Research progress of prelithiation technology for positive electrode of Lithium-ion battery

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Abstract. Lithium-ion batteries (LIBs) deliver cardinal advantages including exceptional energy density and extended cycle durability, establishing their critical function in modern energy storage infrastructures. The formation of a solid electrolyte interface (SEI) on the cathode surface during the first cycle of lithium-ion batteries leads to severe irreversible lithium loss, lowering the initial coulombic efficiency (ICE). Cathode prelithiation technology has become a key strategy to compensate lithium loss and improve first effect due to its strong process compatibility. This paper provides a systematic review of cathode prelithiation technology, with emphasis on the prelithiation mechanism, lithium release kinetics, and performance distinctions of ternary, binary, and nanocomposite lithium additives. The effects, challenges and optimization strategies of various additives in improving initial capacity, coulomb efficiency and cycle stability are described. The feasibility and bottleneck of engineering application of prelithiation technology are further discussed, and the future research direction is prospected, which provides theoretical support and technical path for the development of high energy density and long life Lithium-ion batteries.

Keywords: Lithium-ion batteries, Cathode prelithiation, Initial coulombic efficiency, Energy density.

1. Introduction

With the continuous advancement of the global energy structure transformation, the demand for efficient and sustainable energy storage technologies in the energy storage market is growing, and higher requirements are put forward for the range of electric vehicles, a common energy storage device, and developing high-energy-density batteries demands urgent solutions [1]. Lithium-ion batteries (LIBs) serve as the dominant power source and energy storage medium for new energy vehicles, portable electronics, and renewable energy systems, underpinning the advancement of green transportation and sustainable energy frameworks. This prominence stems from their favorable attributes, including high energy density, long cycle life, and robust safety [2,3].

Li-rich Mn-based (LRM) compounds exhibit significant promise for next-generation high-energy-density batteries. Their appeal lies in high operational voltages and the potential for substantial discharge capacities (200~300 $mAh\cdot g^{-1}$) [4,5]. The practical implementation of LRM, however, is hindered by substantial irreversible lithium loss in initial cycles, caused mainly by two factors: significant volume changes in LRM cathodes and repetitive solid electrolyte interphase (SEI) film breakdown/reformation at the anode. Consequently, the initial coulombic efficiency (ICE) is substantially diminished, typically falling within the 60%–85% range. To mitigate these issues and enhance material performance, strategies such as surface coating and elemental doping have been extensively explored. For example, Wei H et al. [6] employed a $LiMnO_3$ coating on LRM, achieving a reversible capacity of 242.2 $mAh\cdot g^{-1}$ at 1C, but its initial coulomb efficiency was still less than 90%. Ding X et al. [7] constructed a multifunctional structure layer on the $Li_{0.2}Mn_{0.6}Ni_{0.2}O_2$ (LMNO) cathode particles, the ICE of the LMNO electrode reached 81.8 %. Unfortunately, the above methods are not ideal for improving battery usable capacity, overall energy efficiency and cycle life.

For the purpose of solving the problem of lithium loss in the first cycle, prelithiation technology was born and has experienced rapid development in the past five years. According to Web of Science data, the number of related papers with the keyword "prelithiation" has surged from 161 in 2015~2019 to 500 in 2020~2024, and has been cited more than 28, 000 times (Figure 1), highlighting The academic and industrial circles are paying close attention to prelithiation methods.

