

Review

Jahn-Teller Effect in Cathodes for Rechargeable Metal-Ion Batteries: A Review

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Abstract: The Jahn-Teller effect is an origin of structural instability in many cathodes for secondary ion batteries. The primary Jahn-Teller active centers in cathodes are usually high-spin Mn^{3+} , low-spin Ni^{3+} , and high-spin Fe^{4+} , which are usually considered one of the roots of lattice strain, phase transitions, particle cracking, and transition-metal dissolution. However, emerging studies reveal that regulated Jahn-Teller distortion can also facilitate alkali-ion diffusion, mitigate lattice strain, and enhance structural reversibility. Therefore, the underlying mechanisms of the Jahn-Teller effect on the structural stability and ion diffusion of cathodes should be viewed from both perspectives. This review summarizes the current reports of the Jahn-Teller effect across diverse cathodes and highlights its dual role as both detrimental and beneficial. The regulation strategies are summarized, including cationic doping, anionic substitution, structural and interfacial engineering, electrolyte optimization, and the rational harnessing of beneficial distortion. By bridging underlying mechanisms with modification strategies, this review offers a reference for the future design of high-performance cathodes in next-generation secondary ion batteries.

Keywords: Jahn-Teller effect; secondary ion batteries; energy storage materials; cathodes; structural degradation

1. Introduction

Secondary-ion batteries, such as lithium-ion batteries (LIBs) and sodium-ion batteries (SIBs), have played an important role in contemporary energy storage systems, powering portable electronics, electric vehicles, and large-scale storage [1–3]. Their widespread adoption is built on several key properties, including high energy density, extended cycle life, and superior operational safety. The electrode materials fundamentally determine these properties, and the cathode is the most critical component, as it mainly decides the specific capacity and operating voltage of the full cells [4–6]. The development of cathodes that can reversibly store a large quantity of ions over extended cycling without structural degradation is essential for next-generation batteries with improved sustainability and performance.

However, many high-energy cathodes suffer from severe capacity and voltage fading during cycling, which hinders their practical application [7,8]. This degradation primarily originates from the structural instability of their transition-metal oxide frameworks, such as $Mn^{3+}O_6$ octahedra [9,10]. Among the various intrinsic mechanisms, the Jahn-Teller effect is a fundamental and pervasive source of lattice destabilization. First formulated in molecular physics, the Jahn-Teller theorem dictates that any nonlinear molecule or crystal in an electronically degenerate ground state undergoes a spontaneous geometrical distortion (the Jahn-Teller distortion)



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that lifts the degeneracy and lowers the symmetry, thereby reducing the total energy [11,12]. In many cathodes, this distortion becomes especially damaging because it links electronic distribution to long-range lattice strain, often leading to irreversible structural degradation.

The detrimental consequences of the Jahn-Teller distortion are not just symmetry-breaking distortion. The elongation of metal-oxygen bonds weakens the stability of transition metals in the lattice, promoting their dissolution into the electrolyte, a process especially remarkable in cathodes containing Mn^{3+} and Ni^{3+} [13,14]. Dissolved transition-metal ions not only deplete the active element in cathodes but also migrate to the anode, where they catalyze the decomposition of the solid-electrolyte interphase (SEI) and result in capacity loss [15,16]. Furthermore, the Jahn-Teller effect may drive phase transitions, which are often accompanied by large volume changes that induce mechanical stress, thus leading to particle pulverization and electrode failure [17,18]. This microscopic cracking also creates new surfaces for accelerating the side reactions with the electrolyte, exacerbating impedance growth and gas release, and resulting in safety issues [19,20]. However, the Jahn-Teller distortion is not entirely harmful. For instance, Li et al. [21] found that it can assist sodium diffusion for high-performance sodium-ion batteries. Thus, rather than viewing Jahn-Teller distortion solely as a degradation mechanism, a deep understanding that recognizes both its detrimental and potentially beneficial roles will be critical for the rational design of high-performance cathodes. Many studies have focused on specific material classes, often employing advanced characterization techniques and theoretical calculations to elucidate the influence of the distortion. So far, a view that integrates the fundamental theory, experimental results, and materials' manifestations with the emerging mitigation strategies is needed to guide rational cathode design.

In this review, we aim to provide an integrated perspective on the Jahn-Teller effect in energy storage materials for secondary ion batteries. First, the fundamental theory of the Jahn-Teller effect is described, which includes its quantum-mechanical origin and the thermodynamic and kinetic conditions under which it manifests in crystalline solids. A systematic survey of its manifestations in representative cathodes is then conducted, covering not only well-established cases (for example, LiMn_2O_4 spinel and layered LiNiO_2) but also emerging systems such as disordered rock-salts and Prussian blue analogs for rechargeable ion batteries. Finally, we review the strategies developed to suppress or harness the Jahn-Teller effect, including elemental doping, compositionally graded structures, surface coatings, and the design of high-entropy or cation-disordered materials that disrupt the cooperative nature of the distortion [22]. By synthesizing the current understanding and highlighting the remaining challenges, this review can help to guide the rational design of structurally stable, high-performance cathodes for the next generation of secondary ion batteries.

2. Concept of Jahn-Teller Effect and Secondary Ion Batteries

An understanding of the fundamental concept of the Jahn-Teller effect is an essential precondition for analyzing its roles in secondary battery cathodes and developing effective modification strategies.

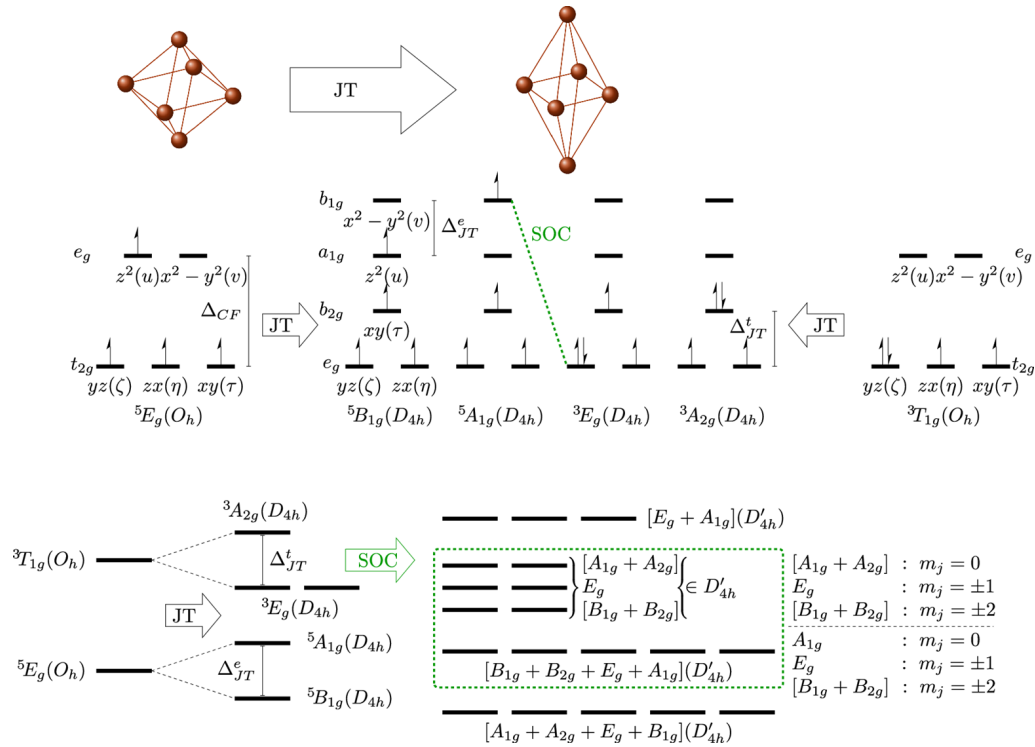
2.1. Concept of the Jahn-Teller Effect

The Jahn-Teller effect was proposed by Jahn and Teller in 1937. In a symmetric nonlinear molecule, if the ground state of the system possesses multiple degenerate energy levels, the system will spontaneously undergo a distortion that lifts the degeneracy, thereby lowering the symmetry and the energy. The d orbitals of a transition metal ion split into t_{2g} and e_g orbitals with an energy difference Δ_0 .

Taking the layered structure as an example, which may have Mn^{3+} (more detailed properties of Jahn-Teller active cations are listed in Table 1), situated in an octahedral coordination environment [23,24]. In an octahedral field, the t_{2g} orbitals are lower in energy than the e_g orbitals due to ligand repulsion. Because oxygen provides a weak-field environment (the orbital splitting energy $\Delta_0 < P$, the pairing energy), the electrons occupy the d orbitals with parallel spins according to Hund's rule, filling the lower-energy t_{2g} orbitals first and then the e_g orbitals. Consequently, as shown in Figure 1, Mn^{3+} adopts a $(t_{2g})^3(e_g)^1$ electronic configuration, which gives rise to four short bonds in the xy plane and two long bonds along the z -axis, causing the regular octahedron to elongate along the z direction. In layered structures, this octahedral distortion induced by the Jahn-Teller effect is referred to as Jahn-Teller distortion and is one of the key factors affecting the structural stability of layered cathodes [25]. Therefore, suppressing Jahn-Teller distortion is a challenge in the modification of layered cathodes.

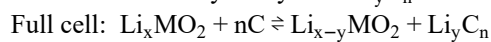
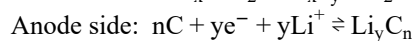
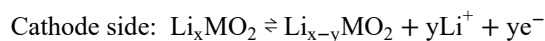
Table 1. Properties of Jahn-Teller active cations.

Cation	3d Orbital Electron Arrangement		Impact on the Microstructure
	e_g	t_{2g}	
Mn ³⁺ (High spin)	1	3	Single e_g electron occupies d_{z^2} , weakening axial σ -antibonding; central cation repels two axial O ²⁻ outward while drawing four equatorial O ²⁻ inward.
Cu ²⁺	3	6	Hole resides in $d_{x^2-y^2}$ axial elongation lowers d_{z^2} energy to fill electrons, pushing two axial O ²⁻ outward
Ni ³⁺ (Low spin)	1	6	Same mechanism as Mn ³⁺ , axial σ^* -antibonding drives central cation to repel axial O ²⁻
Fe ⁴⁺ (High spin)	1	3	Same mechanism as Mn ³⁺ , Dynamic buckling of Fe ⁴⁺ O ₆ octahedra along z-axis
Cr ²⁺ (High spin)	1	3	Same mechanism as Mn ³⁺ , central Cr ²⁺ repels axial O ²⁻

**Figure 1.** Occupation schemes of the 3d-derived orbitals of the relevant electronic states. Reprinted with permission from Ref. [23], copyright 2024, Springer Nature.

2.2. Concept of Secondary Batteries

One unique advantage of secondary-ion batteries is the reversible alkali-ion intercalation/deintercalation from host materials, which allows for the charge and discharge process, often referred to as the rocking chair architecture [26–28]. Usually, taking SIBs and LIBs as examples, the framework of the full cell contains a cathode, an anode, an electrolyte, and a separator. Ions can be deintercalated from the cathode and form new compounds at the anode while charging, and the reverse process happens during discharging. The theoretical maximum work output of a cell is equivalent to the Gibbs free energy change ($-\Delta G$), a thermodynamic quantity that originates directly from the chemical potential difference between the anode and cathode electrodes. As shown in Figure 2, taking a cell with a Li⁺-layered oxide and a carbon-based anode as an example, its working mechanism can be described as follows:



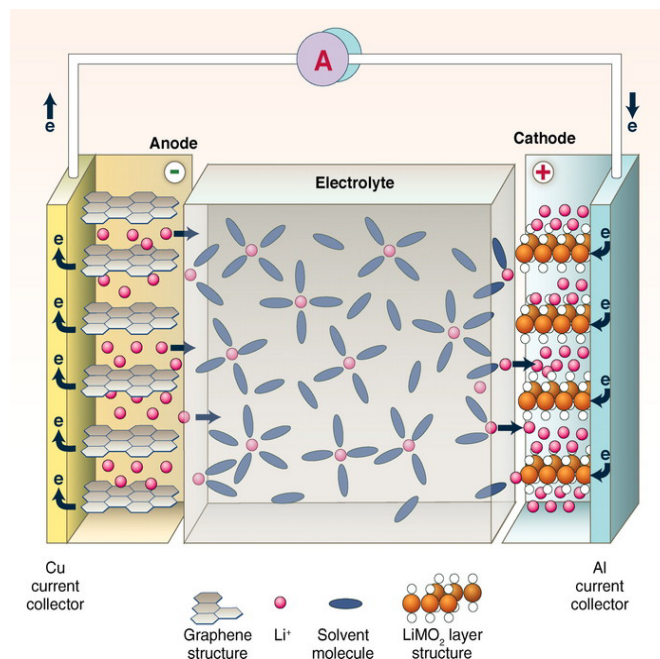


Figure 2. Schematic of Li-ion batteries. Reprinted with permission from Ref. [3], copyright 2011, American Association for the Advancement of Science.

The equations reveal that the cathode plays a decisive role in the capacity of full cells. It is a key determinant of cell voltage and capacity.

