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Enhanced High-Rate Capability of $\text{LiNi}_{0.83}\text{Co}_{0.11}\text{Mn}_{0.06}\text{O}_2$ Cathodes by Homogeneous Sr^{2+} Doping

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Abstract:

To suppress capacity degradation of Ni-rich $\text{LiNi}_{0.83}\text{Co}_{0.11}\text{Mn}_{0.06}\text{O}_2$ (NCM) cathodes with radially oriented primary grains, a homogeneous Sr^{2+} doping strategy was developed by controlling the Sr^{2+} diffusion path from the surface into the bulk. Introducing SrO during sintering enabled Sr^{2+} to penetrate along the radially aligned primary grains, achieving uniform distribution throughout the material. The NCM-Sr cathode with 0.7% Sr^{2+} exhibited superior rate performance, delivering specific capacities of 182.5, 172.3, and 165.7 mAh g⁻¹ at 1C, 5C, and 10C, respectively, compared to 167.3, 148.1, and 143.2 mAh g⁻¹ for the undoped sample. Uniform Sr^{2+} incorporation, facilitated by radial grain alignment, effectively mitigates localized strain accumulation during cycling, enhancing structural stability. For the first time, NCM cathodes were analyzed using deep learning in conjunction with detailed morphological information, confirming that controlled particle orientation and uniform small primary particles significantly improve electrochemical performance. These surface-engineered cathodes demonstrate extended cycle life, stable voltage profiles, and excellent high-rate capabilities, highlighting an effective strategy for advancing Ni-rich ternary cathode materials in high-energy lithium-ion batteries.

Keywords: Ni-rich layered oxides, Sr^{2+} Doping, Lithium ions battery , Lithium-reactive coating

1. Introduction

Layered $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$ ($x \geq 0.8$; NCM) has emerged as a promising cathode material for high-performance electric vehicles (EVs), owing to its high specific capacity, high energy density, and relatively low material cost associated with reduced cobalt content[1–10]. Commercial Ni-rich cathodes generally adopt a polycrystalline architecture to achieve a high tap density and low specific surface area[11–15]. These polycrystalline secondary particles consist of densely packed nanoscale primary particles, which contribute to increased energy density and reduced interfacial side reactions[16–18]. Despite these advantages, polycrystalline NCM is susceptible to detrimental intergranular cracking caused by severe anisotropic mechanical stress, particularly during the H2-H3 phase transition at high voltages, which is accompanied by pronounced contraction along the c-axis. Such cracking further accelerates interfacial side reactions and surface reconstruction during prolonged cycling[19,20]. In addition, the fast-charge and fast-discharge performance of NCM at low and high states of charge (SOCs) is severely constrained by sluggish Li^+ -diffusion kinetics[6,21]. This limitation is primarily associated with the high activation energy resulting from the low concentration of Li vacancies at low SOC and the substantial c-axis contraction induced by the H2-H3 phase transition at high SOC[22,23]. Collectively, these phenomena lead to capacity degradation, lattice-oxygen loss and poor rate capability[24]. Rational structural design and interfacial modification are therefore required to overcome these challenges and facilitate the broader commercialization of Ni-rich NCM cathodes.

Various strategies have been developed to enhance the electrochemical performance of NCM, including elemental doping[25–28], surface modification[29,30], and morphological control, among which elemental doping is considered particularly practical. Elemental doping can improve cathode performance by regulating the atomic and electronic structures, controlling grain size, and suppressing Li/Ni cation disorder[31,32].

Previous studies have primarily focused on trace doping with Nb^{5+} [33,34], Al^{3+} [35], Zr^{4+} [36], Ti^{4+} [37], Ta^{5+} [15,38], etc. By contrast, Sr^{2+} doping in polycrystalline NCM has received comparatively little attention, despite its potential to enhance structural stability, electrochemical reaction kinetics, Li^+ -diffusion kinetics, and rate capability. Although doping can improve the structural and interfacial

durability of NCM cathodes, developing a cost-effective, single-step doping method remains a significant technological challenge.

In this study, Sr^{2+} was introduced into Ni-rich NCM to construct a multifunctional Sr-doped cathode with enhanced structural and interfacial stability and improved electrochemical kinetics. Sr^{2+} was successfully incorporated into the crystal lattice, thereby stabilizing the layered structure and reducing residual lithium species on the particle surface. Moreover, owing to its large ionic radius (1.18 Å), Sr^{2+} acted as a structural pillar that expanded the lattice parameters and increased the O-Li-O slab spacing, thereby facilitating Li^+ diffusion and improving rate capability. A series of Sr-modified NCM samples was synthesized and systematically evaluated. The sample containing 0.7 wt% Sr exhibited excellent cycling stability, retaining 94.4% of its initial capacity after 100 cycles at 5 C and delivering a high discharge capacity of 169.1 mAh g^{-1} at the same rate. These results demonstrate the feasibility of developing Ni-rich cathode materials that simultaneously exhibit high-rate capability and prolonged cycling stability. This work presents a simple synthesis strategy for modifying Ni-rich layered cathodes and provides a promising pathway toward next-generation lithium-ion batteries with lower cost, higher energy density, and extended service life.

