



Strain-insensitively stretchable yarn electrodes for self-powered sensing textiles

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ABSTRACT

Self-powered textiles integrated energy storage and sensors have generated growing interest in the area of wearable electronics. However, most current elastic supercapacitors designed for wearable self-powered systems exhibit insufficient strain-insensitivity to accommodate the intricate deformations of the human body. In this work, MXene is being explored for strain-insensitive one-dimensional (1D) energy storage devices by introduction of reduced graphene oxide (rGO) and polypyrrole (PPy) to create a composite yarn electrode via pre-stretching *in-situ* polymerization strategy, which also can be integrated into self-powered wearable sensing textile. The yarn electrodes incorporating rGO effectively minimize crack formation at high strains, showcasing an enhanced capacitance of $51.35 \text{ mF} \cdot \text{cm}^{-1}$ and retaining $\sim 95\%$ of their original capacitance at 200% strain. The resulting symmetric supercapacitor exhibits strain insensitivity up to 200% and delivers stability of electrochemical behavior under real-time dynamic stretching conditions independent of the strain rate. Furthermore, yarn pressure sensing leveraging the resistance change of stripe coating structure achieves high sensitivity of 1.08 kPa^{-1} . Therefore, the integrated self-powered textiles allow convenient use for transmission of Morse code real-time and provide a feasible routine burgeoning transformative telemedicine diagnosis.

KEYWORDS

self-powered textiles, strain-insensitive supercapacitors, yarn sensor

1 Introduction

The accelerated growth of wearable devices has revolutionized the development of intelligent and integrated textile-based energy devices [1–5]. Self-powered textiles with integrated energy storage and sensors have been the focus of research attention for their prospective use in individual medical care, smart homes, and human-machine interaction [5–8]. For energy storage module, in contrast to the drawbacks of low power density and poor cycle life of batteries, the significant advantages of supercapacitors lie in superior power density, faster charging and discharging, and extended lifetime [9]. Generally, a self-powered textiles are supposed to enable precise measurement of signals without disturbances from body movements, which demands supercapacitor with strain-insensitivity to fit the curvature and activities of human body [10]. Currently, the supercapacitors for the self-powered textiles devices are primarily constructed from large-scale fabric structures, including MWCNT/MoO₃ two-dimensional (2D) knitted textile [11] and MXene/Zn three-

dimensional (3D) woven spacer fabric [12]. Although Su et al. [13] developed a one-dimensional (1D) braided thread using carbon nanotubes, polyester and rubber, the work suffers from difficulty in accurate sensing under stretching. Therefore, new materials and structure designs that have both electrochemical and sensing properties are desirable to increase the supercapacitors' strain-insensitivity and sensors' sensitivity of self-powered textiles for practical wearable applications.

2D transition metal carbonitrides/nitrides (MXene) represent a promising category of electrode materials, featuring large bulk capacitance ($1500 \text{ F} \cdot \text{cm}^{-3}$), excellent conductivity ($6500 \text{ S} \cdot \text{cm}^{-1}$), and distinctive multilayered structure, have demonstrated encouraging prospects for designing supercapacitors and sensors [14, 15]. However, MXene electrodes still encounter several significant challenges: (i) dense stacking of 2D MXene nanosheets renders poor ion diffusion kinetics and relatively low energy density; (ii) high elastic modulus (3–75 GPa in dry state) and micrometer scale sheet size are prone to brittleness and hinder

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their strain-insensitive performance. A series of methods have been proposed to construct stretchable MXene supercapacitor electrode materials, such as MXene/carbon black/sodium alginate films (80%-100% tensile strain) [16], but the added insulating materials may impede the reachable surface area of the MXene flakes, reducing the specific capacitance. Other approaches include MOF/MXene composite films (50% tensile strain) [17] and MXene/rGO films (300% tensile strain) [18] electrode materials, but these are mainly in 2D film form, complicating their integration with textiles. Recent advancements have also been achieved in 1D MXene-based stretchable materials, such as MXene/PU fibers designed for gas sensors can accommodate 30% deformation [19], while MXene/carbon cloth fibers in a helicoid structure realized only 20% reversible tensile strain [20], and CV

curves of the MXene/PANi coaxial-helix yarn electrode changed significantly at 100% strain [21]. However, the limited deformation is insufficient to ensure a stable energy supply in wearable applications where significant human movement occurs. Consequently, enhancing both the strain-insensitivity and electrochemical properties of 1D-MXene-based supercapacitor electrodes for self-powered textiles remains a considerable challenge.

Considering the above-mentioned challenges, in this work, a MXene/reduced graphene oxide (rGO)/polypyrrole (PPy) based self-powered pressure sensor with highly strain-insensitive yarn supercapacitor are successfully realized on a pre-stretched polyurethane (PU) yarn substrate by a simple pre-stretching *in-situ* polymerization strategy (Fig. 1(a)). The MXene/rGO

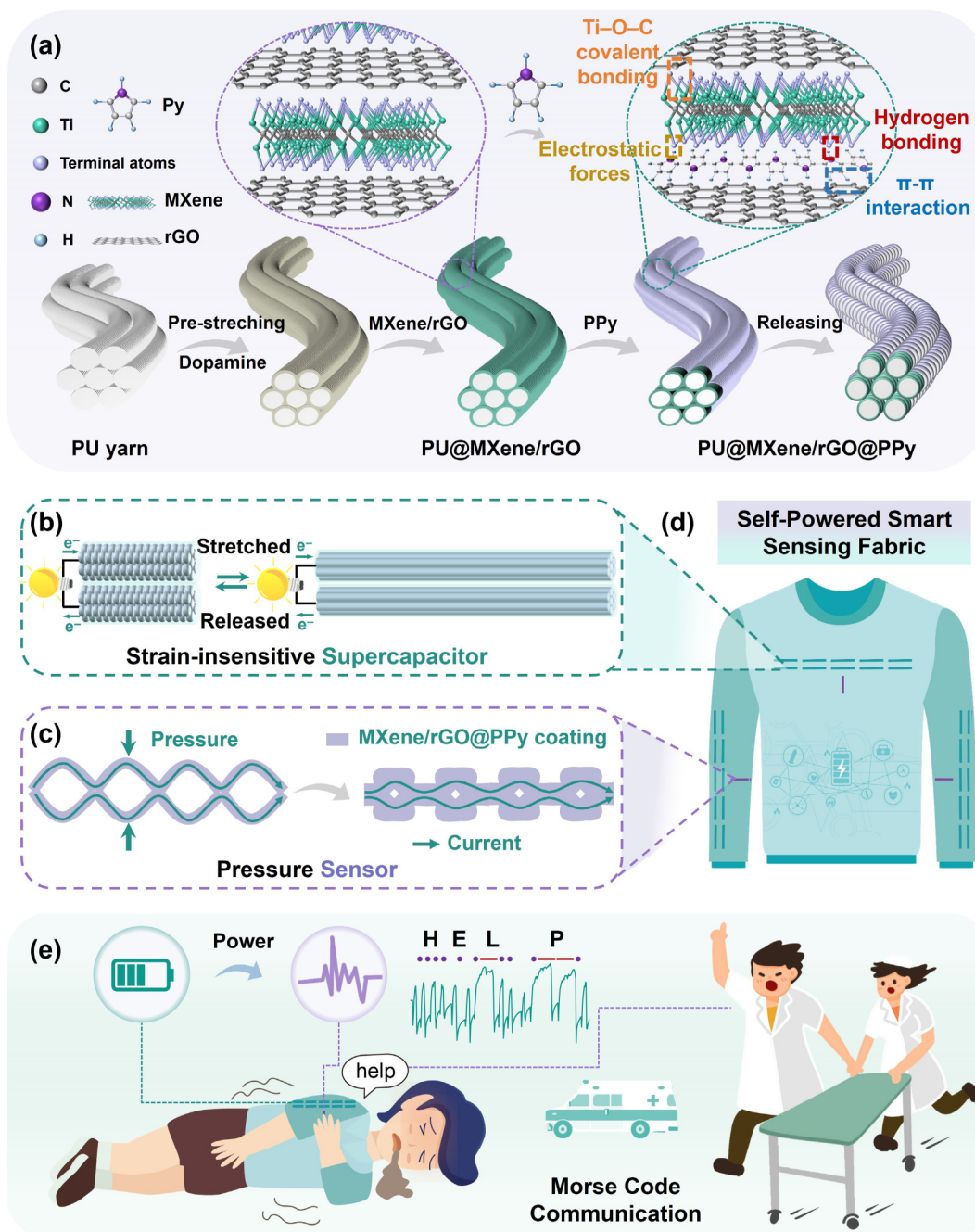


Figure 1 (a) Schematic diagram of the procedure for fabricating strain-insensitive PMGP yarn electrodes. The resulting yarn can be (b) assembled into supercapacitors and (c) utilized as a pressure sensor. (d) Integrated self-powered textile incorporating both energy storage and sensing capabilities. (e) Conceptual illustration of the self-powered sensing textile for remote medicine.

composite coating maintains wrinkled structural integrity and conductivity after releasing the pre-strains of PU, while pure MXene coating fractures into small pieces and develops numerous cracks. Furthermore, the polymerization of PPy intercalate between the MXene/rGO sheets enlarges the interspace among them for faster ion diffusion and creating more conductive paths for charge transport. The obtained wrinkled PU@MXene/rGO@PPy yarn electrodes exhibit excellent strain-insensitivity and high sensing performance, which remain ~95% capacitance retention under 200% tensile strain and show a 1.08 kPa⁻¹ high sensitivity (Fig. 1(b)). In addition, the fabricated fully solid-state strain-insensitive yarn supercapacitor has a length specific capacitance of 50.56 mF·cm⁻¹ (gravimetric capacitance of 146.49 F·g⁻¹, areal capacitance of 101.93 mF·cm⁻²) and exhibits remarkable electrochemical stability even at 200% high static strain or 100% dynamic stretching at high strain rates of 10% s⁻¹ (Fig. 1(c)). It also realizes ~80% capacitance retention after 5000 strain cycles at 50% deformation. Finally, the self-powered pressure sensing textile reliably transmits Morse code in real-time (Figs. 1(d) and 1(e)). This work presents a new approach for linear strain-insensitive electrodes using MXene, providing a scalable framework that can be adapted to various MXene derivatives for next-generation flexible electronics and energy storage. Self-powered textiles capable of signal recognition and communication hold significant potential for applications in remote medicine.

