

Research Progress on the Antifreeze Performance of Water-based Zinc-ion Batteries Using Polyacrylamide as the Gel Electrolyte Base

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Abstract: Compared with lithium batteries, zinc batteries employing quasi-solid-state electrolytes demonstrate superior safety profiles, positioning them as pivotal components for flexible electronics development. Nevertheless, sub-zero temperature-induced electrolyte freezing remains a critical barrier to commercialization. Building upon conventional hydrogel matrices, this study systematically investigates polyacrylamide (PAM)-based gel electrolytes, critically evaluates existing strategies for low-temperature performance enhancement, and establishes standardized assessment protocols for zinc-ion battery performance. The perspective concludes with an outlook on hydrogel electrolyte innovations for next-generation aqueous zinc-ion batteries.

Keywords: aquifer zinc-ion battery; water gel Electrolyte; PAM gel battery; extreme temperatures.

1 Introduction

The growing demand for portable/wearable electronics in healthcare, defense, and fitness has intensified focus on flexible energy storage. While lithium-ion batteries dominate, their flammable organic electrolytes pose safety risks, limiting stability across operating conditions. This drives urgent need for cost-effective, high-performance alternatives with enhanced safety and electrochemical properties for next-generation flexible energy systems.

In recent years, zinc metal has gained significant attention due to its high theoretical capacity (approximately $820 \text{ mA}\cdot\text{h}\cdot\text{g}^{-1}$ gravimetrically and $5851 \text{ mA}\cdot\text{h}\cdot\text{cm}^{-3}$ volumetrically). With substantially larger resource reserves than lithium and lower cost, zinc exhibits low reactivity under normal conditions, offering enhanced chemical stability and high safety.

Hydrogel electrolytes form 3D networks through cross-linking, which enhances Zn^{2+} ion flow uniformity and transmission efficiency while mechanically suppressing dendrite growth. Their hydrogen-bond networks – modifiable by antifreeze additives – enable stable operation across wide temperature ranges. Functional groups (e.g., carboxyl, hydroxyl) facilitate directional ion regulation. Traditional hydrogel systems primarily utilize synthetic polymers such as polyacrylamide (PAM), polyacrylic acid (PAA), and polyvinyl alcohol (PVA), alongside natural polymers including carboxymethyl cellulose (CMC), guar gum (GG), xanthan gum, and gelatin.

In summary, the hydrogel electrolyte simultaneously possesses solid and liquid characteristics. The solid characteristics enable it to act as a separator, providing sufficient mechanical strength and stability during deformation, while the liquid characteristics help to provide flexibility and facilitate ion transport. Therefore, theoretically, it can be used as an integrated combination of the battery separator and electrolyte.

In recent years, researchers have been working to enhance the performance of water-based zinc-ion battery electrolytes for water-based hydrogels, and have extensively studied has been conducted on the optimization strategies of hydrogel electrolytes. Although there is abundant literature in this field, the anti-freezing properties of flexible hydrogel electrolytes is insufficient, which severely limits their application at low temperatures (LT). This paper focuses on discussing the achievements of PAM hydrogel electrolytes in breaking through temperature limitations, and summarizes the interaction mechanism of their performance. (Figure. 1) It also looks forward to the potential value of this hydrogel electrolyte in more extensive practical applications in the future.

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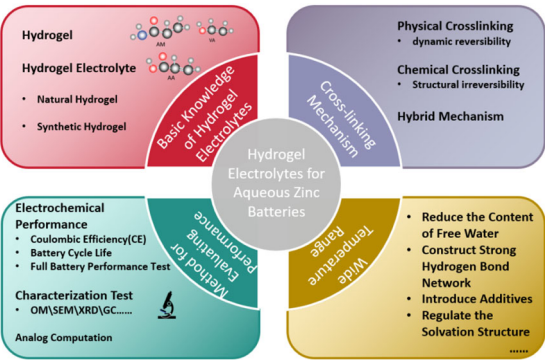


Figure 1. It summarizes the main points of the article.

2 Basic knowledge of hydrogel electrolytes

2.1 Hydrogel electrolyte

Hydrogels, with their multi-functional and controllable gel chemical properties (Table 1), provide a reliable platform for designing novel zinc-based battery hydrogel electrolytes (HPEs) Researchers have proposed various classification methods for hydrogels, including those based on cross-linking techniques, structural characteristics, product properties, or application scenarios.

