \quad (1)$$

where l was the electrolyte thickness, and S was the contact area between the electrolyte and electrode.

The Zn^{2+} transference numbers ($t_{Zn^{2+}}$) were evaluated by combining the chronoamperometry tests with EIS plots in the Zn||Zn symmetric cells with various electrolytes, and calculated by the following equation:

$$t_{Zn^{2+}} = \frac{I_s(\Delta V - I_0 R_0)}{I_0(\Delta V - I_s R_s)} \quad (2)$$

where ΔV is the applied polarization voltage (10 mV), I_0 and R_0 are the initial current and resistance, I_s and R_s are the steady-state current and resistance after polarization.

The relative texture coefficients (RTCs) of different crystal planes were calculated according to the following equation:

$$RTC_{(hkl)} = \frac{I_{(hkl)}/I_{0(hkl)}}{\sum (I_{(hkl)}/I_{0(hkl)})} \times 100\% \quad (3)$$

where $I_{(hkl)}$ is the intensity of the (hkl) plane, and $\sum I_{(hkl)}$ is the sum of the intensity in XRD profile. The $I_{0(hkl)}$ is the (hkl) plane intensity of standard Zn sample.

The discharge of depth (DOD) of the fiber Zn//Zn cells were calculated according to the following equation:

$$DOD = \frac{C_{(actual)}}{C_{(theoretical)}} \times 100\% = \frac{C_{(area)} \times \pi \times d \times l}{C_{(mass)} \times \rho \times \pi \left(\frac{d}{2}\right)^2 \times l} \times 100\% \quad (4)$$

where $C_{(actual)}$ (mAh) is the actual capacity of the deposition/stripping Zn and $C_{(theoretical)}$ (mAh) is the theoretical capacity of the fiber Zn electrode. $C_{(area)}$ is the actual area capacity (mAh cm⁻²), $C_{(mass)}$ (mAh g⁻¹) is the theoretical mass capacity of Zn, d (cm), ρ (g cm⁻³) and l (cm) are the density and length of fiber Zn electrode, respectively. For the fiber Zn electrode with a diameter of 0.3 mm, an area capacity of 37.5 mAh cm⁻² for fiber Zn//Zn cells corresponds to a DOD of 85.4%.

The energy density (E) and power density (P) of the Zn//PANI fiber full cells were calculated according to the following equations:

$$E = \int_{t_1}^{t_2} \frac{I}{m} V dt \quad (5)$$

where I is the discharge current, V is the discharge voltage, t_1 and t_2 are the start and end time of discharge, t is the discharge time, m is the mass of cathode material.

$$P = \frac{E}{\Delta t} \quad (6)$$

where P is the power density, E is the energy density, and Δt is the discharge time.

3. Materials Characterizations

The X-ray diffractometer with Cu K α radiation (XRD, Bruker D8 ADVANCE) was used to explore the crystal structure of the materials. The field emission scanning electron microscopy (FESEM, Zeiss, SIGMA500) was applied to observe the micromorphology of the electrode materials. The X-ray photoelectron spectroscopy (XPS, Thermo Fisher Scientific, ESCALAB 250Xi) was employed to characterize the chemical valence states and elemental composition of the electrode materials. The Fourier-transform infrared (FTIR) spectra were acquired with a Fourier Infrared spectrometer (Thermo Fisher Scientific, Nicolet iS10).

Peeling-force and shear-strength measurements of different hydrogel electrolytes on Zn metal were conducted with a universal tensile testing machine (Instron 68TM-10). The optical microscope images were captured using a metallurgical microscope (M330-3M180).

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Supplementary Figures



Figure S1. Optical photograph of a fiber Zn electrode covered with electro-induced in-situ hybrid hydrogel electrolyte.

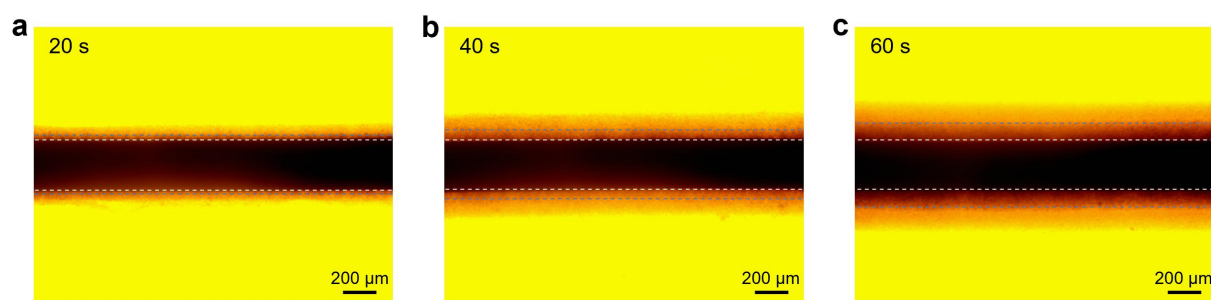


Figure S2. Optical microscope images of the electro-induced in-situ hydrogel electrolytes covered on the fiber electrode after different durations of polymerization, demonstrating the interfacial enrichment process during electro-induced polymerization.

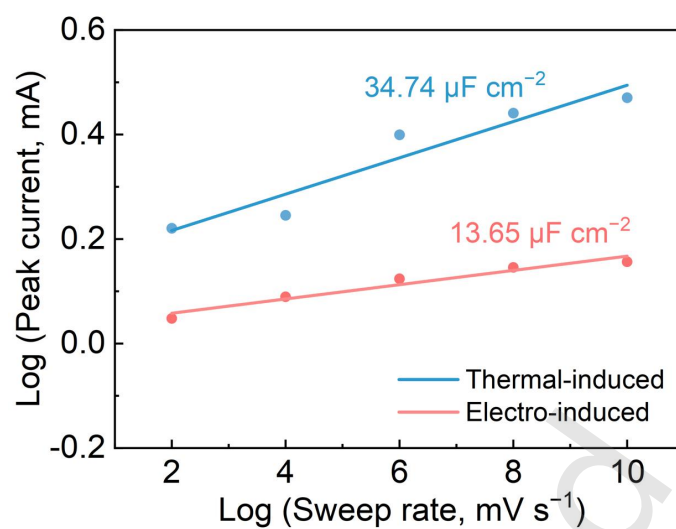


Figure S3. Electric double-layer capacitance of Zn anodes with thermal-induced and electro-induced in-situ hydrogel electrolytes.

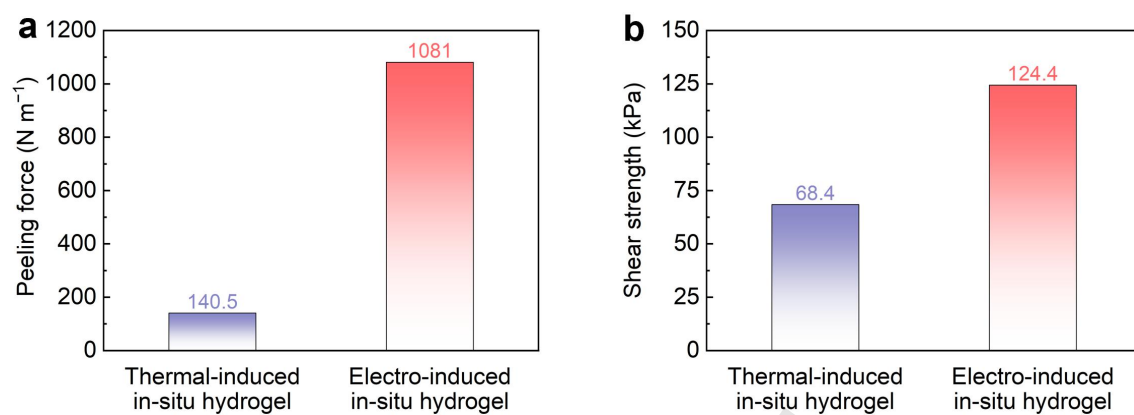


Figure S4. Peeling force and shear strength of the thermal-induced and electro-induced in-situ hybrid hydrogel electrolytes on Zn electrode, respectively.

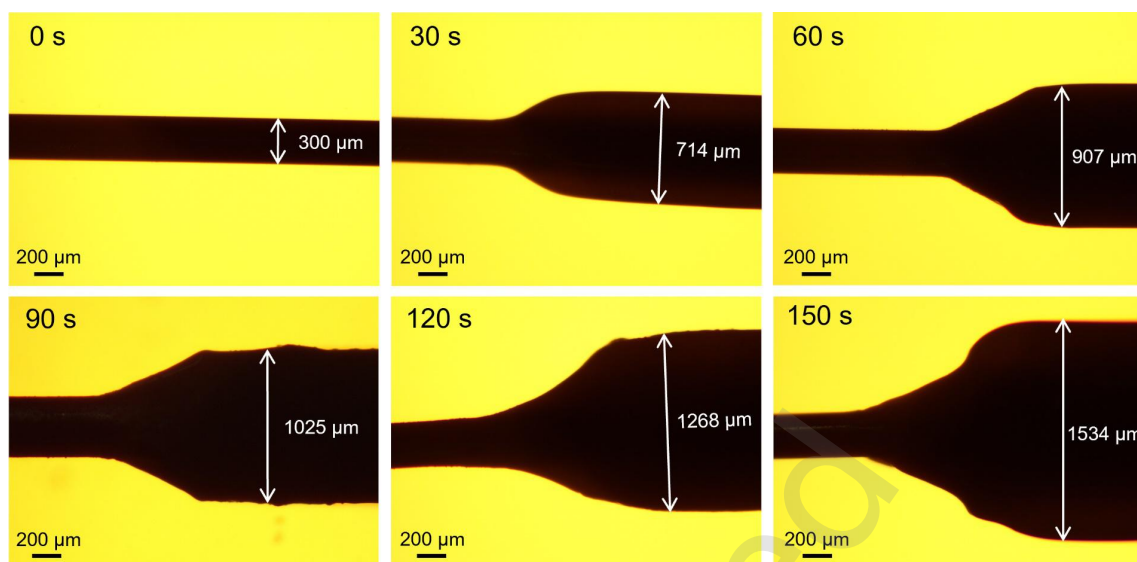


Figure S5. Optical microscope images of the flexible fiber electrodes before and after different durations of electro-induced interfacial enrichment polymerization reaction to prepare the in-situ hybrid hydrogel electrolyte.

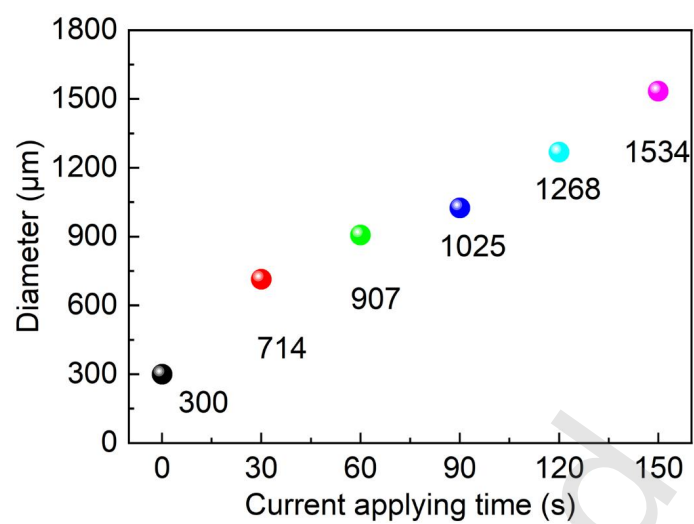


Figure S6. The relationship between the applying time of electric current during polymerization reaction and the diameter of fiber electrode coated with electro-induced in-situ hybrid hydrogel electrolyte.