2 Results and discussion

2.1 Fabrication and characterization of the yarn electrodes

To prepare the composite yarn electrodes, we first produced well-dispersed MXene and rGO solution (Fig. 2(a)). The Ti₃AlC₂ precursor presents a tightly packed layered structure with no gaps (Fig. S1(a) in the Electronic Supplementary Material (ESM)). After etching, aluminum (Al) components are removed, resulting in a looser laminar architecture (Fig. S1(b) in the ESM). It also reflects from the inexistence of the (104) lattice in the MXene XRD pattern. The diffraction peak (002) shifts from 9.58° to a lower angle of 6.28° (Fig. 2(c)), indicating an increase in interlayer spaces [22–26]. Ti₃C₂T_x MXene shows ultrathin and transparent 2D lamellar architecture with a lateral size of ~0.5 μm (Fig. 2(b)). The nanoscale dimension of MXene sheets is essential for facilitating ionic transport and ensuring excellent electrochemical performance [18]. The rGO flakes obtained from chemical reduction exhibit a gentle wrinkled shape with a transverse dimension of 1–2 μm (Fig. 2(d)). This wrinkling is attributed to the higher flexibility of rGO flakes relative to the stiffer MXene nanosheets [27], and the larger size of rGO nanosheets leads to stronger intersheet interactions with MXene [18]. Mixing dispersion of the two for further adhesion modification and many small rigid MXene nanosheets are effectively connected by rGO

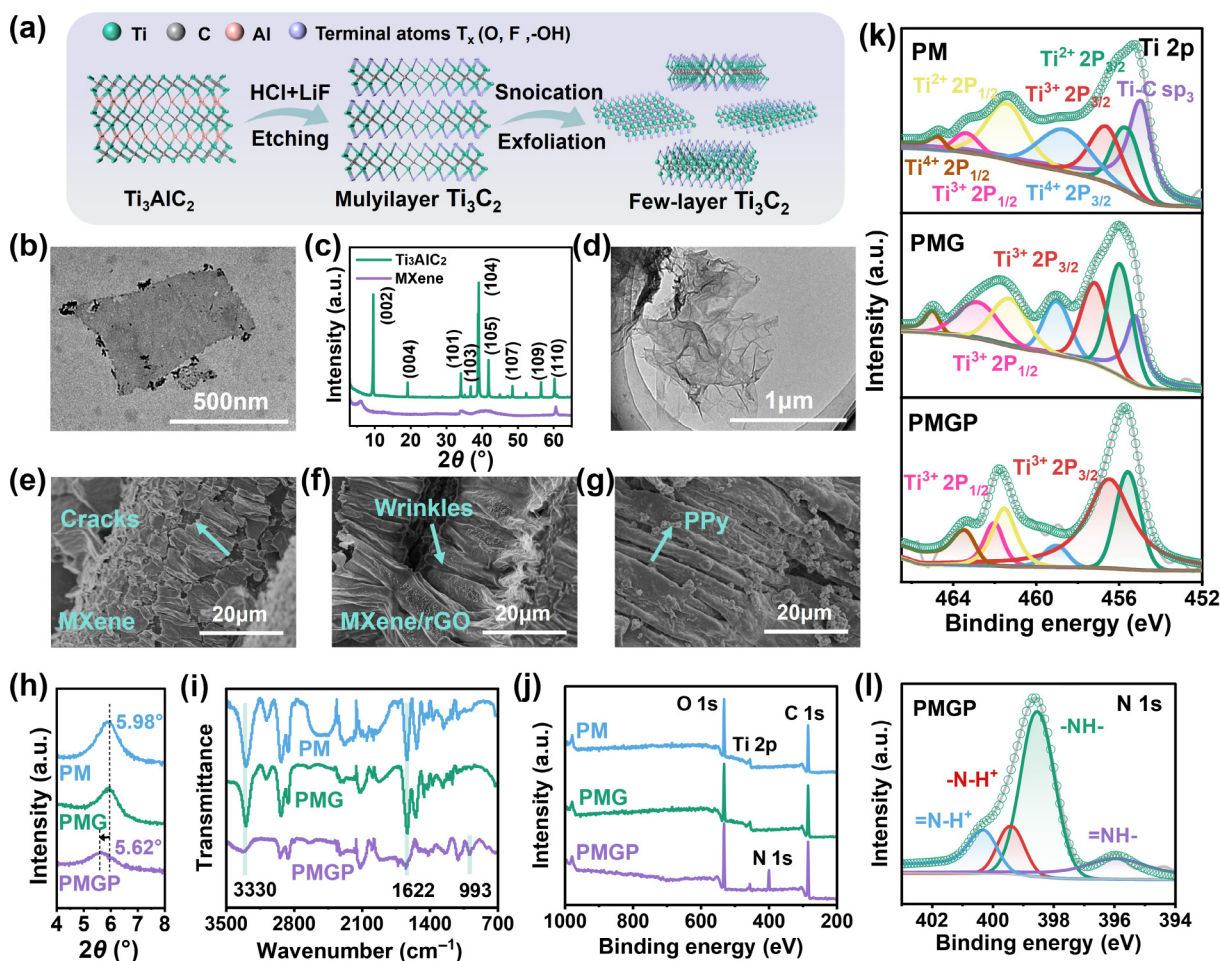


Figure 2 (a) Schematic diagram of the synthesis process for MXene. (b) TEM image of MXene nanosheets. (c) XRD patterns of MXene and MAX precursor. (d) TEM image of rGO sheets. SEM images of (e) PM_{200%}, (f) PMG_{200%} and (g) PMGP_{200%}. (h) XRD patterns, (i) FTIR spectra, (j) XPS full spectra for the PM, PMG, and PMGP. (k) High-resolution spectra of Ti 2p and (l) N 1s for the samples.

nanosheets (Fig. S1(d) in the ESM). Figure 1(a) demonstrates the production process of the PU@MXene/rGO@PPy_x (PMGP_x) composite yarn electrode. Briefly, the pristine PU yarn was stretched under the fabrication strain x and treated firstly with dopamine for adhesion modification, then dipped into the MXene/rGO inks for capillary absorption and finally coated with low-temperature interfacial polymerization PPy. Releasing the pre-strain loaded on the PU yarn to form a wrinkled microlayer.

The scanning electron microscopy (SEM) images of PU@MXene_x (PM_x) prepared by applying different pre-strains reveal that the pure MXene coating electrodes exhibit cracking and fragmentation (Fig. 2(e) and Fig. S2 in the ESM). Cracks observed during relaxation result from the high mechanical modulus and limited flake sizes of MXene films [16, 28, 29]. On the contrary, SEM images for the PMG_x in Fig. 2(f) and Fig. S3 in the ESM show that the yarn maintains its original structure with the wrinkled composite coating arranged in the yarn surface by the same procedures from all different pre-strains, while PMG_{0%} exhibits a smooth surface coating. The ratio of MXene to rGO was optimized, as described in Fig. S2 in the ESM for more details. Unlike MXene, rGO films feature reduced Young's modulus (<1.5 GPa) [30], more robust intersheet interaction, and increased elongation-at-break, enabling them to better endure extreme mechanical stresses during relaxation [18]. Furthermore, PPy was interfacial polymerized on the PMG_x fiber surface to further improve its conductivity and structural stability (Fig. 2(g) and Fig. S4 in the ESM). The PPy nanoparticles result a uniform layer on the PMG_x surface and the density of wrinkles/ridges increases with increasing pre-strains (Fig. S5 in the ESM). For comparison, PU@PPy (PP) (Fig. S6(a) in the ESM) without MXene layer and PU@MXene@PPy (PMP) (Fig. S6(b) in the ESM) without rGO were synthesized by interfacial polymerization of PPy on the PU and PM surface as well. The dense network of PPy strands formed within the inter-yarn spaces plays a critical role in establishing efficient electrolyte ion diffusion channels and providing abundant active sites, ultimately leading to low charge transfer resistance and enhanced capacitance of the yarn electrodes (Fig. S7 in the ESM). The coating treatment has minimal impact for mechanical performance (Fig. S8 in the ESM). The decreasing load observed in PMGP_x under higher pre-strains can be explained by the elongation of yarns and the challenge faced by PPy in penetrating the tightly packed PU yarn under such conditions (Fig. S9 in the ESM).

