

RESEARCH ARTICLE

Structural and Interfacial Engineering of $\text{Ti}_3\text{C}_2\text{T}_x$ Reinforced Polyacrylamide–Polyvinyl Alcohol Double-Network Hydrogel Electrolyte for Long-Durable Flexible Zinc-Ion Batteries

Aakash Carthick Radjendirane^{1,2} | Narendhar Chandrasekar³  | Saradh Prasad Rajendra⁴ | Ju Hyun Oh² | Khalid Eidah Alzhrani⁴ | Seung Jun Lee²  | Subramania Angaiah^{1,2} 

¹Electro-Materials Research Lab, Centre for Nanoscience and Technology, Pondicherry University, Puducherry, India | ²Department of IT -Energy Convergence (BK21 FOUR), Korea National University of Transportation, Chungju, Republic of Korea | ³Department of BioNanoTechnology, Gachon University, Seongnam-si, Republic of Korea | ⁴Research Chair for Laser Diagnosis of Cancers, Department of Physics and Astronomy, College of Science, King Saud University, Saudi Arabia

Correspondence: Narendhar Chandrasekar (narendhar.nano@gmail.com) | Seung Jun Lee (sjlee@ut.ac.kr) | Subramania Angaiah (a.subramania@gmail.com)

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ABSTRACT

Hydrogel electrolytes hold a prominent interest in large-scale Zn-based energy storage systems due to their inherent safety, low cost, and stable cycling performance. However, achieving simultaneous regulation of electrolyte structure and interface stabilization for uniform Zn^{2+} deposition, dendrite suppression, and long-term cycling stability remains an optimal challenge. Herein, the current study emphasizes the advancement of 2D- $\text{Ti}_3\text{C}_2\text{T}_x$ (Ti-MXene) incorporated polyacrylamide (PAM)–polyvinyl alcohol (PVA) double network (DN)-based composite hydrogel electrolytes (CHGEs) for flexible zinc-ion batteries (FZIBs). As a result, the developed PAM–PVA/Ti-MXene (0.1 wt.%) displays excellent mechanical strength of 3.6 MPa and ionic conductivity of 22.9 mS cm^{-1} . In addition, the Zn//Zn symmetrical cell also exhibits a stable stripping/plating of 1000 h at 0.1 mA cm^{-2} as well as exceptional reversibility behavior of Zn//Cu assymetric cell with average coulombic efficiency of 99.4% for over 500 cycles. Furthermore, the fabricated Zn// V_2O_5 battery exhibits a high discharge capacity of 178 mAh g^{-1} at 0.1 A g^{-1} and excellent capacity retention of 84% for 500 cycles with a coulombic efficiency of 100%. Overall, the present investigation of DN-based CHGEs will provide new insights into a stable, dendrite-free Zn anode, enabling high-performance flexible devices and beyond.

1 | Introduction

The development of double network (DN)-based composite hydrogel electrolytes (CHGEs) has recently emerged as a breakthrough for flexible zinc-ion batteries (FZIBs) owing to their excellent mechanical properties and high ionic conductivity. Further, the optimum wearable and stretchable batteries must ensure bending, stretching, and twisting without breaking. The DN-based CHGE can easily withstand these configurations by significantly enhancing flexibility and toughness [1, 2]. Generally, chemically cross-linked single-network (SN) hydrogel electrolytes result in low

ionic conductivity, making them unsuitable for high-performance FZIBs. Therefore, the DN-based CHGE is a vital option for enhancing both the mechanical properties and ionic conductivity of the FZIBs. Furthermore, the excellent mechanical behavior of DN-based CHGE can dissipate external stress and suppress dendrite growth during repeated Zn stripping/plating cycles, thereby enhancing battery life. In general, DN hydrogel comprises two polymeric networks: the first is the primary network, attributed to a rigid structure that provides structural integrity and mechanical strength, whereas the second network is loosely cross-linked to provide flexibility and increased toughness. During repeated

cycles, the primary network breaks down locally due to energy dissipation. In such cases, the second network of DN holds the structure together by preventing deformation [3, 4]. Therefore, DN-based CHGE offers the ideal combination of mechanical robustness and ionic conductivity, as well as electrochemical stability required for next-generation FZIBs, especially in flexible and wearable electronics [5–7].

Generally, the DN-based hydrogel is designed so that the primary network dissipates mechanical stress, thereby preventing extreme material failure. In this regard, polyacrylamide is the preferred primary network, as it provides excellent mechanical strength and the ability to sacrificially break and dissipate energy, making it highly effective at toughening the hydrogel network. For the secondary network, polymers such as carboxymethyl cellulose (CMC), xanthan gum (XG), gelatin, sodium alginate (SA), polyacrylic acid (PAA) [8], polyethylene oxide (PEO), polyvinyl alcohol (PVA), etc., have been widely used [9, 10]. Among the DN hydrogels, polyacrylamide (PAM)–polyvinyl alcohol (PVA) is typically the most suitable DN for ultra-stable CHGEs required for high-performance FZIBs [11–13]. This is because PVA forms strong hydrogen bonds with PAM, thereby enhancing mechanical strength and flexibility. Similarly, PVA's increased hydrophilicity ensures a high water content, which is essential for improving ionic conductivity [14, 15]. In recent times, significant advancements have been made in developing DN-based CHGEs for highly stable FZIBs. For instance, Huihui Zheng et al. developed a polyacrylamide-k-carrageenan-based DN hydrogel exhibiting a tensile strength of 226 kPa and an ionic conductivity of 22.6 mS cm^{−1} [16]. Xie et al. constructed a dual network of polyacrylamide-bacterial cellulose hydrogel electrolytes that displayed ionic conductivity of 18.1 mS cm^{−1} and mechanical strength of 260 kPa [2]. Furthermore, Zhang et al. developed a polyacrylamide-agarose double-network hydrogel electrolyte with a low ionic conductivity of 11.53 mS cm^{−1}, leading to capacity fading [17]. Similarly, Zeng et al. developed a zwitterionic group-based dual network of polyacrylamide-sodium acrylate, which is endowed with high ionic conductivity (33.61 mS cm^{−1}) and low mechanical properties (77 kPa) [18]. Overall, from this aspect, the important drawbacks of the DN-based CHGE are either low ionic conductivity or low mechanical properties. So, developing both parameters in the hydrogel is a difficult task.

To tackle these issues, incorporating 2D nanofiller Ti-MXene into the DN hydrogel will enhance mechanical and ionic conductivity. Notably, the unique characteristics of Ti-MXene, with its abundant surface groups (−OH, −F, −Cl), enhance ionic mobility by providing more active sites for ion hopping and facilitating ion transport. Furthermore, the Ti-MXene synergizes with the polymer networks, distributing stress more uniformly and enhancing the hydrogel's mechanical robustness [19, 20]. Moreover, the synergistic effects of the DN structure and Ti-MXene enhance the hydrogel's flexibility without compromising its mechanical strength and ionic conductivity, which are highly critical for high-performance FZIBs. Previously, our group developed a series of Ti-MXene incorporated CHGEs, such as polyacrylamide (PAM) [21], poly(acrylamide-co-acrylic acid) P(AM-AA) [22], and polyacrylamide-polyethylene glycol (PAM-PEG) [23] IPN-based systems for FZIBs. These systems exhibited progressive improvements in either ionic conductivity or mechanical strength; however, further enhancement of long-cycle durability remained necessary for practical applications.

