

A Solvent-Free, Dry-Processed Li-Ion Battery Enabled by Dual Binders and Nanostructured Aluminum Current Collectors

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Conventional dry-processed electrode (DPE) fabrication using polytetrafluoroethylene (PTFE) or polyvinylidene fluoride (PVDF) offers a solvent-free alternative to wet-coating processes that rely on toxic *N*-methyl-2-pyrrolidone. However, traditional DPE systems often struggle with a trade-off between internal cohesion and interfacial adhesion, limiting electrode integrity and electrochemical performance under high-loading conditions. Herein, an advanced dual-binder DPE (DB-DPE) architecture that integrates PTFE and PVDF with a nanostructured aluminum current collector (NSA) is presented. This synergistic system leverages PTFE's fibrillation-induced cohesion and PVDF's adhesive strength, while the NSA's nanoporous morphology enables robust mechanical interlocking at the electrode–collector interface. The resulting electrodes, fabricated with an ultrahigh active material content of 96 wt.% and minimal binder content of 2 wt.%, exhibit reduced interfacial resistance, excellent mechanical integrity, and enhanced ionic and electronic conductivity. This architecture enables the fabrication of high-mass-loading cathodes (up to 64 mg cm^{−2}, 12.5 mAh cm^{−2}) with superior performance, achieving a high volumetric energy density of 712.7 Wh L^{−1}.

1. Introduction

Lithium-ion batteries (LIBs) have become the dominant energy storage technology for applications ranging from portable electronics to electric vehicles and grid-scale storage, owing to their high energy density and long cycle life.^[1–3] As the demand for higher energy density increases, the development of thick, high-loading electrodes has become crucial.^[4–6]

In conventional LIB manufacturing, the slurry-coated electrode (SCE) method is widely employed, where active materials, conductive additives, and binders are dispersed in a solvent and coated onto a current collector. *N*-methyl-2-pyrrolidone, the solvent commonly used, necessitates a dedicated recovery system, consuming considerable energy, space, and time.^[7,8] Moreover, increasing electrode thickness often leads to challenges such as cracking and binder migration to the electrode surface, hampering the realization of high-energy-density configurations.^[9–11]

To overcome these limitations, dry-processed electrodes (DPEs) have emerged as a promising alternative. DPEs eliminate solvent use entirely, enabling direct deposition of dry components onto the current collector. This approach significantly reduces both energy consumption and processing time by removing the need for solvent recovery and drying. Among dry-processing techniques, binder fibrillation and powder compression are the most extensively studied.^[12–14]

From the perspective of binder design, polytetrafluoroethylene (PTFE) is widely adopted in DPEs because shear-induced fibrillation forms an extended fibrous network that imparts mechanical cohesion even at very low binder contents.^[14–16] Yet, PTFE's intrinsically nonpolar nature leads to poor adhesion with aluminum (Al) current collectors, often causing interfacial delamination and compromising reliability.^[17] To mitigate this, carbon-coated current collectors have been introduced to enhance interfacial bonding.^[18,19] However, these coatings are typically over 1 μm thick, electrochemically inactive, and consequently reduce the overall specific energy of the electrode.

To address the weak interface between the DPE composites and the current collector, powder compression with

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DOI: 10.1002/adfm.202518597

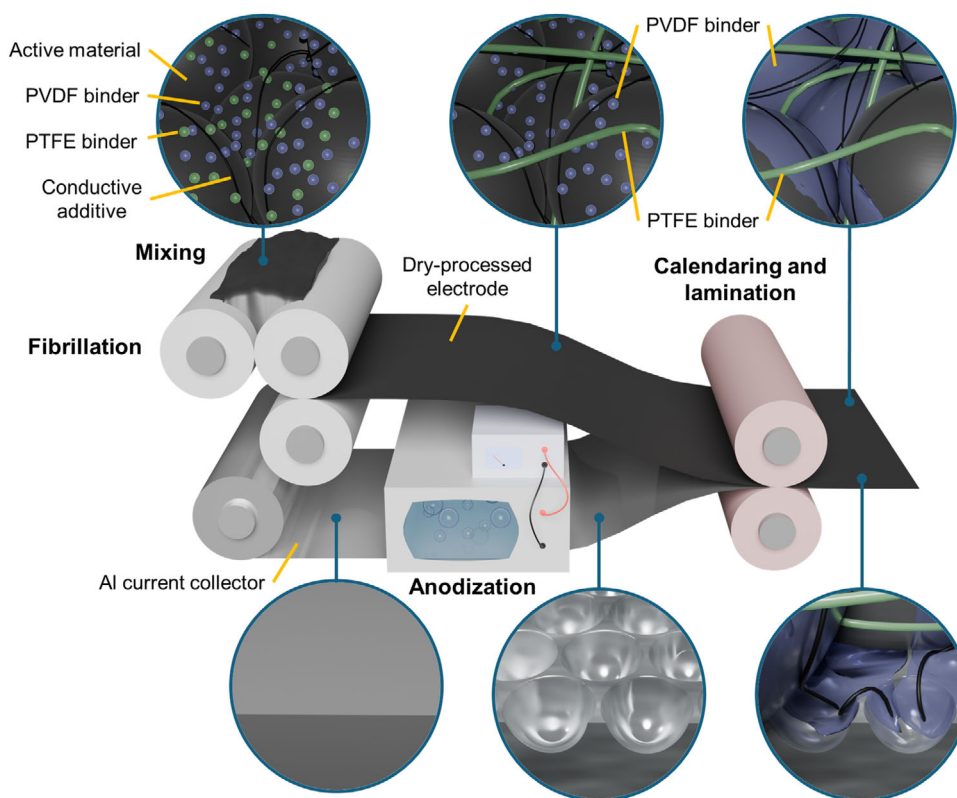


Figure 1. Schematic illustration of the fabrication process and structural configuration of the dual-binder dry-processed electrode (DB-DPE) with nanostructured Al current collector (NSA).

polyvinylidene fluoride (PVDF) binders has been employed. PVDF, owing to the high dipole moment of its $-(CH_2-CF_2)-$ backbone, promotes intermolecular interactions and improves wettability with the native oxide layer present on current collectors, thereby enhancing initial adhesion and interfacial contact.^[20] Our group recently demonstrated a DPE using PVDF/multi-walled carbon nanotube (MWNT) composite as the active material host.^[12] Hot pressing at 180 °C thermally activated PVDF near its melting range, increasing chain mobility and interfacial wetting, and formed bonding at both particle–particle and particle–current collector interfaces, yielding strong adhesion and cohesion. However, when used as the sole binder, PVDF required elevated binder/carbon contents to ensure cohesion, lowering the active material fraction and resulting in thick electrodes with limited volumetric performance.

Beyond binder strategies, significant advances have been reported in current collector engineering, including crystal orientation tuning, surface texturing, and conductive coatings to improve electrochemical stability.^[21,22] These developments, however, have primarily targeted wet-processed electrodes, where solvent-assisted chemical wetting governs adhesion. In contrast, dry electrode adhesion relies almost entirely on mechanical bonding, demanding current collector designs tailored specifically for DPEs.^[23] Despite the rapid industrial momentum toward dry processing, systematic exploration of current collectors optimized for this paradigm remains scarce.

In this study, we report an adhesion-enhanced DPE architecture that integrates a dual-binder system (PTFE–PVDF) with a

nanostructured Al current collector (NSA) (Figure 1). The NSA is fabricated by a two-step process; 1) anodization of Al foil to create an upper vertically oriented Al_2O_3 nanotube array, 2) removing the upper layer for a nano-embossed porous surface. Concurrently, the dual-binder system combines PTFE's fibrillated fibrous network for cohesion with PVDF's polar functionality for adhesion, thereby addressing the inherent trade-off between the two. Even at ultrahigh active material loading (96 wt.%) and minimal binder content (2 wt.%), the NSA-based dual-binder DPEs (DB-DPEs) exhibit markedly improved interfacial contact, electrical conductivity, and electrochemical performance compared with bare Al counterparts. This study thus establishes a compact, mechanically robust, and high-active-material electrode platform that overcomes the limitations of prior DPE designs.

2. Results and Discussion

PVDF exhibits strong adhesion to current collectors but suffers from insufficient internal cohesion, making the electrode composite prone to mechanical failure. In contrast, PTFE forms a fibrous network that reinforces the internal cohesion of cathode particles but has poor adhesion to Al foils and limited electrolyte wettability due to its nonpolar nature. To address these complementary limitations, we developed a DB-DPE architecture combining PTFE and PVDF to synergically improve both interfacial adhesion and internal cohesion (Figure 2a).