3. Influence of Jahn-Teller Effect on Cathodes

The Jahn-Teller effect manifests distinctly in different cathodes, driven by the electronic configuration of transition-metal ions and their local coordination environments. In octahedral oxide lattices, Jahn-Teller active ions such as high-spin Mn^{3+} , low-spin Ni^{3+} , and high-spin Fe^{4+} lift electronic degeneracy through tetragonal distortion, elongating axial metal-oxygen bonds while shortening equatorial ones [29]. This bond anisotropy renders axial oxygen atoms more electron-rich and Lewis basic, leading to such mechanisms for the possible material failure pathway:

(1) Chemical dissolution [30,31]. In all cathode systems containing Jahn-Teller active ions, the strongly basic axial oxygen atoms are prone to proton attack by HF, which is commonly generated from the hydrolysis of PF_6^- based electrolytes. Protonation of axial oxygen is accompanied by charge transfer from the metal d orbitals to oxygen, oxidizing Mn^{3+} to Mn^{4+} (or analogous oxidation of other Jahn-Teller ions). The resulting high-valent metal ion is unstable and undergoes a disproportionation-like reduction to a soluble lower-valent state (Mn^{3+} to Mn^{4+} and Mn^{2+}), which then dissolves into the electrolyte. Dissolved transition-metal ions migrate to the anode and disrupt the SEI. Furthermore, water generated as a byproduct of protonation promotes further salt hydrolysis, creating a vicious cycle. This acid-induced dissolution cascade is common to all Jahn-Teller-active cathodes, regardless of their crystal structure.

(2) Structure degradation [32,33]. Besides chemical dissolution, the local Jahn-Teller distortion can also trigger collective structural instabilities when the concentration of active ions exceeds a threshold. In spinel LiMn_2O_4 , for example, cooperative Jahn-Teller distortion transforms the cubic lattice into a tetragonal phase, causing a large volume change and particle fracture. In layered oxides (including LiNiO_2 , its derivatives, and Li-rich layered oxides), local non-cooperative distortions generate anisotropic strain that leads to intergranular cracks and exposes fresh surfaces to electrolyte attack.

(3) Migration and dissolution [34,35]. Layered cathodes suffer from an additional degradation mechanism enabled by their unique two-dimensional architecture. Jahn-Teller distortions can lower the migration barrier for transition metal ions, promoting their migration from the transition metal layer into the alkali metal layer. Once these ions reside in the alkali metal layer, they become more accessible to the electrolyte and are then dissolved via the universal chemical pathway described above (HF attack, oxidation, and disproportionation) [36,37]. This migration-dissolution coupling is particularly pronounced for Cr^{2+} , Fe^{4+} , Mn^{3+} , and other Jahn-Teller-active ions under high-voltage operation and can be partially suppressed by controlled doping. The process thickens the SEI, increases internal resistance, and shortens cycle life.

In this section, a detailed discussion is conducted on the impact of the Jahn-Teller effect on today's typical electrode materials.

3.1. On Spinel Oxide Cathodes

Spinel-structured LiMn_2O_4 and its derivatives suffer severely from Jahn-Teller distortion, especially under deep discharge or elevated temperatures [38–40]. When the temperature decreases, the entropic contribution to the system diminishes, and the relative importance of strain energy increases, thereby inducing the Jahn-Teller distortion in samples with high Mn^{3+} content (average oxidation state around +3.5) [41]. As the Li content increases, the average oxidation state of Mn rises, which causes the onset temperature of the Jahn-Teller distortion to shift to lower temperatures and its exothermic peak intensity to diminish. For samples with $x > 1.00$ (where the average Mn valence is about 3.54), the distortion is completely suppressed, even when the material is cooled down to $-50\text{ }^\circ\text{C}$. Furthermore, repeated thermal cycling around the transition region leads to a progressive decrease in the onset temperature and peak magnitude during the first few cycles; after about four cycles, these values stabilize at a lower level. However, further cycling results in a gradual recovery of the peak intensity and sharpness, indicating a thermal hysteresis behavior that is not simply reversible.

When in a deep discharge state, Mn^{4+} is reduced to Mn^{3+} during cycling; the high-spin d^4 configuration ($t_{2g}^3 e_g^1$) spontaneously breaks the local octahedral symmetry. Axial Mn-O bonds elongate while equatorial bonds shorten, relieving the electronic degeneracy. When the Mn^{3+} content exceeds a threshold, cooperative Jahn-Teller distortion propagates across the lattice, triggering a cubic-to-tetragonal phase transition accompanied by a large volume change. This anisotropic lattice strain causes particle microcracking, structural collapse, and electrical disconnection. Meanwhile, as described above, elongated and weakly bonded axial oxygen atoms exhibit enhanced Lewis basicity, making them vulnerable to proton attack by HF generated from electrolyte hydrolysis. The resulting Mn dissolution and subsequent SEI degradation on the anode accelerate capacity fading in spinel cathodes. Therefore, Jahn-Teller distortion in spinel links structural instability, chemo-mechanical failure, and interfacial side reactions into a self-reinforcing degradation pathway.

The electrochemical performance of the spinel LiMn_2O_4 cathode can be significantly improved after the Jahn-Teller distortion is suppressed. For example, Haruna et al. [42] used microwave irradiation to suppress the Jahn-Teller distortion in the spinel LiMn_2O_4 cathode. Microwave irradiation converts Jahn-Teller active Mn^{3+} to inactive Mn^{4+} in LiMn_2O_4 , increasing the $\text{Mn}^{4+}/\text{Mn}^{3+}$ ratio from 42:58 to 57:43 and the average Mn oxidation state from +3.42 to +3.57. It also reduces the lattice parameter and increases the BET surface area. These changes effectively suppress the Jahn-Teller distortion caused by the cubic to tetragonal phase transition during charge-discharge processes. This mechanism significantly improves the electrochemical performance. At 0.1 C, the specific capacity increases from 120.5 to 145.9 mAh g^{-1} . After 100 cycles, the capacity loss rate decreases from 0.034% to 0.022% per cycle. The charge transfer resistance is nearly halved, and the exchange current density increase twofold (from 1.3×10^{-6} to 6.6×10^{-7} A cm^{-2}). Moreover, the characteristic peaks of Jahn-Teller distortion in cyclic voltammetry do not intensify with increasing cycle number.

In another case, as shown in Figure 3, Zhang et al. [43] addressed the issue that, in this spinel cathode, the Jahn-Teller effect induced by Mn^{3+} leads to manganese dissolution and interfacial side reactions. Through surface modification with boric acid (H_3BO_3) to construct a boron-based polyanions, the Mn^{3+} content on the material surface is effectively reduced (from 54.35% to 50.05%), thereby alleviating the Jahn-Teller distortion, suppressing the distortion of MnO_6 octahedra, and the dissolution of manganese ions. Meanwhile, boron species react with PF_6^- in the electrolyte to form stable BF_4^- , reducing HF generation and forming a uniform and stable cathode-electrolyte interphase (CEI), further enhancing interfacial stability. The modified BO3-LMO achieves a capacity retention of 80.6% after 900 cycles at 1 C (compared to 61.1% for the unmodified sample), 83.2% after 1000 cycles at 5 C, and 85.1% after 200 cycles at 1 C under elevated temperature ($55\text{ }^\circ\text{C}$). DFT calculations further reveal that boron modification strengthens Mn-O bonding, reduces the number of antibonding states, and lowers the Li^+ migration energy barrier, thereby suppressing the Jahn-Teller effect from the electronic structure perspective and improving structural stability.

Therefore, suppressing the Jahn-Teller distortion is required to stabilize the MnO_6 octahedral framework and maintain cycling stability in spinel cathodes. The two examples above, microwave irradiation and boric acid surface modification, show that suppression can be achieved either by increasing the Mn valence state in the bulk or by stabilizing the surface against HF attack. Both strategies reduce the concentration or reactivity of Mn^{3+} , the primary Jahn-Teller center in spinel. Microwave treatment alters the bulk composition but is difficult to scale. Surface modification preserves bulk capacity but does not remove the distortion driving force within the particle interior. Thus, for spinel modification, combining bulk and surface stabilization strategies may address both the

cooperative phase transition and the chemical dissolution cascade, rather than mitigating one degradation pathway at the expense of the other.

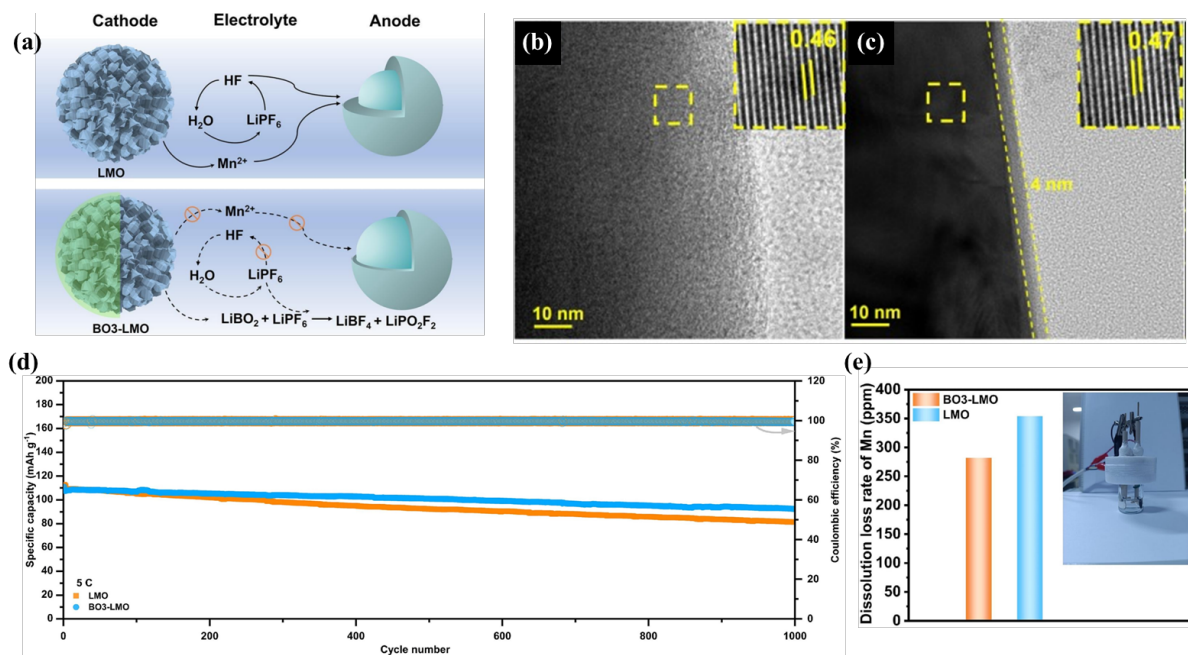


Figure 3. (a) Schematic of the surface reconstruction method. (b,c) HRTEM morphology of samples with (left) and without (right) surface layer. (d) Cycling performance of LMO and BO₃-LMO cathode. (e) Manganese dissolution of LiMn₂O₄ and BO₃-LMO. Reprinted with permission from Ref. [43], copyright 2025, Elsevier Inc.

3.2. On Layered Transition-Metal Oxide Cathodes

Layered oxides, including O3-type Li-ion cathodes, P2/O3-type Na-ion cathodes, and P3-type K-ion cathodes (Figure 4), usually exhibit system-dependent Jahn-Teller distortion [44–47]. Their structural feature is the periodic alternating arrangement of transition metal layers and alkali metal layers. The transition metal ions in the transition metal layer exhibit the Jahn-Teller effect at specific valence states, leading to Jahn-Teller distortion, such as the distortion of a certain transition metal-oxygen octahedron. The primary Jahn-Teller-active centers are high-spin Mn³⁺, low-spin Ni³⁺, and high-spin Fe⁴⁺, all carrying asymmetrically occupied orbitals. Usually, the Jahn-Teller distortion of the MnO₆ octahedron can trigger irreversible phase transitions, Mn dissolution/disproportionation reactions, induced interface degradation, and crack formation in the layered cathode.

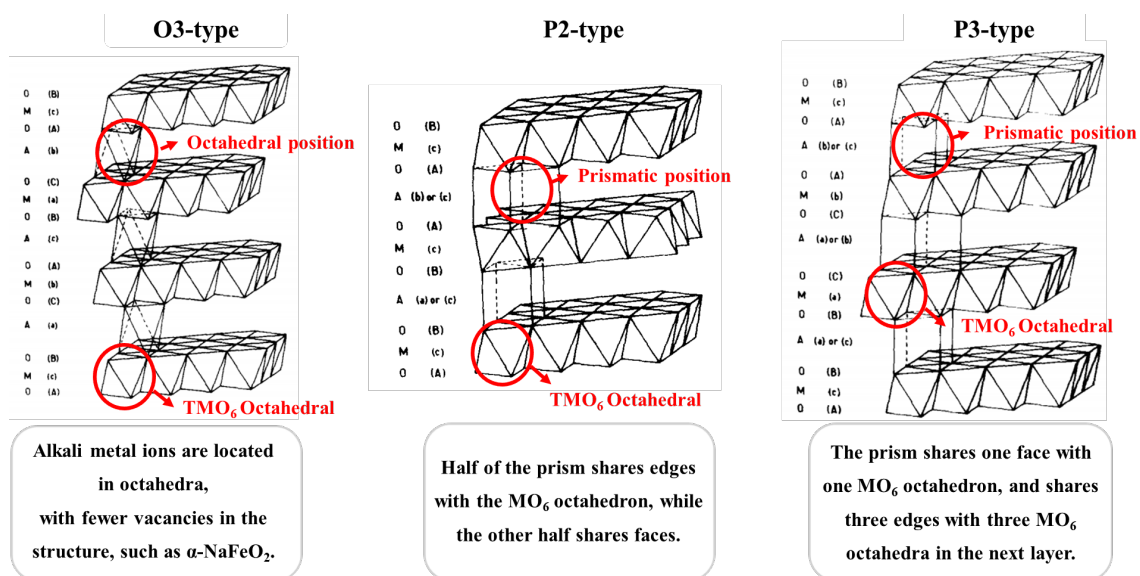


Figure 4. Structure schematic of the O3, P2, and P3-type cathode. Reprinted with permission from Ref. [48], copyright 1980, with permission from Elsevier B.V.

