



Achieving monolithic electrode integration via chemically unified elastomeric current collectors for corrosion immune, high rate lithium-ion batteries

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ABSTRACT

Fast charging lithium-ion batteries are increasingly limited by interfacial mechanical failure between electrodes and current collectors, which leads to contact degradation, impedance growth, and accelerated performance decay under repeated electrochemical cycling. This work reports a chemically continuous and mechanically adaptive current collector architecture that addresses this overlooked bottleneck through the integration of polymethyl methacrylate-grafted natural rubber (MG30) and multi-walled carbon nanotubes (MWCNTs). The polar polymethyl methacrylate (PMMA) segments promote induced dipole interactions with the nanotube network, enabling the formation of a homogeneous conductive framework that combines high electrical conductivity with mechanical compliance. More importantly, the use of MG30 as both the current-collector matrix and electrode binder establishes a chemically continuous interface across the electrode-collector junction, resulting in enhanced adhesion, reduced interfacial resistance, and improved tolerance to cycling-induced stress accumulation. Unlike conventional aluminum current collectors, the elastomeric architecture exhibits intrinsic resistance to fluoride-induced corrosion while maintaining structural integrity during long-term operation. Consequently, lithium iron phosphate (LiFePO₄, LFP)||Li cells deliver stable high-rate performance and exceptional cycling durability, sustaining 4000 cycles at 10 C and retaining 80.3% capacity after 2000 cycles. The architecture further demonstrates compatibility with high mass loading electrodes and flexible pouch-cell configurations.

KEYWORDS

current collector, chemically unified interface, MWCNTs, corrosion resistant electrodes, high rate lithium-ion batteries

1 Introduction

Lithium-ion batteries (LIBs) have enabled transformative advances in modern energy storage due to their high energy density, long cycle life, and mature manufacturing infrastructure. As the technology evolves toward high-voltage cathodes, high areal capacity, and mechanically adaptive systems, performance bottlenecks increasingly originate from interfacial instabilities rather than intrinsic limitations of active materials [1–3]. In particular, the electrode/current collector interface has emerged as a critical yet underexplored factor governing charge transport, structural integrity, and long-term durability [4, 5]. While the electrochemical principles of LIBs are well established, the growing disparity in mechanical modulus, chemical reactivity, and structural continuity among electrode components introduces interfacial heterogeneities that accelerate impedance growth and capacity decay under practical conditions [6].

The cathode current collector, typically aluminum foil, plays a dual role as both an electronic conductor and a mechanical support [7–9]. Despite its widespread use, aluminum exhibits intrinsic limitations under aggressive electrochemical environments. Its native oxide layer (Al₂O₃), though passivating under mild conditions, becomes unstable in the presence of electrolyte decomposition products such as HF and fluorinated anions derived from LiPF₆, leading to localized corrosion, increased interfacial resistance, and eventual electrical failure [10–12]. Moreover, the smooth and chemically inert surface of Al foil results in weak adhesion with composite cathodes. The resulting mismatch creates a distinct heterogeneous boundary between the active layer and the metal substrate—an interface that is particularly susceptible to stress accumulation, interfacial debonding, and contact loss under repeated high-rate cycling, eventually driving increased electronic resistance and accelerated

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capacity decay [10, 13]. Surface modification strategies—including etching, carbon coating, and porous structuring [14–16] have been explored to enhance interfacial contact, yet they do not fundamentally resolve corrosion susceptibility or the mechanical mismatch between rigid metallic collectors and viscoelastic electrode composites [17, 18].

Carbon-based current collectors, such as carbon nanotube (CNT) films and graphene frameworks, offer attractive alternatives due to their low density, high surface area, and excellent electrochemical stability [19, 20]. These materials facilitate improved interfacial contact and reduced charge-transfer resistance through their porous and rough architectures [21]. However, their practical implementation is hindered by insufficient mechanical integrity, poor film-forming capability, and limited cohesion within thick electrodes, especially under repeated cycling or mechanical deformation [22]. These challenges highlight a fundamental limitation in current electrode design paradigms: the lack of chemical and mechanical continuity across the current collector, binder, and active material domains. Recent advances in elastomeric functional materials suggest a promising pathway to address this issue. Functionalized rubber matrices, particularly those incorporating polar side chains [23–25], have demonstrated strong interfacial adhesion, tunable mechanical compliance, and enhanced ion/electron transport when used as binders [26] or solid electrolytes [27]. In this context, polymethyl methacrylate (PMMA)-grafted natural rubber (MG30) represents a unique platform that integrates a deformable backbone with polar functional groups, enabling synergistic electro-chemo-mechanical performance.

Inspired by these insights, we propose a chemically unified electrode architecture in which the same elastomer—MG30 is employed across both the current collector and the cathode binder, thereby eliminating interfacial discontinuities and establishing intrinsic compatibility (Fig. 1). A rubber-based conductive current collector is constructed by integrating MG30 with multi-walled carbon nanotubes (MWCNTs), where the polar PMMA side chains modulate CNT dispersion through interfacial interactions with the π -electron system, suppressing aggregation and enabling the formation of a robust percolated conductive network. More importantly, the shared chemical identity between the collector and binder creates strong interfacial affinity, significantly enhancing adhesion and reducing contact resistance. This integrated design simultaneously addresses the long-standing

challenges of aluminum corrosion, weak electrode–collector adhesion, and structural instability. As a result, the assembled LFP||Li cells exhibit exceptional rate capability and long-term cycling stability, even under high mass loading and practical configurations. This work establishes a new paradigm for battery electrode design, shifting from conventional multi-component assembly toward elastomeric, chemically coherent systems, and provides a scalable strategy for next-generation high-performance and mechanically adaptive energy storage devices.

2 Experimental

2.1 Materials

Polymethyl methacrylate-grafted natural rubber (MG30, PMMA grafting ratio: 30 wt.%) was supplied by Jiangsu Qixiang Advanced Materials Co., Ltd. LFP and polyvinylidene fluoride (PVDF) were obtained from Canrud (Dongguan) New Energy Technology Co., Ltd. Multi-walled carbon nanotubes (MWCNTs, diameter: 10–15 nm, length $\sim 10\ \mu\text{m}$) were purchased from Shenzhen Zhongke Nano Technology Co., Ltd. Conductive carbon black (Super P) was supplied by Shenzhen Kejingzhida Technology Co., Ltd. Tetrahydrofuran (THF, chromatographic grade, 99.9%) and N-methyl-2-pyrrolidone (NMP, electronic grade, 99.9%) were purchased from Shanghai Macklin Biochemical Technology Co., Ltd. The liquid electrolyte (1 M LiPF_6 in ethylene carbonate (EC)/diethyl carbonate (DEC), EC:DEC = 1:1 by volume, with 5 wt.% fluoroethylene carbonate (FEC)) was obtained from Suzhou Duoduo Chemical Technology Co., Ltd. In addition, CR2025 coin cell components, lithium metal foil, spacers, and spring sheets were purchased from Canrud (Dongguan) New Energy Technology Co., Ltd.

2.2 Preparation of elastomeric current collector

MG30 was first dissolved/swollen in THF under magnetic stirring for 12 h at room temperature. Subsequently, MWCNTs were added to the solution, followed by continuous stirring for 72 h. To promote homogeneous dispersion, the mixture was ultrasonicated for 20 min every 24 h during the stirring process. The resulting slurry was cast onto a polytetrafluoroethylene (PTFE) substrate using a four-sided applicator (gap: 800 μm), followed by drying at 60 $^\circ\text{C}$ for 5 h to remove solvent. The dried film was then hot-pressed at 60 $^\circ\text{C}$ for 20 min to improve film densification and

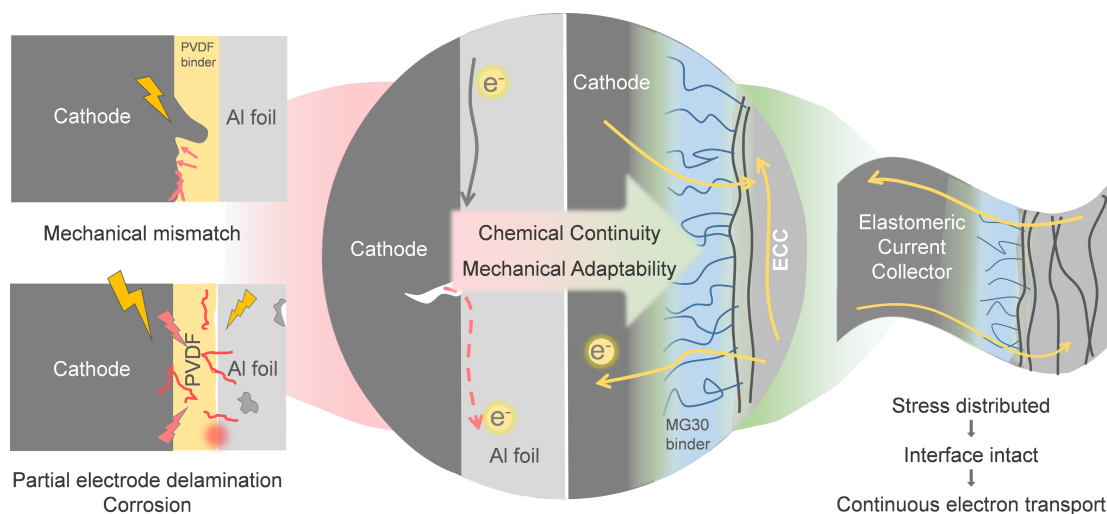


Figure 1 Conceptual design of a chemically continuous and mechanically adaptive current collection architecture for fast charging lithium ion batteries.

interfacial contact between MG30 and MWCNTs. Finally, a freestanding elastomeric current collector (denoted as MC) was obtained by peeling from the PTFE substrate. A series of current collectors with different MG30/MWCNTs weight ratios (4:4, 4:5, 4:6, 4:7, and 4:8) were prepared and labeled as MC44, MC45, MC46, MC47, and MC48, respectively. The resulting membranes exhibited a uniform thickness of $\sim 32 \mu\text{m}$, as verified by digital micrometer measurements (Fig. S1 in the Electronic Supplementary Material (ESM)).