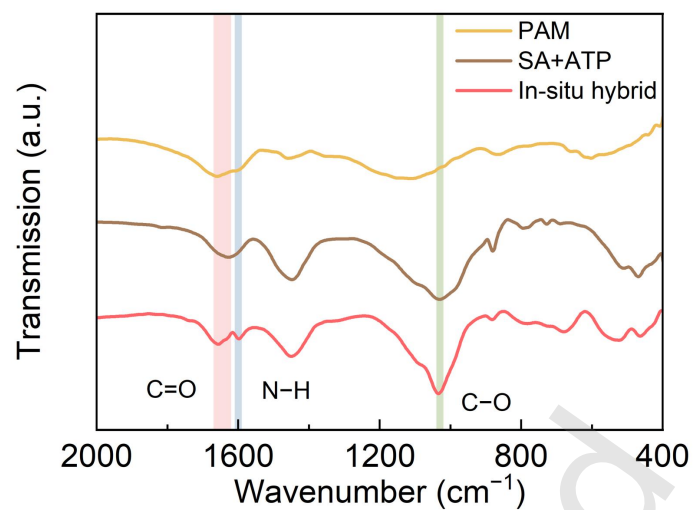


Figure S7. FTIR spectra of different hydrogel electrolytes.

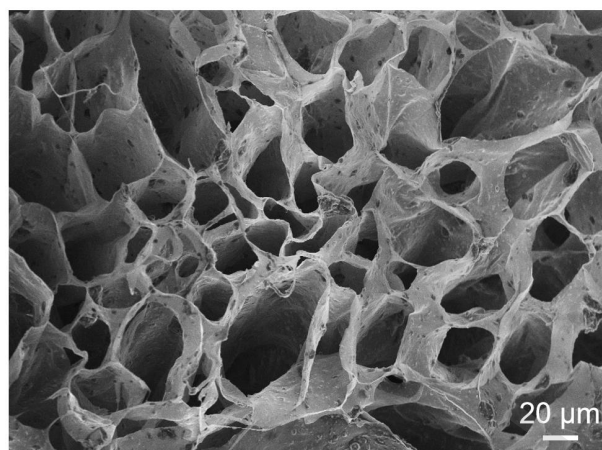


Figure S8. SEM image of the freeze-dried in-situ hybrid hydrogel.

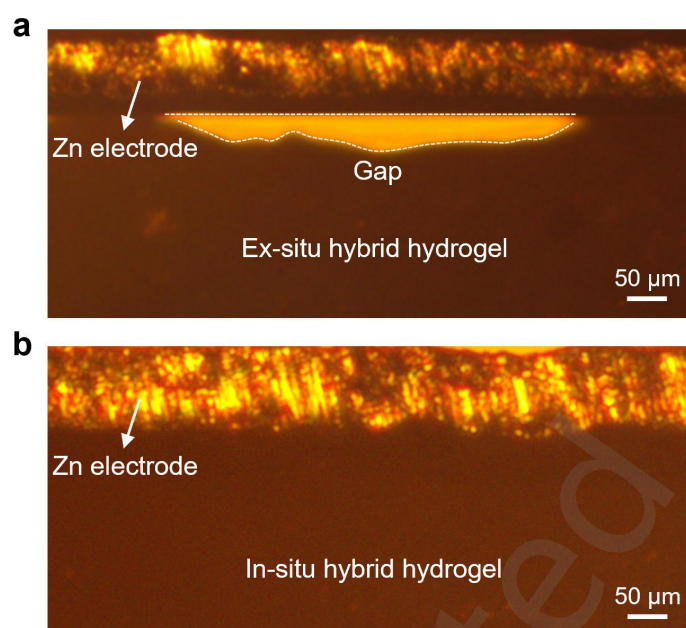


Figure S9. Optical microscope images of the flexible Zn electrodes covered with different hybrid hydrogel electrolytes after repeated bending deformations.

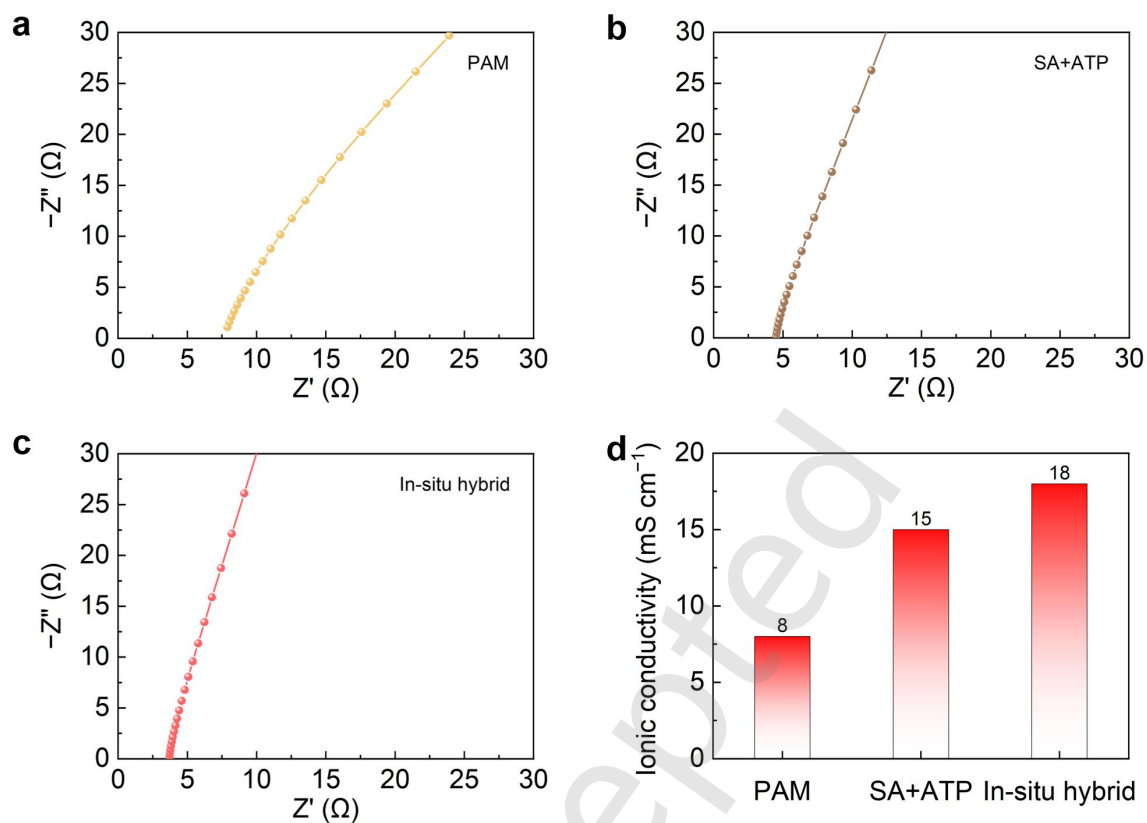


Figure S10. (a-c) EIS curves of the PAM, SA+ATP, and in-situ hybrid hydrogel electrolytes, respectively. (d) Ionic conductivity comparison of various hydrogel electrolytes.

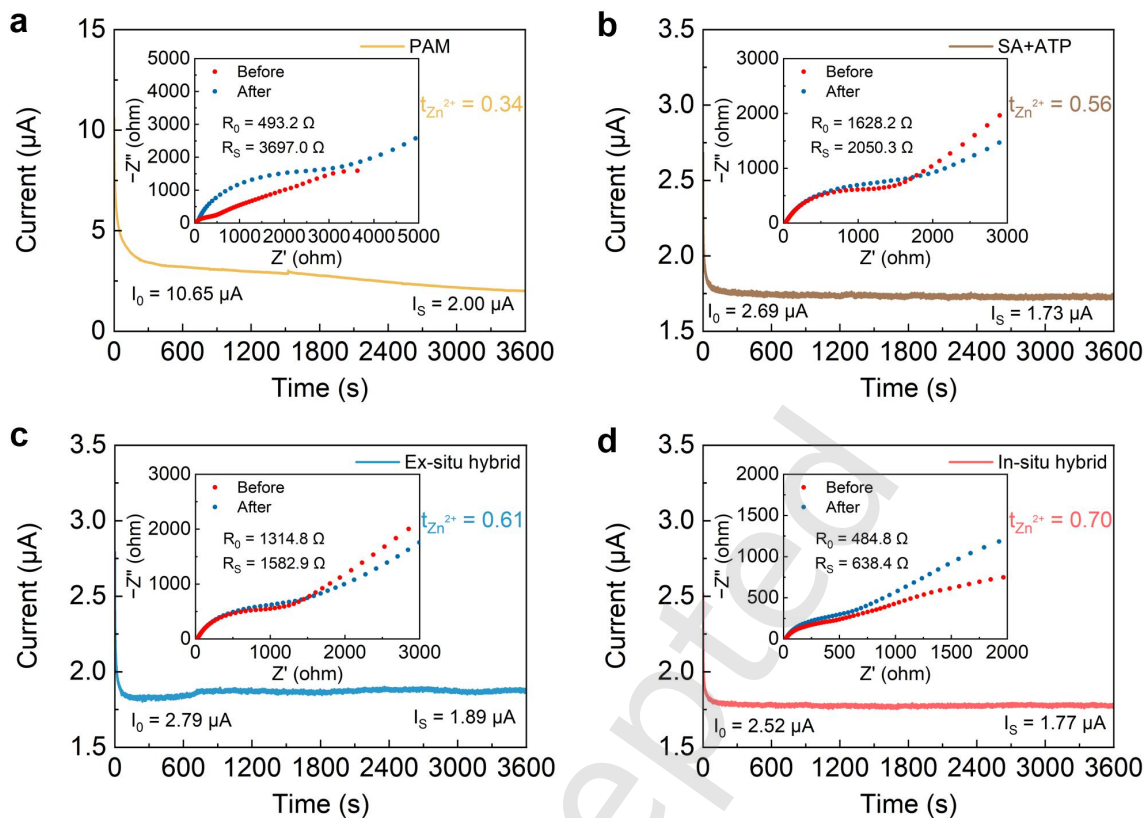


Figure S11. Polarization curves of the Zn//Zn symmetric cells using PAM (a), SA+ATP (b), ex-situ hybrid (c), and in-situ hybrid (d) hydrogel electrolytes. Insets are the EIS curves before and after polarization.