Table 1. Summary of the most significant constraints and notable advantages

Hydrogel Substrate	Advantages	Disadvantages
Starch	Affordable,eco-friendly, good ion transport, dendrite inhibitionn	Low conductivity, weak mechanical properties
Gelatin	Affordable, environmentally safe, excellent ion mobility	Moderate mechanical strength
Alg	Affordable, simple preparation, adjustable properties	Sensitive to environment, low conductivity
Agarose	Strong mechanical properties, good thermal stability	Expensive, humidity sensitive
GG	High water retention, flexible	Poor LT performance
Chitosan	Biocompatible, safe, good mechanical strength	Low ion conductivity
Xanthan gum	Excellent flame retardancy	Complex preparation, costly
CMC	Great mechanical properties, frost resistant	Difficult to dissolve
PAM	Flexible, adhesive, controllable	Poor extreme condition tolerancepreparation process-
PAA	High ionic conductivity	Weak electrochemical stability
PVA	Biocompatible, good water retention	Limited thermal stability

Single-component hydrogels classified mainly into two categories: natural hydrogels, such as CMC, GG, gelatin,

and alginate. Synthetic hydrogels formed vis physical/chemical cross-linking methods, such as PAM, PVA, and PAA. (Figure.2) Natural hydrogels have advantages high water content, high biocompatibility, environmental friendliness, and abundant sources. In contrast synthetic hydrogels owing to their customizable properties, can achieve enhanced mechanical strength, high chemical stability, better self-healing ability, and ionic conductivity through corresponding molecular design. [1]

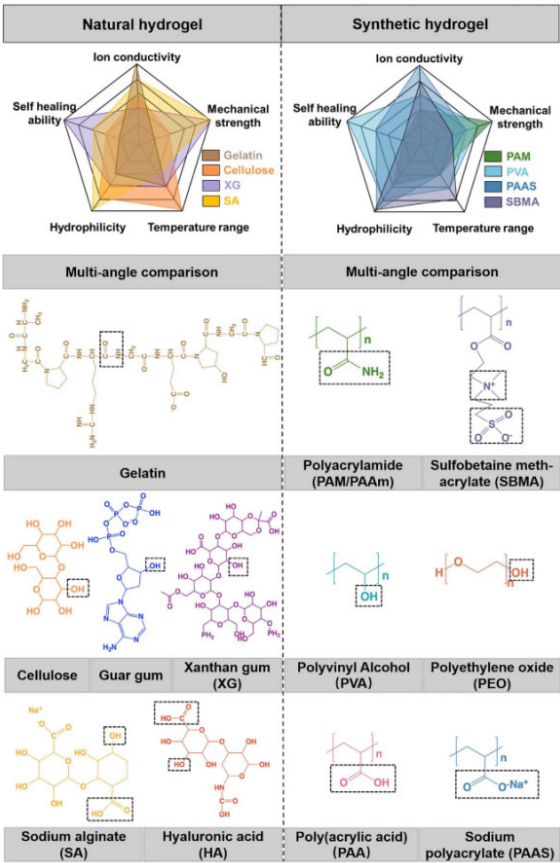


Figure. 2 Natural hydrogel species with their multi-angle comparison: gelatin, GG, xanthan gum, CMC, sodium alginate, and hyaluronic acid; and synthetic hydrogel species with their multi-angle comparison: PAM, PAA, PVA. Reproduced with permission. Copyright 2025 Springer Nature.

The gel-based PAM will be discussed in this article. It is a synthetic polymer with a controllable network structure, exhibiting excellent ionic transport properties primarily attributed to the abundant amide groups (-CONH₂) on the molecular chain. This groups form a dynamic hydrogen bond network with water molecules, enhancing the material’s hydrophilicity and ionic conductivity (up to 10⁻² S·cm⁻¹ level). However, the mechanical strength of pure PAM gel is usually insufficient. Studies have shown that by introducing crosslinking agents to construct a three-dimensional covalent network, simultaneously achieves high elastic modulus and rapid ion migration capability, making it valuable for applications in flexible energy storage devices.

3 Crosslinking mechanism

The crosslinking mechanisms of hydrogels are categorized into two primary modes: non-covalent (physical) and covalent (chemical) bonding. In practical fabrication, distinct advantages offered by either physical or chemical crosslinking drive the prevalent adoption of combined methodologies.

3.1 Single crosslinking mechanism

Physically crosslinked hydrogels based on non-covalent interactions involve multiple intermolecular forces, including ionic bonds/electrostatic attractions, hydrophobic interactions, and crystalline domain formation. These mechanisms provide fundamental insights for developing functional hydrogel materials with self-repair or stimuli-responsive properties.

Covalent crosslinking occurs when active groups on polymer chains (e.g., hydroxyl, carboxyl, amino) undergo condensation, addition, or ring-opening reactions with crosslinking agents, forming irreversible networks. This process is classifiable into four strategies: free radical polymerization, aldehyde-mediated crosslinking, click chemistry, and condensation reactions.

3.2 Hybrid Mechanism

Physically crosslinked hydrogels enable in situ gelation under mild conditions, generating dynamically reversible networks that stabilize interfaces to suppress dendritic growth and minimize side reactions. Chemically

crosslinked hydrogels exhibit superior mechanical strength and long-term stability through irreversible covalent bonds. Hybrid crosslinking strategies synergistically optimize hydrogel performance by integrating these complementary mechanisms for practical applications.