2.3 Cathode fabrication

Slurry-coated cathode: LFP, Super P, and MG30 binder were mixed in a mass ratio of 8:1:1 in THF to form a homogeneous slurry. The slurry was coated onto the prepared current collector using a four-sided applicator (gap: $100 \mu\text{m}$), followed by drying to obtain electrodes with an active material loading of ~ 1.5 – $2.5 \text{ mg}\cdot\text{cm}^{-2}$. For comparison, control electrodes were prepared using PVDF as the binder (dissolved in NMP) under identical conditions.

Freestanding cathode: LFP, a hybrid conductive agent (MWCNTs:Super P = 7:3 by weight), and MG30 binder were mixed in a mass ratio of 8:1:1 in THF. The slurry was cast onto a PTFE substrate (gap: $800 \mu\text{m}$) and dried to obtain a freestanding electrode film with high active material loading (~ 13 – $15 \text{ mg}\cdot\text{cm}^{-2}$).

2.4 Cell assembly

CR2025-type coin cells were assembled in an argon-filled glovebox with oxygen and moisture levels below 0.01 ppm. The cells consisted of a LFP cathode (diameter: 12 mm), lithium metal anode (diameter: 16 mm), and a polyethylene separator (diameter: 16 mm).

2.5 Characterization

Fourier-transform infrared (FTIR) spectra were recorded on a Nicolet iS50 spectrometer (ATR mode, 4000 – 500 cm^{-1} , resolution: 4 cm^{-1}). Raman spectra were collected using a Zolix Finder 930 spectrometer with a 532 nm laser. X-ray diffraction (XRD) measurements were conducted on a Rigaku Miniflex 600 using $\text{Cu K}\alpha$ radiation ($\lambda = 1.5418 \text{ \AA}$) over a 2θ range of 10° – 70° . Surface morphology was characterized using scanning electron microscopy (SEM, Gemini 500, Zeiss) and atomic force microscopy (AFM). SEM samples were transferred under an inert atmosphere to minimize air exposure. AFM was performed in tapping mode (Asylum Research MFP-3D-S). In addition, three-dimensional surface profiles were obtained using a confocal optical profiler (SENSOFA S neox). Contact angle measurements of electrode slurry on MC and Al substrates were conducted using a goniometer (OCA-20, DataPhysics). X-ray photoelectron spectroscopy (XPS) measurements were performed using the NAP-XPS EnviroESCA instrument with an $\text{Al K}\alpha$ source. XPS data fitting was conducted using Avantage software. Electrostatic potential distribution of MG30 was calculated using Gaussian 16. All-atom molecular dynamics (MD) simulations were performed using LAMMPS to examine the interfacial interactions between MG30 and MWCNTs. An atomistic model consisting of a MWCNTs embedded in an NR-g-PMMA matrix was constructed in BIOVIA Materials Studio 2024 and parameterized using the Polymer Consistent Force Field (PCFF). After energy minimization and equilibration, production simulations were carried out in the NPT ensemble at 550 K and 1 atm for 8.0 ns . Radial distribution functions (RDFs), normalized polymer-

MWCNT contact ratios, and interfacial interaction energies were calculated from the production trajectories to qualitatively evaluate the preferential interactions between different polymer segments and the MWCNT surface.

Mechanical properties were evaluated using a universal testing machine (HZ-1007C). Tensile tests were conducted on rectangular specimens ($10 \text{ mm}\times 80 \text{ mm}$) at a strain rate of $20 \text{ mm}\cdot\text{min}^{-1}$. Peel strength was measured using a 180° peel configuration. Cathode strips ($10 \text{ mm}\times 80 \text{ mm}$) were adhered to the current collector, with both ends fixed using adhesive tape. The peel test was conducted at a constant rate of $20 \text{ mm}\cdot\text{min}^{-1}$. Each measurement was repeated at least three times.

Sheet resistance (ρ_s) was measured using a four-point probe system (RTS-8). Through-plane resistance was determined using a digital multimeter in a customized configuration. The resistivity (ρ) was calculated using $\rho = R\cdot A/L$, where R is resistance, A is cross-sectional area, and L is thickness. The electronic conductivity (σ) was calculated as $\sigma = 1/\rho$.

Electrochemical impedance spectroscopy (EIS) was performed over a frequency range from 0.1 Hz to 10^6 Hz with an amplitude of 10 mV . Galvanostatic charge-discharge tests were conducted using a LAND-CT3002A system within a voltage window of 2.5 – 4.0 V (vs. Li^+/Li). A specific capacity of $170 \text{ mAh}\cdot\text{g}^{-1}$ was defined as 1 C .

For cyclic voltammetry (CV) measurements, cells were assembled using the MC47 current collector as the cathode and Li foil as the anode. The scan rate was set to $50 \text{ mV}\cdot\text{s}^{-1}$ over a voltage range of 2.5 – 4.0 V , and the scanning was performed in the positive direction.

3 Results and discussion

3.1 Molecular design of the chemically unified elastomeric conductive network

The design of the elastomeric current collector (ECC) centers on the construction of a synergistically reinforced network, integrating the high-performance MG30 elastomer with multi-walled carbon nanotubes (MWCNTs) to achieve a balance of mechanical compliance and robust electronic conduction [26, 27]. Among the investigated compositions (MC44–MC48), MC47 is identified as the optimum formulation (optimized MWCNTs loading, with its morphological evolution across different contents detailed in Fig. S2 in the ESM). The composite exhibits a compact and continuous morphology (Fig. 2(a)), where MWCNTs are uniformly dispersed within the MG30 matrix without noticeable aggregation. This homogeneous dispersion contrasts sharply with typical CNT bundling behavior. Such interfacial affinity between polymer and carbon nanotubes is essential for forming an effective percolated conductive network at relatively high filler loadings. The structural integration is further confirmed by XRD (Fig. 2(b)), where the amorphous halo of MG30 ($\sim 19^\circ$) coexists with the (002) diffraction peak of MWCNTs ($\sim 26^\circ$). The reduced intensity and slight broadening of the CNT peak suggest disruption of graphitic stacking, consistent with improved dispersion induced by surface functionalization [28, 29]. Compared to pristine CNT systems, this behavior indicates that MWCNTs are not merely physically embedded but are molecularly coupled with the polymer matrix, leading to a more homogeneous composite structure.

To clarify the origin of this enhanced compatibility, the electronic structure of MG30 was analyzed. The electrostatic potential map (Fig. 2(c)) reveals that the ester groups of PMMA