A mixture of $LiNi_{0.7}Co_{0.1}Mn_{0.2}O_2$ (NCM712), MWNT, PTFE, and PVDF was kneaded on a hot plate at 80 °C and then

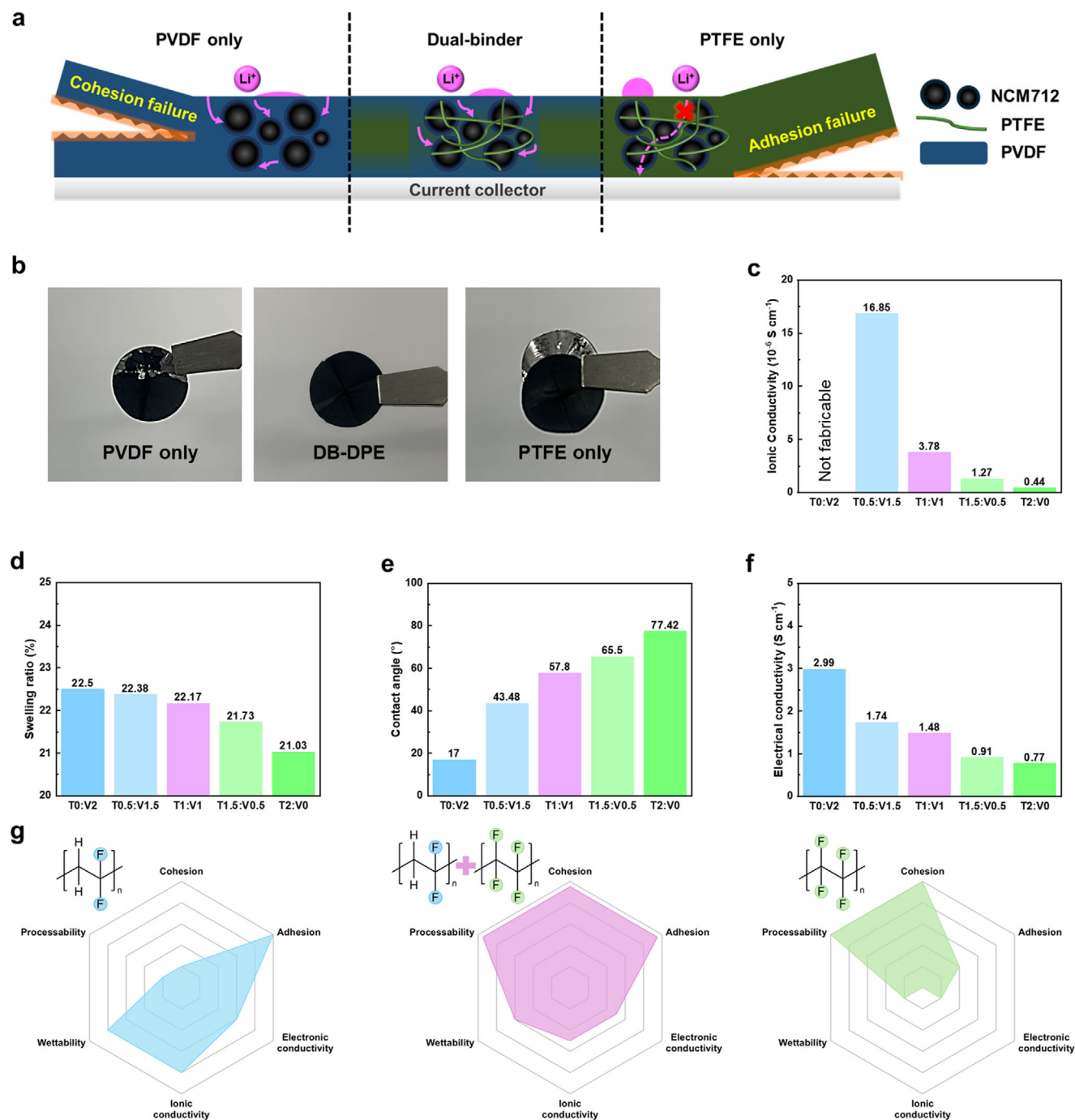


Figure 2. Optimization of DB-DPEs. a) Schematic illustration of the dual-binder strategy. b) Representative failure modes observed during the folding test of thick DPE. c) Ionic conductivity, d) swelling ratio, e) contact angle, and f) electrical conductivity of DB-DPEs. g) Radar chart summarizing overall binder performances for dry process based on metrics from Figure 2b–f.

calendared to achieve the target mass loading. The resulting free-standing film was hot-pressed at 180 °C, above the melting point of PVDF, to enhance binder softening, mechanical integration, and ensure contact with the current collector (see Experimental Section for details). To optimize the binder ratio, DB-DPEs were fabricated with varying PTFE:PVDF mass ratios of 0:2, 0.5:1.5, 1:1, 1.5:0.5, and 2:0. The corresponding samples were la-

beled T0:V2, T0.5:V1.5, T1:V1, T1.5:V0.5, and T2:V0, respectively (Figure S1, Supporting Information). Electrode integrity was first evaluated using a folding test. The T0:V2 sample exhibited poor cohesion within the electrode composite, while T0.5:V1.5 showed cracking upon bending (Figure 2b; Figure S2, Supporting Information). The severe electrode fracture observed in T0:V2 was attributed to the absence of PTFE fibrillation effect, which resulted

in a lack of internal cathode particle cohesion (Figure S3, Supporting Information). In contrast, when the PVDF content was below 0.5 wt.% (T1.5:V0.5 and T2:V0), the cathode electrodes delaminated from the bare Al, indicating insufficient adhesion to the current collector. The T1:V1 sample, however, maintained its structural integrity after two folding cycles, suggesting it achieved an optimal balance between cohesion and adhesion.

To further assess the mechanical robustness of each formulation under external stress, a 5-min ultrasonication test was conducted (Figure S4, Supporting Information). In this test, the electrolyte became visibly discolored in the T0:V2 and T0.5:V1.5 samples, indicating detachment and dispersion of active material particles into the electrolyte. In the T1.5:V0.5 and T2:V0 samples, the electrode composite detached from the current collector, further confirming poor interfacial stability under mechanical agitation. In contrast, the T1:V1 sample maintained its structural integrity, showing neither electrolyte discoloration nor delamination, thereby demonstrating superior mechanical robustness under these test conditions.

To investigate the effects of binder composition on ionic and electronic transport in DB-DPEs, ionic conductivity, swelling ratio, wettability, and sheet resistance were evaluated (Figure 2c–f). For ionic conductivity measurements, freestanding binder films were prepared by blending PTFE and PVDF at specific ratios and compressing them into uniform sheets suitable for electrochemical impedance spectroscopy (EIS) (Figure S5, Supporting Information). The ionic conductivity, calculated from the bulk resistance obtained via EIS, increased significantly with higher PVDF content (Figure 2c). The T2:V0 sample (PTFE-only) showed the lowest conductivity at $0.44 \times 10^{-6} \text{ S cm}^{-1}$, while the values rose progressively with PVDF incorporation. T1.5:V0.5 and T1:V1 exhibited conductivities of 1.27×10^{-6} and $3.78 \times 10^{-6} \text{ S cm}^{-1}$, respectively, indicating that even moderate PVDF content substantially improves ionic pathways. Although the T0.5:V1.5 showed the highest value ($16.85 \times 10^{-6} \text{ S cm}^{-1}$), its structural stability under electrolyte exposure was relatively poor. In contrast, the T1:V1 composition provided both adequate ionic conductivity and robust structural integrity, suggesting it represents a practical and well-balanced formulation for DB-DPEs. It is important to note, however, that the T0:V2 composition (PVDF-only) could not be processed into a freestanding film due to poor mechanical integrity in the dry fabrication process. This limitation highlights the complementary role of PTFE, which improves film formability and mechanical robustness. While PVDF enhances ionic conductivity, PTFE is essential for maintaining structural integrity and ensuring reliable processability in DB-DPEs.

