



Architectural reconfiguration of foil-dependent cathodes via percolating carbon nanotube scaffolds

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ABSTRACT

Conventional lithium-ion battery electrodes rely on metallic current collectors and polymeric binders to ensure interparticle contact and reduce resistance. However, such foil-dependent architectures are inherently incompatible with repeated mechanical deformation, leading to delamination and performance degradation. Here, we report a single-walled carbon nanotubes (SWCNTs)-integrated collector-free (CNT-CF) electrode enabled by monodispersed SWCNTs dispersion. The CNT-CF electrode is fabricated by homogenizing the active material with the SWCNTs dispersion, followed by slurry casting onto a substrate and drying to form a dense self-supporting electrode, while eliminating both metallic foils and polymeric binders. Due to their high aspect ratio ($> 10,000$), SWCNTs form a continuous three-dimensional percolating framework that conformally wraps active material particles, establishing line-contact-based electronic pathways throughout the electrode. This interconnected scaffold homogenizes electron distribution, mitigates localized chemo-mechanical stress accumulation, and suppresses electrode-level degradation. A dense and self-supporting CNT-CF electrode is achieved with only 1.5 wt% SWCNTs, delivering a high mass loading of $\sim 11.1 \text{ mg cm}^{-2}$ and an areal capacity of $\sim 2.19 \text{ mAh cm}^{-2}$ while maintaining an electrode-level volumetric energy density exceeding $\sim 2500 \text{ Wh L}^{-1}$. Furthermore, the CNT-CF electrode retains over 93% of its initial capacity after 1000 bending cycles without delamination. This work provides a practical pathway toward lightweight, flexible, and high-energy-density lithium-ion batteries.

1. Introduction

The rapid emergence of flexible and wearable electronics, including roll-up displays, smart clothing, and implantable medical sensors, has driven an urgent demand for compliant energy storage systems [1–3]. Although Lithium-ion batteries (LIBs) remain the dominant power source for portable electronics, their conventional rigid architectures are fundamentally incompatible with the repeated bending, stretching, and twisting required for soft and deformable interfaces [4,5]. To address this mismatch, extensive efforts have been devoted to structural innovations such as one-dimensional cable-type batteries and two-dimensional planar configurations designed to accommodate mechanical deformation [6–9]. Despite these advances, a persistent challenge remains simultaneously achieving both high energy density and mechanical durability, as most conventional battery components are intrinsically designed for static, rigid operation rather than complex and repeated mechanical stress. [10–13]

At the core of this trade-off lies the conventional electrode

architecture that relies on metallic current collectors. In commercial LIBs, the metallic current collectors (typically Al or Cu foils) account for $\sim 15\%$ – 20% of the total cell weight while providing zero capacity [14,15]. More importantly, this “dead-weight” drawback is accompanied by a second limitation in flexible operation. Under repeated mechanical deformation, rigid metallic foils are prone to irreversible creasing, progressively leading to localized strain and accelerating catastrophic detachment of the active material from the collector [16]. Furthermore, beyond mechanical mismatch, long-term electrolyte exposure can also compromise collector integrity; Al is susceptible to pitting corrosion, whereas Cu can undergo environmentally assisted cracking, further destabilizing electrical contact with internal ohmic resistance growth [17]. To address these instabilities, extensive efforts have been devoted to free-standing and binder-free electrodes that remove inactive components. However, many prior designs still rely on pre-formed carbon substrates produced by chemical vapor deposition (CVD) or vacuum filtration-based assembly, which limits scalability and compatibility with manufacturing-relevant coating processes [18–20].

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In addition, even when high mass loading is demonstrated, it is often achieved through substrate-first or batch-type routes like freeze-drying, making it difficult to achieve simultaneously high areal capacity and reliable mechanical integrity [21]. More importantly, beyond processing limitations, the intrinsic architecture of conventional slurry-cast electrodes introduces fundamental constraints. Particulate conductive additives like carbon black form heterogeneous point-contact pathways that concentrate local current density and induce non-uniform lithiation [22,23]. This heterogeneity amplifies chemo-mechanical stress accumulation, leading to particle-level cracking associated with “electrochemical indentations” (ECIs); these localized failures are further exacerbated by mechanical stiffness mismatches, eventually leading to progressive delamination under repeated bending [24]. Therefore, these limitations highlight the need for a scalable free-standing electrode strategy that enables commercially relevant areal capacity, uniform particle-scale electronic/ionic reaction fields, and delamination-free mechanical reliability under repeated deformation. Notably, existing free-standing electrode studies have primarily focused on improving mechanical flexibility and structural robustness [18,20,25]. However, little attention has been paid to how electrode architecture governs particle-scale reaction heterogeneity and the resulting chemo-mechanical degradation. In addition, many reported fabrication methods remain incompatible with conventional slurry-based electrode manufacturing. Therefore, a scalable free-standing electrode strategy that simultaneously addresses reaction homogeneity, mechanical reliability, and manufacturing compatibility remains highly desirable.

In this study, we report a scalable strategy for high-performance single-walled carbon nanotubes (SWCNTs)-integrated collector-free (CNT-CF) electrode enabled by a monodispersed SWCNTs ink through a manufacturing-relevant slurry casting–drying route, eliminating both metallic current collectors and polymeric binders. Owing to their exceptionally high aspect ratio ($> 10,000$) [26], SWCNTs form a continuous three-dimensional interconnected electronic framework that conformally wraps active material particles, establishing line-contact-based percolation pathways throughout the electrode [27–29]. This interconnected scaffold homogenizes current distribution and mitigates localized chemo-mechanical stress accumulation, thereby suppressing electrochemical indentation-induced particle cracking, while allowing the SWCNTs to simultaneously function as the conductive additive, mechanical binder, and current collector surrogate [30]. Remarkably, a dense and self-supporting electrode is achieved with only 1.5 wt% SWCNTs, delivering a high mass loading of $\sim 11.1 \text{ mg cm}^{-2}$ (areal capacity of $\sim 2.19 \text{ mAh cm}^{-2}$) and an electrode-level volumetric energy density exceeding $\sim 2500 \text{ Wh L}^{-1}$. Moreover, this architecture maintains stable electrochemical kinetics and structural integrity under repeated mechanical deformation, retaining over 93% of its initial capacity even after 1000 bending cycles without delamination. In contrast to freeze-drying-based electrode fabrication, the coating-compatible nature of this process is directly aligned with conventional battery manufacturing and enables controllable electrode densification, providing a practical pathway toward mechanically deformable, long-cycle LIBs [31–33].

