



Synergistic PAM@MXene hydrogel interlayer regulates Sn deposition behavior for highly stable Sn anodes in aqueous tin-ion batteries

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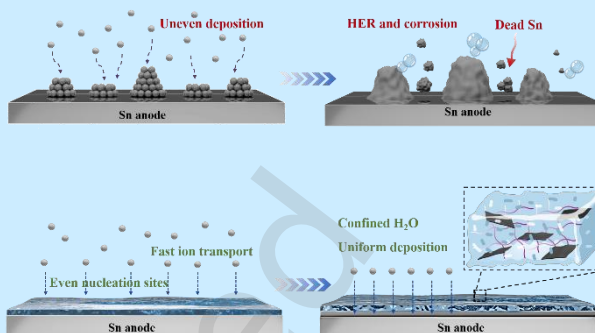
Synergistic PAM@MXene hydrogel interlayer regulates Sn deposition behavior for highly stable Sn anodes in aqueous tin-ion batteries

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The PAM@MXene hydrogel interlayer is constructed on Sn anode to engineer a stable interface, where PAM guides Sn^{2+} migration and confines water, while MXene provides nucleation sites, homogenizes electric field and facilitates ion transport. Their synergy effectively suppresses “dead Sn” formation and side reactions, enabling a highly stable Sn anode and offering a systematic strategy for high-performance aqueous tin-ion batteries.



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ABSTRACT

Aqueous tin-ion batteries represent a promising candidate for safe and cost-effective energy storage, owing to the dendrite-free property of Sn metal anode, but their development is hindered by interfacial instability that leads to the “dead Sn” accumulation and severe side reactions. Herein, an integrated organic-inorganic PAM@MXene composite hydrogel interlayer is constructed on the Sn metal anode to engineer a stable interface. The hydrophilic PAM network guides Sn²⁺ migration and confines interfacial water, while the MXene homogenizes the electric field, promotes ion transport and provides uniform nucleation sites. Their synergy fundamentally optimizes the nucleation kinetics, deposition morphology and interfacial reactivity, leading to the effective inhibition of “dead Sn” accumulation and side reactions. As a result, the symmetric cell assembled with PAM@MXene-Sn electrodes exhibits stable operation for 2,600 h, and the PAM@MXene-Sn||Cu asymmetric cell achieves an ultra-long life of 20,000 cycles with an average Coulombic efficiency of 99.99%. Also, the PAM@MXene-Sn||MnO₂ full cell exhibits negligible capacity decay after 6,000 cycles at 20 mA cm⁻². This work not only presents an effective interfacial design for stable Sn anodes, but also demonstrates a generalizable strategy of organic-inorganic synergy for regulating interfacial behavior for developing highly reversible tin-ion batteries.

KEYWORDS

Sn anode, hydrogel interlayer, MXene, “dead Sn”, aqueous tin-ion batteries

1 Introduction

High-performance electrochemical energy storage is critical for enabling the widespread use of intermittent renewable energy sources [1, 2], yet the safety and cost concerns of the widely used lithium-ion batteries necessitate the development of inherently safe and low-cost alternatives [3-5]. Aqueous rechargeable batteries based on metal anodes, particularly aqueous zinc-ion batteries, have emerged as promising candidates due to their intrinsic safety, environmental friendliness, and cost-effectiveness [6]. However, the commonly studied metal anodes, such as Zn [7-9], Al [10, 11], and Fe [12, 13], suffer from thermodynamic instability and highly anisotropic deposition behavior during plating/stripping, which often lead to uncontrolled dendritic growth and potential internal short-circuit failures. Moreover,

parasitic reactions at the metal anode/electrolyte interface, including hydrogen evolution (HER) and corrosion, result in low Coulombic efficiency (CE), gas generation, and the accumulation of passivating byproducts [14]. These issues are closely interrelated and collectively severely limit the cycling stability and efficiency of aqueous battery systems [15]. Therefore, the development of highly stable, dendrite-free metal anodes is essential for advancing the practical aqueous battery technologies.

Recently, aqueous tin-ion batteries have gained attention due to the unique properties of the Sn metal anode. Sn presents an ideal redox potential (-0.14 V vs. SHE), a high theoretical capacity (451.6 mAh g⁻¹ based on 2e⁻ transfer reaction), and natural abundance [16, 17]. Notably, its body-centered tetragonal crystal structure promotes isotropic deposition, facilitating the formation of nodules rather than

dendrites, which is a key feature for achieving dendrite-free plating behavior. Furthermore, its relatively low Young's modulus (~42 GPa) is beneficial for alleviating mechanical stress from volume changes during cycling, in favor of good mechanical durability [18]. These intrinsic properties, especially its dendrite-free deposition characteristic, collectively render aqueous tin-ion batteries highly competitive against other aqueous systems in terms of safety and long-term cycling stability, positioning them as a very promising candidate for next-generation aqueous batteries.

Despite these advantages, the practical application of Sn anodes remains severely constrained by the “dead Sn” accumulation and side reactions, which fundamentally originate from the uneven interfacial electric field, unfavorable Sn^{2+} deposition kinetics, and the unstable interfacial environment. The inhomogeneous charge distribution and excessively fast deposition result in rampant and non-uniform Sn nucleation and localized overgrowth [19]. During subsequent stripping, these loosely attached grains preferentially dissolve from the root and eventually evolve into electrochemically inactive “dead Sn” [20]. The resultant rough morphology significantly exacerbates the parasitic HER and corrosion, and the generated gas bubbles continuously scour the anode surface, further accelerating the “dead Sn” formation and undermining the interfacial stability [21]. Therefore, achieving stable Sn anodes necessitates a multifunctional interfacial engineering strategy that homogenizes electric field and ion flux, regulates deposition kinetics, suppresses side reactions and maintains structural integrity [22–24]. However, research on this crucial interfacial stabilization remains at an early stage, with only about a dozen studies reported to date primarily focusing on alloy design [20] and electrolyte modulation [25, 26]. For instance, Lu et al. [27] introduced triethyl phosphate as an electrolyte additive to reconstruct the Sn^{2+} solvation structure and adsorb on the Sn surface through P=O groups, regulating the deposition kinetics and enabling the symmetric cell to stably cycle for 760 h. The development of novel interfacial designs that can synergistically address the interlinked challenges is essential to stabilize the Sn metal anode and realize its full practical potential.

Notably, hydrogel-based interfacial layers, inspired by their utilization in other aqueous systems [28–30], provide a foundational approach for regulating interfaces through physical barriers and ion guidance. While their insulating property and insufficient mechanical strength may lead to uneven charge distribution and structural degradation [31–34], the integration of conductive fillers to construct organic-inorganic composite interfacial layers is an effective strategy to enhance ion transport and mechanical robustness [35–36]. Therefore, to engineer a multifunctional interface that can synergistically homogenize charge distribution, optimize deposition kinetics, and suppress side reactions, this work designs and constructs a composite hydrogel consisting of polyacrylamide (PAM) matrix and MXene nanosheets,

denoted as PAM@MXene, on the Sn electrode as an interfacial layer. In this design, the flexible and hydrophilic PAM matrix acts as a physical barrier against the bulk electrolyte while guiding Sn^{2+} migration and facilitating its de-solvation, and the uniformly dispersed MXene nanosheets with abundant terminations adsorb Sn^{2+} and confine interfacial H_2O , provide homogeneous nucleation sites, and ensure the structural integrity of the composite interlayer. This integrated design effectively enables uniform Sn deposition and suppresses the formation of “dead Sn” and associated side reactions. Consequently, the PAM@MXene-Sn electrode exhibits highly reversible plating/stripping behavior with a cycling life of over 2,600 h in the symmetric cell and an average CE of 99.99% for 20,000 cycles in the Sn|Cu asymmetric cell. Beyond presenting an effective multifunctional interlayer for Sn anodes, this work demonstrates a synergistic strategy for multifaceted interfacial regulation, offering a versatile interface design for high-performance aqueous Sn-based batteries.

2 Results and discussion

To regulate the interfacial environment and Sn^{2+} deposition behavior, the PAM@MXene composite hydrogel interlayer was constructed on the Sn electrode surface (denoted as PAM@MXene-Sn) via in situ UV-induced polymerization (Figure 1a and Figure S1). The polymerization process directly yields a well-adhered hydrogel interfacial layer with a hydrophilic and interconnected network, which enables the spontaneous confinement of water molecules, thereby creating a water-depleted interfacial microenvironment, conducive to subsequent ion migration and side reaction suppression. For comparison, an electrode modified with the pure PAM interlayer was also prepared and denoted as PAM-Sn. The top-view and cross-sectional SEM images in Figure 1b and Figure S2 show homogeneous and continuous coatings on both PAM-Sn and PAM@MXene-Sn electrodes. The corresponding EDS mappings (Figure 1c) verify the uniform dispersion of MXene nanosheets within the PAM matrix. Moreover, the PAM@MXene hydrogel interlayer adheres tightly to the Sn metal substrate with a thickness of approximately 35 μm (Figure 1d), indicating good structural integrity that is beneficial for mitigating active Sn delamination, accommodating volume changes, and maintaining stable interfacial microenvironment. Furthermore, the XRD patterns (Figure 1e and Figure S3) of the PAM-Sn and PAM@MXene-Sn electrodes reveal the sharp diffraction peaks corresponding to the metallic Sn substrate (PDF#04-6073) and a broad halo characteristic of the amorphous PAM hydrogel, confirming that the construction of the interfacial hydrogel layer does not alter the bulk crystal structure of the Sn substrate [37].

To investigate the molecular interactions within the interlayers, Raman and FTIR spectra were performed for

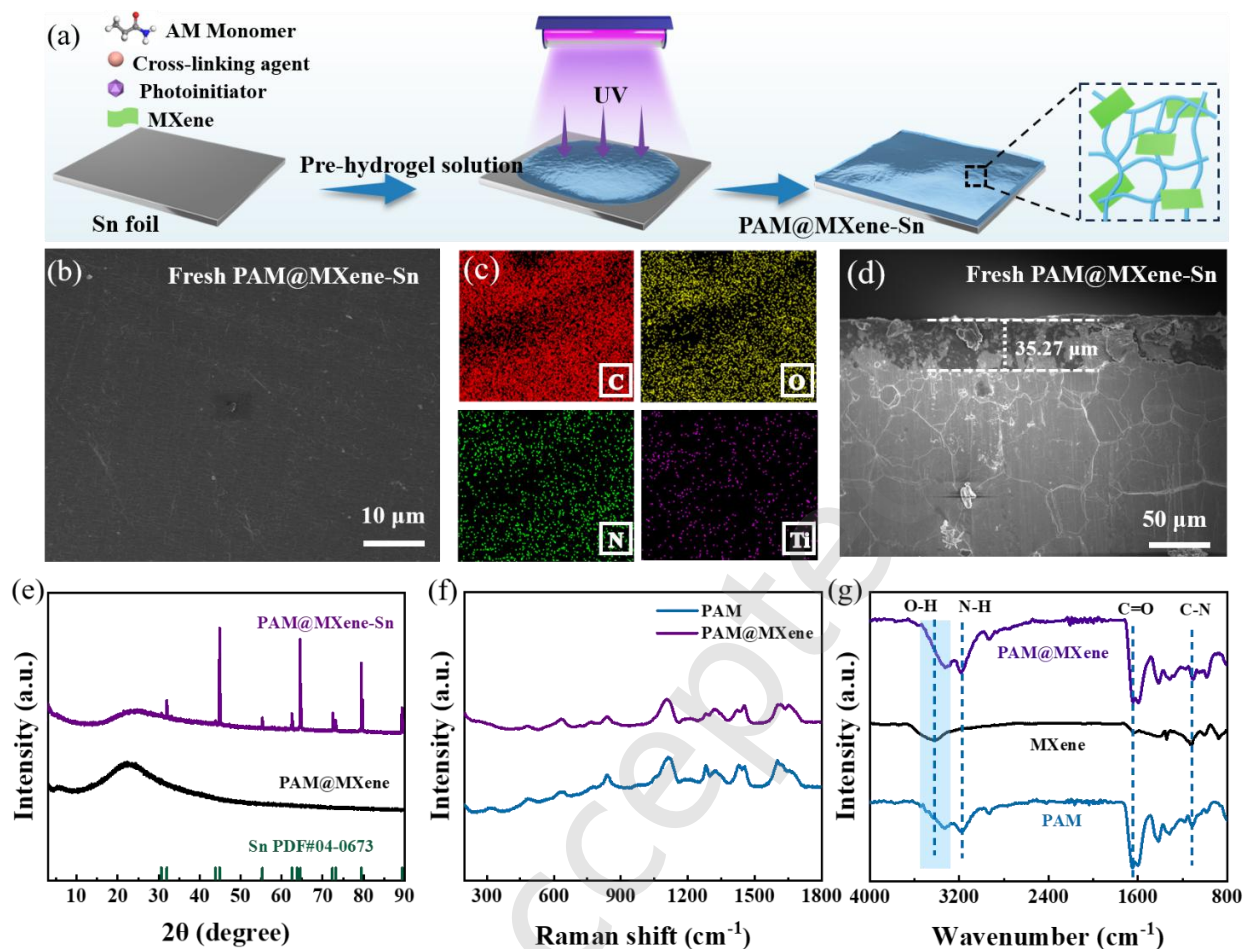


Figure 1 Preparation and characterizations of PAM@MXene-Sn electrode. (a) Schematic illustration of preparation of PAM@MXene-Sn electrode. (b) Top-view SEM image with (c) EDS elemental mappings and (d) cross-sectional SEM image of PAM@MXene-Sn electrode. (e) XRD patterns of PAM@MXene-Sn electrode and PAM@MXene hydrogel. (f) Raman spectra of PAM and PAM@MXene hydrogels. (g) FTIR spectra of PAM@MXene, PAM and MXene.

the PAM and PAM@MXene hydrogels. The Raman spectrum of PAM@MXene (Figure 1f) retains all characteristic vibration modes of pure PAM at 350, 835, 1104, 1290, and 1425 cm^{-1} , corresponding to δ (C-C-C), ν (C-C) side-chain, C-C skeletal stretching, C-H bending, and C-N stretching, respectively [38-39], indicating the integrity of the polymer framework after MXene integration. The O-H stretching region (3000-3800 cm^{-1}), which can be deconvoluted into three distinct characteristic peaks representing strong, moderate, and weak hydrogen bonding interactions, elucidates the evolution of hydrogen-bonding networks within the hydrogel electrolyte. As shown in Figure S4, the proportion of strong hydrogen bonds progressively decreases, while that of weak hydrogen bonds becomes more prominent with increasing MXene content, indicating that the MXene incorporation disrupts the native hydrogen bonding network

of water molecules. This evolution is further supported by ^2H nuclear magnetic resonance (NMR) (Figure S5), where the addition of MXene induces the ^2H signal of H_2O to shift to a higher field, indicating increased electron density around the hydrogen atoms due to the formation of new intermolecular hydrogen bonds between MXene and water molecules. In the FTIR spectra (Figure 1g), the characteristic peaks at 3179, 1648, and 1102 cm^{-1} , corresponding to N-H, C=O, and C-N stretching vibrations, are observed for the PAM hydrogel, while the peaks at 3421 and 1107 cm^{-1} , related to O-H and C=O stretching vibrations, are present for MXene. For the PAM@MXene hydrogel, the characteristic peaks of PAM shift to 3183 cm^{-1} (N-H), 1651 cm^{-1} (C=O), and 1110 cm^{-1} (C-N), indicating the formation of hydrogen-bonding interactions between the oxygen-containing groups on the MXene and the $-\text{NH}_2$ groups on the