Hence, the present work relies on the advancement of 2D inorganic nanofiller, Ti-MXene-incorporated PAM–PVA DN-based composite hydrogel electrolytes for high-performance FZIBs. Herein, various concentrations of Ti₃C₂T_x MXene (0.1, 0.2, and 0.3) incorporated PAM–PVA-based CHGEs were subjected into their physiochemical and electrochemical properties. Among these different wt.% hydrogels, 0.1 wt.% of Ti-MXene incorporated PAM–PVA exhibits increased ionic conductivity of 22.9 mS cm^{−1} and excellent mechanical behavior of 3.6 MPa. Furthermore, V₂O₅ is used here as an active cathode material due to its economical, high theoretical capacity, and excellent structural stability, which help maintain the material during repeated charge–discharge cycles, a crucial factor for the long-term cycling stability of the battery. Furthermore, the as-fabricated FZIB using 0.1 wt.% Ti-MXene-incorporated PAM–PVA-based CHGEs delivers a high specific capacity of 178 mAh g^{−1} at 0.1 A g^{−1}, along with excellent capacity retention over 500 cycles. This Ti-MXene-incorporated PAM–PVA-based CHGE also helps suppress zinc dendrite growth by altering the distribution of Zn ions at the electrode interface, thereby enhancing battery safety and longevity. Overall, this work demonstrates a novel approach for developing a highly safe, low-cost, and pliable-based CHGE for high-performance, flexible, and wearable energy storage devices.

2 | Experimental Section

2.1 | Materials

The Ti-MAX (Ti₃AlC₂) phase (98%, 400 mesh) was purchased from Nanoshel Chemical Company. Further, materials such as acrylamide (Mw 71.08), N, N-methylene bis-acrylamide (GR 99%+, Mw 154.17) were obtained from Otto Chemie Pvt. Ltd. Ammonium persulfate (extrapure AR, N₂H₈S₂O₈) and vanadium pentoxide, (99.5%, V₂O₅) were purchased from Sisco Research Laboratories (SRL) Pvt. Ltd. Polyvinyl alcohol (99%+, Mw 85,000–1,25,000) and lithium fluoride (300 mesh, LiF) were obtained from Sigma Aldrich. The electrolyte salt, zinc sulfate (Hi-AR, ZnSO₄), was obtained from Hi-Media. Zinc foil (thickness: 100 microns) was obtained from Sigma Aldrich.

2.2 | Preparation of Delaminated 2D Ti₃C₂T_x MXene Nanosheets

The delaminated 2D-Ti₃C₂T_x MXene nanosheets were prepared as described in the author's previous work [24, 25]. First, 2g of Ti-MAX (Ti₃AlC₂) powder was slowly added to the 40 mL mixture (2g LiF / 9 M HCl). The solution was then etched at 27°C for 24 h under continuous stirring. Further, the etched products were washed via multiple cycles of centrifugation in DI water until the pH approached neutrality. For the delamination process, the subsequent mixture was sonicated for 2 h, and then shaken well and centrifuged to collect the supernatant, which contained a single layer of Ti-MXene nanosheets.

2.3 | Preparation of Ti₃C₂T_x MXene @ Double Network-Based CHGEs

First, 2 g of AM and 0.2 g of hot water-soluble PVA were dissolved in DI water. To this, various concentrations of Ti₃C₂T_x

MXene (0.1, 0.2, and 0.3 wt.%) were added and sonicated for 1 h. Furthermore, the initiator, ammonium persulfate, and the cross-linker N, N'-methylene bis-acrylamide were added, and the mixture was stirred continuously for 24 h at room temperature (25°C). Then, the mixture was transferred onto the tissue culture plate and dried in a vacuum oven at 60°C for 4 h [26]. Finally, the hydrogels were soaked in 2 M ZnSO₄ electrolyte solution for 24 h to obtain PAM-PVA/Ti-MXene DN-based CHGEs. Herein, the sample codes PAM-PVA/Ti-MXene, PAM-PVA/Ti-MXene (0.1 wt.%), PAM-PVA/Ti-MXene (0.2 wt.%), and PAM-PVA/Ti-MXene (0.3 wt.%) were used throughout the manuscript to represent pristine, and 0.1, 0.2, and 0.3 wt.% Ti-MXene incorporated PAM-PVA based CHGEs.

2.4 | Fabrication of Zn//CHGE//V₂O₅ Battery

For the fabrication of ZIB, Zn foil is used as the anode, V₂O₅ as the cathode, and PAM-PVA/Ti-MXene (0.1 wt.%) CHGE as the electrolyte using an Al pouch cell case. The active material was prepared via ultrasonication bath treatment of V₂O₅/CNT in a 9:1 ratio. Further, the cathode was prepared by mixing the active material, acetylene black, and binder (PVDF) in a 7:2:1 ratio using N-methyl-2-pyrrolidone (NMP) solvent. Then, the as-prepared slurry was coated on the graphite sheet and vacuum-dried at 85°C for 8 h [27, 28]. The mass loading of the active cathode material was 1.5–2.0 mg cm⁻². Herein, the V₂O₅ sample code was used throughout the manuscript to specify the cathode material.

2.5 | Characterizations

Physicochemical characterizations, including X-ray diffraction (XRD), Fourier transform infrared (FTIR) spectroscopy, thermogravimetric analysis (TGA), mechanical studies, and X-ray photoelectron spectroscopy (XPS), have been performed on the as-prepared pristine and Ti-MXene (0.1, 0.2, and 0.3 wt.%) incorporated PAM-PVA hydrogels. Electrolyte uptake (EU) studies have also been conducted by immersing the hydrogel samples in a 2 M ZnSO₄ electrolyte solution for 48 h. The electrolyte uptake was calculated using the equation [29]:

$$EU(\%) = \frac{W - W_0}{W_0} \times 100$$

where (*W*₀) represents the weight of the fully dried hydrogel membrane, while (*W*) corresponds to the weight of the membrane after immersion in the electrolyte solution. Before the electrolyte uptake measurement, the as-prepared hydrogel samples were vacuum-dried to remove residual water and solvent, ensuring that the initial weight (*W*₀) corresponded to the dry polymer network, free of absorbed solvent. The dried membranes were then immersed in 2 M ZnSO₄ electrolyte solution for 48 h to allow complete electrolyte absorption. After immersion, the samples were removed, gently wiped to eliminate surface electrolyte, and weighed immediately to obtain (*W*) [21, 22].

The post-cycling behavior of the Zn anode has been analyzed using XRD and FESEM studies for the galvanostatic charge-discharge (GCD) stability cells.

Electrochemical characterizations of electrochemical impedance spectroscopy (EIS) have been performed to determine the ionic

conductivity of the as-prepared pristine and Ti-MXene incorporated PAM-PVA based CHGEs. The ionic conductivity of the CHGEs was calculated using the equation [30]:

$$\sigma = \frac{l}{R \times A}$$

Herein, *l* refers to the thickness (0.9 mm), *R* denotes resistance, and *A* refers to the area (15 × 15 mm²) of the CHGEs.

Linear sweep voltammetry (LSV) studies have been performed to understand the stability window of PAM-PVA/Ti-MXene (0.1 wt.%) CHGE. Moreover, the transference number (*t*⁺) is calculated by a Bruce-Vincent method using a relation [31]:

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

whereas *R*₀ and *R*_s are the initial and steady-state resistance and *I*₀ and *I*_s are the initial and steady-state currents.