2. Experimental

2.1 Preparation of Sr-Modified NCM

The commercial $\text{Ni}_{0.83}\text{Co}_{0.11}\text{Mn}_{0.06}(\text{OH})_2$ and analytical-grade $\text{LiOH}\cdot\text{H}_2\text{O}$ and SrO were purchased from Aladdin Biochemical Technology Co., Ltd., without further purification. To synthesize the Sr-doped cathode material, the prepared precursor was thoroughly mixed with $\text{LiOH}\cdot\text{H}_2\text{O}$ and SrO using a high-speed mixer. This mixture was then calcined at 765°C for 10 hours in an oxygen atmosphere. The molar ratio of the precursor to the lithium source was 1:1.03, and the weight ratio of SrO to the precursor was 0.007:1 (0.3 wt%). For simplicity, the resulting cathode material was named NCM-Sr-0.7. To investigate the influence of SrO on crystal growth and electrochemical performance, three additional cathode materials were synthesized under the same calcination conditions, with 0 wt%, 0.3 wt%, 0.7 wt%, and 1 wt% SrO added to the precursor. These were named NCM-Sr-0, NCM-Sr-0.3,

NCM-Sr-0.7, and NCM-Sr-1, respectively.

2.2 Material characterization

The crystal structure of the prepared cathode powders was characterized using X-ray diffraction (XRD, Rigaku Ultima IV, Japan). The morphology of the samples was observed using a field emission scanning electron microscope (FE-SEM, SUPRA 55, Zeiss, Germany). Additionally, the valence states of Ni and Sr were analyzed using X-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha spectrometer).

2.3 Electrochemical characterizations

2.3.1 Half cells

First, the freshly prepared cathode powder was mixed thoroughly with acetylene black and polyvinylidene fluoride (PVDF) at a weight ratio of 8:1:1 using an agate mortar. Then, a suitable amount of N-methylpyrrolidone (NMP) was added to the mixture to form a flowable slurry. This slurry was then evenly coated onto aluminum foil using a scraper. The coated aluminum foil was then vacuum-dried at 110°C for 10-13 hours to ensure complete drying, after which it was cut into disc-shaped electrodes with a diameter of 12 mm. The active material loading on each electrode was approximately 1.2 mg/cm². The electrodes were assembled in a glovebox filled with argon (Mikrouna). Lithium metal discs (16 mm in diameter) were used as counter electrodes in CR2032 coin cells, and the electrolyte consisted of 1 M LiPF₆ dissolved in a 1:1:1 mixture of solvents: ethylene carbonate (EC), ethyl methyl carbonate (EMC), and dimethyl carbonate (DMC). The electrolyte also contained 2% fluoroethylene carbonate (FEC) and 1% vinylene carbonate (VC). Galvanostatic charge-discharge tests were conducted within a voltage range of 2.8-4.3 V (relative to Li⁺/Li) using a battery testing system (Neware). Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) tests were performed on an electrochemical workstation (Shanghai Chenhua CHI 760).

3 Results and Discussion

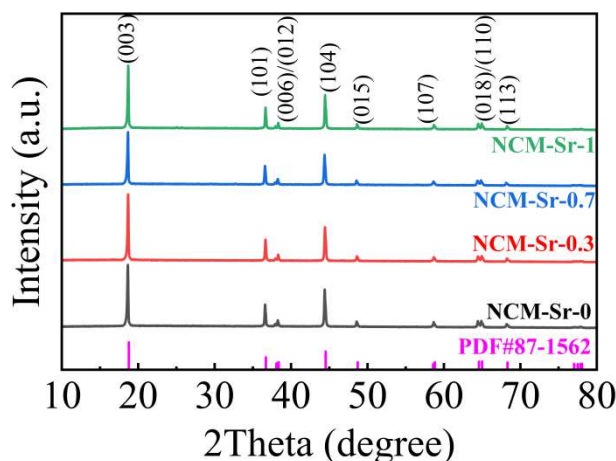


Fig. 1 XRD spectra of pristine NCM-Sr-0, NCM-Sr-0.3, NCM-Sr-0.7, and NCM-Sr-1

The XRD spectra can be attributed to the hexagonal α -NaFeO₂ (R-3m) structure without impurity phases, which agrees well with the standard diffraction pattern of LiNiO₂ (PDF#87–1562), as shown in Fig. 1. The distinct splitting of the (006)/(012) and (018)/(110) diffraction peaks indicates the presence of an ordered layered structure in NCM-Sr-X. No characteristic peaks of Sr-containing impurities were observed for NCM-Sr-X (X = 0.3, 0.7, 1). Therefore, a small amount of SrO significantly impacts the microstructure during the calcination, but does not affect the crystal structure of the layered cathode. The R-values marked in Table 1 represent the intensity ratio of the (003)/ (104) peak and reflect the degree of cation mixing. The R-values for the samples are greater than 1.2, indicating that the cationic order is well maintained. In Fig. S1(a-b), the distinct splitting of the (006)/(102) and (018)/(110) doublets at approximately 38°-38.6° and 64°-65°, respectively, is shown. This further supports the well-ordered hexagonal layered crystalline structure after Sr²⁺ doping.

