



High-Ni layered cathodes for wide-temperature operation: From coupled degradation mechanisms to temperature-adaptive stabilization

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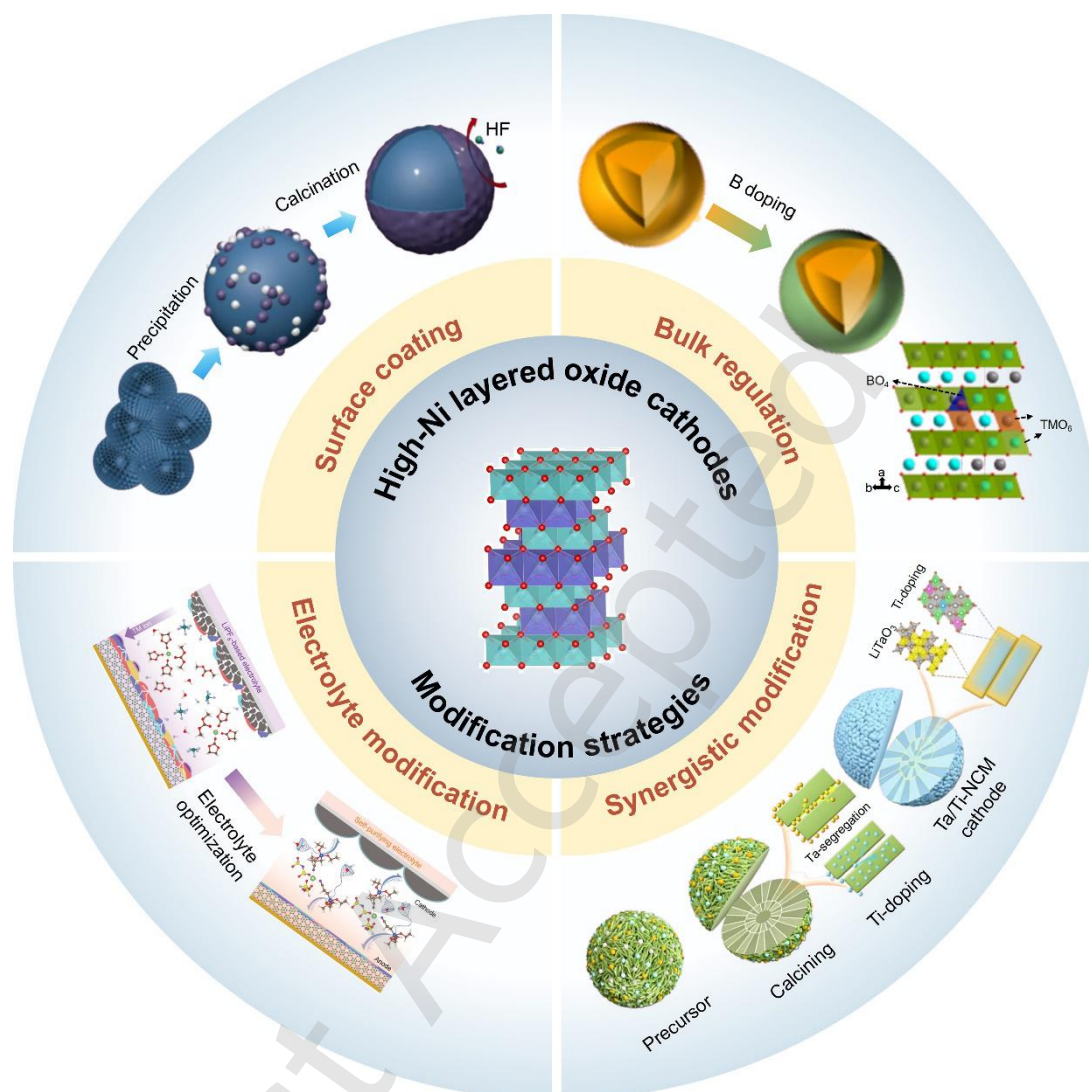
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This review summarizes the coupled degradation mechanisms of high- Ni layered cathodes under wide- temperature conditions and reviews recent advances in optimization strategies. Apart from conventional doping and coating, functional interfacial layers with intrinsic thermal- responsive properties are highlighted as an emerging strategy to regulate interfacial charge transfer and lattice- strain evolution. These findings provide guidance for developing wide- temperature high- Ni cathodes with enhanced safety and durability.

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ABSTRACT

Wide-temperature operation is becoming essential for high-energy Li-ion batteries deployed in extreme environments, thereby requiring cathode materials capable of sustaining high energy density and stable electrochemical reactions under thermal fluctuations. Accordingly, high-Ni layered oxide cathodes have attracted considerable attention due to the high specific capacity and elevated operating voltage. However, their practical application across a wide temperature range remains constrained by temperature-induced degradation. At elevated temperatures, lattice expansion, oxygen release, and irreversible structural evolution within the cathode bulk undermine structural stability. Meanwhile, accelerated electrolyte decomposition further promotes surface reconstruction, transition-metal dissolution, and uncontrolled interfacial layer growth at the cathode/electrolyte interface. At low temperatures, sluggish Li-ion transport throughout the cathode bulk and electrolyte is accompanied by poor interfacial desolvation/charge-transfer kinetics. Understanding these coupled degradation processes is essential for improving the wide-temperature performance of high-Ni layered cathodes. This review therefore elucidates their temperature-dependent degradation mechanisms and discusses recent progress in optimization strategies. Beyond conventional doping and coating, functional interfacial layers with intrinsic physical responses to service-temperature variations is emphasized as an emerging approach for regulating interfacial charge transfer and lattice strain evolution. These insights are expected to guide the design of wide temperature high-Ni cathodes with improved safety and durability.

KEYWORDS

Li-ion battery, layered oxide cathode, wide temperature operation

1 Introduction

With the expanding deployment of Li-ion batteries (LIBs) in extreme environments, such as aerospace systems, polar exploration, high-altitude energy storage, and deep-sea exploration, the demand for stronger environmental adaptability, higher energy density and long-term stability continues to increase [1–7]. These extreme environments often involve both large temperature fluctuations and prolonged exposure to low or elevated temperatures. Consequently, reliable battery operation is required across wide-temperature regions beyond conventional room-temperature conditions [8–11]. Current studies on

wide-temperature battery operation generally cover temperature windows of approximately -40 to 60 °C or broader, while more extreme temperatures have also been explored in recent work.

Meeting these requirements is particularly dependent on cathode materials, which largely determine the energy density, rate and cycling performances of LIBs under wide-temperature conditions. Among the various cathodes, high-Ni layered oxides $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$ are widely regarded as promising candidates for high-specific-energy LIBs due to their high theoretical specific capacity of approximately $280 \text{ mAh}\cdot\text{g}^{-1}$ and high operating voltage of $3.8 \text{ V vs. Li/Li}^+$. Nevertheless, their wide-temperature operation remains challenging, as low and high temperatures trigger the different degradation pathways involving sluggish reaction kinetics, interfacial instability, and structural deterioration [12–16]. Under high-temperature conditions, degradation occurs simultaneously within the cathode bulk, at the cathode/electrolyte interface, and in the electrolyte. Bulk degradation mainly involves lattice thermal expansion, strain accumulation, lattice oxygen release, irreversible phase transitions, and microcrack formation. At the cathode/electrolyte interface, surface reconstruction, transition-metal dissolution, CEI evolution, and increasing interfacial resistance progressively deteriorate electrochemical reversibility. Meanwhile, elevated temperatures accelerate electrolyte decomposition and generate reactive species that further intensify interfacial degradation [17–24]. Under low-temperature conditions, degradation is dominated by the kinetic limitations. Within the cathode bulk, lattice contraction and reduced solid-state Li-ion diffusivity restrict Li-ion transport. At the interface, sluggish desolvation and charge transfer increase electrochemical polarization. In the electrolyte, increased viscosity and reduced ionic conductivity further hinder Li-ion transport [25–39]. These processes are strongly coupled and collectively determine the wide-temperature performance of high-Ni layered cathodes.

To mitigate these limitations, surface coating, elemental doping, and electrolyte optimization have been widely developed. Surface coatings, including inert compounds [40], fast ionic conductors [41, 42], and conductive polymers [43–45], can protect cathode particles from electrolyte attack, thereby suppressing parasitic reactions during high-temperature cycling. Elemental doping [46–56] can stabilize the layered framework and modulate Li-ion migration pathways, which can improve the structural robustness at high temperatures or lower Li-ion diffusion barriers under low-temperature conditions. Electrolyte engineering, including film-forming additives [57–61], and weak-solvation regulation [62–68], can reduce electrolyte viscosity and desolvation barriers at low-temperatures, or promote stable and thin cathode-electrolyte interfacial film formation during high-temperature cycling. Despite these significant advances, most existing strategies remain effective only within specific temperature regimes and fail to simultaneously address high-temperature interfacial/bulk structure degradation, and low-temperature kinetic retardation.

Given the limitations of conventional strategies, functional interfacial layers with intrinsic physical responses to service-temperature variations have emerged as a promising route to integrate electrochemical regulation with interfacial multiphysics coupling involving thermal, electrical, and mechanical fields. For examples, ferroelectric layers can generate built-in electric fields across CEI, thereby accelerating Li-ion migration, suppressing side-reaction product accumulation, and blocking aggressive species [69–71]. Negative-thermal-expansion layers can generate reverse thermal strain in response to service-temperature variations, thereby compensating for lattice expansion or contraction in high-Ni cathodes, reducing interfacial thermal stress, and improving thermomechanical stability [72, 73]. Thermoelectric and piezoelectric materials further offer opportunities to regulate local temperature

gradients, interfacial ion transport, and strain evolution. Unlike conventional surface modifications, these functional interfacial layers enable coupled regulation of Li-ion transport, interfacial chemical stability, and structural integrity through mechanical, electrical, and thermal field interactions. Therefore, they hold great promise for improving the wide-temperature performance of high-Ni layered oxide cathodes. However, reports on the wide-temperature regulation of high-Ni cathodes through multiphysics-coupled interfacial design remain scarce, mainly due to the insufficient targeted modulation of service-temperature-dependent functional parameters in interfacial layers, which limits their adaptive regulation under wide-temperature operating conditions. Moreover, existing reviews mainly focus on room-temperature or single-temperature degradation and modification strategies, whereas systematic summaries of wide-temperature failure behaviors and modification strategies remain limited. In particular, the intrinsic mechanisms and application pathways of ferroelectric, negative thermal expansion, thermoelectric, and piezoelectric interfacial layers for coordinated high and low-temperature regulation require further clarification.

Based on the above research background, this review systematically summarizes the performance degradation mechanisms and modification strategies of high-Ni layered oxide cathodes under wide-temperature operating conditions. This review further highlights the regulation potential and future prospects of functional interfacial layers with intrinsic physical responses to service-temperature variations. First, the structural, interfacial, and kinetic degradation mechanisms of high-Ni layered cathodes at high and low-temperatures are discussed, followed by a summary of existing modification strategies and their optimization mechanisms along with an analysis of their limitations in wide-temperature-range applications. Subsequently, functional interfacial layers, such as ferroelectric materials and negative-thermal-expansion materials, are discussed in terms of their current applications and multiphysics-coupled regulation mechanisms in high-Ni layered oxide cathode. Their potential pathways for enhancing the high and low-temperature performance through coupled thermal, electrical, mechanical, and electrochemical fields are further proposed. Building on these discussions, this review aims to provide theoretical support and forward-looking guidance for the rational design of high-Ni layered oxide cathodes toward all-climate and extreme-environment applications.

2 Structure and temperature-dependent degradation mechanisms

2.1 Structural features of high-Ni layered oxides

High-Ni layered oxide cathodes generally crystallize in an α -NaFeO₂ type hexagonal layered structure with the $R\bar{3}m$ space group. In this structure, Li ions occupy the 3a octahedral sites, whereas transition-metal ions occupy the 3b octahedral sites, forming alternating Li layers and transition-metal oxide slabs along the *c*-axis (Fig. 1(a)) [74–78]. This layered structure provides quasi-two-dimensional Li-ion diffusion channels, facilitating ion transport and high-rate electrochemical reactions [79–81].

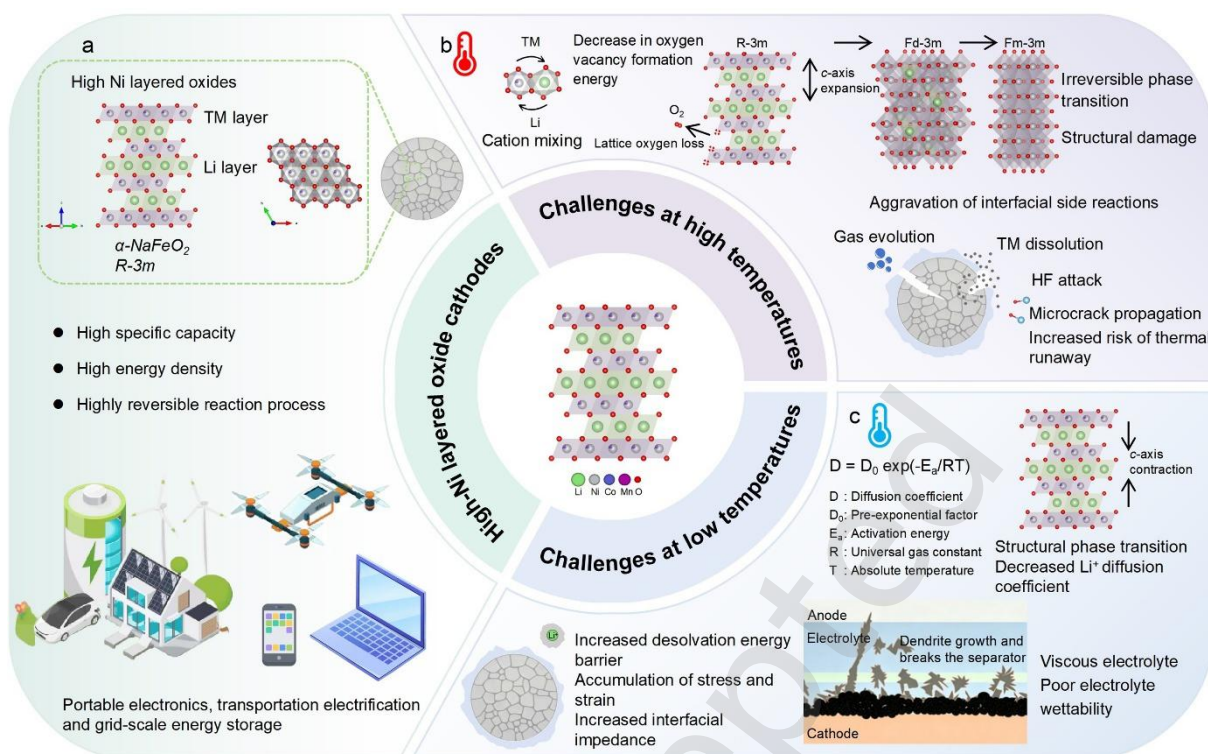


Figure 1 (a) Structural characteristics of high-Ni layered oxides, (b) high and (c) low temperature degradation mechanisms.