Furthermore, stripping/plating behavior is performed using Zn//Zn configurations for the PAM-PVA/Ti-MXene (0.1 wt.%) CHGE at 0.1 mA cm⁻² for 1000 h. The rate capability behavior of the stripping/plating cell was evaluated for PAM-PVA/Ti-MXene (0.1 wt.%) CHGE at current densities of 0.1, 0.5, 1.0, 2.0, and 5.0 mA cm⁻² for 60 h. Furthermore, the Zn//Cu stripping-plating studies were performed at 0.1 mA cm⁻² over 500 cycles to understand the reversibility behavior of the PAM-PVA/Ti-MXene (0.1 wt.%) CHGE.

Zinc-ion storage performance (Zn//V₂O₅ pouch cell) was evaluated using galvanostatic charge-discharge (GCD) studies over a voltage window of 0.2–1.8 V. Further, the rate capability was assessed at current densities of 0.1, 0.2, 0.5, 1.0, 2.0, 5.0, and again 0.1 A g⁻¹ for 35 cycles [32]. The long-cycle stability was evaluated at 0.5 A g⁻¹ over 500 cycles. Finally, flexibility tests were performed using different bending angles (0°, 45°, 90°, 180°) on the pouch cell at 1 A g⁻¹ over 40 cycles. The bending states were applied sequentially rather than continuously or dynamically to evaluate electrochemical performance under different deformation configurations. The bending radius of the device was approximately ~5 mm, which was limited by the mechanical constraints of the flexible cell configuration.

3 | Results and Discussion

In the present work, the PAM-PVA/Ti₃C₂T_x CHGE is designed based on a typical DN hydrogel architecture. In this structure, PAM forms the primary chemically cross-linked rigid network through free-radical polymerization, which provides structural integrity. Meanwhile, PVA acts as the secondary flexible network, formed primarily through physical interactions such as extensive hydrogen bonding and chain entanglement. Specifically, the hydroxyl (—OH) groups of PVA interact strongly with the amide (—CONH₂) groups of PAM via hydrogen bonding, forming an interpenetrating yet cooperative network structure. This dual-network configuration effectively combines the rigidity of the chemically cross-linked PAM network with the elasticity of the physically cross-linked PVA network, which is responsible for the enhanced mechanical strength and improved ionic transport properties observed in the hydrogel electrolyte [33, 34].

Therefore, the chemically crosslinked PAM provides structural rigidity, while the physically crosslinked PVA contributes flexibility and water retention, forming a tough, stretchable matrix. Furthermore, the incorporation of $\text{Ti}_3\text{C}_2\text{T}_x$ MXene nanosheets into the DN introduces continuous ion-conducting pathways, increasing ionic conductivity by facilitating rapid Zn^{2+} transport. Generally, $\text{Ti}_3\text{C}_2\text{T}_x$ MXene possesses high intrinsic electronic conductivity. However, the amount of $\text{Ti}_3\text{C}_2\text{T}_x$ used is relatively low and it is uniformly dispersed within the polymer hydrogel matrix. Under these conditions, the $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets remain spatially isolated and do not form a continuous electronic percolation network. Therefore, they do not create an internal electronic pathway that could cause short-circuiting or significantly increase self-discharge. Instead, the incorporated $\text{Ti}_3\text{C}_2\text{T}_x$ mainly functions as an ion-transport facilitator and structural reinforcement within the hydrogel electrolyte. The hydrophilic surface terminations of $\text{Ti}_3\text{C}_2\text{T}_x$ ($-\text{OH}$, $-\text{F}$, and $-\text{Cl}$) enhance electrolyte affinity and promote the formation of interconnected ionic transport pathways, thereby facilitating efficient Zn^{2+} migration and improving electrode-electrolyte interfacial stability. In addition, the negatively charged surface of $\text{Ti}_3\text{C}_2\text{T}_x$ stabilize Zn^{2+} ions, suppresses ion pairing, and enhances ionic mobility within the hydrogel matrix [20, 35]. As a result, the PAM-PVA/Ti-MXene CHGE exhibits improved ionic conductivity and electrochemical stability without significantly increasing electronic leakage. Overall, the combined structure of Ti-MXene with PAM-PVA DN interacts via a strong hydrogen bond (Figure 1), resulting in an exceptional ionic conductivity and mechanical behavior, making it ideal for stable, durable, and flexible ZIBs.

Most importantly, the XRD pattern of $\text{Ti}_3\text{C}_2\text{T}_x$ MXene confirms the successful removal of Al layer from the Ti_3AlC_2 MAX phase, as evidenced by the disappearance of the characteristic MAX peak (104) and the emergence of a pronounced (002) diffraction

peak around $2\theta \approx 6^\circ$ [36], confirming the increased interlayer spacing and effective delamination, as shown in Figure S1. Furthermore, the XRD patterns of PAM-PVA and Ti-MXene-incorporated PAM-PVA DN-hydrogels are shown in Figure 2a. The broadened peak at $\sim 26^\circ$ (2θ) indicates the amorphous nature of the as-prepared PAM-PVA hydrogel. Furthermore, the addition of Ti-MXene into PAM-PVA retains its amorphous nature, thereby enhancing ionic conductivity. The broadened peak, along with a slight shift observed in Ti-MXene of various concentrations, can be attributed to the strong interactions between the PAM-PVA hydrogel matrix and the Ti-MXene layers [19].

The chemical interactions of the PAM-PVA and Ti-MXene-incorporated PAM-PVA DN-hydrogels are characterized by the FTIR spectroscopy technique, shown in Figure 2b. In the pristine PAM-PVA hydrogel, the peak at $\sim 1095\text{ cm}^{-1}$ corresponds to the C—O stretching vibration of PVA. Further, the absorption peak at $\sim 1658\text{ cm}^{-1}$ is associated with the stretching vibrations of the carbonyl (C=O) group in the amide linkage of PAM. The bands near ~ 1450 and $\sim 1418\text{ cm}^{-1}$ correspond to C—N stretching and N—H deformation vibrations, respectively. Then, the broad absorption band around $3186\text{--}3420\text{ cm}^{-1}$ is attributed to the stretching vibrations of hydroxyl groups ($-\text{OH}$) present in both PVA and PAM. Moreover, all these peaks are reminiscent of the Ti-MXene-incorporated PAM-PVA DN-hydrogels, with minimal shifts, confirming the presence of strong hydrogen-bonding interactions between the hydroxyl groups of PAM-PVA and the surface functional groups of Ti-MXene.