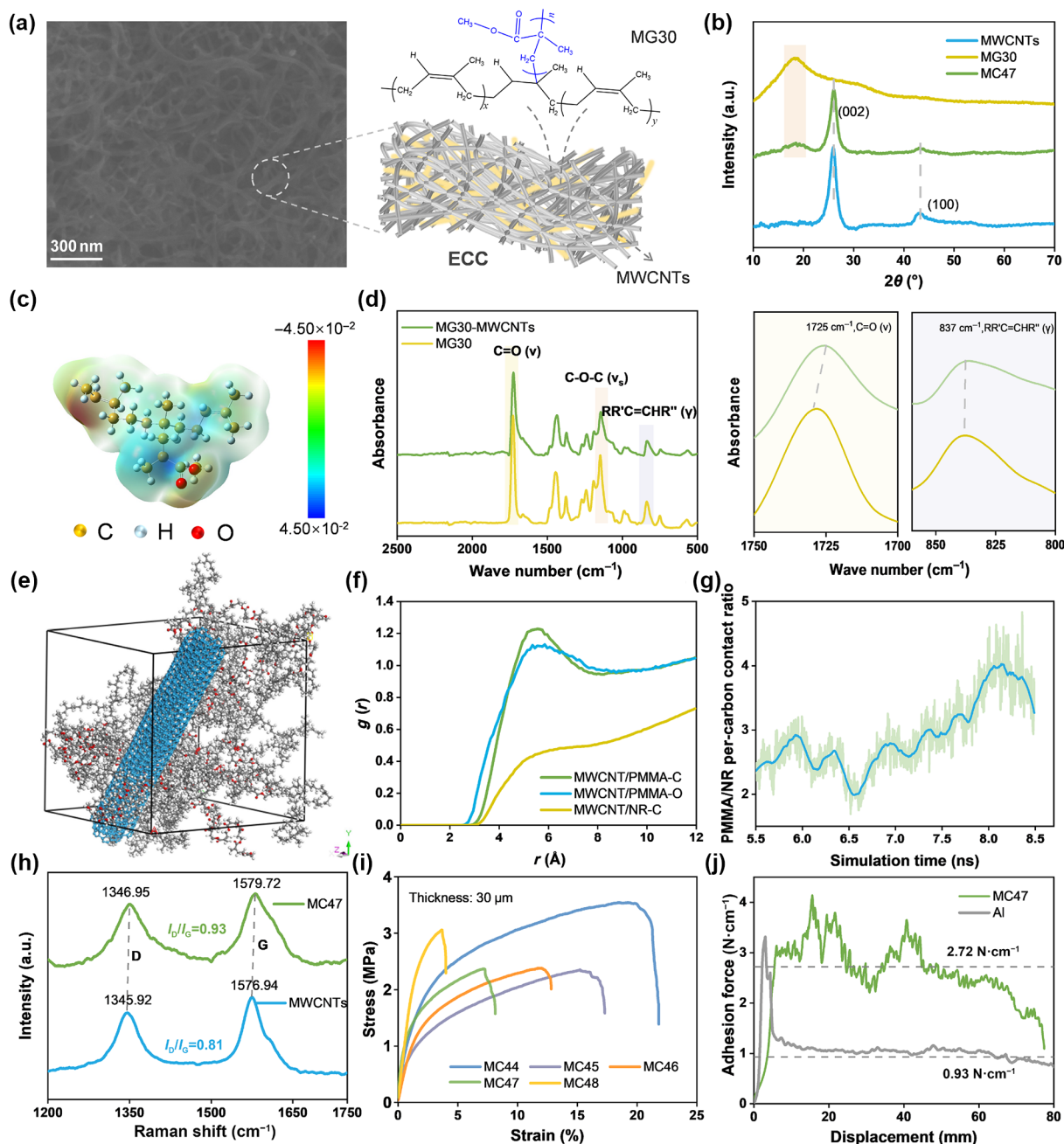


Figure 2 (a) Molecular structure of polymethyl methacrylate-grafted natural rubber (MG30) and surface SEM image of the elastomeric current collector (MCx), showing homogeneous dispersion of MWCNTs within the rubber matrix. (b) XRD patterns of MG30, MWCNTs, and MC47 composites. (c) Electrostatic potential map of MG30 monomer unit. (d) FTIR spectra of MG30 and MG30-MWCNTs hybrid. (e) Diagrammatic figure of molecular dynamics (MD) simulation of the interaction between MG30 and multi-walled carbon nanotubes. (f) Radial distribution functions (RDF) between the MWCNT and NR or PMMA. (g) Segment carbon number normalized contact ratio of PMMA/NR with MWCNT within 5 Å. (h) Raman spectra of MWCNTs and MC47. (i) Tensile stress-strain curves of MC with varying compositions. (j) 180° peel strength comparison between MC and Al foil.

side chains and unsaturated C=C bonds along the rubber backbone provide electron-rich regions. These sites are predicted to dominate the interaction with the sp^2 -hybridized carbon network of the MWCNTs [30]. This hypothesis is supported by FTIR spectra (Fig. 2(d) and Fig. S3 in the ESM), where the C=O ($\sim 1726 \text{ cm}^{-1}$) stretching vibration of MG30 shifts to lower wavenumbers upon composite formation, accompanied by subtle broadening of the $>\text{C}=\text{CH}-$ stretching region ($\sim 835 \text{ cm}^{-1}$) [31]. This shift indicates a slight decrease in the C=O bond order and an alteration in its electron density distribution. Specifically, it suggests a partial overlap between the lone pair (n) orbital of the carbonyl oxygen and the π -electron cloud of the MWCNTs

surface. The highly polar ester group locally polarizes the π electrons of MWCNTs, simultaneously promoting strong physical adsorption and partially preventing agglomeration by π - π stacking between nanotubes, which is well documented in CNT/polymer systems [28]. Meanwhile, the subtle spectral shift (C=CH-) suggests that the primary mechanism of NR phase-CNT bonding is physical and weak, akin to conventional carbon-filled rubber composites, relying on physical adsorption and mechanical interlocking [32, 33].

To further understand the molecular origin of the interfacial compatibility, all-atom molecular dynamics (MD) simulations were performed for the MG30/MWCNT composite (Fig. 2(e))

[34, 35]. The simulations suggest that the PMMA grafts exhibit a higher tendency to approach the MWCNT surface than the NR backbone, as indicated by the radial distribution functions and the evolution of the normalized polymer–MWCNT contact ratio (Figs. 2(f) and 2(g)). Consistently, the calculated polymer–MWCNT interaction energies indicate a stronger attractive interaction between the PMMA segments and the MWCNT surface than that of the NR segments (Fig. S4 in the ESM). Although the simulated interface continues to undergo gradual structural relaxation within the accessible simulation timescale, these results qualitatively support that the polar PMMA grafts preferentially associate with the graphitic surface of MWCNTs, whereas the NR chains mainly provide a flexible surrounding matrix. Such molecular-scale interfacial organization is consistent with the experimentally observed homogeneous dispersion of MWCNTs and contributes to stabilizing the conductive network against aggregation.

In addition to the permanently induced dipoles discussed above, dispersive interactions arising from transient polarization further contribute to the interfacial affinity between MG30 and MWCNTs. Raman spectra (Fig. 2(h) and Fig. S5 in the ESM) show that the characteristic D and G bands of MWCNTs are well preserved after incorporation into the MG30 matrix, exhibiting only slight shifts in peak position (from 1345.92 and 1576.94 cm^{-1} for pristine MWCNTs to 1346.95 and 1579.92 cm^{-1} for MC47) [36]. These subtle changes suggest only minor perturbations of the local chemical environment while confirming that the graphitic framework remains structurally intact. Meanwhile, the moderately increased I_D/I_G ratio is consistent with enhanced interfacial interactions between MG30 and MWCNTs rather than significant structural modification. Together, these physical interactions suppress MWCNT aggregation and stabilize the conductive network under mechanical deformation [28].

As a result, the composite exhibits a unique combination of elasticity and mechanical robustness. As shown in Fig. S6 in the ESM, the MCx films can be bent and folded repeatedly without permanent deformation. The presence of MG30 enhances the ductility of the current collector itself. As its content increases, the elongation at break of the cathode sheet incorporating the current collector increases. Tensile measurements (Fig. 2(i)) show that increasing MG30 content enhances ductility, with elongation at break reaching ~22% at the MC44 composition while maintaining tensile strength above 3 MPa [37]. This performance originates from the synergistic effect of the elastomeric matrix and the well-dispersed CNT network, where strong interfacial interactions enable efficient stress transfer—a hallmark of CNT-reinforced elastomers [38].

More importantly, the elastomeric collector delivers markedly improved interfacial adhesion with the cathode. As shown in Fig. 2(j), the peel strength reaches 2.72 $\text{N}\cdot\text{cm}^{-1}$, approximately three times that of Al foil (0.93 $\text{N}\cdot\text{cm}^{-1}$). Failure analysis (Fig. S7 in the ESM) indicates cohesive failure within the electrode for MC, rather than interfacial delamination observed for Al foil. This superior adhesion can be attributed to two key factors: (i) chemical coupling enabled by MWCNTs and polar MG30 chains, and (ii) mechanical interpenetration of the elastomeric network into the electrode layer. These features establish a conformal and robust interface, which is fundamentally inaccessible to conventional metallic current collectors.

3.2 Eliminating interfacial discontinuities via elastomeric co-diffusion

To fully exploit the potential of the ECC, we implemented a 'monolithic' design strategy by utilizing the same MG30 elastomer as both the current collector matrix and the cathode binder. This chemical unification is specifically engineered to eliminate the thermodynamic and mechanical incompatibility typically found at the current collector/electrode junction. In standard battery architectures, the mismatch between the metallic Al foil and the organic binder-rich cathode leads to heterogeneous charge-transfer pathways and localized mechanical stress concentrations [9]. In contrast, the ECC/cathode interface introduced here is chemically coherent and mechanically adaptive. By bridging the collector and the electrode with a shared polymer phase, we facilitate molecular inter-diffusion across the boundary, fundamentally altering the interfacial behavior from a discrete junction to a continuous, integrated electronic framework.

Cross-sectional SEM analysis (Fig. 3(a)) reveals a clear gap between the Al foil and the cathode layer, indicative of incomplete wetting and weak adhesion. In contrast, the MC-based electrode exhibits seamless contact without discernible voids, confirming intimate interfacial integration [39]. This difference is directly linked to both the hierarchical surface structure and the chemical functionality of MCx. From a structural perspective, SEM (Fig. S8 in the ESM) and 3D profilometry (Fig. 3(b) and Fig. S9 in the ESM) show that MCx possesses pronounced surface undulations (~10 μm height variation), significantly larger than those of Al foil (~2 μm). At the nanoscale, AFM measurements (Fig. 3(c) and Fig. S10 in the ESM) reveal a roughness (R_q) up to 141 nm for MC47, compared to only 2.37 nm for Al foil. This hierarchical roughness enhances the effective contact area and facilitates mechanical interlocking with the cathode layer, consistent with classical adhesion theory [40].

The unique surface structure of MC was further corroborated by wettability tests, which directly reflect its interfacial affinity with the cathode materials. Using diluted cathode slurry as the test liquid, droplets were deposited on both Al foil and MC. The contact angles were measured after the droplets reached equilibrium. The results demonstrate a striking difference: the slurry rapidly spread and wetted the MC surface, while it remained as a discrete droplet with a contact angle of 19.2° on the Al foil (Fig. 3(d)). This superior wettability of MC is attributed to its micro/nano-rough structure and favorable surface chemistry, which promote intimate contact with the cathode slurry, thereby facilitating a more uniform coating and stronger interfacial adhesion. The wetting behavior can be analyzed using the Young's equation:

$$\gamma_{LV} \cos \theta = \gamma_{SV} - \gamma_{SL} \quad (1)$$

For a given slurry (constant γ_{LV}), a smaller contact angle (θ) indicates either an increase in the solid surface energy (γ_{SV}) or a decrease in the solid-liquid interfacial tension (γ_{SL}). In the system, the micro/nano rough surface of MC enhances solid-liquid affinity, thereby reducing γ_{SL} . Concurrently, the presence of polar functional groups in the collector likely increases γ_{SV} [29, 41]. Both factors contribute to the observed superior wettability. To decouple the effects of the current collector's surface chemistry from those of the binder, a control experiment was performed using a slurry with commercial PVDF binder instead of MG30. On the same rubber collector, this PVDF-based slurry formed a droplet with a contact angle of 12.7° (Fig. S11 in the ESM), which is significantly higher than that of the MG30-based slurry. This result underscores that specific chemical interactions between the