The electrochemical performance of $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$ is strongly governed by the composition-dependent balance among Ni, Co, and Mn. Ni provides the dominant redox contribution in high-Ni layered oxides, and a higher Ni content strengthens the $\text{Ni}^{2+}/\text{Ni}^{3+}/\text{Ni}^{4+}$ redox activity, thereby increasing reversible capacity and energy density [82–84]. Co contributes to electronic conductivity and cation ordering, which helps mitigate $\text{Li}^+/\text{Ni}^{2+}$ disorder, while Mn mainly exists as Mn^{4+} under conventional operating conditions and contributes to structural stabilization through strong Mn-O bonding [85–89]. Therefore, Ni enrichment represents the most direct route to increasing energy density, and high-Ni NCM cathodes with Ni contents above 60% usually deliver reversible capacities exceeding $200 \text{ mAh}\cdot\text{g}^{-1}$. However, excessive Ni contents will reduce the structural and interfacial stability. At high states of charge, reactive Ni^{4+} species promote oxygen release, electrolyte decomposition, surface reconstruction, and transition-metal migration. Meanwhile, deep delithiation induces the H2-H3 phase transition and anisotropic lattice contraction, leading to microcrack formation and continuous exposure of fresh surfaces to the electrolyte [90–95]. Consequently, the capacity advantage enabled by Ni enrichment is accompanied by accelerated structural and interfacial degradation, constituting an intrinsic trade-off between high energy density and long-term stability in high-Ni layered oxide cathodes.

This intrinsic trade-off becomes more pronounced under wide-temperature operating conditions, where the structural and interfacial instabilities associated with Ni enrichment are further amplified. At elevated temperatures, highly reactive Ni^{4+} species accelerate lattice oxygen release, electrolyte oxidation, transition-metal dissolution, and surface reconstruction, resulting in rapid interfacial impedance growth and structural degradation. At low-temperatures, sluggish Li-ion diffusion, increased charge-transfer

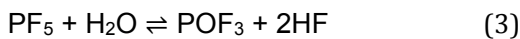
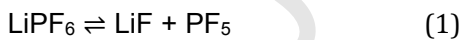
resistance, and aggravated polarization further magnify the adverse effects of $\text{Li}^+/\text{Ni}^{2+}$ cation mixing and blocked diffusion channels. In addition, temperature fluctuations can intensify anisotropic lattice strain and intergranular stress accumulation, thereby promoting microcrack propagation and continuous electrolyte penetration. Therefore, an in-depth understanding of the temperature-dependent failure mechanisms at both high and low-temperatures is essential for establishing rational modification strategies toward wide-temperature high-Ni layered oxide cathodes.

2.2 Challenges at high-temperatures

The elevated-temperature degradation of high-Ni layered oxide cathodes involves a cascade of coupled processes in the cathode bulk, at the cathode/electrolyte interface, and within the electrolyte. Their mutual reinforcement progressively compromises structural integrity and thermal safety (Fig. 1(b)). Under high-temperature and highly charged conditions, thermally induced lattice expansion is coupled with deep-delithiation driven anisotropic lattice variation, which generates severe strain within the Ni-rich layered framework. The thermally aggravated lattice strain becomes more pronounced with increasing Ni content, especially during the H2-H3 phase transition. The abrupt c-axis contraction and anisotropic volume variation during this transition will accelerate stress accumulation. The accumulated stress further initiates and propagates microcracks, exposing fresh surfaces to the electrolyte and accelerating subsequent interfacial degradation [17, 22, 24].

Severe lattice distortion also weakens the stability of lattice oxygen. Highly oxidized Ni^{4+} species strongly interact with lattice oxygen, facilitating oxygen-vacancy formation and accelerating oxygen release at elevated temperatures. The loss of lattice oxygen drives irreversible surface reconstruction from the layered phase to spinel and rock-salt phases, leading to blocked Li-ion diffusion channels and increased interfacial impedance. Meanwhile, the reactive oxygen species can oxidize electrolyte components and further aggravate interfacial side reactions. Therefore, lattice distortion, oxygen release, and phase reconstruction are closely coupled structural degradation pathways in high-Ni cathodes at elevated temperatures [20, 23].

In addition, the thermal stability of the electrolyte progressively decreases at elevated temperatures. LiPF_6 decomposition and its reaction with trace moisture are accelerated, producing LiF , PF_5 , POF_3 , and corrosive HF according to the following reactions [96–98]



These electrolyte-derived reactive species directly affect the cathode/electrolyte interface. For example, the generated HF continuously attacks the CEI and corrodes the exposed cathode surface, resulting in transition-metal dissolution, surface structural damage, and additional lattice oxygen loss [18, 21]. Besides, highly reactive Ni^{4+} species at high states of charge promote oxidative electrolyte decomposition and gas evolution, resulting in the continuous CEI thickening [19]. These processes increase charge-transfer resistance and further accelerate the degradation of surface structure.

Overall, high-temperature degradation of high-Ni layered oxide cathodes arises from coupled deterioration of the cathode bulk, electrolyte, and cathode/electrolyte interface. Within the cathode bulk, lattice strain, oxygen instability, irreversible phase evolution, and microcracking progressively destabilize the layered structure. Concurrently, accelerated electrolyte decomposition generates reactive and corrosive species while promoting gas evolution. These species further attack the cathode/electrolyte interface, driving CEI growth, surface corrosion, transition-metal dissolution, and structural reconstruction, thereby increasing interfacial resistance. These degradation processes continuously interact and amplify each other during cycling, ultimately leading to rapid capacity fading, shortened cycle life, and increased thermal runaway risk.

2.3 Challenges at low-temperatures

Under low-temperature conditions, the performance degradation of high-Ni layered oxide cathodes arises from concurrently deteriorated Li-ion transport within the cathode bulk, electrolyte transport properties, and cathode/electrolyte interfacial kinetics (Fig. 1(c)) [26, 28, 36]. As temperature decreases, the contraction of the Li slabs reduces the interlayer spacing available for Li-ion transport, thereby lowering the bulk Li-ion diffusion coefficient and weakening the reversibility of Li-ion insertion/extraction [32]. This lattice-contraction-induced transport limitation becomes more pronounced in high-Ni cathodes with long Li-ion diffusion pathways or reduced Co content, where insufficient electronic/ionic transport further enlarges electrochemical polarization at low-temperatures [25, 27, 29–31]. According to the Arrhenius relationship [99]:

$$D = D_0 \exp(-E_a/RT) \quad (4)$$

where D is the Li-ion diffusion coefficient, D_0 is the pre-exponential factor, E_a is the activation energy, R is the gas constant, and T is the absolute temperature, respectively [99]. A decrease in temperature therefore leads to reduced Li-ion diffusivity. Consequently, Li-ion insertion and extraction become spatially nonuniform within cathode particles, resulting in incomplete phase transitions, weakened H2-H3 phase reversibility, and intensified local Li-ion concentration gradients.

The restricted bulk Li-ion diffusion further induces anisotropic lattice strain and stress accumulation during repeated charge/discharge processes. Especially under low-temperature and high-rate operation, sluggish Li-ion transport fails to match the imposed electrochemical reaction rate, leading to local over-lithiation or over-delithiation. The resulting reaction heterogeneity promotes lattice dislocations, intragranular cracking, and intergranular microcrack formation. These structural defects disrupt particle integrity, weaken electrical contact between primary particles, and lengthen Li-ion transport pathways. Meanwhile, newly exposed internal surfaces provide additional reactive sites for electrolyte penetration and interfacial side reactions, further accelerating structural degradation of high-Ni layered cathodes.

In addition to the Li-ion transport limitation within the bulk structure, low temperatures simultaneously deteriorate electrolyte transport and cathode/electrolyte interfacial kinetics. The increased electrolyte viscosity and reduced ionic conductivity hinder Li-ion transport through the electrolyte, while temperature-dependent changes in the solvation environment can further influence ion transport behavior. At the cathode/electrolyte interface, Li-ion desolvation becomes more difficult, leading to sluggish interfacial charge transfer [33, 34]. Under more extreme low-temperature conditions, partial solidification of some

electrolyte formulations may further reduce ionic conductivity and hinder ion transport through the electrode, resulting in severe polarization and poor rate capability [35, 37–39].

Therefore, low-temperature degradation of high-Ni layered oxide cathodes results from the coupling of sluggish solid-state Li-ion transport, deteriorated electrolyte transport properties, and poor cathode/electrolyte interfacial kinetics. Reduced bulk Li-ion diffusivity promotes heterogeneous lithiation/delithiation, incomplete phase evolution, and local stress accumulation. Meanwhile, increased electrolyte viscosity and reduced ionic conductivity restrict ion transport, while sluggish desolvation and charge transfer further increase interfacial polarization. These coupled kinetic limitations lead to capacity loss, rate deterioration, and unstable cycling under low-temperature operating conditions.

In summary, high- and low-temperature degradation of high-Ni layered cathodes is governed by distinct yet coupled mechanisms. Elevated temperatures mainly accelerate bulk structural instability, electrolyte decomposition, and cathode/electrolyte interfacial degradation. In contrast, low temperatures primarily impose kinetic limitations on Li-ion transport in the cathode and electrolyte, together with interfacial desolvation/charge-transfer processes. The resulting reaction heterogeneity and Li-ion concentration gradients can further induce nonuniform lattice evolution and local stress accumulation, particularly under severe low-temperature or high-rate conditions. Therefore, effective wide-temperature regulation (Fig. 2) requires coordinated optimization of the cathode bulk, electrolyte, and interface while maintaining efficient Li-ion transport across a broad temperature range.

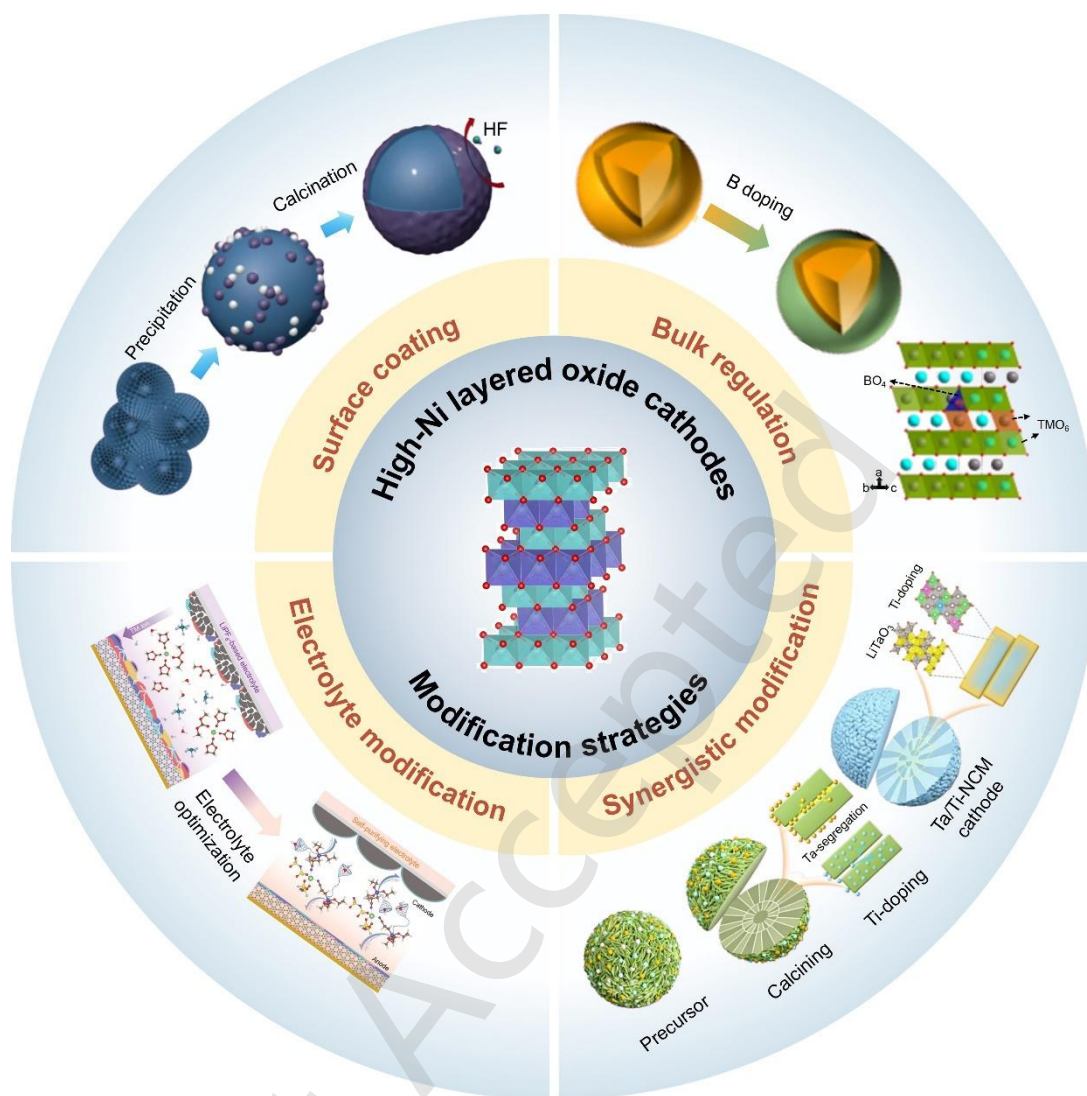


Figure 2 Modification strategies for wide-temperature high-Ni layered oxide cathodes.

3 Wide-temperature modification strategies

3.1 Surface coating

Surface coating is widely regarded as a simple and effective strategy for protecting cathode materials [40–45, 60, 100–113]. Specifically, the protective layer formed on the cathode surface suppresses parasitic interfacial reactions, reduces transition-metal dissolution, and stabilizes the surface structure, thereby improving electrochemical performance.