2. Experimental

2.1. Electrode preparation

Single-crystalline $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ (SC-NCM811) powder was used for electrode fabrication (MTI Korea). SWCNTs-integrated collector-free (CNT-CF) electrode was prepared using a SWCNTs dispersion in *N*-methyl-2-pyrrolidone (NMP, 0.5 wt% SWCNTs in NMP, S2104NE, BETTERIAL). SWCNTs served as a continuous conductive framework while simultaneously providing mechanical integrity without polymeric binders. Slurry mixture was prepared with 98.5 wt% SC-NCM811 and 1.5 wt% SWCNTs, and mixed and homogenized at 2000 rpm for 15 min via the planetary centrifugal mixer (ARE-310, Thinky). Doctor-blade was used to apply the slurry onto a glass substrate, followed by a 2-h

drying step at 110°C for NMP removal. Upon drying, the electrode spontaneously delaminated from the substrate due to the absence of strong adhesion between the SWCNTs network and the glass surface. For the conventional foil-dependent (FD) cathode electrode, SC-NCM811, Super P, and Polyvinylidene Fluoride (PVDF) were blended in a 94:3:3 weight ratio within an NMP (99.9%) solution, functioning as the active mass, conductor, and binder, respectively. A planetary centrifugal system (THINKY ARE-310) was employed to homogenize the slurry at a speed of 2000 rpm for a 30-min duration. The slurry was cast on Al foil via a doctor-blade technique, followed by thermal treatment in a convection oven (110°C , 2 h) for NMP removal. The cathode electrodes applied to coin-cell were cast to $11.1 \pm 0.3 \text{ mg cm}^{-2}$ and $15.0 \pm 0.3 \text{ mg cm}^{-2}$, and calendered to a density of $\sim 3.2 \text{ g cc}^{-1}$, followed by an overnight vacuum-drying process at 110°C prior to battery assembly. Graphite anodes were prepared in both FD and CNT-CF configurations. For the conventional FD anode electrode, Meso Carbon Micro Beads (MCMB) graphite (96 wt%) served as the active material, incorporated with 1 wt% Super P and a binary binder system composed of 1 wt% Carboxymethyl Cellulose (CMC) and 2 wt% Styrene-Butadiene Rubber (SBR). The CNT-CF anode was fabricated following a similar processing strategy for CNT-CF cathode electrode, while using the same MCMB graphite as the conventional anode. The areal loading level of prepared anodes was adjusted to $\sim 7 \text{ mg cm}^{-2}$.

2.2. Cell assembly

For half-cell tests, the CR 2032 coin-type cells were assembled in an Ar glove box using with the 12-mm cathodes and 14-mm lithium metal foils serving as both reference and counter electrodes. The electrolyte composition was 1.3 M LiPF₆ in ethylene carbonate (EC)/ethyl methyl carbonate (EMC)/ diethyl carbonate (DEC) (EC:EMC:DEC = 3:5:2 Vol%) with Polystyrene Sulfonate (PS) 1%, Vinyl Carbonate (VC) 1% and Fluoroethylene Carbonate (FEC) FE10% (Enchem Co.,Ltd) and 0.2 g of electrolyte was injected per cell. As the separator, the Celgard 2320 (Celgard) with diameter 19 mm was used. For full cell tests, SC-NCM811 cathodes and MCMB graphite anodes were utilized to assemble coin-type and pouch-type full cells. The negative/positive capacity ratio was set at 1.11 ± 0.01 per unit area. Single-sided coatings were applied to both electrodes, targeting areal mass loadings of $\sim 10 \text{ mg cm}^{-2}$ and $\sim 7 \text{ mg cm}^{-2}$ for the cathode and anode, respectively. Each pouch-type cell was then injected with 500 μL of electrolyte. The full cells were assembled in a dry-room environment (relative humidity $< 1\%$), utilizing the same electrolyte and separator specifications as described for the half-cell evaluations.

2.3. Material and electrode characterization

Morphological investigations of the active materials and electrodes were carried out using scanning electron microscopy (SEM; SU8600, Hitachi), which was integrated with an energy-dispersive X-ray spectroscopy (EDX; Oxford Instruments) system. To investigate the elemental composition and oxidation states, X-ray photoelectron spectroscopy (XPS; K-Alpha, Thermo Fisher Scientific) was employed. The Raman spectra were obtained using a micro-Raman system (Olympus BX51) with a 633 nm He–Ne laser. Additionally, thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) were performed with a TA Instruments SDT Q600 system, where the temperature was increased to 600°C at $10^\circ\text{C min}^{-1}$ under a 100 mL min^{-1} N_2 atmosphere. X-ray diffraction (XRD; SmartLab, Rigaku) was utilized to investigate the crystallographic structures of the samples. Data collection was performed using monochromatic Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$) across a 2θ span of 20° – 80° , maintaining a constant step size of 0.03° . In-situ XRD measurements were conducted using cathodes assembled in a battery cell kit (Bruker) inside an Ar-filled glove box. The assembled cells were mounted on the XRD stage and electrochemically cycled between 2.5 and 4.3 V vs. Li/Li⁺ at 0.05C, while XRD patterns were

continuously collected using Cu K α radiation on a D8 Advance diffractometer (Bruker). Electrode resistance was measured using a HIOKI RM2611 resistance meter equipped with RM9004 fixtures. To evaluate the mechanical flexibility of the pouch-type full cells, an Instron universal testing machine (Model 34SC-1; Instron Corporation) was employed at a 1000 mm min⁻¹ crosshead speed, with a bending radius of $\sim$ 40 mm. During the 100 charge–discharge cycles of the pouch cells, a bending test consisting of 1000 repetitions was performed at every 20-cycle interval using the Instron system. Time-of-flight secondary-ion mass spectrometry (TOF-SIMS) measurement was conducted by using ION-TOF TOF-SIMS 5, and the analysis chamber pressure was maintained lower than 1×10^{-9} mbar. Surface analysis was conducted in negative polarity using a Bi⁺ primary ion beam (high-current bunched mode) over a $400 \times 400 \mu\text{m}^2$ area for 300 s without preliminary etching. High-resolution chemical imaging was obtained using a Bi₃²⁺ primary ion beam (burst-alignment mode) over a $50 \times 50 \mu\text{m}^2$ area. Depth-resolved profiles were obtained by employing a Bi⁺ analytical beam combined with Cs⁺ sputtering (0.5 keV, $\sim$ 35 nA), with analysis and sputter areas of $50 \times 50 \mu\text{m}^2$ and $250 \times 250 \mu\text{m}^2$, respectively, for a total profiling time of 3600 s.

2.4. Electrochemical measurements

Electrochemical evaluations of the coin-type half-cells were conducted between 2.8 and 4.3 V vs. Li/Li⁺ at temperatures of 25 °C and 45 °C. The formation process consisted of cycles at 0.1C (1C $\approx$ 200 mA g⁻¹). Cycling stability was first assessed at 25 °C using a 0.5C charge/1.0C discharge protocol over 200 cycles with $11.1 \pm 0.3 \text{ mg cm}^{-2}$ electrode loading level. In addition, the electrochemical performance at a high mass loading of 15 mg cm^{-2} was evaluated at 25 °C under a 0.5C charge/1.0C discharge protocol for 100 cycles within the voltage range of 2.8–4.3 V vs. Li/Li⁺. Rate capability was subsequently examined with a constant 0.99 mA cm^{-2} charge while incrementally increasing discharge rates from 0.99 to 9.90 mA cm^{-2} (0.99, 1.98, 3.96, 5.94, and 9.90 mA cm^{-2}) for five cycles at each step before returning to 0.99 mA cm^{-2} . Upon completing the formation, 50th, and 100th cycle, electrochemical impedance spectroscopy (EIS) measurements were recorded. These were performed with a BioLogic VSP-3e workstation, spanning a frequency from 1 MHz down to 10 mHz at a 10 mV amplitude. For the galvanostatic intermittent titration technique (GITT) within 2.8–4.3 V, 0.3C current pulses were applied for 10 min, each followed by a 2 h rest period to reach equilibrium. Apart from the EIS measurements, all electrochemical cycling was performed using a NEWARE battery testing system (CT-4004-15V30A). High-temperature (45 °C) cycling stability