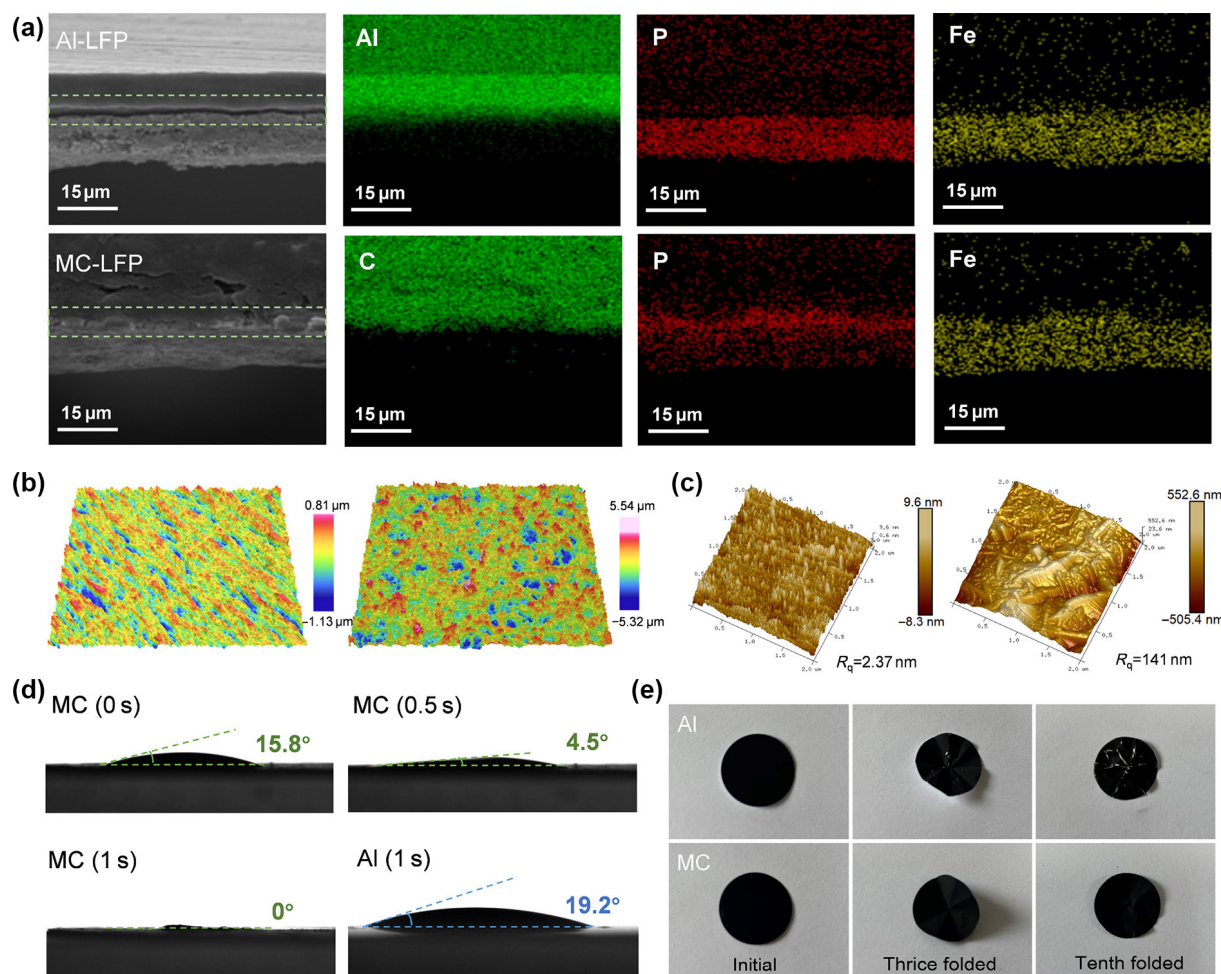


Figure 3 (a) Cross-sectional SEM images and corresponding EDS elemental mapping of cathodes on MC47 and Al foil. (b) 3D optical profilometry images showing surface topography: Al (left) and MC47 (right). (c) AFM topography images and roughness analysis (R_q): Al (left) and MC47 (right). (d) Contact angle measurements of cathode slurry on MC47 and Al foil. (e) Optical images of electrodes after repeated bending deformation.

collector and the binder–active materials in the cathode also play a critical role in determining ultimate interfacial contact. Therefore, the superior interfacial adhesion of MC over Al foil can be attributed to three synergistic factors. First, its textured morphology with optimal roughness provides a larger effective surface area and enhances mechanical interlocking with the cathode layer. Second, the polar ester groups in MG30 establish strong interactions (e.g., charge-induced and van der Waals forces) with the cathode materials. Third, the inherent chemical affinity between the collector and cathode components such as between MWCNTs and Super P, and between MG30 in the collector and MG30 serving as the cathode binder, creates a "like-dissolves" compatibility effect, further promoting interfacial integration [42]. Collectively, these features ensure robust bonding, which is critical for maintaining structural integrity during cycling.

The practical viability of this monolithic architecture was validated under extreme mechanical and environmental stress tests. As demonstrated in Fig. 3(e), cathode sheets based on the ECC design could be subjected to repeated folding—fully recovering their original geometry without crack propagation or active material loss. Conversely, the Al foil-based counterparts exhibited immediate structural failure; the emergence of metallic luster from surface scratches indicated severe coating detachment and irreversible damage to the electronic network. Furthermore, under high-power ultrasonication (Fig. S12 in the ESM), the ECC-

based cathode remained flawlessly intact, whereas the coating on the Al foil completely delaminated. This exceptional resilience confirms that the chemically coherent interface provides a level of robust bonding and structural integrity that is essential for flexible energy storage and high-power applications.

3.3 Inherent chemical immunity against fluoride-induced electrolyte corrosion

Current collector corrosion represents a catastrophic failure mode in high-rate lithium-ion batteries, as structural degradation simultaneously weakens active material adhesion and dismantles the electronic percolation network [10, 11]. To evaluate the electrochemical–chemical resilience of our design, current collectors were subjected to accelerated degradation in a simulated corrosive environment (dilute acid/electrolyte mixture under applied potential). As illustrated in Fig. 4(a), commercial Al foil undergoes severe pitting corrosion after 36 hours, contrasting sharply with its initial smooth morphology. This degradation stems from the chemical breakdown of the native Al_2O_3 passivation layer. In LiPF_6 -based electrolytes, trace moisture triggers the formation of hydrofluoric acid (HF), which converts the oxide layer into AlF_3 via: $\text{Al}_2\text{O}_3 + 6\text{HF} = 2\text{AlF}_3 + 3\text{H}_2\text{O}$ [43–45]. While AlF_3 provides transient protection, its susceptibility to localized fracture under sustained high-voltage or high-temperature operation initiates irreversible pitting. Furthermore,

in LiTFSI-based systems, the formation of soluble $\text{Al}(\text{TFSI})_3$ complexes leads to continuous foil dissolution [12]. Control experiments in salt-free acidic solutions (Fig. S13 in the ESM) revealed significantly fewer pits, confirming that lithium salts act as potent accelerators for metallic corrosion. In striking contrast, the ECC current collector remained entirely intact under identical conditions (Fig. 4(b)), demonstrating an inherent chemical immunity to both acidic and electrolyte-mediated attack.

Post-cycling morphological analysis provides definitive evidence of the ECC's superior stability. After 2000 cycles at 10 C, Al foil collectors exhibit extensive pitting and surface cratering (Fig. 4(c)). Conversely, the ECC not only maintains its structural integrity but undergoes a beneficial 'architectural consolidation', where initial surface imperfections are suppressed to form a denser, more cohesive framework (Fig. 4(d)). Correspondingly, cross-sectional SEM and energy dispersive spectroscopy (EDS) analyses after 2000 cycles (Fig. S14 in the ESM) reveal an intact and continuous interface between the LFP cathode and the MC47 current collector without discernible gaps or delamination, demonstrating sustained interfacial conformity throughout prolonged cycling.

The failure mechanism of Al foil is a synergistic three-pathway process (Fig. 4(h)): (i) Al^{3+} out-diffusion creates interfacial voids

that destabilize the passive film; (ii) F^- in-diffusion-induced volume expansion generates tensile stress and subsequent crack propagation; and (iii) locally concentrated HF species directly compromise the passivation layer [44]. In contrast, the ECC circumvents these degradation pathways through its unique composition. Composed entirely of chemically stable MG30 and MWCNTs, the collector is inherently resistant to fluoride-induced corrosion and maintains its structural integrity under battery operating conditions. Electrolyte-soaking tests reveal negligible thickness variation and essentially unchanged electrical conductivity after drying, indicating minimal irreversible solvent uptake despite good electrolyte wettability (Fig. S15 in the ESM). In addition, cyclic voltammetry of MC47||Li cells exhibits stable and reproducible responses without additional redox features within the operating voltage window of the LFP cathode (Fig. 4(e)), while Raman spectra collected before and after electrolyte exposure and prolonged cycling preserve the characteristic D and G bands of MWCNTs without the appearance of degradation-related features (Fig. 4(f)) [28]. These results collectively confirm the excellent chemical robustness and dimensional stability of the elastomeric current collector. In addition, the intrinsic elasticity of the MG30 matrix accommodates the chemo-mechanical stresses arising from