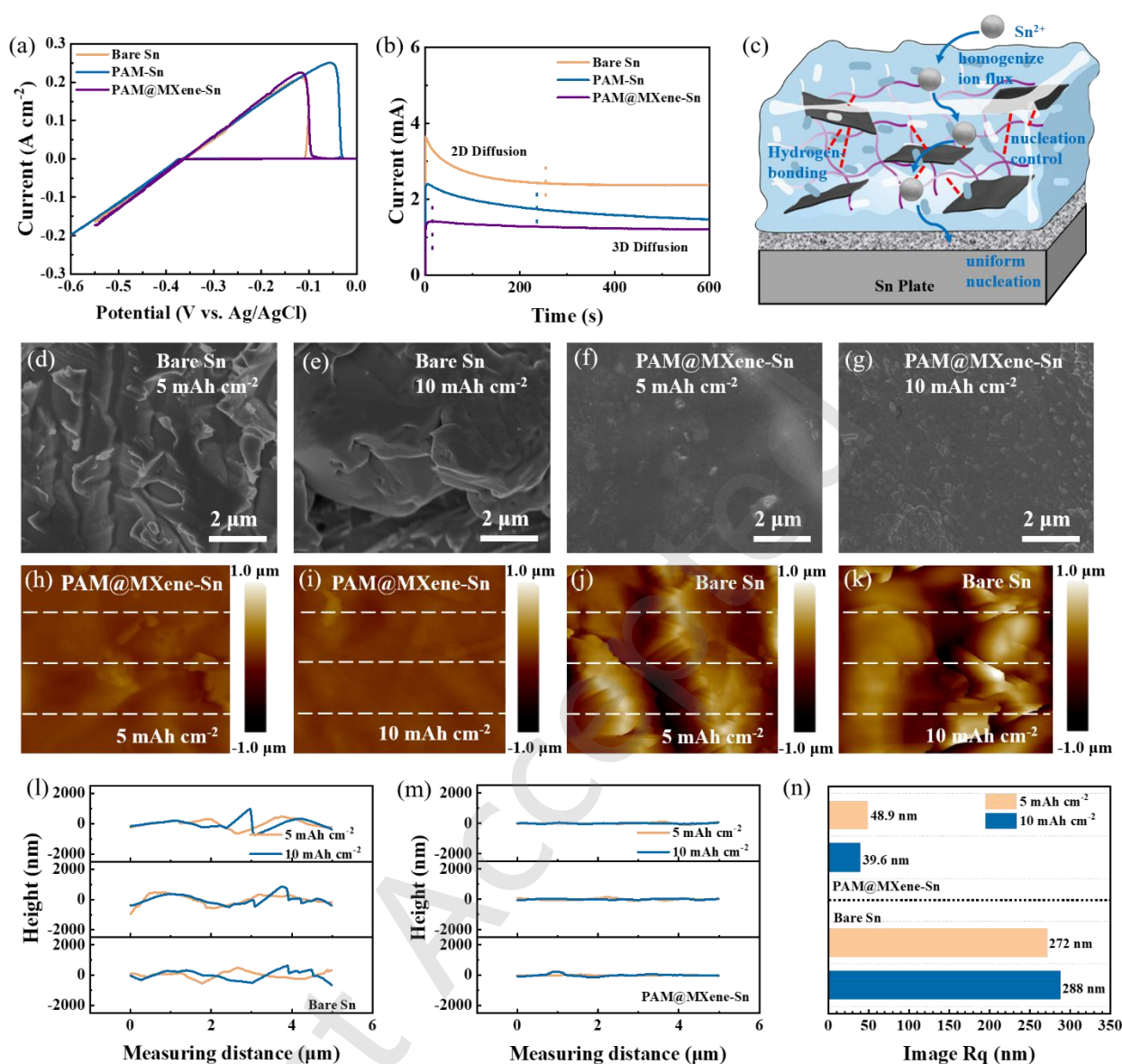


Figure 2 Deposition behavior and morphology of bare Sn, PAM-Sn, and PAM@MXene-Sn electrodes. (a) CV curves of Sn||Cu asymmetric cells at 5 mV s⁻¹. (b) CA curves at an overpotential of 10 mV. (c) Schematics of Sn²⁺ deposition behavior on PAM@MXene-Sn. SEM images of (d, e) bare Sn and (f, g) PAM@MXene-Sn with deposition capacities of 5 mAh cm⁻² and 10 mAh cm⁻². AFM images of (h, i) PAM@MXene-Sn and (j, k) bare Sn. Height distribution of (l) bare Sn and (m) PAM@MXene-Sn and (n) surface roughness with different deposition capacities.

PAM chains [40]. Remarkably, these hydrogen-bonding interactions not only enhance the dispersion of the MXene nanosheets within the PAM matrix and strengthen the interfacial adhesion, thereby ensuring the structural integrity of the composite interlayer, but also contribute to a tailored interfacial environment by affecting the solvation structure of Sn²⁺, facilitating the de-solvation process and moderating the subsequent deposition kinetics [41].

To analyze the chemical states and interactions of the components in the hydrogels, XPS spectra were employed, revealing that the intensities of the C 1s and O 1s peaks are significantly enhanced in the PAM@MXene hydrogel compared to pure MXene (Figure S6). Notably, the

high-resolution C 1s spectra exhibit a shift of the Ti-C peak from 281.8 eV in MXene to 283.4 eV in PAM@MXene hydrogel [42]. This shift suggests the redistribution of the electron density resulting from the hydrogen-bonding interactions between PAM and MXene, which both reinforces the mechanical integrity of the organic-inorganic composite network and facilitates interfacial charge transfer for achieving uniform and dense Sn deposition.

The Sn²⁺ deposition behavior modulated by the PAM@MXene composite hydrogel interlayer is systematically explored, including nucleation kinetics, interfacial ion transport, deposition morphology, and the side reactions. The nucleation kinetics on bare Sn, PAM-Sn and

PAM@MXene-Sn electrodes are obtained by cyclic voltammetry (CV) as shown in Figure 2a and Figure S7. The PAM@MXene-Sn electrode exhibits a moderate nucleation overpotential of 7.15 mV at a scan rate of 5 mV s⁻¹, which lies between the lower value of bare Sn (4.86 mV) and the higher value of PAM-Sn (8.61 mV). An excessively low overpotential promotes rapid and localized nucleation prone to “dead Sn” formation, whereas an overly high overpotential may reflect increased kinetic resistance and polarization. Therefore, this moderate overpotential enabled by the PAM@MXene composite hydrogel corresponds to optimized nucleation kinetics that are critical for uniform dense Sn deposition alongside efficient interfacial charge transport.

Furthermore, as displayed in the chronoamperometry (CA) curves (Figure 2b), the current density for the bare Sn electrode decreases continuously over time, suggesting a 2D diffusion-controlled process that leads to uneven Sn deposition. In contrast, the PAM@MXene-Sn electrode exhibits rapid current stabilization following a brief nucleation period of only ~15 s, implying progressive 3D nucleation and growth mode that favors homogeneous and densely packed Sn plating. The improved deposition behavior is underpinned by enhanced interfacial ion transport, as evidenced by the enhanced ionic conductivity of 36.51 mS cm⁻¹ for PAM@MXene hydrogel (Figure S8), and the increased Sn²⁺ transfer number of 0.806 for the PAM@MXene-Sn electrode (Figure S9), confirming that the composite hydrogel interlayer significantly facilitates the Sn²⁺ diffusion toward the electrode surface.

The underlying mechanism for the optimized ion migration and deposition kinetics as well as the resultant improved deposition uniformity is depicted in Figure 2c. On bare Sn, the ultra-fast deposition kinetics promotes random growth from initial nuclei, forming large particles that readily detach into “dead Sn”. The pure PAM hydrogel interlayer, though beneficial for interfacial stability by confining water molecules and accommodating volume changes, suffers from high nucleation barrier due to its insulating property. In contrast, the PAM@MXene composite hydrogel interlayer achieves a synergistic regulation of the interface, in which the PAM matrix maintains its role in guiding ion migration, confining water and buffering volume changes, while the incorporated MXene nanosheets provide numerous uniform nucleation sites, ensure sufficient Sn²⁺ supply, and homogenize the interfacial electric field. Besides, the hydrogen-bonding interactions between PAM and MXene reinforce the structural integrity of the composite interlayer and improve its adhesion with the electrode. These coordinated functions collectively homogenize the Sn²⁺ flux, modulate the nucleation kinetics, promote uniform Sn deposition and suppress side reactions, which is expected to endow the Sn electrode with outstanding performance.

The uniformity of Sn deposition is directly visualized by

examining the morphologies of different electrodes after plating at a current density of 1 mA cm⁻² for 5 h and 10 h. The bare Sn electrode is covered by irregular Sn particles at 5 mAh cm⁻² (Figure 2d and Figure S10), which continue to grow and evolve into inactive “dead Sn” with the capacity further increasing to 10 mAh cm⁻² (Figure 2e). By comparison, the Sn electrodes modified with PAM and PAM@MXene hydrogel interlayers display much more uniform surfaces at both deposition stages (Figure 2f-g and Figure S11-S12). Especially, the PAM@MXene-Sn electrode maintains a remarkably smooth and flat surface even at 10 mAh cm⁻² (Figure 2g), demonstrating the effectiveness of the composite hydrogel interlayer in suppressing “dead Sn” formation. This superior uniformity of Sn deposition is further corroborated by atomic force microscopy (AFM), which reveals exceptionally smooth and defect-free surfaces on the PAM@MXene-Sn electrode after 5 and 10 mAh cm⁻² of plating (Figure 2h-i and Figure S13), in stark contrast to the irregular, rough surfaces of bare Sn and PAM-Sn electrodes (Figure 2j-k and Figure S14-S15). Additionally, height profiles confirm the uniformity on the PAM@MXene-Sn electrode (Figure 2l-m), with the quantitative roughness analysis showing an average height variation of only 39.6 nm, significantly lower than that of PAM-Sn (173 nm) and bare Sn (288 nm) (Figure 2n and Figure S16-S17). In situ optical microscopy of the plating process at 10 mA cm⁻² provides additional evidence (Figure S18). While the irregular protrusions rapidly form on the bare Sn electrode within 10 min, the PAM@MXene-Sn electrode exhibits a compact morphology throughout the entire observation period of 30 min. These results demonstrate that the PAM@MXene interlayer effectively regulates the Sn²⁺ nucleation and growth behavior and establishes stable interfacial environment with efficient ion transport and mechanical stability, thereby promoting uniform Sn deposition, and minimizing the formation of coarse Sn particles and “dead Sn”.

To elucidate the interfacial mechanism of the composite hydrogel interlayer on regulating Sn²⁺ deposition behavior and alleviating H₂O-induced side reaction, density functional theory (DFT) calculations were performed to evaluate the binding energies of Sn²⁺ and H₂O on PAM and MXene. As shown in Figure 3a and Figure S19, the significantly stronger binding energy of Sn²⁺ on MXene (-0.270 eV) compared to that on PAM fragments (-0.151 eV) and H₂O (-0.229 eV) indicates a pronounced thermodynamic preference of Sn²⁺ for the MXene surface, which provides abundant and uniform active nucleation sites. This preferential adsorption, combined with the ability of the PAM matrix to guide and homogenize ion flux through its hydrophilic network, ensures uniform supply of Sn²⁺ to the electrode surface, thereby promoting even nucleation and deposition. Regarding the water-induced side reactions, the binding energies of H₂O on MXene

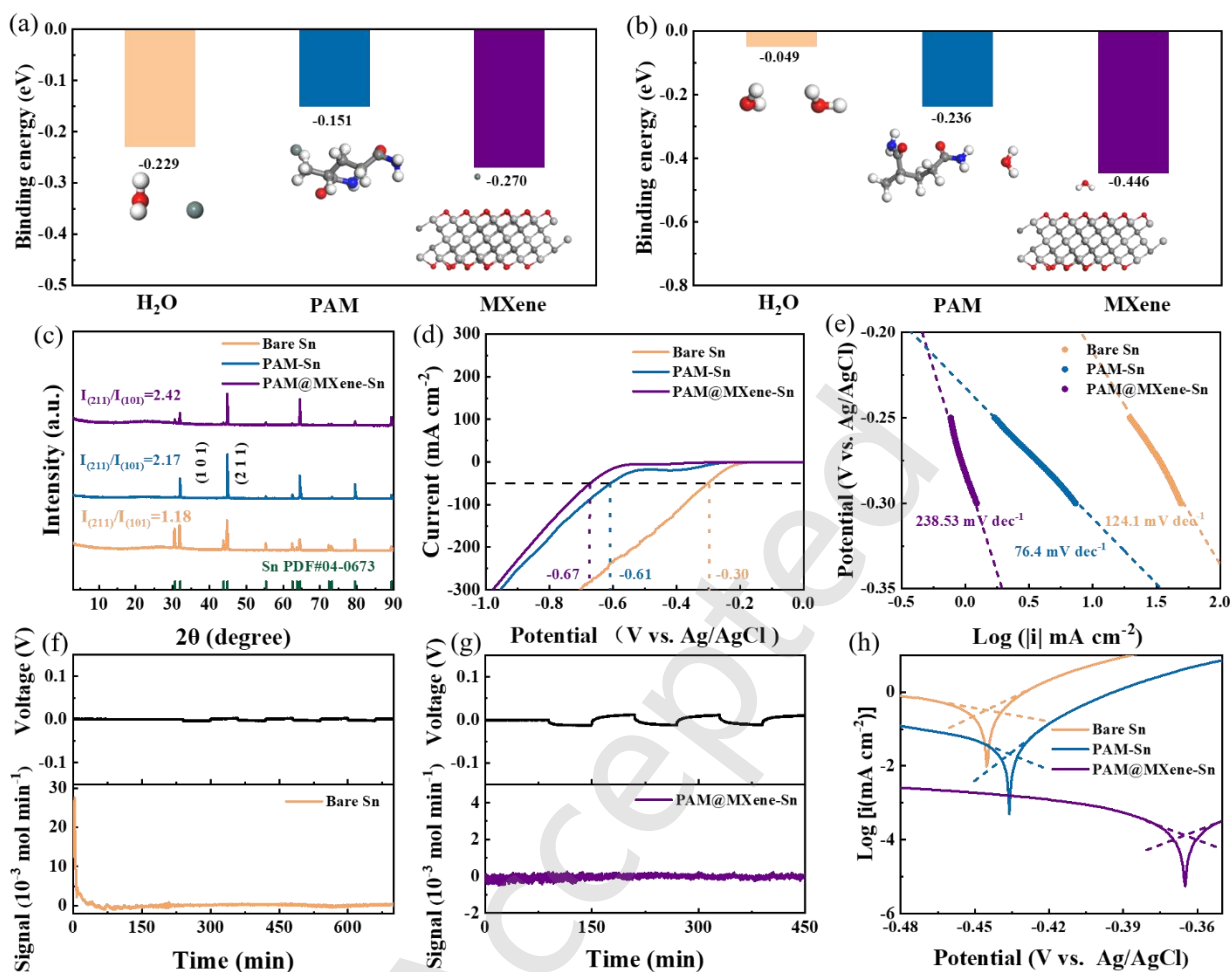


Figure 3 Side reactions on bare Sn, PAM-Sn and PAM@MXene-Sn electrodes. Binding energies of H₂O, PAM, and MXene with (a) Sn²⁺ and (b) H₂O. (c) XRD patterns of different electrodes after 100 h of cycling. (d) LSV curves and (e) Tafel slopes of different electrodes. In situ DEMS signals of (f) bare Sn and (g) PAM@MXene-Sn electrodes. (h) Linear polarization curves.

(-0.446 eV) and on PAM (-0.236 eV) are substantially stronger than the intermolecular H₂O⋯H₂O interaction (-0.049 eV), suggesting that both components can immobilize interfacial H₂O molecules with MXene exhibiting superior confinement capability (Figure 3b and Figure S20). Combined with the Raman-evidenced increase in weak hydrogen bond proportion upon MXene incorporation, these results reveal the disruption of the original H₂O⋯H₂O interactions and the reconstruction of the hydrogen-bond network. It not only facilitates Sn²⁺ de-solvation and transport, but also converts highly reactive bulk-like water into confined, low-activity water at the interface, thereby suppressing the associated interfacial side reactions.