In layered cathodes for LIBs, the most widely studied cathodes are O3-LiNiO₂, O3-LiCoO₂, and their derivative materials. Owing to the Ni³⁺, the LiNiO₂ cathode is significantly affected by the Jahn-Teller distortion. In the LiNiO₂ cathode, this type of distortion has the property of being temperature-sensitive [49,50]. By combining ab initio molecular dynamics (AIMD) simulations with variable-temperature synchrotron X-ray diffraction (VT-XRD), Genreith-Schrieffer et al. [51] revealed the true temperature-dependent evolution of Jahn-Teller distortions and the nature of the phase transition in lithium nickelate (LiNiO₂). Anti-site defects (Ni/Li interchange) pin the undistorted domains at low temperatures, effectively preventing the establishment of long-range cooperative distortions, thereby explaining why real LiNiO₂, unlike NaNiO₂, does not show macroscopic symmetry reduction. As shown in Figure 5b, the VT-XRD results (the broadening of the (104)/(113) peaks, the monoclinic lattice parameter ratio $a_{\text{mon}}/b_{\text{mon}}$, and the changes in the 2θ angle) fully support this phase transition path and indicate that the monoclinic distortion exists only within short-range domains. In Figure 5c,d, it is found that at low temperatures (<250 K), LiNiO₂ exhibits static, cooperative Jahn-Teller distortions, characterized by the 4 short and 2 long Ni-O bond configurations in the NiO₆ octahedra. In the room-temperature range of approximately 250–350 K, the system enters a unique mixed-phase regime, where distorted and undistorted domains dynamically coexist, and the “pseudorotations” of the distortion axes do not originate from a conventional order-disorder transition but rather emerge as a side effect of the formation and disappearance of undistorted domains. When the temperature exceeds 350 K, the system enters a high-temperature displacive phase, where the octahedra no longer exhibit local Jahn-Teller distortions (van Vleck analysis shows that the most probable state is undistorted (Figure 5a), the Ni-O bond lengths display a single broad distribution, and the residual asymmetry mainly arises from anisotropic thermal vibrations. This work not only clarifies the dynamic mechanism of Jahn-Teller distortions in LiNiO₂ but also proposes that the mixed-phase regime of a displacive phase transition can exhibit pseudo-order-disorder characteristics, providing an important paradigm for understanding other Jahn-Teller-active materials.

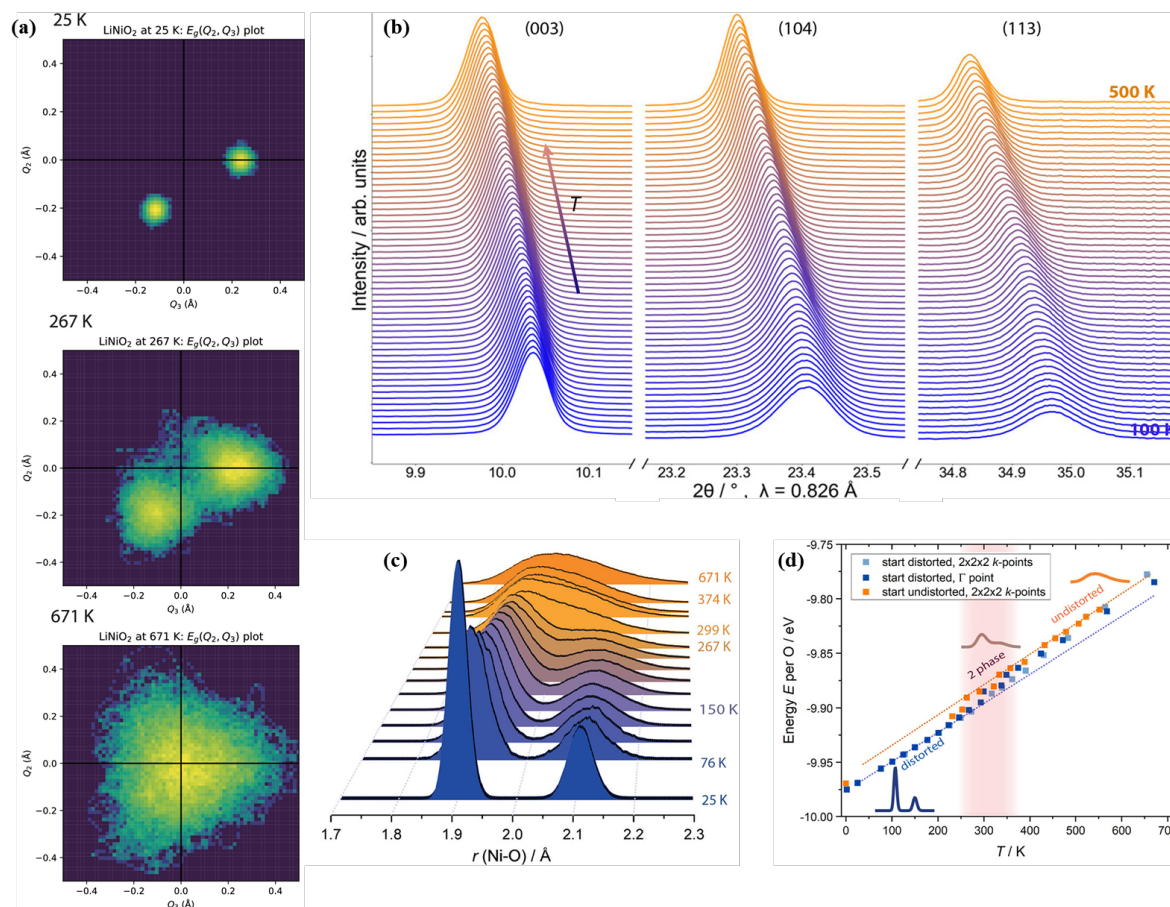


Figure 5. (a) Heat map of a van Vleck analysis of the octahedral distortion modes of AIMD trajectories (0° , 120° , and 240° correspond to three possible orientations of the long axes of quadratically elongated octahedra in LiNiO₂, and brighter colors denote a higher probability of a distortion). (b) In-situ XRD curves of LiNiO₂ at different temperatures. (c) Simulated Ni-O pair distribution function (PDF) at different temperatures. (d) Internal energy of

the distorted and undistorted phase as a function of temperature. Reprinted with permission from Ref. [51], copyright 2024, the American Chemical Society.

The influence of Jahn-Teller distortion on LiNiO_2 is not only reflected in the structural regulation of its phase transition pathway but also directly involved in the failure mechanism of the material [49,52,53]. For example, local non-cooperative Jahn-Teller distortion generates lattice inhomogeneity and intragranular cracks, promoting surface reconstruction and impedance growth. The pristine LiNiO_2 (which contains more Ni^{3+}) is O3-type (R-3m space group) and does not exhibit a monoclinic distortion. However, as shown in Figure 6, under the Jahn-Teller effect, the O3 phase will transform into the O'3 phase (C2/m space group) under the Li-deficient condition. During delithiation (Li_xNiO_2 , $x \approx 0.75\text{--}0.5$), Ni^{3+} cooperatively triggers a collective Jahn-Teller distortion in conjunction with Li/vacancy ordering, stabilizing highly monoclinic phases (M phases) such as $\text{Li}_{0.75}\text{NiO}_2$ and $\text{Li}_{0.4}\text{NiO}_2$. When x decreases to ≤ 0.4 , the Ni oxidation state approaches +4, eliminating the cooperative Jahn-Teller distortion and driving the system from the M phase to the H2 phase. The long-range Li-O-TM-O-Li ordering is concurrently disrupted due to the rearrangement of Li/vacancy and transition-metal ordering, rather than simply by a lack of Ni^{3+} . More importantly, the two-phase transitions mediated by the Jahn-Teller distortion, such as $\text{H1} \rightarrow \text{M}$ and $\text{M} \rightarrow \text{H2}$, create phase boundaries inside the particles. When the lattice parameter difference between the two phases is large, stress and strain are generated. The accumulation of such local stress promotes intragranular cracking of primary particles, which in turn leads to electrolyte infiltration, surface side reactions, and loss of electrical contact, ultimately accelerating capacity fading. Meanwhile, the high-voltage phase sequence involving Jahn-Teller distortion in LiNiO_2 , especially the $\text{H2} \rightarrow \text{H3}$ transition accompanied by a drastic collapse of the c axis, is also an important cause of structural failure at high voltages. In contrast, in Ni-rich NMC or NCA (such as NMC-811), the random distribution of dopants (Mn, Co, or Al) disrupts the ordering of Ni^{3+} , suppressing long-range Li/vacancy ordering and collective Jahn-Teller distortion [54–56]. As a result, the originally pronounced $\text{H1} \rightarrow \text{M} \rightarrow \text{H2}$ phase sequence in LiNiO_2 disappears and is replaced by solid-solution behavior. Through further component regulation, such as doping with high-valent metal ions like W and Mo, or rare earth metals, the structural stability of the cathode can be significantly enhanced, thereby improving its lithium storage performance [57,58]. Therefore, Jahn-Teller distortion not only dictates the phase transition pathway and structural evolution of LiNiO_2 but also serves as a key driving force for its mechanical degradation and electrochemical performance fading by inducing two-phase reactions and lattice mismatch.

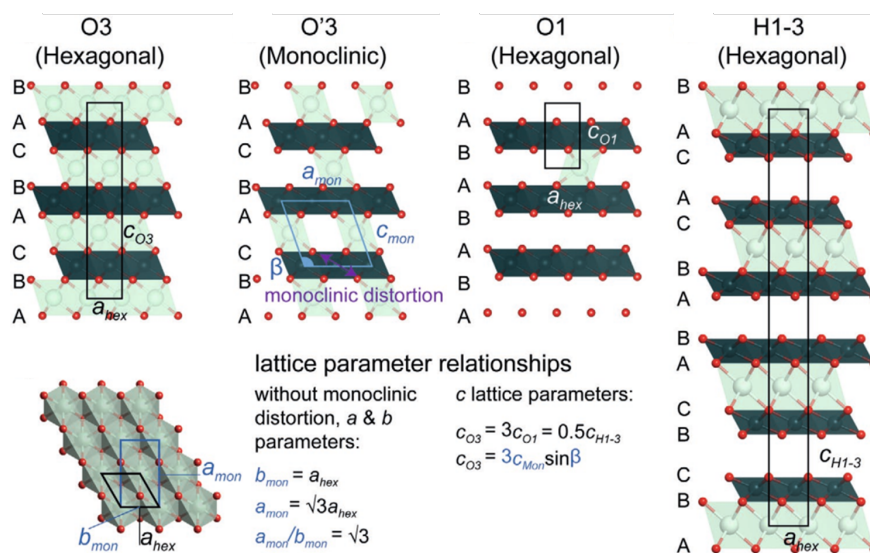


Figure 6. Phase transition of LiNiO_2 cathode during cycling. Reprinted with permission from [52], copyright 2020, Wiley-VCH GmbH.

Among layered cathodes, another one severely affected by Jahn-Teller distortion is Li-rich Mn-based layered oxides. These cathodes are considered promising cathodes for LIBs because of their high specific capacity (exceeding 280 mAh g^{-1}). However, extensive Mn^{3+} results in Jahn-Teller distortion and surface densification, leading to performance degradation [59]. In this cathode, Jahn-Teller distortion generates severe local lattice strain, inducing microcracks on the surface, which expose the material to electrolyte attack and accelerate electrolyte decomposition. Meanwhile, transition metal ion dissolution, particularly Mn^{2+} ions generated via Jahn-Teller distortion-induced disproportionation, becomes pronounced. These side reactions collectively lead to the

formation of a thick and inhomogeneous cathode electrolyte interphase, as well as an electrochemically inert rock-salt phase. Consequently, the degraded interface impedes Li^+ transport and increases the impedance at the interface, ultimately resulting in continuous capacity decay.