The electrolyte swelling behavior and surface wettability of the DB-DPEs were evaluated to investigate how binder composition affects electrolyte interactions in dry-processed systems (Figure 2d,e). As PVDF content increased, the swelling ratio of the DB-DPEs gradually rose from 21.03% for T2:V0 (PTFE-only) to 22.50% for T0:V2 (PVDF-only) (Figure 2d; Figure S6, Supporting Information). Electrolyte wettability, evaluated by contact angle measurements, also improved with increasing PVDF content (Figure 2e; Figure S7, Supporting Information). The contact angle decreased from 77.42° (T2:V0) to 17.0° (T0:V2), confirming enhanced surface polarity and better electrolyte spreading. These trends reflect a greater affinity of PVDF for electrolyte, attributed to its polar functional groups. Among the compositions,

T1:V1 exhibited a moderate swelling ratio of 22.17%, indicating sufficient electrolyte uptake for ionic conduction while minimizing the risk of dimensional instability associated with excessive swelling.^[24] Additionally, T1:V1 showed an intermediate contact angle of 57.8° , which is adequate for electrolyte infiltration but avoids the potential over-wettability that may compromise electrode cohesion or cause binder redistribution.

The electrical conductivity of DB-DPEs was evaluated using a four-point probe method to understand how binder composition influences electronic transport (Figure 2f). As the PTFE content increased, the electrical conductivity generally decreased. Although T0:V2 (PVDF-only) exhibited the highest initial conductivity (2.99 S cm^{-1}), it failed to form a freestanding film due to poor mechanical cohesion, making it impractical for dry-processed fabrication. In contrast, PTFE-rich compositions such as T1.5:V0.5 and T2:V0 exhibited lower electrical conductivities (0.91 and 0.77 S cm^{-1} , respectively), due to the insulating and nonpolar nature of PTFE, which hinders efficient electron transport. The T1:V1 composition, however, achieved sufficiently high electrical conductivity (1.48 S cm^{-1}) while maintaining excellent film formability, mechanical cohesion, and compatibility with electrolyte. Its ability to retain both structure and conductivity after electrolyte exposure underscores the advantage of the 1:1 dual-binder system in providing stable and reliable performance.

To evaluate the effect of binder compositions, half-cells consisting of DB-DPEs and Li metal anodes were fabricated under an areal capacity of 10 mAh cm^{-2} (Figure S8, Supporting Information). The T2:V0 (PTFE-only) electrode exhibited severe polarization, whereas the T0:V2 (PVDF-only) electrode suffered from poor cohesion and consequently delivered reduced capacities. In contrast, the dual-binder electrode achieved both low polarization and stable discharge capacities. EIS conducted before cycling revealed that the T1:V1 electrode exhibited the smallest charge-transfer resistance, attributable to the superior ionic conductivity of PVDF. Furthermore, the T1:V1 electrode delivered the highest discharge capacity at 0.5C compared to the single-binder counterparts. These results clearly demonstrate that the dual-binder formulation effectively resolves the trade-off between cohesion and adhesion.

The comprehensive evaluation of each binder formulation was summarized in a radar chart (Figure 2g). T1:V1, with equal amounts of PTFE and PVDF, exhibited the most balanced performance across all critical parameters, including cohesion, adhesion, ionic/electronic conductivity, wettability, and processability. PVDF-rich formulations provided superior ionic conductivity and adhesion to the current collector, while PTFE-rich systems excelled in cohesion and processability. Only the DB-DPE system successfully integrated the strengths of both components. This well-balanced formulation makes T1:V1 the most practical and optimized binder system for high-performance DB-DPEs.

The NSA was fabricated through a two-step process consisting of anodization and subsequent etching (Figure 3a; see Experimental Section for details). Anodization was performed in 0.3 M oxalic acid under a constant voltage, producing a highly ordered anodized aluminum oxide (AAO) layer with a nanoporous structure. This oxide layer was then selectively removed by chemical etching in a mixed solution of chromic acid and phosphoric acid, exposing a thin and concave nanopattern on the Al surface. The resulting structure exhibited a uniformly arranged

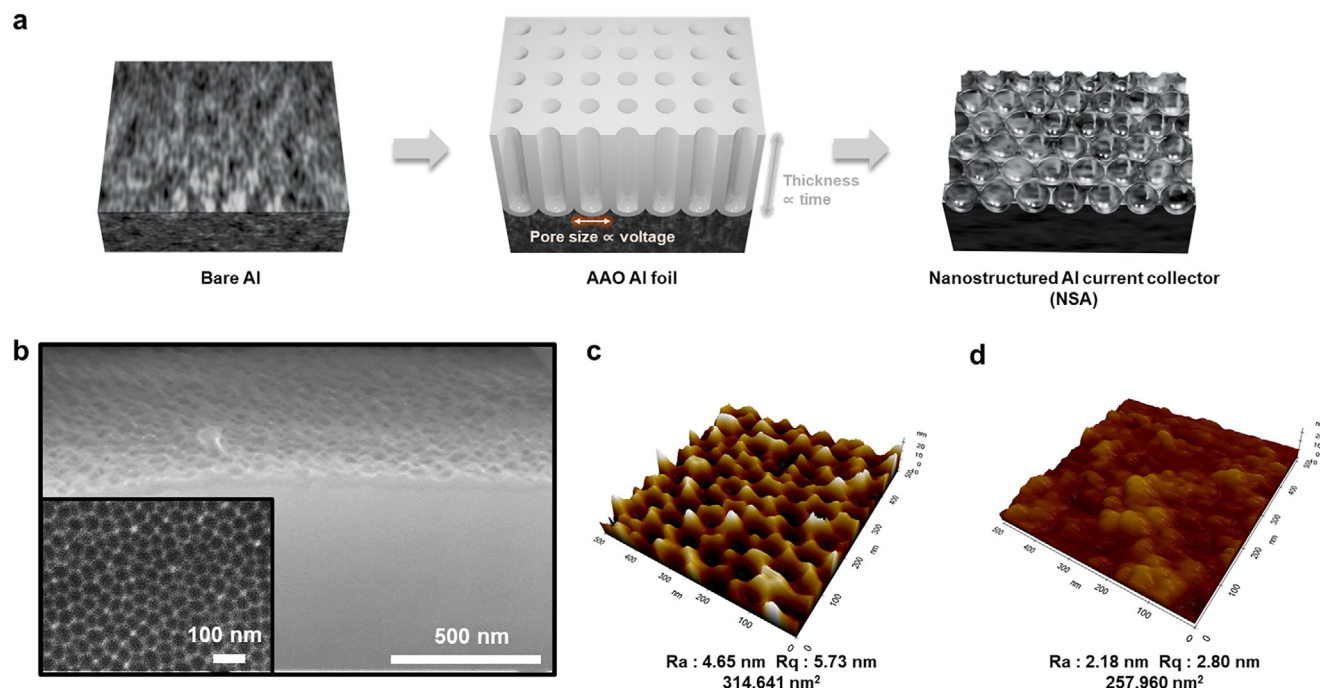


Figure 3. Surface characterization of NSA. a) Schematic illustration of the fabrication process of the NSA via anodization and chemical etching. b) Representative top-view and cross-sectional SEM images showing the ordered nanoconcave array on the NSA surface. c) AFM image of the NSA surface, revealing increased nanoscale roughness and surface area. d) AFM image of bare Al surface.

hexagonal array of nanoconcaves, as shown in the scanning electron microscopy (SEM) images (Figure 3b; Figure S9, Supporting Information).

To optimize the nanostructure for use in LIB current collectors, anodization was carried out under various durations and voltages. The aim was to determine the optimal parameters that would yield a nanostructured Al surface with the best combination of mechanical, electrical, and electrochemical properties. The anodization process was carried out for varying durations (from 5 s to 10 min) to investigate the formation and growth behavior of the tubular nanopore structures (Figure S10a, Supporting Information). A 5-s anodization time was insufficient for the sufficient nanopore development of the AAO layer, while anodization durations longer than 5 min resulted in minimal changes in pore structure, as observed in SEM images. This behavior can be attributed to the multi-step growth process of AAO, as demonstrated by the current density profile in Figure S10b (Supporting Information). Initially, a barrier oxide layer forms, leading to a rapid decrease in current density. As anodization progresses, the oxide barrier begins to crack, slowing the rate of current drop. Subsequently, the current density gradually increases due to pore nucleation and eventually stabilizes at a steady value, indicating steady-state nanopore growth.