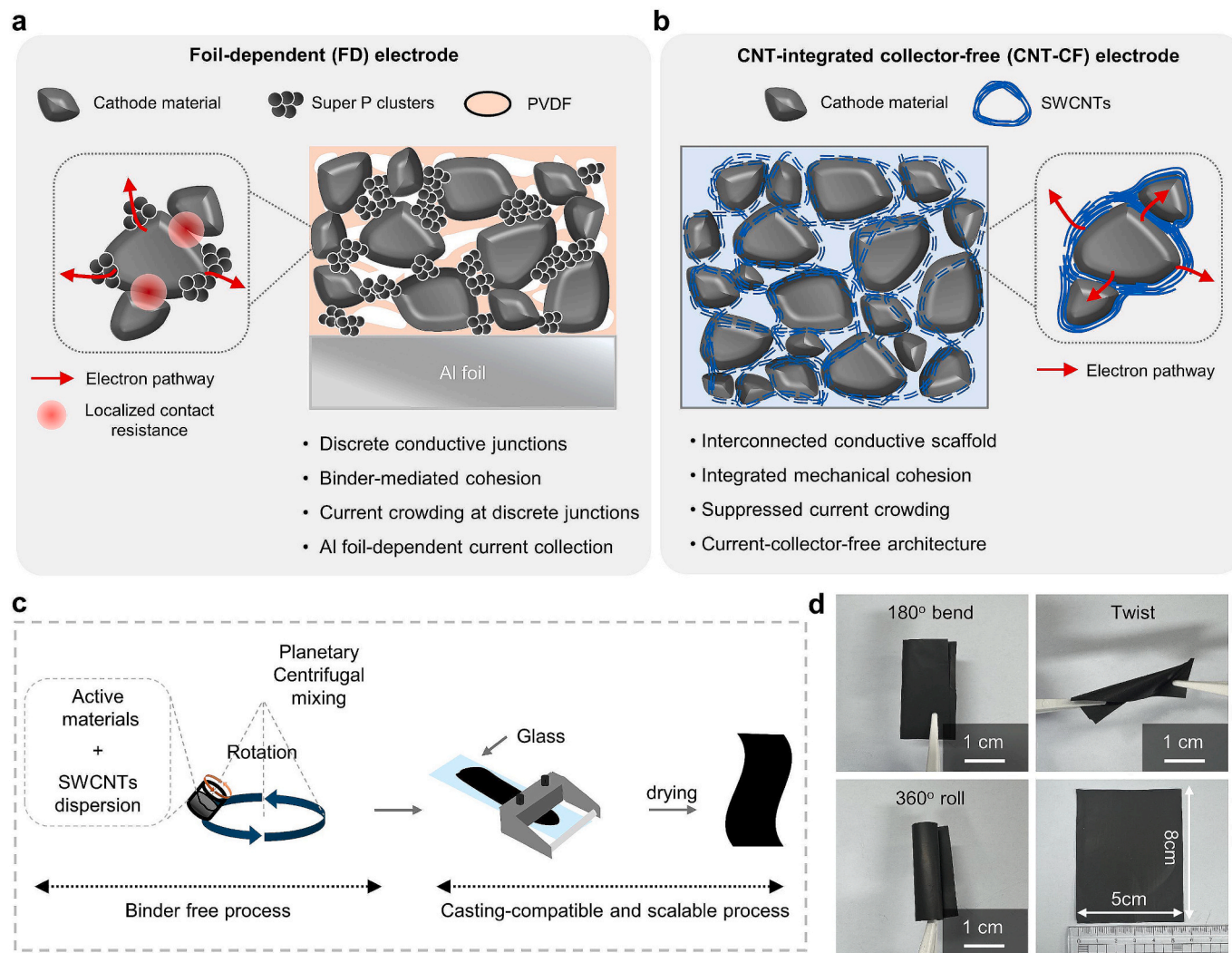


Fig. 1. Schematic illustration of the structural differences between (a) the FD electrode and (b) the CNT-CF electrode. (c) Schematic illustration of the fabrication process of the CNT-CF electrode. (d) Photographs of the CNT-CF electrode film (5 cm $\times$ 8 cm) demonstrating its mechanical flexibility under 180° bending, twisting, and 360° rolling.

was then evaluated using the same 0.5C charge/1.0C discharge protocol. Finally, high-voltage cycling stability at 25 °C was assessed over 100 cycles at 0.5C charge/1.0C discharge within a 2.8–4.6 V vs. Li/Li⁺. Full cells were evaluated within the voltage range of 2.5–4.3 V. To determine the direct current internal resistance (DC-IR), current pulses ranging from 0.5 to 4.0C (0.5, 1.0, 2.0, 3.0, and 4.0C, 1C $\approx$ 200 mA g⁻¹) were applied for 10 s at every 20% state-of-charge (SOC) interval, allowing for a 10-min rest between each step. Following the bending tests, electrochemical stability was assessed under a 0.5C charge/1.0C discharge protocol within 2.5–4.3 V.

3. Results and discussion

Fig. 1 shows the structural and processing differences between a conventional FD electrode and the proposed CNT-CF electrode architecture. In conventional FD electrode (Fig. 1a), single-crystalline cathode particles are embedded within a PVDF binder matrix containing dispersed Super P carbon clusters. PVDF is electrically insulating, thus it contributes to less electronic conduction [34,35]. Electronic transport is therefore established through discrete conductive carbon junctions between adjacent active material particles, while continuous current extraction relies on the metallic Al foil [36]. In particular, the discrete nature of the conductive network further leads to localized contact resistance and current crowding at particle-particle interfaces [37]. In the absence of the current collector, the PVDF-Super P composite framework lacks both sufficient electronic connectivity and mechanical integrity to sustain a self-supporting electrode. Thus, foil integration is intrinsically required in PVDF-Super P electrode architecture.

To overcome this intrinsic limitation, a CNT-CF electrode was designed (Fig. 1b). In this configuration, an interconnected SWCNTs network directly bridges adjacent cathode particles, forming a continuous conductive scaffold that spans the entire electrode thickness. Unlike discrete carbon clusters relying on foil-supported current extraction, the percolated three-dimension nanotube framework distributes electronic pathways throughout the electrode, thereby suppressing localized current crowding and reducing contact-resistance concentration [38]. Simultaneously, the flexible SWCNTs network provides integrated mechanical cohesion, enabling a structurally self-supporting configuration without polymeric binder or metallic current collector [20,39]. The SWCNTs-based design could redefine the role of the conductive phase; rather than acting as a passive additive dispersed within a binder-rich matrix, the SWCNTs network becomes the primary structural and electrical backbone of the electrode. The fabrication route of CNT-CF electrode is presented in Fig. 1c. SC-NCM811 cathode powders were combined with a pre-dispersed SWCNTs solution and homogenized via planetary centrifugal mixing. This high-shear mixing process ensures uniform distribution of the SWCNTs network around the active material without the use of polymeric binder. And the mixture was subsequently cast onto a glass substrate and dried to obtain a binder-free, self-supporting electrode film. The CNT-CF electrode fabrication procedure is compatible with conventional slurry-casting methodology despite the elimination of metallic foil, demonstrating its engineering feasibility and scalability. Thus, the proposed CNT-CF architecture can be implemented in existing electrode fabrication routes.

The macroscopic mechanical integrity of the CNT-CF electrode is demonstrated in Fig. 1d. The CNT-CF electrode maintains structural integrity under various deformation modes, including 180 ° bending, 360 ° rolling, and twisting, without any visible cracking or delamination. Notably, even at the centimeter scale (5 × 8 cm), the electrode preserves continuous morphology and mechanical coherence during repeated deformation. This behavior highlights the effectiveness of the interconnected SWCNTs network in uniformly distributing mechanical stress throughout the electrode. Unlike conventional FD architectures, where mechanical strain is concentrated at the rigid current collector interface, the CNT-CF electrode accommodates deformation through its flexible, percolated nanotube framework. This enables simultaneous

mechanical resilience and electronic continuity, ensuring stable conductive pathways under mechanical stress.