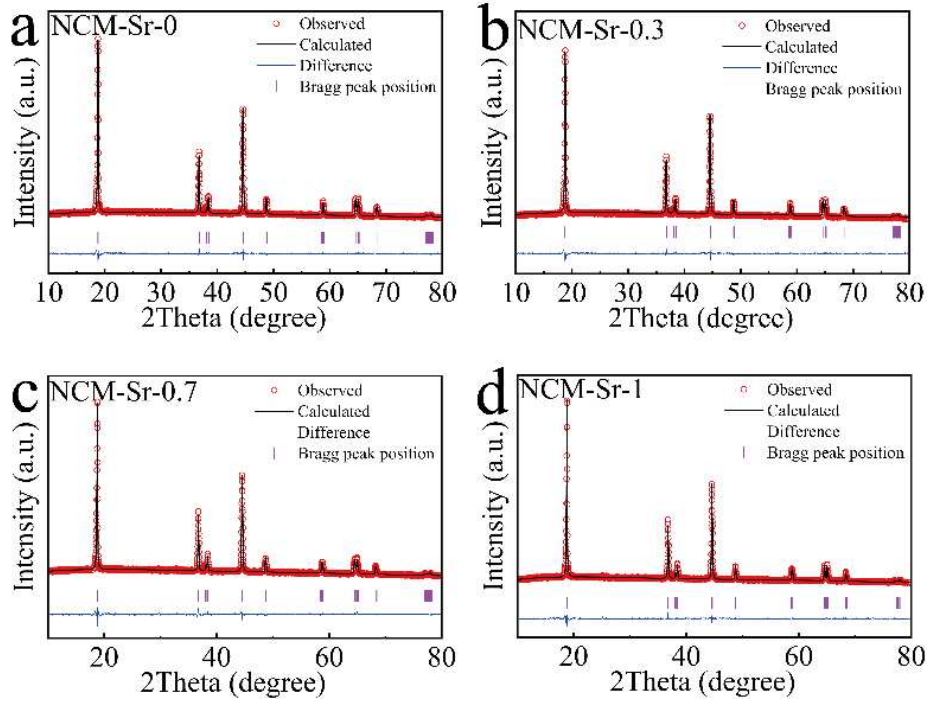


Fig. 2 (a) NCM-Sr-0 refined spectrum; (b) NCM-Sr-0.3 refined spectrum; (c) NCM-Sr-0.7 refined spectrum; (d) NCM-Sr-1 refined spectrum.

The XRD spectra were further refined using the Full Prof Rietveld method and are shown in Fig. 2 (a-d). The lattice parameters of the NCM-Sr-X are a and c , as shown in Table 1. As the amount of Sr^{2+} increases, it is observed that a and c of NCM-Sr-X become larger than those of the undoped materials and remain relatively unchanged. Meanwhile, as the Sr^{2+} content increases from 0.3% to 1%, a , c , and the unit volume all increase. This is because the ionic radius of Sr^{2+} (0.64 Å) is larger than that of Ni^{3+} (0.56 Å), Co^{3+} (0.545 Å), and Mn^{4+} (0.53 Å), but smaller than that of Li^{+} (0.76 Å)[39]. Sr^{2+} can easily replace some of the metal ions, resulting in an increase in the lattice parameters of the material. The c/a ratio of the ideal lattice structure is greater than 4.93[30]. The c/a ratios of NCM-Sr-X in Table 1 are greater than 4.93, which confirms that lattice delamination is not caused by Sr^{2+} doping. Additionally, the $I(003)/I(104)$ ratio indicates the degree of cation mixing in the layered structure. It is well documented that low cation mixing occurs when $I(003)/I(104)$ exceeds 1.2[30], which facilitates Li-ion transfer. The $I(003)/I(104)$ ratio of NCM-Sr-0.7 (1.68) was higher than those of NCM-Sr-0 (1.58), indicating a lower degree of cation mixing and a better crystalline order structure. Furthermore, given that the bond dissociation energy of Sr-O is much stronger than that of Li-O, introducing Sr^{2+} into the lithium layer of the host structure is expected to enhance the structural stability. Therefore, Sr^{2+} doping in the Li^{+} layer

possibly enhances the stability of long-term charge/discharge cycling and improves the material's electrochemical performance. As shown in Table 1, the c-axis value increases with increasing of Sr^{2+} doping amount. The larger the c-axis, the larger the Li^+ migration pathway, indicating that Sr^{2+} enlarges the Li^+ migration path and thus enhances the material's rate performance.

Table 1

Results of Rietveld analysis of NCM-Sr-X (X = 0, 0.3, 0.7, 1).