Since the length of the AAO layer did not significantly affect the structural characteristics of the NSA, no notable morphological changes were observed beyond the nucleation phase. However, excessive anodization durations can weaken mechanical strength, negatively impacting electrode performances. Therefore, a 5-min anodization time was used in subsequent experiments, unless otherwise specified. For dry electrode composites, strong adhesion to the current collector is critical for maintain-

ing both mechanical and electrochemical stability. One theoretical approach to optimizing surface design is the interfacial shear stress model, expressed as $F_{\text{adhesion}} = \tau \cdot A_{\text{real}}$, where τ is the interfacial shear strength and A_{real} is the real contact area. According to this model, increasing the effective contact area between two heterogeneous solid surfaces enhances adhesion, primarily due to cumulative van der Waals interactions and mechanical anchoring effects at the interface.^[25–29]

SEM analysis revealed that increasing the anodization voltage from 20 to 50 V led to an increase in pore size, from 48.5 nm at 20 V to 147.7 nm at 50 V (Figure S11, Supporting Information). Atomic force microscopy (AFM) topography further confirmed the development of surface roughness on NSA, in contrast to the flat morphology of bare Al (Figure 3c,d; Figure S12a, Supporting Information). For a 500 × 500 nm² scan area, the bare Al surface exhibited a calculated surface area of 257 960 nm², nearly identical to its geometric area, indicating a smooth, featureless surface. In comparison, the NSA anodized at 30 V displayed a maximum surface area of 314 610 nm² within the same scan size (Figure S12b, Supporting Information). This corresponds to a 22% increase in the actual contact area, providing more active sites for current collector-binder interactions, thereby improving mechanical anchoring. Details of the surface area calculation method are provided in Figure S13 (Supporting Information).

The elemental composition and chemical states of the synthesized current collector surfaces were characterized by X-ray photoelectron spectroscopy (XPS). Figure S14 (Supporting Information) presents the Al 2p spectra of bare Al, anodized Al, and NSA. The peaks at 72.6 and 75.6 eV correspond to metallic Al and Al³⁺, respectively. In the anodized Al sample, the metallic Al peak is absent, indicating complete surface coverage by the

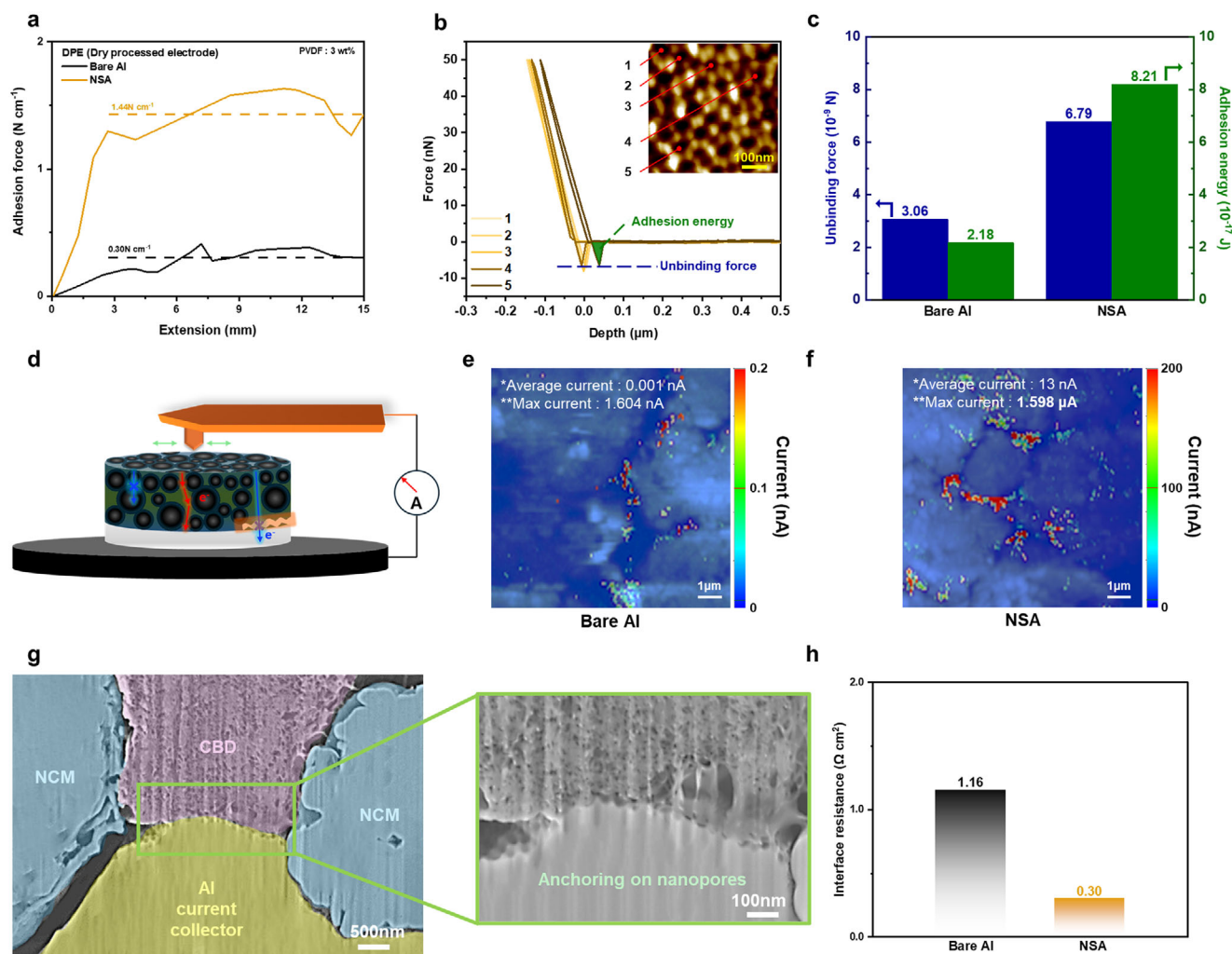


Figure 4. Analysis of adhesion force and interface electrical conductivity of DB-DPEs with various current collectors. a) Adhesion force between the DB-DPE composite and various current collectors. b) F-D curve obtained from NSA. c) Unbinding force and adhesion energy of various current collectors. d) Schematic of the C-AFM to evaluate the top-surface electrical conductivity of DB-DPEs. C-AFM current maps of e) bare Al-based DB-DPE and f) NSA-based DB-DPE. g) Cross-sectional FIB-SEM images of the NSA-based DB-DPE at different magnifications. h) Interface resistance of DB-DPEs with various current collectors.

oxide layer. In contrast, the NSA sample exhibits a metallic Al peak comparable to that of bare Al, confirming the successful removal of the AAO layer through chemical etching. Due to the high reactivity of metallic Al, a thin native oxide layer spontaneously forms upon air exposure.^[30] As a result, the surface chemical composition of NSA remains similar to that of bare Al. Nonetheless, the exposure of a large metallic Al surface area after etching is expected to facilitate improved charge transfer at the current collector–electrode interface, thereby enhancing electrochemical performance. To further confirm that the fabrication process does not compromise the structural robustness of the Al foil, tensile tests were performed on bare Al and NSA, which revealed negligible differences in mechanical strength (Figure S15, Supporting Information).

The peel test, a standard method for evaluating the mechanical properties of battery electrodes,^[31] measures the weaker of the two forces: internal cohesion within the electrode or its adhesion

to the current collector. In commercial-like formulations with high active material content and low binder ratio (e.g., 96:2:1:1 wt.% for NCM712:MWNT:PTFE:PVDF), insufficient cohesion led to premature failure within the composite layer (Figure S16, Supporting Information). Even when PTFE content was increased to 3 wt.%, the peel test still exhibited cohesion failure, preventing meaningful evaluation of interface adhesion (Figure S17a,b, Supporting Information). However, when PTFE content approached ≈ 5 wt.%, the cohesion within the electrode surpassed adhesion to the current collector. Under this condition, clear adhesion failure was observed at the interface (Figure S17c, Supporting Information), allowing reliable quantification of interfacial adhesive strength. Under the identical binder compositions (90:2:5:3 wt.% for NCM712:MWNT:PTFE:PVDF), NSA achieved an adhesion force (1.44 N cm^{-1}), more than four times greater than bare Al (0.38 N cm^{-1}) (Figure 4a), owing to the mechanical interlocking provided by its nanostructured surface. Notably,

similar enhancement was observed even in wet-processed electrodes (5 wt.% PVDF), indicating that the nanostructured surface improves interfacial integrity independent of the processing method (Figure S18, Supporting Information).

To evaluate the interfacial interactions between the electrode and current collector, force-distance (F-D) spectroscopy was performed using AFM. As shown in Figure 4b, F-D curves were collected from five representative positions across the NSA surface. The curves exhibited highly consistent profiles with minimal variation in both unbinding force and adhesion energy, indicating that the nanoscale pores provide uniform and reproducible mechanical anchoring across the surface. In contrast, the F-D curve measured on the bare Al surface is shown separately in Figure S19 (Supporting Information) for comparison.