Fig. 2 presents microstructural and surface-sensitive analyses that validate the fundamentally different conductive network architectures between the FD electrode and the CNT-CF electrode. In the FD electrode, discrete Super P carbon agglomerates are localized at particle-particle junctions (Fig. 2a, c and Fig. S1a), and EDX elemental mapping further reveals spatial overlap between carbon and fluorine signals (Fig. 2b), indicating that conductive carbon clusters are embedded within PVDF binder domains. This segregated distribution of conductive carbon within PVDF domains results in spatially confined and heterogeneous electronic contact between the conductive carbon and oxide particles, thereby inherently limiting effective charge transport. In contrast, the CNT-CF electrode exhibits a fundamentally different morphology, where SWCNTs conformally wrap individual SC-NCM811 particles and continuously bridge neighboring particles, forming an interconnected three-dimensional percolating network (Fig. 2d, f and Fig. S1b). Unlike the localized carbon clusters observed in the FD electrode, the nanotube network establishes line-contact-type conductive pathways that extend throughout the electrode. Also, elemental mapping confirms a homogeneous carbon distribution without detectable fluorine signal (Fig. 2e), consistent with a binder-free architecture and uniform SWCNTs dispersion. This structural distinction indicates that the SWCNTs network is spatially distributed to maximize interparticle connectivity and enlarge the effective electrochemical contact area across the electrode.

Surface-sensitive XPS analysis further reveals these structural differences (Fig. 2g, h). In the FD electrode, a pronounced F 1s peak ($\sim$ 687 eV) confirms the presence of PVDF binder, while the O 1s spectrum is dominated by lattice oxygen (TM–O), indicating substantial exposure of the oxide surface [40]. The C 1s signal consists primarily of C–C contributions associated with conductive additives and binder domains. By contrast, the CNT-CF electrode exhibits a negligible F 1s signal due to the absence of PVDF. The C 1s spectrum is dominated by a strong sp²-rich C–C/C=C peak centered at $\sim$ 284.5 eV, consistent with graphitic SWCNTs conformally covering the particle surfaces [41]. In addition to the dominant sp² carbon signal, a minor C–O contribution is observed in the CNT-CF electrode surface, which could be contributed by surface functional groups on SWCNTs introduced during dispersion and processing (Fig. S2). Notably, the relative intensity of lattice oxygen is reduced in the CNT-CF electrode compared to the FD electrode. Considering the shallow probing depth of XPS, this attenuation indicates that the outermost cathode surfaces are preferentially covered by the high-aspect-ratio SWCNTs (> 10,000) network, effectively screening the underlying oxide signal (Fig. 2h, Fig. S3). [42]. Furthermore, in Raman spectroscopy analysis, the CNT-CF electrode exhibits pronounced D ($\sim$ 1350 cm⁻¹) and G ($\sim$ 1599 cm⁻¹) bands characteristic of graphitic SWCNTs [43], whereas the FD electrode shows significantly weaker graphitic signatures. The strong G-band intensity is consistent with the dominant sp² character observed in C 1s spectra, corroborating the conformal surface localization of the nanotube framework. This enhanced surface carbon signature is observed despite the lower carbon content in the CNT-CF electrode (1.5 wt% SWCNTs) compared to the FD electrode (3 wt% Super P + 3 wt% PVDF), indicating that the observed spectroscopic response originates from conformal nanotube coverage rather than bulk carbon content alone (Fig. S4). Overall, the structural and spectroscopic analyses consistently indicate that the CNT-CF electrode possesses a fundamentally different conductive architecture from the FD electrode: rather than relying on isolated particulate contacts within a binder matrix, it is defined by a continuous carbon network that intimately covers the NCM cathode surface and bridges neighboring particles.

The SWCNTs content was optimized at 1.5 wt% by balancing electrical conductivity and volumetric energy density (Table S1, S2). We then examined how the percolating SWCNT framework, which provides conformal surface coverage and enhanced interparticle connectivity,

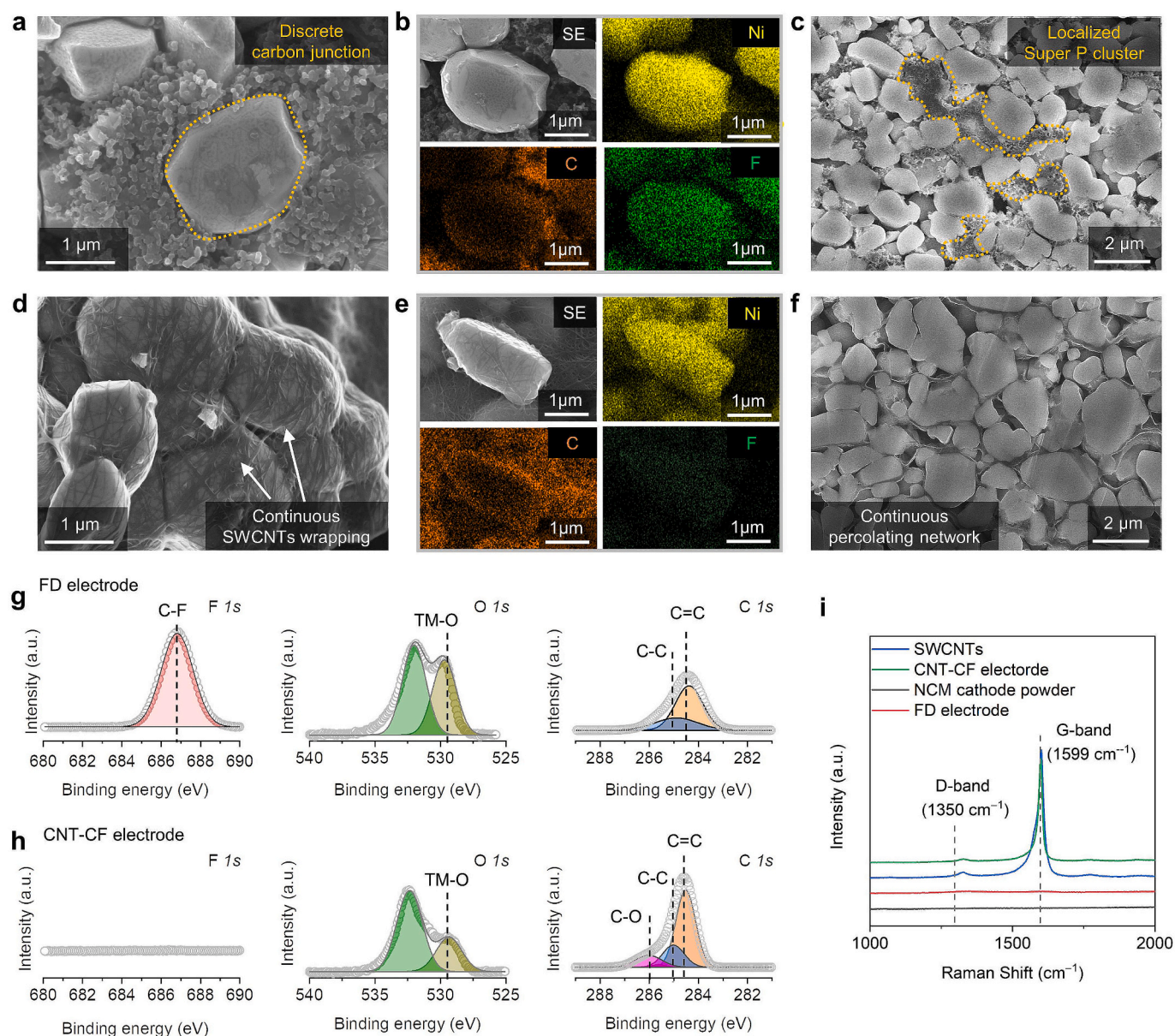


Fig. 2. Morphological, elemental, and surface characterizations of the FD and CNT-CF electrodes: (a) SEM image, (b) EDS elemental mapping, and (c) cross-sectional SEM image of the FD electrode; (d) SEM image, (e) EDS elemental mapping, and (f) cross-sectional SEM image of the CNT-CF electrode. (g,h) XPS spectra (F 1 s, O 1 s, C 1 s) of the FD and CNT-CF electrodes, respectively. (i) Raman spectra comparing SWCNTs, CNT-CF electrode, NCM cathode powder, and FD electrode.