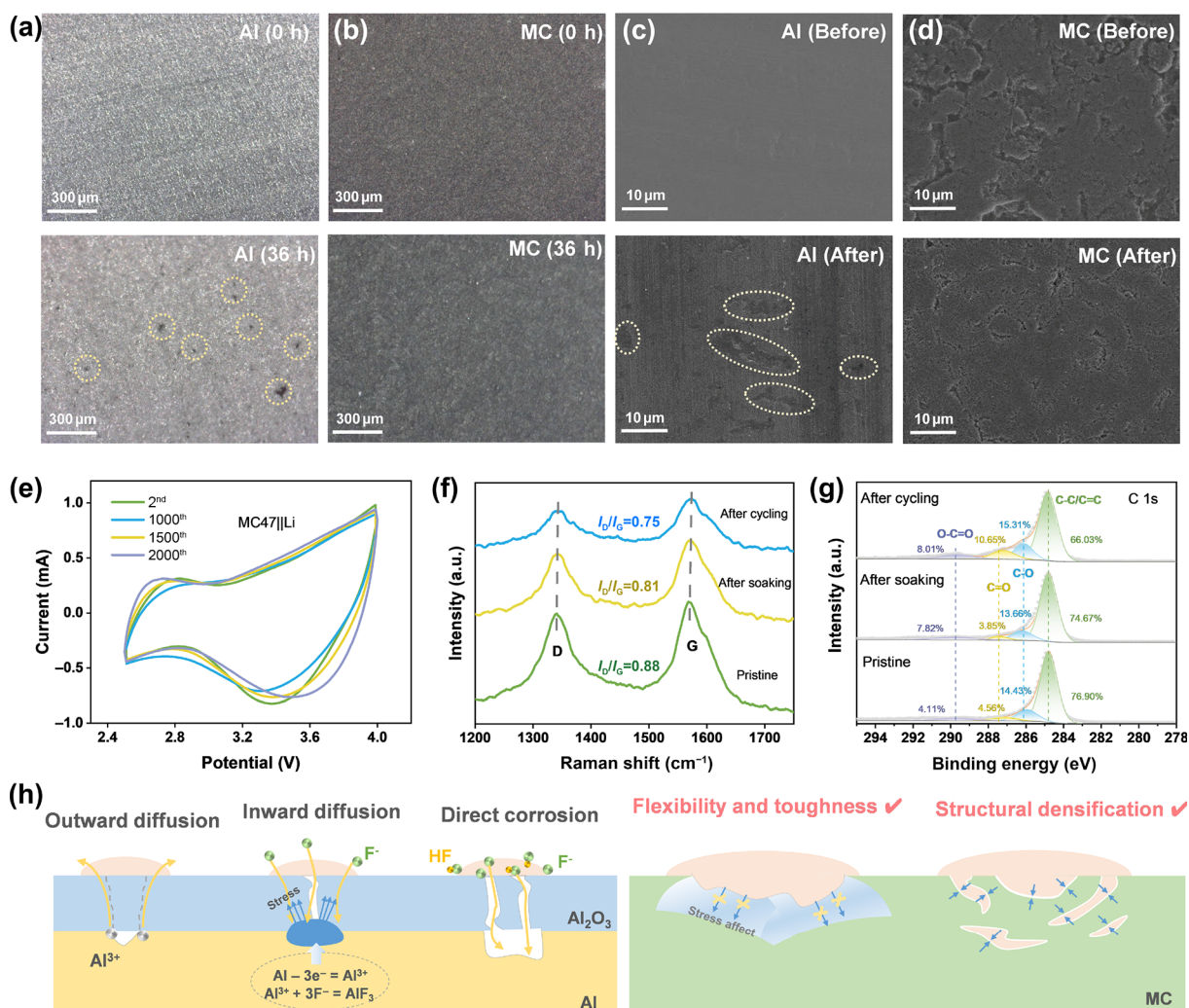


Figure 4 (a, b) Optical images of Al foil and MC before and after corrosion testing. (c, d) SEM images of Al foil and MC before and after 2000 cycles at 10 C. (e) The CV cycling curve of the MC47||Li cell. (f) The Raman spectra of MC47 current collector under different experimental conditions. (g) X-ray photoelectron spectroscopy of MC47 current collector under different experimental conditions. (h) Schematic illustration of corrosion mechanisms for Al and stability mechanism for MC.

repeated volume changes of the active material, thereby suppressing crack initiation and interfacial debonding. Meanwhile, the polar PMMA grafts promote strong interfacial affinity, enabling persistent conformal contact and gradual structural densification during cycling rather than mechanical deterioration. X-ray photoelectron spectroscopy (XPS) analysis (Fig. 4(g)) confirms that this evolution is not accompanied by chemical reconstruction of the MG30 matrix, while only the expected formation of a stable cathode electrolyte interphase is observed. The synergistic combination of elastic stress accommodation, stable interfacial contact, and robust chemical stability preserves efficient electron transport and underpins the exceptional cycling durability of the ECC (Fig. 4(h)) [46].

3.4 Synergistic enhancement of high-rate kinetics and structural longevity

The superior electrochemical kinetics of the ECC system originate from its capacity to establish high-efficiency, through-thickness electron transport pathways. Unlike metallic foils, where electronic transport is often bottlenecked by high contact resistance at the rigid collector/electrode junction [20], the ECC facilitates a continuous electronic bridge. As demonstrated in Fig. 5(a), the through-plane conductivity of the integrated cathode/collector assembly scales monotonically with MWCNTs content [47–49]. Remarkably, even at moderate filler loadings, the through-plane conductivity of the ECC-based electrode surpasses that of the Al

foil-based counterpart ($0.07 \text{ S}\cdot\text{m}^{-1}$), highlighting that the total electrode resistance is dominated by interfacial contact rather than the intrinsic conductivity of the metal itself [50]. At an optimized MG30:MWCNTs ratio (MC48), the conductivity reaches a maximum, exceeding the Al-based system by an order of magnitude. This enhancement is attributed to the 'interfacial merging' discussed previously, which replaces a discrete, high-impedance contact with a percolated, chemically unified electronic network.

This behavior correlates with the intrinsic conductivity of the freestanding collectors (Fig. S16 in the ESM), which exhibits a non-monotonic dependence on MWCNTs content [51, 52]. The optimal composition (MC47, $11.11 \text{ S}\cdot\text{cm}^{-1}$) reflects a balance between network formation and structural integrity. At lower MWCNTs contents, the percolation network is incomplete, while excessive CNT loading weakens the elastomeric matrix and induces aggregation, even for nanotubes [53, 54]. These results highlight that MWCNTs not only improves dispersion but also stabilizes the conductive network via interfacial coupling with MG30, which is critical for maintaining conductivity under mechanical and electrochemical stress. Long-term cycling tests at 3 C were also conducted on current collectors with different compositions. The results indicated that the sample of MC47 exhibited the highest capacity retention of 93.33% after 1000 cycles (Figs. S17 and S18 in the ESM). The term "MC" denotes this optimal composite with a mass ratio of 4:7 in the subsequent

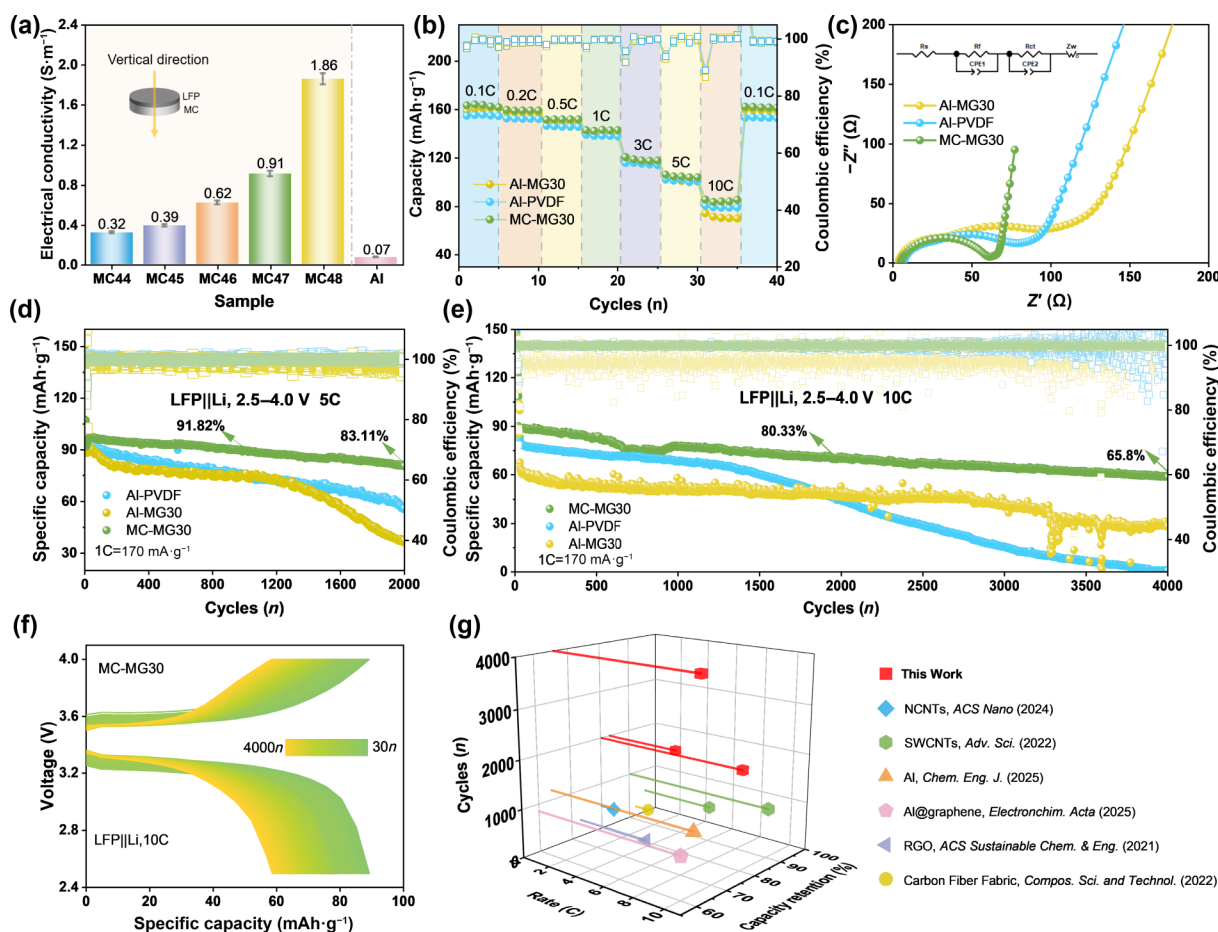


Figure 5 (a) Through-plane electrical conductivity of cathodes assembled with MC at different MG30/MWCNTs ratios, compared with Al foil. (b) Rate capability of LFP||Li cells with different collector/binder combinations. (c) Nyquist plots and fitted impedance parameters. (d) Long-term cycling at 5 C. (e) Long-term cycling at 10 C. (f) Galvanostatic charge-discharge profiles during prolonged cycling at 10 C. (g) Comparison with the reported performance of the relevant batteries with different current collectors. (Notes: "Al-PVDF": Al foil+PVDF binder, "50n": 50 cycles.)

discussion.