Quantitative analysis (Figure 4c) further confirmed the enhanced interfacial adhesion on NSA. The unbinding force increased from 3.06×10^{-9} N (bare Al) to 6.79×10^{-9} N (NSA), while the adhesion energy increased approximately fourfold, from 2.18×10^{-17} to 8.21×10^{-17} J. These results demonstrate that the nanostructured surface of NSA not only increases the effective contact area but also strengthens and homogenizes the interfacial interactions, thereby improving the overall bonding stability of DB-DPEs.

As shown in Figure S20 (Supporting Information), electrodes laminated on NSA remained well attached under both cold and hot pressing, unlike those on bare Al, which showed poor adhesion, especially in the cold-pressed state. Additional evidence for enhanced interfacial contact was shown in the cross-sectional SEM image. The bare Al-based DB-DPE exhibited delamination at the interface, likely due to ion beam-induced damage during cross-sectioning, indicating weak interfacial bonding. In contrast, NSA-based DB-DPE maintained intimate interfacial contact attributed to the strong adhesion enabled by anchoring on nanostructured surface.

To understand the impact of interfacial adhesion on electrical conductivity in DB-DPEs, it is notable that unlike SCEs, where conformal contact minimizes interfacial resistance, DPEs rely on lamination, which often leads to poor interface contact.^[4,32] Therefore, detailed evaluation of the interfacial electronic pathways and contact resistance is essential to understanding and optimizing overall electrode performance in DB-DPE systems.^[33] Accordingly, we performed conductive AFM (C-AFM) on the surface of the DB-DPE, as shown in Figure 4d. The corresponding current maps demonstrate that electrical conduction predominantly occurs when the conductive AFM tip contacts the carbon-binder domain (CBD) rather than the active material itself (Figure 4e,f).^[34–36] This indicates that the MWNT and binder composite, which constitutes the CBD, forms the main conductive network within the thick electrode. MWNT offers superior conductivity, while the binder enhances mechanical stability and adhesion to the current collector. These observations highlight that, for electrodes fabricated by identical freestanding films, the key factor determining overall electrical conductivity is largely governed by the quality of the interface between the CBD and the current collector, rather than direct contact with the active material. Compared to the bare Al-based DB-DPE (average current: 0.001 nA, maximum current: 1.604 nA), the NSA-based DB-DPE showed more than 1000-fold increase in average current (13 nA) and a maximum current of 1.598 μ A (Figure 4e,f), confirm-

ing vastly improved interfacial conduction through nanopore anchoring.

Focused ion beam SEM (FIB-SEM) analysis revealed that both NSA and bare Al exhibited weak adhesion at the interface with NCM particles, consistent with previous reports indicating that NCM-metal adhesion is intrinsically poor and that effective interfacial bonding primarily occurs at the CBD-current collector interface (Figure 4g; Figures S21–23, Supporting Information).^[23,37,38] Closer inspection showed that the CBD network was well integrated into the nanopores of the NSA, while no such interfacial bonding was observed on the flat surface of bare Al. Furthermore, NSA-based DB-DPEs retained better composite adhesion when subjected to 1 h ultrasonication in the electrolyte, demonstrating superior mechanical integrity under external stress (Figure S24, Supporting Information).

Additionally, when both interface resistance and composite resistance were measured using specialized resistance equipment,^[39,40] the composite resistance and composite surface resistivity were nearly identical (Figure 4h; Figure S25 and Table S1, Supporting Information). However, the interface resistance at the electrode-current collector junction differed significantly, with the NSA interface showing approximately four times lower resistance than that of the bare Al, further validating the improved contact quality. Collectively, the results highlight that efficient charge transport in thick DB-DPEs is primarily governed by the interfacial quality between CBD and the current collector, rather than by direct contact with the active material.

The electrochemical performance of the DB-DPEs with various current collectors was evaluated over a range of high areal capacities from 5 to 12 mAh cm⁻² (Figure 5a–c). All cells exhibited initial discharge capacities approaching the theoretical value of NCM712 (195 mAh g⁻¹), with notable differences emerging at elevated current densities. The bare Al-based DB-DPE exhibited a marked capacity drop above ≈ 10 mA cm⁻², whereas the NSA-based DB-DPE retained over 80% of its theoretical capacity under the same conditions. To further probe electrode robustness, we included a recovery step at 0.3C after the rate test. The bare-Al based DB-DPE (12 mAh cm⁻²) showed significant capacity loss upon recovery, whereas the NSA-based DB-DPE retained nearly full capacity, confirming its structural stability under high-rate stress (Figure S26, Supporting Information). Additionally, as shown in dQ/dV plots (Figure S27, Supporting Information), the initial charge peak voltage for the NSA-based electrode (3.660 V) was slightly lower than that of the bare Al-based electrode (3.675 V), indicating lower polarization and faster electrochemical activity in the former. These results suggest that the NSA enhances charge-transfer kinetics, particularly at the upper regions of thick electrodes, as previously demonstrated in studies showing enhanced rate capability and reduced polarization through current collector surface modifications.^[19,41,42] Notably, the NSA-based SCE also outperformed the conventional bare Al-based SCE at high current densities, further highlighting the interfacial benefits of the nanostructured current collector even under slurry-based processing conditions (Figure S28, Supporting Information).

To verify the practical applicability of the DB-DPEs in a full-cell configuration, high-loading DB-DPEs (5 mAh cm⁻²) were

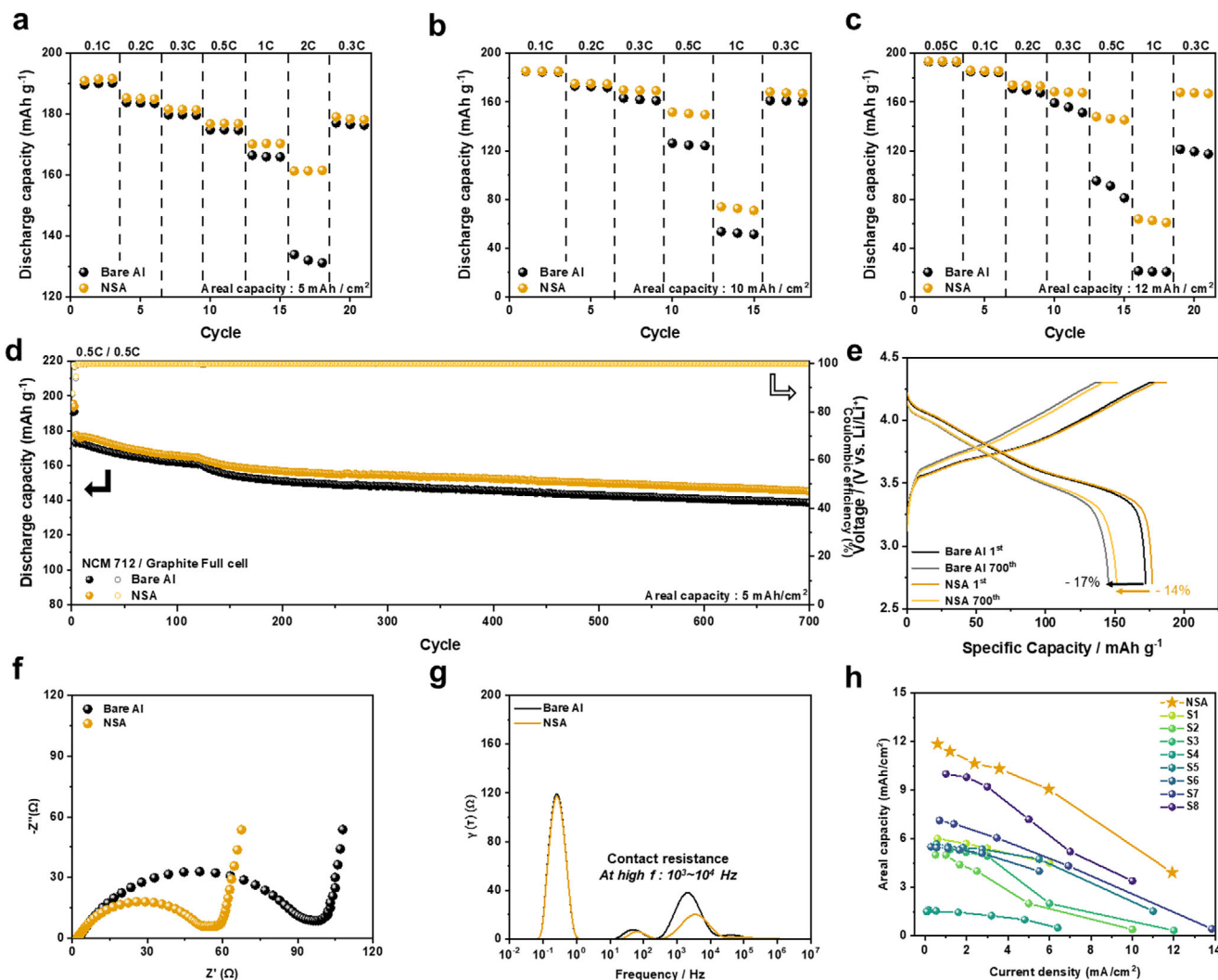


Figure 5. The electrochemical performance comparison of high-loading DB-DPEs with various current collectors. Rate capability with a) high and b), c) ultrahigh-loading DB-DPEs. d) High-loading NCM/graphite full cell cycle performance at charge/discharge current density of 0.5/0.5C at a voltage range of 2.7–4.3 V. e) Charge/discharge voltage profile comparison. f) Nyquist plots of DB-DPEs fabricated with various current collectors before cycling. g) Corresponding DRT analysis and EIS results. h) Comparison of areal capacity versus current density between this work and previously reported DPEs.