The chemical composition and intermolecular structures of the composite electrodes were further investigated. The XRD patterns all exhibit a strong (002) peaks, which is typical for Ti₃C₂T_x MXene (Fig. 2(h)). The observed shift of the (002) peak to 5.62° in PMGP, compared to 5.98° in PM, confirming that PPy was successfully intercalated into the interval between Ti₃C₂T_x and rGO sheets [27]. The FTIR spectra of PMGP (Fig. 2(i)) can corroborate the formation of the MXene, rGO and PPy. The PM shows bands at 3330 cm⁻¹ (–OH), 1622 cm⁻¹ (–C=O) that are attributable to Ti₃C₂T_x MXene flakes. PMG also exhibits the same peaks at 3330 and 1622 cm⁻¹, correlating to –OH and –C=O of rGO, respectively. These oxygen-containing groups can form hydrogen bonds between rGO and MXene flakes on yarns, enabling successful binding of MXene and rGO [31]. Furthermore, bands at 993 cm⁻¹ are related to the plane stretching vibration of –C–H in PPy [32]. Moreover, the XPS spectra of PM, PMG and PMGP composite yarns in Fig. 2(j), confirming the existence of C, O, Ti, and N elements, aligning well with the energy dispersive spectroscopy (EDS) mapping results (Fig. S10 in

the ESM). The enhancement in the C–O–C and C–Ti–O_x bonds in C 1s and O 1s spectrums of PMG and PMGP demonstrates the effective interaction among the MXene and rGO species [33], and the C–N peak derived from PPy appeared at 286.7 eV in C 1s spectrums of PMGP (Fig. S11 in the ESM). The relatively high intensity of the Ti³⁺ bond (Ti–O–C) in PMG and PMGP compared to PM further suggests the reaction between MXene layers and rGO sheets (Fig. 2(k)) [31]. For N 1s in Fig. 2(l), the high N*/N ratio of ~32.52% confirms a significant doping level in PPy, which favors the conductivity and rate properties [32].

2.2 Electrical properties of the yarn electrodes

The electrical properties of the fabricated yarns are shown in Fig. 3. The electrical conductivity gradually decreases as x increases, primarily due to the longer wrinkle coating resulting from the pre-stretching process (Fig. 3(a)). Thereinto, the electrical conductivity of the PMGP_x yarn increases owing to the addition of rGO, which can be also seen in the initial resistance of electrical resistant curves in Fig. S12(a) in the ESM. The strain-dependent relative resistance change ($\Delta R/R_0$) is also analyzed in Fig. 3(b). All modified PU yarn electrodes exhibit a monotonic increase in $\Delta R/R_0$ with strain, but the rate of increase is notably slower at higher pre-strain levels. When PMGP_{0%} is stretched to 90%, the resistance increased by 40 times with a fractured coating, whereas the resistance increase for the PMGP_{200%} yarn electrode is only 3.20 times at 300%. This could be attributed to the emergence of microcracks in both the MXene/rGO and PPy coatings when the yarns stretch exceeding the pre-strain value, as shown in the *in-situ* tensile SEM images (Fig. 3(d) and Fig. S13 in the ESM). The SEM movies documenting the stretching process of PMGP_x are also available in Movie S1. Conversely, under the same pre-strain ratio treatment, the resistance change of PMP is more pronounced than PMGP, potentially due to damage to the internal conductive pathways within MXene. Furthermore, the electrical stability of the PMP_{200%} and PMGP_{200%} yarn under 200% strain for 1000 stretching-relaxation cycles is shown in Fig. 3(c). PMGP_{200%} exhibit steady $\Delta R/R_0$ with the robust stretching cycles while PMP_{200%} shows a gradual increase with mechanical cycling, indicating the detachment or disconnection of MXene coating fragments. Therefore, the PMGP yarns are more favorable for strain-insensitive supercapacitor than the pure MXene coating, which shows $\Delta R/R_0$ of less than 0.5 after 40,000 cycles at 20% strain (Fig. S12(b) in the ESM).

The PMGP_{300%} yarn is composed of multiple fibers featuring wrinkled coatings. Perpendicular pressure applied to the fibers compresses the wrinkled coatings, enhancing the contact area between neighboring fibers and lowering overall resistance [32]. This deformation enables PMGP_{300%} to be used in pressure sensors, as shown in Fig. 3(e). The PMGP_{300%} exhibits an outstanding sensitivity of 1.08 kPa⁻¹ within a pressure region of 0 to 12 kPa, as depicted in Fig. 3(f). This higher sensitivity was compared with other sensors [34–41] (Fig. 3(g)). Additionally, the sensor provides rapid response and recovery time of 208 ms (Fig. 3(h)). The great sensitivity and rapid response time of our yarn electrodes is due to the large contact interface area provided by the wrinkle and integral coatings, as well as the superior elasticity of the PU substrate.

2.3 Electrochemical and strain-insensitive performance of the yarn electrodes

The electrochemical properties of the fabricated yarns were

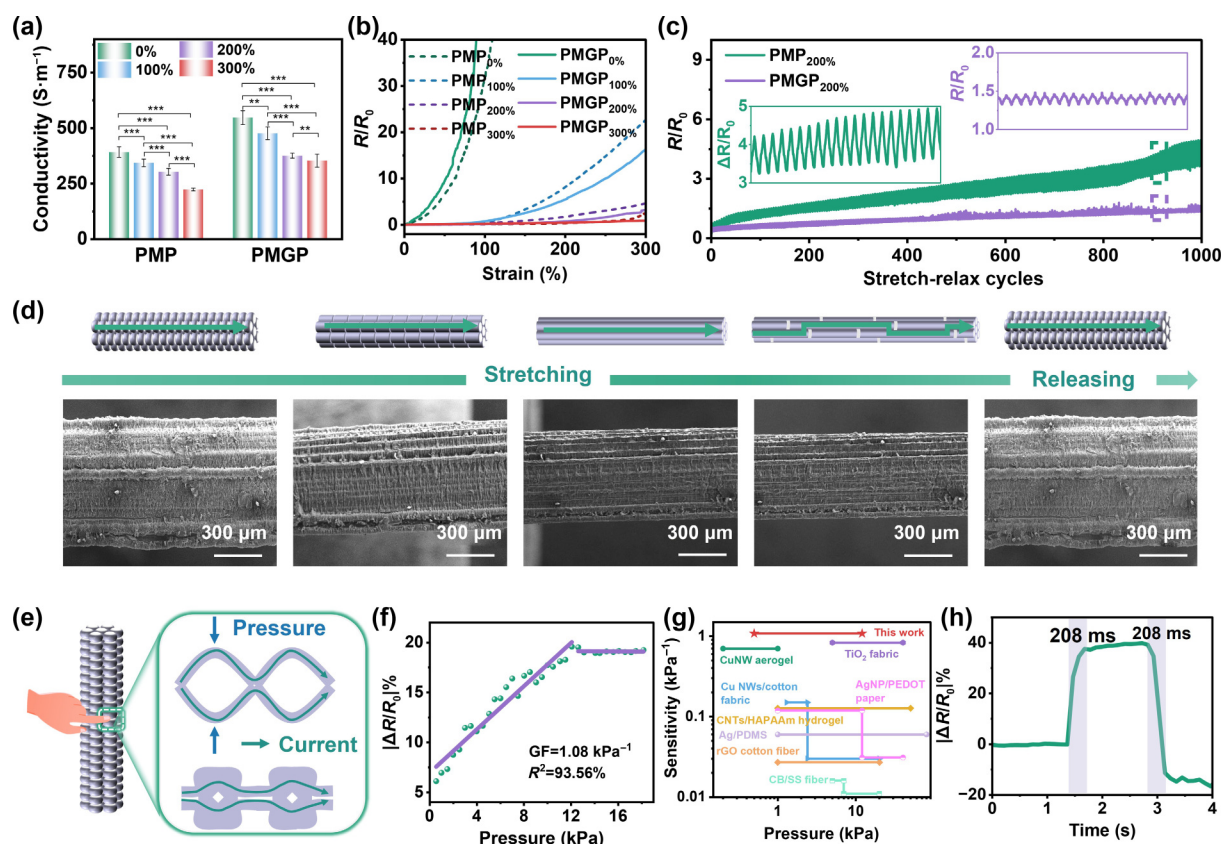


Figure 3 (a) Electrical conductivities of PMP_x and PMGP_x electrodes. (b) Normalized resistance change ($\Delta R/R_0$) plotted during stretching the yarn electrodes. (c) Stretching-releasing cyclic measurement of $\text{PMP}_{200\%}$ and $\text{PMGP}_{200\%}$ at 200% strain. (d) Stretching mechanism diagram and SEM images of the $\text{PMGP}_{200\%}$ during stretching. (e) Pressure mechanism pattern of $\text{PMGP}_{300\%}$ to be as pressure sensors. (f) Relative resistance response, (g) comparison of sensitivity with other pressure sensors, and (h) response time of the pressure sensor.