After suppressing the Jahn-Teller distortion, the lithium storage performance of the lithium-rich manganese cathode is typically significantly improved. As Song et al. [60] reported, the MgAl_2O_4 coating significantly suppresses the Jahn-Teller distortion in lithium-rich layered oxide cathodes through synergistic surface reinforcement and bulk doping (Figure 7a–c). Specifically, the surface MgAl_2O_4 spinel coating induces migration of Mg^{2+} into the bulk lattice during discharge, thereby reducing the electrostatic potential difference between the transition-metal layer and the alkali-metal layer. This effectively alleviates the asymmetric stretching of MnO_6 octahedra (the Mn-O bond-length ratio approaches 1), mitigating the structural distortion and Mn dissolution driven by the Jahn-Teller effect. Electrochemically, the modified material achieves a high-capacity retention of 99.2% after 500 cycles at 1C (compared to only 57.9% for the unmodified sample). At the same time, Mn dissolution decreases from 0.44 to $0.21 \mu\text{g mg}^{-1}$, and the degree of Mn-O bond distortion after cycling is significantly reduced. This mechanism allows the Jahn-Teller effect of Mn ions under high voltage to be mitigated and suppressed, thereby greatly enhancing the cycling stability of the material.

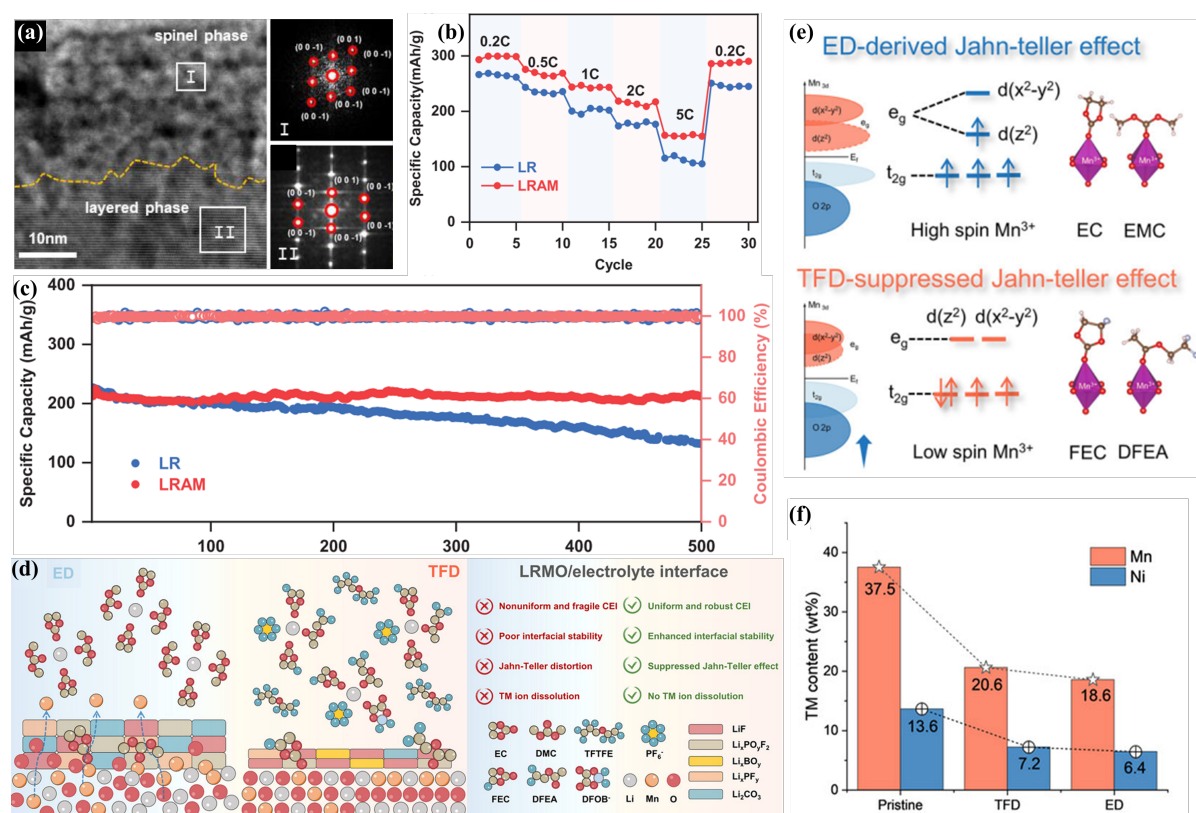


Figure 7. (a) HRTEM images of the Li-rich cathode with MgAl_2O_4 coating. (b) Rate capabilities of the coated and uncoated Li-rich cathodes. (c) Cycling performance of the coated and uncoated Li-rich cathodes. Reprinted with permission from Ref. [60], copyright 2025, Wiley-VCH GmbH. (d) Solvation structure of EC-based carbonate electrolytes (ED) and TFD-based (1.2 M LiPF_6 in 1,1,2,2-Tetrafluoroethyl 2,2,2-trifluoroethyl ether/Fluoroethylene carbonate/2,2-Difluoroethyl acetate (TFTFE/FEC/DFEA) (v/v/v = 1:1:2)) electrolyte. (e) The mechanism of the TFD-based electrolyte suppresses the Jahn-Teller effect. (f) Mn and Ni dissolution content in TFD-based electrolyte. Reprinted with permission from Ref. [61], copyright 2026, Wiley-VCH GmbH.

In this type of cathode, Jahn-Teller distortion can be directly suppressed by tailoring the electrolyte and the interface. Yang et al. [61] designed a fluorinated electrolyte (TFD-based) as shown in Figure 7d, in which the preferential decomposition of lithium difluoro(oxalato)borate (LiDFOB) constructs a thin, uniform, and mechanically robust CEI rich in LiF/LiBO_2 on the lithium-rich manganese-based oxide (LRMO) surface, effectively inhibiting interfacial side reactions. Meanwhile, as shown in Figure 7e,f, the fluorinated solvent molecules (FEC, DFEA) modulate the electronic structure of Mn through their strong electronegativity, converting high-spin Mn^{3+} into a Jahn-Teller inactive Mn^{4+} state, thereby eliminating Jahn-Teller distortion at its source; thus the Mn-ion dissolution is significantly suppressed. Benefiting from this interfacial/bulk dual regulation, the

LRMO||TFD||Li cell achieves a capacity retention of 97.6% after 400 cycles, exhibits smaller volume variation and a thinner surface phase transformation layer, and demonstrates excellent stability under high rates, lean electrolyte conditions, low temperature, and in pouch cells.

Similar detrimental effects have been revealed in Mn-based layered oxides for Na-ion and K-ion batteries. For example, in P2/O3-type Na-ion layered oxides, especially Mn-based compositions, Mn^{3+} -driven Jahn-Teller distortion induces a characteristic P2 to P'2 (O3 to O'3) phase transition at low discharge voltages [62,63]. This transition introduces intense anisotropic strain, layer gliding, transition metal dissolution, and particle fracture. In all cases, Jahn-Teller distortion modulates layer stability, phase transition, strain distribution, and transition-metal dissolution, thereby dominating cycling stability and rate capability.

In this type of cathode, strategies such as elemental doping/substitution, surface reconstruction (coating), multiphase structure design, and vacancy regulation (oxygen vacancies, TM vacancies, Na^+ /vacancy ordering) can effectively stabilize the crystal structure and enhance its cycle life and rate capability by reducing the concentration of Jahn-Teller active ions, strengthening TM-O bonds, introducing phase boundaries to suppress cooperative distortion, or disrupting local symmetry. For example, by introducing non-Jahn-Teller active ions (such as Al, Zn, Li, Ti, Sn, B) into the TM sites, the concentration of Jahn-Teller active ions such as Mn^{3+} , Fe^{4+} , or Ni^{3+} is diluted, while the covalency of TM-O bonds is enhanced. This eliminates the degeneracy of e_g orbitals and suppresses the cooperative distortion of TMO_6 octahedra. Taking Al-doped P2-type $\text{Na}_{0.67}\text{MnO}_2$ as an example (Figure 8), the undoped $\text{Na}_{0.67}\text{MnO}_2$ undergoes a severe P2–P'2 phase transition in the low-voltage range, accompanied by significant volume expansion and poor cycling stability [64]. In contrast, after Al doping, the Mn^{3+} concentration is reduced, the P2–P'2 phase transition is substantially suppressed, the volume change is only 3%, and the capacity retention after 100 cycles increases from about 70% to 86%.

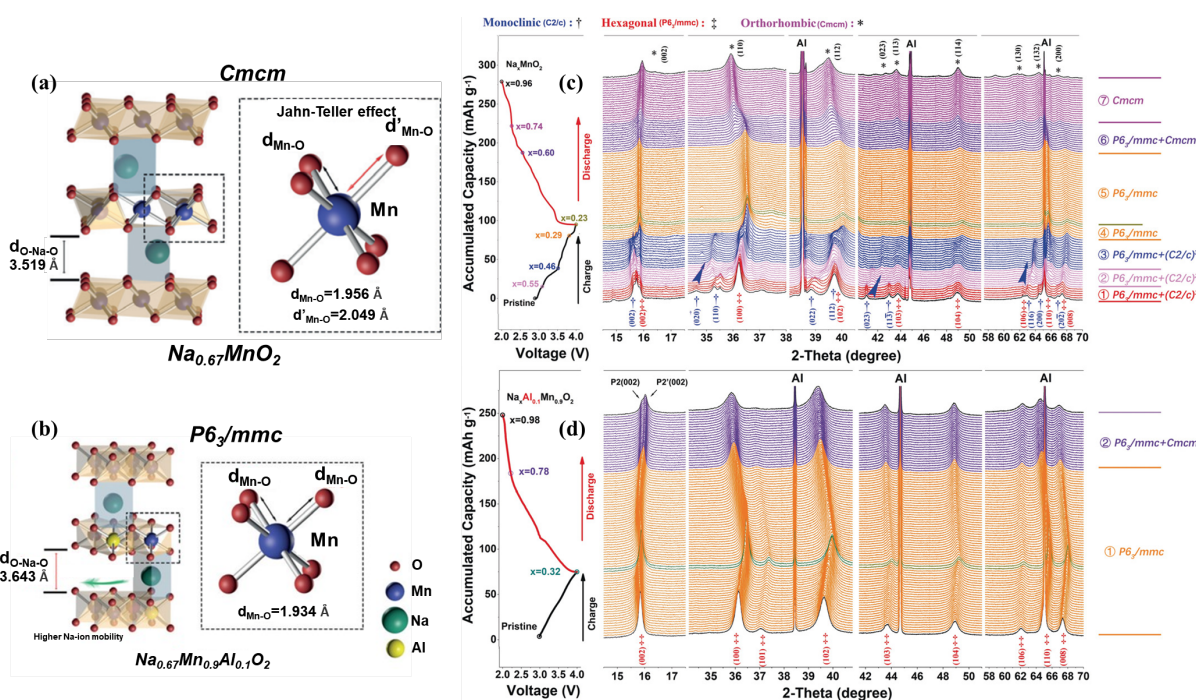


Figure 8. (a,b) Structure schematic of the $\text{Na}_{0.67}\text{MnO}_2$ and $\text{Na}_{0.67}\text{Al}_{0.1}\text{Mn}_{0.9}\text{O}_2$ cathodes. (c,d) In-situ XRD curves of the $\text{Na}_{0.67}\text{MnO}_2$ and $\text{Na}_{0.67}\text{Al}_{0.1}\text{Mn}_{0.9}\text{O}_2$ cathodes. Reprinted with permission from Ref. [64], copyright 2019, Wiley-VCH Verlag GmbH & Co. Weinheim.

Multiphase structure design (such as P2/O3, P2/P3, P2/tunnel phase) constructs intimately connected heterointerfaces, exploiting grain boundary pinning effects and lattice mismatch at the phase boundaries to physically interrupt the propagation path of long-range cooperative Jahn-Teller distortion, while integrating the advantages of different phases (high sodium content of the O3 phase and fast ionic transport of the P2 phase). Taking P2/O3 biphasic $\text{Na}_{0.8}\text{Ni}_{0.23}\text{Fe}_{0.34}\text{Mn}_{0.43}\text{O}_2$ as an example, single-phase P2 or O3 materials exhibit severe irreversible P2–P'2 or O3–P3 phase transitions due to the Jahn-Teller effect of Mn^{3+} , leading to rapid capacity decay [65]. In contrast, in the biphasic material, phase boundaries block the cooperative distortion, allowing only reversible phase transitions of $\text{P2} + \text{O3} \rightarrow \text{P2} + \text{P3} + \text{O3} \rightarrow \text{P2} + \text{P3}$ within 2–4 V, with minimal volume fluctuation. The discharge capacity reaches as high as 146.4 mAh g^{-1} , and after 2000 cycles at a high rate of 10 C, the capacity retention remains 71.7%, far superior to single-phase samples.

Vacancy engineering (including TM vacancies, oxygen vacancies, and Na⁺/vacancy ordering) weakens Jahn-Teller activity by disrupting the local coordination symmetry around Jahn-Teller active ions, forcing them to transition from a high-spin to a low-spin state. Moreover, TM vacancies can trigger reversible anionic redox processes, thereby enhancing capacity. Taking P2-type Na_{0.615}Mg_{0.154}Ti_{0.154}Mn_{0.615}□_{0.077}O₂ (□ indicates TM vacancies) as an example, the vacancy-free Na_{2/3}Mg_{1/3}Mn_{2/3}O₂ exhibits a detrimental P2-O2 phase transition at the end of charging, whereas the TM-vacant sample completely avoids both P2-O2 and P2-P'2 phase transitions over a wide voltage range of 1.5–4.5 V, showing minimal volume change and a significantly increased low-spin/high-spin Mn³⁺ ratio [66]. After 100 cycles, the capacity retention reaches up to 95.2% (the vacancy-free control sample retains only ~70% and undergoes severe phase transitions).