Inorganic and composite coatings have been demonstrated to enhance the cycling stability through reducing the surface residual Li species and suppressing lattice oxygen release. For example, Chu et al. [100] constructed a thermodynamically stable LiF/FeF_3 dual modification layer through a one-step *in-situ*

method, and the residual Li on the surface was reduced and the release of lattice oxygen was suppressed (Fig. 3(a)). The LiF/FeF₃ composite coating layer was uniformly and densely coated onto the surface of the NCM811 cathode material, with the $R\bar{3}m$ layered structure being well preserved and the crystal structure remaining intact and free of impurity phases after coating (Figs. 3(b)–3(i)). The capacity of the NCM811 cathode decayed rapidly during cycling at a high rate of 5 C, with a capacity retention of only 62.4% after 300 cycles, whereas a capacity retention of 86.4% was retained by NCM@LFF3 (Fig. 3(j)). This improvement was attributed to the reduced degree of Li⁺/Ni²⁺ mixing and the enhanced lattice oxygen and structural stability. The cycling performance of the NCM811 and NCM@LFF3 cathodes at a high-temperature of 60 °C with a capacity retention of 91.3% after 150 cycles by NCM@LFF3 (Fig. 3(k)). Compared with the performance at room temperature, the increased degree of Li/Ni mixing was found to accelerate the destruction of layered structure, internal cracking and pulverization of secondary particles, as well as release of oxygen, leading to increased electrolyte consumption and internal resistance. These were identified as the primary factors responsible for the degradation of high-temperature cycling performance. As a comparison, NCM811 exhibited 155.2 mAh·g⁻¹ after 150 cycles, with a capacity retention of 79%, whereas a specific capacity of 159.9 mAh·g⁻¹ was still maintained by NCM@LFF3 after 150 cycles, corresponding to a capacity retention of 91.3%. Liu et al. [41] constructed a Li_{1.3}In_{0.3}Ti_{1.7}(PO₄)₃ fast-ion-conducting layer on single-crystal high-Ni cathodes to homogenize surface Li-ion distribution and mitigate diffusion-induced stress. After 400 cycles in a full cell at a high-temperature of 45 °C, a capacity retention of 88% was achieved. The structural stability and electrochemical performance of the material under high-voltage and high-temperature conditions were significantly enhanced. Furthermore, Zhao et al. [108] proposed an epitaxial entropy-assisted coating strategy, in which an epitaxial coating with high-entropy characteristics was *in-situ* constructed on the particle surface through the orientation-attachment reaction between Wadsley-Roth phase oxides (Nb₁₂WO₃₃ and ZrO₂) and ultra-high-Ni cathodes. By this coating, lattice dislocations, anisotropic strain accumulation, and oxygen release were significantly suppressed, thus enhancing the structural stability under fast-charging and wide-temperature-range conditions.

Conductive polymer coating layers are widely applied to high-Ni cathodes owing to their favorable electronic conductivity, flexibility, and ease of film formation. For instance, an approximately 10 nm ultraconformal poly(3,4-ethylenedioxythiophene) coating was constructed by Liu et al. [45] through oxidative chemical vapor deposition, effectively mitigating electrolyte-induced degradation and inhibiting the detrimental H2-H3 phase transition. After being stored at a high-temperature of 45 °C for 72 h, a capacity retention of 85.3% was still maintained. Liu et al. [43] further built an ultra-conformal poly(3,4-ethylenedioxythiophene) coating through chemical vapor deposition, a protective layer that was endowed with HF-scavenging capability and mechanical buffering effects. By this coating, the dissolution of transition metals, the intragranular dislocations, and the propagation of intergranular cracks were effectively inhibited. Consequently, 88.7% retention was achieved after 100 cycles at 55 °C.

Beyond conductive polymers, functional polymer coatings with tailored functional groups can protect high-Ni cathodes through multiple mechanisms, including chemical passivation and moisture resistance. Inspired by the sustained-release capsule mechanism, Shi et al. [106] constructed a polydimethylsiloxane coating with excellent hydrophobicity and chemical inertness, and thus the moisture attack was effectively blocked and the accumulation of residual lithium was suppressed. Besides, HF was continuously captured by the siloxane groups and the formation of a stable CEI was promoted, and thereby the thermal stability

of the high-Ni layered cathode was improved. Moreover, Yang et al. [44] constructed a self-adaptive polymer interface based on polyrotaxane-co-poly(acrylic acid). The slide-ring structure of polyrotaxane endowed the protective layer with high toughness and effective stress-dissipation capability through the sliding motion of cyclic molecules, while the carboxyl groups chelated transition-metal ions and suppressed their dissolution. As a result, markedly improved cycling stability was achieved at 55 °C.

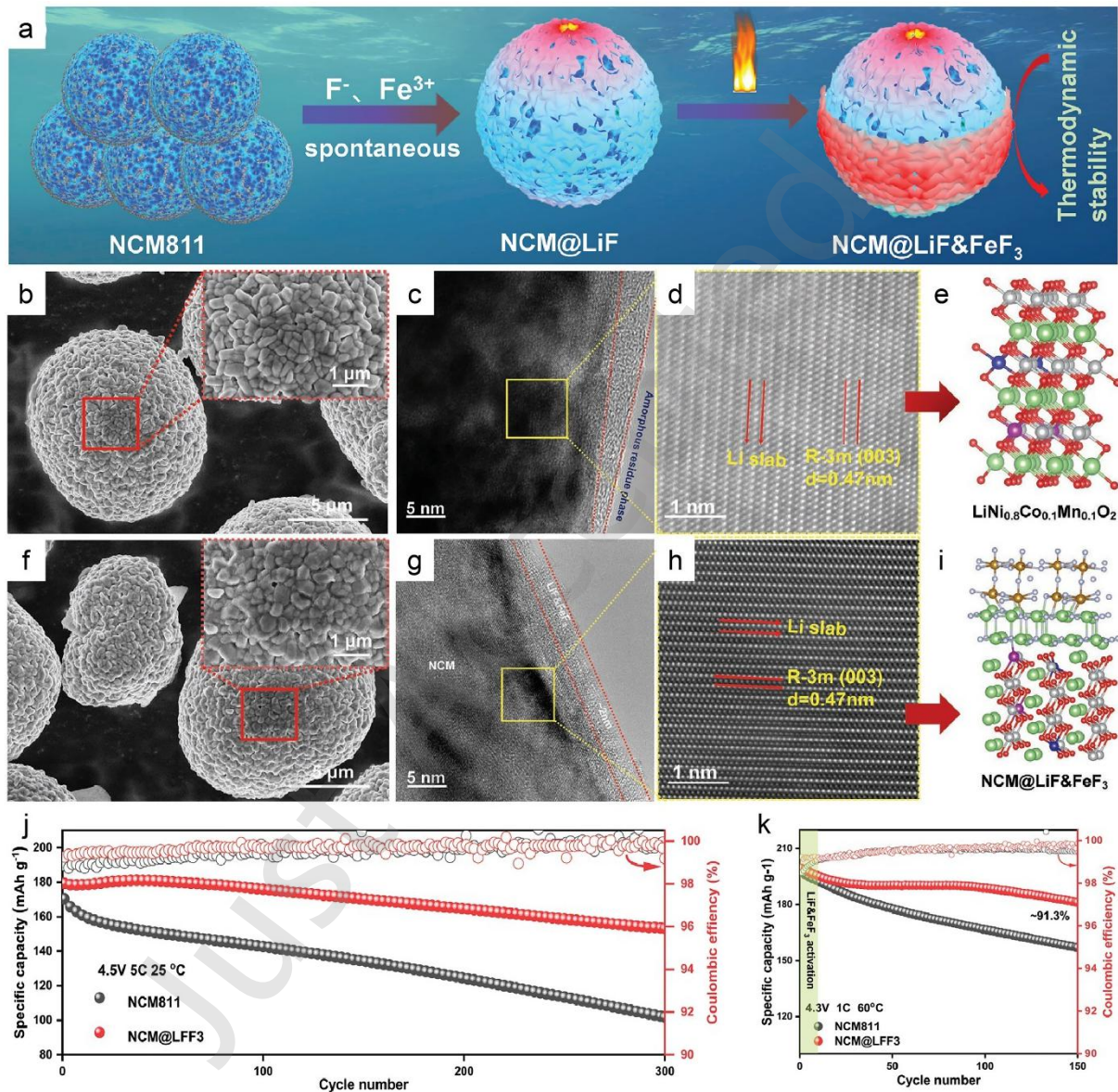


Figure 3 (a) Schematic illustration of the preparation procedure of NCM@LFF material. (b) SEM images of NCM811. (c) HRTEM images and (d) the corresponding SAED patterns of NCM811. (e) Diagram of NCM811 structure. (f) SEM images of NCM@LFF3. (g) HRTEM images and (h) the corresponding SAED patterns of NCM@LFF3. (i) Diagram of NCM@LFF3 structure. (j) Cycling performance of NCM and NCM@LFF3 under 5 C between 2.7–4.3 V at 25 °C. (k) High-temperature performance of NCM811 and NCM@LFF3. (a)–(k) Reproduced with permission from Ref. [100], © Wiley-VCH GmbH 2023.

Overall, surface coating is an effective strategy for improving the wide-temperature stability of high-Ni layered cathodes. By constructing protective layers, parasitic reactions can be suppressed, interfacial stability can be enhanced, and lattice oxygen release together with layered-to-rock-salt phase transition

can be retarded. When fast ionic conductors are used as coating materials, interfacial Li-ion transport at low-temperatures can also be improved, while microcrack propagation induced by volume variation can be alleviated. Nevertheless, surface coating mainly regulates the cathode surface and interface, while its influence on bulk cation mixing, lattice stress accumulation, and internal strain evolution remains limited. Therefore, the complex degradation of high-Ni layered oxide cathodes across a wide temperature range cannot be fully addressed by surface coating alone.

3.2 Bulk regulation

Bulk regulation has been widely investigated to improve the wide-temperature performance of high-Ni layered oxide cathodes [46–56, 114–127]. It encompasses regulation of the internal lattice, compositional distribution, primary-grain architecture, internal defects, and grain boundaries of cathode particles. These strategies operate across the different scales to stabilize local coordination and the layered framework, suppress detrimental phase transitions and lattice oxygen release, regulate Li-ion transport, as well as alleviate heterogeneous strain and stress accumulation.

To suppress the lattice oxygen release and detrimental phase transitions of high-Ni cathode materials in the delithiated state at elevated temperatures, the thermodynamic and kinetic stability are enhanced through strategies such as strengthening the metal-oxygen bond energy, raising the cation migration barrier, or inducing lattice distortion. For example, Shen et al. [55] designed a cobalt-free single-crystal $\text{LiNi}_{0.8}\text{Mn}_{0.18}\text{Fe}_{0.02}\text{O}_2$ cathode material through a FeMn synergistic doping strategy, in which the operating voltage plateau was elevated by Fe doping, while the instability of lattice oxygen was suppressed by Mn doping owing to the larger energy gap with the oxygen p-band. Through the synergistic effect of the dopants, a capacity retention of 84.5% was delivered after 100 cycles under 55 °C and 4.6 V, the thermal decomposition temperature was raised to 234.5 °C, and the cost was reduced by more than 15%. Wang et al. [52] elucidated the structural degradation mechanism of high-Ni cathode materials at elevated temperatures, and it was revealed that the c-axis thermal expansion in the delithiated state was aggravated by high-temperatures and predominantly occurred in the lithium layers, becoming more severe with increasing Ni content and consequently accelerating oxygen release with irreversible phase transitions. Based on these, large-radius Na and La elements were introduced into the Li and Ni sites to induce lattice distortion, and a dislocation-pinning-endowed $\text{Li}_{0.99}\text{Na}_{0.01}\text{Ni}_{0.98}\text{La}_{0.02}\text{O}_2$ was constructed. Consequently, the lattice distortion effectively suppressed the thermal expansion (Figs. 4(a) and 4(b)), and 78.9% retention was thus achieved after 500 cycles at 50 °C (Fig. 4(c)).

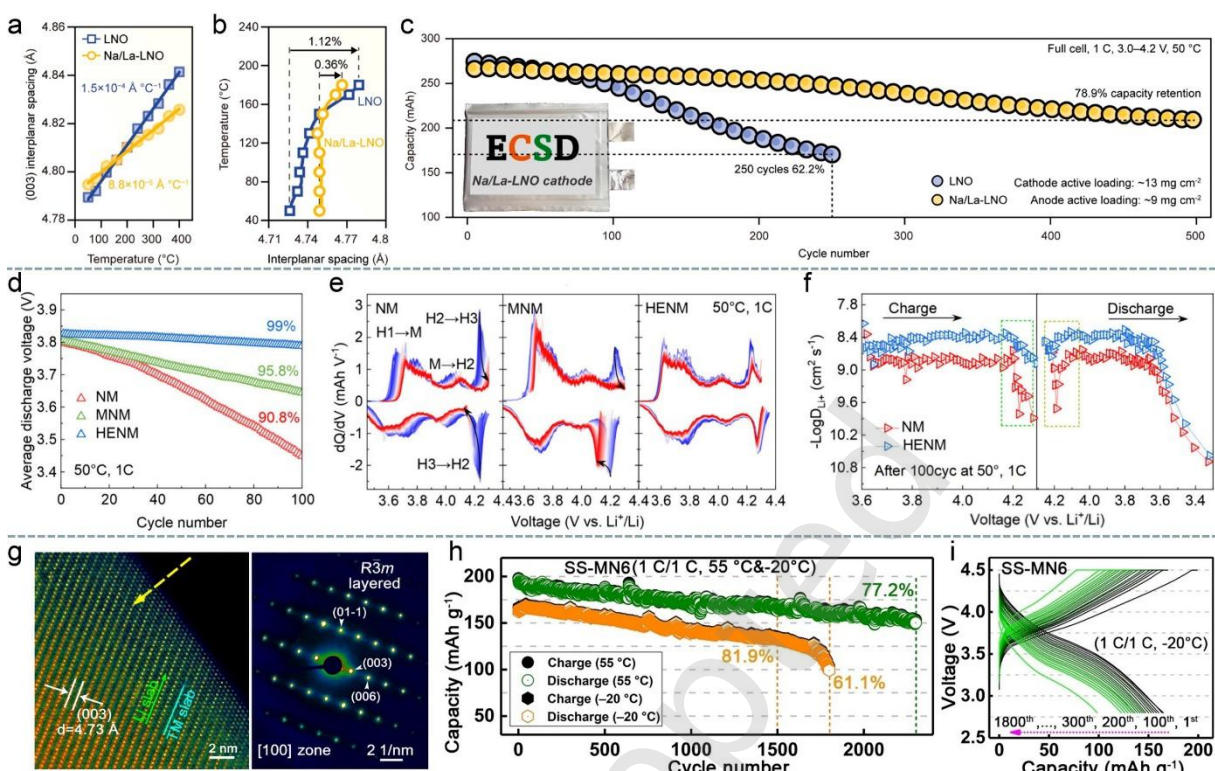


Figure 4 The change of (003) interplane spacing for (a) lithiated and (b) delithiated samples during heating. (c) Cycling performance of LNO and Na/La-LNO at 1 C in pouch-type full cells using graphite as the anode at 50 °C. (a)–(c) Reproduced with permission from Ref. [52], © Elsevier 2025. Cycling performance and the corresponding (d) voltage drop comparison, and (e) dQ/dV plots of NM, MNM, and HENM cathode at 1 C and 50 °C. (f) Li diffusion constant of NM and HENM after 100 cycles at 1 C and 50 °C obtained by the GITT. (d)–(f) Reproduced with permission from Ref. [121], © American Chemical Society 2025. (g) STEM-HAADF image and SAED pattern of SS-MN6. (h) Cycling stability at 1 C under -20 and 55 °C and voltage profiles of SS-MN6 at -20 °C. (g)–(i) Reproduced with permission from Ref. [47], © Wiley-VCH GmbH 2023.