Building on the enhanced confinement of interfacial H₂O molecules and the spatial shielding of the PAM@MXene hydrogel interlayer, its role in alleviating side reactions is evaluated experimentally. The bare Sn electrode exhibits a contact angle of 74.1° with 3 M methanesulfonic acid (MSA)

+ 1 M tin trifluoromethanesulfonate (Sn(MSA)₂) electrolyte, which decreases to 63.5° for the PAM-Sn electrode owing to the intrinsic hydrophilicity of the abundant amide groups on PAM chains (Figure S21). Remarkably, the PAM@MXene-Sn electrode delivers an ultralow contact angle of 35.3°, ascribed to the synergistic contribution of the highly hydrophilic surface terminations on the MXene nanosheets and the increased water accessibility at the organic-inorganic interface. This superior wettability is critical for establishing continuous and efficient ion transport pathways, which is beneficial for homogenizing the interfacial ion flux and alleviating local concentration gradients. Furthermore, the electric double-layer capacitance (C_{dl}), derived from the CV curves at various scan rates, decreases from 9.51 μF cm⁻² for bare Sn to 4.49 μF cm⁻² for PAM-Sn and further to 3.31 μF cm⁻² for PAM@MXene-Sn (Figure S22). It reflects that the hydrogel interlayer decreases the apparent electrochemically

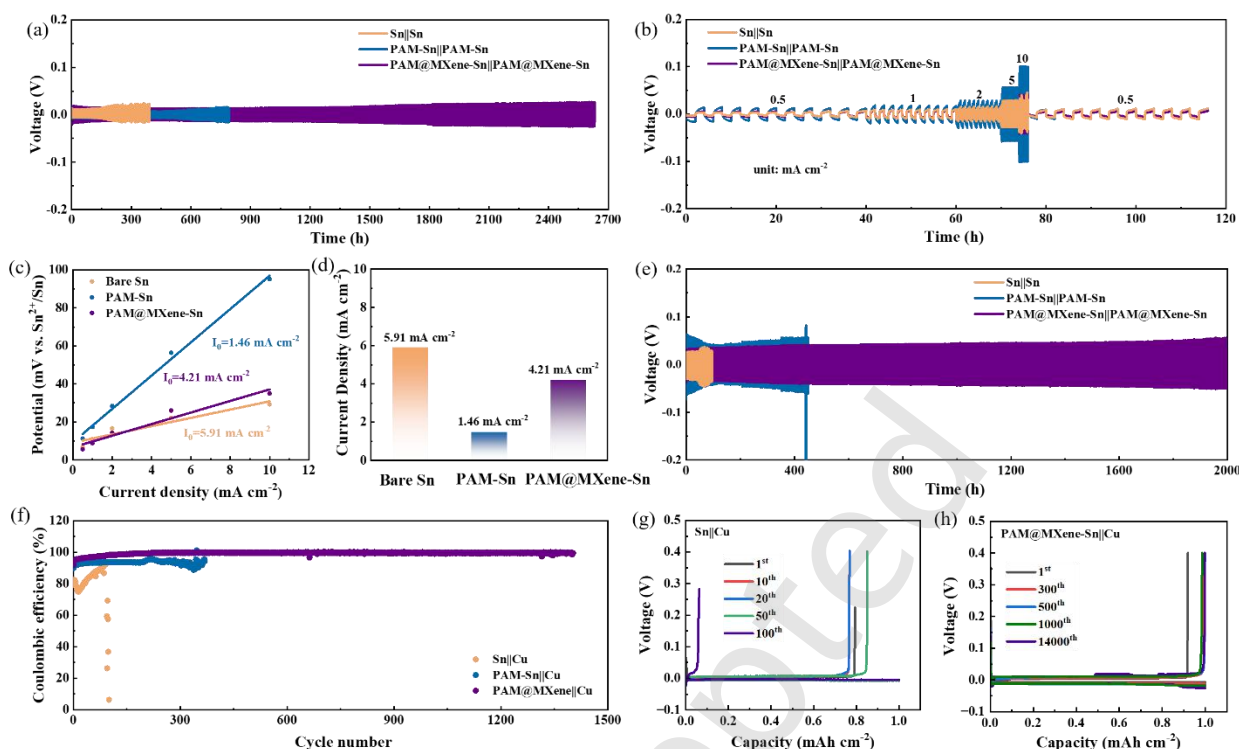


Figure 4 Sn plating/stripping behavior in (a-e) symmetric and (f-h) asymmetric cells based on bare Sn, PAM-Sn, and PAM@MXene-Sn electrodes. (a) Cycling performance at 1 mA cm^{-2} , 1 mAh cm^{-2} , (b) rate performance, (c) exchange current density and (d) the corresponding bar chart, (e) cycling performance at 10 mA cm^{-2} , 0.5 mAh cm^{-2} of symmetric cells. (f) CEs of Sn||Cu asymmetric cells at 1 mA cm^{-2} , 1 mAh cm^{-2} , and charge-discharge curves of (g) bare Sn||Cu and (h) PAM@MXene-Sn||Cu asymmetric cells.

accessible area of the Sn electrode, thereby effectively reducing the available sites for side reactions and facilitates uniform Sn deposition.

To further verify the suppression of side reactions by the hydrogel interlayer, the crystallographic orientation of deposited Sn on different electrodes is analyzed by XRD. After 100 h of Sn plating/stripping, the intensity ratio of the diffraction peaks corresponding to Sn (211) and Sn (101) planes ($I_{(211)}/I_{(101)}$) increases from 1.18 for bare Sn, to 2.17 for PAM-Sn and 2.42 for PAM@MXene-Sn (Figure 3c), demonstrating that the hydrogel interlayers, particularly the PAM@MXene interlayer, effectively directs Sn^{2+} deposition along the (211) plane. Compared to Sn (101) surface, the Sn (211) plane favors more homogeneous interfacial electric field distribution and offers low self-diffusion energy barrier of Sn, thereby promoting dense and planar deposition [43]. Moreover, the Sn (211) surface possesses a significantly higher Gibbs free energy for H^* formation (ΔG_{H^*} , 1.34 eV) than that on Sn (101) surface (0.25 eV), indicating extremely low HER activity. The induction of this favorable Sn (211) orientation by the PAM@MXene interlayer is therefore a key factor contributing to uniform deposition and suppressed HER.

The HER activity of different electrodes is exclusively evaluated using linear sweep voltammetry (LSV) in Sn^{2+} -free 3 M MSA electrolyte to avoid the interference from

concurrent Sn^{2+} deposition. At a current density of 50 mA cm^{-2} , the HER overpotential of the PAM@MXene-Sn and the PAM-Sn reaches -0.67 V and -0.61 V (vs. Ag/AgCl), respectively, significantly more negative than that of bare Sn (-0.30 V vs. Ag/AgCl), verifying the substantial suppression of HER (Figure 3d), in good agreement with the preferential Sn (211) orientation induced by the hydrogel interlayer. Consistent with this trend, the higher Tafel slopes observed for the modified electrodes also confirm the effective mitigation of HER kinetics (Figure 3e). Furthermore, the HER process was directly monitored in symmetric cells using in situ differential electrochemical mass spectrometry (DEMS). For bare Sn, a pronounced H_2 signal is detected even in the initial open-circuit stage, which intensifies substantially during subsequent cycling (Figure 3f). In contrast, the H_2 signal remarkably decreases for the PAM-Sn and further diminishes to a nearly undetectable level for the PAM@MXene-Sn throughout the entire measurement (Figure 3g and Figure S23), directly evidencing the efficient HER suppression achieved by the composite hydrogel interlayer at the interface.

Moreover, the corrosion resistance of the different Sn electrodes is explored by linear polarization curves (Figure 3h). The corrosion potential of the PAM@MXene-Sn (-0.365 V vs. Ag/AgCl) is remarkably higher than that of bare Sn (-0.445 V vs. Ag/AgCl) and the PAM-Sn (-0.436 V vs.

Ag/AgCl), while its corrosion current is obviously lower than the other two electrodes, demonstrating the highest corrosion resistance and the much decreased corrosion rate owing to the multiple synergistic functions of the composite PAM@MXene hydrogel interlayer. It acts as a physical barrier that minimizes direct exposure of the Sn electrode to the electrolyte, while the hydrogen-bonding network within the composite facilitates Sn^{2+} de-solvation and confines interfacial H_2O molecules, thereby reducing their activity and availability for side reactions. Together with the induced preferential deposition along the Sn (211) plane, the interfacial side reactions are significantly suppressed, contributing to the much-improved overall stability of the Sn electrode.

The improved stability and reversibility enabled by the PAM@MXene hydrogel interlayer, which effectively suppresses “dead Sn” formation and side reactions, are further validated through electrochemical plating/stripping cycling in Sn||Sn symmetric cells. As shown in Figure 4a and Figure S24, under a condition of 1 mA cm^{-2} and 1 mAh cm^{-2} , the bare Sn symmetric cell suffers from a short circuit after merely 400 h, while the PAM-Sn cell, despite initially exhibiting low polarization, fails after ~ 800 h of cycling, indicating the inadequate protection offered by the pure PAM hydrogel. However, the cell equipped with the PAM@MXene-Sn electrode retains a stable polarization voltage of ~ 30.7 mV for 2,600 h, revealing highly stable plating/stripping behavior. The SEM characterization of the cycled electrodes provides direct morphological evidence for this enhanced stability (Figure S25). After 50 h and 100 h of cycling, the bare Sn electrode becomes progressively rougher due to the accumulation of “dead Sn” from the heterogeneous deposition/dissolution processes. Both hydrogel-modified Sn electrodes show flat surfaces with no obvious protrusions after cycling. Notably, the PAM@MXene-Sn electrode exhibits an exceptionally smooth and dense morphology, consistent with its superior electrochemical stability and confirming the efficacy of the composite layer in guiding uniform deposition.

The electrochemical kinetics influenced by the PAM@MXene hydrogel interlayer are evaluated through electrochemical impedance spectroscopy (EIS) and rate capability tests. The EIS spectra of symmetric cells show that the PAM@MXene-Sn electrode possesses the lowest charge transfer resistance (Figure S26), demonstrating the effectiveness of the composite hydrogel interlayer in facilitating interfacial charge transfer and ion transport during Sn plating/stripping. Moreover, the PAM@MXene-Sn remains stable charge-transfer resistance over cycling, whereas that of the bare Sn increases markedly due to heterogeneous deposition, “dead Sn” accumulation, and continuous interfacial deterioration, confirming the stability of the regulated interface (Figure S27). This improved kinetics directly contribute to outstanding rate performance. As the current density progressively increases from 0.5 to 10

mA cm^{-2} , the symmetric cell with the PAM@MXene-Sn electrode maintains stable voltage profiles (Figure 4b), which is attributed to the high ion flux and promoted nucleation at the regulated interface.

To gain deeper insight into the deposition kinetics, the exchange current density (i_0) is quantified. As displayed in Figure 4c-d, the bare Sn electrode exhibits an excessively high i_0 of 5.91 mA cm^{-2} , which tends to induce rapid, uncontrolled growth of large Sn particles and results in irregular, unstable deposits. In contrast, the PAM-Sn electrode shows a relatively low i_0 of only 1.46 mA cm^{-2} , indicating sluggish reaction kinetics and ultra-high nucleation barrier that leads to sparse and inefficient deposition. Significantly, the PAM@MXene-Sn electrode exhibits a moderate and optimized i_0 (4.21 mA cm^{-2}). This balanced kinetics, enabled by the organic-inorganic composite hydrogel design, is most favorable for facilitating compact and adherent Sn plating, thereby enhancing the structural integrity and electrochemical cycling stability of

the electrode. Consequently, even at a high current density of 10 mA cm^{-2} , the PAM@MXene-Sn cell still achieves a long cycling life of 2,000 h with a steady polarization voltage of ~ 34.5 mV (Figure 4e and Figure S28). Collectively, the evidence from EIS, rate performance, and kinetic parameter analysis consistently demonstrates that the PAM@MXene hydrogel interlayer establishes superior interfacial charge transfer kinetics and effectively mitigates the accumulation of “dead Sn”, providing a robust foundation for highly stable and reversible Sn metal anodes.

The reversibility of the Sn plating/stripping process in different anodes is quantitatively explored in Sn||Cu asymmetric cells. The bare Sn||Cu asymmetric cell undergoes a pronounced decline in CE after only 100 cycles at 1 mA cm^{-2} and 1 mAh cm^{-2} (Figure 4f), accompanied by a clear short-circuit signature in the galvanostatic charge/discharge profiles (Figure 4g). The PAM-Sn||Cu asymmetric cell, benefiting from the interfacial regulation of the pure PAM hydrogel interlayer, extends the operation to nearly 350 cycles (Figure S29). Remarkably, the PAM@MXene-Sn||Cu asymmetric cell reveals significantly enhanced reversibility, delivering a high average CE of 99.41% for 1,400 cycles with stable voltage hysteresis under the identical conditions (Figure 4h). The beneficial effect of the PAM@MXene hydrogel interlayer becomes even more pronounced under high-rate cycling. At 10 mA cm^{-2} and 0.5 mAh cm^{-2} , the bare Sn||Cu asymmetric cell fails after $\sim 1,300$ cycles and the PAM-Sn||Cu asymmetric cell sustains 6,000 cycles with an average CE of 99.98% (Figure S30). However, the PAM@MXene-Sn||Cu cell possesses an ultra-long cycling life of 20,000 cycles with a stabilized CE of 99.99%. Additionally, the charge/discharge voltage gap of the PAM@MXene-Sn||Cu asymmetric cell is only 37.52 mV, significantly lower than those of the PAM-Sn||Cu (55.18 mV) and bare Sn||Cu (150.82 mV) cells (Figure S31), suggesting excellent interfacial kinetics and exceptional

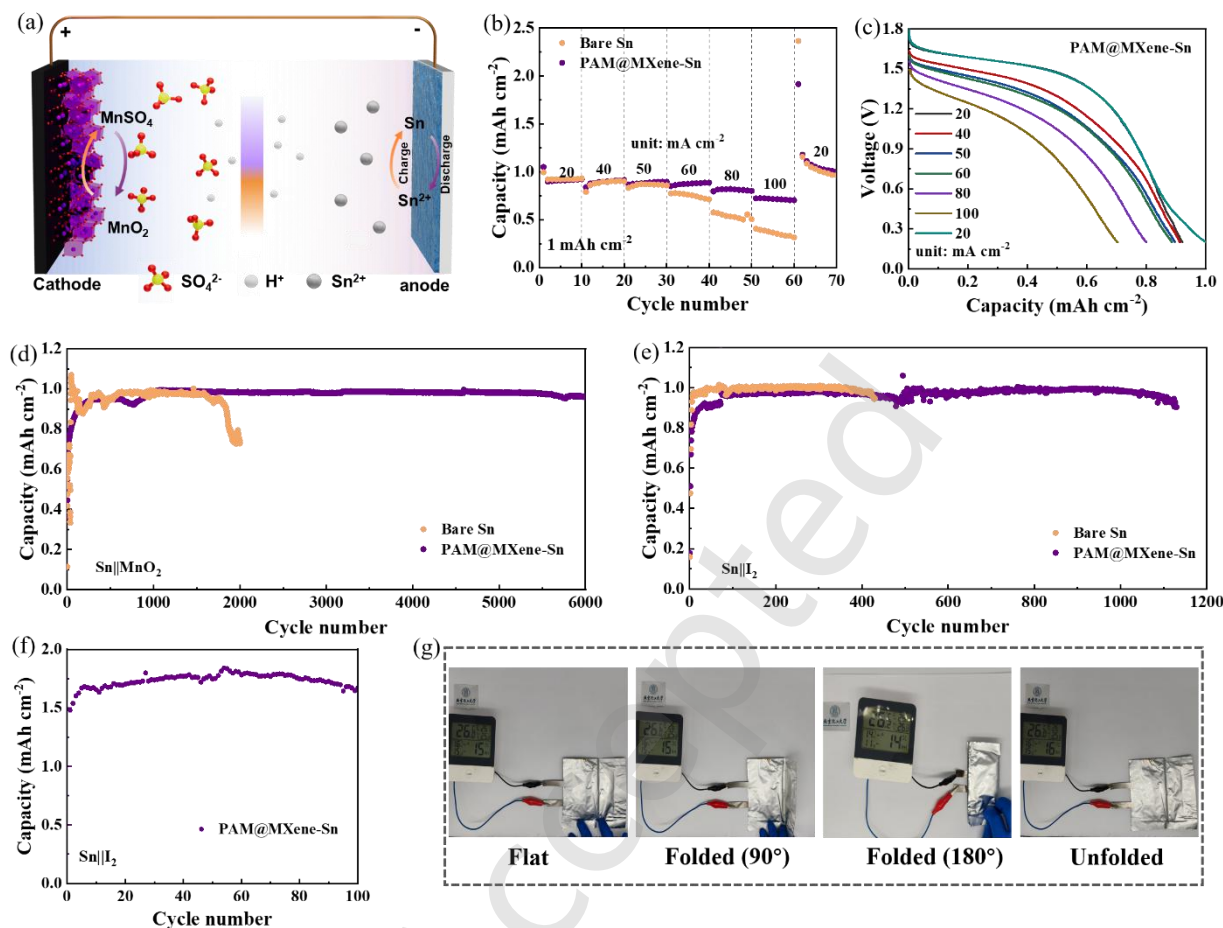


Figure 5 Electrochemical performance of (a-d) Sn||MnO₂ and (e-g) Sn||I₂ full cells based on bare Sn and PAM@MXene-Sn anodes. (a) Schematic illustration of Sn||MnO₂ cell. (b) Rate performance of bare Sn||MnO₂ and PAM@MXene-Sn||MnO₂ cells. Discharge profiles of (c) PAM@MXene-Sn||MnO₂ cell at different current densities. (d) Cycling performance of Sn||MnO₂ cells at 20 mA cm⁻². (e) Cycling performance of Sn||I₂ cells at 10 mA cm⁻². (f) Cycling performance of Sn||I₂ pouch cell and (g) photographs of the cell operating at different bending angles.

reversibility during prolonged cycling.