affects charge-transport characteristics and electrochemical performance (Fig. 3). As shown in Fig. 3a and Table S3, the electrode containing 1.5 wt% SWCNTs exhibited significantly higher electrical conductivity than the electrodes containing 3–15 wt% Super P. Unlike the discrete point-contact pathways formed by particulate Super P additives, the line-contact interactions between SWCNTs and active material particles provide continuous electron-transport pathways even at substantially lower carbon contents [44]. Consequently, the reduced fraction of inactive conductive material contributed to a significant enhancement in volumetric energy density (Table S4). To evaluate the structural stability of the SWCNT percolating network during electrode densification, SEM analyses were conducted before and after calendaring (Fig. S5). The electrode thickness decreased from 71.3 μm to 32.6 μm , corresponding to a densification of approximately 54%. Despite the substantial compression, the interconnected SWCNT framework remained observable between active material particles without evidence of network fracture. The improved conductive network also enhanced its cycling stability. The CNT-CF electrode maintained higher discharge

capacity and cycling stability than the FD electrode over 200 cycles of 0.5C charge/1.0C discharge within the voltage range of 2.8–4.3 V cycling test (Fig. 3b and Fig. S6, at 25 °C, $1\text{C} \approx 200\text{ mA g}^{-1}$). The CNT-CF electrode continuously maintained superior capacity retention compared with the FD electrode throughout the long-term cycling period. In addition, the electrochemical performance was further evaluated under a practically relevant high areal mass loading exceeding 15 mg cm^{-2} (Fig. S7). Even at increased loading level, the CNT-CF electrode maintained stable cycling behavior and superior capacity retention compared with the FD electrode. Also, to exclude the effect of different active material fractions, the rate capability was further evaluated on an areal-current-density basis. Consistently, the CNT-CF electrode exhibited higher specific capacities than the FD electrode at all tested rates from 0.99 mA cm^{-2} to 9.90 mA cm^{-2} at intervals of 1.98 mA cm^{-2} in current density. After the current rate returned to 0.99 mA cm^{-2} , the CNT-CF electrode also exhibited higher recovery capacity (Fig. 3c and Fig. S8). Although the CNT-CF electrode contains a slightly higher active material fraction than the FD electrode, the CNT-CF electrode still

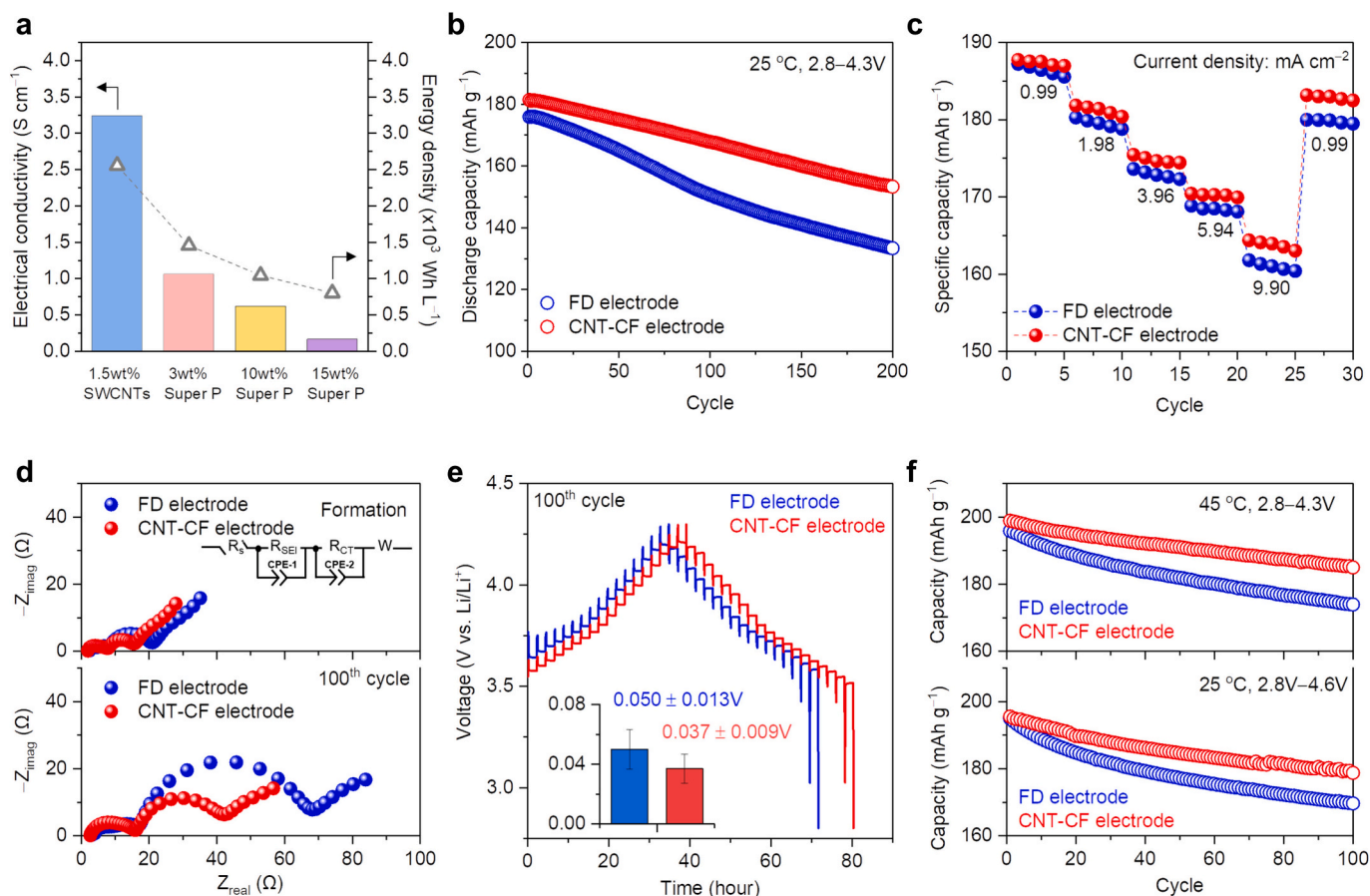


Fig. 3. Electrochemical comparison of FD and CNT-CF electrodes. (a) Electrical conductivity and volumetric energy density of electrodes containing 1.5 wt% SWCNTs and 3, 10, and 15 wt% Super P. (b) Cycling performance of FD and CNT-CF electrodes at 25 °C under 0.5C charge and 1C discharge for 200 cycles in the voltage window of 2.8–4.3 V versus Li/Li⁺. (c) Rate capability of FD and CNT-CF electrodes at 25 °C under discharge current densities of 0.99, 1.98, 3.96, 5.94, and 9.90 mA cm⁻², followed by 0.99 mA cm⁻² (2.8–4.3 V versus Li/Li⁺). (d) Nyquist plots of FD and CNT-CF electrodes after formation (top) and after 100 cycles (bottom). (e) GITT profiles of FD and CNT-CF electrodes after 100 cycles; inset shows the average ohmic voltage loss (IR drop) extracted from the corresponding GITT steps. (f) Cycling performance of FD and CNT-CF electrodes at 45 °C (top, 2.8–4.3 V versus Li/Li⁺) and at high voltage of 2.8–4.6 V at 25 °C (bottom) (1C ≈ 200 mA g⁻¹).