studied in 1 M H_2SO_4 electrolyte. The performance of PMGP_x electrodes with varying pre-strains was first investigated. The cyclic voltammetry (CV) curves show a decreasing trend in current density from $\text{PMGP}_0\%$ to $\text{PMGP}_{300\%}$ (Figs. S14 and S15(a) in the ESM), which is attributed to the decrease in active load per unit area on PU yarn with increasing pre-strain ratio. The galvanostatic charge-discharge (GCD) curves at a current density of $0.1 \text{ mA}\cdot\text{cm}^{-2}$ confirm that $\text{PMGP}_0\%$ exhibits the highest capacitance (Fig. 4(a)). The electrochemical impedance spectroscopy (EIS) Nyquist plots of PMGP_x show small concave semicircles in the high-frequency area and linear parts in the low-frequency zone. The values of charge transfer resistance (R_{ct}) are 4.89, 12.46, 14.50 and 29.17Ω for $\text{PMGP}_0\%$, $\text{PMGP}_{100\%}$, $\text{PMGP}_{200\%}$, and $\text{PMGP}_{300\%}$, respectively. Besides, the increase in real and imaginary impedance suggests a reduction in ionic accessibility and a decline in yarn conductivity, in agreement with the conductivity test results (Fig. S15(b) in the ESM). The Warburg impedance coefficient (σ), determined from the slope of the low-frequency zone in the EIS plots, increases from 6.45 for $\text{PMGP}_0\%$ to 39.11 for $\text{PMGP}_{300\%}$ (Fig. S15(c) in the ESM). Figure S15(d) in the ESM compares the PMGP_x capacitance derived from GCD results. The capacitance of $\text{PMGP}_0\%$ varied between 74.35 and $51.93 \text{ mF}\cdot\text{cm}^{-1}$, higher than $\text{PMGP}_{100\%}$ (67.82 to $51.35 \text{ mF}\cdot\text{cm}^{-1}$), $\text{PMGP}_{200\%}$ (51.35 to $39.63 \text{ mF}\cdot\text{cm}^{-1}$), and $\text{PMGP}_{300\%}$ (32.63 to $19.16 \text{ mF}\cdot\text{cm}^{-1}$). The electrochemical performance of PMGP_x electrode decreases with the increase of pre-strain ratio, indicating a trade-off between strain-insensitivity and conductivity. The PMGP composite electrode with pre-strain of 200% possesses the best combination of strain-insensitivity and electrochemical

performance, which is selected for further comparing to $\text{PP}_{200\%}$ and $\text{PMP}_{200\%}$.

To assess the contributions of individual components to the electrochemical performance, three modified PU yarn electrodes were tested. Figure 4(b) shows the quasi-rectangular CV curves of all electrodes reflect the low internal resistance and outstanding rate capability. The $\text{PMGP}_{200\%}$ exhibits a much larger current response and longer discharge time in comparison with $\text{PP}_{200\%}$ and $\text{PMP}_{200\%}$ at the same scan rate (Fig. 4(b) and Figs. S16 and S17(a) in the ESM), indicating the contributions of MXene and rGO toward the total capacitance. EIS was conducted to further explore the outstanding performance of the yarn electrodes. The R_{ct} is 29.67, 15.62, and 14.50Ω for the $\text{PP}_{200\%}$, $\text{PMP}_{200\%}$ and $\text{PMGP}_{200\%}$ electrodes, respectively. The R_{ct} of $\text{PMGP}_{200\%}$ is evidently less than $\text{PP}_{200\%}$, indicating that the MXene/rGO layer between the yarn fibers promotes faster electron transfer (Fig. 4(c)). The σ and diffusion coefficient (D) also be calculated to confirm that intercalation of highly conductive MXene and rGO between the PU yarn and the active PPy increased the ion diffusion rate (Fig. S18 in the ESM). Figure 4(d) illustrates the length specific capacitance of various electrode derived from GCD measurements. Notably, the $\text{PMGP}_{200\%}$ delivered a maximal length capacitance of $51.35 \text{ mF}\cdot\text{cm}^{-1}$ at $0.1 \text{ mA}\cdot\text{cm}^{-2}$ and $39.63 \text{ mF}\cdot\text{cm}^{-1}$ is maintained at $0.5 \text{ mA}\cdot\text{cm}^{-2}$. Additionally, this difference of length capacitance for the three electrodes widens with increasing current density. The superior rate performance of $\text{PMGP}_{200\%}$ can be explained by the improved ion diffusion kinetics facilitated by the intercalated structure. The high length capacitance value of $\text{PMGP}_{200\%}$ are comparable to numerous other

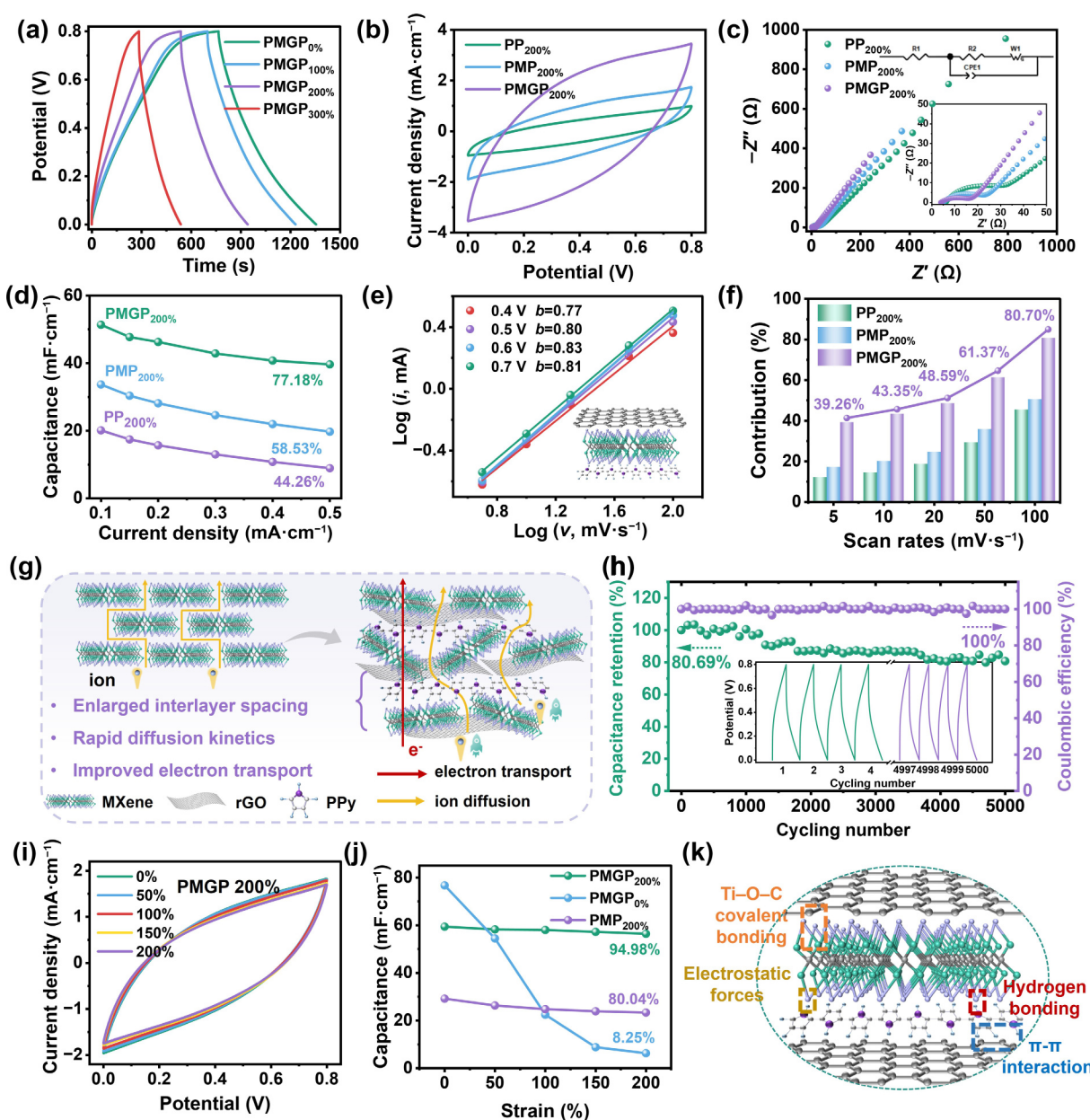


Figure 4 (a) GCD curves of various pre-strain-dependent PMGP_x electrodes at a current density of 0.1 mA·cm⁻¹. (b) CV curves at a scan rate of 100 mV·s⁻¹. (c) EIS plots, and (d) specific capacitances of the PP_{200%}, PMP_{200%} and PMGP_{200%} electrodes. (e) The *b* values of PMGP_{200%} calculated through linear fitting of current response versus scan rate at different voltages. (f) The surface-controlled capacitive contribution as a function of scan rate. (g) Schematic illustration of energy-storage mechanism of PMGP electrodes. (h) Cyclic stability of the PMGP_{200%} yarn electrodes. (i) CV curves of the PMGP_{200%} electrode measured at strains from 0 to 200%. (j) Specific capacitances of PMGP_{200%}, PMGP_{0%}, and PMP_{200%} electrode at strains from 0 to 200%. (k) The proposed strain-insensitive mechanism of the PMGP_{200%}.

reports of MXene-based electrodes, as detailed in Table S1 in the ESM.