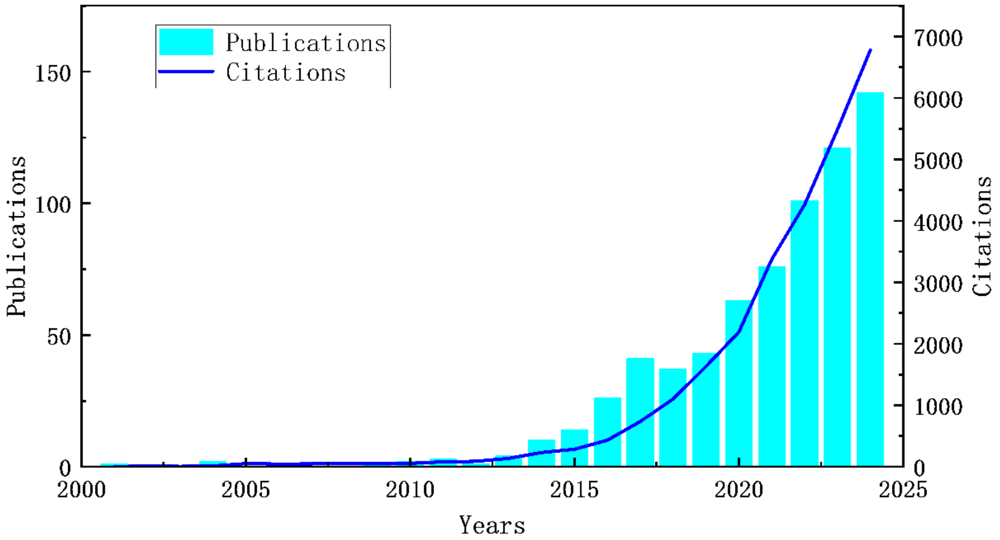

Figure 1. Number of paper publications and citations from March 2001 to March 2025
(Data source: Web of Science)

Prelithiation aims to provide additional lithium source for Lithium-ion batteries to offset irreversible losses such as SEI film formation, expansion of large particle active materials and oxide decomposition, improve ICE, thereby enhancing batteries' initial capacity and energy density. [8,9]. At present, prelithiation technology mainly covers three routes: anode prelithiation, electrolyte prelithiation and cathode prelithiation [10]. Anode prelithiation includes physical, electrochemical and chemical prelithiation, with the characteristics of large prelithiation capacity per unit mass, weak controllability or high side reaction risk [11]; Electrolyte prelithiation supplements lithium source by adding lithium-containing compounds to electrolyte, which is easy to operate but needs to consider the chemical compatibility of additives and electrolyte; The cathode prelithiation is divided into electrochemical lithiation and adding prelithiation additive in cathode. The former can control lithium intercalation but is difficult to scale, while the latter has good compatibility with existing production line, but puts forward higher requirements for chemical properties and cost of additive. In summary, all kinds of prelithiation technologies have their own advantages and challenges, and their continuous optimization is of great significance to promote the industrialization of high-energy-density and long-cycle-life lithium-ion batteries [12]. Cathode prelithiation, in particular, offers a relatively straightforward process with good compatibility to the existing battery manufacturing process, positioning it as a promising candidate for industrial-scale adoption. However, cathode prelithiation still faces challenges such as compatibility between prelithiation agent and cathode material, possible side reactions and product stability.

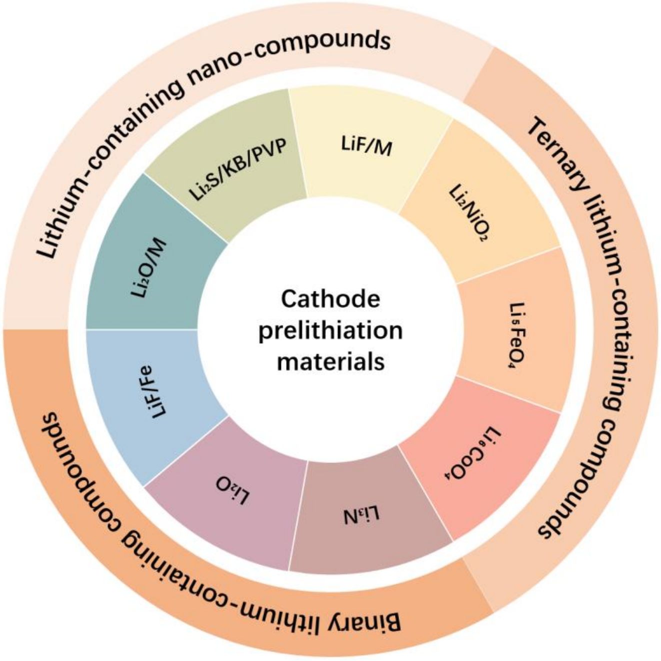

Figure 2. Classification of cathode as additives

Cathode prelithiation additives are categorized into ternary, binary, and nanocomposite types as shown in Figure 2. Ternary additives are primarily utilized in lithium nickel cobalt manganese oxide (NCM) or lithium nickel cobalt aluminum oxide (NCA) systems to enhance material stability and cycle performance through the optimization of lithium source compensation strategies [13]. Binary prelithiation focuses on binary material systems like nickel lithium permanganate, aiming to explore more efficient prelithiation methods for minimizing side reactions [14]. Nanometer prelithiation technology employs nanocomposite materials to boost reactivity and uniformity, thereby enhancing prelithiation effectiveness and material stability [15]. The research and refinement of these technologies are critical for advancing high-performance Lithium-ion batteries.