delivered higher capacity retention at comparable areal current densities. This result indicates that the improved rate capability originates primarily from the continuous SWCNT-mediated electronic percolation rather than from the active-mass difference alone. To gain a better understanding of the improved cycling stability and rate capability, we conducted EIS and GITT analyses after formation and 100 cycles. As shown in the Nyquist plots in Fig. 3d and Table S5, a larger charge-transfer resistance developed in cycled FD electrode than in cycled CNT-CF electrode. This result was further supported by the GITT analysis, in which the CNT-CF electrode exhibited a smaller polarization than the FD electrode at the 100th cycle, and the average voltage loss was ~27% smaller in the former (Fig. 3e). To quantitatively compare ionic transport behavior, the apparent D_{Li^+} were calculated from the GITT transients using the standard method based on transient and steady-state voltage responses. The CNT-CF electrode exhibited higher and more stable D_{Li^+} values than the FD electrode over a broad SOC range after cycling, indicating that the SWCNT-mediated percolating network effectively mitigates transport degradation and preserves electrochemical reaction uniformity during repeated cycling (Fig. S9). A more detailed analysis of the GITT response in Fig. S10 further suggests that the cycled CNT-CF electrode undergoes limited growth in voltage loss. By contrast, the FD electrode exhibits substantial growth in the form of ohmic loss, indicating severe degradation in electron transport at the electrode level. These results suggest that the more uniform conductive framework of the CNT-CF electrode not only improves initial

electron transport but also suppresses the gradual evolution of transport limitations during repeated cycling.

The benefit of the CNT-CF architecture became more evident under harsher operating conditions. At 45 °C, within the voltage range of 2.8–4.3 V, the CNT-CF electrode maintained a higher capacity than the FD electrode during extended cycling (Fig. 3f and Fig. S11). Also, even in the widened voltage range of 2.8–4.6 V, the CNT-CF electrode still exhibited improved cycling stability (Fig. 3f and Fig. S12). Under both thermal and voltage stress, parasitic reactions and structural instability are generally amplified [45]. This indicates that the enhanced stability of the CNT-CF electrode indicates that the SWCNTs network not only improves bulk conduction but also mitigates degradation pathways under aggressive operating conditions. The continuous SWCNTs framework enables effective electron transport at low carbon content and suppresses impedance growth and polarization during prolonged cycling. Such multifunctional contributions underlie the superior electrochemical performance of the CNT-CF electrode over the FD electrode architecture.

To elucidate how electrode architecture governs degradation pathways during cycling, we performed a comprehensive post-mortem analysis integrating morphological, structural, and surface-chemical characterization (Fig. 4). After 100 cycles, the cycled FD electrode exhibits a particle-level degradation, including severe surface roughening and intragranular cracking (Fig. 4a). In contrast, cathode particles in the CNT-CF electrode well preserve their structural integrity, with minimal

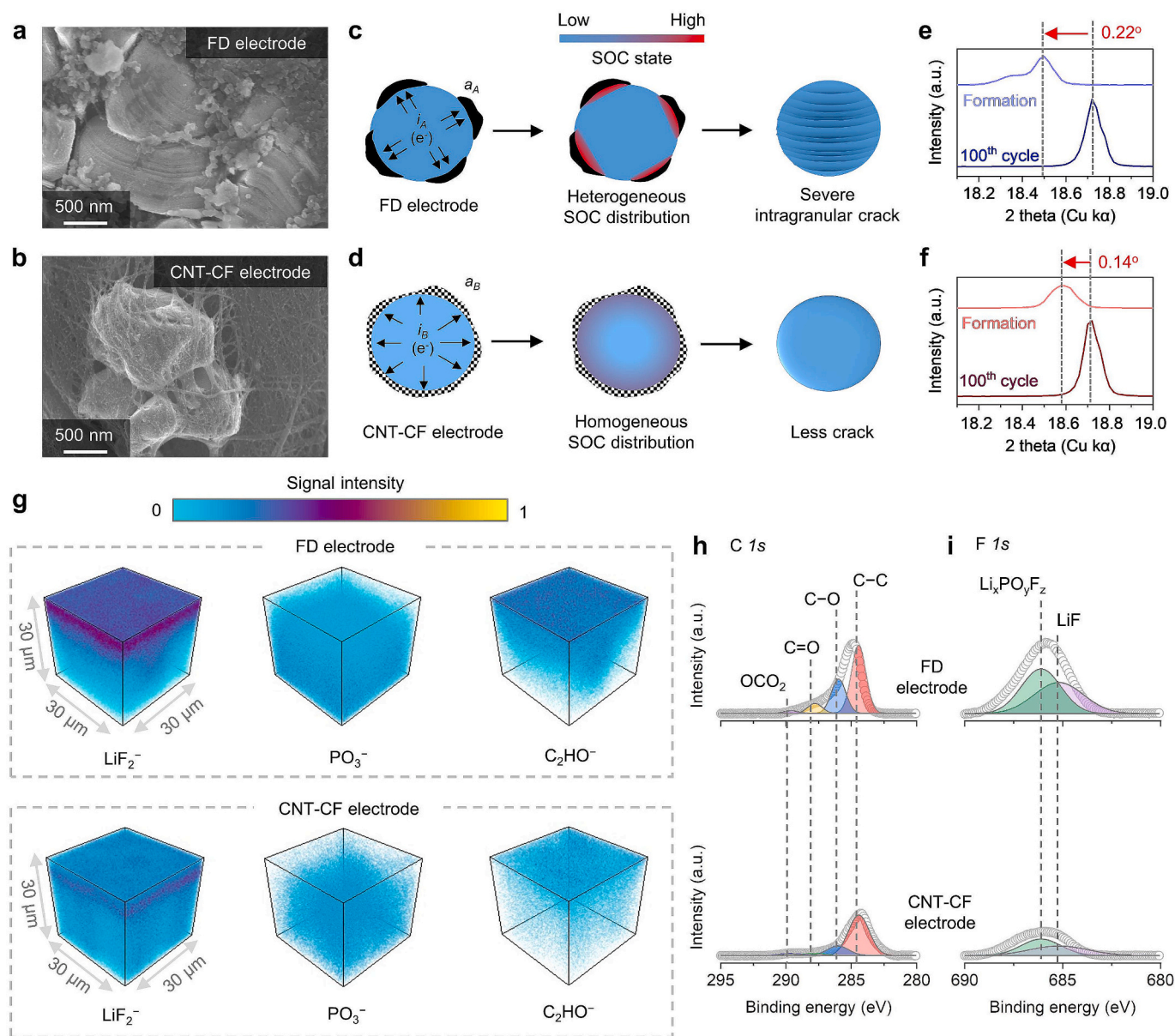


Fig. 4. Structural and chemical analyses of the electrodes after cycling: (a,b) SEM images of the FD and CNT-CF electrodes after 100th cycle. (c,d) Schematic illustrations of the state-of-charge (SOC) distribution in the FD and CNT-CF electrodes during cycling. (e,f) XRD patterns of the FD and CNT-CF electrodes before cycling and after the 100th cycle. (g) Three-dimensional TOF-SIMS reconstruction maps ($30 \times 30 \times 30 \mu\text{m}^3$) comparing the spatial distribution of representative CEI components in the FD (top) and CNT-CF (bottom) electrodes after the 100th cycle. (h,i) XPS spectra (C 1 s and F 1 s) of the FD (top) and CNT-CF (bottom) electrodes after 100th cycle.