For thermal behavior, the weight loss occurs via three stages: $26\text{--}270^\circ\text{C}$, $270\text{--}420^\circ\text{C}$, and $420\text{--}600^\circ\text{C}$. The first weight loss, occurring between 26°C and 270°C , is attributed to the evaporation of both bound and free water within the hydrogel network, resulting in a 20% weight loss. The second weight loss, occurring between $270\text{--}420^\circ\text{C}$, corresponds to the degradation of the side

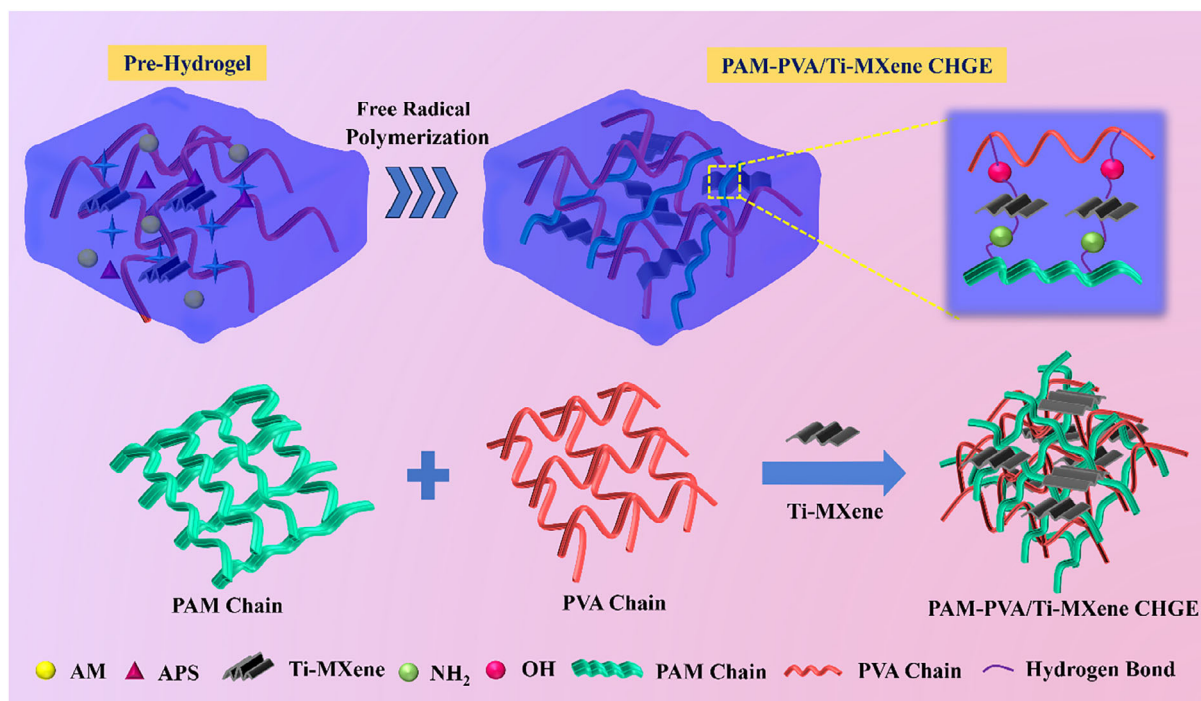


FIGURE 1 | Schematic illustration for the preparation of PAM-PVA/Ti-MXene CHGE.

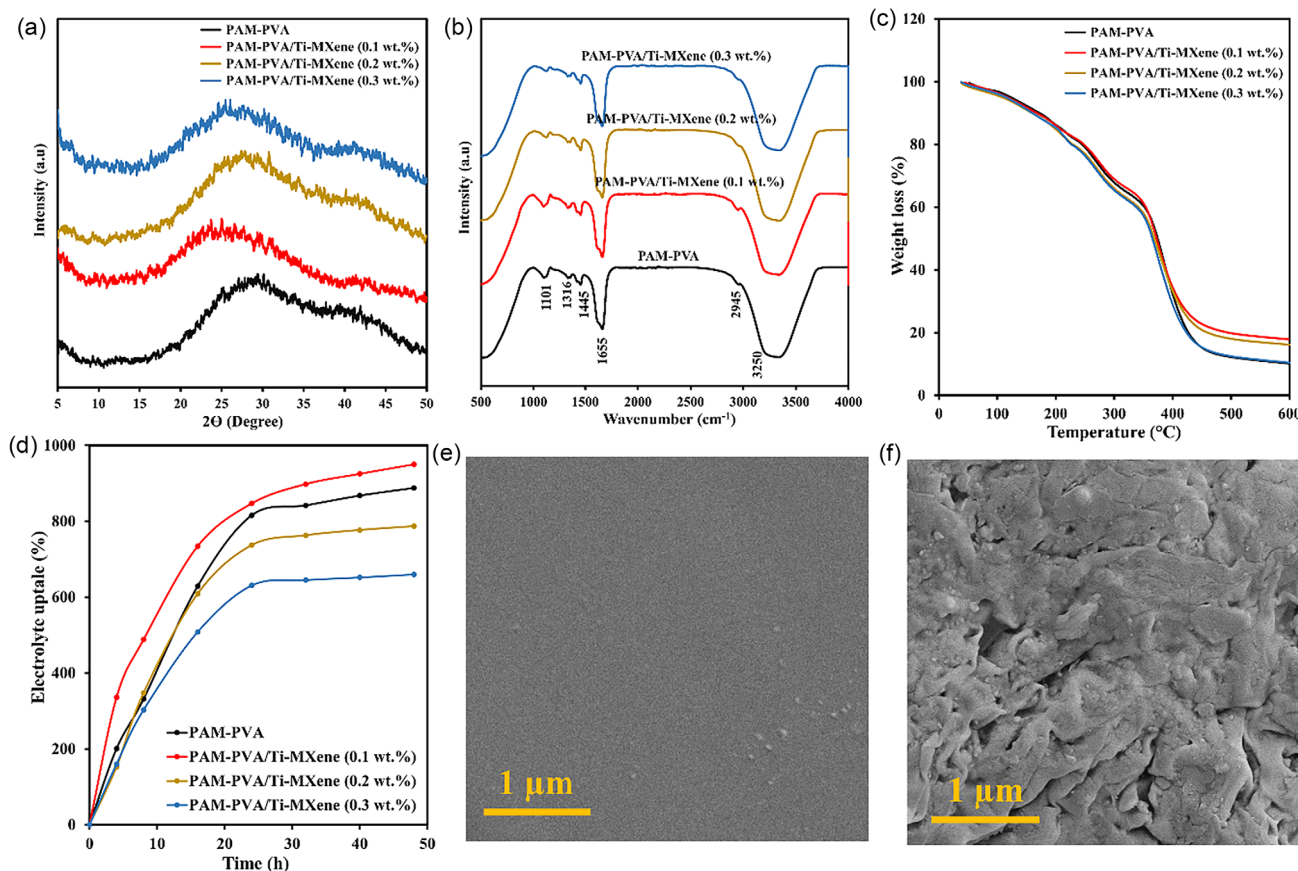


FIGURE 2 | (a) XRD patterns, (b) FTIR spectra, (c) TGA studies, (d) electrolyte uptake of pristine PAM-PVA and PAM-PVA/Ti-MXene hydrogels, FESEM micrographs of (e) PAM-PVA and (f) PAM-PVA/Ti-MXene (0.1 wt.%) DN hydrogels.

chain of the PAM-PVA DN structure. Furthermore, the third-stage weight loss from 420°C to 600°C is associated with the complete decomposition of the PAM-PVA DN backbone. Furthermore, it is clearly apparent from Figure 2c that the weight loss rate of the Ti-MXene-incorporated PAM-PVA DN hydrogel is lower than that of the pristine one, indicating that Ti-MXene nanosheets act as an effective thermal barrier, thereby delaying polymer decomposition.

Figure 2d clearly shows that the electrolyte uptake of PAM-PVA/Ti-MXene DN-hydrogel exhibits higher electrolyte uptake than the pristine PAM-PVA hydrogel. This is owing to the functional groups of Ti-MXene (—OH, —O, and —F), and its high surface area helps trap and retain water molecules within its interlayers. Moreover, the negatively charged Ti-MXene surface attracts Zn²⁺ ions, promoting their migration within the polymer network and thereby improving ionic mobility and electrolyte uptake compared to the pristine PAM-PVA DN hydrogel. Among the prepared samples, the PAM-PVA/Ti-MXene (0.1 wt.%) DN hydrogel shows an electrolyte uptake of 928% after 48 h. However, at higher Ti-MXene concentrations (0.2 and 0.3 wt%), nanosheet aggregation occurs within the polymer matrix, reducing the available free water volume for electrolyte storage and thereby decreasing electrolyte uptake.