Sample	a (Å)	c (Å)	c/a	V (Å)	I(003)/I(104)	Rwp (%)
NCM-Sr-0	2.86544	14.17406	4.9465	100.788	1.58	3.82%
NCM-Sr-0.3	2.86515	14.18192	4.9498	100.824	1.65	3.37%
NCM-Sr-0.7	2.86715	14.2044	4.9515	101.132	1.68	3.21%
NCM-Sr-1	2.86563	14.17909	4.9479	100.837	1.64	3.33%

The morphology or microstructure of NCM-Sr-X (X = 0, 0.3, 0.7, 1) was investigated using SEM, as shown in Fig. 3(a-d). The samples display spherical particles with a diameter of about 9-10 μm , composed of many small particles. They gradually become larger, smoother, and denser with an increase in the Sr^{2+} doping ratio, but retained a spherical morphology, indicating that the Sr^{2+} doping cations did not affect the morphology. SEM images of (a) NCM-Sr-0 and (b) NCM-Sr-0.7 after 100 cycles at 1 C are shown in Fig. S5(a, b). The surface of NCM-Sr-0.7 has no microcracks, which are attributed to the Sr^{2+} doping. In comparison, the change in particles could shorten the Li^+ diffusion path and reduce the volume expansion during charging and discharging.

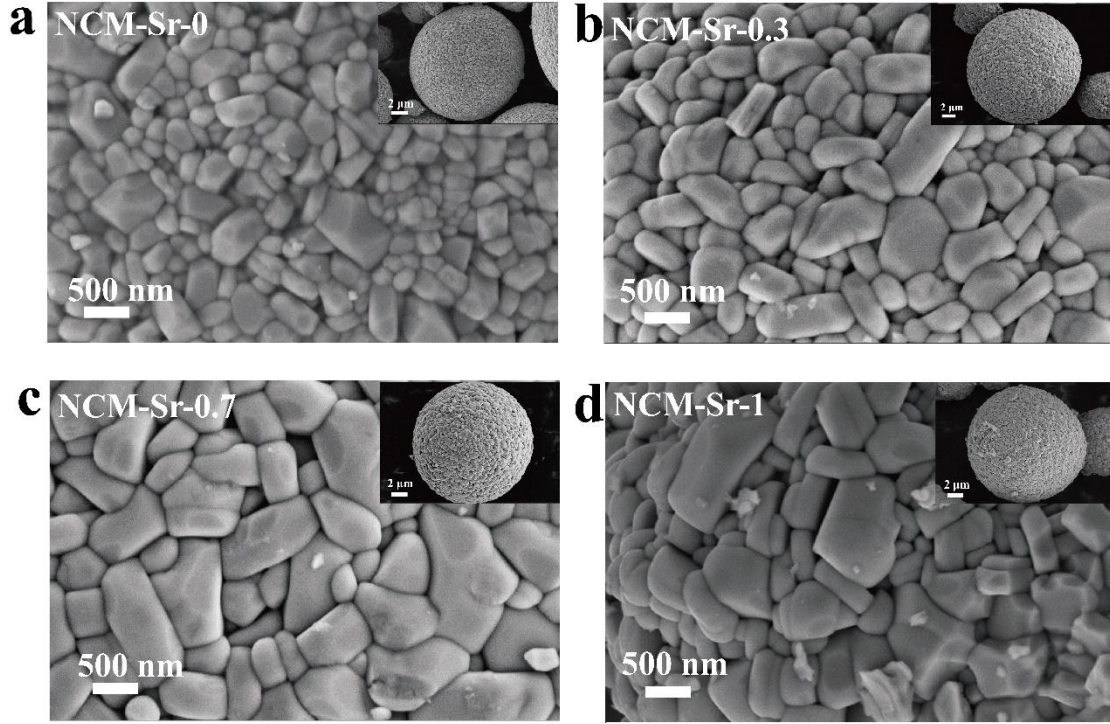


Fig. 3 SEM images of NCM-Sr-X (X = 0 (a), 0.3 (b), 0.7 (c), 1 (d)), respectively.

The particle morphology and microstructure were further characterized by TEM and HRTEM in Fig. 4(a-b). Fig. 4(b) shows that the interplanar spacing of the (003) plane in the NCM-Sr-0.7 sample is about 0.473 nm, which is larger than the 0.470 nm observed in Fig. 4(a) for the sample with fewer Li/Ni disorder defects[40]. This is consistent with the results of XRD Rietveld refinement (see Fig. S1 and Table 1). To further clarify the elemental distribution of the particles, especially the dopant element Sr, the TEM mapping is shown in Fig. S2(a-b). The Sr element was uniformly distributed (red color) and no element aggregation occurred in Fig. S2(b). All elements (Li, Ni, Co, and Mn) were uniformly distributed, as shown in Fig. S2, contributing to the stability of the structure and cyclic stability[41].

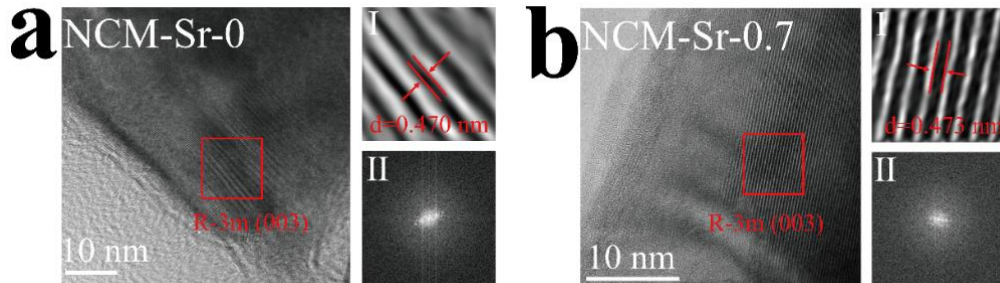


Fig. 4 Cross-sectional HRTEM images (inserts I) of TEM images and the corresponding FFT conversions (inserts II) of (a) NCM-Sr-0 and (b) NCM-Sr-0.7.