This paper examines prelithiation technology, including its mechanism, lithium release behavior, and performance variations among ternary, binary, and nanocomposite materials at both the mechanism and material levels. It also evaluates the potential of prelithiation technology within the current industrialization trajectory of Lithium-ion batteries from an engineering application standpoint. By offering a theoretical foundation and practical solution to address the issue of irreversible lithium depletion in high-energy-density batteries, this paper aims to promotes the industrial adoption of high-energy-density batteries

2. Mechanism and Application of Cathode Prelithiation

Cathode prelithiation technology plays a significant role in enhancing the efficiency of Lithium-ion batteries by incorporating extra lithium sources into cathode materials. This compensates for lithium depletion during initial cycles. The process involves the liberation of Lithium-ions from lithium compounds within cathode materials under specific conditions. These ions are then intercalated into negative electrodes to supplement active lithium [16]. While this technology holds promise for enhancing battery energy density and lifespan, it encounters obstacles related to the stability of prelithiated additives and decomposition voltage.

2.1. Ternary Lithium Compounds

Ternary lithium compounds, exemplified by lithium salts of transition metal oxides like Li_2NiO_2, Li_5FeO_4, and Li_6CoO_4, are denoted by the general formula $Li_aM_bO_c$, where M is a transition metal element. A key advantage of these materials is their high theoretical specific capacity, stemming from the incorporation of multiple lithium-ions, as demonstrated by Li_5FeO_4's 867 $mAh\cdot g^{-1}$. They exhibit chemical stability and can release active Lithium-ions within the operational voltage range to compensate for Lithium-ion depletion caused by SEI film formation during the initial charging cycle [17]. However, certain ternary lithium compounds display inadequate air stability, for instance, Li_2NiO_2 is susceptible to air instability and electrolyte reactions. Li_2NiO_2 undergoes Lithium-ion desorption during charging, leading to enhanced lithium content in the anode and improved the ICE of the battery [18]. Subsequently, these additional Lithium-ions partake in the discharge process, releasing energy and enhancing the overall battery performance.

During battery charging, ternary lithium compounds act as prelithiation additives, Lithium-ions in their crystal structure will be detached, migrated to the surface of the anode through the electrolyte, and embedded in the anode material, thus offsetting the lithium consumed during the formation of the SEI film at the anode [19]. Ternary lithium compounds exhibit different application effects in different electrode material systems. Li_6CoO_4 as additive in lithium cobaltate system can effectively improve the initial discharge capacity of batteries. Studies have shown that adding a proper amount of Li_2CoO_6 to $LiCoO_4$ cathode elevates the initial discharge capacity from 77 $mAh\cdot g^{-1}$ to 133 $mAh\cdot g^{-1}$, representing a 72.7% enhancement [20]. In the lithium iron phosphate system, the addition of Li_5FeO_4 can significantly improve the energy density and cycle life of the battery. When Li_5FeO_4 was added to the electrode, the first charge specific capacity of the full battery increased by 11%, the first cycle coulomb efficiency was 67.8%, and the capacity retention rate increased significantly from 90.94% to 98.92% after 50 cycles.

To improve the prelithiation efficiency and cycle stability of ternary systems, researchers have adopted various methods. On the one hand, the crystal structure and properties of materials can be improved by optimizing the preparation process, such as controlling the synthesis temperature and adjusting the ratio of raw materials. When Li_5FeO_4 is synthesized, the material with good crystallinity and excellent properties can be obtained by accurately controlling the temperature and time of high temperature calcination. On the other hand, ternary lithium compounds are surface-modified or compounded with other materials. For example, coating Al_2O_2 thin layer on the surface of Li_2NiO_3 particles makes it a cathode additive with high air stability, which not only improves the first charge specific capacity, but also enhances the stability of the material in air, thus improving the cycle stability of the battery.

The preparation of ternary lithium compounds faces many challenges. The synthesis conditions of some compounds are harsh, and the requirements on temperature and atmosphere are strict. For example, the synthesis of Li_6CoO_4 requires high temperature solid state reaction, and the selection and storage conditions of cobalt source are harsh. CoO is easy to deliquesce and oxygen denaturation, which has potential safety hazards [21]. In addition, the preparation process is often energy-intensive and costly, limiting its large-scale production. From the point of view of industrialization, the poor air stability of ternary lithium compounds is a key problem, which is easy to react with atmospheric moisture and carbon dioxide during manufacturing, storage, and operation, compromising product quality and electrochemical performance.. At the same time, it is difficult to achieve a high degree of consistency and controllability in the current preparation process, resulting in uneven product quality. Addressing these challenges is crucial to enable the broader utilization of ternary lithium compounds within lithium-ion battery technology.