The FESEM image of the synthesized Ti-MXene reveals the exfoliated sheet-like morphology, confirming the delamination of Ti₃C₂T_x MXene from the Ti-MAX phase, which is clearly displayed in Figure S2. Furthermore, FESEM micrographs of PAM-PVA hydrogel and PAM-PVA/Ti-MXene (0.1 wt.%) DN

samples are displayed in Figure 2e,f. The PAM-PVA/Ti-MXene (0.1 wt.%) DN sample exhibits denser and rougher pore walls due to the strong interaction between Ti-MXene nanosheets and PAM-PVA chains. Furthermore, incorporating Ti-MXene into the DN makes the hydrogel more structured, and its denser network accelerates the migration of Zn²⁺ ions, thereby significantly increasing ionic conductivity. In contrast, the PAM-PVA hydrogel forms smooth polymer walls with uniform, slightly swollen pores, resulting in low ionic conductivity.

Most importantly, ionic conductivity is a key parameter for evaluating the endurance and performance of CHGEs in FZIBs, as it reflects the efficiency of Zn²⁺ ion transport through the electrolyte. As shown in Figure 3a, the Ti-MXene-incorporated PAM-PVA DN-based CHGEs exhibit significantly higher ionic conductivity than the PAM-PVA CHGE. The measured ionic conductivity values for the pristine PAM-PVA and PAM-PVA/Ti-MXene (0.1, 0.2, and 0.3 wt%) DN-based CHGEs are 13.6, 22.9, 19.6, and 17.2 mS cm⁻¹, respectively. The enhanced ionic conductivity observed for the PAM-PVA/Ti-MXene (0.1 wt.%) DN-CHGE is believed to arise from two synergistic mechanisms. First, the two-dimensional Ti₃C₂T_x MXene nanosheets introduce interfacial ion-transport pathways, where Zn²⁺ ions can migrate along the MXene surface. The abundant surface functional groups of Ti₃C₂T_x (—OH, —O, and —F) can interact electrostatically with Zn²⁺ ions, facilitating surface-assisted ion migration (ion-hopping) along the MXene nanosheets. This interfacial transport pathway reduces ion diffusion resistance within the hydrogel electrolyte. Second, incorporating

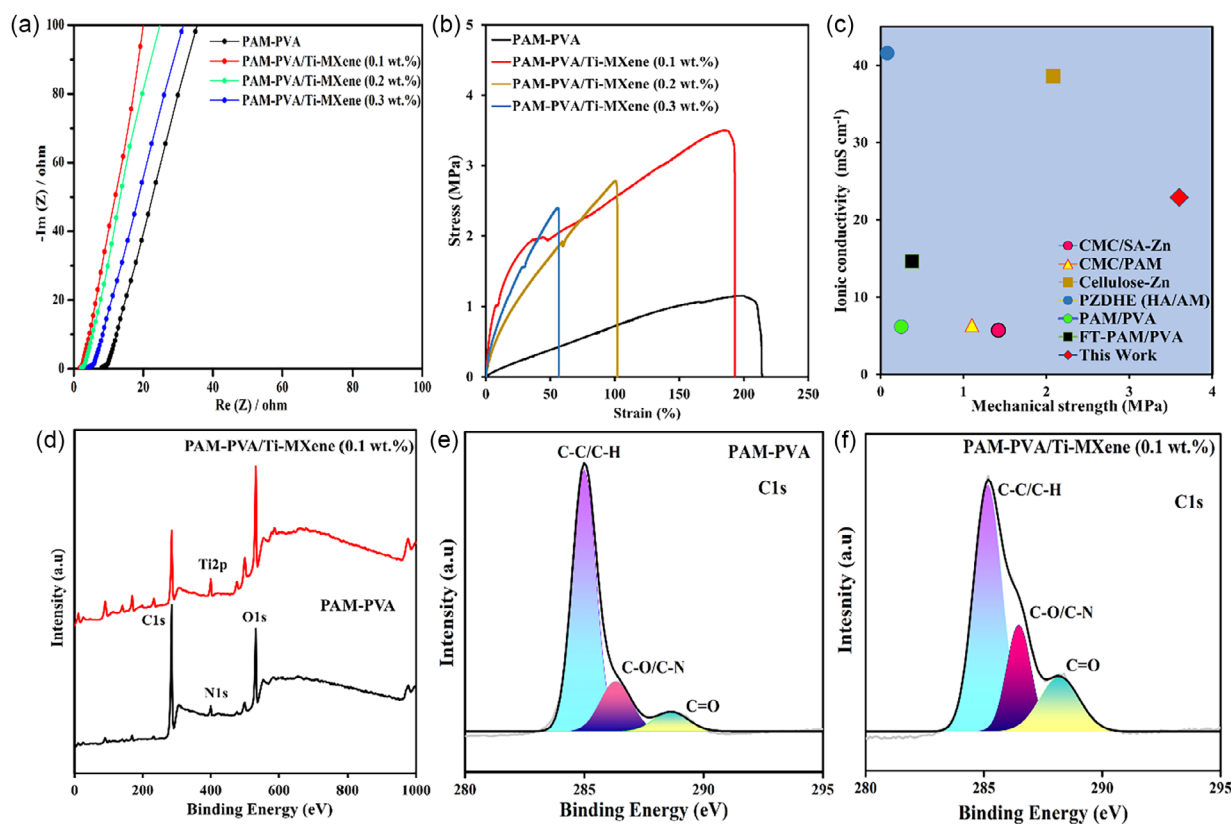


FIGURE 3 | (a) Ionic conductivity studies of the prepared CHGEs, (b) tensile strength of PAM-PVA and PAM-PVA/Ti-MXene hydrogels, (c) comparison plot of Ionic conductivity versus tensile strength with the recent DN systems, (d) XPS survey spectra, deconvoluted C1s spectra of (e) PAM-PVA, and (f) PAM-PVA/Ti-MXene hydrogels.

a small amount of Ti-MXene alters the bulk polymer network structure. The nanosheets act as physical spacers within the PAM-PVA matrix, thereby reducing excessive polymer chain entanglement and increasing free volume and interconnected ionic channels within the hydrogel network. This structural modulation improves electrolyte uptake to 928% and enhances bulk Zn^{2+} mobility through the hydrogel matrix. Therefore, the observed improvement in ionic conductivity results from a combined effect of surface-assisted ion transport and enhanced bulk ion diffusion pathways. Furthermore, beyond 0.2 and 0.3 wt.%, Ti-MXene begins to agglomerate, blocking ion diffusion, increasing tortuosity, hindering ion mobility, and reducing ionic conductivity.