To elucidate the underlying reasons, the impact of MXene and rGO sheets on the electrochemical properties of PMGP yarn composites was comprehensively examined using CV method, which follows a power-law relationship between current response and scan rate [42]. The calculated *b* values at various potentials in Fig. 4(e) range from 0.77 to 0.83, suggesting that the charge/discharge process of PMGP_{200%} is predominantly governed by surface-controlled capacitive behavior. The contributions of capacitive-controlled and diffusion-controlled processes to charge storage were analyzed using Dunn's method [43]. PMGP_{200%} demonstrated a higher pseudocapacitive contribution over all scan rates compared to PMP_{200%} and PP_{200%} (Fig. 4(f) and Fig. S19 in the ESM), highlighting the role of MXene and rGO layers in

enabling a unique kinetic pathway for the insertion and extraction of electrolyte ions. The enhanced electrochemical performance of the PMGP_{200%} is due to an integrated layout involving the direct loading of MXene/rGO on the PU yarns followed by the uniform deposition of pseudocapacitive PPy: (i) the unstacked rGO nanosheets increase the electric double-layer capacitance; (ii) the intercalated MXene/rGO provides plentiful active sites for oriented deposition of PPy to maximize utilization of pseudocapacitive; (iii) MXene/rGO nanosheets bridge neighboring highly conductive PPy chains, and PPy chains intercalate between the MXene/rGO layers expanding the space between them simultaneously, resulting to a formation of 3D interconnected conducting channels with enhanced electron transport and a relatively porous structure with improved electrolyte infiltration (Fig. 4(g)). As shown in Fig. 4(h), the

specific capacitance for PMGP_{200%} drops to ~80.69% of its initial capacity and maintain 100% Coulombic efficiency after 5000 GCD cycles. In addition, the capacitance can be maintained after 18 times of washing and 1200 times of folding, demonstrating its excellent electrochemical stability (Fig. S20 in the ESM).

We further investigated the electrochemical behavior of the electrodes at stretch state to demonstrate the contribution of wrinkled structure and rGO to strain-insensitive performance. The CV and GCD curves almost identical under varying stretching strains of PMGP_{200%} due to the architectural integrity of hybrid coating maintained under large deformations (Fig. 4(i) and Fig. S21 in the ESM). Further insights into the strain-insensitive characteristics of the PMGP_{200%} electrode can be obtained from EIS measurements (Fig. S22(a) in the ESM). On the contrary, the PMP_{200%} electrode with pure MXene inlayer shows an increased separation between cracked MXene flakes, leading to decreased conductivity and a gradual loss of electrochemical performance. PMGP_{0%} shows a dramatically decreases electrochemical performance, due to cracks appearing in the initial stretching stage (Fig. S23 in the ESM). Figure 4(j) presents the length specific capacitance of the yarn electrodes derived from the GCD results at the tensile strain from 0%-200%. The MXene/rGO@PPy composite structure improves capacitance and minimizes the decrease in specific capacitance with increasing tensile strain, unlike the MXene/PPy coating, which shows a loss of active material and higher resistance due to cracking of the inner MXene coating. Although the length specific capacitance of PMGP_{0%} reaches 76.69 mF·cm⁻¹ but falls to 6.33 mF·cm⁻¹ representing ~8.25% capacitance retention when a 200% strain is applied, while PMGP_{200%} could maintain 94.98% (56.40 mF·cm⁻¹ at ε =200% from 59.38 mF·cm⁻¹ at ε =0%) (Fig. 4(j)), indicating the whole cracked coating is electrically disconnected during the deformation. Meanwhile, the XPS spectra of C 1s, Ti 2p, and N 1s for PMGP_{200%} yarn before and after 200% strain shows that all peaks for the MXene, rGO, and PPy coating do not present an obvious shift (Fig. S24 in the ESM). The consistent electrochemical performance and excellent structural stability of PMGP_{200%} at different strain states demonstrates its potential as a strain-insensitive supercapacitor electrodes compared with other reported electrodes (Fig. S24(b) and Table S2 in the ESM).

According to the analyses above, the effects of MXene/rGO@PPy coating greatly enhanced strain-insensitive can be summarized as follows: first, MXene sheets can be assembled into a continuous conductive framework through the robust Ti-O-C covalent bonds of the microscale rGO sheets [31]. Furthermore, the flake structure and the hydrophilicity of MXene/rGO sheets enable their tight and evenly attachment to yarn fibers via co-assembly with dopamine-adhered yarn, which form a wrinkle structure and alleviate the tensile stress on the active coating induced by deformation [32]. Moreover, the inserted PPy is forms strong bonds with MXene flakes via hydrogen bonding among the N-H groups of the pyrrole rings and the oxygen-containing terminal groups on the surfaces of MXene [44], as well as electrostatic forces generated by the positively charged PPy and negatively charged MXene nanosheets [45] and a π - π conjugated bond established among the PPy and rGO nanosheets [46]. Finally, the robust interactions between the PU ("rebar") and MXene/rGO ("bricks") and PPy ("mortar") components endows the PMGP_{200%} ("buildings") with excellent strain-insensitive electrochemical performance (Fig. 4(k) and Fig. S24 in the ESM).

2.4 Electrochemical and performance of the yarn supercapacitors

To further assess the practical applicability of yarn electrodes in energy storage, a strain-insensitive supercapacitor with two PMGP_{200%} yarn electrodes and PVA-H₃PO₄ electrolyte between them were fabricated (Fig. 5(a)). The CV curves show quasi-rectangular profiles at scan rates up to 50 mV·s⁻¹ (Fig. 5(b)), while the GCD curves display triangular profiles in Fig. 5(c). The Nyquist plot in Fig. 5(d) reveals an ESR of 256.00 Ω and a R_{ct} of 82.94 Ω . The maximum gravimetric and length capacitances are calculated to be 146.49 F·g⁻¹ and 50.56 mF·cm⁻¹ at a current density of 0.04 A·g⁻¹ and 13.81 μ A·cm⁻¹, respectively (Fig. 5(e)). Additionally, the supercapacitor achieves a maximum areal capacitance of 101.93 mF·cm⁻² (0.03 mA·cm⁻²). The Ragone plot in Fig. 5(f) demonstrates that the supercapacitor delivers an energy density (E) of 13.02 Wh·kg⁻¹ at a power density (P) of 74.05 W·kg⁻¹ and a P of 217.19 W·kg⁻¹ at an E of 7.32 Wh·kg⁻¹. These values are competitive with or superior to other supercapacitors, as summarized in in Table S3 in the ESM. Notably, the supercapacitor maintains 98.1% of its original capacitance after 3000 CV cycles, confirming the remarkable electrochemical stability (Fig. 5(g)). Figures 5(h) and 5(i) and Fig. S25 in the ESM display the normalized capacitance and recorded CV and GCD curves under pristine, bent and twisted deformations, highlighting the excellent mechanical stability of the yarn supercapacitor.

2.5 Strain-insensitive performance of the yarn supercapacitors

The strain-insensitive performance of PMGP yarn supercapacitor was further tested. The CV and GCD curves in Figs. 6(a) and 6(b) show no significant variation at applied strains up to 200%, demonstrating the outstanding strain-insensitivity of the supercapacitor. The inset of Fig. 6(a) shows the yarn supercapacitor under original and different stretching states. The supercapacitor in series can light up a red LED and power the digital display electronic thermometer under a repeated stretching strain (inset of Fig. 6(b) and Movies S2 and S3), indicating its potential application in the strain-insensitive energy storage. The capacitances calculated from GCD curves indicate that the normalized capacitance remained unchanged at 200% strain, which is higher than that reported for most stretchable fiber or yarn supercapacitors (Fig. S26 in the ESM). In addition, Fig. 6(c) shows the CV curves of the PMGP_{200%} tested under dynamic deformation at tensile rates of 5 and 10%·s⁻¹ and a strain of 100%. The CV curves recorded under dynamic strain conditions align closely with the static-state, proving that the supercapacitor possesses good adaptability to various deformations (Movie S4). This strain insensitivity makes it more suitable than conventional stretchable supercapacitors for providing stable power output under body deformation conditions (Fig. 6(d)). Furthermore, the CV (Fig. 6(e)) and GCD curves (Fig. 6(f)) taken after different stretching/releasing cycles, with strain gradually increased from 0% to 50%, show nearly overlapping profiles. The inset of Fig. 6(e) shows the images of the supercapacitor in both its relaxed and 50% stretched conditions. The device retained ~79.55% of its initial capacitance which is exceeded previous reports of stretchable supercapacitors (Fig. 6(f)). The Radar diagram clearly confirms that the PMGP_{200%} yarn supercapacitor has higher capacitance retention in terms of static and dynamic stretching deformation even under more stretching cycles, larger strains and faster stretching rates (Fig. 6(g) and Table S4 in the ESM). The