To validate the practical feasibility of the PAM@MXene-Sn anode, full cells paired with MnO₂ cathode were assembled and electrochemically evaluated. The PAM@MXene-Sn||MnO₂ cell configuration is illustrated in Figure 5a, in which the MnO₂ cathode undergoes a dissolution/deposition mechanism involving a two-electron transfer process in acidic electrolyte [40]. CV curves show that both bare Sn||MnO₂ and PAM@MXene-Sn||MnO₂ full cells exhibit well-defined oxidation and reduction peaks (Figure S32). The redox peak gap remarkably decreases from 357.3 mV for bare Sn||MnO₂ to 278.7 mV for PAM@MXene-Sn||MnO₂, indicating reduced electrochemical polarization. At a fixed charging capacity of 1 mAh cm⁻², the PAM@MXene-Sn||MnO₂ cell consistently maintains high areal capacities at various current densities and sustains ~ 0.70 mAh cm⁻² even at 100 mA cm⁻² (Figure 5b). When the current density is reduced to 20 mA cm⁻², the capacity recovers to ~1.00 mAh cm⁻², indicating superior rate performance. Moreover, the discharge voltage plateau of the

bare Sn||MnO₂ cell shortens or even disappears at high current densities (Figure S33), whereas the PAM@MXene-Sn||MnO₂ cell retains a distinct plateau even at 100 mA cm⁻² (Figure 5c), demonstrating its ability to sustain efficient electrochemical reactions under high-rate conditions.

Figure 5d illustrates the cycling performance of the bare Sn||MnO₂ and PAM@MXene-Sn||MnO₂ cells at 20 mA cm⁻². Both the cells exhibit low initial capacity due to the activation process for the progressive electrolyte infiltration and the dissolution/deposition of the MnO₂ cathode [44]. In the subsequent cycling, the bare Sn||MnO₂ cell shows considerable capacity fluctuations and fails at 2,000 cycles, implying the performance degradation caused by the “dead Sn” accumulation. However, the PAM@MXene-Sn||MnO₂ cell delivers a stable capacity close to 1 mAh cm⁻² for 6,000 cycles, with flat discharge plateau near 1.5 V maintaining from the 1,000th to the 6,000th cycle (Figure S34). Besides, at a low current density of 6 mA cm⁻², the cell retains a discharge capacity of ~0.97 mAh cm⁻² after 1,750 cycles (Figure S35), substantiating its improved cycling stability.

Furthermore, the cycled bare Sn anode exhibits substantial “dead Sn” accumulation and rough surface, whereas the PAM@MXene-Sn anode remains dense and smooth morphology, providing direct evidence of the PAM@MXene interlayer’s protection effect (Figure S36). Consistently, the EIS spectra, fitted with the equivalent circuit, reveal that the charge-transfer resistance increases severely for the cycled bare Sn anode but remains low for the PAM@MXene-Sn anode (Figure S37), which is ascribed to the suppression of HER and the prevention of irregular deposition by the PAM@MXene hydrogel interlayer.

The practical viability of the PAM@MXene-Sn anode is further extended to Sn||I₂ full cell [45–46]. At a current density of 10 mA cm⁻², the PAM@MXene-Sn||I₂ cell maintains a stable discharge capacity of ~0.95 mAh cm⁻² after 1,100 cycles, demonstrating high reversibility and good cycling stability (Figure 5e and Figure S38). To better align with practical application scenarios, the system was configured into a pouch cell. Under 10 mA cm⁻², the PAM@MXene-Sn||I₂ pouch cell operates stably, delivering a capacity of ~1.67 mAh cm⁻² with negligible decay after 100 cycles (Figure 5f). Additionally, the pouch cell successfully powers an electronic thermometer and maintains normal operation under multi-angle bending (Figure 5g), highlighting the excellent mechanical adaptability and structural integrity enabled by the composite hydrogel interlayer.

3 Conclusions

The organic-inorganic composite hydrogel interlayer, comprising PAM matrix and uniformly dispersed MXene nanosheets, is designed and constructed on Sn metal anode via in-situ UV-induced polymerization. Within this architecture, the hydrophilic PAM network functions as a physical barrier, guides Sn²⁺ migration, confines interfacial water molecules, and accommodates volume changes. The incorporated MXene nanosheets provide homogeneous nucleation sites, ensure uniform interfacial electric field, and facilitate fast ion transport. Additionally, the hydrogen-bonding interactions between PAM and MXene reinforce

the structural integrity and improve its adhesion with the electrode. This integrated hydrogel thereby acts as a multifunctional interfacial regulator for Sn electrodes, which enables uniform and dense Sn deposition, suppresses “dead Sn” accumulation, and mitigates side reactions. Consequently, the PAM@MXene-Sn electrode exhibits excellent electrochemical performance, delivering a cycling life of 2,600 h at 1 mA cm⁻² and 1 mAh cm⁻² in the symmetric cell, and sustaining an average CE of 99.99% for 20,000 cycles in the asymmetric cell. Furthermore, the practical viability of this electrode is validated in the PAM@MXene-Sn||MnO₂ and PAM@MXene-Sn||I₂ full cells. This work not only presents the PAM@MXene composite hydrogel as an efficient interfacial layer for high-performance Sn metal anodes, but also provides a design strategy of constructing organic-inorganic synergistic interfaces, which is promising for stabilizing Sn metal anodes in aqueous batteries.

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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 SEM images, XRD patterns, XPS spectra, Raman spectra, ²H NMR, CV curves, EIS plots, AFM images, in-situ optical microscope images, adsorption models, contact angles, DEMS, charge-discharge profiles) is available in the online version of this article at <https://doi.org/10.26599/NRE.2026.9120262>.

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Electronic Supplementary Material

Synergistic PAM@MXene hydrogel Interlayer regulates Sn deposition behavior for highly stable Sn anodes in aqueous tin-ion batteries

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1 Experimental Section

1.1 Materials

Sn foil was supplied by HaoXuan Metal Materials Co., Ltd. Acrylamide (AM, 99.9%), 2-hydroxy-1-[4-(2-hydroxyethoxyphenyl)]-2-methyl-1-propanone (HMEA, 98%), N, N'-methylenebisacrylamide (MBA, 97%), tin (II) methanesulfonate (Sn (MSA)_2 , 50 wt% in H_2O), and methanesulfonic acid (MSA, 50 wt% in H_2O) were obtained from Aladdin. $\text{MnSO}_4 \cdot \text{H}_2\text{O}$ (99%) and LiF (99%) were sourced from Macklin. Concentrated H_2SO_4 (95.0-98.0%) and HCl (12 M) were acquired from Sinopharm Chemical Reagent Co., Ltd. Ti_3AlC_2 MAX phase was provided by Jilin 11 Technology Co., Ltd. $\delta\text{-MnO}_2$ was obtained from Future (Jilin) Materials Technology Co., Ltd. Ketjen black and PVDF were sourced from Canrd, and Nafion 117 proton exchange membrane was purchased from SCI Materials Hub.

1.2 Preparation of Pre-Hydrogel Solution

First, the MXene dispersion was prepared by stirring 1 g of Ti_3AlC_2 MAX phase powder and 0.99 g of LiF in 10 mL of 12 M HCl at 35 °C for 24 h. The product was then washed via centrifugation until the pH of the supernatant approached neutral. The obtained sediment was thoroughly washed with deionized water via centrifugation and then redispersed to obtain the MXene dispersion with a concentration of about 6 mg mL^{-1} . The pre-hydrogel solution was prepared by dissolving AM monomer (2 g), MBA crosslinker (1 mg), and HMEA photoinitiator (7.5 mg) in 10 mL of deionized water under stirring until a homogeneous solution was obtained. Subsequently, 5 mg of the MXene dispersion was added under continuous stirring to ensure uniform dispersion, yielding a homogeneous and stable pre-hydrogel solution. For comparison, the pre-hydrogel solution without the addition of MXene was also prepared.

1.3 Construction of the PAM@MXene Hydrogel Interlayer

The Sn foil was sequentially cleaned by sonication in ethanol and deionized water, and then dried under vacuum. The prepared pre-hydrogel solution was uniformly drop-cast onto the Sn foil surface, and a glass slide was pressed on top to ensure thin and uniform coverage on the Sn substrate. The assembly was then irradiated under 365 nm UV light for 1 h to induce in situ polymerization, resulting in a tightly adhered composite hydrogel interlayer on the Sn substrate (denoted as PAM@MXene-Sn). For comparison, the pure PAM hydrogel-modified Sn electrode (denoted as PAM-Sn) was prepared using the pre-hydrogel solution without MXene with the same procedure.

1.4 Preparation of MnO_2 and I_2 Cathodes

The MnO_2 cathode was fabricated by thoroughly mixing $\delta\text{-MnO}_2$ powder, Ketjen black, and PVDF in a mass ratio of 7:2:1 in N-methyl-2-pyrrolidone (NMP) to form a homogeneous slurry, which was then uniformly coated onto carbon paper and vacuum-dried at 60 °C for 10 h. The I_2 cathode was prepared by uniformly mixing activated carbon, Ketjen black, and polytetrafluoroethylene (PTFE) in a mass ratio of 7:2:1 in ethanol. The obtained homogeneous paste was roll-pressed onto titanium mesh, and then the electrode was immersed in a ZnI_2 solution (1 g mL^{-1}) for 12 h. After immersion, the electrode was rinsed with deionized water and dried at 60 °C for 10 h. The resulting $\delta\text{-MnO}_2$ and I_2 cathodes were cut into discs with a diameter of 8 mm. The areal mass loading of $\delta\text{-MnO}_2$ or I_2 in the cathodes was maintained at 0.8-1.2 mg cm^{-2} .

1.5 Fabrication of Sn||Sn Symmetric Cells, Sn||Cu Asymmetric Cells, and Sn|| MnO_2 and Sn|| I_2 Full Cells

The aqueous electrolyte was prepared by dissolving 1 M Sn (MSA) $_2$ in 3 M MSA. For Sn||Sn symmetric cells, two pieces of Sn foil electrodes were assembled in CR2032 coin cells with 120 μL of the electrolyte, using a glass fiber separator (Whatman). The Sn||Cu asymmetric cells were fabricated with a Sn foil and a Cu foil as the electrodes with unchanged electrolytes and separators. All the cells were assembled in ambient air. The Sn|| MnO_2 full cell was assembled with a Sn foil anode and a MnO_2 cathode, using glass fiber separator. The Nafion 117 membrane was placed between the anode and the cathode to mitigate ion crossover. The anode side contained 120 μL of 3 M MSA + 1 M Sn (MSA) $_2$ electrolyte, and the cathode side contained 120 μL of 0.3 M H_2SO_4 + 3 M MnSO_4 electrolyte. The Sn|| I_2 full cell was assembled

following the same procedure, except that the cathode-side electrolyte was replaced with 0.3 M H₂SO₄.

1.6 Fabrication of the PAM@MXene-Sn||I₂ Pouch Cell

The PAM@MXene-Sn||I₂ pouch cell was assembled using a PAM@MXene-Sn anode (5 cm × 5 cm) and an I₂ cathode (4 cm × 4 cm). Glass-fiber separators and a Nafion 117 membrane were placed between the anode and cathode. The anode and cathode sides were filled with 1 mL of 3 M MSA+1 M Sn(MSA)₂ electrolyte and 1 mL of 0.3 M H₂SO₄ electrolyte, respectively. The components were stacked in the sequence of PAM@MXene-Sn anode/glass-fiber separator/Nafion 117 membrane/glass-fiber separator/I₂ cathode, packaged in an aluminum-laminated pouch, and vacuum heat-sealed. The assembled pouch cell was rested for 8 h before electrochemical testing.

1.7 Material Characterizations

The X-ray diffraction (XRD) patterns were recorded using a Bruker D8 Advance diffractometer. The morphology and elemental distribution were examined via scanning electron microscopy (SEM, Hitachi S-4700) at an acceleration voltage of 5.0 kV by energy dispersive X-ray spectroscopy (EDS) at 20.0 kV. The chemical bonding and functional groups were analyzed by Raman spectrometer (inVia Reflex, Renishaw) with 633 nm laser source and Fourier transform infrared spectroscopy (FTIR, Nicolet 6700) in the range of 4000-800 cm⁻¹ with a resolution of 2 cm⁻¹. The electrode roughness was assessed using atomic force microscopy (AFM, Bruker Dimension Icon) with a sharp probe featuring a tip radius of 8 nm and a spring constant of 26 N m⁻¹. Before SEM and AFM characterization of the deposited or cycled PAM@MXene-Sn electrodes, the PAM@MXene hydrogel interlayer was removed to expose the underlying Sn deposits. The chemical composition of the samples was determined by X-ray photoelectron spectroscopy (XPS, ESCALAB 250, THERMO VG) in a high vacuum chamber (9.8 × 10⁻¹⁰ Torr) with Al K α radiation ($h\nu$ = 1486.6 eV).

1.8 Electrochemical Measurements

The electrochemical performance of the Sn electrodes with and without hydrogel interlayers was systematically evaluated. The galvanostatic charge-discharge tests of Sn||Sn symmetric cells and Sn||Cu asymmetric cells (with a cut-off voltage of 0.4 V) were performed using LAND battery testing system. Electrochemical impedance spectroscopy (EIS) was carried out on symmetric cells in a frequency range of 100 kHz to 10 mHz. Chronoamperometry (CA) was conducted in symmetric cells with a voltage hysteresis of 10 mV for 600 s. Cyclic voltammetry (CV) of Sn||Cu asymmetric cells was performed in a potential range of -0.6 V to 0 V at a scan rate of 5 mV s⁻¹. The exchange current density (i_0) was calculated from the polarization curves of symmetric cells at various current densities using the following relationship:

$$i = i_0 \frac{F \eta}{RT} 2$$

where T represents the absolute temperature, R denotes the gas constant, F signifies the Faraday constant, η indicates the total overpotential, and i refers to the applied current density.

A three-electrode system was employed for further interfacial analysis, with the Sn-based electrodes as the working electrode, the Pt plate as the counter electrode, and an Ag/AgCl electrode as the reference electrode. Linear sweep voltammetry (LSV) and linear polarization measurements were performed in the three-electrode system with 3 M MSA electrolyte. The LSV measurements were conducted in -1 to 0 V (vs. Ag/AgCl) at 5 mV s⁻¹, while linear polarization curves were obtained in -0.5 to -0.4 V (vs. Ag/AgCl) at the same scan rate. In situ hydrogen evolution during cycling at 1 mA cm⁻² and 1 mAh cm⁻² was monitored using differential electrochemical mass spectrometry (DEMS, LingLu).