fracture observed under identical cycling conditions (Fig. 4b). This divergence in particle integrity reflects fundamentally different electrochemical environments imposed by the two different architectures. In the FD electrode, the limited effective electrochemical contact area (a_A) concentrates the applied current (I) into a small active interfacial region, such that the local current density can be approximated as $i_A \approx I/a_A$ where I is the applied current. (Fig. 4c). The resulting local overpotential and Li concentration gradient promote heterogeneous evolution within the particle [46,47]. As a result, it amplifies internal lattice mismatch and stress concentration during repeated lithiation and delithiation, leading to intragranular crack formation becoming more severe. In contrast, the CNT-CF electrode provides a percolating conductive framework around the particle surface, allowing electrons to be supplied more uniformly across the active material surface with increased effective contact surface area (a_B) (Fig. 4d). Thus, the local current density ($i_B \approx I/a_B$) across cathode particle can be reduced, and it is

expected to reduce SOC heterogeneity and suppress localized stress accumulation, thereby mitigating crack initiation and propagation during cycling [24]. In other words, the SWCNTs network acts not only as an electronic conductor, but also as an electrochemically homogenizing framework that alleviates mechanical damage induced by repeated phase evolution [30]. To further investigate structural evolution during cycling, in-situ XRD measurements were conducted by tracking the (003) reflection of the layered structure (Fig. S13). The CNT-CF electrode exhibited a more gradual and continuous peak evolution than the FD electrode, especially in the H2–H3 transition region. This observation suggests a more homogeneous electrochemical reaction environment enabled by the percolating SWCNT network. Such a homogeneous structural evolution is consistent with the significantly reduced intragranular cracking observed after cycling. Consistent with these observations, ex-situ XRD analysis after 100 cycles reveals a larger peak shift in the FD electrode compared to the CNT-CF electrode

(Fig. 4e, f), indicating more significant irreversible structural evolution. The reduced peak displacement in the CNT-CF electrode suggests that lattice distortion and structural degradation are effectively alleviated.

The difference in bulk damage was further reflected in the spatial distribution of side reaction products. TOF-SIMS reconstruction map and depth profiling showed that the FD electrode accumulated more pronounced surface-derived species after cycling, including LiF-related, phosphate-containing, and organic fragment signals, than the CNT-CF electrode (Fig. 4g and Fig. S14, 15). Moreover, these species were more heterogeneously distributed in the FD electrode, implying that parasitic interfacial reactions occurred more actively and less uniformly across the cycled electrode [48,49]. In contrast, the CNT-CF electrode exhibited much weaker and more uniform distributions of side reaction products, indicating that interfacial side reactions were effectively suppressed. This trend was further confirmed by the normalized TOF-

SIMS depth-profile peak areas in Table S6, where LiF_2^- , PO_3^- , and C_2HO^- signals were all substantially lower for the CNT-CF electrode than for the FD electrode. Considering the much less cracking observed in the CNT-CF electrode, this difference strongly suggests that SWCNTs can suppress additional exposure of reactive surfaces and mitigates electrolyte penetration into the particle interior, thereby decelerating electrolyte decomposition and interphase growth. XPS analysis provides further evidence for this interpretation (Fig. 4 h, i). In the cycled FD electrode, the C 1 s spectrum showed stronger contributions from oxygenated carbon species such as C—O, C=O, and OCO_2 components, consistent with increased formation of organic decomposition products on the cycled FD electrode surface. Moreover, the F 1 s spectrum displayed stronger LiF and $\text{Li}_x\text{PO}_y\text{F}_z$ features, indicating more extensive salt decomposition and inorganic interphase formation [50]. By contrast, the CNT-CF electrode exhibited weaker intensities for these products,

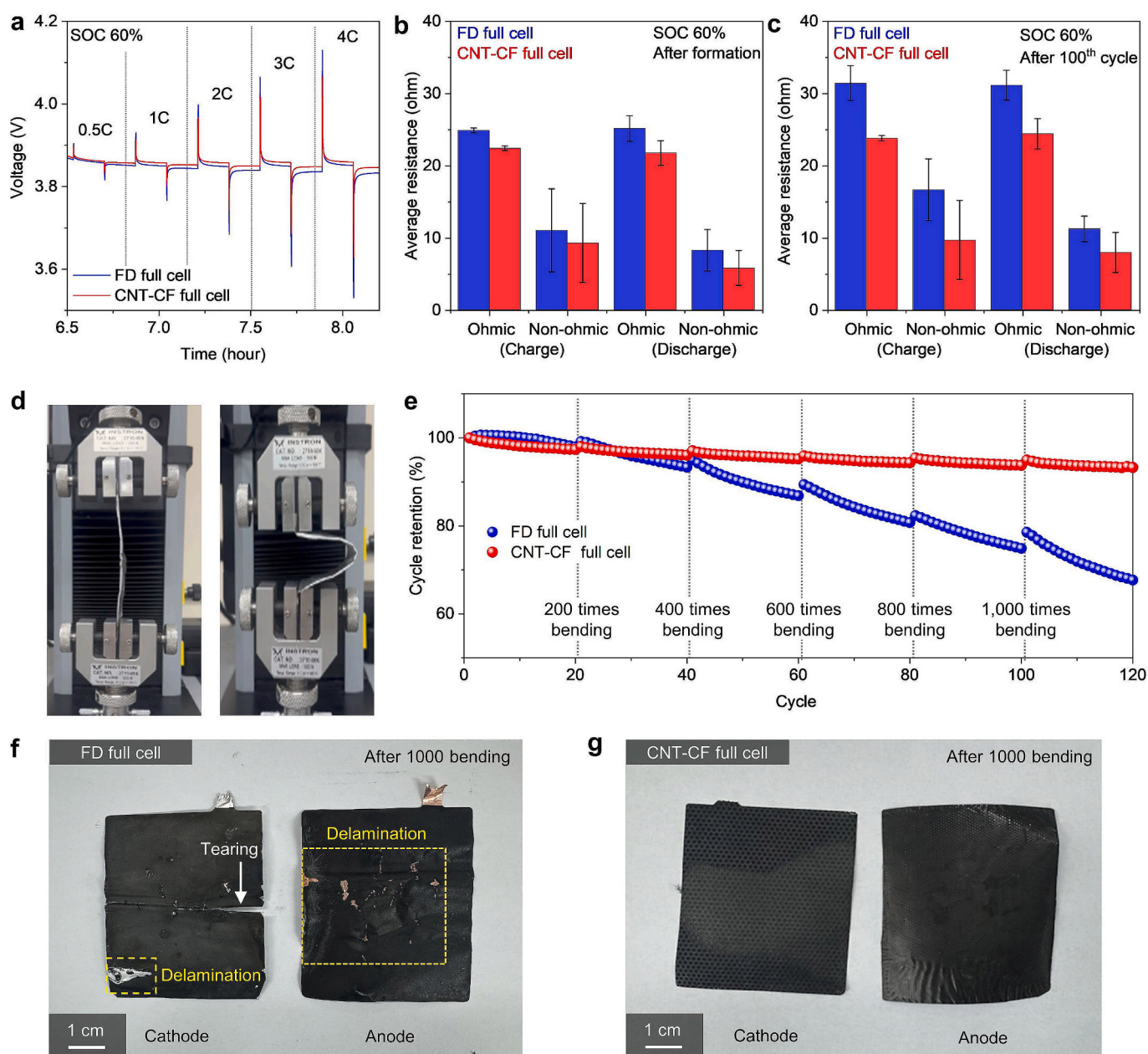


Fig. 5. Mechanical and electrochemical durability of FD and CNT-CF full cells: (a) Direct current internal resistance (DC-IR) voltage profiles of full cells. (b,c) Ohmic and non-ohmic resistance components derived from DC-IR after formation and after 100th cycle, respectively. (d) Photograph of the bending-test setup. (e) Cycling performance of full cells under repeated bending, measured every 20 cycles up to 1000 bending cycles. (f,g) Post-bending photographs of the FD and CNT-CF electrodes extracted from the full cells.