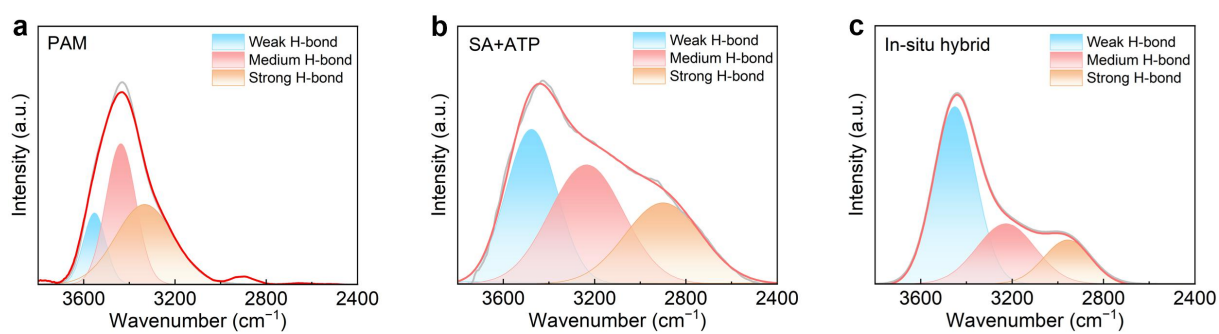


Figure S12. Peak deconvolution of O–H stretching vibrations in the FTIR spectra of PAM (a), SA+ATP (b) and in-situ hybrid (c) hydrogel electrolytes.

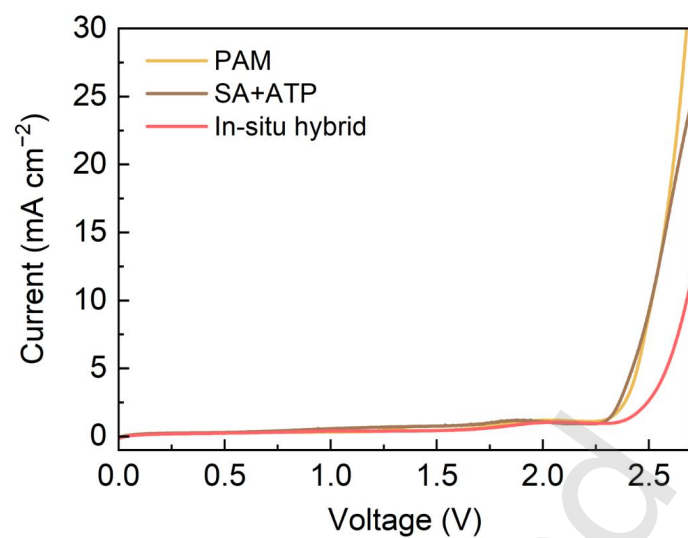


Figure S13. LSV profiles of various hydrogel electrolytes at a scan rate of 10 mV s⁻¹.



Figure S14. Optical photograph of a fiber Zn//Zn symmetric cell with electro-induced in-situ hybrid hydrogel electrolyte.

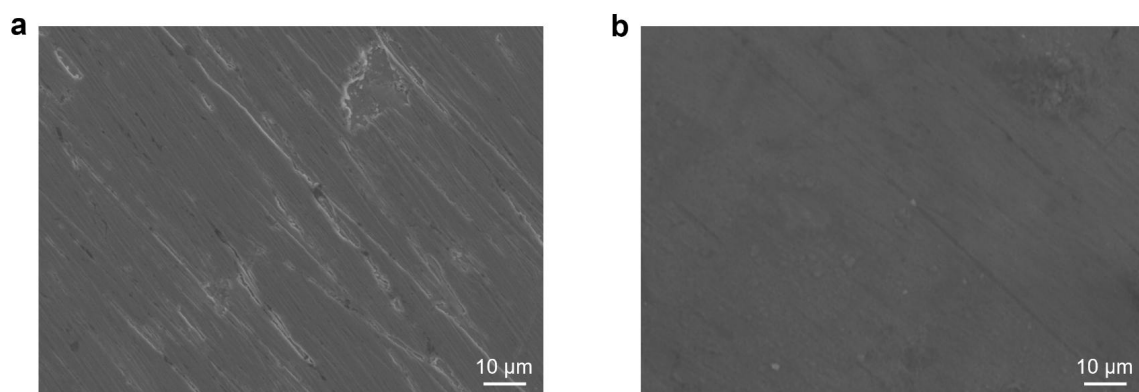


Figure S15. SEM images of the pristine Zn electrode (a) and the Zn electrode after applying electric current during the synthesis process of electro-induced in-situ hybrid hydrogel electrolyte (b).

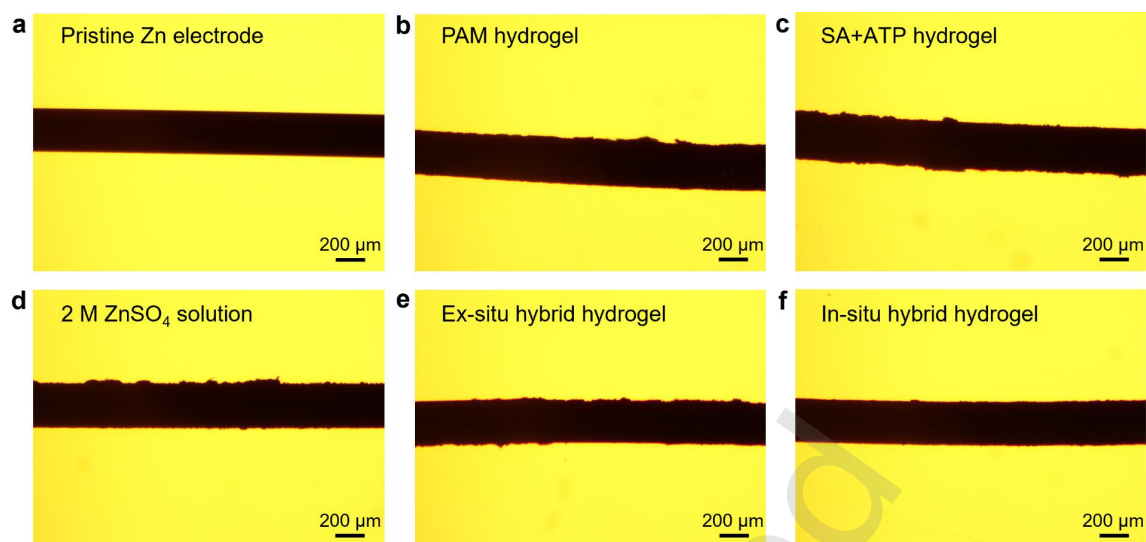


Figure S16. Optical microscope images of the pristine fiber Zn electrode (a) and the Zn electrodes after cycling in various electrolytes (b-f).

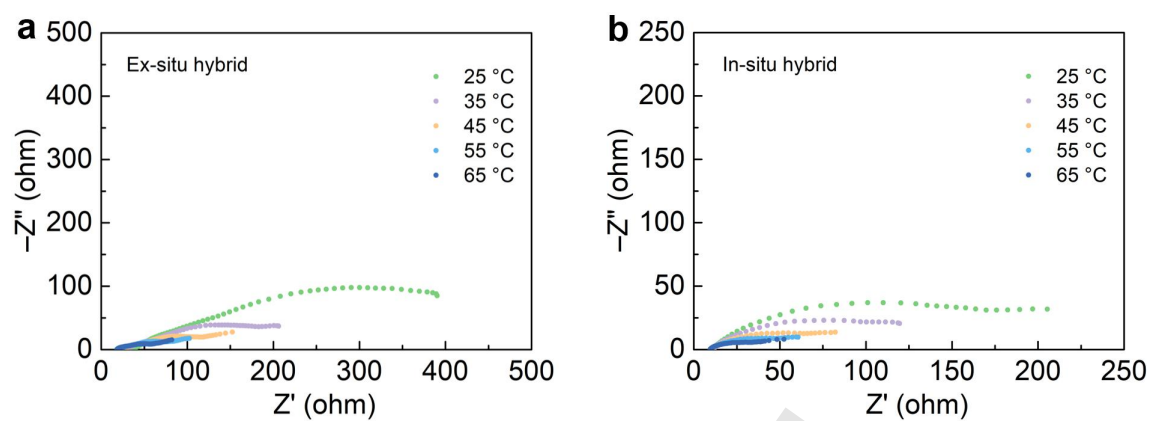


Figure S17. EIS curves of the Zn//Zn symmetric cells with ex-situ (a) and in-situ (b) hybrid hydrogel electrolytes at different temperatures.

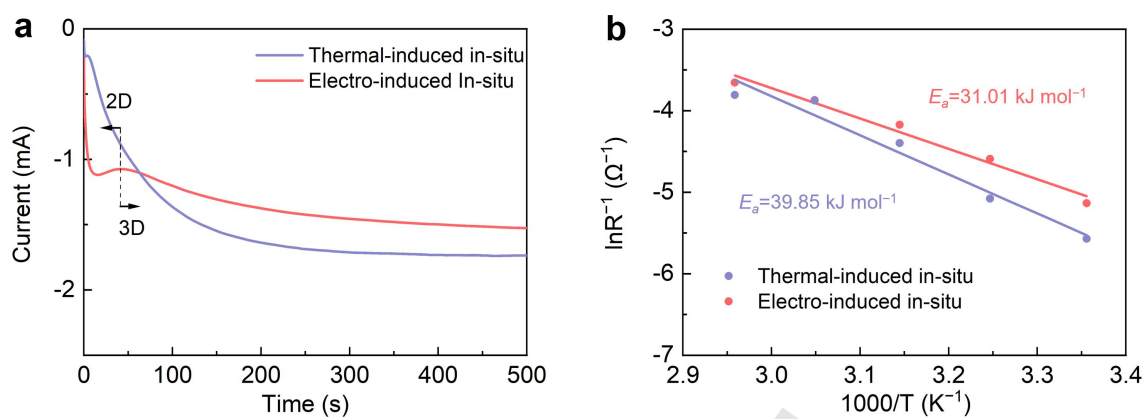


Figure S18. CA curves (a) and calculated desolvation energy barriers (b) of the fiber Zn//Zn cells with thermal-induced and electro-induced in-situ hybrid hydrogel electrolytes.