The CV measurements of the bare Sn||MnO₂ and PAM@MXene-Sn||MnO₂ full cells were conducted over a voltage range of 0-2 V at a scan rate of 0.5 mV s⁻¹. The cycling performance and rate capability of Sn||MnO₂ and Sn||I₂ full cells were tested on Neware battery testing system at a fixed charging capacity of 1 mAh cm⁻², within voltage windows of 0.2-2.0 V and 0.5-1.9 V, respectively. EIS measurement of Sn||MnO₂ cell was carried out in the same manner as for the symmetric cells.

1.9 Density Functional Theory (DFT) Calculations

The DFT calculations on the binding energies were carried out using the DMol3 module in Materials Studio 2020. For the exchange and correlation functional, the generalized gradient approximation (GGA) in the Perdew-Burke-Ernzerhof (PBE) format was used. The self-consistent field (SCF) density convergence criterion was set to 1.0×10^{-4} , and a thermal smearing of 0.005 Ha was applied to facilitate SCF convergence. The convergence thresholds for energy, gradient, and displacement during geometry optimization were set to 1.0×10^{-4} Ha, 0.02 Ha $\text{\AA}^{-1}$, and 0.05 $\text{\AA}$, respectively. The binding energies between Sn^{2+} , H_2O , PAM fragment, and MXene surface were calculated using the following equation:

$$E_{\text{binding}} = E_{(AB)} - E_{(A)} - E_{(B)}$$

where $E_{(A)}$ and $E_{(B)}$ represents the energy of the individual species (e.g., Sn^{2+} , H_2O , PAM fragment, and MXene surface), and $E_{(AB)}$ denotes the total energy of the combined system.

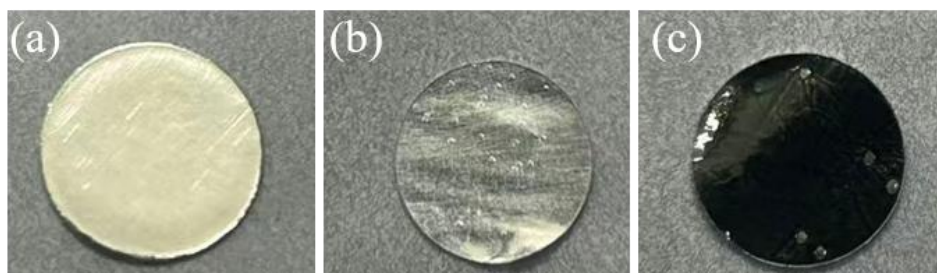


Figure S1 Digital photographs of (a) bare Sn, (b) PAM-Sn and (c) PAM@MXene-Sn electrodes.

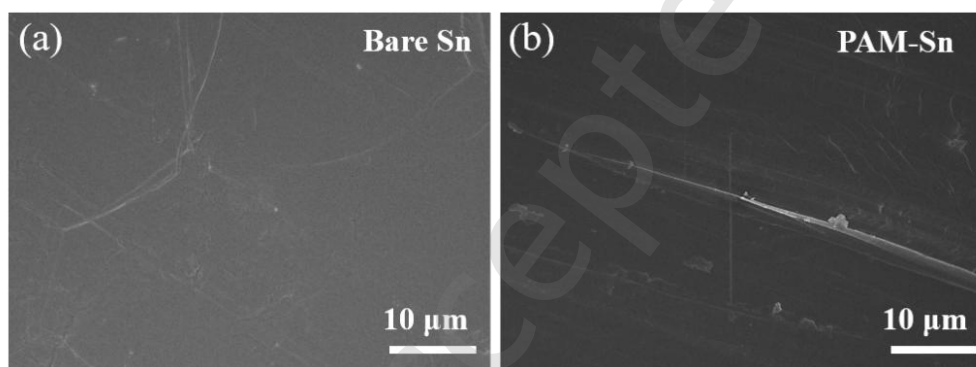


Figure S2 SEM images of (a) bare Sn and (b) PAM-Sn electrodes.

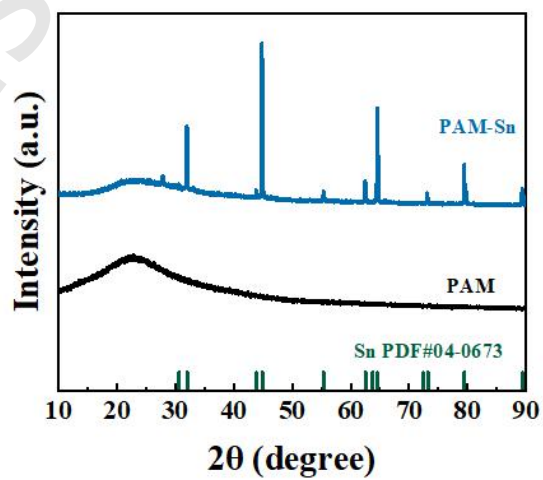


Figure S3 XRD patterns of PAM-Sn electrode and PAM hydrogel.

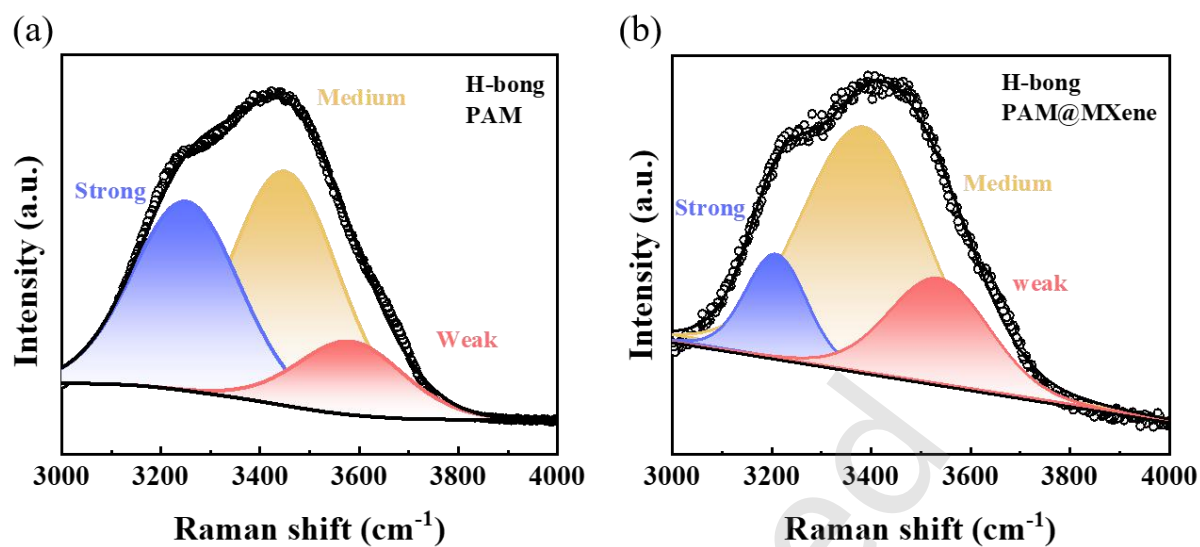


Figure S4 Raman spectra of (a) PAM and (b) PAM@MXene hydrogels in the O-H stretching region.

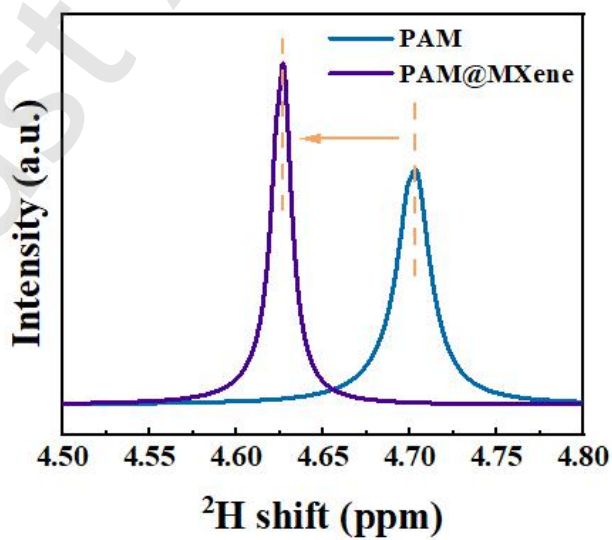


Figure S5 ^2H NMR spectra of PAM and PAM@MXene hydrogels.

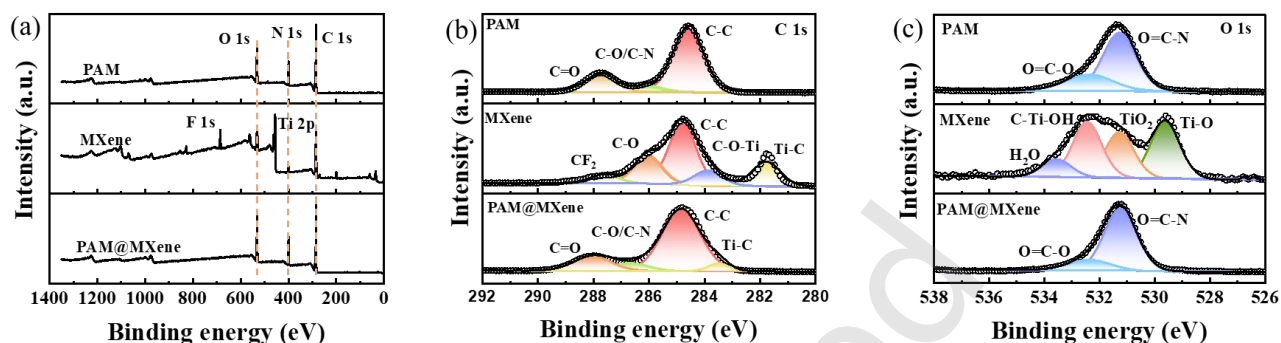


Figure S6 (a) XPS survey spectra with (b) high-resolution C 1s and (c) O 1s spectra of PAM, MXene, and PAM@MXene hydrogel.

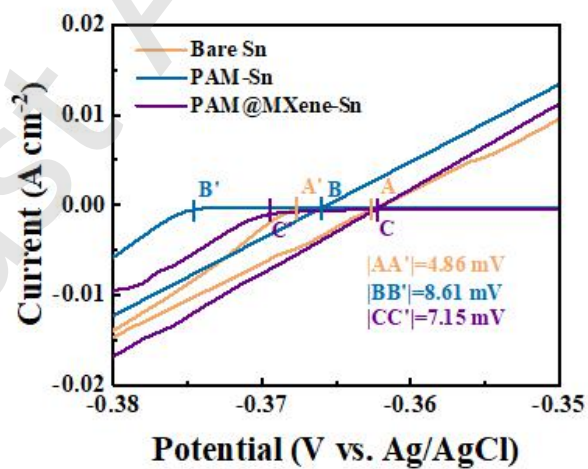


Figure S7 Enlarged view of CV curves (in Figure 2a) of bare Sn, PAM-Sn and PAM@MXene-Sn electrodes.

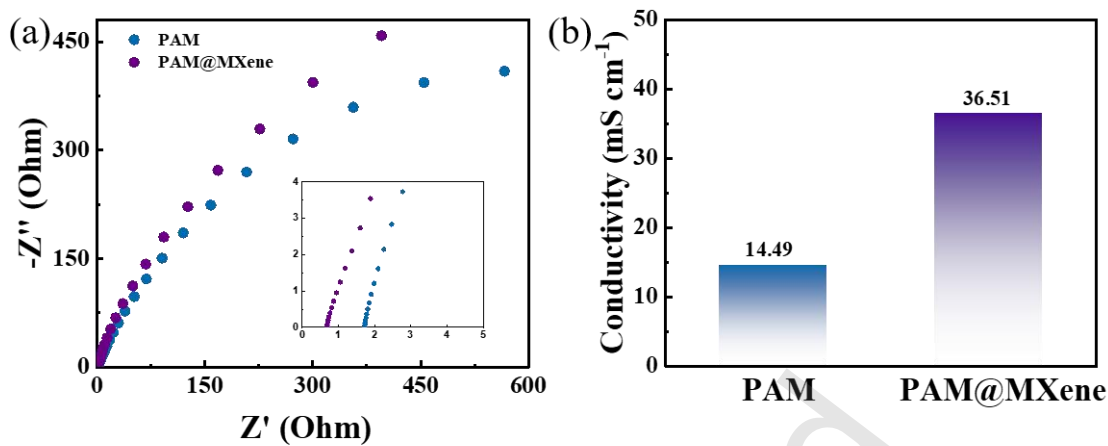


Figure S8 (a) EIS spectra and (b) ionic conductivities of PAM and PAM@MXene hydrogels.

The ionic conductivity (σ) of PAM and PAM@MXene hydrogels that was tested in the stainless-steel (SS) symmetric cells, are calculated as follows:

$$\sigma = \frac{L}{SR}$$

where L is the distance between two parallel SS plates ($\sim 500 \mu\text{m}$), S is the area of the stainless-steel plate ($\sim 2.01 \text{ cm}^2$) and R is the ohmic impedance.

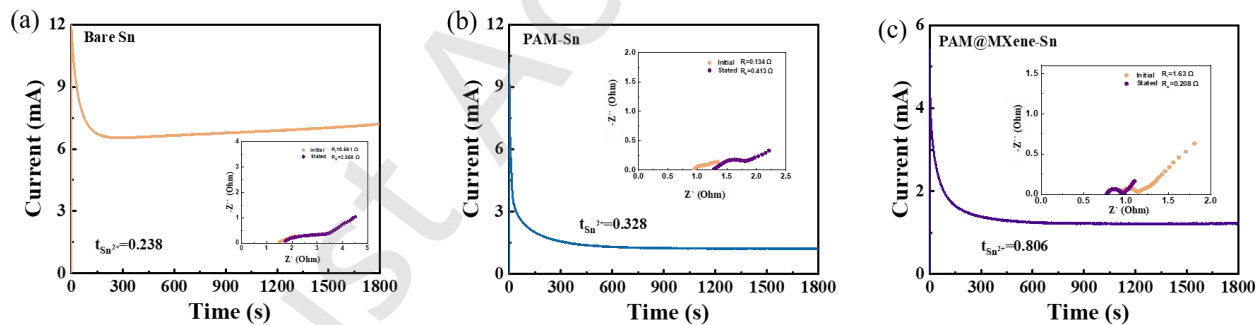


Figure S9 I - t curves of symmetric cells with (a) bare Sn, (b) PAM-Sn and (c) PAM@MXene-Sn electrodes with the Nyquist plots before and after polarization in the inset.

To quantify the interfacial ion-transport behavior, the transfer number of Sn^{2+} ions ($t_{\text{Sn}^{2+}}$) were calculated by analyzing the relationship between the charge transfer resistance and the response current changes before and after the constant potential polarization. The value was calculated using the following equation:

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

where I_0 represents the initial current, I_s stands for the steady-state current, R_0 and R_s is the charge transfer resistance before and after polarization, and ΔV is the applied constant potential.

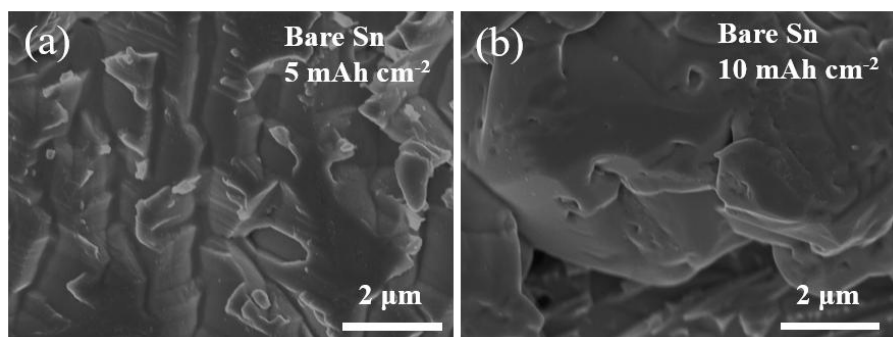


Figure S10 SEM images of bare Sn electrode with deposition capacities of (a) 5 mAh cm⁻² and (b) 10 mAh cm⁻².