Employing a multi-element high-entropy doping strategy, Zhou et al. [121] successfully prepared $\text{LiNi}_{0.87}\text{Mn}_{0.1}\text{Mg}_{0.01}\text{Al}_{0.01}\text{Fe}_{0.005}\text{Ti}_{0.005}\text{O}_2$, in which Mg, Al, Fe, and Ti were incorporated. The charge density around Ni atoms was localized and the Ni-O bond covalency was enhanced by the high-entropy doping. Thus, the out-of-plane migration barrier of Ni^{2+} was increased from 1.30 to 1.59 eV and the energy barrier for the layered-to-spinel-to-rock-salt phase transition was raised from 0.66 to 1.37 eV. Consequently, the structural degradation and surface reconstruction during high-temperature cycling were inhibited. A capacity retention of 91.3% and a voltage retention of 99% were delivered after 100 cycles at 1 C and 50 °C (Fig. 4(d)). Further investigations indicate that the high-entropy strategy reduces polarization and suppresses pronounced variation of the H2-H3 phase transition (Fig. 4(e)), thereby enhancing phase-transition reversibility and Li-ion transport kinetics. This improvement is further reflected by the increased Li-ion diffusion coefficient (Fig. 4(f)). The thermal decomposition temperature was elevated to 257.5 °C, and the heat release was decreased from 1254.3 to 317.9 J·g⁻¹.

Zhu et al. [56] realized the ultradispersed doping of Nb in $\text{LiNi}_{0.9}\text{Co}_{0.1}\text{O}_2$ through an *in-situ* coprecipitation strategy, and the Nb^{5+} was occupied at the octahedral sites of the transition metal layer. The Li/Ni disorder and lattice oxygen release were suppressed by the strong Nb-O bonds, and the radial growth of primary particles was induced to form a densely packed microstructure, significantly homogenizing the strain distribution. As a result, a capacity retention of 95.1% was achieved after 100 cycles at 0.5 C at an elevated temperature of 55 °C, and the temperature of the thermally induced layered-to-spinel phase transition was raised to 200 °C. Zou et al. [49] employed a hafnium gradient doping strategy, and the Ni sites on the surface lattice of $\text{LiNi}_{0.9}\text{Co}_{0.05}\text{Mn}_{0.05}\text{O}_2$ were precisely regulated through the large ionic radius and high atomic mass of Hf^{4+} . The abrupt lattice contraction caused by the H2-H3 phase transition was effectively suppressed, and the c-axis variation was reduced from 0.79% to 0.69%. Furthermore, the formation energy of oxygen vacancies was increased through the strong Hf-O bonds, and the lattice oxygen release was remarkably reduced. As a consequence, a capacity retention of 95.0% was delivered after 100 cycles at 1 C and 30 °C, 93.2% retention was achieved after 100 cycles at 50 °C, and the thermally induced phase transition temperatures were elevated to 186 °C for the layered-to-spinel transition and 274 °C for the spinel-to-rock salt transition.

To address the microcracking issues caused by anisotropic lattice strain, the mechanical stress is effectively reduced through regulating the primary particle morphology and constructing single-crystal structures. For example, Han et al. [46] proposed a multistage co-doping strategy, in which Ti was uniformly doped into the lattice of $\text{Li}[\text{Ni}_{0.90}\text{Co}_{0.05}\text{Mn}_{0.05}]\text{O}_2$ to achieve structural stabilization, and Ta was subsequently introduced to induce grain-boundary segregation. Consequently, a capacity retention of 93.0% after 100 cycles was achieved at a high-temperature of 60 °C, and the exothermic peak temperature was elevated to 208.2 °C.

Mo-doped single-crystal $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$ particles with a size of approximately 230 nm were constructed by Liang et al. [47] using an electrospinning technique combined with an *in-situ* chelation strategy. Mo was uniformly incorporated into the bulk lattice while the layered structure was well preserved. The small single crystals reduced stress concentration and suppressed cracking, whereas Mo^{6+} doping further stabilized the lattice. As a result, a capacity retention of 77.2% was achieved after 2300 cycles at 55 °C (Fig. 4(h)), a capacity retention of 81.9% was delivered after 1500 cycles at -20 °C, and the exothermic peak temperature reached 289 °C (Fig. 4(i)).

Sb5 doping was introduced into $\text{LiNi}_{0.91}\text{Co}_{0.06}\text{Al}_{0.03}\text{O}_2$ by Lv et al. [54] through a high temperature solid-phase method, and no significant morphological change was induced in the Sb NCA91 (Figs. 5(a) and 5(b)). The primary particles of Sb NCA91 were radially aligned (Figs. 5(c) and 5(d)), and a well defined layered structure was exhibited after doping (Figs. 5(e)–5(h)). All elements were uniformly distributed throughout the Sb NCA91 particles, and no impurity phases were detected in the X ray diffraction pattern. Distinct peak splitting was exhibited by both the NCA91 and Sb NCA91 cathodes in the ranges of 37.5°–39.0° and 64.0°–65.5°, which corresponded to the (006)/(102) and (018)/(110) reflections, respectively. This indicates that a well developed layered structure and a high degree of crystallinity are possessed by both the NCA91 and Sb NCA91 samples. The Sb NCA91 cathode delivered a capacity retention of 88% after 200 cycles at 1 C in the voltage range of 3–4.3 V (Fig. 5(i)). A capacity retention of 84% was still maintained after 200 cycles under a high cut off voltage of 2.7–4.5 V (Fig. 5(j)), and a high capacity of 168.6 $\text{mAh}\cdot\text{g}^{-1}$ was still delivered by the Sb NCA91 cathode at 0.2 C when the temperature was lowered

to -20°C . The capacity retention at 0.5C could be elevated to approximately 88% over 100 cycles (Fig. 5(k)). Further simulation indicates that the reduced surface energy of the (003) facet induced by Sb doping (Figs. 5(l)–5(n)) promoted radial grain alignment. Such a microstructure shortened Li-ion transport pathways and mitigated stress concentration associated with the H2-H3 transition, resulting in suppressed cracking and improved structural stability.

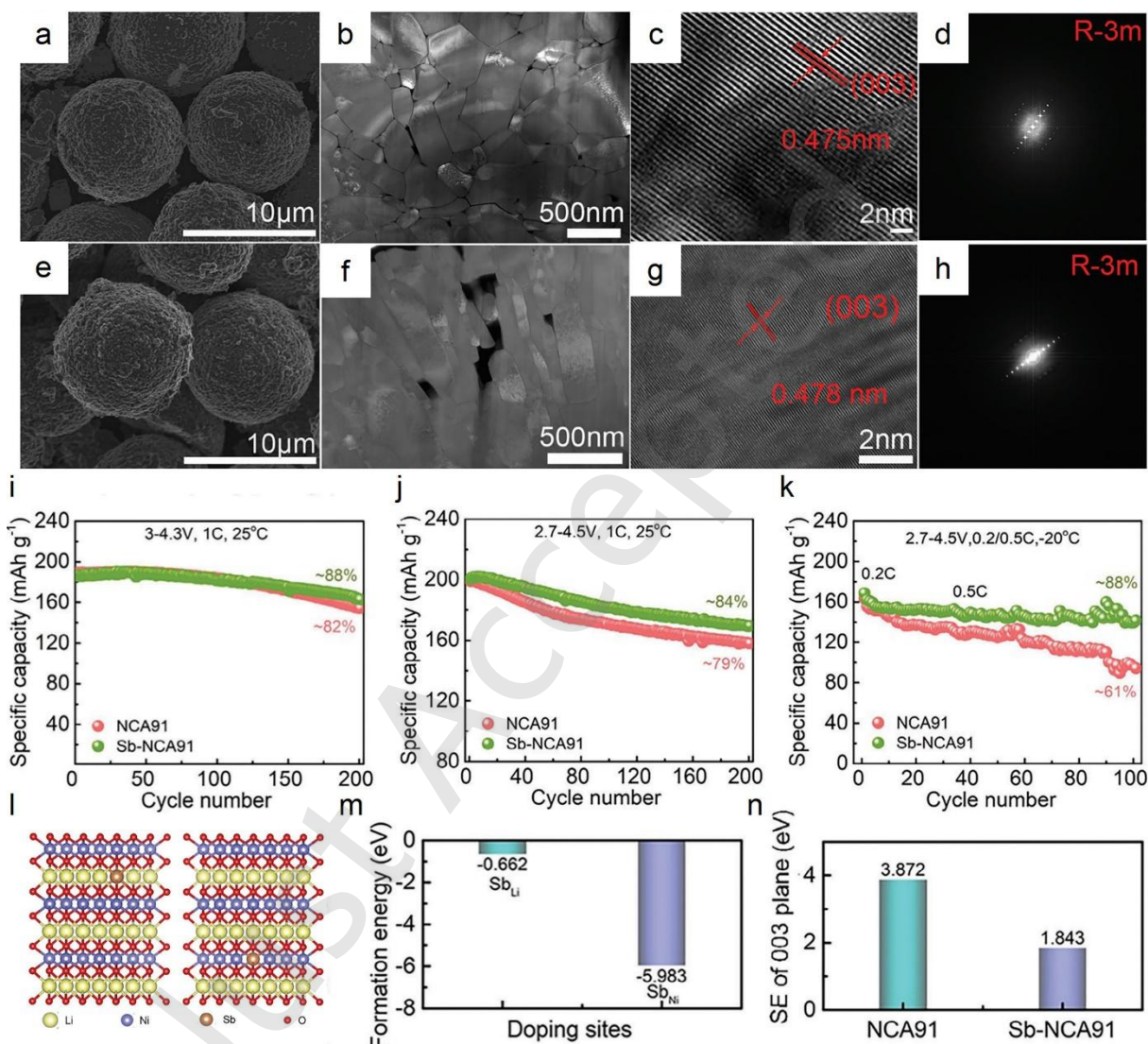


Figure 5 (a) SEM image of the NCA91 cathode. (b) SEM image of the Sb-NCA91 cathode. (c) Cross-sectional STEM image of the NCA91 cathode. (d) Cross-sectional STEM image of the Sb-NCA91 cathode. (e, f) Cross-sectional HRTEM images of the NCA91 cathode and corresponding FFT pattern. (g, h) Cross-sectional STEM image of the Sb-NCA91 cathode. (i, j) Cross-sectional HRTEM images of the Sb-NCA91 cathode and corresponding FFT pattern. (k)–(n) Cross-sectional EDS mapping images of the Sb-NCA91 cathode. (o) Cycling performance of NCA91 and Sb-NCA91 cells in 3–4.3 V. (p) Cycling performance of NCA91 and Sb-NCA91 cells in 2.7–4.5 V. (q) Cycling performance of NCA91 and Sb-NCA91 cells in 2.7–4.5 V at -20°C . (r) Structure of NCA91 cathodes with Sb at Li site and Ni site. (s) The formation energies of Sb substitution at different sites (Li and Ni). (t) Surface energy (SE) of (003) plane before and after Sb doping. (a)–(n) Reproduced with permission from Ref. [54], © Wiley-VCH GmbH 2024.

An excess of Mn was introduced into $\text{Li}[\text{Ni}_{0.910}\text{Co}_{0.079}\text{Al}_{0.011}]\text{O}_2$ by Park et al. [117], and the segregation of Mn at grain boundaries and the formation of secondary phases during high-temperature calcination were exploited to modulate the primary particle morphology and microstructure. As a result, a capacity retention of 76.5% was achieved after 1000 cycles under 3 C and an elevated temperature of 45 °C. Zhang et al. [120] proposed a high entropy doping strategy, and high valence doping elements were introduced into $\text{LiNi}_{0.8}\text{Mn}_{0.13}\text{Ti}_{0.02}\text{Mg}_{0.02}\text{Nb}_{0.01}\text{Mo}_{0.02}\text{O}_2$. A near-zero volume change (less than 0.3%) was realized during cycling, and the lattice strain along with microcrack formation can be significantly suppressed. As a consequence, a capacity retention of 96.6% was achieved after 50 cycles at an elevated temperature of 50 °C, the exothermic peak temperature was raised to 286 °C, and the total heat release was remarkably reduced to 530.0 g·g⁻¹.

The regulation of crystal facet orientation to increase active sites was reported as an effective strategy. Employing a wet co-doping strategy, Xiao et al. [50] simultaneously achieved uniform Al bulk doping and W surface enrichment in single-crystal $\text{LiNi}_{0.718}\text{Mn}_{0.271}\text{Al}_{0.010}\text{W}_{0.001}\text{O}_2$. Through surface energy regulation, a core-shell structure was successfully constructed, in which the (003)-oriented core was encapsulated by a (104)-faceted shell. The exposed (104) facets provided enlarged active sites for Li-ion storage, increasing the Li-ion occupancy ratio from 78.3% to 87.6% with alleviated interfacial polarization. Benefiting from this facet-regulated structure, the cathode delivered 92% retention of after 100 cycles at 1 C and 45 °C.

The relationship between Ni concentration gradients and primary-grain structures in ultrahigh-Ni layered cathodes was systematically investigated by Lu's group [128]. By tailoring the Ni distribution during precursor preparation, different primary-grain structures were obtained after sintering (Figs. 6(a)–6(e)). In the optimized material, internal stress was alleviated by the modified grain structure (Figs. 6(f) and 6(g)), while surface chemical stability was improved by the compositional gradient. Meanwhile, enhanced thermal stability was also achieved (Fig. 6(h)), indicating suppressed thermal decomposition and lattice oxygen release under high-voltage conditions. As a result, a specific capacity of 237.4 mAh·g⁻¹ was delivered at 0.1 C, and 176.2 mAh·g⁻¹ was retained after 500 cycles within 3.0–4.5 V. A high capacity of 138.7 mAh·g⁻¹ was also maintained at 10 C. These findings establish a clear relationship between concentration-gradient design and grain-structure evolution under industrial processing conditions, providing a feasible route to improve the high-voltage electrochemical performances of ultrahigh-Ni cathodes.