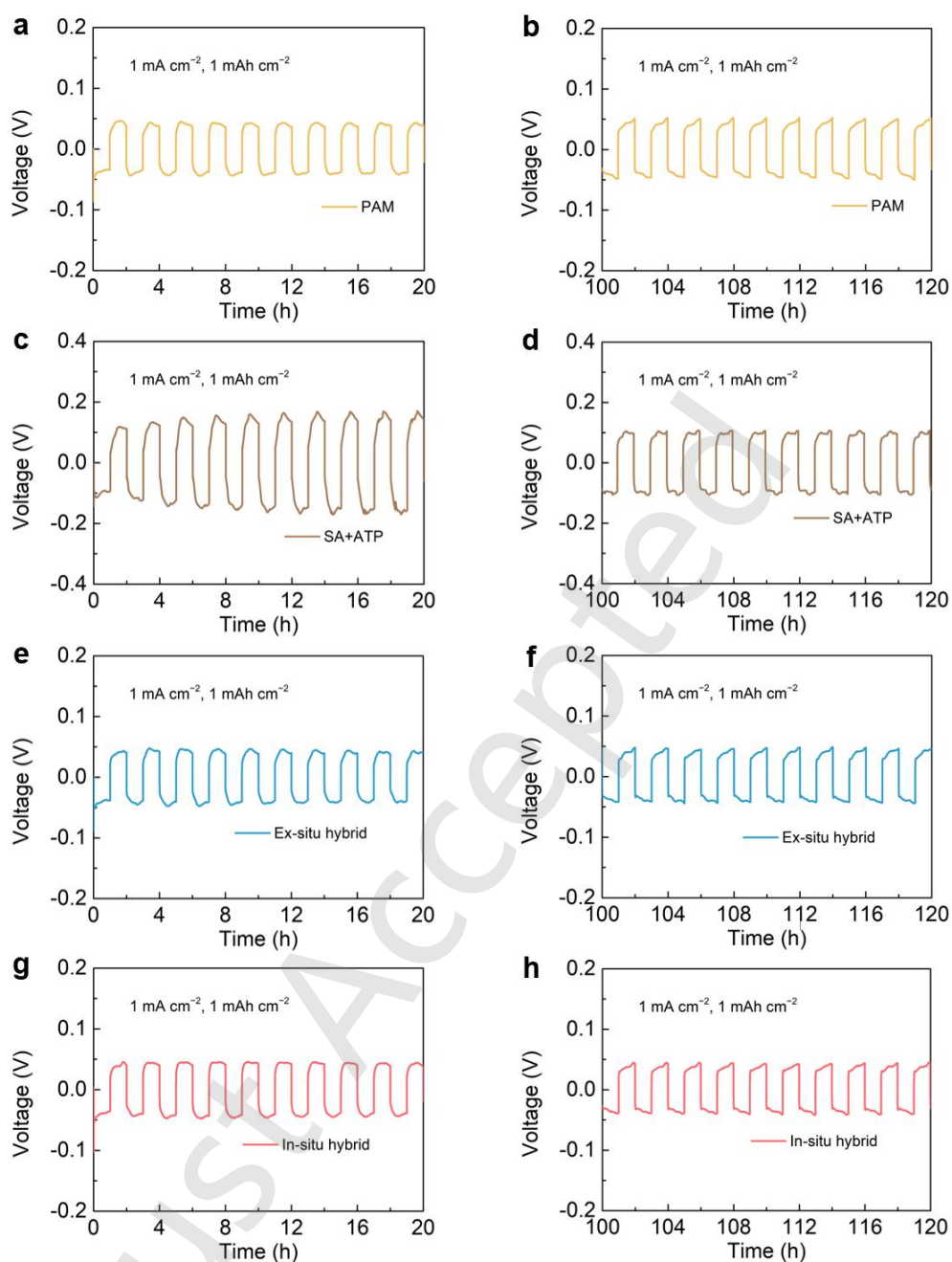


Figure S19. Magnified voltage profiles of the fiber Zn//Zn symmetric cells with various hydrogel electrolytes at different cycling stages.

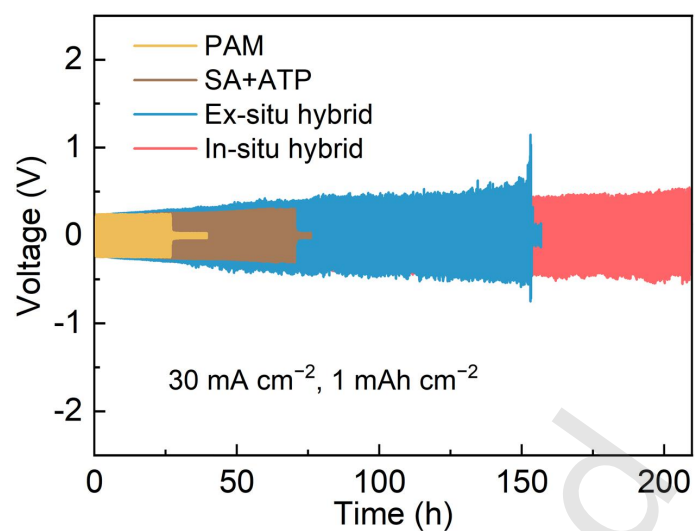


Figure S20. Cycling performance of fiber Zn//Zn cells with various hydrogel electrolytes at 30 mA cm⁻² and 1 mAh cm⁻².

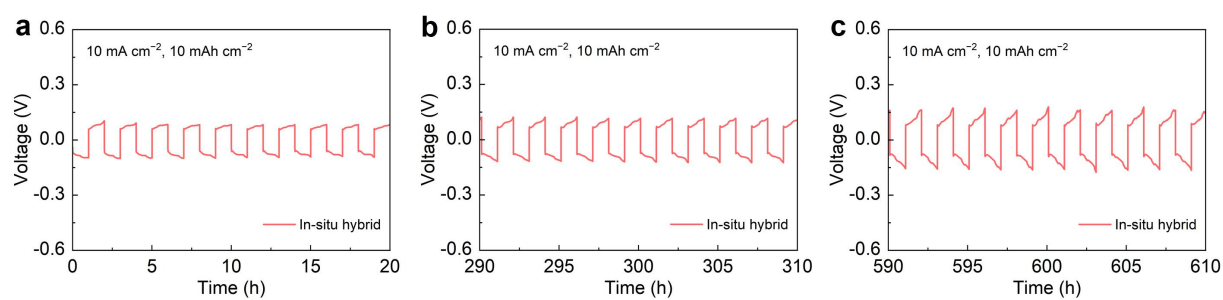


Figure S21. Detailed voltage profiles of the fiber Zn//Zn symmetric cells with in-situ hydrogel electrolytes at 10 mA cm^{-2} and 10 mAh cm^{-2} .

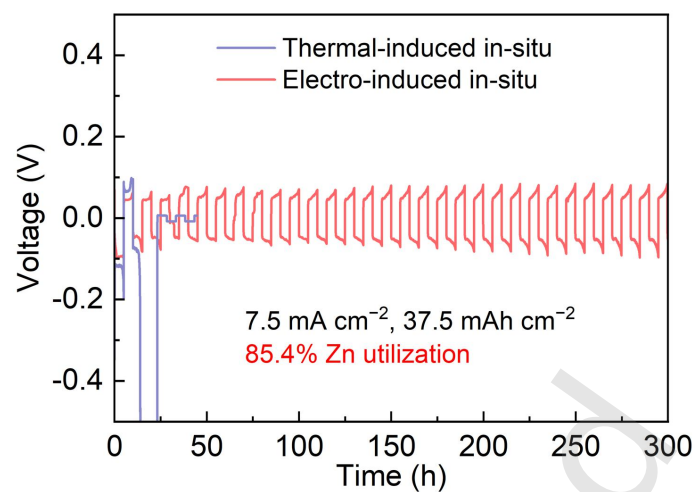


Figure S22. Cycling stability of fiber Zn//Zn cells with thermal-induced and electro-induced in-situ hybrid hydrogel electrolytes under an ultrahigh Zn utilization of 85.4%.

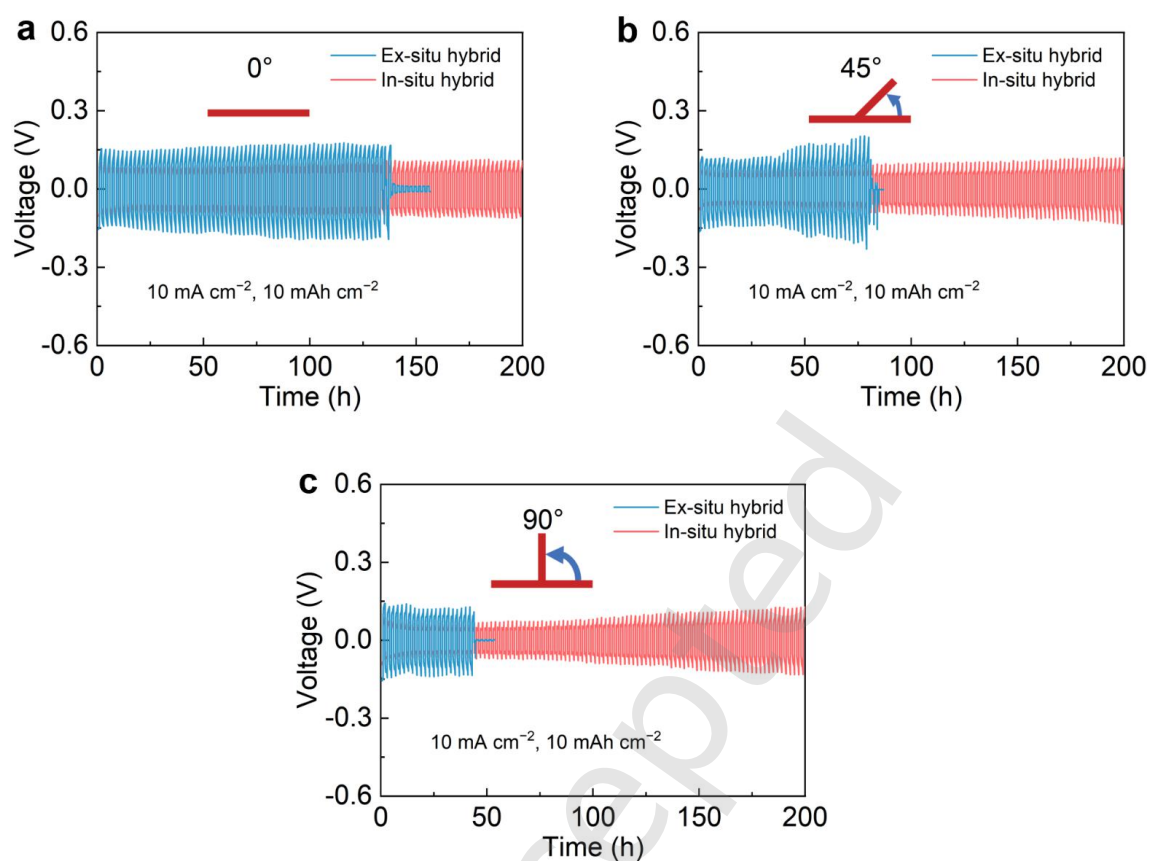


Figure S23. Cycling performance of the fiber Zn//Zn symmetric cells with different hydrogel electrolytes under various bending angles at 10 mA cm^{-2} and 10 mAh cm^{-2} .

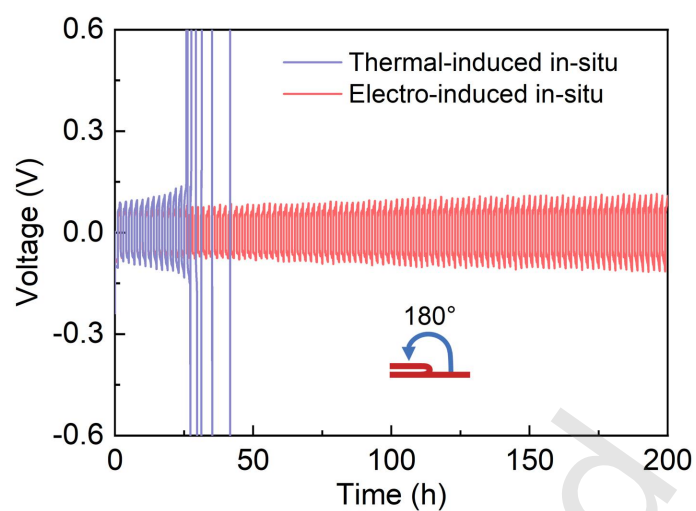


Figure S24. Cycling stability of fiber Zn//Zn cells with thermal-induced and electro-induced in-situ hybrid hydrogel electrolytes under a bending angle of 180°.

