



Review

Suppressing dendritic metallic Li formation on graphite anode under battery fast charging

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ABSTRACT

Lithium-ion batteries (LIBs) with fast-charging capability are essential for enhancing consumer experience and accelerating the global market adoption of electric vehicles. However, achieving fast-charging capability without compromising energy density, cycling lifespan, and safety of LIBs remains a significant challenge due to the formation of dendritic Li metal on graphite anode under fast charging condition. In view of this, the fundamentals for the dendritic metallic Li formation and the strategies for suppressing metallic Li plating based on analyzing the entire Li⁺ transport pathway at the anode including electrolyte, pore structure of electrode, and surface and bulk of materials are summarized and discussed in this review. Besides, we highlight the importance of designing thick electrodes with fast Li⁺ transport kinetics and comprehensively understanding the interaction between solid electrolyte interphase (SEI) and Li⁺ migration in order to avoid the formation of dendritic Li metal in practical fast-charging batteries. Finally, the regulation of Li metal plating with plane morphology, instead of dendritic structure, on the surface of graphite electrode under fast-charging condition is analyzed as a future direction to achieve higher energy density of batteries without safety concerns.

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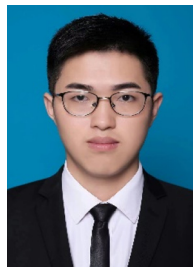
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1. Introduction

The global climate change and energy resources crisis have made it imperative to develop renewable energy sources (e.g., wind, biomass, and solar) and promote the electrification of transportation [1]. Lithium-ion batteries (LIBs) have been widely considered as a dominating energy storage technology for the storing renewable energy and powering sources for electric vehicles (EVs, alternatives of conventional vehicles) [2]. Despite the high energy density and low cost of the-state-of-the-art LIBs with graphite anode, their inferior fast charging capability cannot meet the increasing demand for various applications [3]. In particular, the fast-charging capability of LIBs is a major concern for widespread consumer adoption of EVs, and achieving superior fast-charging capability has become a significant goal for both the research community and the industry. Typically, the United States Department of Energy (US DOE) set a goal in 2018 for EV batteries, aiming to achieve 80% state of charge (SOC) within 15 min and a discharge specific energy of $>200 \text{ Wh kg}^{-1}$, referred to as extreme fast charging (XFC) [4].

To meet the XFC goal, it is necessary to achieve reasonably high energy density and short charging time simultaneously for LIBs. Although numerous works on fast charging have been conducted and have reported promising data, most of these researches have focused on evaluating single battery components and structures, such as anode materials [5–7], electrolytes [8,9], and electrode structures [10], rather than practicality for industrial application. For instance, while 10 C charging with 84% SOC has been achieved for graphite anodes, the low mass loading ($<3 \text{ mg cm}^{-2}$) results in a

significantly lower energy density for the batteries [11]. To date, only a few literatures have reported the cell-level energy density achieved under fast-charging condition [12,13]. Three strategies were widely adopted to improve the fast-charging capability of state-of-the-art LIBs, including electrolyte regulation [14,15], electrode structure design [16,17], and working temperature modulation [15,18–20]. One of the most impressive examples was the combination of asymmetric temperature modulation (ATM) method with a dual-salt electrolyte and larger porosity graphite anode. The cell delivered a specific energy of 199 Wh kg^{-1} within 13 min of charging time based on 50 Ah-LiNi_{0.8}Mn_{0.1}Co_{0.1}O₂ (NMC811)||graphite cell [15].

To enhance the energy density of LIBs under fast charging, the most direct approach is to increase the mass loading of electrodes and areal capacity. Regrettably, when utilizing standard constant current methods for fast charging conventional LIBs within 15 min, it is likely to result in substantial capacity decay of the batteries and the formation of lithium plating on the graphite anode [21]. With mass loading of 7.3, 12.5, and 15.9 mg cm^{-2} for graphite anode (with thickness of 58, 97, and $127 \mu\text{m}$), the LiNi_{0.6}Mn_{0.2}Co_{0.2}O₂ (NMC622)||graphite pouch cells delivered discharge capacities of ~ 2.0 , ~ 3.0 , and $\sim 4.2 \text{ mA h cm}^{-2}$ with charging rate of C/3. However, when increasing the charging rate to 1 C, the discharge capacities of the cells (~ 1.9 , ~ 2.8 , and $\sim 2.4 \text{ mA h cm}^{-2}$ at the 335th cycle) decreased significantly. Notably, a significantly faster capacity decay was observed at 1 C in comparison to that at C/3, as shown in Fig. 1(b). The rapid capacity decay observed in electrodes with high mass loading is attributed to the formation of metallic lithium dendrites during fast charging. Fig. 1(a) illustrates that a