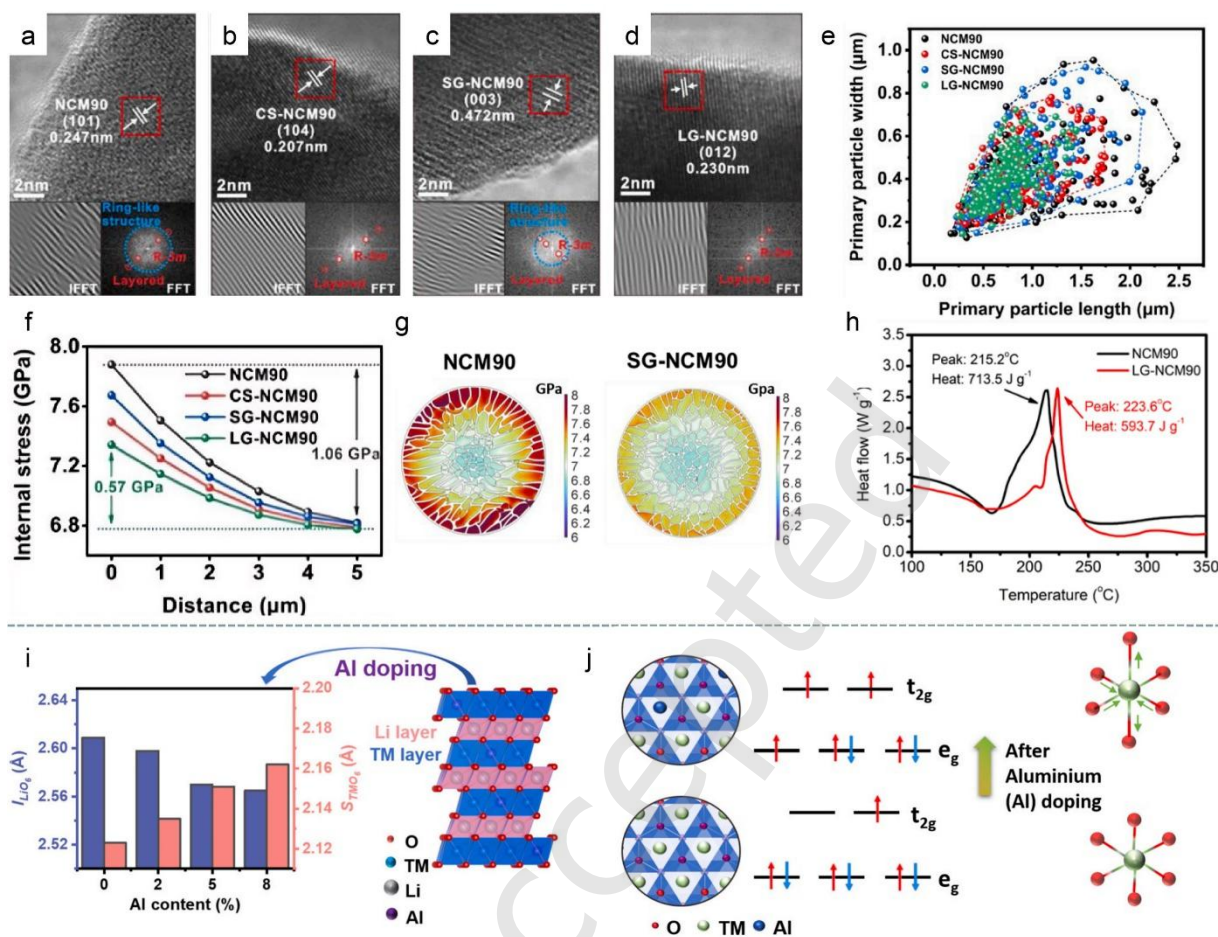


Figure 6 HRTEM images of (a) NCM90, (b) CS-NCM90, (c) SG-NCM90 and (d) LG-NCM90, with the insets showing the corresponding FFT and IFFT images obtained from the marked regions. (e) Length-width distribution of the primary grains. (f) Calculated average internal stress distribution. (g) Simulated cross-sectional stress profiles. (h) DSC profiles with corresponding oxygen release of at charged state of 4.5 V. (a)–(h) Reproduced with permission from Ref. [128], © Elsevier B.V. 2026. (i) Changes in I_{LiO_6} and S_{TMO_6} at the different Al doping amounts. (j) Illustration of the Ni 3d band structures after Al doping. (i–j) Reproduced with permission from Ref. [129], © Elsevier B.V. 2026.

In addition, they systematically investigated the effect of Al doping on the local structure, microstructural evolution, and mechanical stability of ultra-high-Ni cathodes [129]. With the Al content varied from 2 to 8 mol.%, pronounced changes were induced in the primary-grain morphology, microporous architecture, and local structural characteristics. Al incorporation redistributed the thicknesses of the Li and TM slabs (Fig. 6(i)), while also modulating the Ni 3d electronic structure together with the local Ni-O coordination (Fig. 6(j)). At an optimized Al content of 5 mol.%, the NiO₆ framework was reinforced, while intragranular micropores effectively buffered the volume strain generated during cycling. Consequently, the optimized cathode delivered an initial capacity of 198.7 mAh·g⁻¹ at 1 C, retained 86.6% of its initial capacity after 500 cycles within 3.0–4.5 V, and maintained 175.1 mAh·g⁻¹ at 10 C. These results demonstrate that the electrochemical performance can be effectively improved through the coordinated regulation of microstructural evolution and local chemical coordination, providing a practical approach for scalable cathode manufacturing.

Overall, bulk regulation has evolved from conventional atomic-scale doping toward multiscale control of local coordination, compositional distribution, primary-grain architecture, internal defects, and grain boundaries. These approaches can stabilize the layered framework, homogenize strain distribution, suppress crack initiation and propagation, as well as preserve Li-ion transport pathways. However, bulk regulation alone cannot fully control electrolyte decomposition and CEI evolution. Moreover, excessive dopant incorporation, defect generation, or secondary-phase formation may reduce electrochemical activity, disturb structural ordering, or impede Li-ion transport. Therefore, effective bulk regulation requires a balance among chemical stabilization, mechanical strain accommodation, and transport kinetics, while further coupling with surface/interfacial protection is often necessary for wide-temperature operation.

3.3 Synergistic modification

To address the poor cycling stability and severe structural degradation of high-Ni layered oxide cathodes under wide-temperature operating conditions, increasing attention has been focused on synergistic modification strategies. The synergistic integrate bulk regulation with surface/interfacial engineering, enabling coordinated control of the lattice, particle microstructure, surface chemistry, and CEI across a wide temperature range [80, 130–139].

For instance, Chu et al. [130] applied a one-step calcination method, and Sn^{4+}/F co-doping with a LiF surface coating were concurrently introduced into $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$. Through Sn^{4+} doping, the layered-to-spinel phase transformation was promoted and the local electronic structure was tailored, while through the reaction of F^- with surface residual lithium, an LiF coating was formed and TM-F bonding was created. Thus, the Li/Ni cation mixing was effectively reduced and the HF-induced erosion was restrained. Within the temperature range of 130–170 °C, the Ni^{2+} -O concentration in the near-surface structure is increased, and the disordered spinel phase is further transformed into the rock-salt phase (Figs. 7(a) and 7(b)). Compared with the pristine NCM811, the critical temperature at which the phase transformation occurs in NCM811-1SF is higher, a result that further confirms the enhanced lattice oxygen stability conferred by the Sn/F dual modification (Figs. 7(c) and 7(d)). As a result, a capacity retention of 85.4% was achieved after 300 cycles at 2.8–4.5 V and 1 C, a retention of 78.3% was delivered after 200 cycles at 55 °C, and a retention of 94.8% was obtained after 500 cycles in pouch full-cell. Zhang et al. [80] employed a one-step phytic acid coating treatment (Fig. 7(e)). A tripartite synergistic structure composed of an amorphous lithium phosphate coating layer, a subsurface reconstructed NiO layer, and a bulk gradient phosphorus doping was constructed from the surface to the bulk of $\text{LiNi}_{0.83}\text{Co}_{0.06}\text{Mn}_{0.11}\text{O}_2$ (Figs 7(f)–7(h)). In this architecture, rapid Li-ion transport pathways were provided and interfacial side reactions were blocked by the amorphous lithium phosphate layer, the subsurface structural stability was reinforced by the reconstructed NiO layer, and the interlayer spacing of LiO_6 slabs was expanded by the gradient phosphorus doping. Thus, a capacity retention of 72.4% was delivered after 300 cycles within 2.8–4.45 V at 1 C, a retention of 69.8% was achieved after 200 cycles at an elevated temperature of 45 °C (Fig. 7(i)), and a discharge capacity of 168.8 $\text{mAh}\cdot\text{g}^{-1}$ was obtained at a rate of 5 C. A synergistic doping and surface-modification strategy using Y was reported by Lu's group for ultra-high-Ni cathodes [140]. Y was found to primarily substitute Ni sites, thereby stabilizing the O framework. In addition, the surface-coated solid electrolyte layer can reduce interfacial side reactions and help maintain stable cycling at high potential and elevated temperature. Consequently, the modified cathode delivered 228.5 $\text{mAh}\cdot\text{g}^{-1}$ within

3.0–4.5 V and retained 90.7% of its capacity after 200 cycles. At 55 °C, a capacity retention of 80.4% was maintained after 200 cycles.

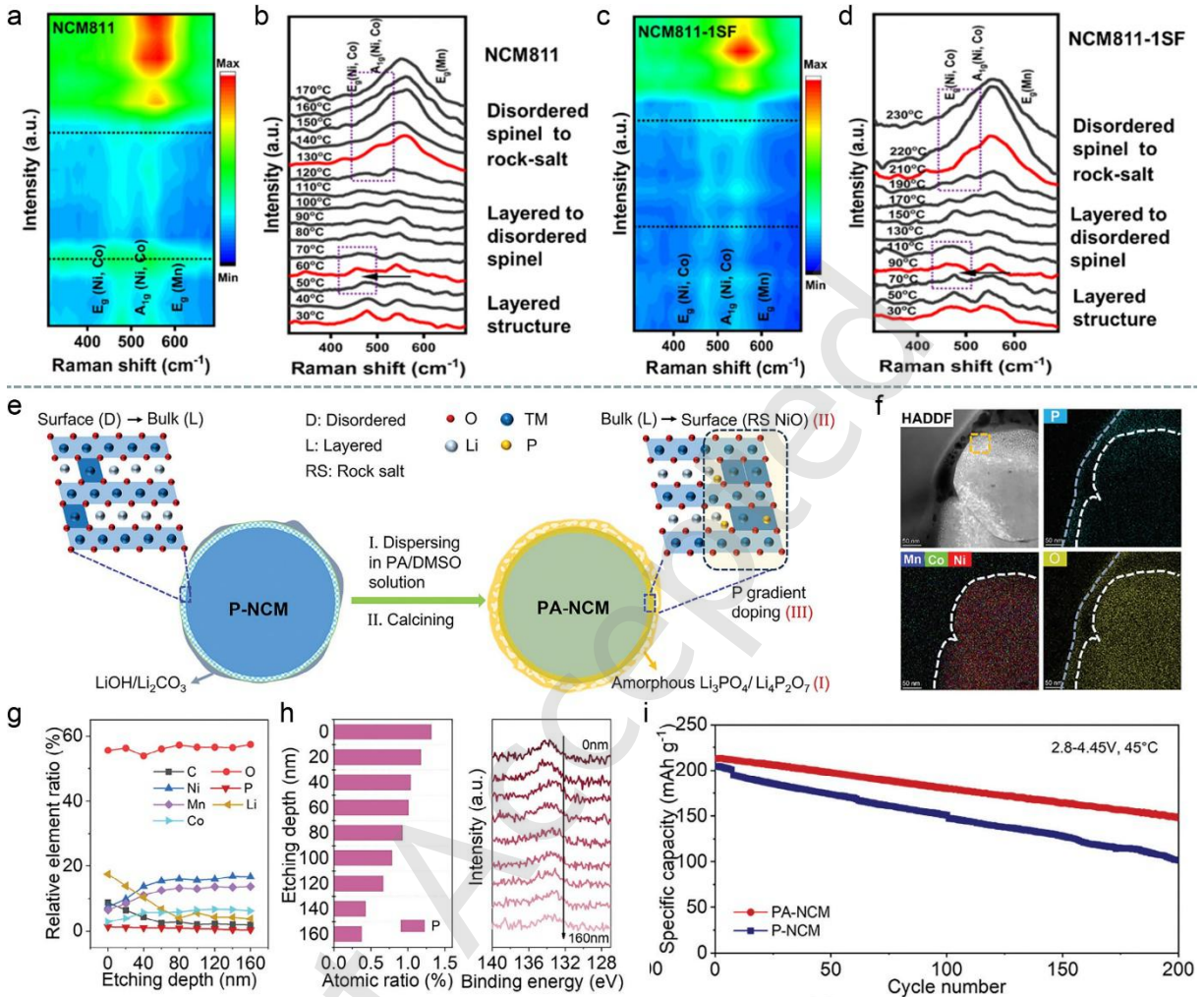


Figure 7 *In situ* Raman results of fully charged (a, b) NCM811 and (c, d) NCM811-1SF electrodes in the 5th cycle during the heating experiment. (a)–(d) Reproduced with permission from Ref. [130], © American Chemical Society 2024. (e) Surface to bulk synergistic modulation of Ni-rich cathode materials with phytic acid. Surface structure analysis of Ni-rich cathode materials with and without surface modification. (f) HADDF-STEM and EDS mapping (P, Mn/Co/Ni, and O) images of PA-NCM. (g) Element atomic ratios of C, O, Ni, Co, Mn, Li, and P in PA-NCM with different Ar-ion etching depths from XPS results. (h) Atomic ratios of P element obtained from XPS results and XPS depth profile of P 2p peaks of PA-NCM sample. Electrochemical performance of P-NCM and PA-NCM samples based on half cells. (i) Cycle performance at 1 C rate at 45 °C. (e)–(h) Reproduced with permission from Ref. [80], © Wiley-VCH GmbH 2024.

Overall, synergistic modification integrates bulk regulation with surface/interfacial engineering to concurrently suppress lattice instability, particle-level mechanical degradation, and interfacial deterioration. Coordinated control of local lattice chemistry, particle-scale strain evolution, surface reactivity, and CEI stability can simultaneously mitigate structural degradation and parasitic interfacial reactions. However,

the spatial distribution and relative amount of each regulating component are critical, since excessive bulk defects, segregated phases, or surface products may compromise structural ordering and Li-ion transport. Therefore, rational bulk and interface matching, rather than the simple superposition of individual modification strategies, is essential for effective wide-temperature stabilization.

3.4 Electrolyte optimization

As the medium for Li-ion transport and a direct participant in interfacial reactions, the electrolyte plays a critical role in determining the wide-temperature performance of high-Ni layered oxide cathodes. Under high-voltage and high-temperature conditions, electrolyte oxidative decomposition is accelerated, generating corrosive by-products that aggravate surface phase reconstruction, transition-metal dissolution, and interfacial impedance growth. Under low-temperature conditions, increased electrolyte viscosity and reduced ionic conductivity hinder Li-ion migration and desolvation, leading to sluggish interfacial charge transfer and enlarged polarization. To address these coupled challenges, electrolyte optimization has been widely explored through solvent-system design, Li-salt regulation, functional additive engineering, and localized high-concentration electrolyte construction. These strategies can suppress parasitic interfacial reactions at elevated temperatures while improving ionic transport and interfacial kinetics at low-temperatures, thereby enhancing the electrochemical stability of high-Ni cathodes across a wide temperature range [57, 58, 61, 63, 64, 68, 141–155].