In Na-layered cathodes, there exists a synergistic effect between distortions caused by Jahn-Teller-active centers, which is known as the cooperative Jahn-Teller effect (CJTE). Li et al. [9] directly visualized the CJTE (Figure 9) and its coupling to alkali-ion ordering at the atomic scale, providing new insights into structural evolution in layered transition-metal oxide cathodes. Before this work, superstructure formation in Na_xTMO₂ was universally attributed to electrostatic interactions between Na⁺ ions and vacancies, while the role of electronic-lattice coupling remained speculative due to the lack of direct experimental evidence. Using a combination of aberration-corrected scanning transmission electron microscopy (STEM) with annular bright-field (ABF) imaging, which can resolve light alkali atoms, together with electron energy-loss spectroscopy (EELS) for quantitative valence mapping, synchrotron X-ray diffraction (XRD) refinement, neutron diffraction, and density functional theory (DFT) calculations, the authors unambiguously demonstrated that the thermodynamically stable Na_{5/8}MnO₂ phase formed during electrochemical desodiation is governed entirely by the CJTE, not by electrostatic forces. The study directly imaged periodic charge-ordered stripes consisting of Jahn-Teller-active Mn³⁺, Jahn-Teller-inactive Mn⁴⁺, and mixed Mn³⁺/Mn⁴⁺ within the transition-metal layers, and the Mn³⁺-induced octahedral elongation produces a measurable rippling of the Mn planes. Most strikingly, a subset of Na ions was observed to shift by a large shift of about 1.4 Å from normal edge-sharing octahedral sites into highly distorted face-sharing octahedral sites, a rearrangement exclusively driven by the CJTE, as confirmed by control DFT calculations showing that replacing Mn with non-Jahn-Teller-active Cr eliminates this displacement. This anomalous Na migration originates from long-range 180° Na-O-Mn³⁺-O-Na s-p-d orbital hybridization, which creates repulsive interactions in unstable vacancy-O-Mn³⁺-O-Na configurations and drives Na redistribution to lower the system energy. Furthermore, this coupled charge-orbital-alkali ordering generates a unique “magnetic stripe sandwich” structure below 60 K, in which antiferromagnetic Mn³⁺ stripes are interleaved with ferrimagnetic Mn⁴⁺ and mixed-valence stripes. Beyond its fundamental significance in condensed matter physics, this work established that the CJTE exerts significant influence over alkali-ion site occupancy, diffusion pathways, and structural stability in Mn-based layered oxides, providing an atomic-level mechanistic foundation for the rational design of high-performance sodium-ion battery cathodes and inspiring a generation of in situ and operando studies of Jahn-Teller dynamics during electrochemical cycling.

As a typical Mn-based layered cathode for potassium-ion batteries, P3-type K_xMnO₂ (KMO) exhibits considerable reversible capacity but suffers from unsatisfactory cycling stability and rapid capacity fading, especially when charged to high voltages [67–69]. Kim et al. [70] report, using in situ XRD and first principles calculations, that this work reveals that K_{0.5}MnO₂ undergoes reversible P3 to O3 phase transitions (Figure 10) during electrochemical cycling, while excessive deintercalation of potassium ions causes structural amorphization and stacking faults. These structural degradations are closely associated with the Jahn-Teller effect originating from high-spin Mn³⁺. The asymmetric electron occupation of Mn³⁺ gives rise to inherent octahedral distortion and anisotropic lattice strain, which reduce structural stability and aggravate irreversible structure evolution upon repeated K-K-ion extraction and insertion. The K_{0.5}MnO₂ cathode exhibits poor cycling stability in PF₆[−]-based electrolyte within the voltage window of 1.5–3.9 V, with an average Coulombic efficiency of only 95%. Based on the above analysis, this phenomenon may be related to interfacial side reactions triggered by the combination of Jahn-Teller distortion and electrolyte salt hydrolysis. Therefore, the poor cycling durability of K_{0.5}MnO₂ can be reasonably attributed to the Jahn-Teller distortion of Mn³⁺, which provides direct guidance for subsequent structural stabilization strategies.

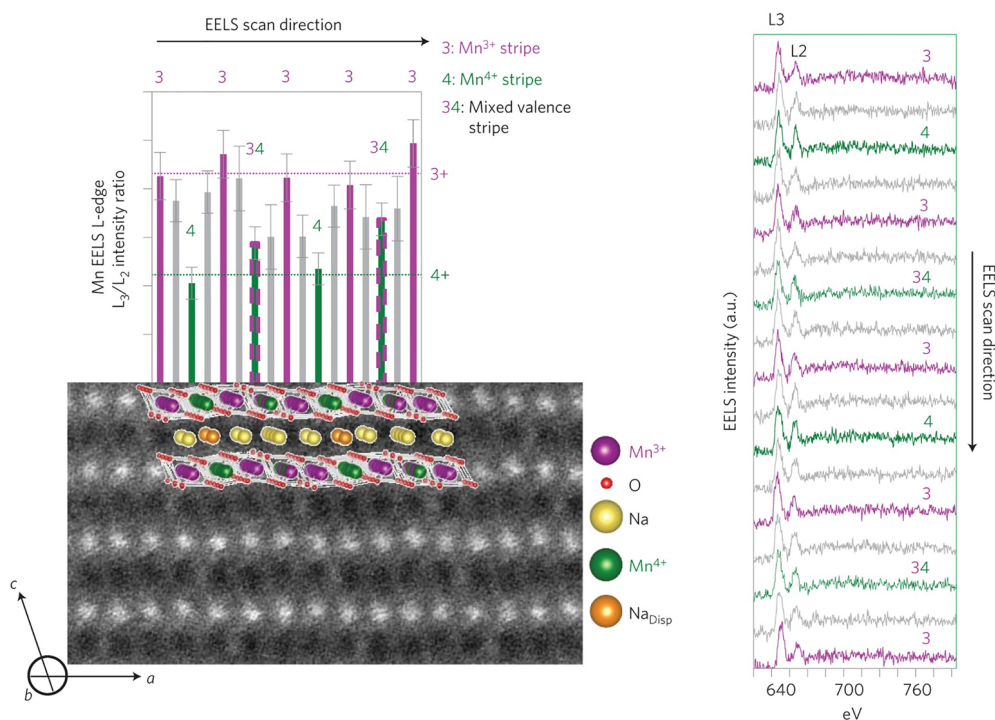


Figure 9. Atomic-resolution STEM image visualizes CJTE and STEM-EELS. Reprinted with permission from Ref. [9], copyright 2014, with permission from Macmillan Publishers Limited.

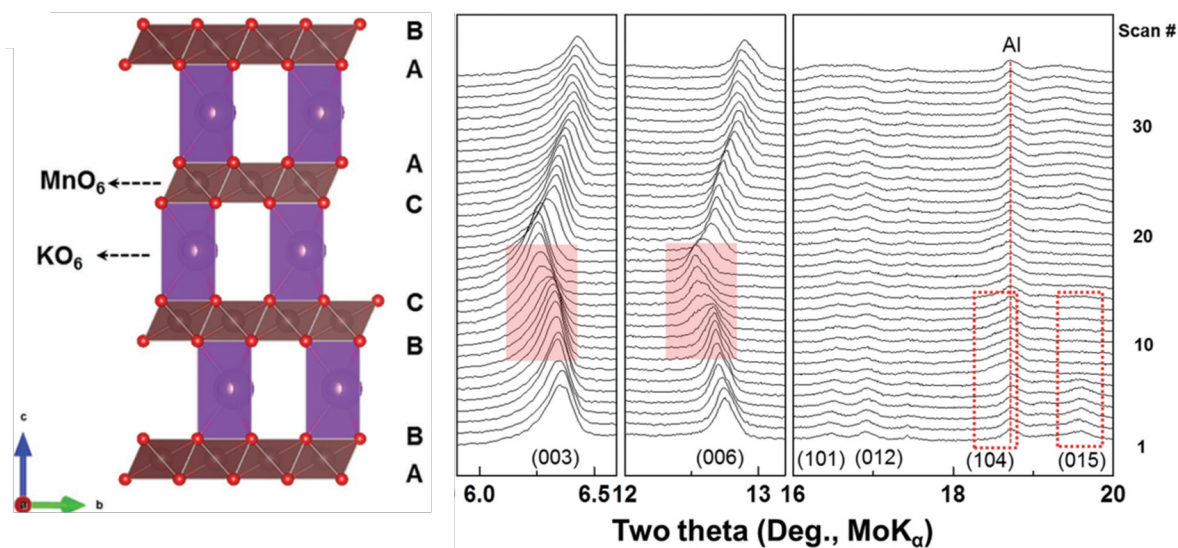


Figure 10. XRD and in situ XRD curves of the $K_{0.5}MnO_2$ cathode. Reprinted with permission from Ref. [70], copyright 2017, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

Nevertheless, the Jahn-Teller effect can be effectively regulated through rational doping strategies to achieve enhanced structural stability and electrochemical performance in Mn-based layered cathodes for potassium-ion batteries. Recent studies have demonstrated that both transition-metal-site substitution and alkali-metal-site engineering can suppress detrimental Jahn-Teller distortion and inhibit irreversible phase transitions. Luo et al. [71] proposed a distinct stabilization route by substituting Mg into the K layers of $K_{0.5}MnO_2$. The Mg ions located in the alkali-metal layers serve as structural pillars to suppress layer sliding and lattice collapse along the c-axis. This design effectively eliminates Jahn-Teller distortion by elevating the Mn valence state and completely blocks the P3 to O3 phase transition. The optimal $K_{0.5}Mg_{0.15}[Mn_{0.8}Mg_{0.05}]O_2$ cathode delivers significantly improved cycling stability with 48.2% capacity retention after 400 cycles at 1 C and enhanced rate capability up to 5 C. Synchrotron X-ray absorption spectroscopy demonstrates highly reversible Mn³⁺/Mn⁴⁺ redox chemistry, while theoretical calculations confirm lower K⁺ diffusion barriers and reduced activation energy. Together, these studies establish that controlled lattice distortion via either transition-metal doping or alkali-site pillar engineering can effectively mitigate Jahn-Teller distortion, suppress harmful phase transitions, and improve ion transport kinetics, providing

a reliable materials design platform for high-performance Mn-based layered cathodes in potassium-ion batteries. Zhou et al. [72] introduced low-valent Cu^{2+} into the Mn sites of P3-type $\text{K}_{0.5}\text{MnO}_2$ to adjust the average valence of Mn, reduce the content of Jahn-Teller-active Mn^{3+} , and construct elastically distorted $[\text{MnO}_6]$ octahedra. The optimized $\text{K}_{0.5}\text{Mn}_{0.9}\text{Cu}_{0.1}\text{O}_2$ material shows outstanding long-cycle stability, with 70% capacity retention over 500 cycles at 1 A g^{-1} . In situ XRD and pair distribution function analysis confirm that Cu doping prevents the harmful P3 to O3 phase transition and homogenizes lattice strain during cycling. First-principles calculations further reveal that Cu-3d orbitals fill the electronic gap, reduce K^+ migration barriers, and strengthen Mn-O covalency.

A multiple-ion doping strategy can more effectively suppress the Jahn-Teller effect. As Tang et al. [73] reported, the incorporation of Li and Fe doping preserves the symmetry of the MnO_6 octahedra with Mn-O bond lengths of $1.94 \text{ \AA}$ in all directions. Compared with bare KMO, the Jahn-Teller-suppressed $\text{K}_{0.4}\text{Li}_{0.1}\text{Fe}_{0.1}\text{Mn}_{0.8}\text{O}_2$ (KLFMO) cathode demonstrates significantly improved performance. It delivers a reversible capacity of 110.2 mAh g^{-1} at 0.2 C , higher than the 101.8 mAh g^{-1} of KMO. After 150 cycles at 0.6C , it retains 84.2% of its capacity, far exceeding the 41.0% of KMO. In terms of rate capability, KLFMO still outputs 57.6 mAh g^{-1} at 10C , whereas KMO only delivers 37.3 mAh g^{-1} . During cycling, it does not undergo the P3 to O3 phase transition but only a reversible P3–P'3 transformation, with small and homogeneous lattice strain. After cycling, the particles remain intact without cracks, and both surface reconstruction and manganese dissolution are effectively suppressed.