4 Evaluation methods for the Performance of Aqueous Zinc-ion Batteries

4.1 Electrochemical Method

Zn/electrolyte interface research remains nascent, resulting in immature methodologies for evaluating electrochemical performance.

4.1.1 Coulombic Efficiency(CE)

This method measures the ratio of discharged to charged capacity (in ampere-hours), serving as a key indicator of zinc deposition/stripping reversibility. Higher CE values indicate improved zinc-ion utilization and suppressed side reactions. Although modification strategies inhibiting hydrogen evolution and dendrite growth can enhance CE, test conditions significantly affect results: elevated current densities may artificially inflate CE due to denser zinc deposition. For accurate evaluation, multi-rate testing is recommended to assess cathode interlayer effectiveness.

$$CE = \frac{\text{stripping capacity}}{\text{plating capacity}} = \frac{\text{electric charge for Zn}^{2+}\text{stripping from active metallic Zn(mA}\cdot\text{h)}}{\text{electric charge for Zn}^{2+}\text{plating+irreversible electrochemical reaction(mA}\cdot\text{h)}} \quad (1)$$

4.1.2 Cycle Life

Monitoring battery capacity fading across cycles reveals inflection points indicating significant depletion of zinc anode active material or intensified side reactions. The cycle count preceding this inflection serves as a reliable cycle life indicator. When combined with actual coulombic efficiency calculations at high zinc utilization rates (ZUR), this parameter more accurately reflects zinc anode reversibility. Symmetric cell cycle life under specific current densities (e.g., 1 mA·cm⁻²) and areal capacities (e.g., 1 mA·h·cm⁻²) directly quantifies zinc anode stability. For instance, advanced zinc anodes achieve >1000 cycles under 1 mA·cm⁻²/1 mA·h·cm⁻² conditions.

The cycle count preceding the inflection point serves as a reliable indicator of cycle life. When combined with coulombic efficiency measurements at high zinc utilization rates (ZUR), this parameter more accurately reflects zinc anode reversibility. Under standardized current density and areal capacity conditions, symmetric cell cycle life directly quantifies zinc anode stability. For

example, some optimized zinc anodes can achieve thousands of cycles under the conditions of 1mA·cm⁻² and 1mA·h·cm⁻².

4.1.3 Full Battery Performance Test

Aqueous zinc-ion battery (ZIB) full-cell testing requires evaluation methods closely mimicking real-world conditions. While traditional half-cell (Zn||Cu) Coulombic efficiency and symmetric cell (Zn||Zn) cycling stability are important metrics, they often fail to represent actual electrochemical performance due to voltage window deviations from practical operating environments. The current research has two key issues: Firstly, excessive use of electrolyte will mask the effects of dendrite growth and side reactions; Secondly, an excessively high N:P ratio (usually > 15:1) leads to low utilization of the zinc negative electrode, thereby overestimating the battery performance.

4.2 Characterization Test

Effective modification of zinc electrode/electrolyte interfaces requires simultaneous resolution of three core challenges: hydrogen evolution suppression, dendrite prevention, and corrosion mitigation. To qualitatively or quantitatively evaluate the effectiveness of the interface modification strategy, different characterization techniques can be employed to monitor the changes in the interface phase before and after modification. According to previous studies, the commonly used characterization techniques include optical microscopes (OP), X-ray, scanning electron microscope (SEM), gas chromatography (GC) etc. Different characterization methods are used to detect different issues. For instance, an optical microscope can be used for in-situ observation of dendrite growth and for detecting hydrogen gas precipitation. (Figure.3) [8]

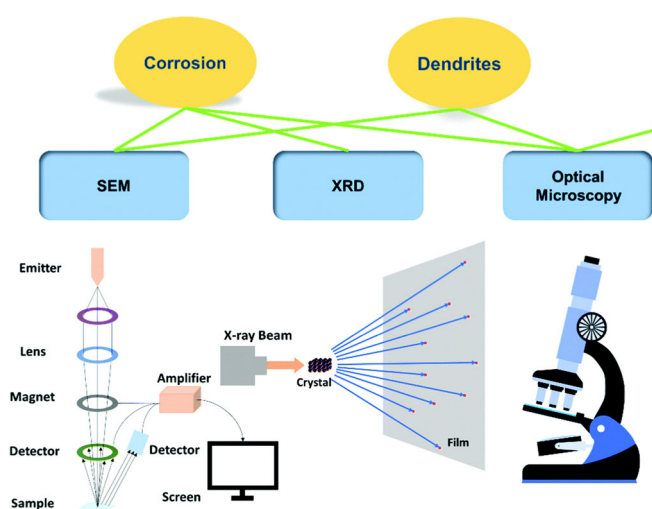


Figure.3 Characterization techniques to evaluate the effectiveness of interphase modification strategies. Reproduced with permission. Copyright 2021, Royal Society of Chemistry.