To investigate the effect of Sr^{2+} doping on the chemical states of the elements in the NCM material, XPS spectra were obtained for the NCM-Sr-0 and NCM-Sr-0.7 in

Fig. 4(a-f). The spectra were fitted using XPS Advantage software. Fig. 4(a, b) shows the Ni 2p valence state. The spectra of the samples were well fitted to peaks corresponding to Ni 2p_{3/2} at 861.1 eV (satellite peak), 856.1 eV (Ni³⁺), and 854.9 eV (Ni²⁺). The relative content of Ni²⁺ is about 55% in the target sample NCM-Sr-0.7, which is significantly higher than the 43.45% content of the optimized layered structure in NCM-Sr-0. Some Sr²⁺ ions diffused into the lattice of the NCM particles, acting as stabilizers and causing partial reduction of Ni³⁺ to Ni²⁺, thereby achieving charge compensation^{16, 21}. Although an increased Ni²⁺/Ni³⁺ ratio typically induces cation mixing, the degree of cation mixing decreased after Sr²⁺ doping. This is likely because Sr²⁺ preferentially occupies lithium sites, and the large repulsive force inhibits Ni²⁺ migration into the lithium layer, thereby reducing the extent of Li/Ni cation mixing[42,43].

The C 1s spectra of NCM-Sr-0 and NCM-Sr-0.7 in Fig. 4 (c, d) exhibit four peaks that correspond to adventitious carbon[44] (at ~284.8 eV and 286.0 eV), O-C=O (at ~288.0 eV), and carbonate (at ~289.8 eV), which are derived from Li₂CO₃. The CO₃²⁻ peak at ~289.8 eV indicates that the content of this component decreased significantly from 19.1% to 8.0% after Sr²⁺ doping, suggesting that Sr²⁺ effectively restrained the generation of Li₂CO₃[14]. Fig. 4(f) shows a strong signal peak for Sr 3d in NCM-Sr-0.7 compared to NCM-Sr-0, as shown in Fig. 4(e), indicating the presence of Sr elements. The signal peak spectra were well fitted to two peaks corresponding to Sr 3d_{5/2} and Sr 3d_{3/2}, located at 135.4 eV and 133.5 eV, respectively[30]. This result is consistent with the XRD.

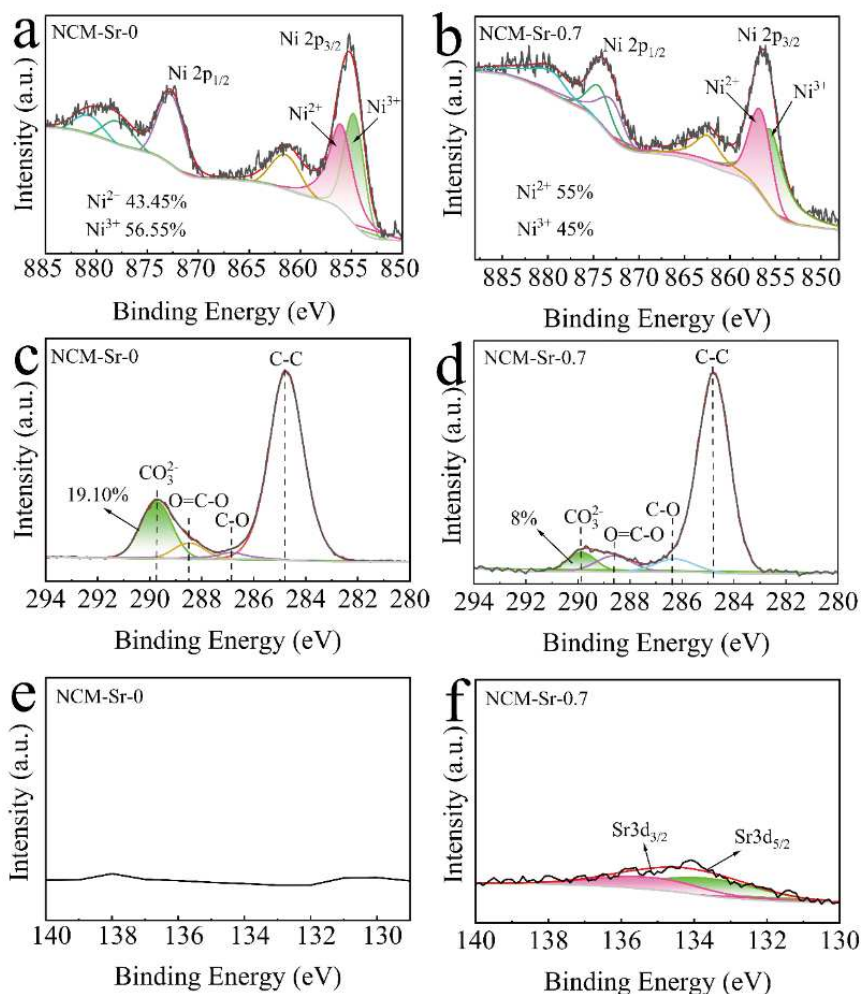


Fig. 5 (a) XPS spectra and fitting results for the surfaces of NCM-Sr-0 Ni 2p, (b) NCM-Sr-0.7 Ni 2p (c) NCM-Sr-0 C 1 s, (d) NCM-Sr-0.7 C 1 s, (e) NCM-Sr-0 Sr 3d, and (f) NCM-Sr-0.7 Sr 3d.