The construction of stable CEI layers through the preferential decomposition of functional additives is recognized as one of the effective strategies to enhance the wide-temperature-range performance of high-Ni cathodes. For example, Chen et al. [61] designed a tris(2,2-difluoroethyl) borate additive, which was oxidatively decomposed at approximately 3.8 V and participated in the construction of a CEI rich in fluorinated organic species (Fig. 8(a)). Therefore, a capacity retention of 79.9% was achieved by the NCM811 | Li cell after 180 cycles at 0.5 C and 60 °C (Fig. 8(b)). Li et al. [151] introduced vinyltrimethylsilane (VTMS) as a multifunctional additive, and the HF was effectively scavenged. This contributes to constructing a robust CEI film, thus enabling the graphite | NCM811 full cell to retain 92.7% of its capacity after 100 cycles at 0 °C. Min et al. [154] proposed an "all-in-one" strategy, in which triallyl isocyanurate (TAC) was adopted as a multifunctional additive, and the high-temperature cycling life of pouch cells at 45 °C was significantly extended. Park et al. [156] synthesized lithium tetrafluoro (fluoromalonato) phosphate (LFMP) as a bifunctional electrolyte additive, which was decomposed on the NCM811 cathode to derive a stable CEI. Thus, the electrolyte decomposition and gas generation were suppressed, and a Li-rich SEI film was simultaneously induced on the graphite anode to resist PF₅ attack. As a result, the cycling performance of the NCM811 | graphite cell at 60 °C was enhanced. Zhang et al. [157] used 3-fluoro-5-(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)picolionitrile (FTDP) as an additive, and the NCM811 | Li cell was enabled to exhibit excellent cycling performance under an elevated temperature of 60 °C.

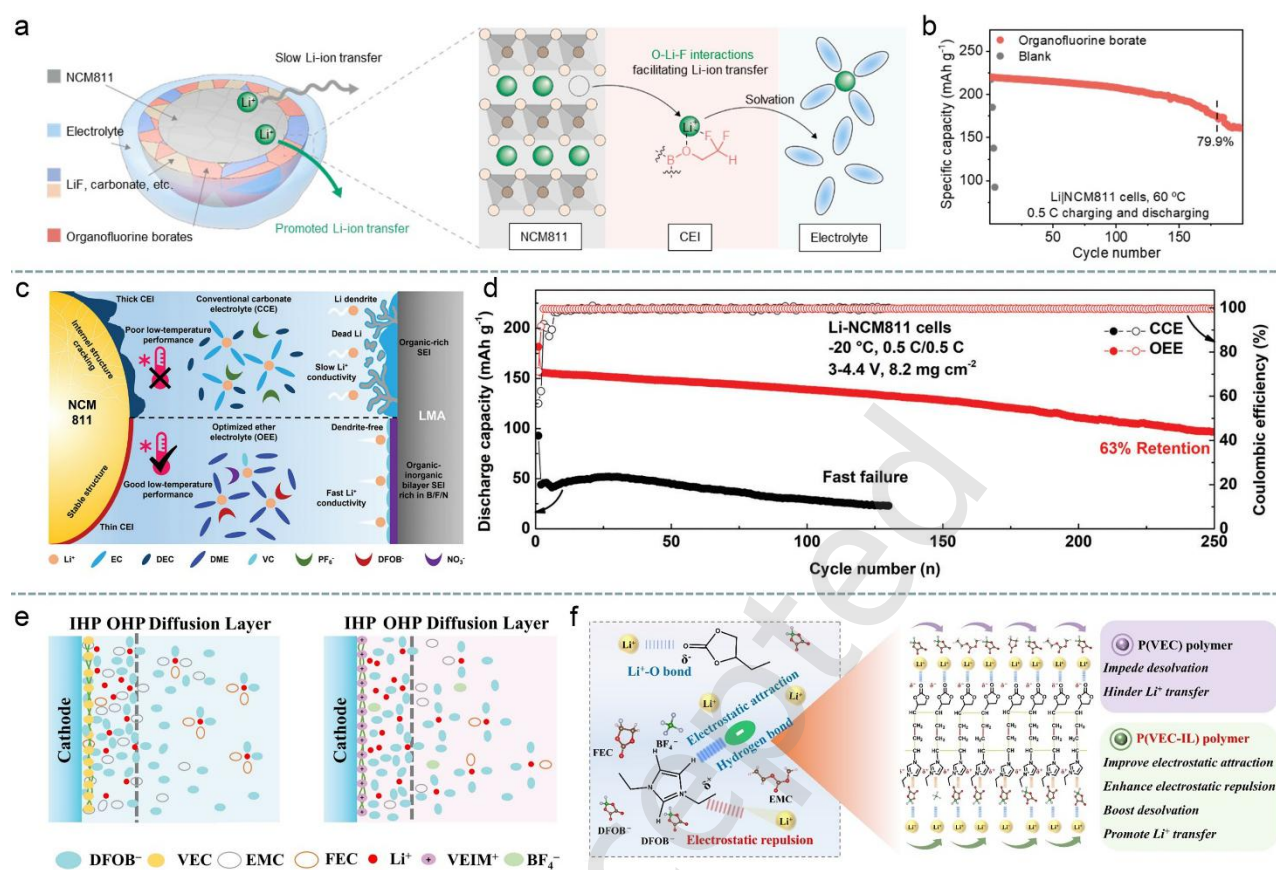


Figure 8 (a) Schematic of the organofluorine borate-involved interphase of NCM cathodes. (b) Cycling performance of Li NCM811 cells with and without organofluorine borate at 60 °C. (a, b) Reproduced with permission from Ref. [61], © Elsevier B.V. 2025. (c) Schematic diagram of the solvation structure and the corresponding interface chemistry of Li-NCM811 batteries with CCE or OEE. (d) Cycling performance comparison of Li-NCM811 full cell using different electrolytes at -20 °C (test conditions: 3–4.4 V, 0.5 C Charge and 0.5 C discharge, 1 C = 185 mA·g⁻¹, 8.2 mg·cm⁻²). (c, d) Reproduced with permission from Ref. [141], © Wiley-VCH GmbH 2023. (e) Schematic illustrations of EDL structure before and after introducing [VEIM][BF₄]. (f) Schematic illustration of the effects of P(VEC-IL) polymer skeletons on the Li⁺ transport and desolvation process. (e, f) Reproduced with permission from Ref. [67], © Wiley-VCH GmbH 2024.

Another effective approach is to regulate the Li ion solvation structure together with the properties of the electrolyte. Solvent optimization, diluent introduction, and electric double-layer modulation can reduce ion-transport barriers and facilitate interfacial kinetics. For instance, Chen et al. [68] optimized the Li-ion solvation environment with triethyl phosphate (TEP) and fluoroethylene carbonate (FEC), and the solvation number was reduced and the energy barrier for Li-ion transport was decreased. As a result, a capacity retention of 81.3% was delivered by the single-crystal NCM83 cathode after 200 cycles at 1 C and 45 °C. The introduction of a non-flammable multi-cyclic cosolvent, pentafluorophenyl diphenyl phosphate (PDP), by Zeng et al. [62] led to remarkable performance of the Li|NCM811 cell. An 80% capacity retention after 200 cycles at 4.4 V, a capacity of 126 mAh·g⁻¹ at -50 °C, and 93.4% capacity retention after 80 cycles at -20 °C were achieved. Moreover, a retention of 87.7% was attained after 100 cycles at an elevated temperature of 50 °C, demonstrating outstanding wide-temperature performance. Jiang et al. [141] optimized an ether-based electrolyte through the co-addition of LiNO₃ and vinylene

carbonate (VC) (Fig. 8(c)), and a discharge capacity exceeding $150 \text{ mAh}\cdot\text{g}^{-1}$ was delivered by the Li|NCM811 cell at 0.5 C and -20°C (Fig. 8(d)). Liu et al. [153] employed ethoxy (pentafluoro) cyclotriphosphazene (PFPN) as a diluent to construct a localized high-concentration electrolyte, and a capacity retention of 73.8% was retained by the NCM622|Li cell after 200 cycles at 50°C . Terreblanche et al. [158] adopted an EC-free electrolyte (1.5 M LiPF₆ in EMC with 10 wt.% FEC), and the lattice oxygen release was suppressed and the cycle life of the NMC811/graphite full cell was extended. Zhou et al. [67] proposed an electrolyte engineering strategy, and the electric double layer (EDL) structure was regulated through specific cation adsorption (Fig. 8(e)). In addition, a positively charged polymer layer was constructed within the inner Helmholtz plane (IHP) through 1-vinyl-3-ethylimidazolium cations (VEIM), and the cations were preferentially adsorbed on the NCM83 surface and a positively charged polymer protective layer was *in-situ* formed within the IHP, thereby inducing the formation of an anion-derived CEI with fast interfacial kinetics. Meanwhile, a positive electric field was provided by the polymer framework, and the transport and desolvation of Li were promoted (Fig. 8(f)). As a result, a reversible capacity of $130 \text{ mAh}\cdot\text{g}^{-1}$ was still maintained by the Li|NCM83 cell after 1000 cycles at 25°C with excellent capacity retention, and a capacity retention as high as 100% was achieved after 100 cycles at a low-temperature of -20°C . The visualized images of the NCM particles cycled with the P(VEC-IL) electrolyte revealed that the Li-ion concentration and stress distribution were uniform, and thus the formation of intergranular microcracks was effectively mitigated. As a consequence, the excellent electrochemical performance was exhibited by the Li|NCM83 cell under a high voltage of 4.5 V and over a wide temperature range from 30 to 70°C . Piao et al. [159] designed a quasi-solvated single-solvent electrolyte (QSE), and the Li|NCM811 cell was enabled to operate stably over a wide temperature range from -20°C to 60°C with a capacity retention of 82.8% after 180 cycles at a high voltage of 4.7 V.

By designing novel lithium salts or salt-blend systems, the high-temperature interfacial stability and the low-temperature ionic conductivity can be simultaneously improved through the multifunctional design of salt anions, such as film-forming capability, ionic conduction, and thermal stability, and their synergistic effects. For example, Cheng et al. [146] designed a ternary fluorinated lithium-salt electrolyte (TLE) consisting of LiDFOB, LiDFP, and LiPF₆, and thus an excellent cycling stability was exhibited under a high voltage of 4.7 V, an elevated temperature of 45°C , and a low-temperature of 0°C . Zhang et al. [65] constructed an intrinsically non-flammable LiDFOB/TMP electrolyte, and the full cell was endowed with excellent high-temperature stability and thermal runaway tolerance across a wide range from -20°C to 60°C . Moreover, Li et al. [64] designed a localized medium-concentration ionic liquid electrolyte with a monofluorinated piperidinium cation skeleton, and 90.3% retention was delivered by the NCM90|Li cell after 200 cycles at 1 C and 60°C .

The wide-temperature safety and interfacial compatibility of high-Ni cathodes are significantly improved when polymer-based electrolytes are employed. The mechanical compliance, adaptive capability, *in-situ* polymerizability, and inherent flame retardancy are possessed by the polymer framework. For instance, Chen et al. [144] prepared an amphiphilic copolymer electrolyte composed of hydrophobic C-F side chains and hydrophilic subunits, which was capable of self-assembling on the surface of high-Ni cathodes and being converted into a stable CEI. By blocking the lattice oxygen release pathway, the anion redox reaction was replaced by $\text{Co}^{3+}/\text{Co}^{4+}$ oxidation, and thereby the irreversible phase transition was suppressed. The NCM811/Li pouch cell was enabled to cycle safely and stably under a high voltage of 4.7 V and an elevated temperature of 120°C , demonstrating superior thermal stability. Lee et al. [149]

developed an anthracene-functionalized cyanoethyl poly (vinyl alcohol) (An-PVA-CN) gel polymer electrolyte, and the capacity retention of the NCM811|Gr full cell was elevated to 75% after 500 cycles at 45 °C. Li et al. [152] constructed a mechanically adaptive supramolecular poly(urethane-ureido-pyrimidinone) CEI, resulting in a capacity retention of 86.4% after 200 cycles at 50 °C and a discharge capacity of 160 mAh·g⁻¹ at -10 °C. Ye et al. [161] constructed a stable poly(ethylene oxide)-based electrolyte through an *in-situ* ultraviolet curing technique. The Li| NCM622 cell was enabled to cycle stably for over 260 cycles at a low-temperature of -20 °C and to operate reliably at a high-temperature of 60 °C. Zhang et al. [161] constructed a solid polymer electrolyte (SPE) with excellent compatibility toward high-loading high-Ni cathodes through the *in-situ* gradient polymerization of a deep eutectic electrolyte. The high-temperature safety of the solid-state electrolyte was significantly enhanced and the thermal runaway temperature was elevated from 198 °C to 312 °C.

Overall, electrolyte regulation is an important approach for improving the wide-temperature performance of high-Ni layered oxide cathodes. At elevated temperatures, preferential decomposition of functional additives can promote the formation of a stable, inorganic-rich CEI, thereby suppressing interfacial side reactions and transition-metal dissolution. At low temperatures, rational optimization of solvents and Li salts can reduce electrolyte viscosity, improve ionic transport, and accelerate interfacial reaction kinetics. These effects contribute to improved cycling stability and rate capability over a wide temperature range. Nevertheless, electrolyte regulation mainly modifies the electrolyte environment and CEI chemistry, with limited ability to directly suppress bulk structural degradation, including cation mixing, lattice distortion, and internal strain accumulation. Moreover, the function of a single additive is often insufficient to simultaneously meet the conflicting requirements of high-temperature interfacial stability and low-temperature ionic transport. Therefore, careful matching of additives, electrolyte formulations, and cathode structures remains essential for reliable wide-temperature operation.

3.5 Multi-field coupling modification

Beyond conventional modification strategies, multi-field coupling modification provides another active and adaptive pathways for stabilizing high-Ni layered oxide cathodes. This strategy aims to couple chemical bonding regulation with physical-field responses, including stress-induced electric fields and temperature-induced reverse thermal strain, thereby simultaneously modulating lattice oxygen stability, Li-ion transport kinetics, stress evolution, and interfacial structural integrity [105, 131, 162–165]. In particular, piezoelectric interfacial layers can convert cycling-induced mechanical stress into localized polarization electric fields, which reduce Li-ion migration barriers and alleviate polarization. Negative-thermal-expansion layers can generate reverse thermal strain in response to temperature variations, thereby compensating for lattice expansion, suppressing thermal-stress accumulation, and improving thermal safety. These studies indicate that functional interfacial layers can serve not only as passive protective barriers but also as active regulatory components capable of coordinating lattice chemistry, charge transport, mechanical strain, and thermal response in high-Ni cathodes.