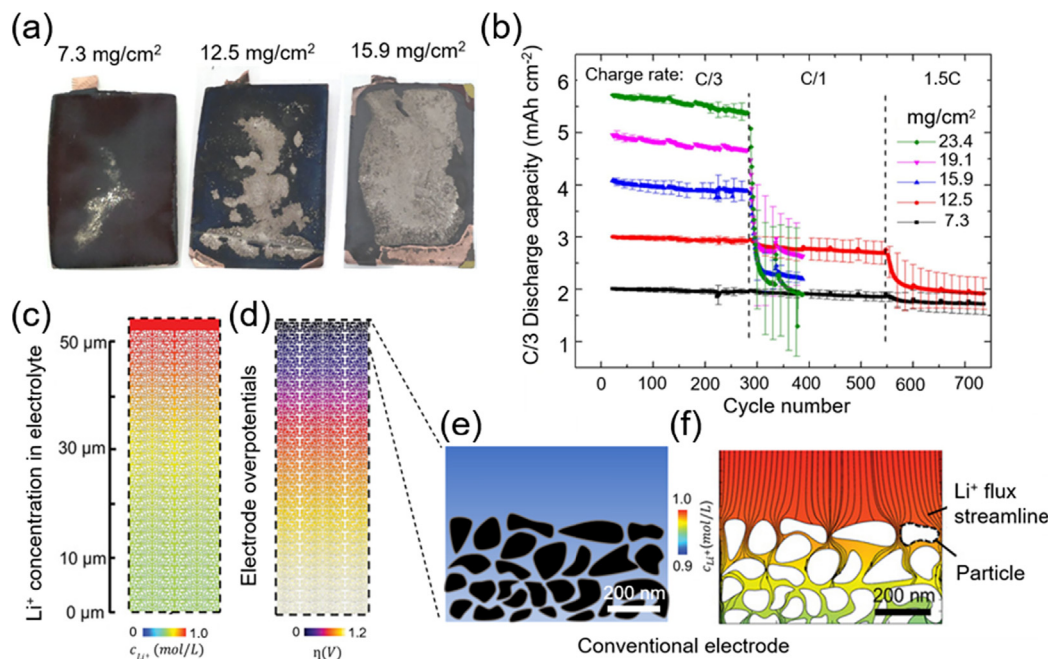


Fig. 1. (a) Images of graphite electrode after aging in NMC622||graphite pouch cells. Metallic Lithium deposits can be observed on the surface of electrode, and their amount increases with mass loading (mg cm^{-2}) as depicted from left to right. (b) NMC622||graphite pouch cells with different mass loading as a function of charge rate. Reproduced with permission from Ref. [21]. Copyright 2017, Elsevier. The simulation diagrams of (c) Li^+ concentration distribution in electrolyte and (d) electrode overpotentials versus depth in electrode. (e) The schematic of graphite electrode/electrolyte interface. (f) The simulation diagram of Li^+ flux in electrolyte at electrode/electrolyte interface. Reproduced with permission from Ref. [22]. Copyright 2022, Wiley-VCH.

thick layer of metallic Li deposition covered almost the entire surface of the graphite anode with a high mass loading of 15.9 mg cm^{-2} after voltage holding for 24 h at 3.75 V. In contrast, the graphite anode with a lower mass loading of 7.3 mg cm^{-2} showed a much smaller area ratio of metallic Li deposits, less than 1/5 of the electrode surface area [21]. The intrinsic low Li^+ intercalation potential of graphite ($\sim 0.1 \text{ V}$ vs. Li^+/Li , close to that of metallic Li plating) and the slow Li^+ intercalation kinetics are the reasons for the occurrence of metallic Li plating and affect the fast-charging performance. Obviously, a thick electrode causes long Li^+ diffusion distance and a large Li^+ concentration gradient. As shown in Fig. 1(c–f), it is emphasized that the Li^+ concentration gradients significantly manifest its effect on overpotential for thick electrode under fast charging, and the depleting of Li^+ concentration within the pores of the thick electrode occurs faster compared to the thin electrode. These factors together cause severe electrochemical Li plating behavior in high-mass-loading electrodes [22]. The high chemical reactivity to corrosive with electrolytes and irreversible metallic Li stripping cause a high accumulated active Li loss, leading to unsatisfactory cycling behavior [23]. Dendritic metallic Li plating has the potential to penetrate the separator, thereby inducing internal short circuits within the batteries and escalating safety concerns [24,25]. More specifically, these internal short circuits can stimulate the spontaneous release of electrochemical energy stored within the materials, leading to substantial heat generation. This internal accumulation of heat can subsequently trigger a cascade of violent side reactions (e.g., electrolyte combustion), culminating in thermal runaway of the cells. In the most severe scenarios, this uncontrollable thermal runaway can propagate throughout the entire battery pack and even to the vehicle itself, resulting in catastrophic fires and explosions. Consequently, the achievement of the XFC goal for current LIBs continues to pose a significant challenge [26].