Mechanical properties are key parameters in defining the optimal hydrogel electrolytes for real-time FZIB applications. As shown in Figure 3b, the tensile strength of PAM-PVA/Ti-MXene (0.1 wt.%) DN hydrogel (3.6 MPa) is 3.2 times higher than the pristine PAM-PVA hydrogel (1.1 MPa). The results demonstrate that the PAM-PVA/Ti-MXene (0.1 wt.%) DN-CHGE exhibits the most balanced mechanical performance, with a maximum tensile strength of 3.6 MPa and an elongation at break of approximately 190%, indicating excellent strength and flexibility. In contrast, when the Ti-MXene content increases to 0.2 wt.% and 0.3 wt.%, the tensile strength decreases to 2.7 and 2.2 MPa, respectively, while the elongation at break decreases to 102% and 58%, respectively. This trend indicates that excessive Ti-MXene loading reduces the flexibility and extensibility of the hydrogel network. This is entirely due to Ti-MXene's intervention in the DN hydrogel. Herein, Ti-MXene nanosheets act as

a nanofiller, reinforcing the DN matrix, and the enhanced interactions between DN and Ti-MXene prevent premature hydrogel breakage. Furthermore, the substantial functional groups ($-\text{OH}$ and $-\text{O}-$) of Ti-MXene, when assisted by PAM-PVA, form additional strong hydrogen bonds, thereby improving interfacial adhesion and making the hydrogel more durable [35]. Therefore, considering the combined effects of ionic conductivity, electrolyte uptake, and mechanical robustness, the 0.1 wt.% Ti-MXene loading provides the optimal composition for achieving mechanically stable and high-performance CHGE for FZIBs. Furthermore, the Comparison plot (ionic conductivity vs. tensile strength) with the recently reported DN systems clearly demonstrates the superiority of the developed DN-CHGE sample (Figure 3c) [33, 37–40].

The XPS survey spectrum confirms the existence of C1s, N1s, O1s, and Ti2p at the respective binding energies of 285.1, 401.3, 532.4, and 460.1 eV for the 0.1 wt.% Ti-MXene hydrogel, as shown in Figure 3d. Further, the deconvoluted C1s spectrum displays the upsurge in the $\text{C}=\text{O}$ peak in the PAM-PVA/Ti-MXene (0.1 wt.%) DN, indicating the enhanced interaction through hydrogen bonding between the carbonyl/amino groups in PAM and the functional groups ($-\text{OH}$, $=\text{O}$) of Ti-MXene seen in Figure 3e,f. These enhanced interactions are highly beneficial in improving network stability, mechanical reinforcement, and ion mobility within the hydrogel matrix.

The electrochemical characterizations of the prepared CHGEs were carried out by symmetric cells, asymmetric cells, and battery device performances. First, the PAM-PVA/Ti-MXene

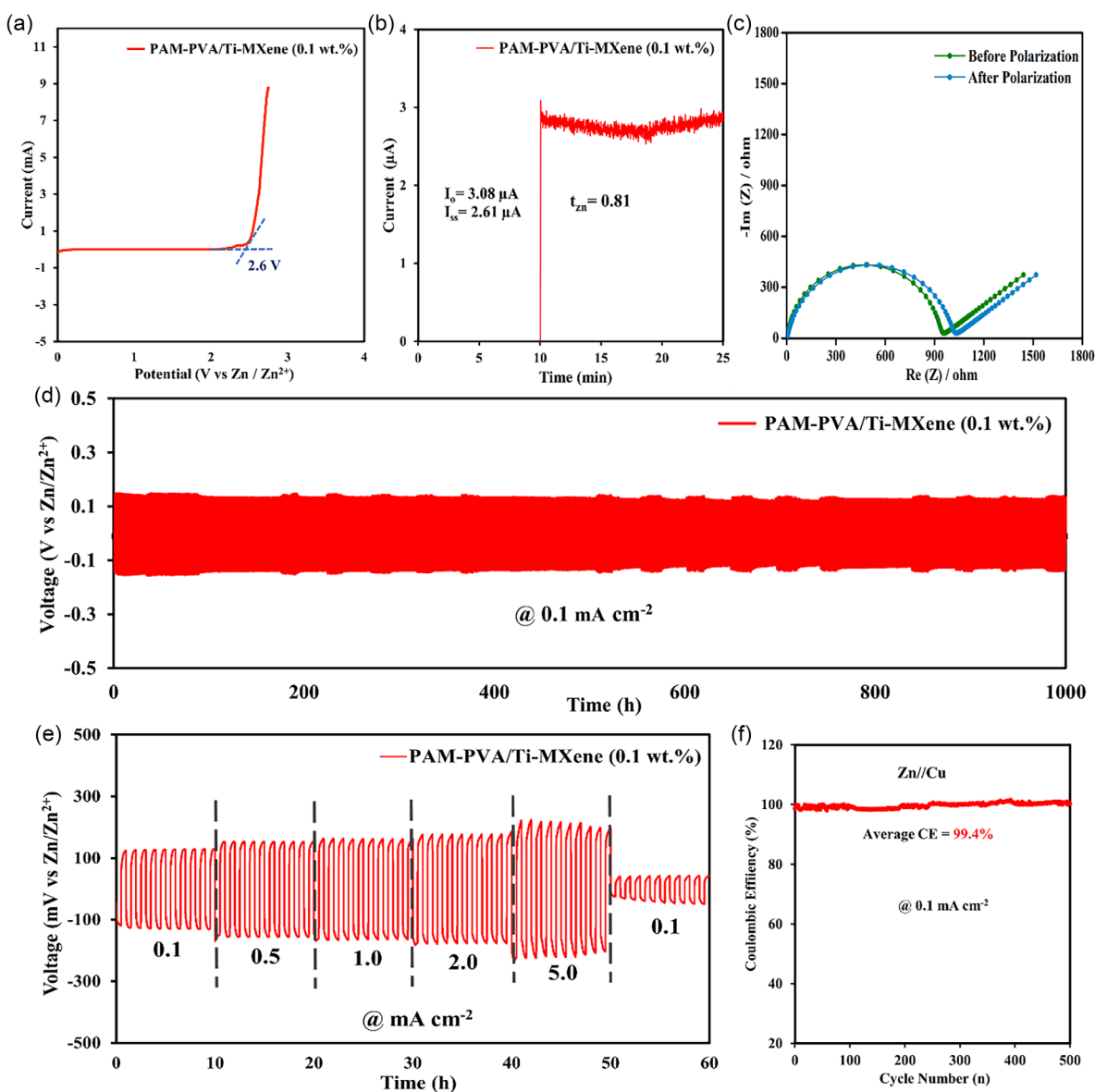


FIGURE 4 | (a) Linear sweep voltammetry plot, (b) chronoamperometry study, (c) EIS before and after CA, (d) stripping/plating behavior of Zn//Zn, (e) rate capability studies of Zn//Zn at various current densities, and (f) CE of asymmetric Zn//Cu studies of the PAM-PVA/Ti-MXene (0.1 wt.%) DN-based CHGE.

(0.1 wt.%) DN-based CHGE shows a wider stability window of 2.6 V (Figure 4a), which is attributed to the Ti-MXene's role in stabilizing water decomposition. Herein, the Ti-MXene aids in binding free water molecules, reducing water electrolysis at high voltage. Overall, the developed PAM-PVA/Ti-MXene (0.1 wt.%) DN-based CHGE with a stable and wider ESW of 2.6 V strongly indicates its potential for high-voltage ZIB applications.

The transference number (t_+) is calculated via the Bruce–Vincent method. The calculated t_+ of the PAM-PVA/Ti-MXene (0.1 wt.%) DN-based CHGE is 0.81, which is significantly higher than the reported literature [41]: PNMA/SA DN (0.55), PAM-SN (0.49), and Zn(ClO₄)₂ LE (0.36). The increased t_+ implies that the incorporation of Ti-MXene into the DN matrix significantly enhances Zn²⁺ diffusion pathways, thereby reducing ion transport resistance, as shown in Figure 4b,c. Additionally, Ti-MXene effectively suppresses anion diffusion, leading to cation-dominance in charge transport and a consequent increase in t_+ .