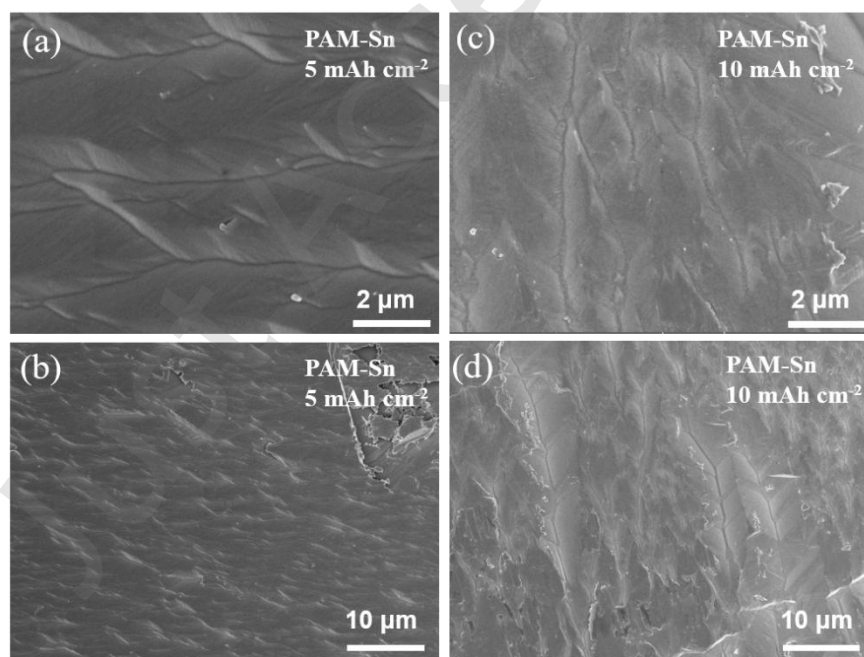


Figure S11 SEM images of PAM-Sn electrode with deposition capacities of (a, b) 5 mAh cm⁻² and (c, d) 10 mAh cm⁻².

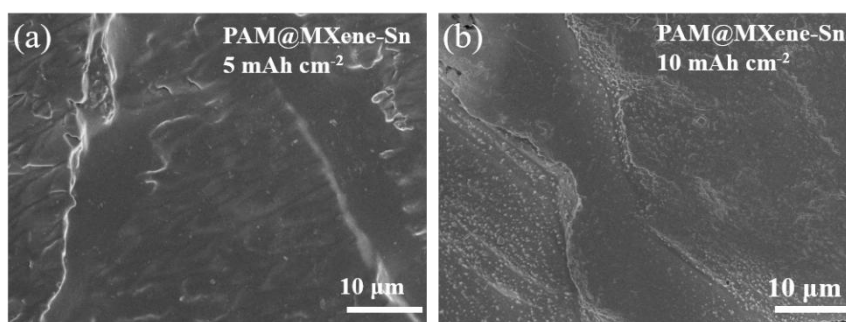


Figure S12 SEM images of PAM@MXene-Sn electrode with deposition capacities of (a) 5 mAh cm⁻² and (b) 10 mAh cm⁻².

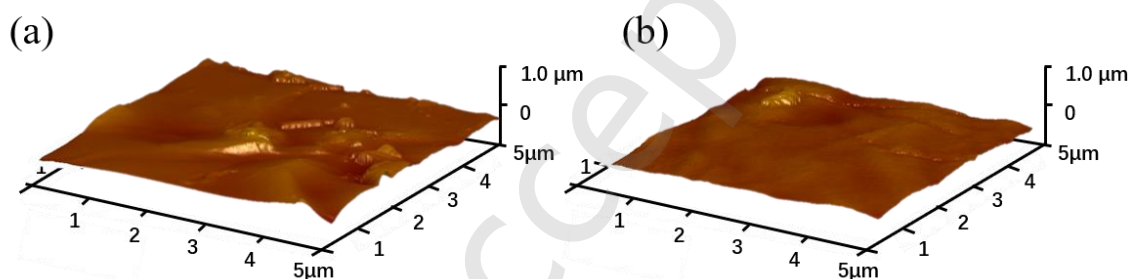


Figure S13 3D AFM images of PAM@MXene-Sn with deposition capacities of (a) 5 mAh cm⁻² and (b) 10 mAh cm⁻².

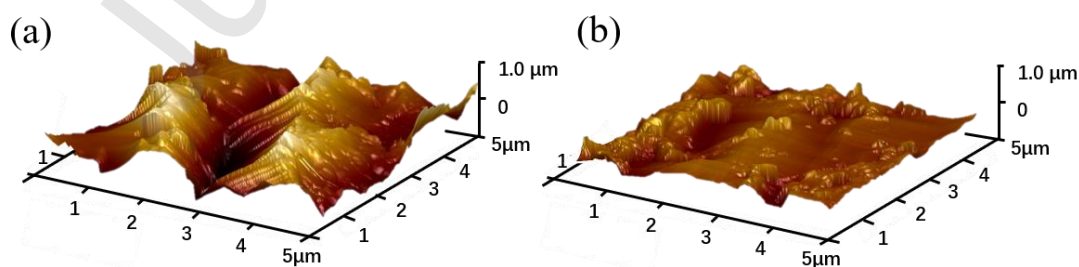


Figure S14 3D AFM images of bare Sn electrode with deposition capacities of (a) 5 mAh cm⁻² and (b) 10 mAh cm⁻².

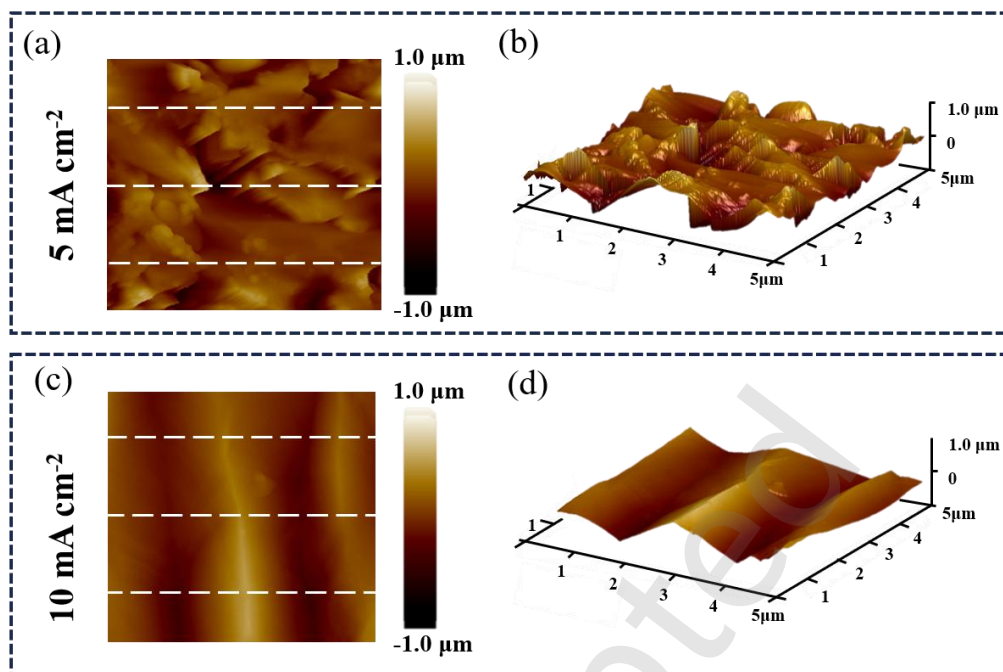


Figure S15 AFM images of PAM-Sn electrode with deposition capacities of (a, b) 5 mAh cm^{-2} and (c, d) 10 mAh cm^{-2} .

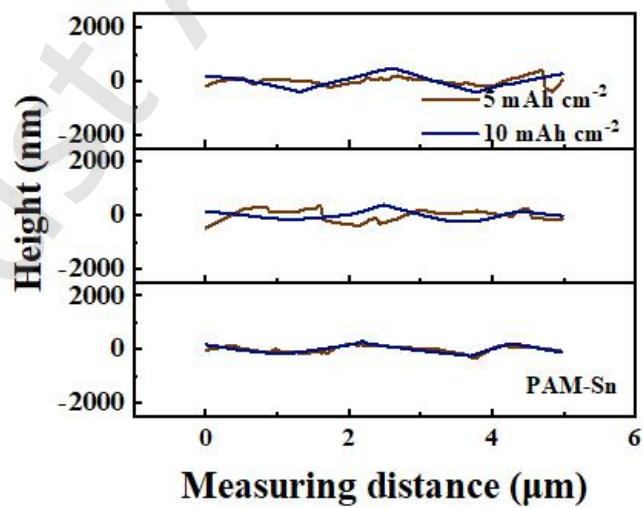


Figure S16 Height distribution of PAM-Sn electrode with deposition capacities of 5 mAh cm^{-2} and 10 mAh cm^{-2} .

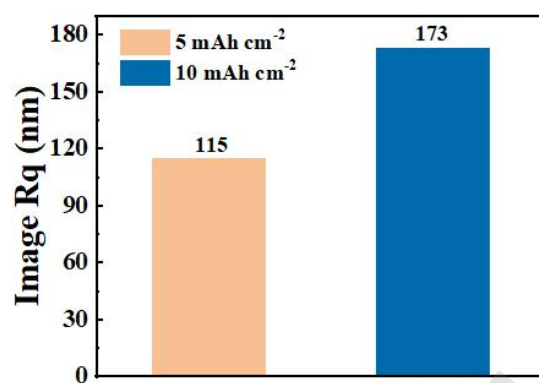


Figure S17 Surface roughness of PAM-Sn electrode with deposition capacities of 5 mAh cm^{-2} and 10 mAh cm^{-2} .

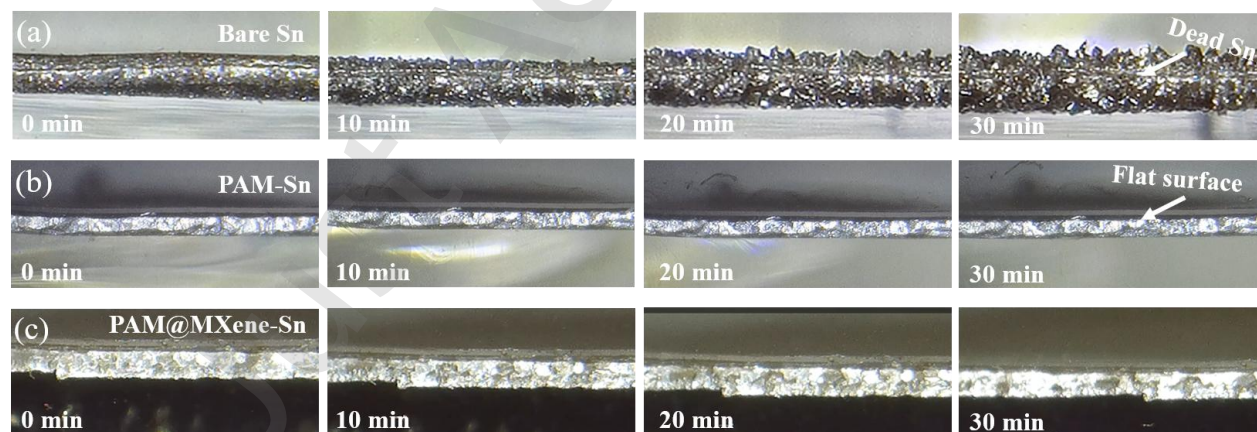


Figure S18 In-situ optical microscope images of (a) bare Sn, (b) PAM-Sn and (c) PAM@MXene-Sn electrodes during Sn deposition at 10 mA cm^{-2} .

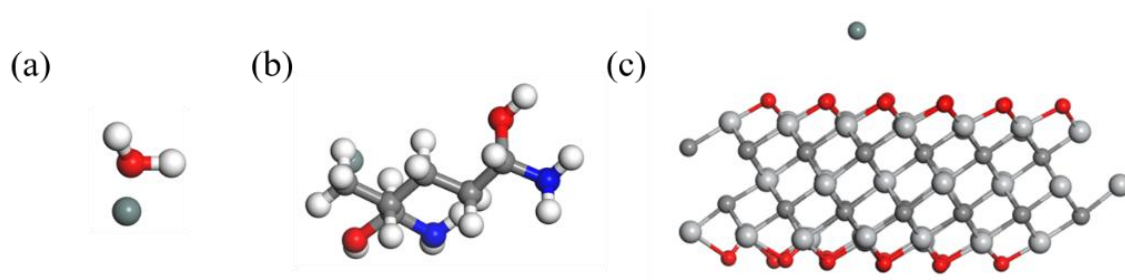


Figure S19 Adsorption models of Sn^{2+} with (a) H_2O , (b) PAM fragment, and (c) MXene surface.

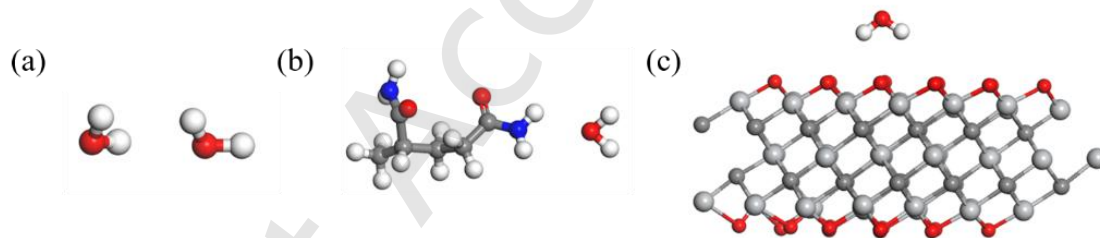


Figure S20 Adsorption models of H_2O with (a) H_2O , (b) PAM fragment, and (c) MXene surface.



Figure S21 Contact angles of (a) bare Sn, (b) PAM-Sn, and (c) PAM@MXene-Sn electrodes with 3M MSA + 1 M Sn (MSA)₂.

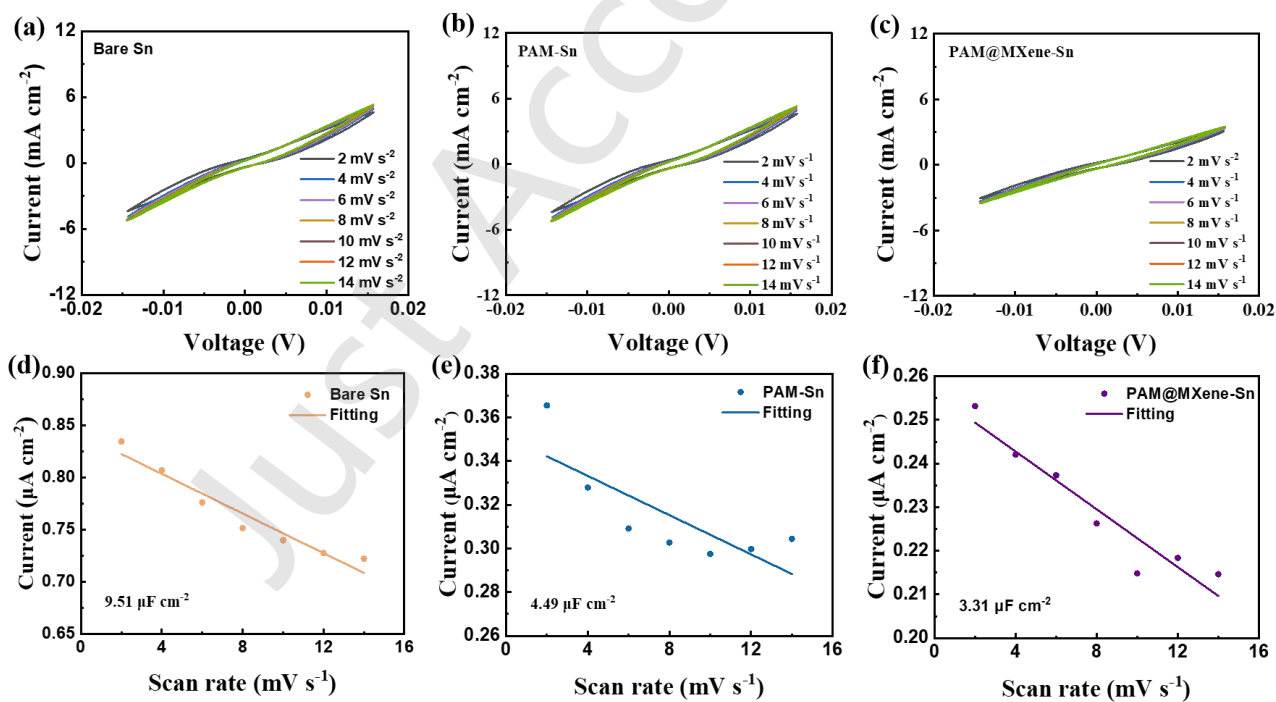


Figure S22 (a-c) CV curves at different scan rates and (d-f) the corresponding electric double-layer capacitances of (a, d) bare Sn, (b, e) PAM-Sn, and (c, f) PAM@MXene-Sn electrodes.