The electrochemical properties of NCM-Sr-X ($X=0, 0.3, 0.7, 1$) were tested using CR2032 button-type lithium metal cathode half-cells. The cells were cycled at 3 th at a cut-off voltage of 2.7-4.3 V and a constant current density of 0.1 C and 1 C under 180 mAh g⁻¹, as shown in Fig. 6(a). The initial discharge capacities of NCM-Sr-X ($X = 0, 0.3, 0.7, 1$) at 0.1 C were 182.1, 184.6, 208.7, and 183.4 mAh g⁻¹, with first-time coulombic efficiencies of 82.3%, 85.7%, 87.2%, and 84.1%, respectively. The charge-discharge graphs at 0.1 C are similar; however, the discharge specific capacity of NCM-Sr-0.7 increased slightly, because SrO possibly reacted with LiOH during calcination to form the fast ionic conductor LiSrO₄.

Fig. 6(b) shows the initial capacities of NCM-Sr-X ($X = 0, 0.3, 0.7, 1$) at the test temperature of 25 °C 1 C and the cutoff voltage of 2.7-4.3 V: 167.3 mAh g⁻¹, 161.8 mAh g⁻¹, 182.5 mAh g⁻¹, and 167.4 mAh g⁻¹, respectively. The capacity retention rates of NCM-Sr-X ($X = 0, 0.3, 0.7, 1$) are 95.5%, 95%, 97.69%, and 96.7%, respectively, after 100 charge/discharge cycles. In the high-Ni cathode material, NCM-Sr-0.7 could

effectively retain cycling stability; the specific capacity increased slightly at 0.1C and more significantly at 1C.

In addition, NCM-Sr-X (X=0, 0.3, 0.7, 1) at 25 °C, with a cutoff voltage of 2.7-4.3 V and a charging rate of 0.2 C, was tested (see Fig. 6(c)). During charge/discharge testing at current densities of 0.1, 0.2, 0.5, 1, 2, and 5 C (1 C = 180 mAh g⁻¹), the NCM-Sr-0.7 electrode retained reversible capacities of 201, 194.3, 186.4, 179.6, 174.5, and 169.1 mAh g⁻¹, while the NCM-Sr-0 electrode possessed specific capacities of 186.1, 173.6, 168.4, 161.4, 156.6, and 148.1 mAh g⁻¹, respectively. Fig. 6(d) shows the cycling performance at a high current density of 5 C. NCM-Sr-0.7 has a capacity of 169.1 mAh g⁻¹, highlighting its excellent performance compared to NCM-Sr-0, which has a capacity of 148 mAh g⁻¹. The discharge capacity of NCM-Sr-0.7 is about 161.1 mAh g⁻¹ higher than that of NCM-Sr-0, which is about 143.2 mAh g⁻¹ at 10 C, as shown in Fig. S3.

Overall, the discharge capacities of NCM-Sr-0.7 are higher than those of NCM-Sr-0 at all of the current densities, and rate performance is further improved after Sr²⁺ doping. This is probably because Sr²⁺ doping reduces the lattice parameter and the extent of Li/Ni cation mixing, thus improving the lithium mobility[45] and the electrochemical performance of NCM-Sr-0.7.