Zn//Zn stripping-plating is a crucial study for assessing the Zn metal reversibility, which is essential for long-life batteries. The PAM-PVA/Ti-MXene (0.1 wt.%) DN-based CHGE exhibits stable cycling for over 1000 h at 0.1 mA cm⁻², without any voltage fluctuations or short circuits, indicating that the Ti-MXene incorporated DN-CHGE effectively stabilizes the Zn//Zn stripping-plating, as shown in Figure 4d. In contrast, the LE undergoes a short circuit at 150 h due to zinc dendrite formation, resulting in poor cyclic stability [18].

Furthermore, the rate capability of PAM-PVA/Ti-MXene (0.1 wt.%) DN-based CHGE was evaluated using a symmetric cell at various current densities, as shown in Figure 4e. The Zn//Zn cell exhibits a stable, persistent voltage plateau over current densities ranging from 0.1 mA cm⁻² to 5.0 mA cm⁻². Even at a high current density of 5 mA cm⁻², the cell maintained reversible Zn stripping/plating behavior with a gradual increase in polarization, without any voltage fluctuation or short-circuit failure.

Importantly, when the current density was returned to 0.1 mA cm^{-2} , the cell recovered its original stable voltage profile, confirming the robust and reversible electrochemical behavior. These results demonstrate that the PAM-PVA/Ti-MXene (0.1 wt.%) DN-CHGE provides stable Zn plating/stripping and maintains excellent interfacial stability even under high current density conditions. These results indicate that the PAM-PVA/Ti-MXene (0.1 wt.%) DN-based CHGE supports faster, more reversible ion migration, as evidenced by both cycling stability and rate capability.

Zn//Cu asymmetric cells were constructed to evaluate the reversibility of the PAM-PVA/Ti-MXene (0.1 wt.%) DN-based CHGE, as shown in Figure 4f. An average coulombic efficiency of 99.4% was achieved over 500 cycles. Specifically, Zn was deposited onto the Cu substrate at a current density of 0.1 mA cm^{-2} with a fixed deposition capacity of $0.022 \text{ mAh cm}^{-2}$, corresponding to a deposition time of 0.5 h for each cycle [42]. After deposition, the Zn was stripped until a cutoff voltage of 1.0 V versus Zn/Zn²⁺ was reached. The Coulombic efficiency was calculated as the ratio of the stripping capacity to the plating capacity for each cycle. Under these conditions, the Zn//Cu cell exhibits a high average Coulombic efficiency of 99.4% over 500 cycles, indicating highly reversible Zn deposition/stripping behavior and excellent interfacial stability provided by the PAM-PVA/Ti-MXene (0.1 wt.%) DN-based CHGE. This enhanced efficiency is attributed to the effective suppression of the hydrogen evolution reaction (HER) and the uniform Zn²⁺ flux, which promote stable Zn deposition and stripping. These results demonstrate that the PAM-PVA/Ti-MXene (0.1 wt.%) DN-based CHGE facilitates uniform zinc deposition, thereby ensuring high cycling reversibility of the Zn anode [31].

To further evaluate its practical applicability, a pouch cell-type ZIB was assembled using PAM-PVA/Ti-MXene (0.1 wt.%) DN-CHGE as the electrolyte, Zn foil as the anode, and V₂O₅ as the cathode. The pouch cell architecture is shown in Figure 5a. The Zn//V₂O₅ pouch cell delivered a high discharge capacity of 178 mAh g^{-1} at 0.1 A g^{-1} and retained nearly the same capacity after three cycles. To examine reaction kinetics, rate performance tests were conducted at current densities of 0.1, 0.2, 0.5, 1.0, 2.0, and 5.0 A g^{-1} . The PAM-PVA/Ti-MXene (0.1 wt.%) DN-CHGE-based cell exhibited reversible capacities of 178, 142, 122, 102, 85, and 68 mAh g^{-1} , respectively. Furthermore, when the current density returned to 0.1 A g^{-1} , the capacity recovered to 165 mAh g^{-1} , demonstrating the excellent redox reversibility of the PAM-PVA/Ti-MXene (0.1 wt.%) DN-CHGE, as shown in Figure 5b,d.

Under these conditions, the Zn//V₂O₅ full cell in this work delivers a high reversible capacity of 178 mAh g^{-1} at 0.1 A g^{-1} , indicating efficient Zn²⁺ transport within the hydrogel electrolyte and favorable electrode/electrolyte interfacial kinetics. Importantly, the 3-D polymeric network of the PAM-PVA/Ti-MXene (0.1 wt.%) DN CHGE provides continuous ion-transport pathways, high electrolyte uptake, and excellent electrode wettability, which are beneficial for maintaining efficient Zn²⁺ diffusion even in thicker electrodes. These structural characteristics suggest that the developed hydrogel electrolyte has strong potential to support electrodes with higher mass loading. In addition, long-cycle performance was evaluated at 0.5 Ag^{-1} for over 500 cycles, as shown in Figure 5e. The Zn//V₂O₅ cell utilizing PAM-PVA/Ti-MXene (0.1 wt.%) DN-CHGE exhibits a high

capacity retention of 84% even after 500 cycles, along with an average Coulombic efficiency of 100%, indicating its excellent cycling stability and minimal side reactions. This superior charge/discharge behavior is attributed to the high ionic conductivity and mechanical robustness of PAM-PVA/Ti-MXene (0.1 wt.%) DN-based CHGE, which facilitates enhanced cyclic stability and outstanding capacity.

The effectiveness of the 0.1 wt.% Ti-MXene DN-CHGE in suppressing dendrite growth and byproduct formation is further demonstrated through post-cycling XRD and FESEM analyses. FESEM images of the Zn anode surface reveal a smooth, dendrite-free morphology without any noticeable protrusions (Figure S3). Subsequently, its corresponding XRD analysis confirms the absence of byproduct formation even after 500 cycles (Figure S4). These outcomes indicate that the PAM-PVA/Ti-MXene (0.1 wt.%) DN-based CHGE effectively inhibits dendrite growth and minimizes side reactions, and enhances the Zn anode stability.

Finally, the practical applicability of FZIBs via PAM-PVA/Ti-MXene (0.1 wt.%) DN-CHGE was evaluated (Figure 5f). The FZIB maintained nearly stable discharge capacity at 1 A g^{-1} over 40 cycles, even under different bending angles (0° , 45° , 90° , and 180°), indicating the exceptional mechanical resilience of the 0.1 wt.% Ti-MXene DN-CHGE. Although a slight capacity drop was observed during bending, the cell subsequently exhibited stable cycling performance, confirming the potential of this CHGE for flexible energy storage applications. Regarding structural integrity, the PAM-PVA/Ti-MXene (0.1 wt.%) DN-CHGE exhibits an interconnected polymeric network with high elasticity and mechanical robustness, enabling it to withstand mechanical deformation without significant cracking or delamination. The relatively stable electrochemical performance observed under different bending angles suggests that the PAM-PVA/Ti-MXene (0.1 wt.%) DN-CHGE maintains continuous ion-transport pathways and stable electrode-electrolyte interfacial contact during deformation. The slight capacity variations observed during bending are therefore more likely associated with temporary changes in interfacial contact resistance or ion-transport pathways induced by mechanical deformation, rather than permanent structural degradation of the hydrogel electrolyte. Furthermore, a comparative table to systematically summarize the mechanical properties and electrochemical performance of the hydrogel electrolyte developed in this work with those reported in recent studies on CHGEs for FZIBs is shown in Table S1. The comparison clearly demonstrates that the PAM-PVA/Ti-MXene DN hydrogel exhibits significantly higher mechanical strength while maintaining competitive electrochemical performance compared with previously reported CHGEs.