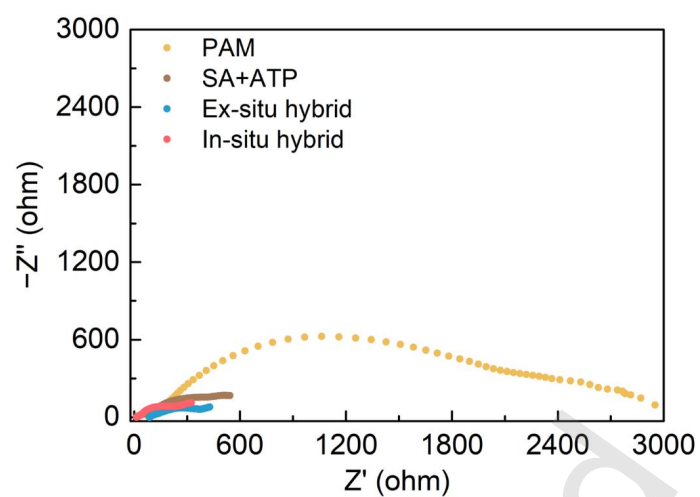


Figure S25. EIS curves of the Zn//Zn symmetric cells with various hydrogel electrolytes after charge-discharge cycles.

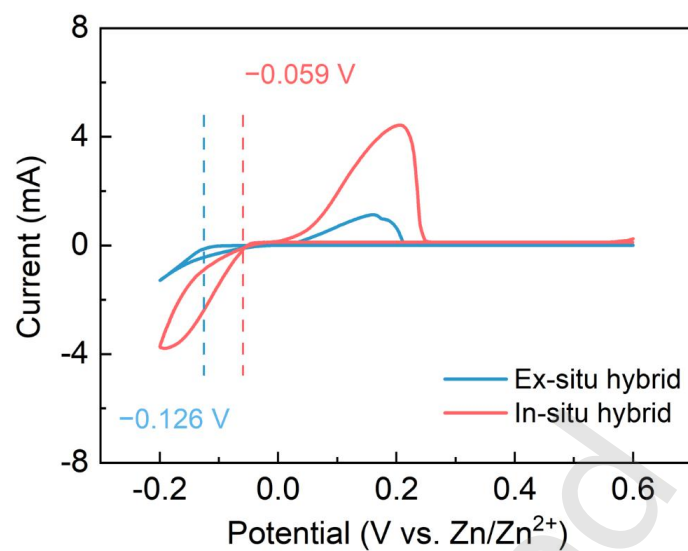


Figure S26. CV curves of the Zn//Cu asymmetric cells with the electro-induced in-situ and ex-situ hybrid hydrogel electrolytes.

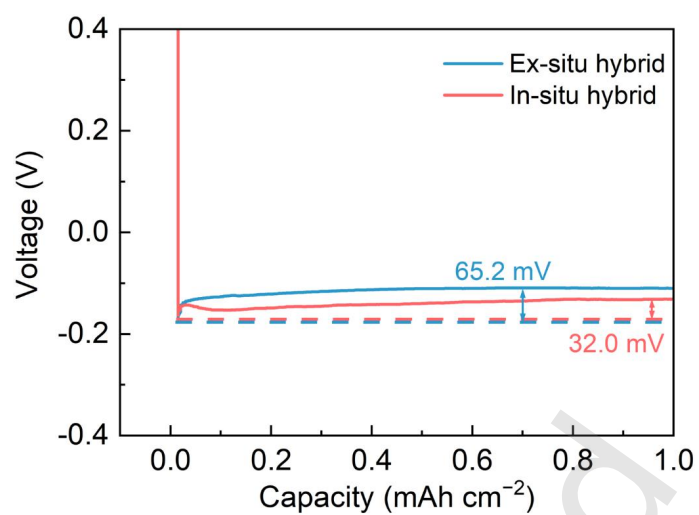


Figure S27. The deposition voltage profile of the Zn//Cu cells with electro-induced in-situ and ex-situ hybrid hydrogel electrolytes.

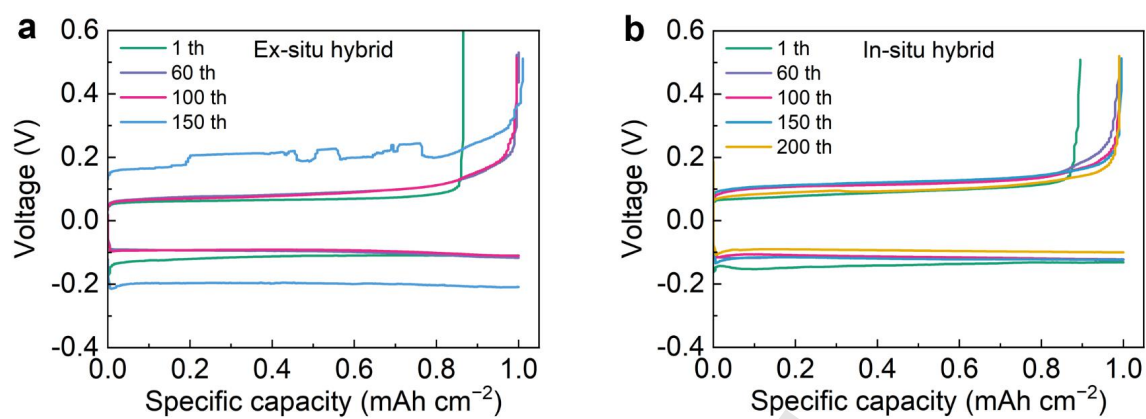


Figure S28. Voltage profiles of the Zn//Cu cells with ex-situ (a) and electro-induced in-situ (b) hybrid hydrogel electrolytes.

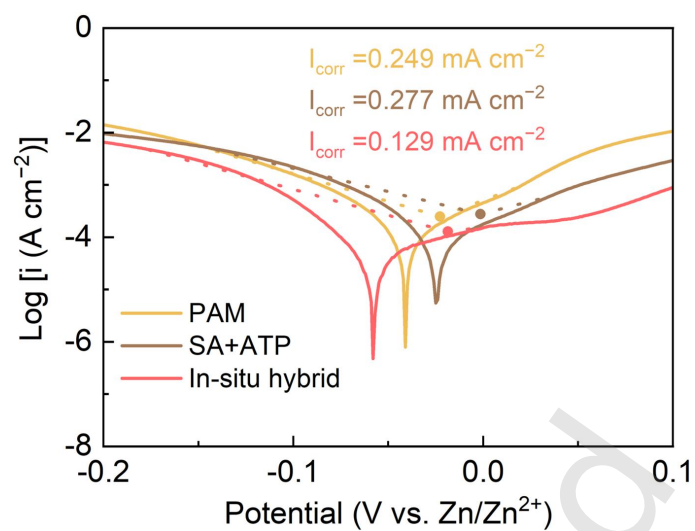


Figure S29. Tafel curves of the metallic Zn electrodes in different hydrogel electrolytes.

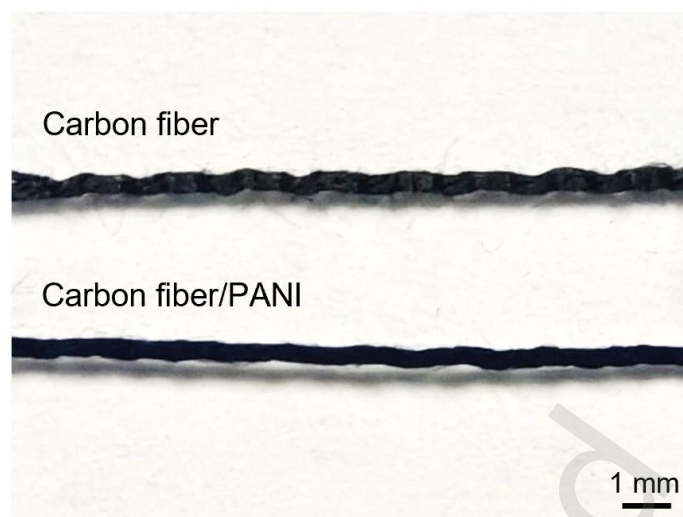


Figure S30. Optical photograph of the pristine carbon fiber and carbon fiber/PANI hybrid fiber cathode.



Figure S31. Photograph of a Zn//PANI fiber battery with a diameter of 1.93 mm.

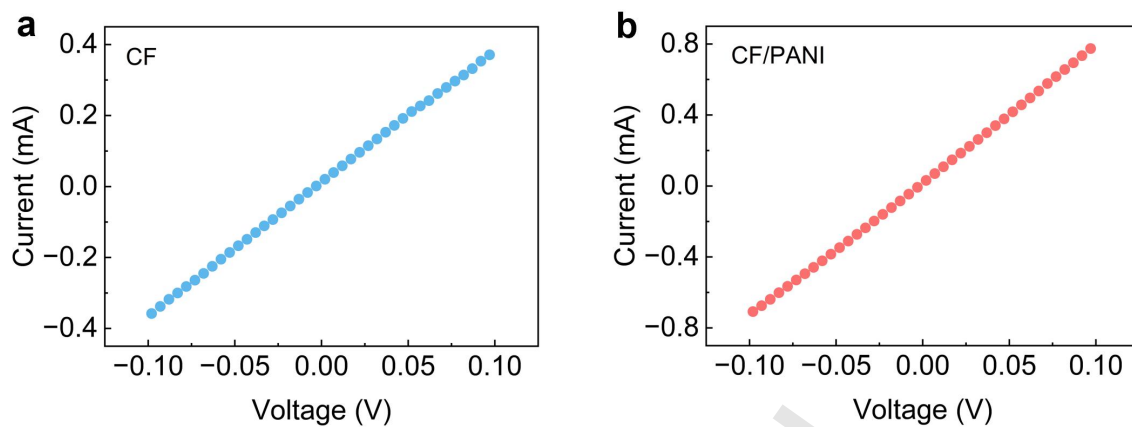


Figure S32. Current-voltage (I-V) characteristics of CF (a) and CF/PANI fiber cathode (b) with a length of 2 cm.