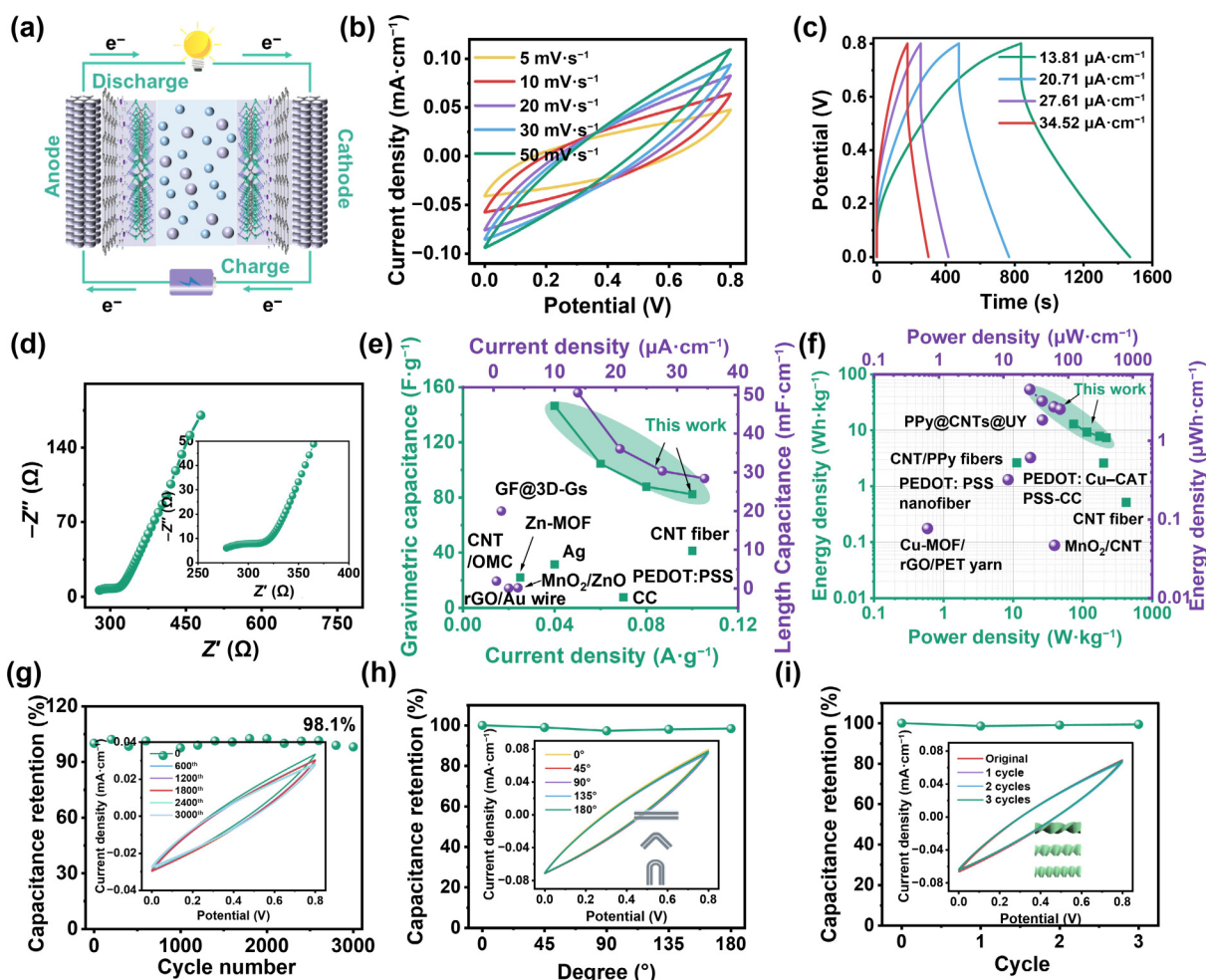


Figure 5 (a) Schematic illustration of the strain-insensitive PMGP_{200%} yarn supercapacitor. (b) CV curves at various scan rates from 5 to 50 mV·s⁻¹. (c) GCD curves at various current densities from 13.81 to 34.52 μA·cm⁻¹. (d) EIS plot, the inset shows the high frequency area. (e) Rate performance and (f) Ragone plots of the device and other reported supercapacitors. (g) Cycling stability of supercapacitor, with corresponding CV curves in the inset. Capacitance retention under different (h) bending and (i) twisting conditions, with corresponding CV curves in the inset.

integrated wrinkles unfold to counteract the impact of macroscopic elongation on the nanolayers and ensure a secure grip on the elastic substrate, retaining the high capacitance under stretching conditions and highlighting the significant promises of the supercapacitor for use in wearable electronics as strain-insensitive energy storage devices (Fig. 6(h)). Meanwhile, benefiting from the gel electrolyte buffering effect and striped coating structure, the CV curves of the supercapacitor nearly overlapped under various pressures (Fig. S27 in the ESM), indicating that the yarn supercapacitor maintained stable electrochemical performance under load bearing.

2.6 Application of the self-powered smart sensing fabric

Integration of energy storage and sensors has significantly advanced the feasibility of multifunctional electronic products [47–49]. 1D yarns, owing to their softness, lightweight nature, and ease of weaving, are particularly well-suited to accommodate the deformations and distortions inherent in wearable textiles [50]. To address the requirements of actual usages, the current and voltage window of the supercapacitor can be enhanced through parallel and series configurations. When two supercapacitors are connected in parallel, the output current and discharge time are doubled relative to an individual supercapacitor, while the voltage window remains unchanged (Figs. 7(a) and 7(b)). Conversely,

connecting two supercapacitors in series increases the voltage from 0.8 to 1.6 V at a constant current density. These results align with fundamental principles of electrical circuits and demonstrate the excellent assemblability and scalability of the devices. The phenomenon of self-discharge is a significant issue in the practical utilization of supercapacitors [51]. Figure 7(c) shows a self-discharge curve of individual supercapacitor. From the 118 s at which charging was stopped, the potential of the supercapacitor drops to 0.38 V within ~1000 s. Figure S28 in the ESM shows the circuit connections for self-discharge profile testing. Meanwhile, the resistance changes of our yarn under pressure make it highly appropriate to be used as pressure sensor. To assess the actual usage in self-powered wearable sensing textiles, 10 supercapacitors connected in five series and two parallel and sensor were sewn onto the fabric and electrically connected via Cu tape, as shown in Fig. 7(d). The supercapacitors are charged with a power supply and the connected sensor is driven by the energy stored in the charged supercapacitors (Fig. S29 in the ESM). Figure 7(e) shows the schematic diagram of potential application scenarios where the self-powered textiles are communicates information by transmitting the English alphabet in Morse code, which can distinguish a range of different words (Movie S5). The decrease in resistance of the sensor at finger pressure can be output by an increased current signal when the supercapacitor is supplied with

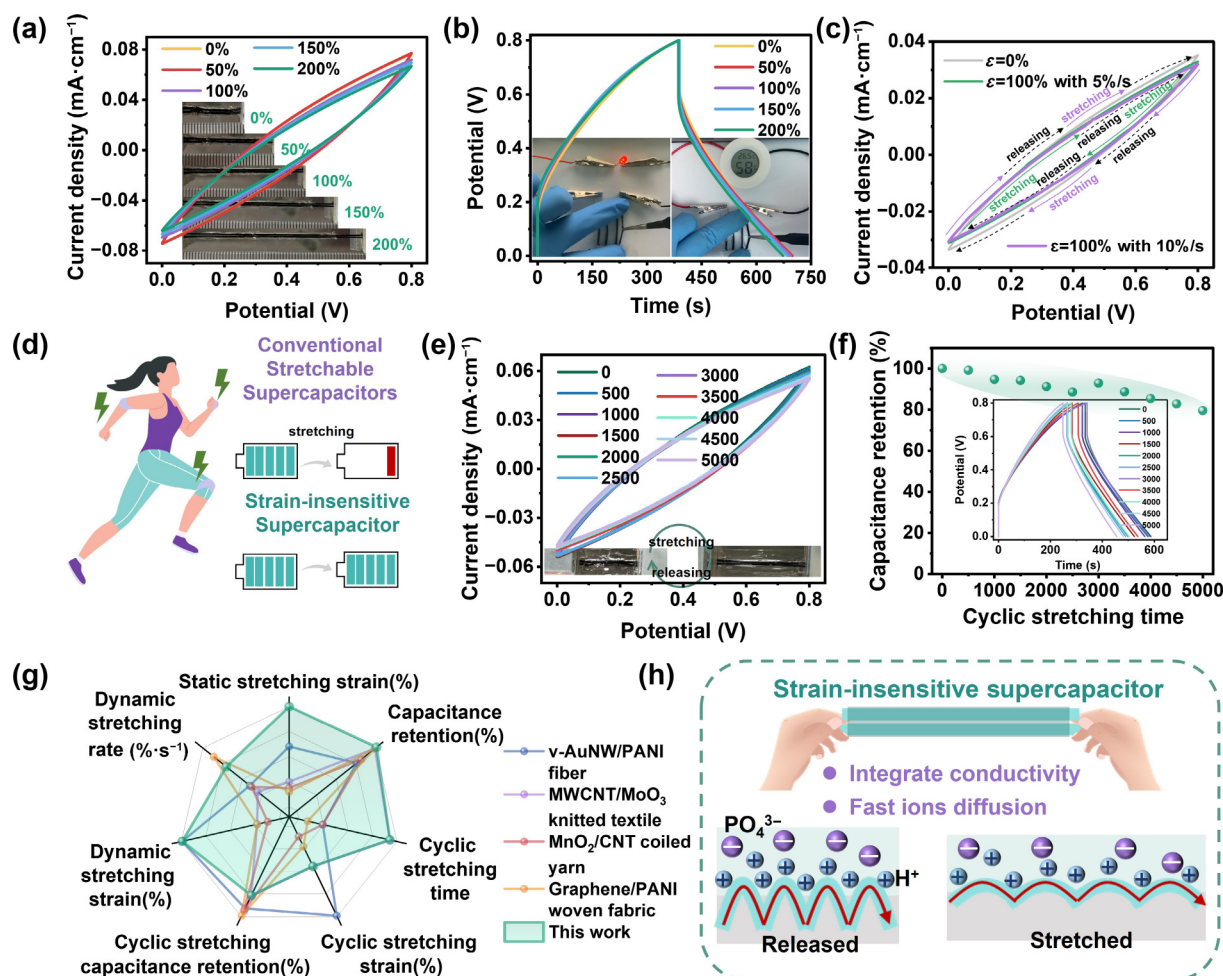


Figure 6 (a) CV and (b) GCD curves under different strain conditions. (c) CV curves measured during dynamic stretching/releasing cycles at 100% strain with different strain rates of 5 and 10 %·s⁻¹. (d) Application advantages of strain-insensitive supercapacitors. (e) CV curves taken after the 5000th stretching by 50% strains, and the image of the stretched supercapacitor. (f) Capacitances retention under repetitive stretching cycles and GCD curves. (g) Radar plots of PMGP yarn supercapacitor compared with other previous reports. (h) Schematic illustration of proposed working principle of strain-insensitive yarn supercapacitor.