Overall, the Jahn-Teller effect in layered oxides depends strongly on the active ion, alkali species, state of charge, and temperature. Its manifestations range from cooperative phase transitions in LiNiO_2 and P2-type Na cathodes to local non-cooperative strain in Li-rich Mn-based oxides. Accordingly, the suppression strategies developed for layered systems are diverse. Cationic doping dilutes Jahn-Teller-active centers and suppresses collective ordering. Multiphase structures introduce phase boundaries that physically block cooperative distortion propagation. Vacancy engineering disrupts local coordination symmetry. Electrolyte and interface engineering reduces surface reactivity. Each approach has distinct applicability. Cationic doping is straightforward but may dilute redox capacity. Multiphase and vacancy engineering are effective but require precise compositional control. Electrolyte engineering is non-destructive to the bulk but demands stable formulations. These strategies also share a common limitation: most of them are demonstrated in specific layered systems and their transferability to other layered cathodes requires further validation. Moreover, controlled distortion in layered oxides can assist ion diffusion or relieve lattice strain, a beneficial aspect that will be discussed in Section 3.5. The diversity of layered oxide chemistry thus necessitates a case-specific regulation strategy, rather than a universal suppression approach.

3.3. On Prussian Blue Analogue Cathodes

Prussian blue analogues (PBAs) possess a robust open framework, but their electrochemical performance is still limited by Jahn-Teller distortion when Mn-based redox centers are employed [74–76]. During charge-discharge cycling, Mn^{2+} is oxidized to Mn^{3+} , which adopts a high-spin d^4 configuration and induces strong Jahn-Teller distortion within MnN_6 octahedra. This local distortion tilts Mn-N-C bonds, deforms the cyanide-bridged framework, and disrupts the uniformity of ion-transport channels, resulting in a multistep phase transition from monoclinic to cubic to tetragonal, accompanied by lattice volume expansion. Unlike in oxides, Jahn-Teller distortion in PBAs rarely triggers long-range phase transitions but increases voltage hysteresis and reduces ionic conductivity. Meanwhile, distorted metal-ligand bonds weaken lattice cohesion and promote transition-metal dissolution. Consequently, suppressing Jahn-Teller distortion in PBAs effectively improves structural integrity, lowers polarization, and extends cyclic life.

Suppressing Jahn-Teller distortion is crucial for enhancing the structural stability of PBA. There are currently many examples where the performance of materials has been significantly improved after suppressing this distortion. For example, Kang et al. [77] synthesized a surface Cu concentration-gradient-doped Cu@MnPBA via a two-step coprecipitation method. The $\sim 200 \text{ nm}$ Cu gradient layer effectively suppresses Jahn-Teller distortion (DFT calculations show shortened axial Mn-N bonds, reduced distortion ratio, and enhanced Mn-N bonding), while the outermost $\sim 10 \text{ nm}$ Cu-rich amorphous layer promotes Na^+ surface transport and inhibits interfacial side reactions (Figure 11a,b). Finite element simulations indicate that the maximum von Mises stress of Cu@MnPBA is reduced from 257 MPa (pristine MnPBA) to 58 MPa , and the damage value decreases from 1.65% to 0.8%. Electrochemically, the Cu@MnPBA half-cell achieves a capacity retention of 78% after 500 cycles at 100 mA g^{-1} (compared to only 20% for MnPBA) and delivers 70.4 mAh g^{-1} at a high rate of 20 C (vs. 47.9 mAh g^{-1} for MnPBA). Furthermore, a pouch cell assembled with hard carbon retains 75% of its capacity over 3000 cycles.

Choi et al. [78] adopted a high-entropy strategy to introduce multiple 3d transition metals (Co, Ni, Cu, Zn) at the Mn sites (Figure 11c), forming an entropically stabilized coordination environment. In this design, Co participates in charge compensation to reduce the proportion of the $\text{Mn}^{3+}/\text{Mn}^{4+}$ redox couple, thereby directly mitigating Jahn-

Teller distortion; Ni stiffens the lattice and promotes K^+ diffusion; Cu and Zn form strong Cu-N/Zn-N bonds to locally stabilize the structure and suppress Mn dissolution. This high-entropy PBA (HEPBA) minimizes the concentration of $Fe(CN)_6^{4-}$ vacancies, induces charge disorder, and converts the original two-phase reaction into a solid-solution behavior, avoiding the detrimental tetragonal phase formation. Electrochemically, the HEPBA half-cell delivers a discharge capacity of 142.4 mAh g^{-1} at 0.2 C and retains 88.2% of its capacity after 5000 cycles. The pouch-type full cell assembled with a Perylene-3,4,9,10-tetracarboxylic diimide (PTCDI) organic anode achieves an energy density of 122.1 Wh kg^{-1} and maintains 84.3% capacity at an ultra-high rate of 20 C , outperforming all reported aqueous potassium-ion battery systems. This work demonstrates that high-entropy engineering can simultaneously address four major challenges of PBAs: Jahn-Teller distortion, lattice water, vacancies, and phase transitions, offering a design paradigm for low-cost, long-life, and high-safety aqueous batteries.

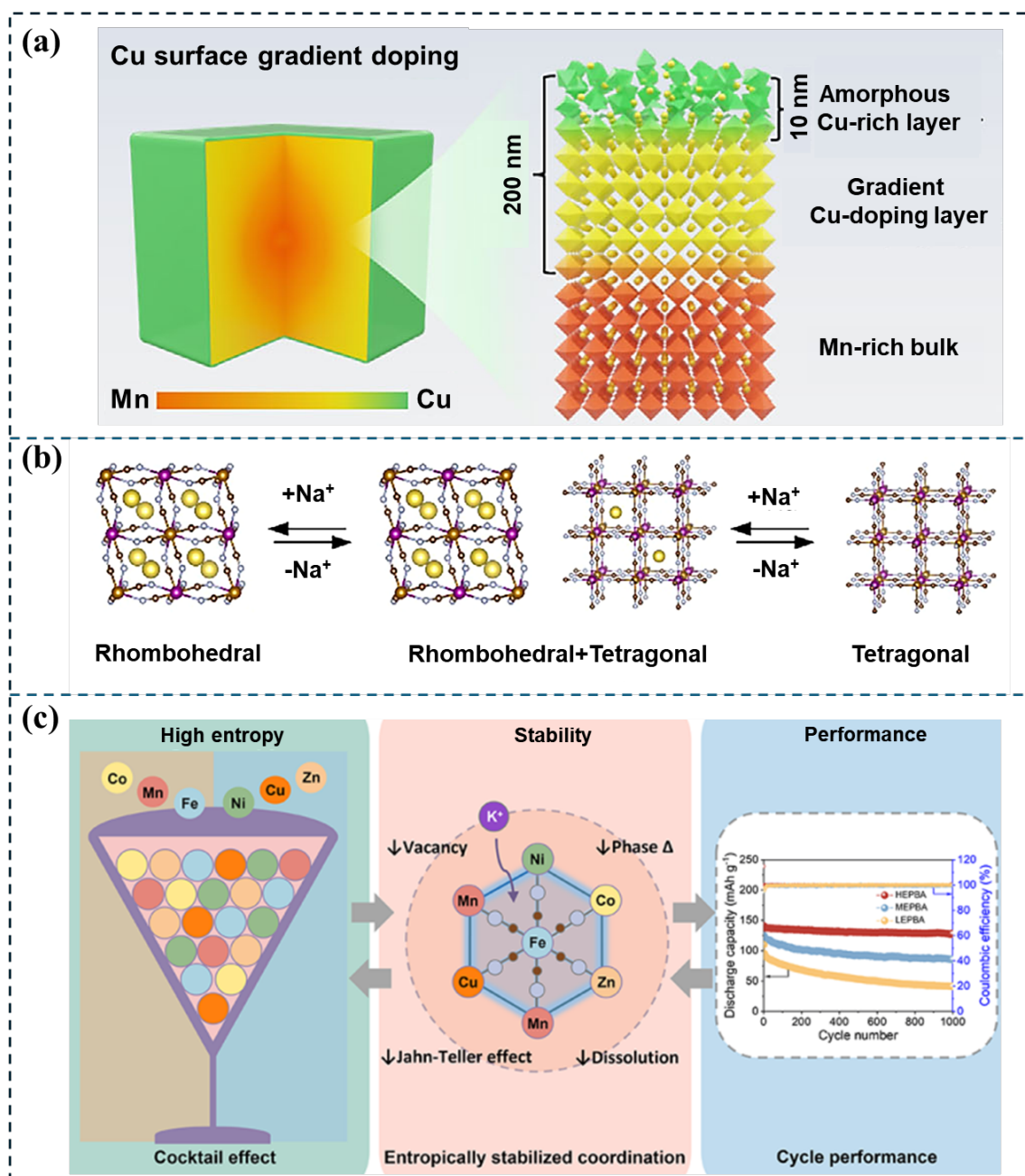


Figure 11. (a) Schematic of Cu doped PBA cathode. (b) structural evolution of Cu doped PBA cathode. Reprinted with permission from Ref. [77], copyright 2026, Wiley-VCH GmbH. (c) Schematic of the high-entropy PBA cathode. Reprinted with permission from Ref. [78], copyright 2026, American Chemical Society.

According to the above content, the Jahn-Teller distortion in Prussian blue analogues arises from MnN_6 octahedra and causes framework tilting, multistep phase transitions, and reduced ionic conductivity. Unlike in oxides, long-range cooperative distortion is rare, but voltage hysteresis and transition metal dissolution remain

serious. Suppressing this distortion through gradient doping or high entropy design has proven effective. The open framework structure of PBAs offers unique opportunities to localize Jahn-Teller effects, making them more amenable to composition and structural engineering than densely packed oxides. Future efforts should focus on eliminating local distortion without compromising the inherent fast ion diffusion pathways.

3.4. On Manganese Oxide Cathodes

In aqueous Zn-ion batteries, α - MnO_2 -based cathodes rely on intercalation-conversion reactions, where Jahn-Teller distortion of Mn^{3+} acts as the dominant degradation mechanism [79,80]. As Mn^{4+} is reduced to Mn^{3+} during discharge, the high-spin Mn^{3+} centers undergo strong Jahn-Teller distortion, elongating axial Mn-O bonds and weakening their bonding strength. These vulnerable sites readily react with protons and water molecules, triggering the disproportionation reaction $2\text{Mn}^{3+} \rightarrow \text{Mn}^{2+} + \text{Mn}^{4+}$. Soluble Mn^{2+} is continuously released into the electrolyte, causing rapid capacity decay and structural amorphization. Repeated lattice distortion also accumulates mechanical strain and microcracks. In aqueous environments, Jahn-Teller distortion is tightly coupled with dissolution and interfacial chemical reactions rather than global phase transitions, making it the decisive factor limiting long-term stability.

To address the Jahn-Teller distortion in this cathode that leads to structural collapse of α - MnO_2 , Mn^{2+} dissolution, and rapid capacity decay, doping offers an effective solution. Li et al. [79] constructed a breathable expanded tunnel structure through Ca doping, which effectively suppressed the distortion, reduced Mn dissolution by an order of magnitude, and halved the polarization. After suppressing Jahn-Teller distortion, the capacity after 200 cycles increased from 53 to 221 mAh g^{-1} . Although Cr doping also expanded the tunnel, its rigid, non-breathable structure could not accommodate the dynamic Jahn-Teller distortion during cycling, leading to a similarly rapid capacity decay. Therefore, for this material, suppressing Jahn-Teller distortion and imparting flexibility to the tunnel structure are key to improving stability. Lu et al. [81] introduced highly electronegative F^- ions into the MnO_2 lattice. This study successfully implements an axial compressive strain engineering strategy in which F^- doping not only shortens the axial Mn-O bond to mechanically suppress the Jahn-Teller distortion but also regulates the electronic configuration of Mn from the unstable high-spin state to the more stable state, thereby greatly reducing Mn^{3+} dissolution and structural distortion. Compared with pristine MnO_2 , the optimized F- MnO_2 cathode increases the reversible capacity from 240.3 mAh g^{-1} to 313.6 mAh g^{-1} at 0.1 A g^{-1} , raises the electrical conductivity by about 70%, and maintains a high-capacity retention of 81% after 1000 cycles at 2 A g^{-1} , while the Zn^{2+} diffusion coefficient is improved by more than one order of magnitude and the undoped material suffers from severe degradation. These results demonstrate that the axial compressive strain strategy effectively suppresses the Jahn-Teller effect and significantly enhances the cycling stability and rate performance of aqueous zinc-ion batteries.

Unlike nonaqueous systems, the aqueous environment tightly couples distortion with dissolution and interfacial reactions. Strategies such as Ca doping to create flexible tunnels or F doping to impose axial compressive strain have successfully suppressed distortion and improved stability. Managing Jahn-Teller activity in aqueous batteries requires not only electronic modification but also mechanical compatibility, as dynamic lattice changes must be accommodated without fracturing the tunnel structure. This dual requirement distinguishes aqueous MnO_2 cathodes from other oxide cathodes with Jahn-Teller distortion.

3.5. Beneficial Jahn-Teller Effects

Many reports have shown another view of the Jahn-Teller effect in layered cathodes. The Jahn-Teller effect is not exclusively detrimental, but rather a phenomenon that only causes structural degradation and electrochemical performance decay in layered oxides. On the contrary, it exhibits multiple beneficial functions that can be rationally harnessed through reasonable compositional design to significantly improve the electrochemical performance of the cathodes.