paired with high-loading graphite anodes (5.5 mAh cm⁻²) prepared via slurry coating, maintaining an N/P ratio (the ratio of the capacity of the negative electrode to that of the positive electrode) of 1.1. As shown in Figure 5d, the full cell assembled with the NSA-based DB-DPE delivered a higher initial discharge capacity (177.62 mAh g⁻¹) compared to the bare Al-based DB-DPE (174.67 mAh g⁻¹). More notably, the NSA-based DB-DPE demonstrated enhanced cycling stability, retaining 85.5% of its initial capacity after 700 cycles, whereas the bare Al-based DB-DPE retained 83.5% over the same period (Figure 5e). These improvements are attributed to the intimate interfacial contact established between the electrode composite and the nanopores on NSA. Repeated lithiation and delithiation generate mechanical stress that, if not countered by strong adhesion at the current collector–composite interface, can cause delamination and structural degradation.^[43] The strong anchoring effect and improved adhesion provided by the nanoporous surface of NSA

effectively mitigate this issue, resulting in superior structural integrity and long-term electrochemical stability under high-loading conditions.^[44]

To further elucidate the interfacial advantages of the NSA, EIS measurements were conducted both immediately after cell fabrication and after 30 cycles (Figure 5f; Figure S29a, Supporting Information). As illustrated by the distribution of relaxation time (DRT) analysis (Figure 5g; Figure S29b, Supporting Information), a significant reduction in the resistance associated with particle–particle contact was observed in the high-frequency region (10³–10⁴ Hz).^[45] Moreover, a noticeable decrease in charge transfer resistance was also detected. These findings suggest that the physical contact between the electrode composite and the NSA is substantially improved compared to that of the bare Al. This enhanced interfacial contact contributes to more efficient electron transport and overall electrochemical performance, further confirming the superior mechanical and

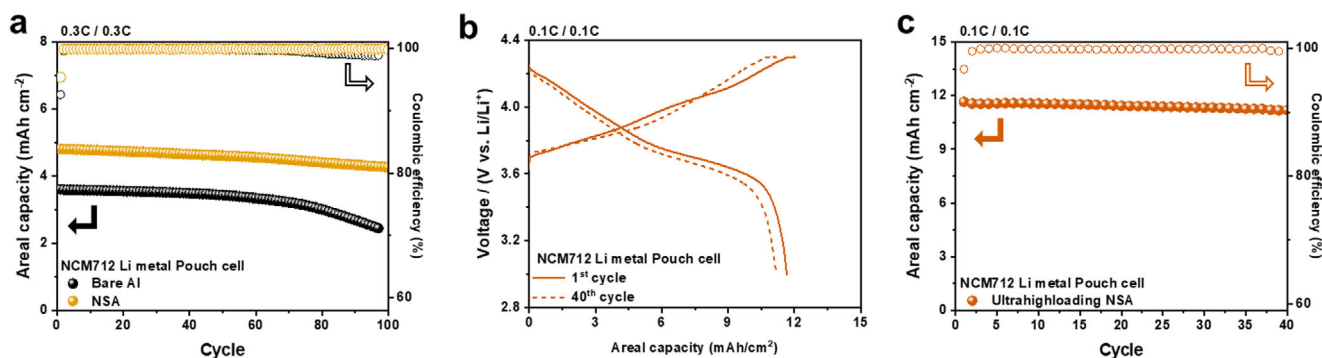


Figure 6. High-loading NSA-based DB-DPEs (Pouch cells). a) Cycling performance of high-loading Li-metal pouch cell with various current collectors at a charge/discharge current density of 0.3/0.3C at a voltage range of 3.0–4.3 V. b) Charge/discharge profiles of ultrahigh-loading Li-metal pouch cell at 0.1/0.1C. c) Cycling performance of ultrahigh-loading Li-metal pouch cell at a charge/discharge current density of 0.1/0.1C at a voltage range of 3.0–4.3 V.

electrochemical stability of the NSA-based DB-DPEs under high-loading conditions.

The rate capability test of the NSA-based DB-DPEs developed in this study, incorporating 96% active material content, demonstrated excellent rate capability even at current densities up to 12 mA cm⁻² (Figure 5h; Table S2, Supporting Information). Compared to previously reported DPEs, the electrode fabricated here exhibited superior performance, highlighting the benefit of enhanced interfacial adhesion provided by the dual-binder dry-process. This robust interfacial contact not only facilitates efficient electronic conduction within thick electrode architectures but also ensures stable operation under high-rate conditions. These results confirm that the high-loading NSA-based DB-DPEs developed in this work are well-suited for coin cell configurations, demonstrating the practical scalability of dry electrode technology for next-generation LIBs.

Furthermore, Li-metal pouch cells (2 × 2 cm²) were assembled using a 200 μm thick Li metal as an anode and DB-DPEs as a cathode to verify their commercial feasibility. As the electrode area increased and binder content decreased, delamination between the current collector and the electrode composite tends to become more critical compared to coin cells. As shown in Figure 6a, the pouch cell employing bare Al exhibited capacity retention of 67.9% after 100 cycles, whereas the NSA-based cell demonstrated improved retention of 88.9%, even at higher loading conditions. This enhancement is attributed to the robust interfacial contact formed by the NSA, which stably maintains electrode structure and ensures reliable electronic pathways throughout cycling.

Using the NSA-based DB-DPEs, an ultrahigh-loading pouch cell with an areal capacity of 12.5 mAh cm⁻² was fabricated and retained 95.9% capacity even after 40 cycles (Figure 6b,c). Notably, the NSA-based DB-DPE architecture enabled the fabrication of high-energy-density cells, with estimated specific energy and volumetric energy density of 305 Wh kg⁻¹ and 712.7 Wh L⁻¹, respectively (Table S3, Supporting Information). The improvement in volumetric energy density is attributed to the reduced electrode thickness and minimized inactive volume, enabled by the high active material content (96 wt.%) and the reduced carbon additive content.

3. Conclusion

In this study, we developed a high-performance DPE by integrating NSA with a dual-binder system of PTFE and PVDF. The NSA, fabricated through a single-step anodization and etching process, provided a uniform nanoporous surface that enhanced interfacial adhesion via mechanical anchoring. The dual-binder approach further improved both cohesion within the electrode and adhesion to the current collector. As a result, the NSA-based DB-DPEs exhibited superior electrical conductivity, structural stability, and electrochemical performance under ultrahigh-loading conditions. Characterizations, including peel testing, C-AFM, and EIS, confirmed reduced interfacial resistance and improved charge transport. Coin cell and pouch cell tests demonstrated excellent rate capability, cycling stability, and practical scalability. Notably, a pouch cell with an areal capacity of 12.5 mAh cm⁻² achieved a volumetric energy density of 712.7 Wh L⁻¹, enabled by the high active material content (96 wt.%) and reduced inactive components, and retained 95.9% of its capacity after 40 cycles. This work demonstrates that combining nanostructured current collectors with dual-binder systems is a viable strategy for scalable, high-energy-density LIBs.