The improved electron transport directly translates into enhanced electrochemical kinetics. As shown in Fig. 5(b), the MC-MG30 configuration exhibits superior rate capability compared to both Al-PVDF and Al-MG30 systems. While capacities are comparable at low rates (0.1–5 C), a clear divergence emerges at 10 C, where MC-MG30 delivers a higher capacity (85.7 mAh·g⁻¹) and nearly complete recovery upon returning to 0.1 C. This behavior indicates minimal kinetic hysteresis and excellent structural reversibility. Electrochemical impedance spectroscopy (Fig. 5(c)) further confirms this advantage, with the MCx-MG30 cell exhibiting a significantly lower charge-transfer resistance (R_{ct} = 63 Ω) than Al-PVDF (92 Ω) and Al-MG30 (113 Ω). This reduction is attributed not only to improved electronic pathways but also to the suppression of interfacial degradation, as strong adhesion prevents microcrack formation and loss of electrical contact during cycling.

The transformative impact of the chemically unified elastomeric interface is most evident during extended high-rate cycling, where traditional metal-foil systems typically succumb to accumulative interfacial fatigue. At moderate rates (3 C and 5 C), the ECC-based cells demonstrate remarkable durability, retaining 93.33% capacity over 1,000 cycles and 83.11% over 2000 cycles, respectively (Fig. S19 in the ESM and Fig. 5(d)). Under ultra-high-rate conditions (10 C, Fig. 5(e)), the performance gap widens significantly; the ECC cell maintains 80.33% capacity retention after 2000 cycles and persists for 4000 cycles with 65.8% retention and a stable Coulombic efficiency (CE) of 99.8%. In stark contrast, the Al-based counterparts exhibit erratic CE fluctuations and rapid capacity decay, with the Al-PVDF system suffering a catastrophic short circuit near 3000 cycles.

The mechanistic origin of this stability is revealed by the evolution of voltage profiles and galvanostatic charge/discharge (GCD) curves (Fig. 5(f) and Fig. S20 in the ESM). The ECC-based cell exhibits remarkably stable discharge plateaus corresponding to the Fe²⁺/Fe³⁺ redox couple, with negligible increase in overpotential even after thousands of cycles [55]. This indicates that the 'monolithic' interface effectively suppresses the growth of interfacial resistance. Conversely, the Al-based cells show a progressive widening of the voltage gap (polarization) and a transition toward sloping profiles, characteristic of hindered charge-transfer kinetics and loss of electronic contact. When benchmarked against contemporary literature (Fig. 5(g) and Table S1 in the ESM), the ECC system ranks among the top performers in terms of simultaneous rate capability and cycle life. This underscores that the 'chemical coherence' strategy does not merely improve adhesion, but fundamentally preserves the electronic integrity of the electrode under the high-flux lithium-ion transport required for next-generation energy storage.

3.5 Demonstrating structural integrity in high-loading and flexible pouch cell formats

To bridge the gap toward practical commercialization and high energy density, it is imperative to increase active material mass loading without compromising mechanical or electrochemical stability. Conventional current collectors typically provide insufficient contact area and mechanical anchoring at high loadings, leading to catastrophic electrode delamination and increased internal resistance [8, 56, 57].

To address these limitations, we extended our elastomeric design to a freestanding, monolithic cathode architecture. By

utilizing the MG30/MWCNTs framework as a 3D conductive scaffold, we eliminated the discrete collector/electrode interface entirely. This monolithic configuration establishes a truly continuous electronic network throughout the high-loading electrode, effectively minimizing interfacial impedance. The resulting electrode maintains a uniform thickness (114 μ m) and a dense, cohesive morphology (Fig. S21 in the ESM). Electrochemical impedance spectroscopy (EIS) (Fig. 6(a)) reveals a remarkably low bulk resistance (R_s < 10 Ω) and a suppressed charge-transfer resistance (R_{ct} = 50 Ω), underscoring the efficiency of the interconnected electronic and ionic transport pathways. This facilitated transport behavior is a direct consequence of the synergistic combination of a robustly percolated conductive network and the enhanced electrolyte wettability afforded by the polar MG30 matrix.

Electrochemical testing (Figs. 6(b) and 6(c)) demonstrates excellent rate capability and reversibility, with capacities fully recovering upon returning to low rates. The electrode maintains stable cycling performance, achieving high capacity retention at 0.5 C (Fig. 6(d)) and 94.8% retention after 150 cycles at 1 C (Fig. 6(e) and Fig. S22 in the ESM), along with near-unity Coulombic efficiency. These results confirm that the unified elastomeric architecture effectively mitigates mechanical and interfacial degradation even at high loading. Finally, the intrinsic flexibility of the system was validated in a pouch cell configuration (Fig. 6(f)). The device delivers stable output (>3 V, Fig. S23 in the ESM) and maintains operation under repeated bending and folding without performance degradation. This robustness highlights the unique advantage of the elastomeric collector in enabling mechanically adaptive energy storage devices, where conventional rigid metal collectors fail [58].

4 Conclusions

This study demonstrates that achieving chemical and mechanical coherence at the current collector/electrode interface is vital for overcoming the limitations of conventional energy storage. By synthesizing an MG30-based elastomeric current collector, we created a robust conductive network where induced dipole interactions and mechanical interlocking ensure both structural integrity and efficient electron transport. The pivotal innovation lies in the use of a unified elastomer chemistry across the collector and binder, which transforms the interface from a discrete junction into a monolithic, integrated unit. This seamless architecture not only stabilizes the electrode against mechanical delamination but also provides intrinsic resistance to the fluoride-induced corrosion typical of metallic foils. The resulting LFP||Li cells exhibit superior rate capability and long-term stability, maintaining high capacity even under rigorous high-current cycling. Furthermore, the successful implementation in high-mass-loading electrodes and flexible pouch cells underscores the practical versatility of this approach. Ultimately, this work provides a blueprint for next-generation battery designs where interfacial coherence enables enhanced electrochemical performance and mechanical resilience.

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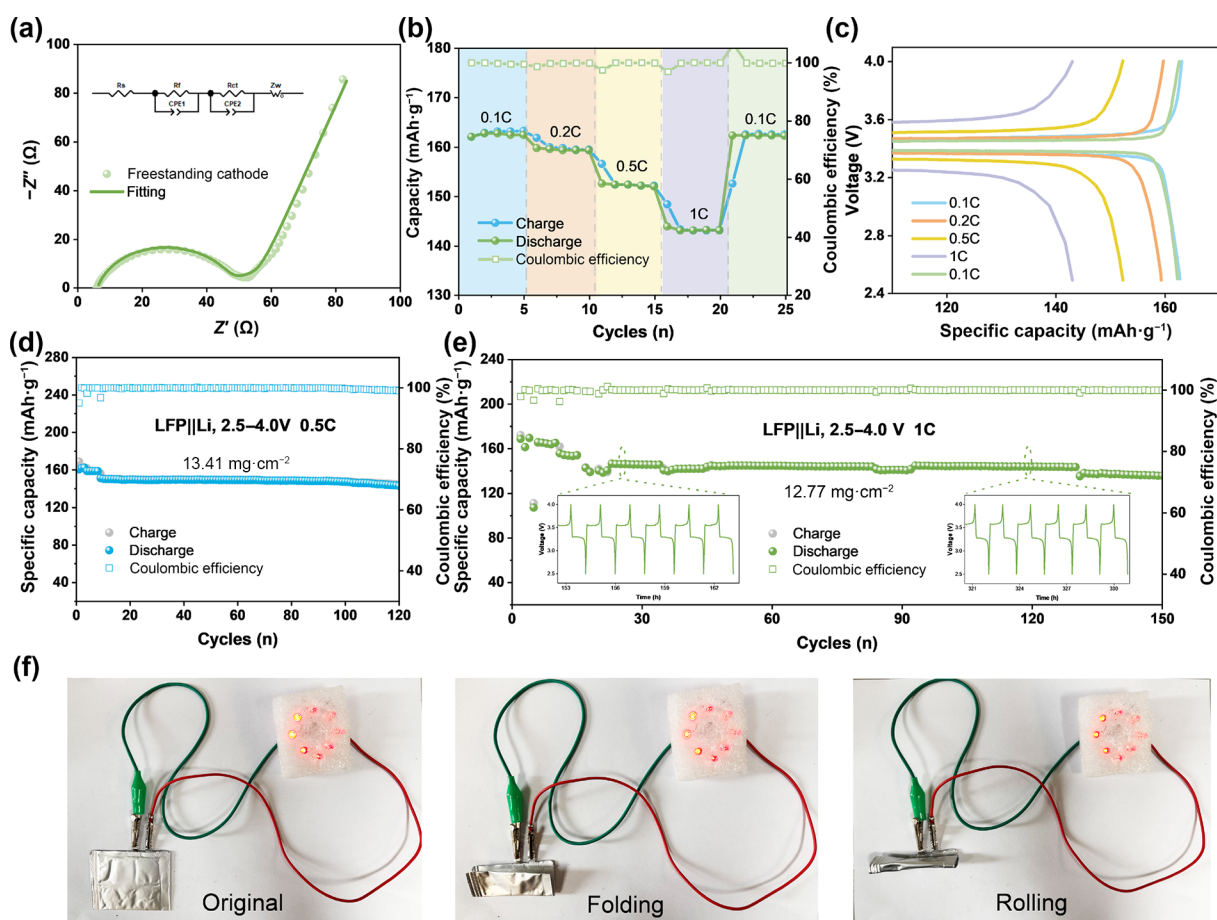


Figure 6 Electrochemical performance and practical demonstrations of high-mass-loading and flexible pouch cell architectures. (a) EIS spectra of the monolithic freestanding electrodes, illustrating the low internal resistance and efficient charge transfer. (b) Rate capability and (c) corresponding galvanostatic charge-discharge profiles of the high-loading cells, demonstrating robust kinetics under increasing current densities. (d, e) Long-term cycling stability and Coulombic efficiency at 0.5 C and 1 C, highlighting the structural integrity of the "chemically unified" interface. (f) Digital photographs of a flexible LFP||Li pouch cell successfully powering an LED array under various extreme mechanical deformations, including original, folded, and rolled states, demonstrating the exceptional mechanical adaptability and reliability of the elastomeric current collector system.