4.3 Analog Computation

Computational simulations complement characterization methods in evaluating electrode/electrolyte control strategies. Molecular dynamics (MD) and density functional theory (DFT) are predominantly employed to verify molecular-level mechanisms. DFT analyzes post-optimization electrode properties (e.g., Zn^{2+} diffusion paths and interfacial energy barriers). MD simulates multi-molecule electrolyte systems (>500 molecules)

5 Aqueous Zinc-ion Battery Achieves Wide Temperature Range

5.1 Strategies for Expanding the Operating Temperature Range of HPES

The traditional gels are mainly composed of polymers such as PAM, PAA, and PVA, as well as natural polymers like CMC, GG, xanthan gum and gelatin. Currently, the methods for expanding the temperature range of gel

electrolytes in aqueous zinc-ion batteries include: reducing the free water content, constructing strong hydrogen bond networks, introducing additives, and regulating the solvation structure, etc. For example, Zhi et al. optimized the hydrophilicity and flexible segments of the polymer framework through molecular engineering, reducing the water content of the hydrogel electrolyte from the traditional high water content (>60 wt%) to 35 wt%, while maintaining efficient zinc ion transport, significantly reducing water activity, inhibiting side reactions at high temperatures (HT), and enabling the electrolyte to achieve a 99% Coulomb efficiency at 90°C^[1]. Wang et al. developed an anti-freezing polyelectrolyte hydrogel by optimizing the ratio of lithium chloride to zinc chloride. They utilized the ionic interaction between the double salt metal ions and the hydrophilic groups in the polymer network to induce the polymer's enhanced co-crystallization behavior, forming a strong hydrogen bond network. This simultaneously enhanced the ionic conductivity, mechanical properties, and anti-freezing ability of the electrolyte, enabling the polyaniline||zinc battery to achieve a high capacity of 118 $\text{mA}\cdot\text{h}\cdot\text{g}^{-1}/0.2\text{ A}\cdot\text{g}^{-1}$ at -70°C^[2]. Zhang et al. introduced diethylene glycol monomethyl ether (dg) as an electrolyte additive to disrupt the original hydrogen bond network, thereby enhancing the performance of zinc over a wide temperature range from -35°C to 65°C^[3]. Li et al. prepared a local aggregation electrolyte that anchors active water using long-chain organic molecules with hydrogen bond acceptors. This enabled the solvent shell of zinc ions to mainly adopt the contact ion pair (CIP) structure, resulting in local aggregation. Water was dynamically anchored by ether bond groups through strong hydrogen bonds, and it neither evaporated nor froze even at 90°C and -30°C. The operating temperature range could reach 120°C^[4].

5.1.1 The Freezing of Water-based Gel Electrolytes at LT

Hydrogel freezing behavior depends on water-polymer hydrogen bonding: at LT, reduced molecular motion strengthens these bonds, promoting ice formation. Disrupting this network lowers the FT. Hydrogel water exists as free water (easily frozen) and bound water (lower FT due to polymer chain attachment). Reducing the free water proportion decreases hydrogel FT. Ions also influence FT—moderate concentrations disrupt water's hydrogen bonds to suppress freezing, while excess ions may crystallize. Polymer network density affects FT; tighter structures restrict water mobility but excessive crosslinking compromises mechanical properties. Optimizing these factors must balance temperature range expansion with overall battery performance.

5.2 Improvement Strategies for Enhancing the LT Limit

By analyzing the performance achieved after modification of the gel electrolytes prepared based on PAM in aqueous zinc-ion batteries, the advantages of each modified gel

electrolyte obtained through different methods are statistically evaluated. (Table 2)

Table 2. Comparison of the working temperature range and performance of batteries prepared with different gel electrolytes.