2.2. Binary Lithium Compounds

Binary lithium compounds, comprising Li and nonmetals like N, O, S, and F, are pivotal for prelithiation in lithium batteries due to their high lithium content, controlled reactivity, and compatibility with electrodes. Compounds like LiN, Li_2O, Li_2S play a crucial role in compensating for capacity loss from positive lithium depletion due to SEI formation by releasing active lithium early in charge-discharge cycles, thereby enhancing the ICE and energy density [22]. Nonetheless, challenges persist in their practical application, including side effects, air sensitivity, and uneven dispersion [23]. Recent advancements in material synthesis, nanostructural design, and process optimization have notably enhanced the stability and lithium release efficiency of binary lithium compounds, facilitating their transition from academic research to industrial applications.

Some binary lithium compounds have high stability in a specific atmosphere, so they can match the working potential of the corresponding positive and negative electrodes to achieve accurate lithium supplementation, which is more suitable for large-scale production and storage processing. However, since the lithium supplementation process of binary lithium compounds often decomposes to produce gases, etc., they often need to be further modified and processed. The basic characteristics of common binary lithium compounds are shown in Table 1.

Table 1 Comparison of the characteristics of typical binary lithium compounds

Compounds	Lithium Content (wt%)	Theoretical Capacity ($mAh \cdot g^{-1}$)	Decomposition Voltage (V vs. Li^+/Li)	Air Stability	Applicable Battery System
Li_3N	59.7	2309	0.4–0.6	Poor	NCM ‖ C NCA ‖ C
Li_3P	30.3	1553	0.5–1.0	Very Poor	LCO ‖ C NCM ‖ C
Li_2O	46.4	897	1.0–1.5	Excellent	LCO ‖ C LNMO ‖ C
Li_2O_2	40.2	1168	2.5–3.0	Poor	LCO ‖ C LNMO ‖ C
Li_2S	30.2	1166	1.4–1.8	Medium	LCO ‖ C NCM ‖ C
Li_2Se	15.0	577	1.5–2.2	Poor	LCO ‖ C NCM ‖ C
LiF	26.8	722	3.8–4.5	Excellent	LNMO ‖ C

The positive binary lithium compound releases active lithium through decomposition reaction to offset lithium loss caused by anode SEI formation during the first-cycle operation and cycle of the positive electrode material, thus boosting the battery's energy density and cycling stability. Generally, binary prelithiation additive can release Lithium-ions through electrochemical decomposition at low voltage. In the first week of charging, positive additive decomposes at voltage higher than delithiation potential of positive electrode material, releases Lithium-ions and embeds into positive electrode host material, directly supplementing lithium source. Specific reaction formula is as follows [24-31] :

$$2Li_3N \rightarrow 6Li^+ + N_2 \uparrow +6e^- \tag{1}$$

$$Li_3P \rightarrow 3Li^+ + P + 6e^- \tag{2}$$

$$2Li_2O \rightarrow 4Li^+ + O_2 \uparrow +4e^- \tag{3}$$

$$Li_2O_2 \rightarrow 2Li^+ + O_2 \uparrow +2e^- \tag{4}$$

$$Li_2Se \rightarrow 2Li^+ + Se + 2e^- \tag{5}$$

$$Li_2S \rightarrow 2Li^+ + S + 2e^- \tag{6}$$

$$xLiF + M \rightarrow xLi^+ + MF_x + xe^- \tag{7}$$

$$2LiF + x + (1 - x)Fe \rightarrow 2Li^+ + Fe_{1-x}Cu_xF_2 + 2e^- \tag{8}$$

Common binary lithium compounds are relatively active and usually need to be doped with graphite or metal to extend storage life and ensure storage quality. LiF is usually combined with transition metals to form LiF/M (M is the corresponding transition metal such as Fe, Co, Mn, etc.) composite materials. When such additives participate in prelithiation, the reaction as shown in Formula (7) mainly occurs.

For binary lithium compounds with high decomposition voltage such as LiF, catalytic auxiliary decomposition strategy can be adopted to accelerate lithium release in the prelithiation process. The specific action mechanism of LiF/Fe/Cu ternary composite prepared by Zhao Y is shown in Formula (8), which mainly uses bimetallic nano-domains to catalyze the decomposition of LiF to release Lithium-ions, which significantly improves the diffusion coefficient of Lithium-ions and accelerates the decomposition kinetics of LiF.