Most importantly, in this work, the PAM-PVA/Ti-MXene DN-CHGE was prepared through a facile free-radical polymerization and solution-casting process, which is simple and readily scalable for large-scale production. The fabrication process involves mild reaction conditions and does not require complex equipment or high-temperature processing, making it suitable for practical manufacturing. In addition, the main precursor materials, including PAM, PVA, and ZnSO₄, are commercially available and inexpensive. Although Ti₃C₂T_x MXene is incorporated into the hydrogel matrix, its content is very low (0.1–0.3 wt.%), which minimizes its overall contribution to the material cost. Therefore,

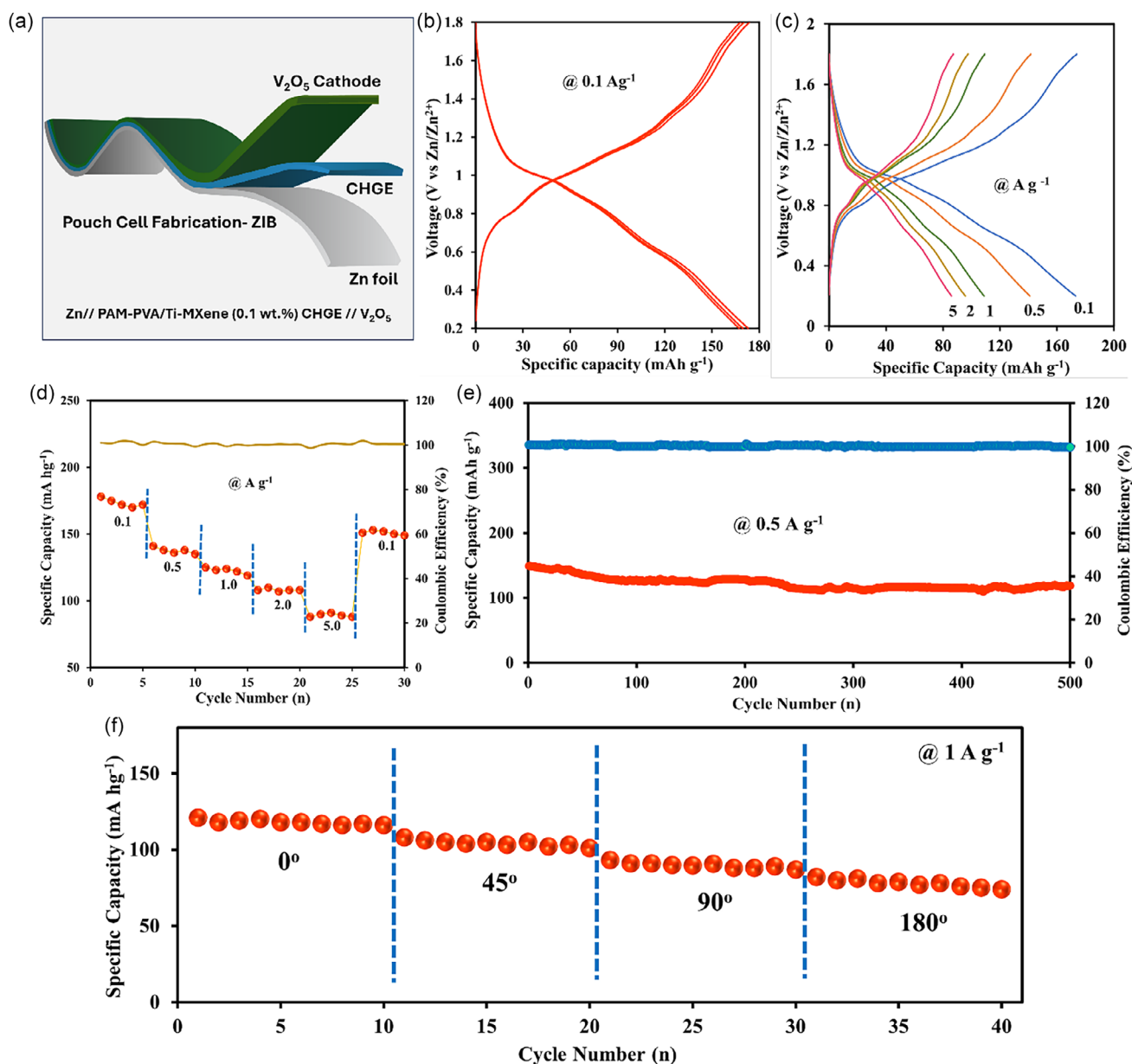


FIGURE 5 | (a) Pouch cell construction of the developed ZIB, (b) GCD plot of the Zn//V₂O₅ for the three cycles, (c) rate performance of the Zn//V₂O₅ at different current densities, (d) rate capability performance of the for 30 cycles at various current densities, (e) long cycling performance of the Zn//V₂O₅ for over 500 cycles and (f) flexibility test at various bending conditions for the fabricated Zn//CHGE//V₂O₅ battery.

the overall fabrication cost remains relatively low. Compared with conventional solid-state electrolytes, which often require high-temperature sintering and sophisticated fabrication processes, the proposed PAM-PVA/Ti-MXene DN-CHGE provides a more cost-effective, scalable, and practical approach for the development of FZIBs.

4 | Conclusion

In conclusion, a Ti-MXene-incorporated PAM-PVA DN-based CHGE was successfully developed via in situ free radical polymerization for high-performance FZIB applications. The synergistic combination of PAM-PVA and Ti-MXene enhances the flexibility of the DN hydrogel without compromising mechanical strength or

ionic conductivity. Among the different Ti-MXene loadings, 0.1 wt% exhibited outstanding mechanical robustness (3.6 MPa) and high ionic conductivity (22.9 mS cm⁻¹). Zn//Zn symmetric cells demonstrated stable cycling for 1000 h at 0.1 mA cm⁻², while Zn//Cu asymmetric cells achieved an excellent coulombic efficiency of 99.4%. Furthermore, the fabricated FZIB exhibited a high initial specific capacity of 178 mAh g⁻¹ at 0.1 A g⁻¹. The combined structural and interfacial engineering strategy significantly contributes to long-term durability, with 84% capacity retention and 100% coulombic efficiency over extended cycling, thereby ensuring the reliable performance of FZIBs. Overall, Ti-MXene-based DN-CHGE presents a promising approach for achieving high ionic conductivity, mechanical strength, dendrite-free Zn anodes, and stable cycling, thereby providing a new pathway for the development of large-scale FZIBs.

Author Contributions

Aakash Carthick Radjendirane: methodology, conceptualization, investigation, formal analysis, data acquisition, writing – original draft; **Narendhar Chandrasekar:** validation, reviewing and editing. **Saradh Prasad Rajendra and Khalid Eidah Alzhirani:** funding acquisition, reviewing and editing. **Ju Hyun Oh:** validation, reviewing and editing. **Seung Jun Lee:** funding acquisition, validation, reviewing and editing. **Subramania Angaiah:** methodology, conceptualization, supervision, validation, resources, writing – reviewing and editing.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

Data will be made available on request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.