Electrolyte Type and Working Temperature Range(°C)	Battery Specific Capacity Under Different Conditions (mA·h·g ⁻¹)	Cycle Performance of battery	Rate Performance of Battery	Other Performance Characteristics	Highlighted Features	Ref
ZS/GL/AN -20~60	-20°C 500laps 138 60°C 500laps 262-251.52	-20°C 500laps 100% 60°C 500laps 96%	5A·g ⁻¹ 10,000laps 185 mA·h·g ⁻¹ , 88%	60°C No short circuit	High temperature working capacity, long life	[5]
Zn(BF ₄) ₂ -PAM -70~25	-70°C 45.4 25°C 110.3	-70°C 41% -60°C 55% -40°C 78%	50 mA·h·g ⁻¹ 45.4 mA·h·g ⁻¹	-	-	[6]
PDC-20 -40~25	-20°C 160.5 25°C 238.4	-40°C 100cycle life -20°C 350laps 0.1A·g ⁻¹ 62.5% 0.2 A·g ⁻¹ 61.6%	Room temperature 2A·g ⁻¹ 238.4mA·h·g ⁻¹ 5000laps 67% 0.2A·g ⁻¹ 151.8mA·h·g ⁻¹	Freeze resistance of gel electrolyte	-	[7]
PAM-BIS-D0.6 -20~25	-20°C 161 25°C 189	-20°C 98.8% 25°C 300laps 99.5%	0.1C 211.5mA·h·g ⁻¹ 0.2C 195.9mA·h·g ⁻¹ 0.5C 172.9mA·h·g ⁻¹ 1.0C 150.2mA·h·g ⁻¹ 2.0C 129.3mA·h·g ⁻¹	Bending state is stable	-20° No loss of work	[8]
PAM-1,2-PG -30~25	-30°C 97 25°C 380	65,000laps 212F·g ⁻¹ -30°C 16,470laps 145F·g ⁻¹	0.5A·g ⁻¹ 380mA·h·g ⁻¹ 1A·g ⁻¹ 350mA·h·g ⁻¹ 3A·g ⁻¹ 312mA·h·g ⁻¹ 5A·g ⁻¹ 291mA·h·g ⁻¹ 10A·g ⁻¹ 261mA·h·g ⁻¹	Super long cycle life 65,000 times	Ultra long cycle life	[9]
GO/LAP-PAM -20~25	20°C 39.76 0.25A·g ⁻¹ 10.38	-20°C 37.02%	-	-	-	[10]
M-DPAM-3 20~25	616	2mA·cm ⁻² : -20°C 72h Room temperature 55 h	-	25°C Power density 136.2 mW·cm ⁻² LT Power density 115.6 mW·cm ⁻²	High power	[11]
PAM-SC -40~80	20°C 746.1	-40°C 700cycle life 20°C 420cycle life	-	It can work in liquid nitrogen environment Maximum power of 20°C: 107.7mW·cm ⁻²	Wide working temperature range	[12]
CSAM -30~25	-30°C 97 25°C 380	1A/g 200laps 73.5% -30°C, 1mA·cm ⁻² 3780h	0.5~10 A·g ⁻¹ 380~261 mA·h·g ⁻¹	Temperature fluctuation stability	LT life, but low capacity, but suitable for high current density work	[13]
PAM/CNF/LiCl -30~50	-30°C 0.2A·g ⁻¹ 124	-30°C 0.2A·g ⁻¹ 500cycle life 30°C 4A·g ⁻¹ 1000laps 87% 50°C 4A·g ⁻¹ 500laps 85.8%	-30°C 0.1~1 A·g ⁻¹ 130~75 mA·h·g ⁻¹ When the current was restored to 0.1 A·g ⁻¹ , the capacity unchanged	Full climate power supply	-30°C with high rate recovery 1A·g ⁻¹ 75mA·h·g ⁻¹	[14]
PBDT -70~0	-70°C 0.2A·g ⁻¹ 118 -40°C~0°C 113~137	-70°C 0.5A·g ⁻¹ 12,000cycle life 99.5%	-70°C 1A·g ⁻¹ 64.61 mA·h·g ⁻¹	-	ExtremeLT High LT capacity, the longest battery life	[2]

5.2.1 PAM-1,2-PG Ultra-long Cycle Life

Using polyacrylamide (PAM) and Zn(OTf)₂ as the main components, 1,2-propanediol (1,2-PG) was introduced as

a co-solvent. (Figure.4a) The hydroxyl groups of 1,2-PG strongly interact with water molecules, breaking the hydrogen bond network among the original water molecules. The DSC test verified the absence of

crystallization peaks, indicating that the water molecule activity was inhibited, and the hydrogen bond network was restructured. The Raman spectrum (Figure.4c) showed an optimized ratio of strong hydrogen bonds (DDAA-OH) and weak hydrogen bonds (DA-OH), reducing the content of free water. Finally, an ultra-long cycle life for the Zn||FSC flexible capacitor was achieved (Figure.4d): a capacity retention of $212 \text{ F} \cdot \text{g}^{-1}$ was maintained after 65,000 cycles at room temperature, and $145 \text{ F} \cdot \text{g}^{-1}$ after 16,470 cycles at -30°C [9].