The prelithiation mechanism of binary lithium compounds is evolving from single decomposition to multi-mechanism coordination. The prelithiation efficiency and cycle stability can be considered by various modification strategies.

Park K et al. [32] reduced the particle size by grinding and exposed the fresh surface, and at the same time, Li_3N was mechanically pressed on the $LiCoO_2$ electrode surface by surface coating to mitigate the insulating effect of Li_3N on electrode conductivity. Wang X et al. [33] reduced graphene oxide (rGO) with high-purity Li_3P, and uniformly dispersed Li_3P@rGO composite material on the electrode surface by surface coating, which improved its conductivity and interface stability, avoided volume expansion and reduced interface impedance. Chen Y et al. [34] mixed micron-level Li_2O with high-energy composite cathode materials such as Li_2MnO_3-$LiMnO_2$ to release Lithium-ions under high voltage, which effectively compensated for the irreversible capacity loss of the first circle of alloy cathode such as SiO-SnCoC. Zhang L et al. [35] embedded Li_2O_2 into PMMA nanofibers by electrospinning technology to form stable P-Li2O2 composite materials, so that the energy density and cycle life of the NCM-P-Li_2O_2||Si-Gra full cell are significantly improved. As shown in Figure 3, The battery capacity retention rate can reach 81.3% after 50 cycles. Ding R et al. [36] compounded Li_2S with super activated carbon (SAC), and uniformly coated Li_2S/SAC slurry on the surface of LFP cathode by spraying process. The full cell test showed that the discharge capacity increased by 11.7%. Fu Y et al. [37] prepared Li_2Se with high crystallinity by chemical lithiation of selenium powder and further high temperature sintering, which significantly improved the stability of Li_2Se. Liu Y et al. [38] prepared LiF/Fe additive by ball milling LiF and Fe, and coated it on NCM811 cathode with double-layer structure to form stable solid-liquid interface and complete cathode structure, thus improving the overall electrochemical performance.

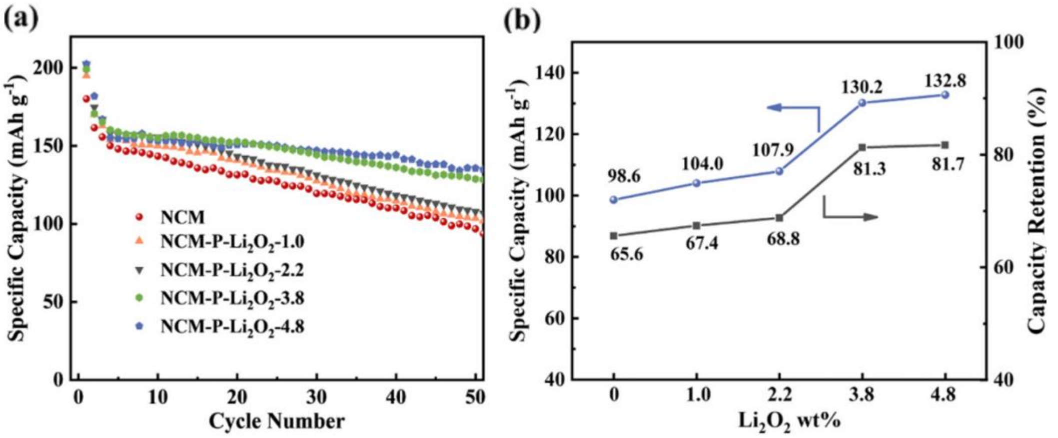

Figure 3. (a) Cycling performances and (b) 50th-cycle capacity retention (vs. 5th cycle) of NCM-P-Li_2O_2||Si–graphite and NCM||Si–graphite full cells.

Binary lithium compounds have demonstrated significant potential as prelithiation additives in Lithium-ion battery technology. However, their widespread industrial implementation encounters several challenges. Firstly, these compounds exhibit low chemical stability, particularly in air and humid conditions, necessitating their handling and utilization in inert atmospheres, thereby incurring additional storage expenses. Secondly, the electrochemical properties of binary lithium compounds are irreversible, leading to the generation of other less conductive compounds during Lithium-ion release for prelithiation, consequently diminishing battery discharge efficiency. Lastly, the complexity of the preparation processes for high-purity binary lithium compounds involves specialized techniques which result in high equipment and preparation costs. Future research directions may include the exploration of novel coating materials and inert atmosphere packaging technologies, the design of multi-stage structures, the incorporation of doping or composite conductive frameworks, the advancement of large-scale preparation methodologies, as well as the establishment of an industrial evaluation framework to enhance the feasibility and cost-effectiveness of large-scale binary lithium compound applications in Lithium-ion batteries.