High-spin Fe^{4+} acts as a unique Jahn-Teller active species in layered sodium oxide cathodes. As Li et al. [21] reported, unlike Mn^{3+} and Ni^{3+} , Fe becomes Jahn-Teller active only when oxidized to Fe^{4+} at high states of charge. The dynamic buckling of Fe^{4+}O_6 octahedra lowers the Na^+ diffusion barrier by enlarging the bottleneck for Na migration (Figure 12a,b). As a result, Fe-containing layered oxides maintain high Na mobility even at extremely low Na contents, enabling unusually high reversible capacity. However, excessive Fe also triggers Fe migration into Na layers via cooperative Jahn-Teller effects. When iron atoms agglomerate, the distortions of adjacent Jahn-Teller active iron ions in the transition metal layer accommodate the shortening of Fe-O bonds in the tetrahedra associated with the migrating iron. This shortening blocks diffusion channels, leading to capacity decay. The optimal Fe content is found to be approximately 30%, where the beneficial Jahn-Teller-assisted diffusion is

maximized while harmful Fe migration is suppressed. This study is the first to demonstrate that the Jahn-Teller effect can be exploited to improve diffusion kinetics rather than only being suppressed.

The Jahn-Teller effect in P2-type Fe/Mn-based layered oxide cathodes for sodium-ion batteries can play a role in mitigating and dispersing the lattice strain. As Gao et al. [17] reported, the P2 to P'2 phase transition driven by Mn^{3+} -induced Jahn-Teller distortion was previously considered harmful, and many studies aimed to suppress it. In sharp contrast, their work demonstrates that properly activated Jahn-Teller distortion can effectively mitigate and disperse lattice strain, thereby releasing internal stress and improving cycling stability. By constructing Al/Ti co-doped P2-type $\text{Na}_{0.67}\text{Fe}_{0.175}\text{Al}_{0.025}\text{Mn}_{0.7}\text{Ti}_{0.1}\text{O}_2$ (NFAMT), the researchers intentionally retained a moderately high content of Mn^{3+} (52.8%), which promotes pronounced Jahn-Teller distortion and the formation of a P'2 phase during discharge, whereas cathode without Al and Ti doped exhibits more less P'2 phase (Figure 12c–h). In situ XRD and geometric phase analysis (GPA) directly reveal that the Jahn-Teller distortion redistributes and homogenizes lattice strain instead of causing destructive stress concentration. Such strain dispersion suppresses intragranular cracking, preserves structural integrity, and maintains rapid Na^+ transport. Benefiting from this mechanism, the NFAMT cathode exhibits outstanding cycling stability: it delivers an initial discharge capacity of 94.4 mAh g^{-1} at 1 C and retains 93.2% after 300 cycles, significantly outperforming the undoped sample (57.6%). This work shows that the Jahn-Teller distortion by showing that it can serve as a strain-release mechanism rather than only a source of structural degradation. It establishes a new design concept: rationally activating rather than eliminating Jahn-Teller distortion can stabilize layered oxides under long-term cycling.

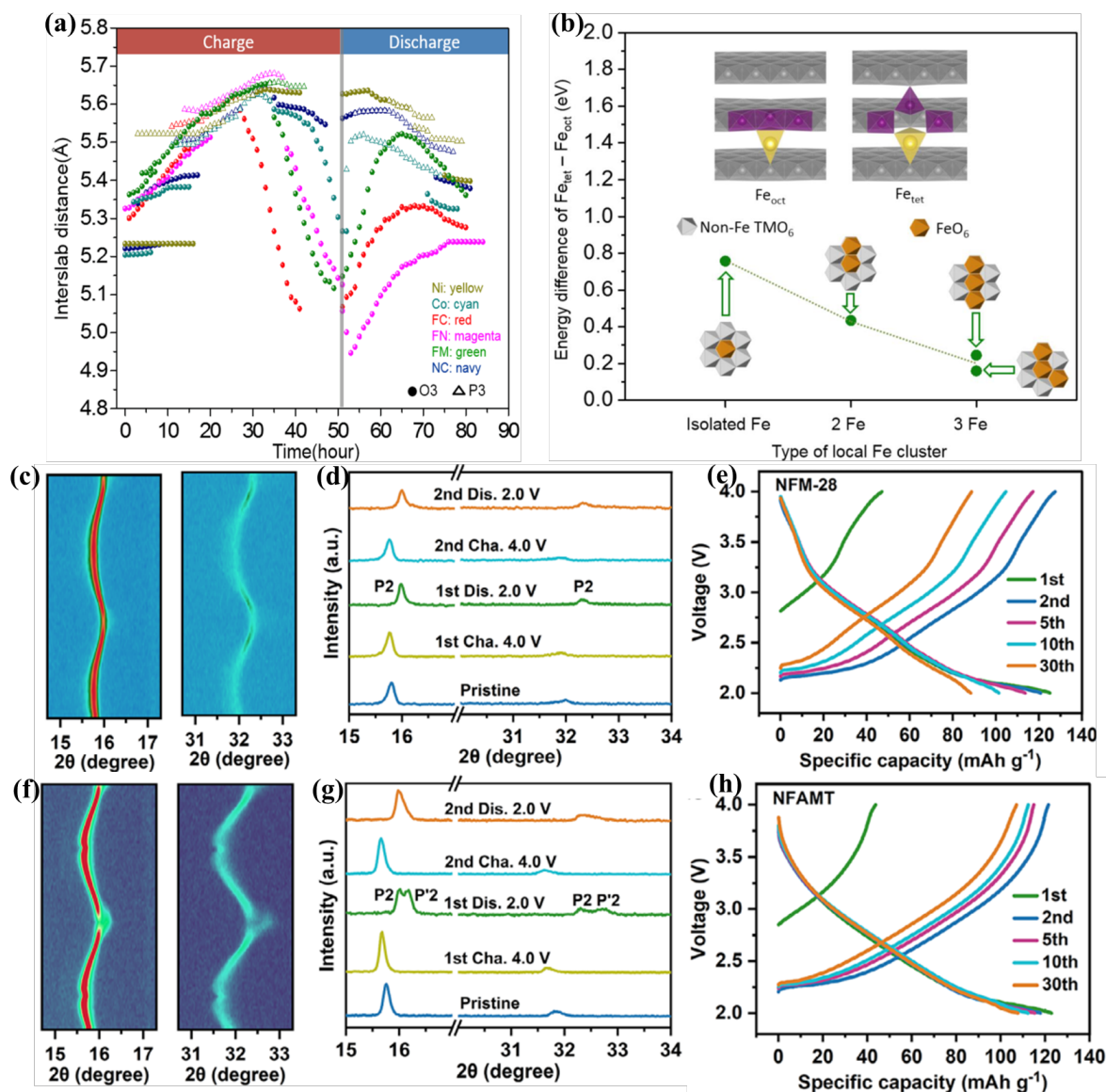


Figure 12. (a) Interplanar distance of layered NaTMO₂ compounds with different binary and single transition metal combinations upon charge and discharge. (b) Calculated energy penalty for Fe migration from an octahedral site in the TM layer to a tetrahedral site in the Na layer of NaTMO₂. Reprinted with permission from [21], copyright 2016,

American Chemical Society. **(c,d)** In situ XRD patterns and **(e)** cycling performance of $\text{Na}_{0.67}\text{Fe}_{0.2}\text{Mn}_{0.8}\text{O}_2$ (NFM) cathode. **(f,g)** In situ XRD patterns and **(h)** cycling performance of NFAMT cathode. Reprinted with permission from Ref. [17], copyright 2026, Wiley-VCH GmbH.

The Jahn-Teller distortion effect of Mn^{3+} and its interaction with electrolyte components can be exploited to induce the formation of a dual interfacial layer. Lei et al. [82] developed a high-concentration electrolyte that significantly enhances the electrochemical performance of KMO, specifically composed of diethylene glycol dimethyl ether solvent (G2) and 6 M KFSI salt, and synthesized a high-K-content P2-type $\text{K}_{0.67}\text{MnO}_2$ (P2-KMO) cathode via a sol-gel method. STEM observations revealed that after cycling in this electrolyte, a dual interfacial layer consisting of a spinel phase and a KF-rich CEI exists at the grain boundaries of the material (Figure 13). This interfacial layer can only be realized when using the high-concentration KFSI-G2 electrolyte and charging to voltages above 3.7 V. Because the uneven occupation of the orbitals of Mn^{3+} triggers the Jahn-Teller distortion, Mn ions can migrate into the K^+ vacancies in the KMO cathode during charging. Mn^{2+} tends to occupy the 8a tetrahedral sites, coordinating with four O^{2-} to form MnO_4 tetrahedra, whereas Mn^{3+} tends to enter the 16d octahedral sites, coordinating with six O^{2-} to form MnO_6 octahedra. In the high-concentration KFSI-G2 electrolyte, the aggregated FSI^- anions exhibit strong electronegativity, and together with the adsorption of G2 solvent molecules, they induce the reconstruction of the oxygen stacking arrangement into an ABCABC-type stacking sequence. On this basis, the MnO_4 tetrahedra and MnO_6 octahedra connect through edge-sharing or corner-sharing to reconstruct into a spinel intermediate layer. Meanwhile, electrolyte decomposition products form a KF-rich interfacial layer on the cathode surface, coating the outer side of the spinel intermediate layer, ultimately resulting in a dual interfacial layer structure composed of a spinel intermediate layer and a KF-rich CEI. In this electrolyte system, the P2-KMO cathode delivers an initial capacity of approximately 90 mAh g^{-1} at a current density of 45.5 mA g^{-1} , exhibits a capacity retention of 90.5% after 300 cycles, and a Coulombic efficiency close to 100%. These results indicate that, after constructing the dual interfacial layer, the cathode exhibits good structural reversibility, the electrolyte chemistry remains stable, and interfacial side reactions are minimal.

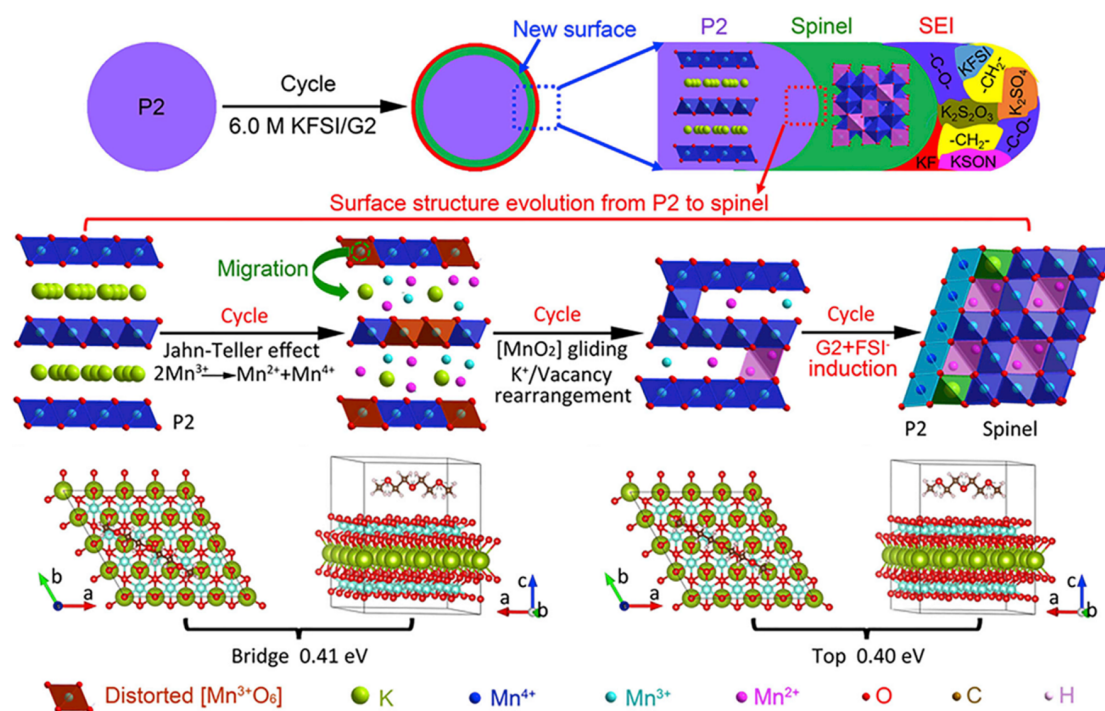


Figure 13. Schematic of the dual interface forming mechanism. Reprinted with permission from Ref. [82], with permission from, copyright 2019, Elsevier Inc.

The key distinction between harmful and helpful regimes lies in the connectivity of Jahn-Teller-active centers. Isolated or diluted centers tend to produce local distortions that may be accommodated elastically. In contrast, concentrated centers that enforce cooperative ordering may lead to undesirable phase transitions. Keeping the density of active ions below the threshold for cooperative distortion preserves enough local activity to assist ion diffusion or release the local stress. This view shifts the focus from eliminating Jahn-Teller ions to controlling

their microenvironment. Future work should develop synthesis methods that achieve atomic-scale regulation of Jahn-Teller active centers, as well as operando techniques to monitor the cooperativity mechanism during cycling.

4. Summary of Strategies on Regulating the Jahn-Teller Effect in Cathodes

Given the critical role of the Jahn-Teller effect in structural instability, transition-metal dissolution, and electrochemical degradation, extensive efforts have been devoted to suppressing harmful distortions or harnessing favorable ones. Currently, the regulation mechanisms of Jahn-Teller distortion are mostly deduced from phenomenological observations, and a systematic summary is still lacking. Representative regulation strategies are summarized in this section, including cationic doping, anionic substitution, structural and interfacial engineering, electrolyte optimization, and strain modulation. All of the strategies are summarized in Table 2.