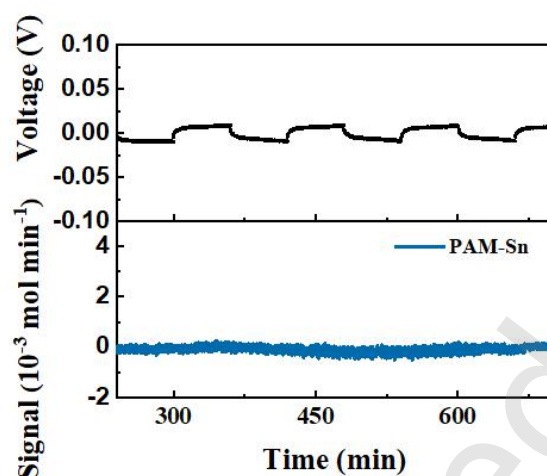


Figure S23 In situ DEMS signals with corresponding galvanostatic cycling profiles for PAM-Sn||PAM-Sn symmetric cell.

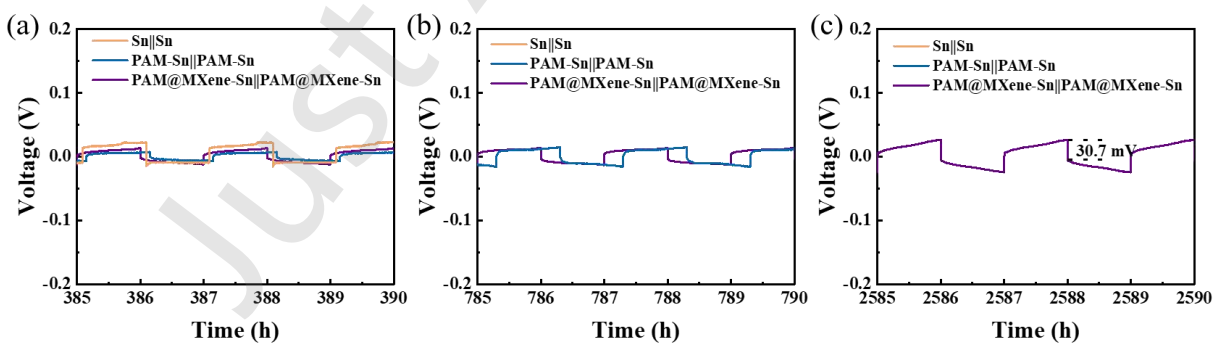


Figure S24 Charge-discharge profiles of symmetrical cell at 1 mA cm^{-2} , 1 mAh cm^{-2} (a) from 385 h to 390 h, (b) from 758 h to 790 h and (c) from 2585 h to 2590 h.

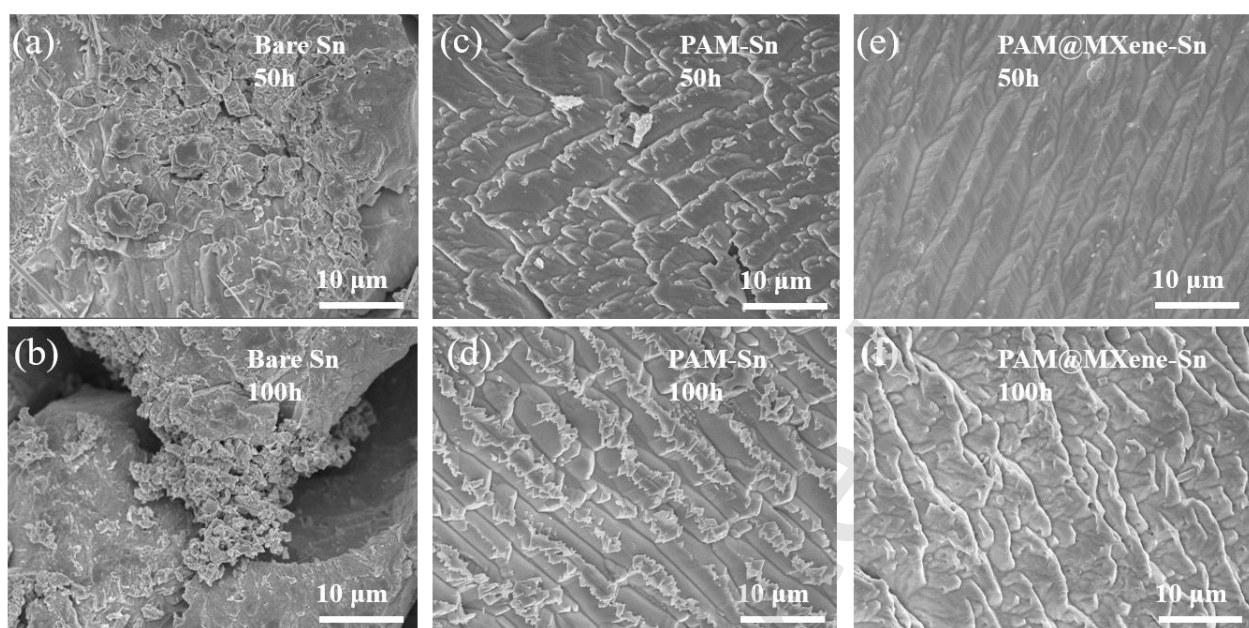


Figure S25 SEM images of (a, b) bare Sn, (c, d) PAM-Sn and (e, f) PAM@MXene-Sn electrodes after cycling 50 and 100 h.

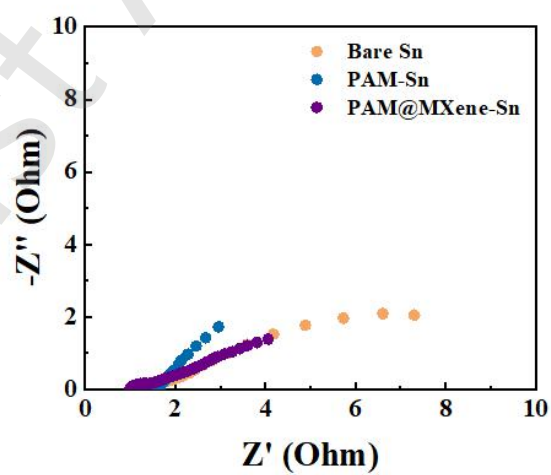


Figure S26 EIS spectra of bare Sn, PAM-Sn and PAM@MXene-Sn.

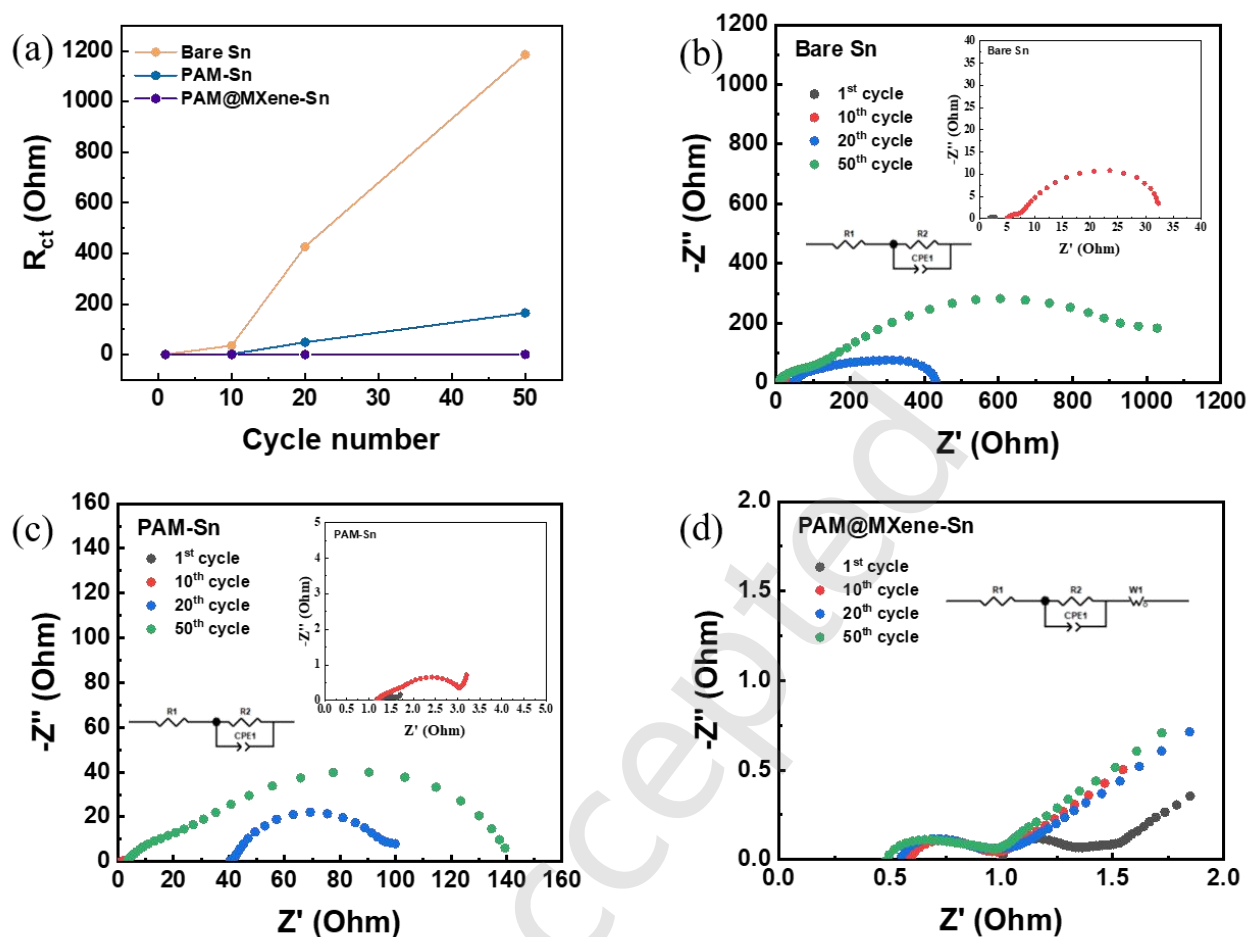


Figure S27 (a) Evolution of interfacial resistance and EIS spectra of symmetric cells with (b) bare Sn, (c) PAM-Sn, and (d) PAM@MXene-Sn electrodes after 1, 10, 20, and 50 cycles.

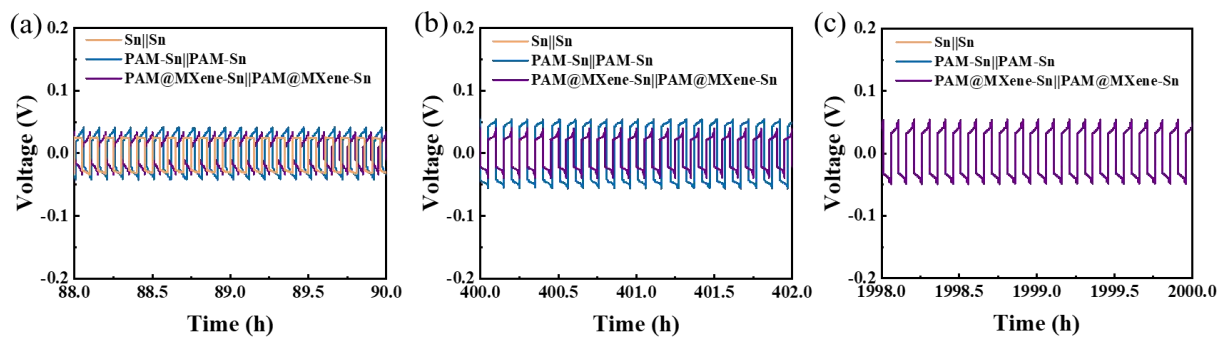


Figure S28 Charge-discharge profiles of symmetrical cell at 10 mA cm^{-2} , 0.5 mAh cm^{-2} (a) from 88 h to 90 h, (b) from 400 h to 402 h and (c) from 1998 h to 2000 h.

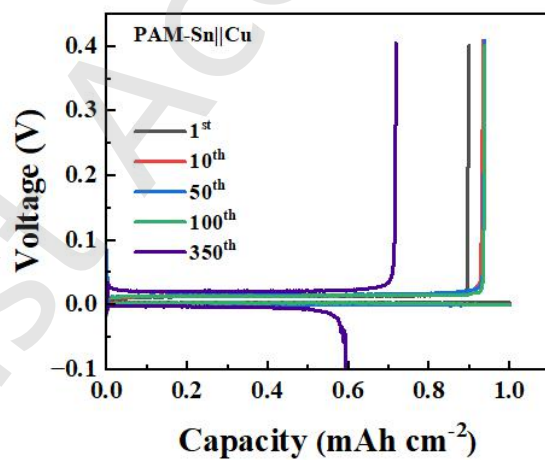


Figure S29 Charge-discharge profiles of PAM-Sn||Cu asymmetric cells at 1 mA cm^{-2} , 1 mAh cm^{-2} .

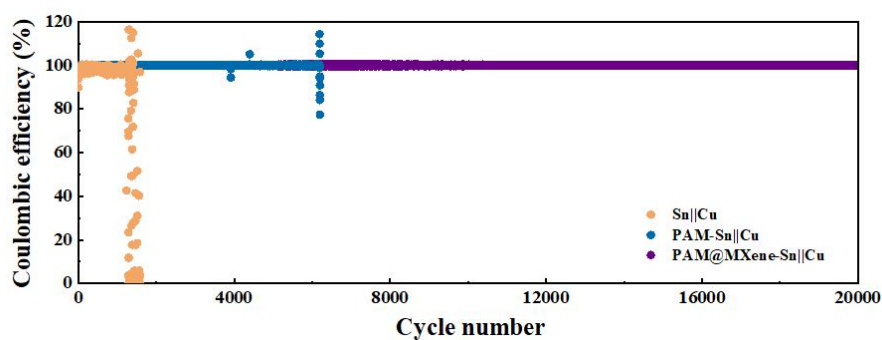


Figure S30 CEs of different Sn||Cu asymmetric cells at 10 mA cm^{-2} , 0.5 mAh cm^{-2} .

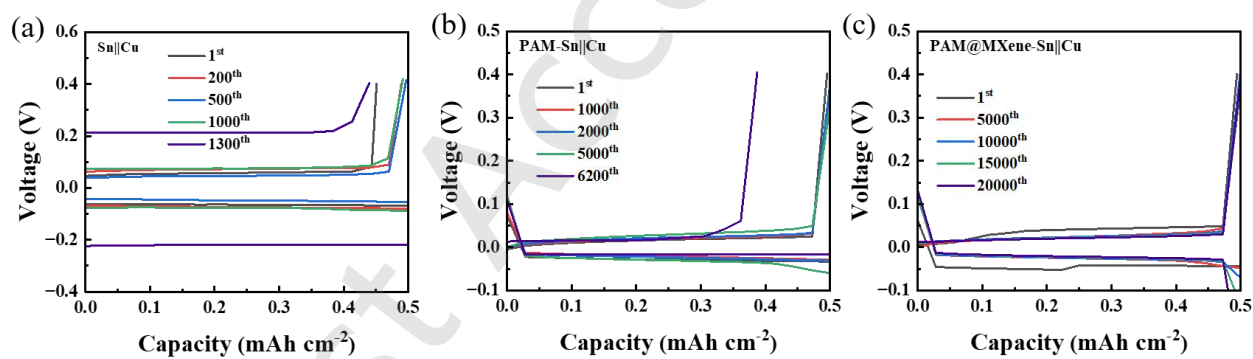


Figure S31 Charge-discharge profiles of (a) bare Sn||Cu, (b) PAM-Sn||Cu and (c) PAM@MXene-Sn||Cu asymmetric cells at 10 mA cm^{-2} , 0.5 mAh cm^{-2} .