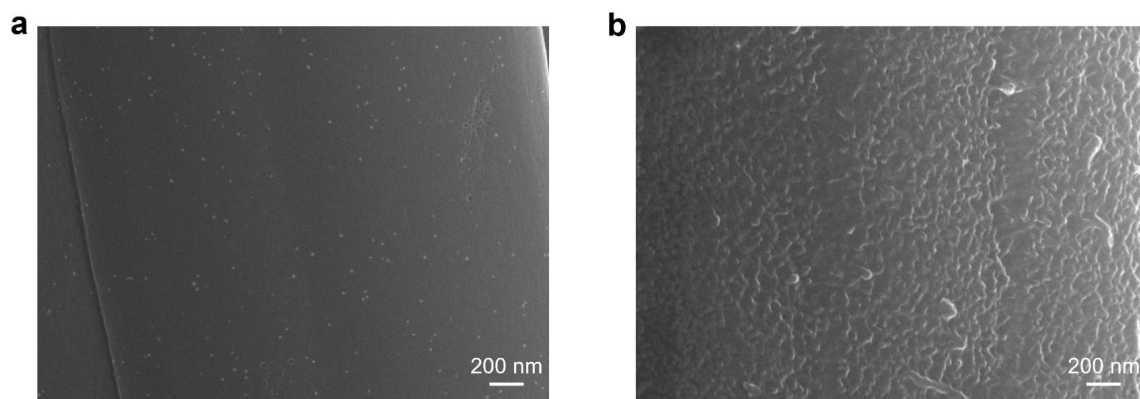


Figure S33. SEM images of the pristine carbon fiber (a) and carbon fiber/PANI cathode (b).

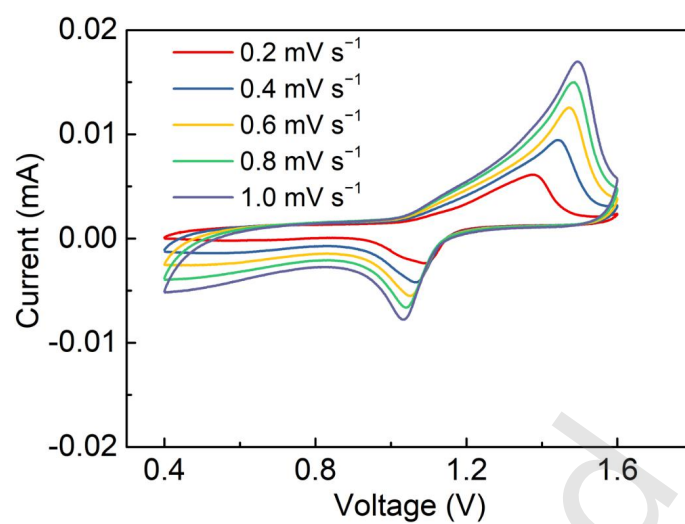


Figure S34. CV curves of the Zn//PANI fiber full cells using electro-induced in-situ hybrid hydrogel electrolyte at various scan rates.

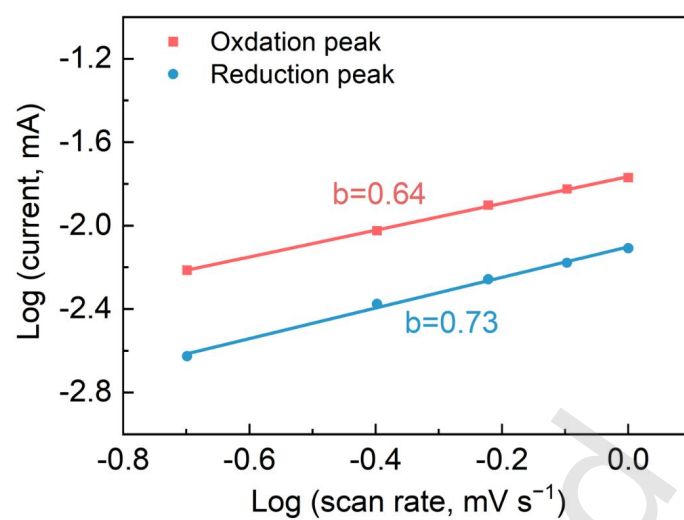


Figure S35. Plots of log (peak current) versus log (scan rate) at cathodic and anodic peaks in CV curves.

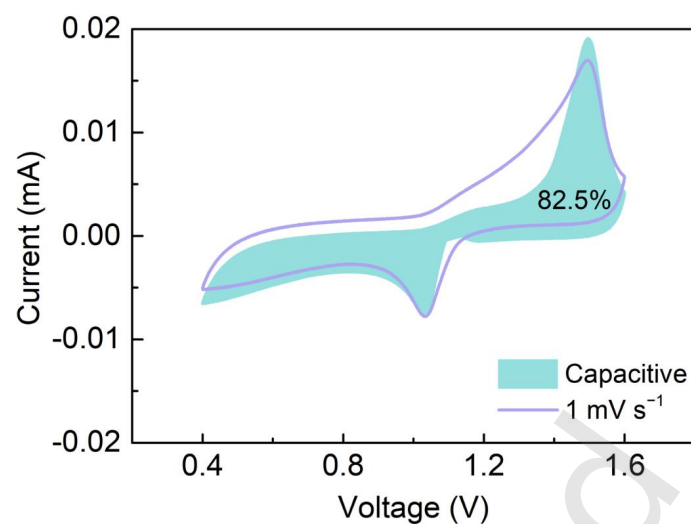


Figure S36. The capacitive contribution curve of the electro-induced flexible Zn//PANI fiber full cell at 1 mV s^{-1} .

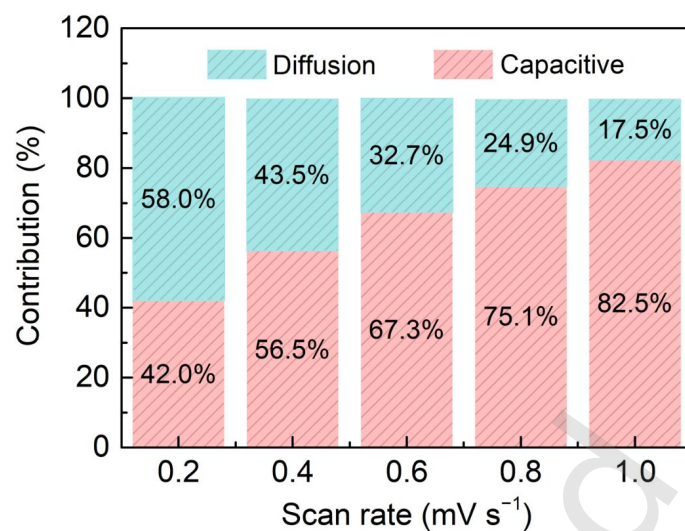


Figure S37. Comparison of capacity contribution of the flexible Zn//PANI fiber full cells at increasing scan rates.

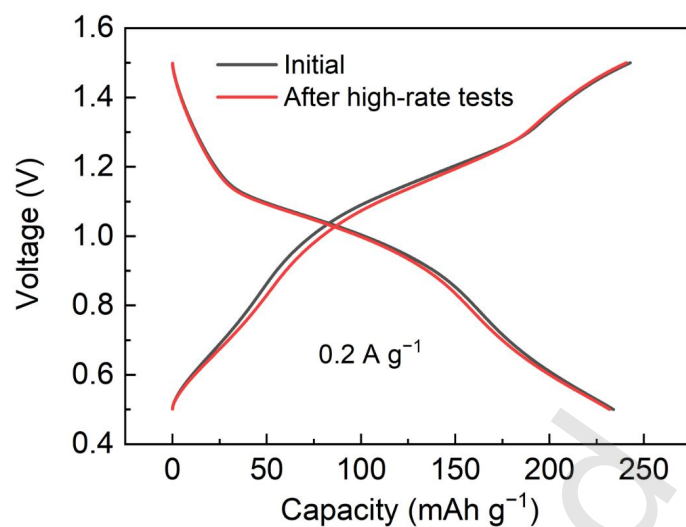


Figure S38. Galvanostatic charge-discharge curves of the electro-induced in-situ Zn//PANI fiber full cells before and after high-rate tests.

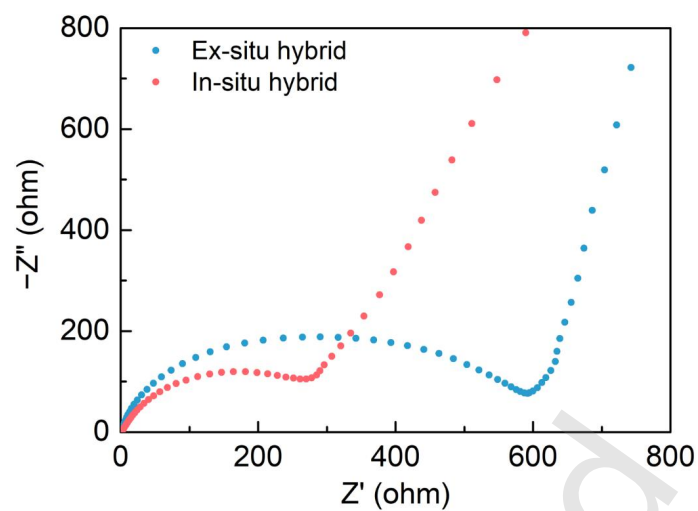


Figure S39. EIS curves of the Zn//PANI fiber full cells employing different hybrid hydrogel electrolytes before cycling.

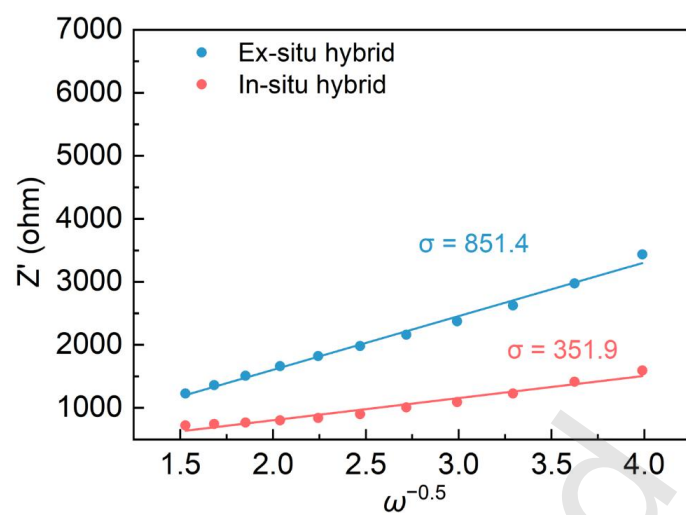


Figure S40. Relationship curves of Z' and $\omega^{-0.5}$ in the diffusion-controlled frequency range before cycling.