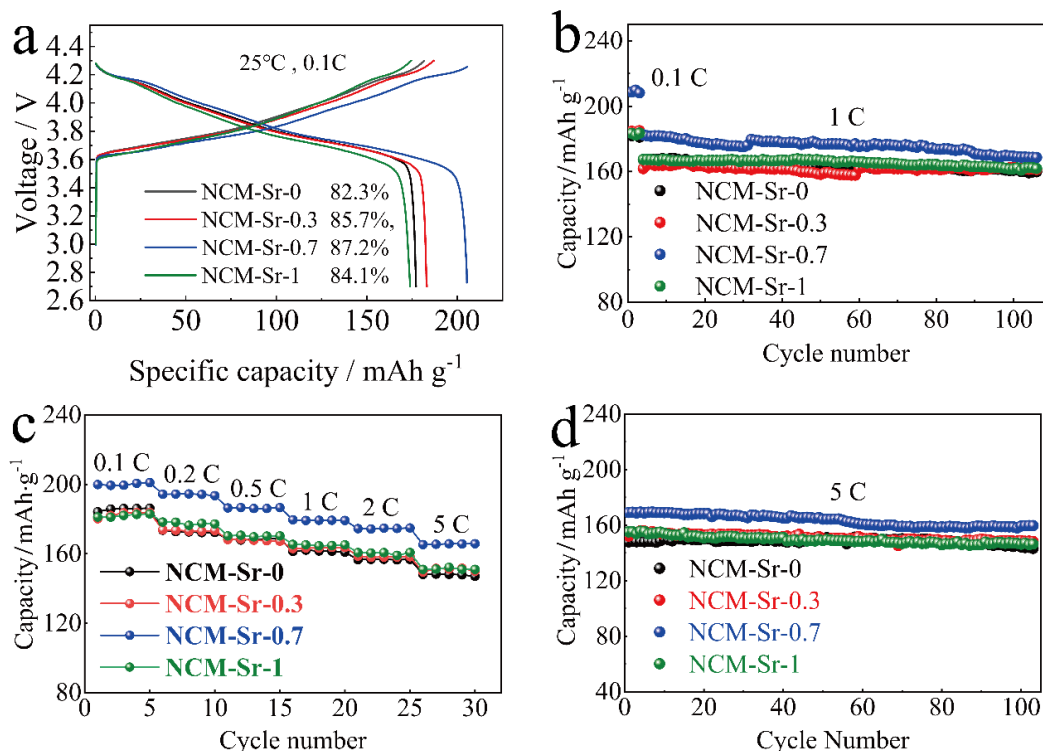


Fig. 6 (a) Initial charge/discharge curves, (b) cycled curves at 0.1 C (3 th) and 1 C (100 th), (c) different current multiplicities curves of NCM-Sr-X; (d) long-multiplicity performance of NCM-Sr-X.

CV was performed to investigate the kinetics of the electrode reaction of NCM-Sr-0 and NCM-Sr-0.7. The CV curves for the first three cycles are shown in Fig. 7(a, b). Both samples showed a pair of distinct peaks between 3.6 V and 4.0 V at the same time, which corresponded to $\text{Ni}^{2+}/\text{Ni}^{3+}$ redox reaction[46,47]. The voltage difference (ΔE) between the oxidation and reduction peaks indicated electrode polarization. For the NCM-Sr-0 electrode, the oxidation and reduction peaks in Fig. 7(a) in the first cycle are 4.022 V and 3.70 V, respectively; the corresponding value of ΔE_1 is 0.322 V. For the NCM-Sr-0.7 electrode, the corresponding values are 3.9 V and 3.69 V in Fig. 7(b), with a reduced value of ΔE_1 of 0.226 V. Similarly, the peak voltage differences (ΔE_2 , ΔE_3 , and ΔE_4) of the NCM-Sr-0.7 electrode decreased, suggesting that the Sr^{2+} doping significantly reduces polarization and improves the kinetics of the electrode reaction[48]. This is consistent with the fact that Sr^{2+} doping reduces the production of surface impurities.

The EIS spectra were recorded for NCM-Sr-0 and NCM-Sr-0.7 at the 3 th, 50 th, and 100 th turns of the charge cycles at 1C, within a voltage range of 2.7-4.3 V at 25 °C, as shown in Fig. 7 (c, d). The first semicircle at high frequency is attributed to the diffusion resistance (R_f) of lithium ions through the interfacial layer of the solid electrolyte. The subsequent semicircle at high frequency represents the charge transfer resistance (R_{ct}) between the electrodes and the electrolyte. The low-frequency diagonal line corresponds to the diffusion of lithium ions in the solid electrode (Z_w)[43]. The fitting results are shown in Table S2. The impedance of all components is reduced by about half after 50 cycles; the increase in impedance is largely suppressed by Sr^{2+} doping, which contributes to the formation of a stable interface between the organic electrolyte and the NCM cathode.