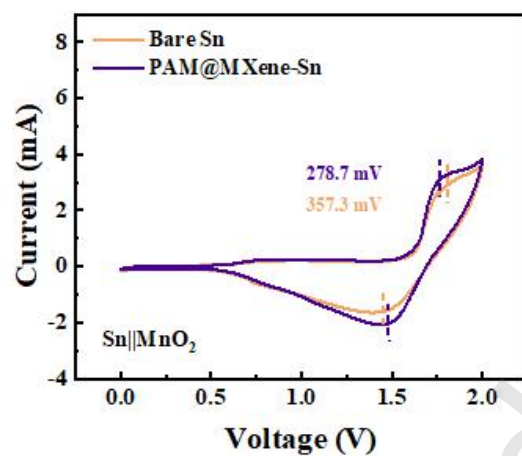


Figure S32 CV curves of bare Sn||MnO₂ and PAM@MXene-Sn||MnO₂ full cells at a scan rate of 0.5 mV s⁻¹.

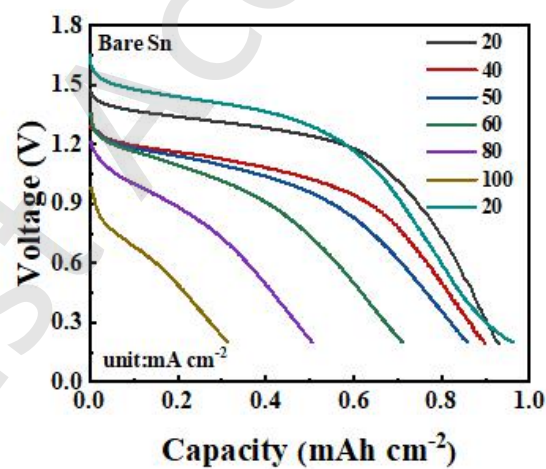


Figure S33 Discharge profiles of bare Sn||MnO₂ cell at different current densities.

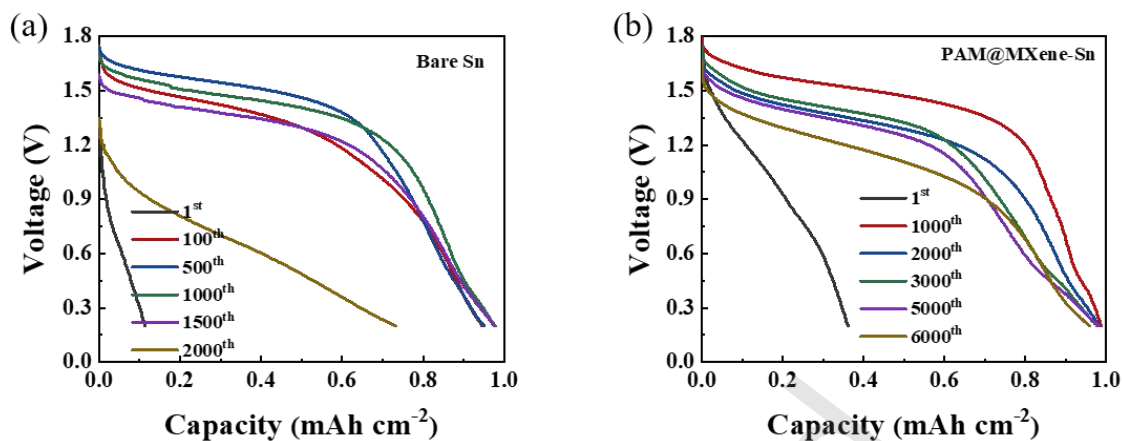


Figure S34 Discharge profiles of (a) bare Sn||MnO₂ and (b) PAM@MXene-Sn||MnO₂ cells at 20 mA cm⁻².

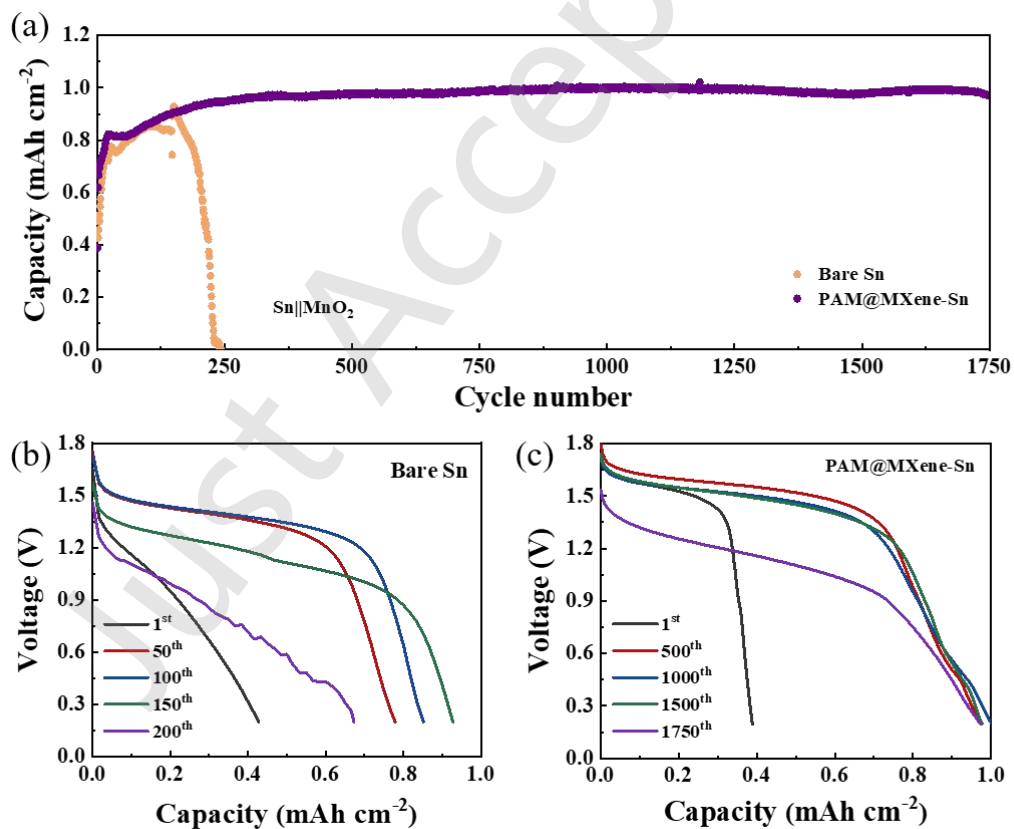


Figure S35 (a) Cycling performance of Sn||MnO₂ and PAM@MXene-Sn||MnO₂ cells at 6 mA cm⁻². Discharge profiles of (b) bare Sn||MnO₂ and (c) PAM@MXene-Sn||MnO₂ cells at 6 mA cm⁻².

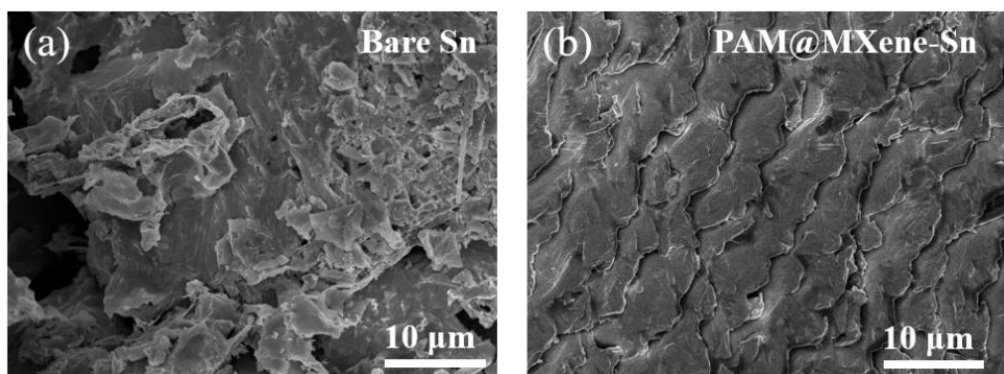


Figure S36 SEM images of (a) bare Sn and (b) PAM@MXene-Sn anodes after 200 cycles at 6 mA cm⁻².

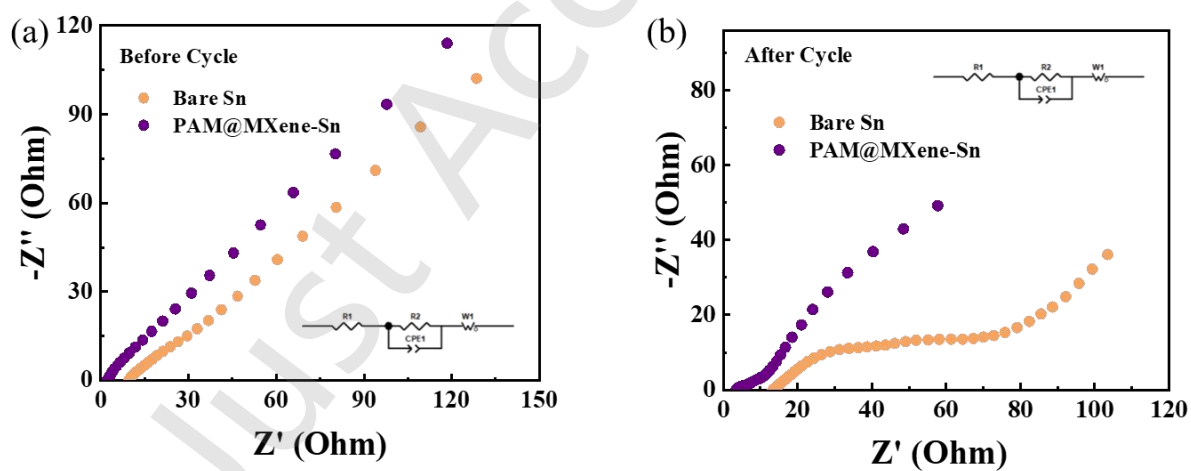


Figure S37 EIS spectra of bare Sn || MnO₂ and PAM@MXene-Sn || MnO₂ cells (a) before and (b) after 200 cycles at 6 mA cm⁻², with the equivalent circuit in the inset.

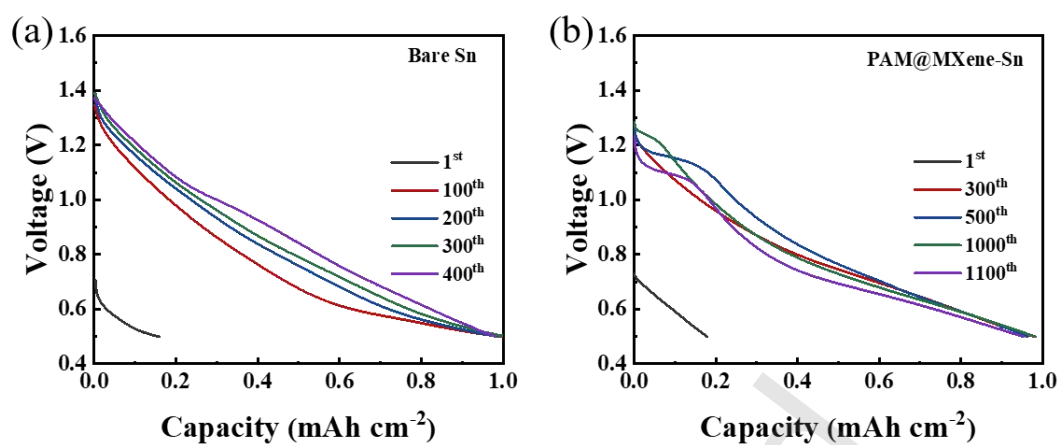


Figure S38 Discharge profiles of (a) bare Sn||I₂ and (b) PAM@MXene-Sn||I₂ cells at 10 mA cm⁻².

Table S1 Performance comparison of Sn||Cu asymmetric cells between this work and recently reported aqueous Sn-based batteries.

Anode	Electrolyte	CE (%)	Cycling condition (h)	Refs.
Sn/Cu _s	2 M H ₂ SO ₄ +0.1 M SnSO ₄	98.00	2 mA cm ⁻² /1 mAh cm ⁻² (260 h)	1
Sn foil	3 M MSA+1 M Sn(MSA) ₂	99.95	10 mA cm ⁻² /1 mAh cm ⁻² (323 h)	2
	2 M H ₂ SO ₄ +0.2 M SnSO ₄ +1 g L ⁻¹ GEL	99.50	1 mA cm ⁻² /1 mAh cm ⁻² (100 h)	
Sn/G _s	2 M H ₂ SO ₄ +0.2 M SnSO ₄ +1% POPE	99.00	6 mA cm ⁻² /5 mAh cm ⁻² (500 h)	3
Sn foil	1 M SnCl ₂	99.93	1 mA cm ⁻² /5 mAh cm ⁻² (960 h)	4
Sn foil	2 M MSA+1 M Sn (MSA) ₂ +50 mM HQ	99.90	2 mA cm ⁻² /2 mAh cm ⁻² (600 h)	5
Sn foil	2 M H ₂ SO ₄ +0.1 M SnSO ₄ +QO ⁺	99.96	3 mA cm ⁻² /3 mAh cm ⁻² (800 h)	6
Sn@Ag-VG	2 M H ₂ SO ₄ +0.1 M SnSO ₄	99.72	1 mA cm ⁻² /1 mAh cm ⁻² (1,000 h)	7
Sn foil	2 M H ₂ SO ₄ +0.2 M SnSO ₄ +0.6 M TPE	99.4	5 mA cm ⁻² /5 mAh cm ⁻² (1,000 h)	8
Sn foil	2 M H ₂ SO ₄ +0.2 M SnSO ₄ +1 mM PVP	99.9	1 mA cm ⁻² /1 mAh cm ⁻² (1,400 h)	9
PAM@MXene-Sn	3 M MSA+1 M Sn (MSA) ₂	99.4	1 mA cm ⁻² /1 mAh cm ⁻² (2,800 h)	This work
		99.9	10 mA cm ⁻² /0.5 mAh cm ⁻² (2,000 h)	

Table S2 Performance comparison of full cells with different cathodes between this work and recently reported aqueous Sn-based batteries.

Anode	Cathode	Capacity	Cycling life (cycles)	Refs.
Sn/Cu _s	PANI	0.15 mAh cm ⁻² (20 mA cm ⁻²)	1,600	1
	MnO ₂	0.5 mAh cm ⁻² (10 mAh cm ⁻²)	2,400	
Sn foil	TCBQ	95 mAh g ⁻¹ (10 C)	2,200	2
Sn/G _s	PbO ₂	5 mA cm ⁻² (1 mAh cm ⁻²)	2,000	3
Sn foil	LMO	70 mAh g ⁻¹ (0.5 A g ⁻¹)	1,000	4
Sn foil	TCBQ	150 mAh g ⁻¹ (1 A g ⁻¹)	700	5
Sn foil	MnO ₂	5 mAh cm ⁻² (20 mA cm ⁻²)	350	6
Sn@Ag-VG	I ₂	179.24 mAh g ⁻¹ (1 C)	2,000	7
	IBr	322.5 mAh g ⁻¹ (0.1 A g ⁻¹)	200	
	LCO	134.2 mAh g ⁻¹ (0.5 C)	600	
Sn foil	MnO ₂	1 mAh cm ⁻² (20 mA cm ⁻²)	90	8
Sn foil	PbO ₂	10 mAh cm ⁻² (5 mA cm ⁻²)	200	9
	MnO ₂	1 mAh cm ⁻² (6 mA cm ⁻²)	3,000	
	MnO ₂	1 mAh cm ⁻² (20 mA cm ⁻²)	6,000	
PAM@MXene-Sn	I ₂	1 mAh cm ⁻² (10 mA cm ⁻²)	1,100	This work

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