2.3. Nano-lithium Compounds

Nanometer-scale lithium compounds, a category of prelithiated additives boasting high theoretical capacity, are crafted using nanotechnology, with particle sizes spanning 1 to 100 nm. This group primarily encompasses nano metal oxides, nano-lithium salts, and composite nanomaterials [39]. Nano-lithium compounds play a crucial role in precisely regulating the initial lithium content of the cathode through direct prelithiation or on-site generation of an active lithium source. Furthermore, leveraging their superior performance stemming from their minute particle size, their stability can be fine-tuned through surface coating, doping, or recombination techniques. Presently, these compounds have emerged as pivotal materials in the advancement of lithium batteries characterized by high energy density and prolonged cycle life.

The theoretical capacity of nano-Li_2O/M [40] (M= Co,Fe) composites can reach 900 mAh·g^{-1} while it has good stability. It has been found through experiments that, as shown in Figure 4(a), after adding 4.8% and 8% Li_2O/Co to the $LiFePO_4$ electrode, the capacity increases by 11% and 14%, respectively. Li_2O/Fe has good compatibility with adhesive and electrolyte, which can concurrently elevate specific energy density (8~13%) and volumetric energy density (11~14%) in LFP/graphite full cells, alongside enhanced cycling stability.The core-shell Li_2S/KB/PVP [41] maintains a capacity retention of nearly 100% after 100 cycles in carbonate electrolytes (Figure 4b), and the specific energy of LFP/graphite full cell can be increased by 5.6%. The decomposition voltage of Li_2S/M [42] (M=Co, Fe) nanocomposites is lower than the cut-off potential of high voltage cathode. Figure 4c shows the charge capacity of the LFP electrode added with 4.8% Li_2S/Co increased by 42 mAh·g^{-1}. Nanometer LiF/M [43] (M=Fe,Co) composites also have high theoretical capacity and can exist stably in air and electrolyte. As validated in Figure 4d, Incorporating LiF/M additives in LFP cathodes elevates first-charge capacity from 164 mAh·g^{-1} to 197 mAh·g^{-1}. Du J et al. [44] prepared nano-scale Fe-LiF-Li_2O composite material, which can reach 550 mAh·g^{-1} in the first circle of electrode when used as positive electrode additive. As shown in Figure 4e, the decomposition voltage of Fe-LiF-Li_2O nanocomposite is 3.1~4.1V, which is lower than that of pure LiF matrix material; Figure 4f shows Fe-LiF-Li_2O nanocomposite can still retain 65% capacity after 48 hours of exposure in air, which has better environmental stability than pure Li_2O matrix composite. The preparation process of nanocomposites based on inverse conversion reaction is relatively simple, usually by mixing M_xN_y and molten Li in argon atmosphere by in-situ solid state method, and the properties are significantly improved compared with the original materials.