voltage. By controlling the time of pressure to combines the “dot” and “dashes” signals to accurately express the messages. As displayed in Figs. 7(f) and 7(g), the words of “HELP” and “DHU” are visualized and recorded via the pressure sensor powered by the supercapacitors. In addition, it can effectively detect a wide range of pressures applied to the skin. When attached to the fingers or leg, the sensor accurately monitors the flexion of the fingers and knee (Fig.S30 in the ESM). Such high-performance finger-pressing sensing technology promises to enable portable health monitoring and telediagnosis.

3 Conclusions

A simple pre-stretching *in-situ* polymerization strategy for fabricating highly strain-insensitive supercapacitor and sensitive pressure sensor based on PU @MXene/rGO@PPy yarn is presented. Notably, this design enables the detection of pressure signal while achieving non-interfering power output of supercapacitor. The composite yarn electrodes retained conductivity and architectural integrity during the fabrication. The measurements show that electrodes have excellent strain-insensitive and high sensing capabilities with a capacitance retention of ~95% under 200% strain coupled with pressure sensitivity of 1.08 kPa⁻¹. The supercapacitor has a specific capacitance of 50.56 mF·cm⁻¹ (146.49 F·g⁻¹ and 101.93 mF·cm⁻²)

and is electrochemically stable under variable stretch tests over 5000 stretching cycles. The excellent strain-insensitive electrochemical performance and high response to pressure endow self-powered textiles with advanced functionalities in single communication. This study demonstrates a practical and promising approach for developing flexible, intelligent wearable electronic warning systems, particularly in the field of telemedicine and remote diagnostics.

4 Experimental section

4.1 Materials

The PU multifilaments (2500D) were provided by Xinhai Textile Co., Ltd. Ti₃AlC₂ MAX phase powders (99%, 400 mesh) obtained from Huawei Ruike Chemical Co., Ltd. Graphene oxide (GO) nanosheets (0.8–1.2 nm thick, 0.5–5 nm in lateral size) purchased from Nanjing Xian-feng Nano Science and Technology Ltd. Pyrrole (Py, ≥98%) and lithium fluoride (LiF) from Shanghai Aladdin Biochemical Technology Co., Ltd. Iron trichloride hexahydrate (FeCl₃·6H₂O), tris(hydroxymethyl)methyl aminomethane (>99%), cyclohexane, hydrochloric acid (HCl, 37%), sulfuric acid (H₂SO₄, 98%), phosphoric acid (H₃PO₄, 99%), ascorbic acid purchased from Sinopharm Chemical Reagent Co., Ltd. Sodium sulfosalicylate (NaSSA) purchased from Rhawn Co.,

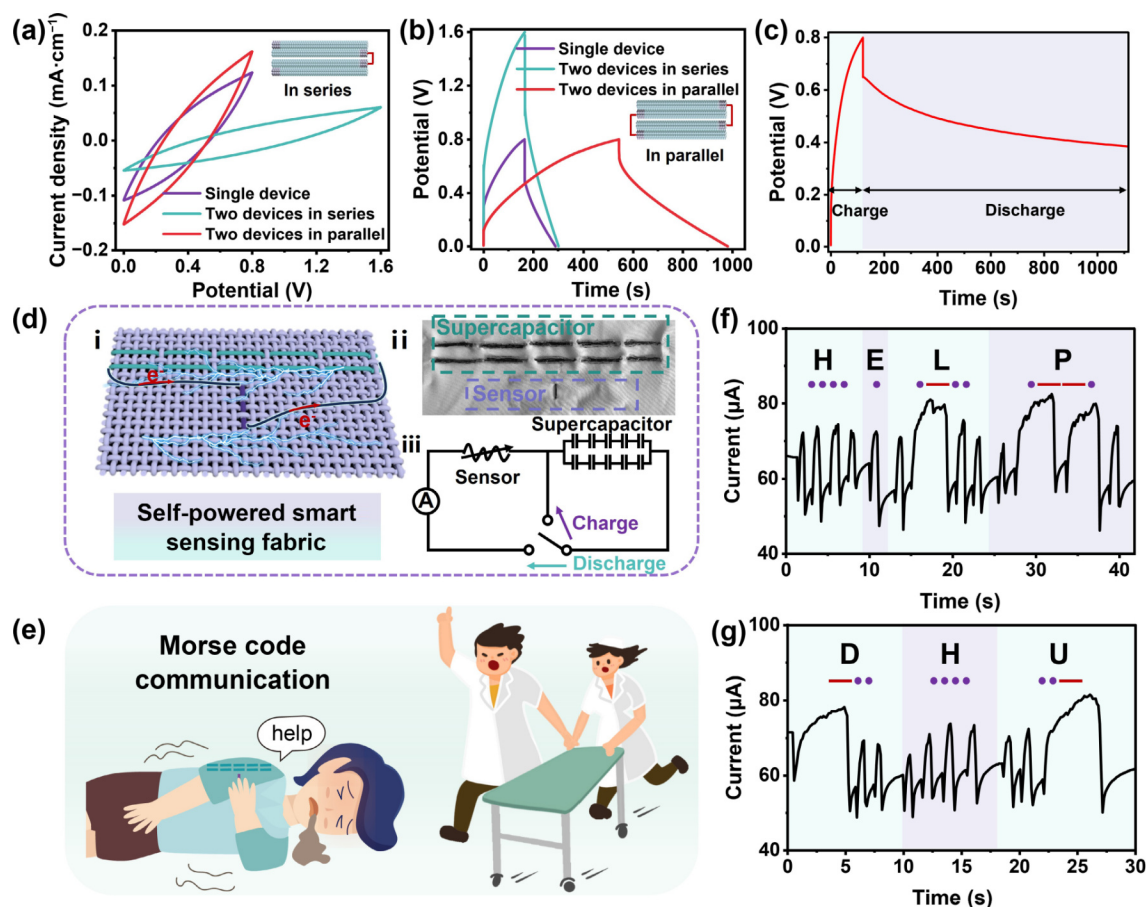


Figure 7 (a) CV and (b) GCD curves of one and two supercapacitors connected in series and parallel. (c) The self-discharge behavior of the individual supercapacitor. (d) (i) Schematic and (ii) optical images of the supercapacitor and sensor with (iii) a circuit diagram of the integrated fabric. (e) Schematic of potential application scenarios where self-powered sensing fabric for single communication. Morse code of (f) "HELP" and (g) "DHU".

Ltd. Dopamine (DA) and polyvinyl alcohol (PVA) was provided by Sigma-Aldrich. The Py monomer was purified through vacuum distillation and stored at 4 °C in the dark.

4.2 Synthesis of $\text{Ti}_3\text{C}_2\text{T}_x$ MXene

The MXene suspension was synthesized by effectively etching the Al atomic layer in the MAX phase, similar to the methods described elsewhere [52]. 1.0 g of Ti_3AlC_2 powder was carefully and slowly dispersed in the impregnant solution (3.2 g of LiF dissolved in 40 mL of 9 M HCl) with continuous mixing at 40 °C for 24 h. The solution was then washed repeatedly with deionized water (DI) and centrifuged at 3500 rpm until the pH was nearly neutral. The resulting precipitate was then redispersed into DI water, ultrasonicated for 2 h under an ice bath, and centrifuged at 3500 rpm for 1 h to yield a well-dispersed MXene dispersion.

4.3 Preparation of MXene/rGO dispersion

100 mg GO powder was dissolved in 20 mL DI water, then ascorbic acid powder was added as reduction agents with the mass ratio with GO of 5:1, the chemical reduction reaction lasts for 3 h at 90 °C to generate rGO powers. The as-prepared MXene dispersion and rGO powder were stirred in DI water and ultrasonicated for 2 h to prepare well-dispersed MXene/rGO mixture with weight ratios of MXene to rGO at 2:1 and 1:1.