4.1. Cationic Doping

Cationic doping is the most widely used and effective strategy to mitigate Jahn-Teller distortion by reducing the concentration of Jahn-Teller-active centers, increasing average transition-metal oxidation states, and stabilizing lattice frameworks. In spinel LiMn_2O_4 and layered Mn-based oxides, doping with electrochemically inactive cations such as Al^{3+} , Mg^{2+} , Zn^{2+} , Ti^{4+} , and Zr^{4+} effectively dilutes Mn^{3+} content and enhances structural robustness. Stronger metal-oxygen bonds formed by doped ions suppress octahedral distortion and inhibit detrimental phase transitions. For instance, Al doping in P2-type $\text{Na}_{0.67}\text{Fe}_{0.5}\text{Mn}_{0.5}\text{O}_2$ eliminates the P2 to P'2 phase transition by reducing the fraction of Mn^{3+} , thereby enhancing cycling stability remarkably [18,83]. Similarly, Al/Co/Mo substitution may regulate electronic structures and lower lattice mismatch, mitigating local non-cooperative Jahn-Teller distortion in Ni-rich layered oxides [58]. High-entropy doping strategy further disrupts cooperative Jahn-Teller behavior by introducing structural disorder and frustration, preventing long-range lattice distortion [84–86].

Table 2. Summary of regulation strategies for the Jahn-Teller (JT) effect.

Strategy	Approach	Mechanism	Cathode Systems	Limitations
Cationic doping	Substitution of JT-active ions (Mn^{3+} , Ni^{3+}) with electrochemically inactive cations (Al^{3+} , Mg^{2+} , Zn^{2+})	Dilutes concentration of JT-active centers; increases average TM valence state; strengthens M-O bonds; suppresses cooperative distortion.	Spinel LiMn_2O_4 , layered oxides (Li/Ni-rich, Na/K capacity layered), PBAs	Sacrifices specific
Multi-element/high-entropy doping	Introduction of multiple TM cations at TM sites	Introduces compositional disorder and local environment diversity; disrupts long-range cooperative JT ordering; induces charge disorder	Layered oxides, PBAs, cation-disordered rock-salts	Precise composition control
Alkali-site doping	Substitution at alkali-metal sites (doping $\text{Mg}^{2+}/\text{Ca}^{2+}$ at alkali sites)	Pillaring effect stabilizes layered structure and raises average TM valence	Layered oxides	May affect alkali-ion diffusion kinetics
Anionic substitution	Fluorine doping (F^- substituting O^{2-})	High electronegativity of F^- increases M-O covalency, stabilizes oxygen orbitals, reduces octahedral symmetry breaking, and suppresses oxygen vacancy formation.	Layered oxides, Li-rich Mn-based oxides	May affect anionic redox activity
Structural engineering	Single-crystal morphology, core-shell architectures, and concentration-gradient structures	Reduces grain boundaries and microcracks, confines JT-active components inside the particle, and buffers volume changes	Spinel, layered oxides, PBAs	Increased manufacturing complexity
Surface coating/interfacial engineering	Oxide (such as Al_2O_3 , MgAl_2O_4), phosphate (such as Na_3PO_4), borate coatings or surface reconstruction	Physical barrier suppresses electrolyte attack and HF-induced dissolution; stabilizes surface against reconstruction.	Spinel, layered oxides	Poor suppression effect on internal stress of particles
Electrolyte/interphase engineering	HF-scavenging additives, fluorinated electrolytes, high-concentration electrolytes, and tailored CEI formation	Forms robust inorganic-rich CEI; isolates cathode from reactive species; modulates spin state and charge distribution of TM ions	Especially Mn-based cathodes	High requirements for the composition and stability of the electrolyte
Harnessing beneficial JT distortion	Controlled retention of JT-active ions (especially iron-based cathodes); dynamic distortion activation	Dynamic octahedral buckling enlarges ion diffusion channels; lowers migration barriers; disperses lattice strain	Fe-containing layered Na oxides, Mn-based P2-type layered oxides	Difficult to quantify the JT-active centers

4.2. Anionic Substitution

Anionic regulation primarily relies on fluorine doping to strengthen transition-metal-oxygen bonds and suppress Jahn-Teller activity. The high electronegativity of F^- increases the covalency of metal-ligand interactions, stabilizes oxygen orbitals, and reduces octahedral symmetry breaking [87,88]. F-doping also reduces the concentration of oxygen vacancies and inhibits electrolyte decomposition, indirectly alleviating lattice distortion and transition-metal dissolution. The anion-based strategy provides additional stabilization beyond cation doping, especially for voltage-decaying and low-stability cathodes.

4.3. Structural and Interfacial Engineering

Structural engineering manipulates morphology, defects, and heterointerfaces to alleviate strain and block cooperative Jahn-Teller distortion. Single-crystal structures reduce grain boundaries and microcracks, avoiding strain accumulation and intragranular fracture induced by Jahn-Teller distortion. Core-shell and concentration-gradient architectures confine Jahn-Teller-active components inside the particle while using stable outer layers to buffer volume changes and prevent transition-metal dissolution [89,90]. Surface coatings such as oxides, phosphates, and fluorides act as physical barriers to suppress electrolyte attack and stabilize surface reconstruction. Moreover, defect and strain engineering can be used to regulate local octahedral symmetry, offset intrinsic Jahn-Teller distortion, and disperse lattice strain. In some cases, moderate lattice strain can even beneficially trap local distortion and prevent long-range cooperative behavior.

4.4. Electrolyte and Interphase Engineering

Electrolyte optimization improves interfacial stability and reduces chemical driving forces for Jahn-Teller-related degradation. Using HF-scavenging additives and hydrolytically stable salts minimizes acid-induced transition-metal dissolution triggered by Jahn-Teller-activated oxygen sites. Fluorinated electrolytes and high-concentration electrolytes form a robust, inorganic-rich CEI, which isolates the cathode from reactive solvents and suppresses surface distortion [91,92]. In addition, advanced electrolytes can also modulate the spin state and charge distribution of transition-metal ions, directly weakening electronic degeneracy and Jahn-Teller activity. Rational electrolyte design thus provides a useful surface reconstruction method to mitigate Jahn-Teller effects without altering the structure of cathodes.

4.5. Harnessing Beneficial Jahn-Teller Effects

Instead of only suppressing Jahn-Teller distortion, emerging studies aim to utilize favorable Jahn-Teller behavior to enhance electrochemical performance. Proper Jahn-Teller distortion can dynamically expand ion-diffusion channels, lower Na^+ migration barriers, and improve rate capability, as demonstrated in Fe-containing layered sodium oxides. The dynamic octahedral buckling induced by high-spin Fe^{4+} creates a more flexible local environment that facilitates ion diffusion at high states of charge. Similarly, moderate local Jahn-Teller distortion can release internal lattice strain and improve structural reversibility. By balancing detrimental and beneficial contributions, rational exploitation of the Jahn-Teller effect opens new opportunities for designing high-power and stable electrode materials.

5. Conclusions and Outlooks

The Jahn-Teller effect in cathodes for rechargeable secondary ion batteries has been systematically examined in this review. In accordance with many reports, the Jahn-Teller effect is not absolutely harmful. Its impact depends on the coordination conditions, playing a role from the root of degradation or performance enhancement.

In most cases, the Jahn-Teller effect significantly impacts the structural stability and electrochemical performance of cathode materials, and its mechanisms of action are relatively similar across different materials. Most of the strategies ultimately aim to reduce the concentration or reactivity of Jahn-Teller active centers, whether through cation dilution, anion strengthening, or physical isolation. In spinel $LiMn_2O_4$, the distortion induced by Mn^{3+} triggers a phase transition from cubic to tetragonal, accompanied by a substantial volume change, which in turn induces particle microcracking, electrical disconnection, and rapid capacity decay. In layered oxide systems (including O3-type Li-ion cathodes, P2/O3-type Na-ion cathodes, and P3-type K-ion cathodes), the primary active centers are high-spin Mn^{3+} , low-spin Ni^{3+} , and high-spin Fe^{4+} , which generate local non-cooperative distortions that cause intragranular cracking, surface reconstruction, and increased interfacial impedance. Beyond structural degradation, the Jahn-Teller effect can also lead to transition metal ion dissolution via a Lewis acid-base mechanism. The axial elongation of metal-oxygen bonds enhances the Lewis basicity of axial oxygen atoms,

making them susceptible to proton attack by HF in the electrolyte, which then triggers metal oxidation and disproportionation reactions, ultimately dissolving Mn^{2+} into the electrolyte, which can migrate to the anode and damage the SEI. In Prussian blue analogues, the distortion of Mn^{3+} tilts the MnN_6 octahedra, leading to distortion of the cyanide-bridged coordination framework and reduced ionic conductivity. In aqueous zinc-ion batteries, the Mn^{3+} distortion in MnO_2 -based cathodes induces a disproportionation reaction ($\text{Mn}^{3+} \rightarrow \text{Mn}^{2+} + \text{Mn}^{4+}$), resulting in Mn^{2+} dissolution and irreversible capacity loss. These examples indicate that the Jahn-Teller distortion is one of the causes of structural degradation and interfacial failure in various cathode systems.

However, the Jahn-Teller effect can play a positive role in layered oxide cathodes, rather than being purely detrimental. With proper regulation, this effect can be beneficial to electrochemical performance. The dynamic distortion of Fe^{4+} at high states of charge can widen sodium-ion diffusion channels and lower the migration barrier, thereby enabling high capacity even under Na-deficient conditions. The distortion of Mn^{3+} effectively disperses lattice strain and suppresses cracking, thus preserving structural integrity. In addition, this effect can drive interfacial reconstruction, forming a protective dual-layer structure that comprises a spinel intermediate layer and an outer KF-rich CEI, which enhances cycling stability and Coulombic efficiency. Therefore, judiciously regulating rather than indiscriminately suppressing the Jahn-Teller distortion offers a new pathway for the design of high-performance layered oxide cathodes.

Future strategies for the Jahn-Teller effect should focus on regulation rather than elimination. Instead of viewing it as inherently harmful, we should consider its potential to enhance performance. This requires clarifying the electronic and atomic origins of distortion–property relationships. Specifically, it mainly includes the following perspective:

(1) Investigation of both detrimental and beneficial Jahn-Teller effects: In previous studies, the impact of this effect has generally been treated qualitatively, with most reports simply showing that suppressing the distortion improves electrochemical performance. Systematic quantitative studies across different material systems remain scarce. Given the dual nature of the Jahn-Teller effect, it is important to clarify under what conditions the distortion facilitates ion transport or relieves lattice strain, and under what conditions it triggers irreversible structural degradation. This would transform the exploitation of beneficial distortion from a serendipitous observation into a verifiable basis for materials design.

(2) Regulation of Jahn-Teller distortion via alkali-site doping: Current modification strategies are mainly focused on transition-metal-site doping. However, the ionic size and electronegativity of alkali ions ($\text{Li}^+/\text{Na}^+/\text{K}^+$) can affect the interlayer spacing and the charge state of axial oxygen atoms. For example, introducing an appropriate number of hetero-valent ions at the alkali site (such as doping Mg^{2+} at the K^+ or Na^+ site) can simultaneously act as the pillaring effect to stabilize the layered structure, increase the average valence state of transition-metal ions, and dilute the concentration of Jahn-Teller active centers.

(3) Exploring beneficial Jahn-Teller distortion in high-entropy structures: High-entropy systems possess compositional disorder and local coordination environment diversity, which can suppress cooperative distortion and potentially retain local structural dynamics that facilitate ion diffusion during cycling. The role of Jahn-Teller distortion under the influence of the four major effects of the high-entropy effect, lattice distortion effect, sluggish diffusion effect, and cocktail effect in high-entropy systems should be explored. This class of materials thus offers a promising database for investigating beneficial Jahn-Teller effects and deserves further investigation as a priority direction.

(4) Establishing descriptors for the Jahn-Teller effect in cathode materials: Currently, the relative reports lack specialized descriptors that can directly reveal the relationship between the type and nature of distortion and the electrochemical performance of electrode materials. Combined high-throughput computational screening and experimental validation could enable the identification of the boundary conditions between beneficial and detrimental distortion across different material families, thereby revealing how specific distortion modes correlate with performance metrics.

(5) Developing a failure model coupled with Jahn-Teller distortion: Material degradation induced by Jahn-Teller distortion is not only a crystallographic issue, but rather the result of coupled electronic structure, mechanical strain, and interfacial chemistry. A computational model that integrates these three aspects is needed to transform the research mode of failure first and analysis later into a predictable and controllable mode, thereby providing theoretical support for the future design of advanced cathode materials. The future work is to unravel the mechanistic link between Jahn-Teller distortion and phase-transition behavior, so that specific local distortions can be directly identified as triggers of structural failure.

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Y.L.: Writing—reviewing and editing. S.L.: Writing—reviewing and editing. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest

The authors declare no conflict of interest.

Use of AI and AI-Assisted Technologies

No AI tools were utilized for this paper.

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