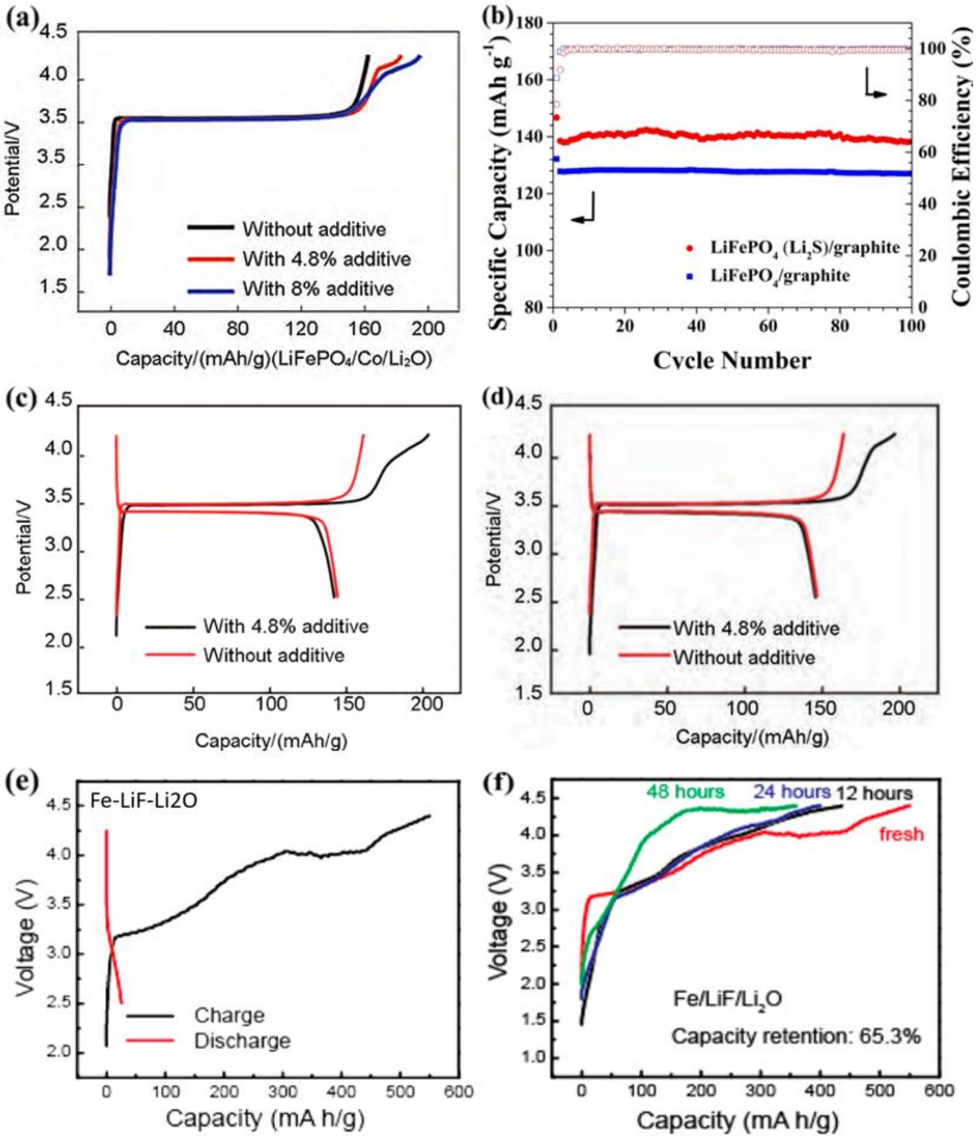

Figure 4. (a) LFP cathodes with 4.8 % and 8 % Li_2O/Co additives. (b) The cycle performance of the $LiFePO_4$ (Li_2S)/graphite and $LiFePO_4$/graphite full cells. The rate for the initial cycle was 0.05C, and that for the following cycles was 0.2C.(c) LFP cathode with 4.8% Li_2S/Co.(d) LFP cathode with 4.8% LiF/Co. (e) Initial charge-discharge profiles of Fe-LiF-Li_2O at a current density of 25 mAh·g^{-1}. (f) Initial charge curve of the Fe-LiF-Li_2O nanocomposite after storage in the ambient atmosphere (20% humidity) for different times. The current density is 25 mAh·g^{-1}.

LCNO [45] replaces Ni^{2+} in Li_2NiO_2 with Cu^{2+} to form solid solution junction, which will irreversibly transform into layered structure after first charging. As shown in Figure 5a, NiO content decreases from 12.57% to 0.86%, indicating that LCNO can effectively eliminate NiO impurity phase in LNO electrode. Electrochemical testing (Figure 5b) demonstrates LCNO delivers a first-charge capacity of 465 mAh·g^{-1}, significantly surpassing LNO's 235.8 mAh·g^{-1}. After battery cycling, the fluoride (LiF) content in the CEI layer on the cathode surface decreases, and the proportion of Li_2CO_3 increases. This transition inhibits electrolyte decomposition and markedly enhances cycling stability.

Li_2S-PAN developed by Zhang H et al. [46] forms nanoscale spherical clusters through sulfide and polyacrylonitrile (PAN) recombination, in which sulfur elements are uniformly embedded in carbon-based frameworks through C-S bonds. Li_2S-PAN releases 695 mAh·g^{-1} active Lithium-ions during the first charge process, and the sulfide formation is less, and the shuttle effect is not strong. Figure 5c shows Li_2S-PAN has good cycle stability while effectively improving the reversible capacity of the electrode in the $LiFePO_4$/SiC half cell, and the capacity retention rate is still as high as 82.8% after 300 charge and discharge cycles.