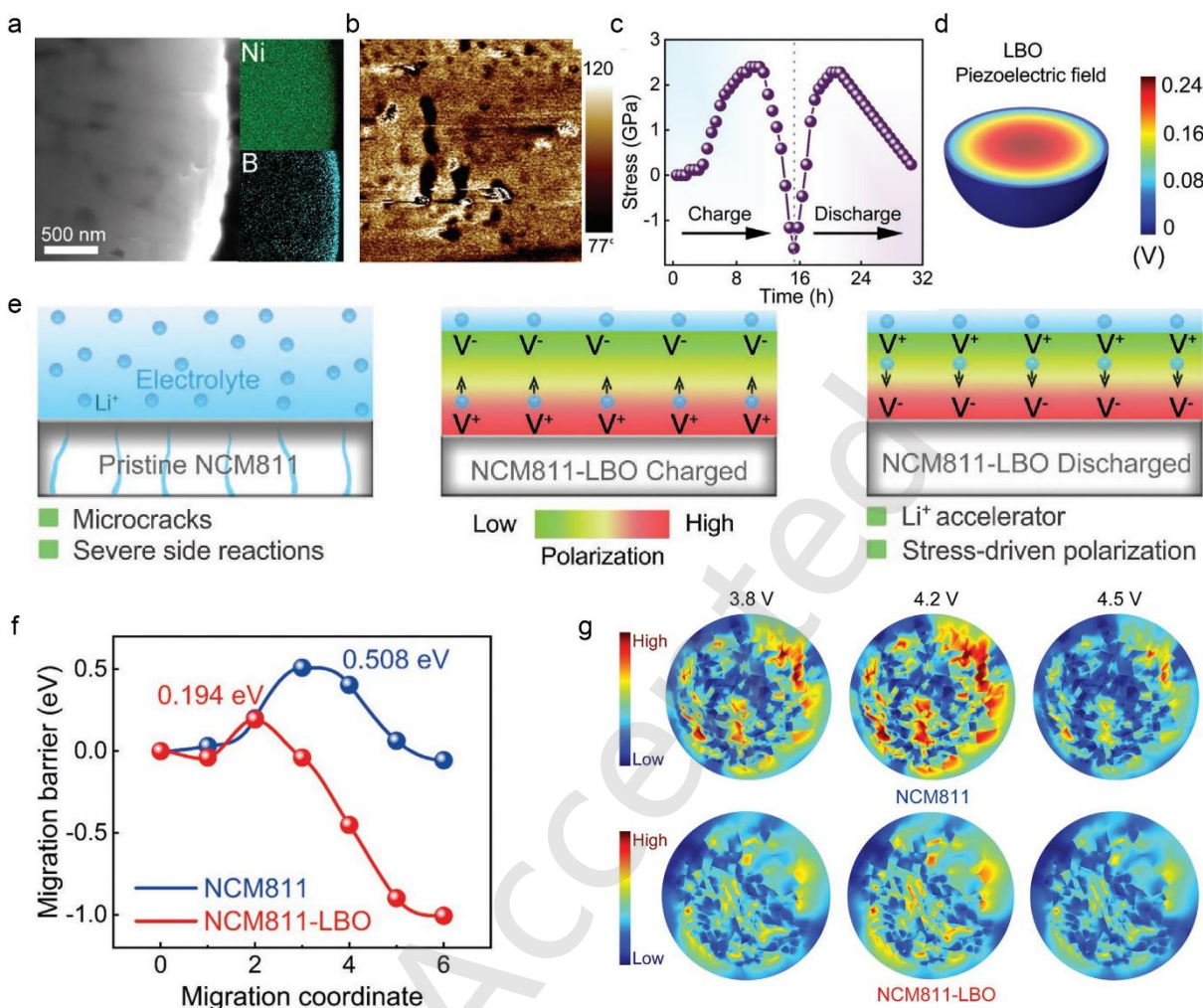


Figure 9 (a) Cross section SEM image with element mappings (Ni and B) of NCM811-LBO. (b) Phase images of LBO detected by PFM technique. (c) Stress curves for NCM811-LBO during initial cycle. (d) Finite element analysis of piezoelectric field distribution in LBO layer. (e) Schematic illustration of the regulation effect of piezoelectric LBO layer on Li⁺ transportation at NCM811 interfacial. (f) Corresponding diffusion energy barriers for NCM811 and NCM811-LBO. (g) NCM811 and NCM811-LBO particles at different voltage. (a)–(g) Reproduced with permission from Ref. [131], © Wiley-VCH GmbH 2022.

Through an *in-situ* construction strategy, a synergistic modification of gradient B doping with piezoelectric Li₂B₄O₇ (LBO) coating was realized in NCM811 cathode materials by our group (Figs. 9(a)–9(d)) [131]. Lattice oxygen release was effectively suppressed by B³⁺ at the electronic structure level. This suppression relied on high energy B–O bonds formed between boron and oxygen atoms. Meanwhile, a localized polarization electric field was induced by the piezoelectric LBO coating layer under cyclic stress. This field acted as an interfacial cation "self accelerator", significantly reducing the migration energy barrier for Li ions (Figs. 9(e) and 9(f)) while reducing stress accumulation (Fig. 9(g)). As a result, a capacity retention of 88.9% was achieved after 300 cycles at 25 °C and 1C. Under a high-temperature of 50 °C, a capacity retention of 87.9% was obtained at 1C. Furthermore, a specific discharge capacity of 112.2 mAh·g^{−1} was still delivered at a high rate of 10 C. Furthermore, a piezoelectric LiTaO₃ (LTO) functional layer was constructed on the surface of NCM811 and NCM622 cathodes by our group using a sol gel method [162]. The stress strain generated during cycling of the high-Ni material itself was utilized by this design. A localized polarization electric field was thereby induced in the LTO layer. An *in-situ*

mechanical electric field coupling modification was thus achieved. As a result, a capacity retention of 71.8% was achieved by NCM811 LTO after 200 cycles at 25 °C and 0.1C. This value was significantly higher than the 59.7% retention observed for the unmodified sample. A specific discharge capacity of 129.9 mAh·g⁻¹ was still attainable for this material, even at a high rate of 10 C.

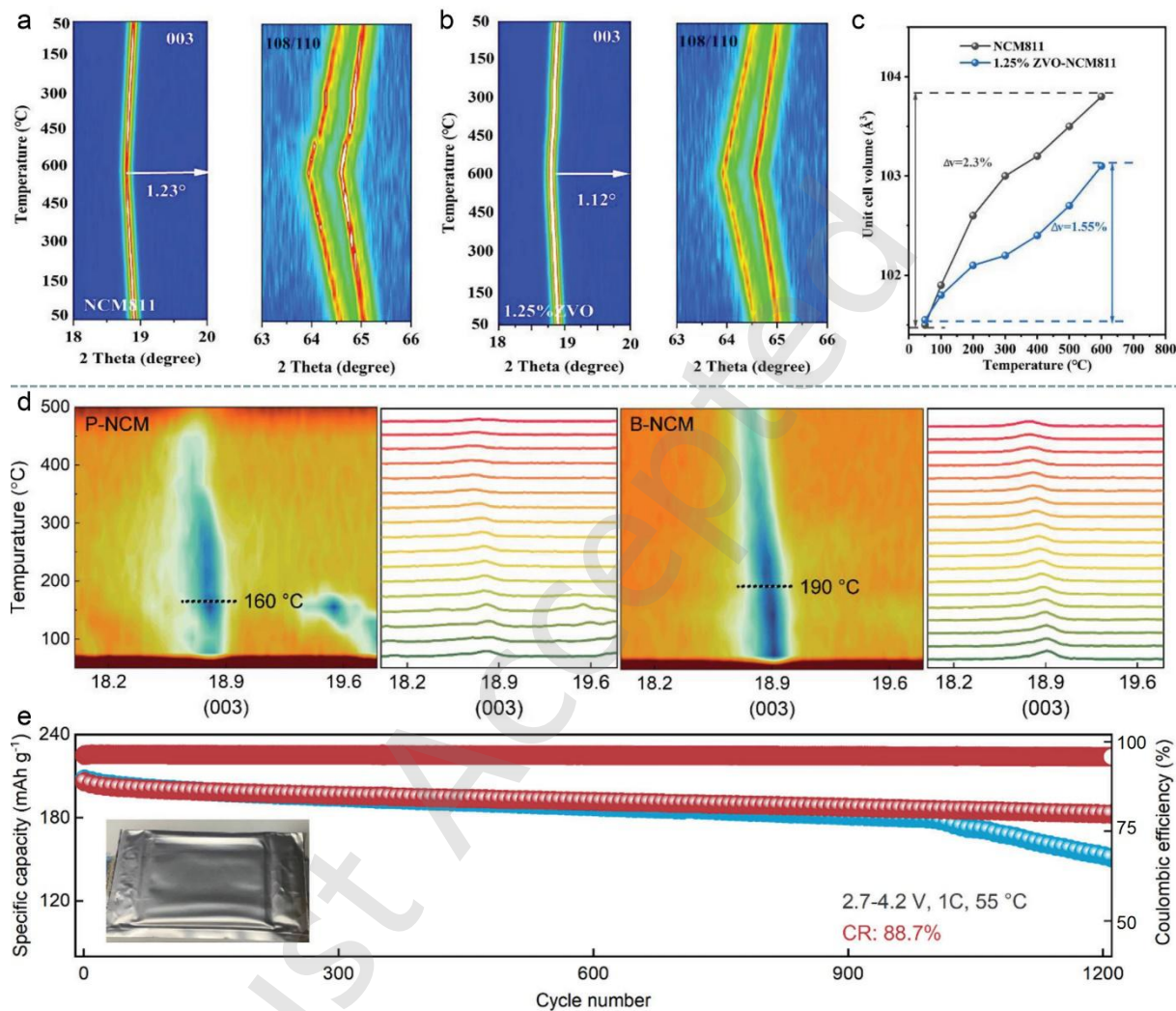


Figure 10 High temperature *in-situ* XRD of (a) NCM811 and (b) 1.25% ZVO-NCM811 at 50–600 °C in the degree of 18°–20° and 63°–66°. (c) The variation of the cell volume parameter during Heating for NCM811 and 1.25% ZVO-NCM811. (a)–(c) Reproduced with permission from Ref. [165], © Wiley-VCH GmbH 2023. (d) *In-situ* HT-XRD patterns for the delithiated cathodes at 4.5 V in the temperature range of 25–500 °C. *In-situ* DEMS data for (e) Cycling performances for P-NCM||Gr and B-NCM||Gr pouch cells at 55 °C under 2.7–4.2 V. (d, e) Reproduced with permission from Ref. [105], © Wiley-VCH GmbH 2024.

Moreover, Huang et al. [164] constructed a piezoelectric LiGaO₂ (LGO) modification layer with lattice compatibility on the surface of NCM811 cathodes. During charge-discharge processes, the lattice stress generated in the high-Ni material induced a built-in electric field within this LGO layer. Simultaneously, subsurface Ga doping was achieved, which stabilized the TM-O bonds. Electrochemical evaluations reveal that the NCM@LGO₂ cathode delivered 78.2% retention after 350 cycles at 25 °C and 5 C and

81.2% after 200 cycles at 55 °C and 1 C (compared to 68.1% for the pristine sample). Moreover, a capacity retention of 75.6% was maintained after 200 cycles at 1 C under a high cutoff voltage of 4.7 V. This piezoelectric modification strategy significantly enhances the cycling stability and high voltage tolerance of high-Ni cathodes under both room temperature and high-temperature conditions, owing to the suppression of lattice oxygen escape and the stabilization of the bulk structure.

In addition to mechanical electric coupling, thermal mechanical matching strategies have also been proven as effective means for enhancing the high-temperature stability of high-Ni cathodes. A coating of ZrV_2O_7 (ZVO) material, known for its negative thermal expansion property, was applied to the surface of NCM622 cathodes by our group [163]. The unique high-temperature volume contraction effect of this material was utilized. Lattice expansion and stress accumulation of the high-Ni material were thereby actively suppressed under cycling and high-temperature conditions. As a result, a capacity retention of 71.0% was achieved by the NCM622@1.0 wt.% ZVO sample after 500 cycles at 25 °C and 0.2 C. A specific capacity of 67.0% was maintained after 200 cycles at a high-temperature of 55 °C and 0.2 C. A capacity retention of 60.7% was obtained at 5 C. Further evidence was provided by *in-situ* XRD and theoretical calculations. Through constrained unit cell volume expansion and increased migration energy barriers for Ni ions, the ZVO coating layer effectively suppressed the formation of Li_2NiO_2 impurities and structural degradation. The structural stability and cycle life of high-Ni cathodes were thereby significantly enhanced under both room temperature and high-temperature conditions.

Furthermore, an *in-situ* constructed protective layer of ZrV_2O_7 (ZVO) with negative thermal expansion properties was applied to both the secondary particle surface and the primary particle grain boundaries of NCM811 cathodes by Nie et al. [165]. Intergranular microcrack propagation was suppressed and the irreversible transformation from the layered phase to spinel/rock salt phases was mitigated through the isotropic thermal contraction characteristics of ZVO. The isotropic NTE nature of ZVO endowed the 1.25% ZVO-NCM811 sample with considerably suppressed high-temperature XRD peak shifts relative to the bare NCM811 (Figs. 10(a) and 10(b)), a suppression that was especially evident for the (003) diffraction peak, where the shift values were 1.12° and 1.23° respectively. As the temperature was raised, the unit cell volume of NCM811 was expanded by 2.3% while that of 1.25% ZVO-NCM811 was expanded by only 1.55% (Fig. 10(c)), demonstrating that the lattice changes and the volumetric swelling were effectively restrained. The oxygen vacancy formation energy was simultaneously increased from 1.43 eV to 1.90 eV, and the lattice oxygen release was inhibited. Under a hazardous temperature of 120 °C, the Li-ion transport channel was actively shut down by this ZVO coating due to its negative thermal expansion behavior, with a charging capacity of only 28 $\text{mAh}\cdot\text{g}^{-1}$. The thermal runaway process of the battery was thereby effectively blocked. A synergistic optimization of structural and thermal safety was consequently achieved under high voltage, high-temperature, and overheating conditions.

A BaZrO_3 (BZO) thermal barrier layer with an ultralow thermal conductivity of $3.42\text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ was deposited on the surface of the NCM811 cathode by our group [105]. A portion of Zr^{4+} were simultaneously incorporated into the lattice, and high bond energy Zr-O bonds were formed with lattice oxygen to enhance the electronegativity of oxygen. *In-situ* high-temperature XRD revealed that the phase transition temperature from the layered structure to the disordered structure was elevated from approximately 160 °C to approximately 190 °C by the BZO modification (Fig. 10(d)). The effective suppression of O_2 release at high voltage by the BZO modification was confirmed by *in-situ* differential

electrochemical mass spectrometry. Electrochemical tests demonstrated that a capacity retention of 92.5% was achieved by the B-NCM| graphite pouch cell after 1400 cycles at 25 °C and 1 C, and a retention of 88.7% was still maintained after 1200 cycles at a high-temperature of 55 °C (Fig. 10(e)), while a capacity retention of 84.3% was retained even after 100 cycles at an extreme temperature of 80 °C. Therefore, the thermal barrier layer can effectively block the rapid heat exchange between the electrode and the electrolyte, and thus the severe surface side reactions at high-temperatures were alleviated.