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 fourier transform infrared spectroscopy, Raman spectra, AFM images, 3D profilometer images, mechanical property tests, and electrochemical performance tests such as electronic conductivity, long-term cycling performance of batteries, and charge-discharge curve data) is available in the online version of this article at <https://doi.org/10.26599/NRE.2026.9120267>.

References

- [1] Wan, H. L.; Xu, J. J.; Wang, C. S. Designing electrolytes and interphases for high-energy lithium batteries. *Nat. Rev. Chem.* **2024**, *8*, 30–44.
- [2] Li, Y. Y.; Li, C. J.; Hu, F. B.; Yu, W. Q.; Dong, H. G.; Meng, Q.; Zhu, Z. Y.; Dong, P. Direct regeneration of spent LiFePO_4 cathode materials through Li^+ supplementation and Sm doping. *Nano Res. Energy* **2025**, *4*, e9120190.
- [3] Chen, S.; Wu, G. B.; Jiang, H. B.; Wang, J. F.; Chen, T. T.; Han, C. Y.; Wang, W. W.; Yang, R. C.; Zhao, J. H.; Tang, Z. H. et al. External Li supply reshapes Li deficiency and lifetime limit of batteries. *Nature* **2025**, *638*, 676–683.
- [4] Hao, Z. M.; Lu, Y.; Yang, G. J.; Zhao, Q.; Yan, Z. H.; Chen, J. Designing current collectors to stabilize Li metal anodes. *Adv. Mater.* **2025**, *37*, 2415258.
- [5] Du, Y. J.; Luo, Y.; Shi, K. Y.; Zuo, P. J.; Zhang, Q. S.; Zheng, Z. Q.; Sun, B. Y.; Xie, J. Y. From metals to polymers: Material evolution and functional advancements in current collectors. *Adv. Mater.* **2025**, *37*, 2502095.
- [6] Xu, K. Electrolytes and interphases in Li-ion batteries and beyond. *Chem. Rev.* **2014**, *114*, 11503–11618.
- [7] Chen, J. Y.; Zhang, S. H.; Huang, Y. H.; Han, X. R.; Li, H. R.; Liu, G. Y.; Shi, L.; Zhang, Y.; Shen, Z.; Wang, Y. Z. et al. Expandable bidirectional-gradient current collector for high-performance Li metal battery. *Adv. Funct. Mater.* **2025**, *35*, 2500531.
- [8] Kim, S. Y.; Liu, T. K.; Kim, H. S.; Wang, D. H.; Park, J. H. A solvent-free, dry-processed Li-ion battery enabled by dual binders and nanostructured aluminum current collectors. *Adv. Funct. Mater.* **2026**, *36*, e18597.
- [9] Park, J.; Jung, J.; Yang, S.; Kim, D. H.; Lee, J. B.; Oh, J. A. S.; Yang, H. D.; Nashiru, M.; Huang, C. J.; Meng, Y. S. Realizing low-

- pressure operation of all-solid-state lithium-sulfur batteries enabled by carbon-coated current collectors. *Adv. Energy Mater.* **2026**, *16*, e04272.
- [10] Zhu, P. C.; Gastol, D.; Marshall, J.; Sommerville, R.; Goodship, V.; Kendrick, E. A review of current collectors for lithium-ion batteries. *J. Power Sources* **2021**, *485*, 229321.
- [11] Offermann, J.; Gayretli, E.; Schmidt, C.; Carstensen, J.; Bremes, H. G.; Würsig, A.; Hansen, S.; Abdollahifar, M.; Adelung, R. Enabling high-performance battery electrodes by surface-structuring of current collectors and crack formation in electrodes: A proof-of-concept. *J. Colloid Interface Sci.* **2024**, *664*, 444–453.
- [12] Wang, M. Z.; Tang, M.; Chen, S. L.; Ci, H. N.; Wang, K. X.; Shi, L. R.; Lin, L.; Ren, H. Y.; Shan, J. Y.; Gao, P. et al. Graphene-armored aluminum foil with enhanced anticorrosion performance as current collectors for lithium-ion battery. *Adv. Mater.* **2017**, *29*, 1703882.
- [13] Nam, G.; Lim, J.; Choi, S.; Hong, R.; Yang, W.; Kwon, M.; Oh, J.; Kang, D.; Lee, Y. M. Revisiting primer layer design for robust dry electrode-current collector interfaces in lithium-ion batteries. *ACS Energy Lett.* **2026**, *11*, 5149–5159.
- [14] Du, X. F.; Wang, Q. W.; Feng, T. Y.; Chen, X. Z.; Li, L.; Li, L.; Meng, X. F.; Xiong, L. L.; Sun, X. F.; Lu, L. et al. One-step preparation of nanoarchitected TiO₂ on porous Al as integrated anode for high-performance lithium-ion batteries. *Sci. Rep.* **2016**, *6*, 20138.
- [15] Kim, S. Y.; Song, Y. I.; Wee, J. H.; Kim, C. H.; Ahn, B. W.; Lee, J. W.; Shu, S. J.; Terrones, M.; Kim, Y. A.; Yang, C. M. Few-layer graphene coated current collectors for safe and powerful lithium ion batteries. *Carbon* **2019**, *153*, 495–503.
- [16] Peng, L.; Huang, X. Y.; Zeng, Q. Y.; Wei, L. J.; Liu, Z. Z.; Lv, X. Y.; Su, J.; Wen, Y. X. Fabrication of a rigid-flexible and dual-conductive interphase on an aluminum current collector for ultra-stable anode-free sodium batteries. *Small* **2025**, *21*, e08205.
- [17] Koerver, R.; Zhang, W. B.; de Biasi, L.; Schweidler, S.; Kondrakov, A. O.; Kolling, S.; Brezesinski, T.; Hartmann, P.; Zeier, W. G.; Janek, J. Chemo-mechanical expansion of lithium electrode materials-on the route to mechanically optimized all-solid-state batteries. *Energy Environ. Sci.* **2018**, *11*, 2142–2158.
- [18] Xu, T.; Zou, L.; Yang, Z. H.; Zhang, W. X. A comparative study on polymer-based cathode composite current collector with single-layer and multi-layer conductive Al layers. *J. Membr. Sci.* **2026**, *744*, 125214.
- [19] Xing, Z. H.; Zou, S. H.; Ma, C.; Qiao, Q. Q.; Cai, X. H.; Yue, K.; Yue, J. X.; Zhou, C. Y.; Zheng, J. L.; Wang, Y. et al. π - π Stacked Nigrosine@Carbon nanotube nanocomposite as an all-in-one additive for high energy flexible batteries. *ACS Nano* **2024**, *18*, 17950–17957.
- [20] Shi, Y.; Wang, Z. X.; Wen, L.; Pei, S. F.; Chen, K.; Li, H. C.; Cheng, H. M.; Li, F. Ultrastable interfacial contacts enabling unimpeded charge transfer and ion diffusion in flexible lithium-ion batteries. *Adv. Sci.* **2022**, *9*, 2105419.
- [21] Yu, D. S.; Goh, K.; Wang, H.; Wei, L.; Jiang, W. C.; Zhang, Q.; Dai, L. M.; Chen, Y. Scalable synthesis of hierarchically structured carbon nanotube-graphene fibres for capacitive energy storage. *Nat. Nanotechnol.* **2014**, *9*, 555–562.
- [22] Liu, Z. X.; Li, H. F.; Zhu, M. S.; Huang, Y.; Tang, Z. J.; Pei, Z. X.; Wang, Z. F.; Shi, Z. C.; Liu, J.; Huang, Y. et al. Towards wearable electronic devices: A quasi-solid-state aqueous lithium-ion battery with outstanding stability, flexibility, safety and breathability. *Nano Energy* **2018**, *44*, 164–173.
- [23] Pire, M.; Norvez, S.; Iliopoulos, I.; Le Rossignol, B.; Leibler, L. Epoxidized natural rubber/dicarboxylic acid self-vulcanized blends. *Polymer* **2010**, *51*, 5903–5909.
- [24] Fu, Y. H.; Zhang, S. X.; Li, B. R.; Wang, L. J.; Zhang, Y. B.; Ye, Z. W.; Zhou, Y. S.; Yi, J.; Qin, H. M.; Shi, Q. et al. Highly ordered rubbers with a giant elastocaloric effect. *Sci. Adv.* **2026**, *12*, eady2745.
- [25] Jeschke, J.; Mutlu, H. From waste rubber to value polybutadiene modification for circular materials. *Nat. Commun.* **2026**, *17*, 1487.
- [26] Xia, K.; Yao, Z. Y.; Zhao, P. F.; Liu, Z.; Zhao, S.; Peng, Z.; Zhang, P. Enabling 10C-rate fast charging in LiFePO₄ based lithium-ion battery by balancing the electronic and ionic conductivity paths in cathode. *Chem. Eng. J.* **2025**, *506*, 160290.
- [27] Zhou, Z. K.; Tao, Z. R.; Chen, R. Y.; Liu, Z.; He, Z. H.; Zhong, L.; Li, X.; Chen, G. X.; Zhang, P. Elastomeric electrolyte for high capacity and long-cycle-life solid-state lithium metal battery. *Small Methods* **2023**, *7*, 2201328.
- [28] Liu, Y. L.; Zhang, S. F.; Li, L.; Li, N. High-performance cellulose nanofibers/carbon nanotubes composite for constructing multifunctional sensors and wearable electronics. *Adv. Fiber Mater.* **2024**, *6*, 758–771.
- [29] Mohamed, S. A. A.; Tohamy, H. A. S.; El-Sakhawy, M.; Othman, A. M.; Kamel, S. Potential application of new antimicrobials biological macromolecule of CNT/ZnO-NPs based on hydroxypropyl methylcellulose/shellac as packaging films. *Int. J. Biol. Macromol.* **2025**, *306*, 141421.
- [30] Zhao, S.; Luo, S. Y.; Zhang, Y. M.; Xie, R. Y.; Liu, Z.; Yao, Z. Y.; He, X. R.; Li, J. B.; Yao, X.; Zhou, Z. et al. Adhesive tape-inspired polymer coatings enable record-stable zinc anodes for high-rate aqueous batteries. *Adv. Sci.* **2026**, *13*, e20648.
- [31] Assumpção, N. R. L.; Lona, L. M. F. Effect of lignin without surface treatment in *in situ* methyl methacrylate miniemulsion polymerization. *ACS Sustain. Chem. Eng.* **2022**, *10*, 3219–3226.
- [32] Wilke, L. A.; Robertson, C. G.; Karsten, D. A.; Hardman, N. J. Detailed understanding of the carbon black-polymer interface in filled rubber composites. *Carbon* **2023**, *201*, 520–528.
- [33] Das, A.; Kasaliwal, G. R.; Jurk, R.; Boldt, R.; Fischer, D.; Stöckelhuber, K. W.; Heinrich, G. Rubber composites based on graphene nanoplatelets, expanded graphite, carbon nanotubes and their combination: A comparative study. *Compos. Sci. Technol.* **2012**, *72*, 1961–1967.
- [34] Skountzou, E. N.; Mermigkis, P. G.; Mavrantzas, V. G. Molecular dynamics study of an atactic poly(methyl methacrylate)-carbon nanotube nanocomposite. *J. Phys. Chem. B* **2018**, *122*, 9007–9021.
- [35] Inoue, A. D.; Yoshida, K.; Ando, T.; Fukura, S.; Yumura, T.; Ajiro, H.; Kawai, T.; Nonoguchi, Y. Dispersion of carbon nanotubes triggered by the helical self-assembly of poly(methyl methacrylate). *Nanoscale* **2025**, *17*, 18105–18111.
- [36] Qin, J. K.; Liao, P. Y.; Si, M. W.; Gao, S. Y.; Qiu, G.; Jian, J.; Wang, Q. X.; Zhang, S. Q.; Huang, S. Y.; Charnas, A. et al. Raman response and transport properties of tellurium atomic chains encapsulated in nanotubes. *Nat. Electron.* **2020**, *3*, 141–147.
- [37] Yao, Z. Y.; Liu, Z.; Xia, K.; Zhao, S.; Xie, H. R.; Li, X. R.; Zhou, S. L.; Sun, D. B.; Zhang, P. Design and preparation of self-adaptive and robust solid-state elastomeric electrolyte for lithium metal battery inspired by rubber tire. *Angew. Chem., Int. Ed.* **2025**, *64*, e202513167.
- [38] Bokobza, L. Multiwall carbon nanotube elastomeric composites: A review. *Polymer* **2007**, *48*, 4907–4920.
- [39] Miao, X.; Guan, S. D.; Ma, C.; Li, L. L.; Nan, C. W. Role of interfaces in solid-state batteries. *Adv. Mater.* **2023**, *35*, 2206402.
- [40] Persson, B. N. J. Theory of rubber friction and contact mechanics. *J. Chem. Phys.* **2001**, *115*, 3840–3861.
- [41] Beharaj, A.; McCaslin, E. Z.; Blessing, W. A.; Grinstaff, M. W. Sustainable polycarbonate adhesives for dry and aqueous conditions with thermoresponsive properties. *Nat. Commun.* **2019**, *10*, 5478.
- [42] Nguyen, V. H.; Wang, W. L.; Jin, E. M.; Gu, H. B. Impacts of different polymer binders on electrochemical properties of LiFePO₄ cathode. *Appl. Surf. Sci.* **2013**, *282*, 444–449.
- [43] Shi, X. W.; Zhang, Y.; Zhang, H. D.; Zhao, Z. H.; Liu, B. H.; Liu, J. M.; Liu, X. C.; Li, L. Graphene enables aluminum current collectors of 5 V class battery. *Nano Lett.* **2024**, *24*, 12398–12405.
- [44] Yoon, E.; Lee, J.; Byun, S.; Kim, D.; Yoon, T. Passivation failure of Al current collector in LiPF₆-based electrolytes for lithium-ion batteries. *Adv. Funct. Mater.* **2022**, *32*, 2200026.
- [45] Yang, S. L.; Li, S. M.; Du, J.; Han, C.; Zhong, J. Y.; Yu, M.; Li, B. Unveiling the passivation and corrosion process of cathode