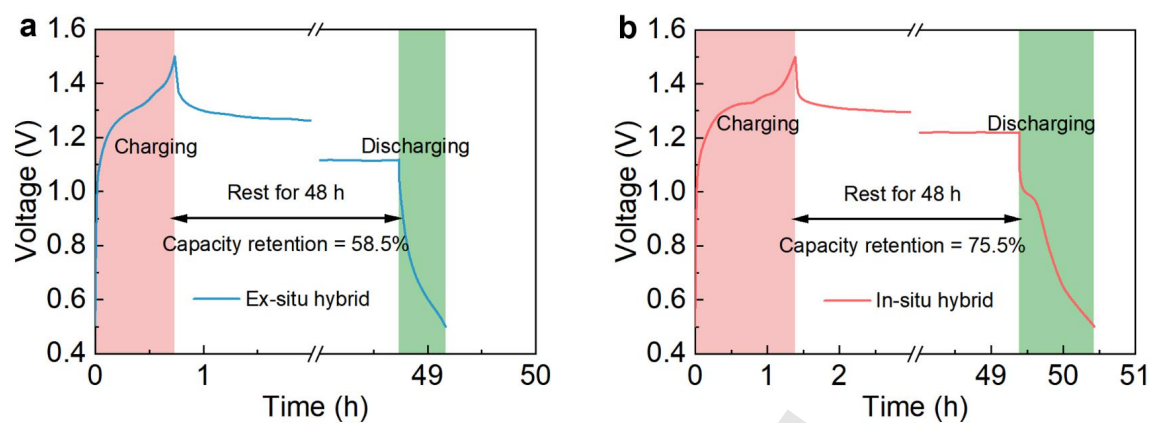


Figure S41. Self-discharge tests of the Zn//PANI fiber cells with ex-situ (a) and electro-induced in-situ (b) hybrid hydrogel electrolytes at 0.2 A g^{-1} .

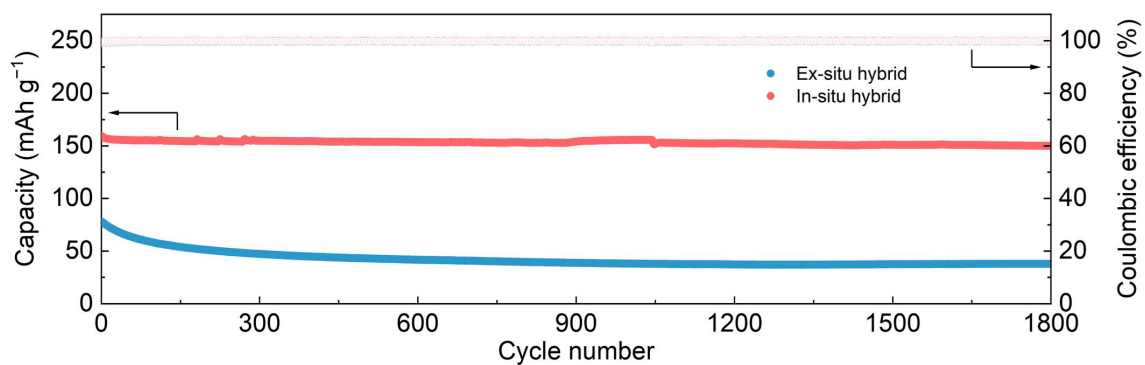


Figure S42. Long-term cycling stability of the flexible Zn//PANI fiber cells with ex-situ and electro-induced in-situ hybrid hydrogel electrolytes at 5 A g^{-1} .

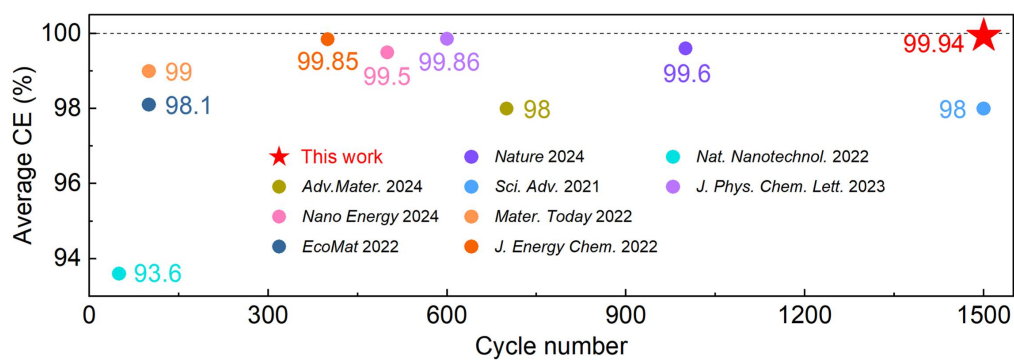


Figure S43. Comparisons of average coulombic efficiency (CE) and cycling performance of the electro-induced in-situ Zn//PANI fiber batteries and those of recently reported fiber batteries [S1-S9].

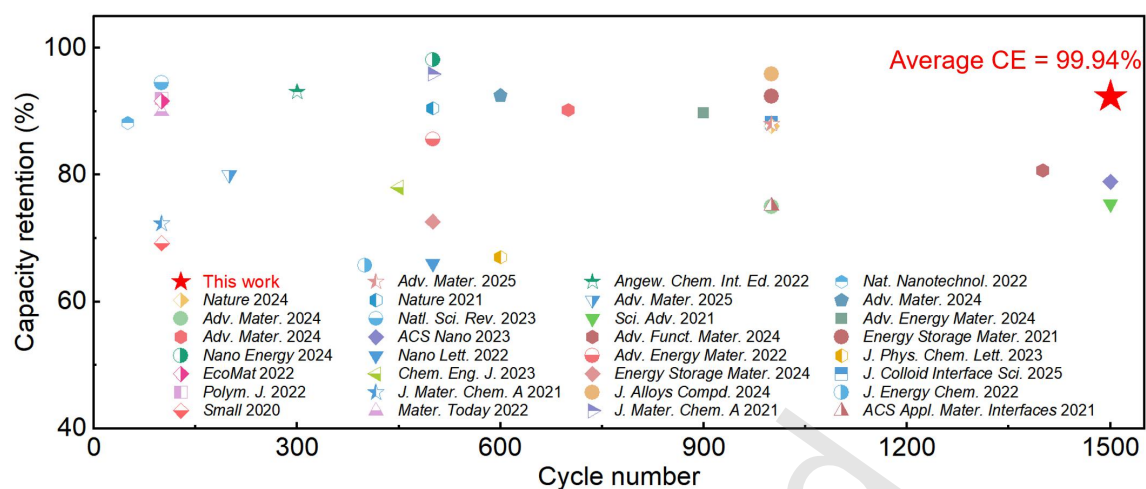


Figure S44. Comparisons of capacity retention rate and cycling lifespan of the electro-induced in-situ fiber batteries and those of previously reported fiber batteries [S10-S31].

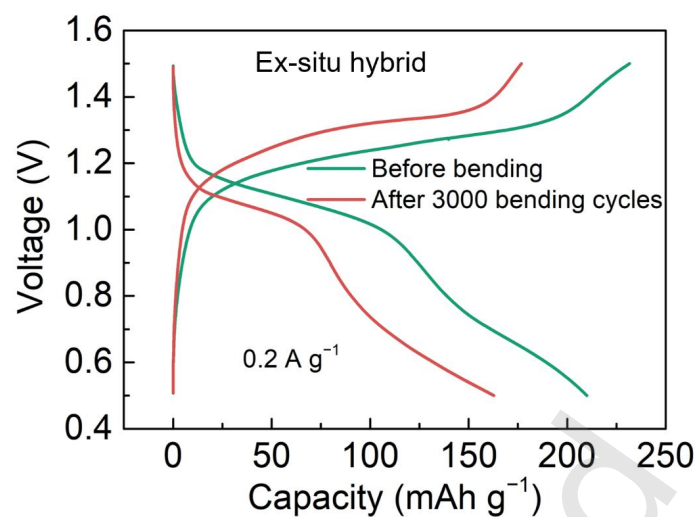


Figure S45. Charge-discharge curves of the electro-induced in-situ Zn//PANI fiber full cells using ex-situ hybrid hydrogel electrolyte before and after 3000 bending cycles at a bending angle of 180°.

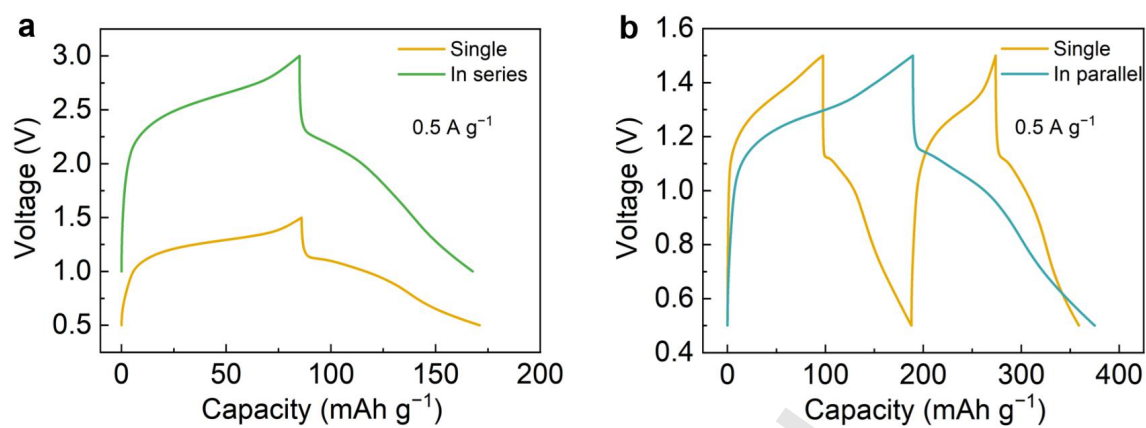


Figure S46. Galvanostatic charge/discharge curve of two flexible aqueous Zn//PANI fiber batteries connected in series (a) and in parallel (b), respectively.

Table S1 The intensities of various crystal planes of the standard Zn sample ($I_{0(hkl)}$) and the experimental Zn samples after cycling for various durations with electro-induced in-situ hybrid hydrogel electrolyte ($I_{(hkl)}$).

Intensity	(002)	(100)	(101)	(102)	(103)	(110)
I_0	53	40	100	28	25	21
$I(0\ h)$	3880	1293	7684	1539	1411	457
$I(20\ h)$	8908	1715	8764	2460	2650	790
$I(50\ h)$	5512	623	3109	1100	1484	560
$I(100\ h)$	4852	533	2195	784	1003	338
$I(300\ h)$	5975	281	1152	460	675	164

Table S2 Comparison of electrochemical properties of our electro-induced in-situ fiber Zn//Zn symmetric cells and previously reported fiber Zn//Zn cells.

Fiber electrodes	Electrolyte	Current density (mA cm ⁻²)	Areal capacity (mAh cm ⁻²)	Zinc utilization (DOD, %)	Cycle life (h)	Ref.
Zn wire	Electro-induced in-situ hybrid hydrogel electrolyte	1 7.5	1 37.5	1.7 85.4	2592 300	This work
Zn wire	2 M ZnSO ₄ + Blue fluorescent carbon dots solution	4	1.33	3	400	[S15]
Zn@liquid metal fiber	2 M ZnSO ₄ solution	1.74	0.35	7.1	800	[S16]
Annealed Zn@carbon nanotubes	1 M ZnSO ₄ solution	1	1	~12.8	800	[S10]
Zn wire	Zn-alginate polymer gel electrolyte	5	1.25	0.9	500	[S14]
Zn@carbon nanotube fibers	Dual-layer gel electrolyte with lysine	1	0.5	2.5	1500	[S2]
Zn wire	Polyacrylamide/sodium alginate hydrogel electrolyte	5	46.8	60	43	[S21]
Zn@carbon fibers	PVA/ethylene glycol/graphene oxide gel polymer electrolyte	0.49	4.9	10	150	[S7]
Zn@carbon nanotube fibers	Gelatin gel electrolyte	2	1	5	500	[S23]
Zn wire	Polydopamine-based all-in-one gel electrolyte	1	0.5	1.14	1200	[S32]
Zn@OCNTF fibers	2.5 M ZnSO ₄ solution	0.1 mA cm ⁻¹	0.1 mAh cm ⁻¹	20	920	[S28]
Carbon fibers/Zn	1 M ZnCl ₂ + 0.5 M NH ₄ Cl aqueous electrolyte	2	1	8.1	1000	[S17]
Zn wire	Poly(vinyl alcohol) hydrogel electrolyte + ZnSO ₄	0.1	0.05	/	600	[S33]

Table S3 Comparison of N/P ratio and cycling performance of our electro-induced in-situ Zn//PANI fiber batteries with recently reported flexible Zn-based full batteries.