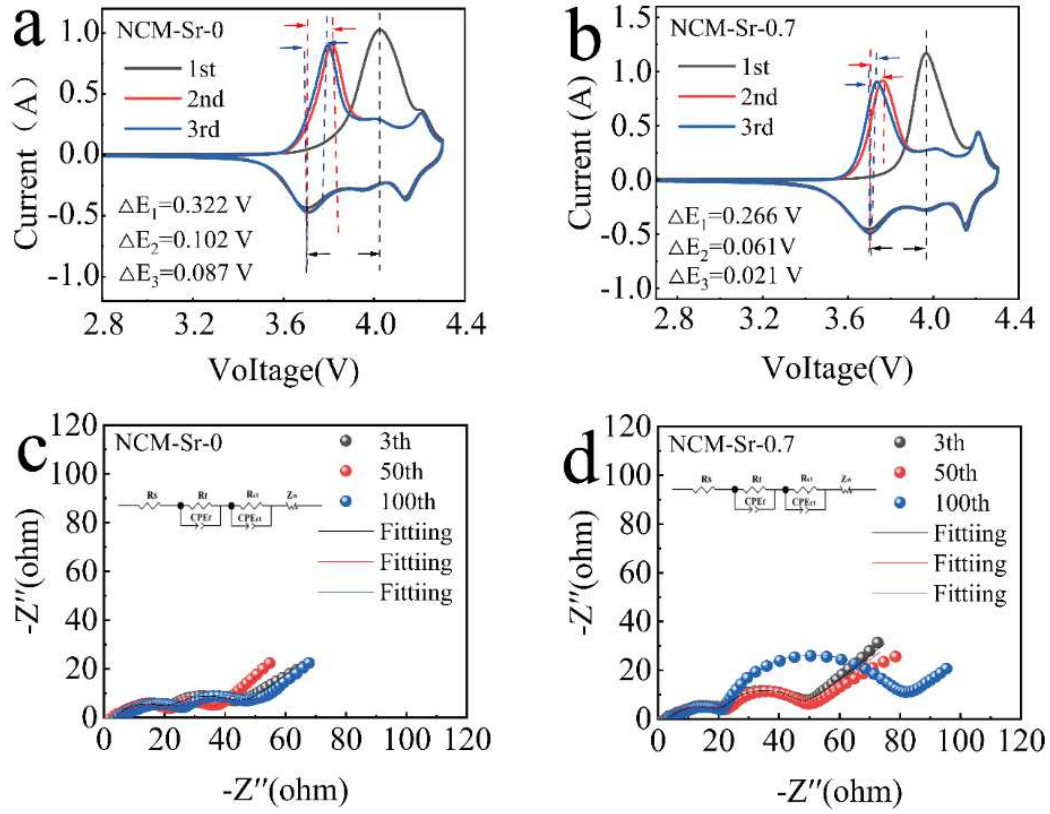


Fig. 7 CV curves of (a) NCM-Sr-0 and (b) NCM-Sr-0.7 recorded at 0.1 mV s⁻¹; Nyquist plots and equivalent electric circuit diagrams (inserts) of (c) NCM-Sr-0 and (d) NCM-Sr-0.7 charge after 3, 50, and 100 cycles.

To further improve Li⁺ diffusivity, the GITT technique was used to evaluate the lithium diffusion coefficients (D_{Li^+}) of NCM-Sr-0 and NCM-Sr-0.7. The GITT curves obtained during charging are shown in Fig. S4(a, b). D_{Li^+} can be calculated using the following equation: $D_{Li^+} = \frac{4}{\pi} \left(\frac{m_B V_m}{M_B A} \right)^2 \left(\frac{\Delta E_s}{\tau \left(\frac{dE}{d\tau} \right)} \right)^2 \left(\tau \ll \frac{L^2}{D_{Li^+}} \right)$, where V and $\tau^{1/2}$, D_{Li^+} are changed as same as ΔE_s . It was observed that there was an initial strong increase at lower voltages, followed by stability and a small increase at higher voltages, as shown in Fig. S5. The average D_{Li^+} is about $3.6 \times 10^{-9} \text{ cm}^2 \text{ s}^{-1}$ for NCM-Sr-0.7, which is significantly higher than $2.0 \times 10^{-9} \text{ cm}^2 \text{ s}^{-1}$ for NCM-Sr-0. This higher D_{Li^+} by Sr²⁺ doping can be attributed to the blocking of impurities and enhanced Li⁺ diffusion.

4. Conclusion

Sr^{2+} doping improves the electrochemical performance of NCM. Target NCM-Sr-0.7 has a broadened diffusion path that accelerates the diffusion rate of lithium ions and plays a role in forming morphology. It also has no detrimental effect on the crystal structure. Such low Sr^{2+} doping does not lead to an impurity phase, indicating successful doping. A one-step surface-to-body modification strategy via in situ precursor engineering successfully addressed the severe Li/Ni disorder and interfacial dynamics challenges in advanced polycrystalline, high-Ni, cobalt-rich cathodes. During, the Sr^{2+} ions in the precursor gradually precipitated and became enriched on the surface of the grains, reducing surface energy and facilitating grain growth. Thanks to this integrated structural design, the obtained cathode significantly reduces Li/Ni mixing and lattice oxygen loss during synthesis, enabling fast lithiumion diffusion kinetics. Coin cells assembled with NCM-Sr-0.7 electrodes provided high reversible capacity.

Consequently, NCM-Sr-0.7 delivers high reversible capacities of 208.7, 169.8, and 161.1 mAh g⁻¹ at 0.1, 5, and 10 C, respectively. After 100 cycles in the voltage range of 2.7-4.3 V, it retains 94.4% of its initial capacity at 5 C. This one-step synthesis strategy, enabled by in situ precursor engineering, effectively mitigates cation mixing and simultaneously enhances the reversible capacity, initial Coulombic efficiency (ICE), cycling stability, and rate capability. It thereby offers a promising technological pathway for the modification of Ni-rich cathodes and provides new insights into the scalable synthesis of high-energy-density layered cathode materials with high Ni content.

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4

5 **Author contributions**

6 Teng Ma and Zhikun Zhang were responsible for conceptualization, investigation, data
7 curation, methodology and writing-original draft; Others curated the data; Baohua
8 Zhang, Fuqiang Guo were involved in conceptualization, resources, funding,
9 supervision and writing-editing and reviewing. Teng Ma and Zhikun Zhang contrib-
10 uted equally to this work and should be considered co-first authors. Baohua Zhang,
11 Fuqiang Guo shall be considered as the corresponding author

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21

22 **Declarations**

23 Conflict of interest: The authors declare that there are no competing interests regarding
24 the publication of this paper. Ethical approval and Consent to participate Not applicable.

25

26 Ethical approval: All the carried out experiments did not require any ethical approval.

27

28 Supplementary Information: Not Applicable.

29

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