- aluminum current collector in lithium-ion battery. *Corros. Sci.* **2024**, 237, 112359.
- [46] Zhang, D. C.; Liu, Y. X.; Li, D. D.; Li, S. M.; Xiong, Q.; Huang, Z. D.; Wang, S. X.; Hong, H.; Zhu, J. X.; Lv, H. M. et al. Salt dissociation and localized high-concentration solvation at the interface of a fluorinated gel and polymer solid electrolyte. *Energy Environ. Sci.* **2025**, 18, 227–235.
- [47] He, Z. Y.; Zhang, C. X.; Zhu, Z. X.; Yu, Y. X.; Zheng, C.; Wei, F. Advances in carbon nanotubes and carbon coatings as conductive networks in silicon-based anodes. *Adv. Funct. Mater.* **2024**, 34, 2408285.
- [48] Kinloch, I. A.; Suhr, J.; Lou, J.; Young, R. J.; Ajayan, P. M. Composites with carbon nanotubes and graphene: An outlook. *Science* **2018**, 362, 547–553.
- [49] Kim, A. Y.; Lee, M.; Shin, H. J.; Kim, H.; Choi, J.; Kim, J. H.; Yoo, J. K.; Lee, M.; Han, J. T.; Ha, J. S. et al. Carbon nanotube-grafted silicon-carbon composite as a highly durable anode material for lithium-ion batteries. *J. Mater. Chem. A* **2026**, 14, 6951–6962.
- [50] Kushima, A.; Koido, T.; Fujiwara, Y.; Kuriyama, N.; Kusumi, N.; Li, J. Charging/discharging nanomorphology asymmetry and rate-dependent capacity degradation in Li-oxygen battery. *Nano Lett.* **2015**, 15, 8260–8265.
- [51] Huang, S. Z.; Zhang, L. L.; Wang, J. Y.; Zhu, J. L.; Shen, P. K. *In situ* carbon nanotube clusters grown from three-dimensional porous graphene networks as efficient sulfur hosts for high-rate ultra-stable Li-S batteries. *Nano Res.* **2018**, 11, 1731–1743.
- [52] Liu, S.; Dong, C.; Che, S.; Ma, G.; Cheng, G.; Tian, Y. C.; Xiao, X.; Huang, Y. N.; Gao, W. Y.; Dang, Y. X. et al. Graphene-coated current collectors for enhanced interfacial stability and electrochemical performance in LiFePO₄/Li batteries. *Electrochim. Acta* **2025**, 540, 147231.
- [53] Guadagno, L.; Raimondo, M.; Vertuccio, L.; Naddeo, C.; Barra, G.; Longo, P.; Lamberti, P.; Spinelli, G.; Nobile, M. R. Morphological, rheological and electrical properties of composites filled with carbon nanotubes functionalized with 1-pyrenebutyric acid. *Compos. Part B: Eng.* **2018**, 147, 12–21.
- [54] Ma, Q.; Hao, B.; Ma, P. C. *In-situ* characterization on the fracture behavior of three dimensional polymer nanocomposites reinforced by CNT sponge. *Compos. Sci. Technol.* **2022**, 217, 109132.
- [55] Tang, S.; Lan, Q.; Xu, L.; Liang, J. Y.; Lou, P.; Liu, C.; Mai, L.; Cao, Y. C.; Cheng, S. J. A novel cross-linked nanocomposite solid-state electrolyte with super flexibility and performance for lithium metal battery. *Nano Energy* **2020**, 71, 104600.
- [56] Ji, T. T.; Zhang, Y. X.; Torres, J.; Mijailovic, A. S.; Tang, Y.; Zhao, X. H.; Bilheux, J. C.; Wang, J. W.; Sheldon, B. W.; Oyediji, O. et al. Operando neutron imaging-guided gradient design of Li-ion solid conductor for high-mass-loading cathodes. *Nat. Commun.* **2025**, 16, 7667.
- [57] Kim, J.; Kim, K. W.; Yoon, H.; Park, S.; Woo, D.; Jo, H.; Jo, C.; Kim, J. K. Ultrahigh mass loading, binder-free synthetic approach for mesoporous graphitic carbon in 3D current collector for high energy lithium-ion batteries. *J. Mater. Chem. A* **2025**, 13, 4521–4529.
- [58] Zhu, X.; Zhang, H. R.; Huang, Y. X.; He, E.; Shen, Y.; Huang, G.; Yuan, S. Y.; Dong, X. L.; Zhang, Y.; Chen, R. J. et al. Recent progress of flexible rechargeable batteries. *Sci. Bull.* **2024**, 69, 3730–3755.



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