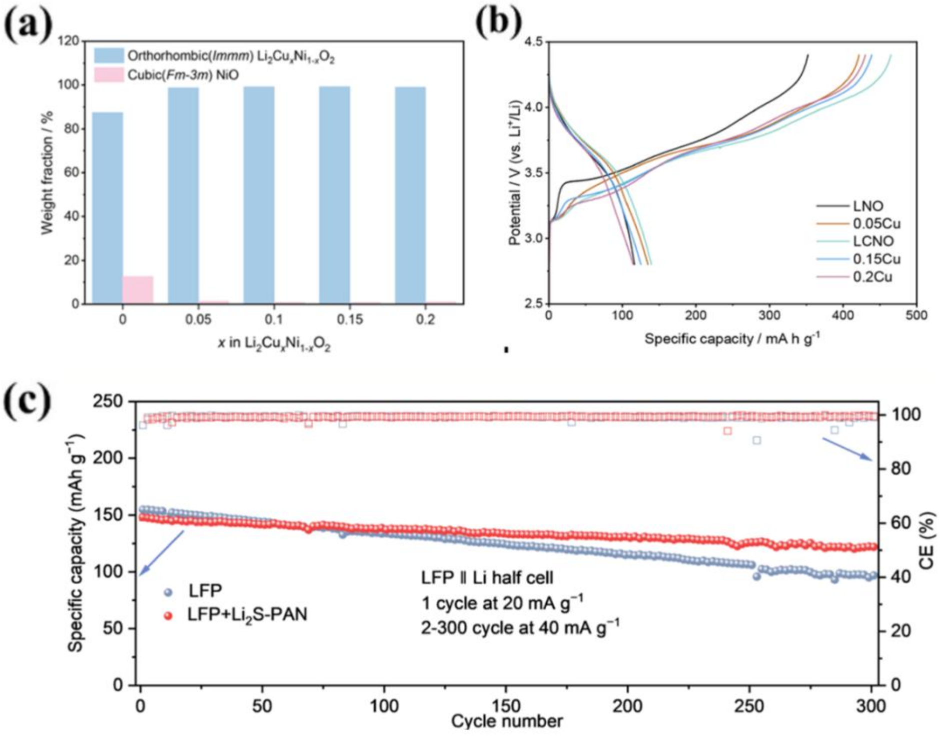

Figure 5. (a) Phase weight fraction in $Li_2Cu_xNi_{1-x}O_2$. (b) Initial charge–discharge voltage profiles of LNO, LCNO, and $Li_2Cu_xNi_{1-x}O_2$ (x= 0.05, 0.15, and 0.2) electrodes. (c) Cycling performance of the LFP||Li half cell with or without Li_2S-PAN at 40 $mAh\cdot g^{-1}$.

Currently, research in nano-lithium compounds is primarily centered on high-capacity nanocomposite materials and lithium-rich materials. Through nanostructure engineering, minute additions can effectively offset lithium depletion in the cathode, suppress transition metal dissolution and uphold structural integrity. Nevertheless, the reaction mechanisms of certain additives remain ambiguous, and certain materials pose risks of gas generation. Consequently, future research should prioritize the optimization of material design, emphasizing substantial compensation capacity, minimal side reactions, and practicality for large-scale production.

3. Summary and Prospect

From what was discussing above, this review systematically synthesizes advancements in cathode prelithiation for lithium-ion batteries, a technology centered on offsetting initial-cycle irreversible lithium loss to enhance first-cycle Coulombic efficiency, energy density, and cycling stability. Current technology relies primarily on ternary, binary and nanocomposite lithium additives, each of which has its own characteristics in key properties. Large-scale adoption of prelithiation additives faces three key limitations: the side reaction in the prelithiation process, the environmental instability of the additive and the high production cost. Future research can focus on the following aspects to break through the bottleneck: deeply reveal the reaction mechanism and by-product evolution law, guide the development of new additives with high lithium content, controllable release and excellent stability; innovate material design, accurately control the reaction path to inhibit side reactions and improve stability by constructing core-shell structure, doping conductive skeleton, optimizing interface and other schemes; Develop low-energy, high-consistency, large-scale preparation and integration processes to reduce production costs and ensure compatibility with existing production lines; establish quantitative models of prelithiation degree, by-products, electrode evolution and long-cycle performance to provide theoretical support for optimization. Through collaborative innovation of materials and processes, this technology will transition from laboratory validation to industrial deployment, providing key support for the development of the next-generation lithium-ion batteries with enhanced energy density and extended cycle life.

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