4. Experimental Section

Fabrication of NSA: High-purity Al foils (≥99.85%, 20 μm in thickness, Welcos corp.) were used as the starting material. The raw Al foils were first ultrasonicated in a mixed solution of acetone and methanol for 5 min to remove surface contaminants. The cleaned Al foils were cut into 4 cm × 3.5 cm pieces and immersed in a 0.3 M oxalic acid solution (C₂H₂O₄, 98%, Sigma-Aldrich) for anodization, using a parallel electrode configuration with Pt foil as the cathode. Mild anodization was performed in potentiostatic mode using a DC power supply (TPE-1520DI, Toyotech), with applied voltages ranging from 20 to 50 V for durations between 5 s and 10 min. The resulting alumina layer was removed by immersing the anodized Al foils in a mixture of 10.8 g chromic oxide (CrO₃, >99%, Sigma-Aldrich) and 6 vol% phosphoric acid (H₃PO₄, >85%, Sigma-Aldrich) in 100 mL deionized (DI) water at room temperature for 18 h. The etched foils were subsequently rinsed with DI water and dried in an oven at 60 °C for at least 6 h.

Fabrication of DB-DPEs: To fabricate the DB-DPE, NCM712 (L&F Co., Korea), multi-walled carbon nanotube (MWNT, 20 nm in diameter; JEIO

Co., Korea), and PVDF (HSV-900, Arkema) binder were first mixed at a mass ratio of 96:2:1 using a planetary centrifugal mixer (AR-100, THINKY Corp., Japan) at 2000 rpm for 5 min. Subsequently, 1 wt.% PTFE (F-106, Daikin) binder was added to the mixture and further mixed under the same conditions. The resulting powder mixture was kneaded on a hot plate preheated to 80 °C to form a cohesive dough. This dough was then rolled using a stainless-steel roller to achieve the desired areal loading (5–12 mAh cm⁻²), producing freestanding electrode films. The freestanding films were placed onto either bare Al or NSA. To prevent adhesion to the pressing plates during hot pressing, stainless-steel foils were attached to both sides of the film assembly. The electrodes were then pressed at 180 °C under 10 MPa for 60 s using a preheated hot plate. Finally, the DB-DPEs were roll-pressed at room temperature to achieve a target electrode density of 2.5 g cm⁻³.

Fabrication of SCEs: For the SCEs used in rate capability tests and peel tests, NCM712 (L&F Co.), carbon black (Super P, Timcal Ltd.), and PVDF (HSV-900, Arkema) were used as the active material, conductive agent, and binder, respectively. The components (90:5:5 wt.% for NCM712:carbon black:PVDF) were dispersed in *N*-methyl-2-pyrrolidone (Sigma-Aldrich) to form a homogeneous slurry, which was cast onto Al foil and dried in a vacuum oven at 110 °C.

Characterization: Focused ion beam scanning electron microscopy (FIB-SEM) was performed using a Crossbeam 540 (ZEISS). Field-emission SEM (FE-SEM, JSM-7800F, JEOL Ltd.) and energy-dispersive X-ray spectroscopy (EDS) were used for surface and compositional analysis. Cross-sectional images were obtained from sample cross-sections prepared using an argon ion beam polisher (Cross Section Polisher CP, IB-19510, JEOL Ltd.). Atomic force microscopy (AFM) was performed using an NX-10 system (Park Systems) in contact mode with a scan area of 500 × 500 nm² and a resolution of 512 × 512 pixels. Conductive AFM (C-AFM) measurements were conducted on the same instrument with a scan area of 20 × 20 μm² at a resolution of 256 × 256 pixels. A sample bias of 2 V was applied during C-AFM analysis. X-ray photoelectron spectroscopy (XPS) measurements were recorded on a K-alpha instrument from Thermo Scientific Inc. equipped with an Al anode (Al Kα = 1486.6 eV). The X-ray power for the measurements was 12 kV with 3 mA.

Peel tests were conducted using a universal testing machine (Lloyd instruments) in a 180° configuration to evaluate the adhesion strength and mechanical integrity of the electrodes. The test was performed at a constant peel rate of 50 mm min⁻¹. To prevent cohesion failure within the electrode composite, the component ratio of DPE was adjusted to 90:2:5:3 wt.% for NCM712:MWNT:PTFE:PVDF. The composite resistance, surface resistivity, and interfacial resistance of the electrodes were measured using a specialized electrode measurement system (RM2610, Hioki).

Electrochemical Measurement: Electrochemical measurements were carried out using a battery cycler (WBCS 3000, WonATech) in a temperature-controlled chamber maintained at 25 °C. Half-cell tests were conducted using 2032 type coin cells with Li metal foil (200 μm, Honjo Metal) as both the reference and counter electrode. Cells were charged using a constant current–constant voltage (CC–CV) procedure in a voltage range of 3.0–4.3 V versus Li/Li⁺. Three precycles at 0.1C were performed as a formation step for all cells before the tests (0.05C for electrodes with areal capacities exceeding 10 mAh cm⁻²). The rate capability test was evaluated by fixing the charge rate at 0.2C while varying the discharge rate from 0.2C to 2C. For full-cell configurations, DB-DPE cathodes (96 wt.% active material) were paired with graphite-based anodes. The anodes were prepared using graphite (LG Energy Solution), carbon black (Super P, Timcal Ltd.), styrene–butadiene rubber (SBR, MTI Corp.), and sodium carboxymethyl cellulose (CMC, Sigma-Aldrich). Full cells were cycled using the CC–CV procedure in the voltage range of 2.7–4.3 V versus Li/Li⁺, with an N/P ratio of 1.1. The electrolyte consisted of 1 M LiPF₆ in a 1:1 (v/v) mixture of ethylene carbonate (EC) and diethyl carbonate (DEC), containing 10 wt.% fluoroethylene carbonate (FEC) and 1 wt.% vinylene carbonate (VC) additives (Soulbrain Co., Korea).

To evaluate ionic conductivity, binder films were prepared by mixing PTFE and PVDF in various ratios. The mixtures were kneaded to form a uniform mass and then punched into circular films with a diameter of 19.0 mm. Measurements were conducted at 25 °C using a symmetric SS

| binder film | liquid electrolyte | SS cell configuration. Ionic conductivity (σ) was calculated using the equation $\sigma = L / (R_b \times S)$, where R_b is the bulk resistance obtained from EIS measurements, L is the combined thickness of the binder film and separator, and S is the contact area of the stainless-steel electrodes. EIS measurement was performed using an electrochemical workstation (ZIVE MP1, WonATech) with a frequency range of 1 MHz to 0.1 Hz. Open-source MATLAB script-based software (DRT Tools) was used to calculate the distribution of relaxation time (DRT) from the impedance data. The data were discretized using the Gaussian method with second-order regularization (regularization parameter = 0.001) and fitted using a radial basis function (RBF) with a full width at half maximum (FWHM) of 0.5.

Pouch Cell Fabrication: The Li metal pouch cell was fabricated using an Al pouch film as a package and Li metal (200 μm) as an anode in a dry room with a dew point of –50 °C. The electrochemical performance of the pouch cell was evaluated after the initial formation for 3 cycles at 0.05C under a fixed pressure set at 500 kPa. The specific energies and volumetric energy densities of the pouch cell were calculated based on the experimentally measured weight and volume of the cell, including the packaging film. Calculation details for the specific energies and specific densities are described in Table S3 (Supporting Information).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

S.Y.K. and T.-K.L. contributed equally to this work. This work was supported by the Technology Innovation Program (RS-2024-00437258) funded by the Ministry of Trade, Industry & Energy (MOTIE, Korea) and the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIT) (Nos. RS-2023-00302697, RS-2024-00354154).

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

current collector engineering, dry-processed electrode, dual binder, lithium-ion battery

Received: July 18, 2025
Revised: September 4, 2025
Published online: October 3, 2025

- [1] Y. Q. Guo, C. Guo, P. H. Li, W. J. Song, W. Y. Huang, J. X. Yan, X. B. Liao, K. He, W. X. Sha, X. M. Zeng, X. Y. Tang, Q. Q. Ren, S. Wang, K. Amine, A. M. Nie, T. C. Liu, Y. F. Yuan, *Nat. Commun.* **2025**, *16*, 2441.
- [2] Y. Cui, Y. S. Ye, *Nat. Energy* **2024**, *9*, 639.
- [3] D. Y. Liu, B. B. Wu, Y. B. Xu, J. Ellis, A. Baranovskiy, D. P. Lu, J. Lochala, C. Anderson, K. Baar, D. Y. Qu, J. H. Yang, D. Galvez-Aranda, K. J. Lopez, P. B. Balbuena, J. M. Seminario, J. Liu, J. Xiao, *Nat. Energy* **2024**, *9*, 559.