consistent with a thinner or less accumulated cathode-electrolyte interphase as observed by TOF-SIMS profiles. This interpretation is further supported by the normalized XPS peak areas in Table S6. The origin of reduced intraparticle cracking and side reaction products in CNT-CF can be ascribed to interconnected SWCNTs network. By promoting more homogeneous electronic access and reaction distribution, the SWCNTs network mitigates local stress concentration, weakens particle cracking, and reduces the degree of irreversible structural evolution during cycling. This mechanically stabilized particle state and suppressed the generation of additional reactive surfaces, thus limiting electrolyte-derived surface reconstruction and decomposition [51]. Therefore, the superior cycling stability of the CNT-CF electrode can be ascribed to a coupled structural–interfacial stabilization by the percolating SWCNTs framework.

The practical applicability of CNT-CF architecture is clearly confirmed in full cell performance and mechanical durability tests. In the CNT-CF architecture-based full cell (CNT-CF full cell) configuration, not only the cathode but also the anode was fabricated using an SWCNTs-based electrode design with areal capacities of ~ 2.12 and ~ 2.36 mAh cm $^{-2}$, respectively, corresponding to an N/P ratio of 1.11 ± 0.01 . (Table S7, S8). Notably, the elimination of metallic current collectors and polymeric binders enabled a higher fraction of electrochemically active components at the cell level, leading to an energy density of ~ 607 Wh L $^{-1}$ (including package: ~ 275 Wh L $^{-1}$) (Table S9). Firstly, the DC-IR of the full cells was measured at 60% SOC by applying current pulses from 0.5 to 4.0C for 10 s with a 10 min relaxation period between steps (Fig. 5a). Even after formation, the CNT-CF full cell exhibited consistently smaller voltage drops than the conventional FD architecture-based full cell (FD full cell) throughout the pulse sequence, indicating lower internal resistance. Quantitative separation of the DC-IR components further showed that both ohmic and non-ohmic resistances during charge and discharge were much lower in the CNT-CF full cell (Fig. 5b and Fig. S16). These results indicate that the CNT-CF electrode pair provides more efficient charge transport and reduced polarization under practical full cell operation. The difference became more increased after prolonged cycling. After the 100th cycle, the FD full cell exhibited a substantial increase in both ohmic and non-ohmic resistance, whereas the CNT-CF full cell still maintained markedly lower resistance values (Fig. 5c and Fig. S17). The suppressed resistance growth in CNT-CF full cell can be ascribed to enhanced structure integrity with suppressed electrolyte decomposition along the electrode surface. This behavior arises not only from the cathode but also from the percolating SWCNTs network in the anode, which maintains continuous conductive pathways and stable interfacial contact during repeated cycling (Fig. S18). At the full cell level, these benefits appear together with the mechanically stable SWCNTs-based architecture anode, enabling the cell to preserve conductive continuity and interfacial stability more effectively during repeated operation.

To assess mechanical robustness at the pouch-cell level, pouch-type full cells were subjected to repeated bending using the setup shown in Fig. 5d and Video S1. Electrochemical stability after bending was then evaluated under a 0.5C charge/1.0C discharge protocol in the same 2.5–4.3 V voltage window. As shown in Fig. 5e, the FD full cell exhibited dramatic capacity decay as bending accumulated, whereas the CNT-CF full cell maintained more stable cycling behavior even after 1000 bending cycles with repeated 100 charge-discharge cycles. The structural origin of enhanced structural and electrochemical properties can be more clarified in post-mortem analysis after 1000 bending cycles (Fig. 5f, g). In the FD full cell, both the cathode and anode showed mechanical damage including tearing and severe delamination from the current collector. By contrast, both electrodes extracted from the CNT-CF full cell relatively well preserved their structural integrity without obvious tearing or large-area delamination. These observations indicate that the superior bending durability of the CNT-CF full cell does not arise from a single electrode component alone, but from the mechanically integrated behavior of the CNT-CF based cathode and anode that

prevents electrode tearing and delamination [52,53]. To verify the reproducibility of cell assembly and electrode pairing, three independently assembled full cells were evaluated for each configuration under an identical N/P ratio of 1.11. All CNT-CF full cells exhibited highly consistent cycling behavior with capacity retention above 92% after 120 cycles, whereas all FD full cells showed substantially accelerated degradation, confirming the reproducibility of the observed performance difference (Fig. S19). The advantages of the CNT-CF architecture extend beyond half-cell demonstration and beyond cathode-only implementation. The SWCNTs framework serves as a multifunctional scaffold that maintains interparticle contact, electrical continuity and improved tolerance to mechanical stress in both electrodes during repeated deformation. These results highlight the practical potential of CNT-CF electrode pairs for flexible and mechanically durable LIBs.

4. Conclusion

In this study, we developed a CNT-CF electrode architecture that functions not only as a conductive-additive replacement, but also as a multifunctional framework for electron transport, mechanical integration, and degradation mitigation. Unlike the conventional FD electrode, where discrete carbon contacts and binder-rich domains limit electronic percolation and concentrate local electrochemical reactions, the CNT-CF design forms a continuous three-dimensional network that uniformly bridges active particles without metallic current collectors or polymeric binders. Unlike many previously reported free-standing or binder-free electrodes that rely on pre-formed carbon substrates or batch-type assembly routes, the present work adopts a manufacturing-relevant slurry-casting and drying process that is compatible with conventional coating-based fabrication. This architecture enabled higher conductivity at lower inactive content, leading to improved rate capability, cycling stability, and reduced polarization. More importantly, the SWCNTs network increased the effective electrochemical contact area, alleviated local current concentration, and suppressed heterogeneous SOC evolution during cycling. As a result, the CNT-CF electrode exhibited less severe intragranular cracking, smaller irreversible structural change, and reduced accumulation of electrolyte-derived surface species after prolonged cycling. These results indicate that the SWCNTs framework stabilizes not only charge transport, but also the coupled chemo-mechanical and interfacial degradation processes of layered oxide cathodes. The practical relevance of this strategy was further validated in full cells employing CNT-CF cathode and anode electrodes, which delivered lower internal resistance and superior bending durability. This work presents a viable route toward lightweight, flexible, and high-energy-density LIBs.

Authorship contribution statement

Jungmin Shin: Idea formulation, data accumulation, and initial blueprint preparation; **Seungmi Lee:** Idea formulation and Full cell assembly; **Jieun Kim:** Electrochemical testing; **Youngmin Kim:** conduct in-situ XRD measurements and analysis; **Jinwoo Park:** Experiment design and supervision; **Moonsu Yoon:** Supervision, resources, and writing–review & editing.

CRediT authorship contribution statement

Jungmin Shin: Writing – original draft, Data curation, Conceptualization. **Seungmi Lee:** Investigation, Data curation. **Jieun Kim:** Investigation, Validation. **Youngmin Kim:** Investigation, Methodology. **Jinwoo Park:** Methodology, Investigation. **Moonsu Yoon:** Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial

interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.cej.2026.179642>.

Data availability

Data will be made available on request.

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