4.4 Preparation of PU@PPy electrode

Approximately one meter of stretched PU yarn was immersed in a

DA solution (2 mg·mL⁻¹ in 50 mM Tris-HCl buffer, pH 8.5) and reacted for 8 hours. The yarn was put into dish (90 mm) containing $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (0.36 M) and NaSSA (0.36 M) aqueous solutions (20 mL) and frozen at -20 °C. Subsequently, a mixture of cyclohexane (19.5 mL) and pyrrole (0.5 mL) was poured over the frozen solution. The *in-situ* polymerization was carried out at 2 °C for 24 hours. After a 5-minute ultrasonic cleaning in water, the pre-strain on the PU yarn was released and air-dried at room temperature. The yarns were named as PP_x , where x denotes the pre-strain ratio.

4.5 Preparation of PU@MXene/rGO@PPy electrode

The PDA modified PU yarns were firstly coated with 5 mg·mL⁻¹ $\text{Ti}_3\text{C}_2\text{T}_x$ MXene solution at room temperature for 10 min and dried in a vacuum oven at 50 °C for 1 h to prepare MXene modified PU yarns. This procedure was replicated for 3 rounds to provide extended loading of MXene, then PPy coating was synthesized by the same procedure of PP_x yarn electrode. The obtained yarns were labeled as PMP_x . Similarly, PMGP_x were fabricated just change the MXene dispersion to as-prepared MXene/rGO dispersion.

4.6 Assembling of yarn supercapacitor

PVA- H_3PO_4 electrolyte was prepared by stirring 1 g PVA powders in 10 mL DI water at 90 °C for 2 h, and then 1 g of H_3PO_4 solution was dropped into the solution. A symmetric $\text{PMGP}_{200\%}$ yarn supercapacitor was assembled with two yarns (4 cm long) in

parallel, imbedded in the gel electrolyte and eventually dried at room temperature for 12 h.

4.7 Integration of the self-powered sensing fabric and testing of self-discharge behavior

On the back of the fabric, two Cu tapes were connected to an electrochemical workstation to facilitate charging the supercapacitor. Once the supercapacitors were fully charged, these Cu tapes were disconnected, and another pair of Cu tapes was linked to a source meter device to measure current changes in response to pressure applied to the sensor. As for the self-discharge performance measurement, two Cu tapes were connected to an electrochemical workstation (CHI660E) to facilitate charging the supercapacitor. The Cu tapes was linked to a source meter (Keithley, 2450; Software: Kick start) device to measure voltage changes versus time to obtain a discharge curve after fully charged.

4.8 Characterization

Morphology of the yarn electrodes and EDS were characterized by scanning electron microscopy (SEM, 13 Hitachi SU8010, Tokyo, Japan) at an accelerating voltage of 5 kV and transmission electron microscopy (TEM, JEM-2100F). *In-situ* tensile testing was performed using the Phenom XL equipped with a tensile 18 holder (Phenom XL, Phenom-Scientific, Shanghai, China). Composition characterization and material detection were carried out by Fourier transform infrared spectrometer (FT-NIR, Antaris II, Thermofisher, Waltham, USA), X-ray diffraction (XRD, D/MAX 2550 V B/PC diffractometer, Rigaku Tokyo, Japan) and X-ray photoelectron spectroscopy (XPS, Escalab 250Xi, 5 Thermofisher, Waltham, USA). For the thermostability test, approximately 3 mg of samples were heated from 50 to 800 °C at a heating rate of 20 °C·min⁻¹ under a nitrogen atmosphere using a thermogravimetric analyzer (TGA8000, Thermofisher, Waltham, USA). The mass of the yarn was measured before and after coating using an electronic balance (ME204T/02, Mettler-Toledo International Trading (Shanghai) Co. Ltd). The mechanical properties of the yarn electrodes were investigated using a single fiber strength tester (LLY-06E, Laizhou 15 Electron Instrument Co., Ltd., Laizhou, China) at 1cm length and a strain rate of 50 mm·min⁻¹. The change in electrical resistance and current of the electrodes was measured by a source meter device (Keithley 2450, Tektronix, USA). The simulated washing process using a method similar to the color fastness washing test (GB/T 3921-2008). For simplicity, the rinsing step was replaced by a more severe warm wash step and the draining step, which did little damage to the yarns, was omitted. During a simulated washing test, therefore, the multifilament underwent a warm washing (800 rpm, 40 °C, 20 min) and a drying (60 °C, 15 min). The separation between electrode ends is reduced to 5 mm from the 1 cm planar electrode to simulate folding process. The electrochemical performance of these electrodes was studied in a three-electrode cell in 1 M H₂SO₄ aqueous electrolytes with as-prepared yarn, Ag/AgCl and Pt plate as the working electrode, reference electrode and counter electrode, respectively. All electrochemical measurements were performed on a CHI660E electrochemical workstation. For this characterization the yarns were cut into 1.5 cm length pieces, and partly clamped with a platinum plate electrode (0.5 cm), while the uncovered section (1 cm) was immersed in the electrolyte. The current density of GCD test was 0.1–0.5 mA·cm⁻¹ respectively, the scanning rate of

CV test was 5, 10, 20, 50, 100 mV·s⁻¹. The potential window was 0–0.8 V.

4.9 Calculation methods

(1) The length conductivities were calculated using the following Eq. (1):

$$\delta = \frac{L}{R \cdot S} \quad (1)$$

where L , R and S are the length, resistance and cross-sectional area of yarn electrode.

(2) The active substance mass loading content (mg·cm⁻¹) were calculated using the following Eq. (2):

$$M = \frac{m - m_0}{L} \quad (2)$$

Where L (cm) is the length, m_0 is the initial mass of the yarn and m is mass of the yarn after coating.

(2) The gravimetric, length and areal capacitances were calculated using the following Eqs. (3)–(8).

Gravimetric capacitance:

$$C_M (F \cdot g^{-1}) = \frac{\int I_1 dU}{2vmU} \quad (3)$$

$$C_M (F \cdot g^{-1}) = \frac{I_2 t}{mU} \quad (4)$$

Length capacitance:

$$C_L (F \cdot cm^{-1}) = \frac{\int I_1 dU}{2vLU} \quad (5)$$

$$C_L (F \cdot cm^{-1}) = \frac{I_2 t}{LU} \quad (6)$$

Areal capacitance:

$$C_A (F \cdot cm^{-2}) = \frac{\int I_1 dU}{2vsU} \quad (7)$$

$$C_A (F \cdot cm^{-2}) = \frac{I_2 t}{sU} \quad (8)$$

where I_1 (A) is the response current of CV, U (V) is voltage window (voltage drop IR drop has been considered for galvanostatic charging/discharging), v (V·s⁻¹) is the scan rate, I_2 (A) is the discharging current, t (s) is the discharging time, and m (g), l (cm) and s (cm²) are the weight, length and area of the electrochemically active substance.

(3) The Warburg impedance coefficient (σ) and diffusion coefficient (D) were calculated using the following Eqs. (9) and (10).

$$Z_{re} = R + \sigma \omega^{-1/2} \quad (9)$$

$$D = (R^2 T^2) / (2S n^4 F^4 C^2 \sigma^2) \quad (10)$$

Where Z_{re} is the the real part of impedance, $\omega^{-1/2}$ is the square root of frequency, R is the gas constant, T is the absolute temperature, S is the surface area of the electrode, n is the number of electronic transfers, F is the Faraday constant, and C is the H⁺ concentration.

(4) The CV method was performed to analyze the electrochemical kinetics using the following Eqs. (11) and (12).

$$i = av^b \quad (11)$$

$$\log(i) = b \log(v) + \log(a) \quad (12)$$

Where i (A) represents the current, v ($V \cdot s^{-1}$) represents the sweep rate, and a as well as b are the adjustable coefficients.

(5) The electrochemical kinetics analysis by Dunn's method using the following Eq. (13).

$$i(v) = k_1 v + k_2 v^{1/2} \quad (13)$$

Where i (A) is the current response under a fixed potential, v ($V \cdot s^{-1}$) is the scan rate, k_1 as well as k_2 are constants, where $k_1 v$ represents the surface-controlled contribution, and $k_2 v^{1/2}$ represents the diffusion-controlled contribution.

(6) The energy density (E) and power density (P) of the SCs device can be calculated using the following Eqs. (14) and (15).

$$E = \frac{CU^2}{2 \times 3600} \quad (14)$$

$$P = E \times \frac{3600}{t} \quad (15)$$

Where C ($mF \cdot cm^{-1}$) is capacitance of the whole cell, U (V) is voltage window (voltage drop IR drop has been considered for galvanostatic charging/discharging), t (s) is the discharging time.

4.10 Statistical analysis

Data were analyzed using one-way analysis of variance (ANOVA) with Tukey post-test in IBM SPSS Statistics 26. The significant difference was marked when $p < 0.001$ (***), $p < 0.01$ (**), $p < 0.05$ (*).

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CRedit authorship contribution statement

Shasha Wang: Writing—original draft, Methodology, Formal analysis, Conceptualization. **Leqian Wei:** Methodology, Investigation. **Fengkai Zhou:** Validation, Methodology. **Yimeng Li:** Validation, Methodology. **Jianhua Zhu:** Methodology. **Qian Zhang:** Methodology. **Lizhen Lan:** Visualization. **Zeyu Wang:** Visualization. **Fujun Wang:** Resources. **Lu Wang:** Supervision. **Jifu Mao:** Writing—review & editing, Supervision, Project administration, Funding acquisition.

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 (additional SEM images, structural characterization, mechanical and electrical results; additional electrochemical data, digital photographs and tables) is available in the online version of this article at <https://doi.org/10.26599/NRE.2025.9120184>.

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