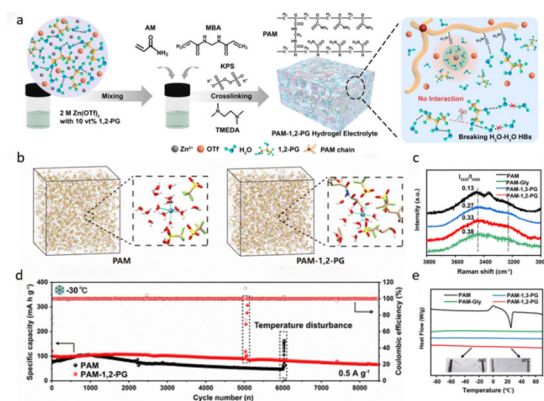


Figure 4. Reproduced with permission [9]. Copyright 2024, Royal Society of Chemistry. PAM-1,2-PG hydrogel electrolyte a) Illustration of the polymerization processes and structure of PAM-1,2-PG. b) MD simulation snapshots of PAM and PAM-1,2-PG hydrogel electrolytes (PAM: $n = 6$). c) Raman spectra of different HPEs in the range of 3000–3800 cm⁻¹. d) Cycling performance of the AZIBs at -30°C . e) DSC test of different HPEs from -90°C to 70°C at a heating rate of 5°C min^{-1} .

5.2.2 PBDT Extreme LT Gel

Gao et al. constructed a dual-crosslinked network by combining the rigid polyelectrolyte (PBDT) with the flexible polyacrylamide (PAAm) containing $-\text{CONH}_2$ groups. (Figure.5a) They regulated the properties of the hydrogel through the synergistic effect of the double salts (ZnCl_2 and LiCl). The optimal ratio of the double salts was 3m ZnCl_2 - 8m LiCl . The interaction between the ions and the hydrophilic groups formed a "polymer-enhanced co-crystallization effect" (PFG-NMR and Raman spectroscopy confirmed this). Li^+ inhibited ice crystal formation by breaking the hydrogen bonds between free water molecules. Additionally, DSC tests showed that the hydrophilic groups ($-\text{SO}_3^-$ and $-\text{CONH}_2$) bound and restrained water, lowering the freezing point to -113.9°C . The battery assembled with this gel electrolyte could achieve good cycling performance at -70°C . For example, the Zn||PANI battery maintained a capacity retention rate of nearly 100% ($97.8 \text{ mA} \cdot \text{h} \cdot \text{g}^{-1}$) after cycling 12,000 times at -70°C at a current density of $0.5 \text{ A} \cdot \text{g}^{-1}$ (Figure5.b) [2], and the Coulombic efficiency reached 100%. This study was the first to achieve stable operation of an aqueous zinc battery at -70°C , expanding the applicability in extreme environments compared to similar studies (with the $\text{LT} \leq -50^\circ\text{C}$) [2].

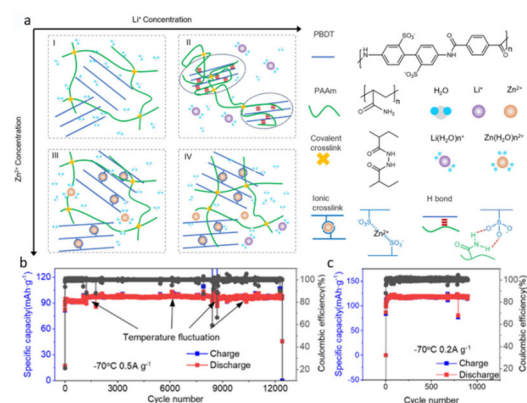


Figure 5. Reproduced with permission. Copyright 2025, Wiley-VCH. a) Synergistic polymer-salt design strategy for the HPEs. b) Cycling performance of Zn||PANI cells at -70°C and $0.5 \text{ A} \cdot \text{g}^{-1}$. c) Cycling performance of Zn||PANI cells at -70°C and $0.2 \text{ A} \cdot \text{g}^{-1}$.

5.2.3 PAM/CNF/LiCl Low-temperature Rate Operation Restored Gel

Ge et al. used cellulose nanofibers (CNF) as the reinforcing phase and physically cross-linked it with PAM through hydrogen bonds. LiCl was added to inhibit the freezing and evaporation of water molecules. (Fig.6a) Li^+ increased the proportion of bound water (accounting for 30.9%) through strong hydrogen bonding. DSC showed that no phase transition peak was generated at -80°C , meaning no ice crystals formed. Using PAM/CNF/ LiCl 50% as the electrolyte, and carbon CNF/polypyrrole (PPy) as the electrode, a flexible device was constructed. The surface capacitance was $36.9 \text{ mF} \cdot \text{cm}^{-2}$ at -20°C ($110.2 \text{ mF} \cdot \text{cm}^{-2}$ at 25°C), and the performance remained stable after temperature cycling. (Figure.6b) Due to the synergistic effect of CNF and LiCl , this hydrogel possesses a stable ion conductive network reconstruction ability. Based on this hydrogel, the supercapacitor exhibited an extremely high capacity recovery rate during extreme temperature cycling (25°C to -40°C) - maintaining 96% of the initial capacity after 10,000 charge-discharge cycles [14].