- [4] J. H. Kim, N. Y. Kim, Z. Y. Ju, Y. K. Hong, K. D. Kang, J. H. Pang, S. J. Lee, S. S. Chae, M. S. Park, J. Y. Kim, G. H. Yu, S. Y. Lee, *Nat. Energy* **2025**, *10*, 295.
- [5] H. Oh, G. S. Kim, J. Bang, S. Kim, K. M. Jeong, *Energy Environ. Sci.* **2025**, *18*, 645.
- [6] Z. Yan, L. Wang, X. He, *Adv. Funct. Mater.* **2025**, *35*, 2415637.
- [7] Y. S. Zhang, N. E. Courtier, Z. Y. Zhang, K. L. Liu, J. J. Bailey, A. M. Boyce, G. Richardson, P. R. Shearing, E. Kendrick, D. J. L. Brett, *Adv. Energy Mater.* **2022**, *12*, 2102233.
- [8] S. Ahmed, P. A. Nelson, K. G. Gallagher, D. W. Dees, *J. Power Sources* **2016**, *322*, 169.
- [9] R. Sahore, D. L. Wood, A. Kukay, K. M. Grady, J. L. Li, I. Belharouak, *ACS Sustain. Chem. Eng.* **2020**, *8*, 3162.
- [10] F. Font, B. Protas, G. Richardson, J. M. Foster, *J. Power Sources* **2018**, *393*, 177.
- [11] Y. Gao, Y. Yang, S. Jia, D. Luo, Z. Chen, *Adv. Funct. Mater.* **2025**, <https://doi.org/10.1002/adfm.202511636>.
- [12] M. Ryu, Y. K. Hong, S. Y. Lee, J. H. Park, *Nat. Commun.* **2023**, *14*, 1316.
- [13] D. J. Lee, J. Jang, J.-P. Lee, J. Wu, Y.-T. Chen, J. Holoubek, K. Yu, S.-Y. Ham, Y. Jeon, T.-H. Kim, J. B. Lee, M.-S. Song, Y. S. Meng, Z. Chen, *Adv. Funct. Mater.* **2023**, *33*, 2301341.
- [14] H. Kim, J. H. Lim, T. Lee, J. An, H. Kim, H. Song, H. Lee, J. W. Choi, J. H. Kang, *ACS Energy Lett.* **2023**, *8*, 3460.
- [15] R. He, W. Zhong, C. Cai, S. Li, S. Cheng, J. Xie, *Adv. Energy Mater.* **2024**, *14*, 2402109.
- [16] G. Matthews, B. Meyer, C. Doerrer, J. Ramírez-González, E. Darnbrough, N. Hallemans, D. Armstrong, P. S. Grant, *J. Mater. Chem. A* **2025**, *13*, 18283.
- [17] S. Jang, M.-H. Ryou, J.-Y. Bae, S.-H. Yu, S.-Y. Lee, J. Bang, *Prog. Polym. Sci.* **2025**, *166*, 101987.
- [18] K. E. Sung, I. Hwan, J. Choi, S. K. Jun, J. Yoon, *Chem. Eng. J.* **2025**, *511*, 161789.
- [19] F. Wang, S. Tang, Q. Han, S. Ji, J. Wang, B. Du, L. Xu, M. Guan, P. Lou, W. Zhang, Y.-c. Cao, S. Cheng, *J. Colloid Interface Sci.* **2025**, *678*, 57.
- [20] W. Jin, G. Song, J.-K. Yoo, S.-K. Jung, T.-H. Kim, J. Kim, *ChemElectroChem* **2024**, *11*, 202400288.
- [21] G. Sun, Q. Lu, W. Pan, L. Xiao, X. Wang, P. Yue, G. Zhang, P. Xiong, *J. Mater. Chem. A* **2025**, *13*, 28682.
- [22] Y. Du, Y. Luo, K. Shi, P. Zuo, Q. Zhang, Z. Zheng, B. Sun, J. Xie, *Adv. Mater.* **2025**, *37*, 2502095.
- [23] B. Ludwig, Z. F. Zheng, W. Shou, Y. Wang, H. Pan, *Sci. Rep.* **2016**, *6*, 23150.
- [24] J. Kang, H. Eom, S. Jang, D. Yoo, H. Lee, M. Kim, M.-L. Seol, J. W. Han, I. Nam, H. Song, *Adv. Mater.* **2025**, *37*, 2416872.
- [25] S. G. Croll, *Prog. Org. Coat.* **2020**, *148*, 105847.
- [26] O. D. Ferreira, E. Gelinck, D. de Graaf, H. Fischer, *Appl. Surf. Sci.* **2010**, *257*, 48.
- [27] J. Offermann, E. Gayretli, C. Schmidt, J. Carstensen, H. G. Bremes, A. Würsig, S. Hansen, M. Abdollahifar, R. Adelung, *J. Colloid Interface Sci.* **2024**, *664*, 444.
- [28] D. Reyter, S. Rousselot, D. Mazouzi, M. Gauthier, P. Moreau, B. Lestriez, D. Guyomard, L. Roué, *J. Power Sources* **2013**, *239*, 308.
- [29] S. H. Kim, P. K. Choi, Y. B. Lee, T. S. Kim, M. S. Jo, S. Y. Lee, H. W. Min, J. B. Yoon, *Nanoscale Adv.* **2024**, *6*, 2013.
- [30] W. Yang, Z. P. Luo, W. K. Bao, H. Xie, Z. S. You, H. J. Jin, *Sci. Adv.* **2021**, *7*, abb9471.
- [31] B. Son, M. H. Ryou, J. Choi, T. Lee, H. K. Yu, J. H. Kim, Y. M. Lee, *ACS Appl. Mater. Interfaces* **2014**, *6*, 526.
- [32] K. Uzun, B. Sharma, B. R. Frieberg, M. Wang, J. Z. Hu, A. Li, X. S. Huang, Y. T. Cheng, *J. Electrochem. Soc.* **2024**, *171*, 020516.
- [33] L. K. Ventrapragada, J. Y. Zhu, S. E. Creager, A. M. Rao, R. Podila, *ACS Omega* **2018**, *3*, 4502.
- [34] J. Entwistle, R. H. Ge, K. Pardikar, R. Smith, D. Cumming, *Renew. Sustain. Energy Rev.* **2022**, *166*, 112624.
- [35] X. F. Bian, Q. Fu, C. G. Qiu, X. F. Bie, F. Du, Y. H. Wang, Y. Q. Zhang, H. L. Qiu, G. Chen, Y. J. Wei, *Mater. Chem. Phys.* **2015**, *156*, 69.
- [36] J. Q. Kang, L. Gu, J. V. Wang, Z. X. Wu, G. R. Zhu, Z. Li, *J. Power Sources* **2022**, *542*, 231746.
- [37] S. P. Zhong, H. Y. Liu, L. Yang, W. Weng, W. Tan, *ACS Appl. Mater. Interfaces* **2025**, *17*, 10593.
- [38] K. T. Chen, Y. C. Yang, Y. H. Yi, X. T. Zheng, H. Y. Tuan, *J. Colloid Interface Sci.* **2021**, *598*, 155.
- [39] M. Z. Wang, H. Yang, K. X. Wang, S. L. Chen, H. N. Ci, L. R. Shi, J. Y. Shan, S. P. Xu, Q. C. Wu, C. Z. Wang, M. Tang, P. Gao, Z. F. Liu, H. L. Peng, *Nano Lett.* **2020**, *20*, 2175.
- [40] M. J. Lain, G. Apachitei, D. E. Dogaru, W. D. Widanage, J. Marco, M. Copley, *RSC Adv.* **2023**, *13*, 33437.
- [41] L. J. Cao, L. L. Li, Z. Xue, W. Yang, H. B. Zou, S. Z. Chen, Z. L. Liu, *J. Porous Mater.* **2020**, *27*, 1677.
- [42] S. J. Chen, C. H. Liu, Q. L. Wang, X. X. Wu, Y. Yang, L. Wang, J. L. Luo, X. Z. Fu, *J. Power Sources* **2023**, *587*, 233680.
- [43] B. Q. Li, J. Chen, W. Li, J. P. Li, J. Y. Chen, L. B. Wang, *J. Power Sources* **2024**, *624*, 235586.
- [44] D. Y. Shin, D. H. Park, H. J. Ahn, *Appl. Surf. Sci.* **2019**, *475*, 519.
- [45] H. Nara, D. Mukoyama, R. Shimizu, T. Momma, T. Osaka, *J. Power Sources* **2019**, *409*, 139.