Structure	Cathode	Anode	Electrolyte	N/P ratio	Current density (A g ⁻¹)	Cycle/ Capacity retention	Ref.
Fibrous	Carbon fiber/PANI	Zn wire	Electro-induced in-situ hybrid hydrogel electrolyte	2.55	2	1500 92.31%	This work
Planar	Carbon cloth@MnO ₂	Zn foil	2 M ZnSO ₄ + 0.1 M MnSO ₄ solution	3.63	1.5	200 62.8%	[S34]
Planar	Ti foil@MnO ₂	Zn foil@COF-320N	2 M ZnSO ₄ solution	3.9	1	800 46.7%	[S35]
Planar	MnO ₂	Zn foil	Oxygen-rich poly(urea-urethane)/polyacrylamide hydrogel electrolyte	3.3	0.2	50 92.3%	[S36]
Planar	Ti mesh@VO ₂ ·0.4H ₂ O	Zn foil	Polyacrylamide- 1,2-propanediol gel electrolyte	3.5	1	200 73.5%	[S37]
Planar	Carbon cloth@MnO ₂ /Carboxylic-MWCNT	Zn foil	Dopamine-grafted sodium alginate/poly(acrylamide-octadecyl methacrylate) gel electrolyte	5.8	0.62	100 59.0%	[S38]
Planar	Carbon cloth@V ₂ O ₅	Polycarbonate-based polyurethane-Zinc powder@ MXene-coated silk fabric	PVA/Zn(CF ₃ SO ₃) ₂ gel electrolyte	11.2	1	1000 84.5%	[S39]
Planar	ZnI ₂	Zn powder	2 M ZnSO ₄ + 1-butyl-3-methylimidazolium methanesulfonate	3	0.5	530 88.8%	[S40]
Planar	VO ₂	Zn foil	2 M ZnSO ₄ + D-pantothenol	2.89	1	475 96.35%	[S41]
Planar	Ti mesh @NH ₄ V ₄ O ₁₀	Zn foil	2 M Zn(CF ₃ SO ₃) ₂ + 50 mM cellobiose solution	2.9	2.2	500 89.2%	[S42]
Planar	V ₂ O ₅	Zn foil	1 M ZnSO ₄ + 50 mM 2-Morpholinoethanesulphonic acid	3.7	0.5	600 75%	[S43]
Planar	VOPO ₄ ·2H ₂ O	Zn foil	0.83 M Zn(TFSI) ₂ in H ₂ O-TFEA-TTE	2.85	/	500 88.4%	[S44]
Planar	VO ₂	Zn@Poly(allylamine hydrochloride)/poly(acrylic acid)	3 M Zn(CF ₃ SO ₃) ₂	5.08	0.43	250 96.3%	[S45]
Planar	NH ₄ V ₄ O ₁₀	Zn foil	2 M ZnSO ₄ + N-Acetoacetylmorpholine	2.63	0.3	130 75.83%	[S46]
Planar	MnO ₂	Zn foil@mechanically adaptive layer	2 M ZnSO ₄ + 0.1 M MnSO ₄ solution	2.5	0.5	110 85%	[S47]

Table S4 Comparison of electrochemical properties of our electro-induced in-situ Zn//PANI fiber batteries with previously reported fiber batteries.

Cathode	Anode	Electrolyte	Current density	Discharge capacity	Cycle number/ Capacity retention	Average CE	Ref.
Carbon fiber/PANI	Zn wire	Electro-induced in-situ hybrid hydrogel electrolyte	2 A g⁻¹	180.73 mAh g⁻¹	1500 92.31%	99.94%	This work
Lithium manganese oxide	Lithium titanium phosphate	Polyvinyl alcohol hydrogel electrolyte with Li ₂ SO ₄	0.05 A g ⁻¹	86 mAh g ⁻¹	50 88.2%	93.6%	[S1]
Zinc hexacyanoferrate	Carbon nanotube fibers/Zn	Dual-layer hydrogel electrolyte with lysine	0.2 A cm ⁻³	84.6 mAh cm ⁻³	700 90.2%	98%	[S2]
LiCoO ₂	Graphite	Dimethacrylate and acrylate-based hydrogel electrolyte	/	/	1000 87.7%	99.6%	[S3]
MnO ₂ rod	Zn microparticles	Polyacrylamide hydrogel with ZnCl ₂ , MnCl ₂ and PVA	0.05 A g ⁻¹	122 mAh g ⁻¹	500 98.14%	99.5%	[S4]
Lithium-iron-phosphate	Lithium titanate	PVDF hydrogel electrolyte with 1 M LiTFSI in PC/EC	25 μA cm ⁻¹	/	100 90%	99%	[S5]
Carbon nanotube/MnHCF	Carbon nanotube/MoO ₃ /graphene oxide	PVA-Al(CF ₃ SO ₃) ₃ hydrogel electrolyte	1 A cm ⁻³	34.6 mAh g ⁻¹	100 91.6%	98.1%	[S6]
Carbon fibers/MnO ₂	Carbon fibers/Zn	PVA/ethylene glycol/graphene oxide hydrogel electrolyte	2 A g ⁻¹	100 mAh g ⁻¹	600 67%	99.86%	[S7]
Carbon wire/MnO ₂	Zn wire	Graphene oxide-embedded polyvinyl alcohol hydrogel electrolyte	0.3 A g ⁻¹	166.7 mAh g ⁻¹	1500 75.4%	98%	[S8]
CNT/MnO ₂	Cellulose nanofibers/Zn	2 M ZnSO ₄ + 0.1 M MnSO ₄ solution	1 A g ⁻¹	126 mAh g ⁻¹	400 65.7%	99.85%	[S9]
V ₂ O ₅ /rGO	Carbon nanotubes/annealed Zn	1 M ZnSO ₄ solution	1 A g ⁻¹	145 mAh g ⁻¹	1000 75%	/	[S10]
LiCoO ₂	Graphite	Triglycidyl ether/tetrahydrofuran hydrogel electrolyte with LiTFSI	/	143.8 mAh g ⁻¹	1000 88%	/	[S11]
Lithium cobalt oxide	Graphite	Poly(ethylene glycol) dimethacrylate hydrogel electrolyte with 1 M LiPF ₆	0.085 A g ⁻¹	/	500 90.5%	/	[S12]
Graphite	Lithium cobalt oxide	1 M LiPF ₆ with EC/DEC/DMC (LB303)	0.075 A g ⁻¹	150 mAh g ⁻¹	200 80%	/	[S13]

MnO ₂	Zn wire	Zn-alginate polymer electrolyte	1 A g ⁻¹	218.1 mAh g ⁻¹	100 94.5%	/	[S14]
K ₂ NiFe(CN) ₆	Zn wire	2 M ZnSO ₄ + Blue fluorescent carbon dots solution	0.4 A cm ⁻³	75.3 mAh cm ⁻³	1500 78.9%	/	[S15]
Carbon fiber/V ₂ O ₅	Liquid metal fiber/Zn	2 M ZnSO ₄ solution	2 A cm ⁻³	179.8 mAh cm ⁻³	600 92.5%	/	[S16]
MoS ₂ /PEDOT	Zn wire	Poly(vinyl alcohol) hydrogel electrolyte + ZnSO ₄	0.1 A g ⁻¹	260.2 mAh g ⁻¹	300 93.1%	/	[S17]
CNT fiber /NC/N-VO ₂	CNT fiber/Zn	CMC/ZnSO ₄ hydrogel electrolyte	20 A cm ⁻³	/	500 85.61%	/	[S18]
MnO ₂	Zn wire	2 M ZnSO ₄ + 0.2 M MnSO ₄ solution	0.163 A g ⁻¹	54.4 mAh g ⁻¹	500 66%	/	[S19]
Ti/LiMn ₂ O ₄	Ti/Li ₄ Ti ₅ O ₁₂	LiCl + sulfolane + trimethyl phosphate solution	0.225 A g ⁻¹	44.9 mAh g ⁻¹	500 72.6%	/	[S20]
MnO ₂	Zn wire	PAAm/SA hydrogel electrolyte	1.248 A g ⁻¹	208 mAh g ⁻¹	1400 80.7%	/	[S21]
PPy/PEDOT:PSS	Carbon fibers/Zn	Carboxymethyl cellulose/ZnSO ₄ hydrogel electrolyte	0.16 A g ⁻¹	108.4 mAh g ⁻¹	1000 92.4%	/	[S22]
CNT fiber /A-Co(OH) ₂ /PEDOT:PSS	CNT fiber/Zn	Gelatin hydrogel electrolyte	0.05 A g ⁻¹	126.5 mAh g ⁻¹	1000 88.2%	/	[S23]
CNT/MoO ₃ /polypyrrole	CNT/Na _{0.44} MnO ₂	Tissue fluid in subcutaneous tissues	1 A g ⁻¹	43.03 mAh g ⁻¹	100 72.3%	/	[S24]
LiMn ₂ O ₄	LiTi ₂ (PO ₄) ₃	PAM/CNT hydrogel electrolyte	0.5 A g ⁻¹	84.8 mAh g ⁻¹	100 92%	/	[S25]
Al ₂ O ₃ / P-MoO ₃ - _x	Zn	Polyvinyl alcohol hydrogel electrolyte	20 A g ⁻¹	/	100 69.2%	/	[S26]
V ₂ O ₅ @Au@Ni Ti alloy wire	Zn wire	Gelatin hydrogel electrolyte	2 A g ⁻¹	/	1000 95.88%	/	[S27]
Carbon nano fibers/PANI	Carbon nano fibers/Zn	PVA hydrogel electrolyte	2 A g ⁻¹	116.0 mAh g ⁻¹	1000 75.1%	/	[S28]
Zn _x MnO ₂	Cu _{2-x} Se nanosheets	1 M ZnSO ₄ solution	1 A g ⁻¹	92.8 mAh g ⁻¹	900 89.8%		[S29]
CNT fiber /V ₆ O ₁₃	CNT fiber/Zn	3 M Zn(CF ₃ SO ₃) ₂	0.5 A g ⁻¹	323.3 mAh g ⁻¹	500 95.9%	/	[S30]
Silver coated nylon fiber @Ni@MOF@NiCo LDHs	Zn wire	PVA-KOH/Zn(CH ₃ COO) ₂ hydrogel electrolyte	2.5 mA cm ⁻¹	/	450 78%	/	[S31]

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