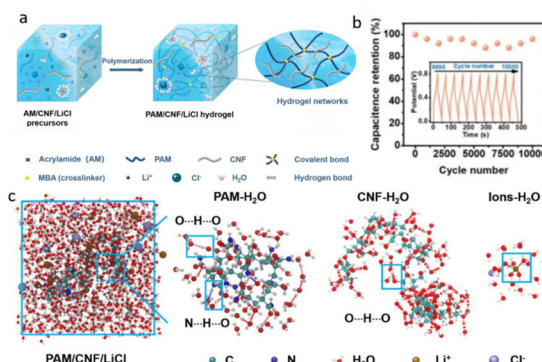


Figure 6. Reproduced with permission. Copyright 2021, Elsevier. a) Schematic illustration of the formation of PAM/CNF/ LiCl hydrogel. b) Cycling stability performance of the supercapacitor at 25°C . c) Equilibrium structures comprising PAM/CNF/ LiCl hydrogel and interactions of each component with water molecules.

5.2.4 M-DPAM-3 High-Power Gel Battery

Zhong et al. introduced high-concentration ZnCl_2 (acting as a pore-forming agent and salt additive) and lignin (acting as a scaffold repair agent) into PAM hydrogel through a one-pot method. They regulated the Zn^{2+} solvation structure with DMSO (dimethyl sulfoxide) to prepare M-DPAM-3 hydrogel. (Figure 7.c) ZnCl_2 reduced free water molecules through the "salt-in-water" mechanism, enhancing its anti-freezing ability. The linear sweep voltammetry (LSV) curve showed that the zinc-air battery assembled with the M-DPAM-3 hydrogel electrolyte could not only withstand higher current density, but also achieved an excellent power density of $136.2 \text{ mW} \cdot \text{cm}^{-2}$ at room temperature. (Figure 7.a) This is beneficial to the effective promotion of rapid ion transport within the gel's regular porous structure, significantly reducing the battery's internal resistance. (Figure 7.b) In terms of improving the anti-freezing performance of M-DPAM-3 hydrogel, it was possible to maintain an ion conductivity of up to $380.95 \text{ mS} \cdot \text{cm}^{-1}$ in LT environments, which was 13.6% lower than that at room temperature[11].

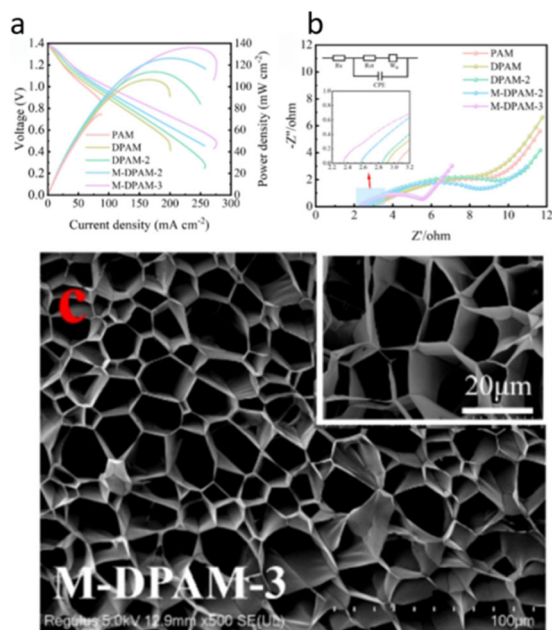


Figure. 7 Reproduced with permission. Copyright 2025, Elsevier. a) Comparison of LSV curves and power density curves. b) Impedance diagram of the flexible batteries. c) Cryo-SEM 500 $\times$ and 2000 $\times$ images of M- DPAM-3 hydrogels.

6 Conclusion

In the future, the research on gel electrolytes for LT aqueous zinc-ion batteries should not only focus on reducing their applicable operating temperature, but also pay attention to their electrochemical performance. This mainly includes aspects such as ion transport, stability, preparation processing. New polymer network structures can be designed to improve ion transport efficiency. Further optimization of the cross-linking method and cross-linking density of polymers can be carried out to achieve the goal of obtaining gels with both high

mechanical properties and flexibility, ensuring the stability and safety of batteries during use. By studying new polymer materials and additives, the activity of the gel electrolyte water can be reduced, and side reactions such as hydrogen evolution can be inhibited, thereby expanding the electrochemical stability window and increasing the working voltage and energy density of the battery.

Currently, the preparation of gel electrolytes usually involves processes such as free radical polymerization. However, this process is often difficult to control, resulting in unstable properties of the prepared gels. Therefore, to achieve mass production, it is urgent to develop more advanced polymerization technologies to achieve precise control over the polymerization process. At the same time, more simple and efficient preparation methods should be explored to reduce production costs and improve production efficiency.