Overall, multi-field coupling modification provides an active and adaptive pathway for improving the electrochemical performance and structural stability of high-Ni layered oxide cathodes. Through the coupling of chemical bonding regulation, stress-induced polarization electric fields, negative-thermal-expansion responses, and thermal-barrier effects, these strategies can simultaneously stabilize lattice oxygen, accelerate interfacial Li-ion transport, alleviate stress accumulation, suppress thermal expansion, and improve thermal safety under room-temperature and high-temperature conditions. Nevertheless, the development of multi-field coupling modification is still at an early stage. First, most reported studies mainly focus on room-temperature or high-temperature performance, while systematic evaluation under low-temperature and temperature-fluctuation conditions remains insufficient. Second, the functional parameters of piezoelectric, thermal-barrier, and negative-thermal-expansion layers, such as polarization intensity, piezoelectric coefficient, thermal conductivity, thermal expansion coefficient, phase-transition temperature, and strain reversibility, have not yet been quantitatively matched with the practical service-temperature range of high-Ni cathodes. Third, the introduction of functional layers may increase interfacial impedance or cause interfacial incompatibility if their thickness, crystallinity, ionic conductivity, and chemical stability are not properly controlled. Therefore, future studies should emphasize temperature-dependent functional-parameter regulation, operando verification of field-coupling mechanisms, and integrated design with doping, coating, and electrolyte optimization. By constructing temperature-field-adaptive functional interfaces with balanced ion transport, structural buffering, and thermal regulation capabilities, multi-field coupling modification is expected to provide a forward-looking strategy for wide-temperature high-Ni layered oxide cathodes.

4 Summary and outlook

The wide-temperature degradation of high-Ni layered oxide cathodes is essentially a coupled process involving thermal, electrical, mechanical, and electrochemical factors. At elevated temperatures, bulk lattice expansion, oxygen instability, and irreversible phase reconstruction are accompanied by accelerated electrolyte decomposition and cathode/electrolyte interfacial degradation, including CEI growth, surface corrosion, transition-metal dissolution, and gas evolution. In contrast, low-temperature degradation is dominated by kinetic limitations arising from reduced solid-state Li-ion transport within the bulk structure, deteriorated electrolyte ionic transport, and sluggish interfacial desolvation/charge-transfer kinetics. To address these distinct degradation characteristics, diverse optimization strategies have been developed, including surface coating, bulk regulation, synergistic modification, and electrolyte optimization. As summarized in Table 1, these efforts have achieved remarkable progress in extending the operating-temperature range and improving the electrochemical performance of high-Ni cathodes. However, most reported strategies still rely on static regulation, and therefore remain effective primarily within specific temperature windows. Such limited adaptability to temperature variations makes it difficult to simultaneously address the distinct degradation mechanisms at high and low temperatures.

Table 1 Summary of representative modification strategies for wide-temperature high-Ni layered oxide cathodes

Cathode	Modification method	Temperature (°C)	Cycling performance	Capacity (mAh·g ⁻¹)	Ref.
LiNi _{0.83} Co _{0.11} Mn _{0.06} O ₂	Coating	45	91.3%/120 th /0.5 C	197.2/0.5 C	[41]
LiNi _{0.8} Co _{0.1} Mn _{0.1} O ₂	Coating	55	86.3%/100 th /0.33 C	~242.0/0.1 C	[43]
LiNi _{0.83} Mn _{0.1} Co _{0.07} O ₂	Coating	45	85.3%/100 th /0.5 C	174.9/0.5 C	[45]
LiNi _{0.8} Co _{0.1} Mn _{0.1} O ₂	Coating	60	91.3%/150 th /1 C	159.9/1 C	[100]
LiNi _{0.90} Co _{0.05} Mn _{0.05} O ₂	Doping	60	~92.0%/100 th /0.5 C	~230.0/0.5 C	[46]
		55	77.2%/2300 th /1 C	~150.0/1 C	
LiNi _{0.6} Co _{0.2} Mn _{0.2} O ₂	Doping	-20	81.9%/1500 th /1 C	~132.0/1 C 116.4/5 C	[47]
LiNi _{0.9} Co _{0.05} Mn _{0.05} O ₂	Doping	50	93.2%/100 th /1 C	174.0/7 C	[49]
LiNi _{0.718} Mn _{0.271} Al _{0.010} W _{0.001} O ₂	Doping	45	92.0%/100 th /1 C	141.7/10 C	[50]
LiNi _{0.91} Co _{0.06} Al _{0.03} O ₂	Doping	-20	88.0%/100 th /0.5 C	~138.0/0.5 C	[54]
LiNi _{0.8} Mn _{0.18} Fe _{0.02} O ₂	Doping	55	84.5%/100 th /1 C	147.3/1 C	[55]
LiNi _{0.9} Co _{0.1} O ₂	Doping	55	95.1%/100 th /0.5 C	229.0/0.1 C 169.1/5 C	[56]
LiNi _{0.874} Co _{0.076} Al _{0.010} Mn _{0.040} O ₂	Doping	45	76.5%/1000 th /3 C	~142.0/3 C	[117]
LiNi _{0.8} Mn _{0.13} Ti _{0.02} Mg _{0.02} Nb _{0.01} Mo _{0.02} O ₂	Doping	50	96.6%/50 th /0.5 C	210.1/0.1 C	[120]
LiNi _{0.87} Mn _{0.1} Mg _{0.01} Al _{0.01} Fe _{0.005} Ti _{0.005} O ₂	Doping	50	99.0%/100 th /1 C	213.3/0.2 C	[121]
LiNi _{0.83} Co _{0.06} Mn _{0.11} O ₂	Synergistic modification	45	69.9%/200 th /1 C	~148.0/1 C 168.8/5 C	[80]
LiNi _{0.8} Co _{0.1} Mn _{0.1} O ₂	Synergistic modification	30	85.4%/300 th /1 C	169.0/1 C 163.5/10 C	[130]

		60	79.9%/180 th /0.5 C	~220.0/0.5 C	
LiNi _{0.8} Co _{0.1} Mn _{0.1} O ₂	Electrolyte optimization	30	80.1%/120 th /0.5 C	--	[61]
		-10	86.4%/100 th /0.5 C	~125.0/0.5 C	
LiNi _{0.8} Co _{0.1} Mn _{0.1} O ₂	Electrolyte optimization	50	87.7%/100 th /1 C	168.0/1 C	[62]
LiNi _{0.9} Co _{0.05} Mn _{0.05} O ₂	Electrolyte optimization	60	~84.8%/80 th /1 C	~200.0/1 C	
		-20 to 30	—	110.0 to 215.0/0.1 C	[65]
NCM83	Electrolyte optimization	70	~69.3%/100 th /2 C	138.0/2 C	
		-20	~91.5%/100 th /0.05 C	~132.0/0.05 C	[67]
NCM83	Electrolyte optimization	45	81.3%/200 th /1 C	162.0/1 C	[68]
LiNi _{0.8} Co _{0.1} Mn _{0.1} O ₂	Electrolyte optimization	-20	63.0%/250 th /0.5 C	~96.0/0.5 C	[141]
LiNi _{0.8} Co _{0.1} Mn _{0.1} O ₂	Electrolyte optimization	45	87.0%/400 th /1 C	~186.0/1 C	
		0	88.0%/500 th /1 C	~148.0/1C	[146]
LiNi _{0.6} Co _{0.2} Mn _{0.2} O ₂	Electrolyte optimization	50	~70.3%/200 th /0.2 C charge-0.5 C discharge	~144.0/0.5 C	[153]
LiNi _{0.8} Co _{0.1} Mn _{0.1} O ₂	Electrolyte optimization	60	60.0%/200 th /1 C	152.0/5 C	[156]
LiNi _{0.8} Co _{0.1} Mn _{0.1} O ₂	Electrolyte optimization	60	~84.2%/200 th /0.5 C	~170.0/0.5 C	[157]
LiNi _{0.8} Co _{0.1} Mn _{0.1} O ₂	Electrolyte optimization	-20 to 60	--	~152.0 to 224.0/0.2 C	[159]
LiNi _{0.8} Co _{0.1} Mn _{0.1} O ₂	Electrolyte Optimization	60	77.5%/200 th /1 C	~168.0/1 C	
		-20 to 60	—	~152.0 to	[64]

				232.0/0.1 C	
LiNi _{0.6} Co _{0.2} Mn _{0.2} O ₂	Electrolyte optimization	30	66.4%/1600 th /1 C	87.5/5 C	[160]
	Multi-field coupling modification	55	82.5%/100 th /1 C	~144.0/1 C	
LiNi _{0.8} Co _{0.1} Mn _{0.1} O ₂		80	80.2%/100 th /1 C	~150.0/1 C	[105]
	Multi-field coupling modification	50	87.9%/100 th /1 C	~164.0/1 C	[131]
LiNi _{0.6} Co _{0.2} Mn _{0.2} O ₂	Multi-field coupling modification	55	66.6%/200 th /0.2 C	115.2/0.2 C	[163]
LiNi _{0.8} Co _{0.1} Mn _{0.1} O ₂	Multi-field coupling modification	55	~79.2%/200 th /1 C	168.7/10 C	[164]
LiNi _{0.8} Co _{0.1} Mn _{0.1} O ₂	Multi-field coupling modification	60	90.5%/100 th /0.5 C	182.8/0.5 C	[165]

In this context, temperature-field-adaptive multiphysics interfaces provide a promising concept for wide-temperature regulation. Unlike conventional passive protective layers, these functional interfaces are expected to respond to service-temperature variations or cycling-induced physical perturbations, and actively regulate Li-ion transport, interfacial charge transfer, lattice strain evolution, CEI stability, and thermal safety. Their effectiveness is closely associated with service-temperature-dependent physical parameters. For ferroelectric and piezoelectric materials, polarization intensity, dielectric response, piezoelectric coefficient, coercive field, and Curie temperature determine whether effective electric-field regulation can be maintained within the target operating-temperature window. For negative-thermal-expansion materials, the thermal expansion coefficient, phase-transition temperature, and strain reversibility govern their ability to compensate for lattice expansion or contraction. Therefore, rational matching between functional-parameter windows and battery service temperatures should be regarded as a prerequisite for applying multiphysics-coupled interfacial regulation to wide-temperature high-Ni cathodes. Future research should be focused on the following aspects (Fig. 11).

4.1 Deepening the understanding of wide-temperature degradation

Wide-temperature degradation of high-Ni layered cathodes involves coupled changes in the bulk lattice, interface, and electrolyte. Future studies should move beyond single-temperature tests to clarify how these processes evolve and interact during temperature variations. Operando characterization, together with AI-assisted analysis of multimodal data, could help correlate lattice strain, oxygen activity, interfacial impedance, electrolyte decomposition, and crack evolution, thereby identifying the dominant degradation factors under different temperature conditions.

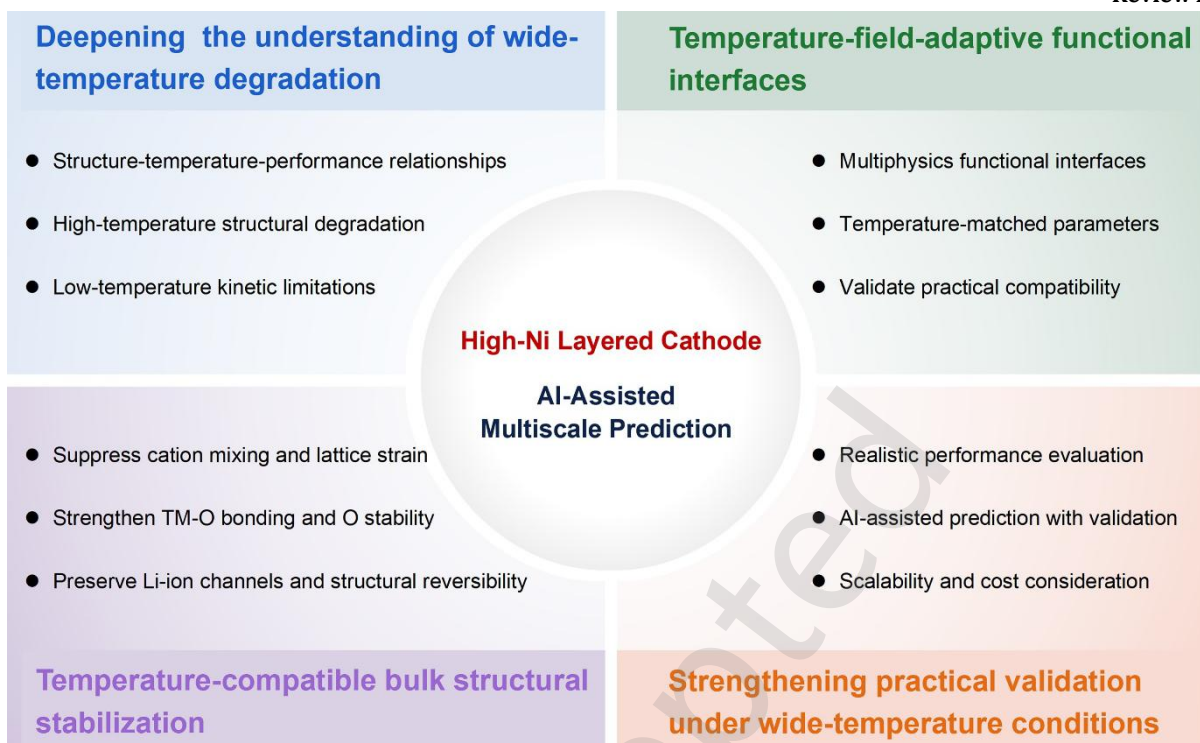


Figure 11 Outlook for the development of wide-temperature high-Ni layered oxide cathodes.

4.2 Designing temperature-adaptive functional interfaces

Temperature-adaptive interfaces should be designed by matching their physical response with the actual operating-temperature range. Key parameters, including polarization intensity, piezoelectric coefficient, thermal expansion coefficient, and Seebeck coefficient, need to be considered together with electrochemical compatibility and Li-ion transport. AI-assisted screening combined with theoretical calculations and experiments could accelerate the selection of interface materials with suitable functional parameters for specific temperature windows.

4.3 Developing temperature-compatible bulk structural stabilization

Bulk structural regulation for wide-temperature operation should account for the different structural requirements at high and low temperatures rather than optimize the cathode for a single temperature. Future structural design should therefore seek a balance between high-temperature lattice stability and low-temperature Li-ion accessibility across the targeted temperature range. AI-assisted screening, combined with theoretical calculations and experimental validation, could further identify structural parameters that satisfy these temperature-dependent requirements.

4.4 Strengthening practical validation under wide-temperature conditions

Wide-temperature performance should be evaluated under conditions closer to practical operation, including high mass loading, lean electrolyte, high voltage, fast charging, temperature fluctuations, long-term cycling, and pouch-cell configurations. Large datasets obtained under the different temperatures and operating conditions could further be used for AI-assisted lifetime and performance prediction, while the resulting models should be continuously checked against realistic cell-level experiments. In addition, the manufacturability, synthesis scalability, and cost should also be included in the evaluation.

Overall, future research should focus on resolving coupled degradation under dynamic temperature conditions, matching functional interfaces with service-temperature windows, improving temperature-compatible bulk structures, and validating these strategies under practical cell conditions. The deep integration of advanced characterization, calculations, AI-assisted analysis, and experimental validation could further improve the efficiency and reliability of design.

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Declaration of conflicting interests

The authors declare no conflicting interests regarding the content of this article.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and/or the Supplementary Materials. Additional data related to this paper may be requested from the corresponding author upon request.

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