References

1. Wang, Y., Liang, B., Li, D., Wang, Y., Li, C., Cui, H., ... & Zhi, C. (2025). Hydrogel electrolyte design for long-lifespan aqueous zinc batteries to realize a 99% Coulombic efficiency at 90° C. *Joule*, 9(6). <https://doi.org/10.1016/j.joule.2025.101944>
2. Gao, R., Wang, J., Song, Y., Li, K., Chen, Z., Shen, Q., & Wang, Y. (2025). Polymer-Salt Effects with Enhanced Eutectic Behavior in Hydrogel Electrolytes for Aqueous Zinc Batteries at- 70° C. *Advanced Functional Materials*, e14585. <https://doi.org/10.1002/adfm.202514585>
3. Wang, R., Ma, Q., Zhang, L., Liu, Z., Wan, J., Mao, J., ... & Zhang, C. (2023). An aqueous electrolyte regulator for highly stable zinc anode under- 35 to 65° C. *Advanced Energy Materials*, 13(40), 2302543. <https://doi.org/10.1002/aenm.202302543>
4. Xu, X., Li, S., Luo, Z., Zhang, C., Zhang, D., Hui, J., ... & Li, B. (2024). Ultra-wide temperature range aqueous electrolyte through local aggregation anchoring active water towards practical aqueous zinc metal battery. *Energy Storage Materials*, 71, 103567. <https://doi.org/10.1016/j.ensm.2024.103567>
5. Wei, T., Ren, Y., Li, Z., Zhang, X., Ji, D., & Hu, L. (2022). Bonding interaction regulation in hydrogel electrolyte enable dendrite-free aqueous zinc-ion batteries from- 20 to 60° C. *Chemical Engineering Journal*, 434, 134646. <https://doi.org/10.1016/j.cej.2022.134646>
6. Shi, Y., Wang, R., Bi, S., Yang, M., Liu, L., & Niu, Z. (2023). An anti-freezing hydrogel electrolyte for flexible zinc-ion batteries operating at- 70° C. *Advanced Functional Materials*, 33(24), 2214546. <https://doi.org/10.1002/adfm.202214546>
7. Huang, S., He, S., Li, Y., Wang, S., & Hou, X. (2023). Hydrogen bond acceptor lined hydrogel electrolyte toward Dendrite-Free aqueous Zn ion batteries with low temperature adaptability. *Chemical Engineering Journal*, 464, 142607.

<https://doi.org/10.1016/j.cej.2023.142607>

8. Zhang, F., Yang, M., Fang, P., Yu, J., Ma, X., Hu, Y., & Yan, F. (2024). Organohydrogel electrolytes with solvated structure regulation for highly reversible low-temperature zinc metal batteries. *Journal of Materials Chemistry A*, 12(6), 3470-3479. <https://doi.org/10.1039/D3TA07246K>
9. Guo, S. J., Yan, M. Y., Xu, D. M., He, P., Yan, K. J., Zhu, J. X., ... & Cao, F. F. (2025). Anti-freezing hydrogel electrolyte with a regulated hydrogen bond network enables high-rate and long cycling zinc batteries. *Energy & Environmental Science*, 18(1), 418-429. DOI: 10.1039/D4EE02772H
10. Zhang, X., Wang, J., Wang, M., Liu, D., & Wang, Z. (2024). In situ reduction strategy towards high conductivity, anti-freezing and super-stretchable rGO based hydrogel for diverse flexible electronics. *Nano Research*, 17(5), 4016-4022. <https://doi.org/10.1007/s12274-023-6267-9>
11. Zhong, D., Wang, K., Wei, M., Wang, H., & Pei, P. (2025). Enhanced Low-Temperature performance of flexible Zinc-Air batteries via High-Concentration ZnCl₂ and lignin modified polyacrylamide hydrogels. *Chemical Engineering Journal*, 510, 161596. <https://doi.org/10.1016/j.cej.2025.161596>
12. Jiao, M., Dai, L., Ren, H. R., Zhang, M., Xiao, X., Wang, B., ... & Cheng, H. M. (2023). A polarized gel electrolyte for wide-temperature flexible zinc-air batteries. *Angewandte Chemie International Edition*, 62(20), e202301114. <https://doi.org/10.1002/anie.202301114>
13. Huang, S., Hou, L., Li, T., Jiao, Y., & Wu, P. (2022). Antifreezing hydrogel electrolyte with ternary hydrogen bonding for high-performance zinc-ion batteries. *Advanced Materials*, 34(14), 2110140. <https://doi.org/10.1002/adma.202110140>
14. Ge, W., Cao, S., Yang, Y., Rojas, O. J., & Wang, X. (2021). Nanocellulose/LiCl systems enable conductive and stretchable electrolyte hydrogels with tolerance to dehydration and extreme cold conditions. *Chemical Engineering Journal*, 408, 127306. <https://doi.org/10.1016/j.cej.2020.127306>