In summary, dendritic metallic Li plating on graphite anodes is one of the most crucial obstacles for practical fast-charging LIBs.

This review aims to thoroughly investigate the dendritic metallic Li plating on graphite anode under fast charging, with specific focuses on its underlying mechanism, current studies on suppressing the formation of Li dendrites, and perspective research directions. Kinetics constraints play a vital role in bridging the thermodynamics gap (potential difference) between metallic Li plating and Li^+ intercalation in graphite anodes. Therefore, the mechanism of Li metal plating on graphite electrode is introduced from both kinetics and thermodynamics aspects. Subsequently, this review analyzes the current status and summarizes strategies for mitigating dendritic metallic Li plating on the basis of various parameters that affect the Li^+ migration and electrochemical reaction kinetics in fast-charging graphite anode, including both materials and electrode-level factors. Finally, it is emphasized that uniform metallic Li plating on graphite anode with high reversibility can be an ideal approach for further rechargeable Li-based batteries with both good fast-charging capability and high energy density. When graphite is effectively utilized as both the active material and hosts/substrates for reversible metallic Li plating, not only the safety concerns of dendritic metallic Li products could be addressed, but also a further increase in the energy density of batteries could be achieved. In addition to solely modulating material, electrodes, and electrolytes, the importance of synergistic interactions across multiple aspects is emphasized to potentially yield better results and achieve fast-charging Li-based batteries in this review, which makes this review different from the previous publications.

2. Analysis of metallic Li plating on graphite

2.1. Analyzing metallic Li plating on graphite

Fig. 2(a) illustrates the Li^+ transport in a LIB. During charging process, Li^+ ions are extracted from the cathode, transferred

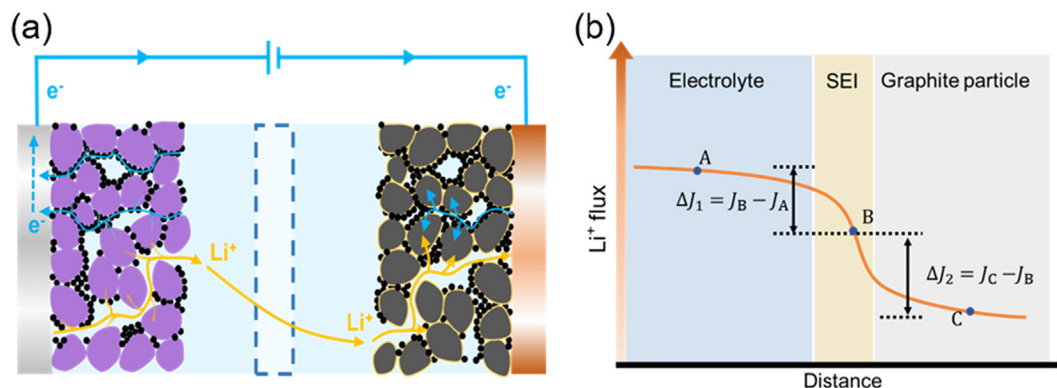


Fig. 2. (a) Illustration of the charge process of a rechargeable LIB consisting of a graphite anode, electrolyte, and transition metal oxides cathode. (b) The schematic of the flux distribution of Li^+ across the graphite/electrolyte interphase. The orange curves represent the distribution of Li^+ flux under fast charging rates (solid lines). Point **A**, **B**, and **C** refer to the typical position in electrolyte, SEI, and graphite particles, respectively. ΔJ_1 indicates the difference flux between electrolyte and SEI and ΔJ_2 indicates the difference flux between SEI and graphite materials.

through the electrolyte, and intercalate into the graphite anode. Simultaneously, electrons migrate via the external circuit in the same direction. The inverse charge transfer process occurs during discharge process. Specifically, graphite possesses a two-dimensional layered structure with a layer spacing of 0.335 nm, and these interlayers are connected by Van der Waals forces [27]. During electrochemical charging, Li^+ can intercalate into the graphite interlayer to form Li_xC_6 ($0 \leq x \leq 1$) as a Li-graphite intercalated compounds, delivering a stoichiometric theoretical specific capacity of 372 mA h g^{-1} (based on LiC_6) [28]. From the perspective of thermodynamics, the potential of graphite electrode of LIBs can be determined by the Nernst equation (Eq. (1)).

$$E = E^0 + \frac{RT}{nF} \ln \left(\frac{[\text{Li}^+]}{[\text{Li}^0]} \right) \quad (1)$$

Where E^0 is the standard electrode potentials, R is the universal gas constant, T is absolute temperature, n is the valence of the Li^+ , and F is the Faraday constant. Ideally, the reaction potential for Li^+ intercalation into graphite (E_{Gr} , 0.1 V vs. Li^+/Li) should be higher than that for metallic Li deposition (E_{Li} , 0 V vs. Li^+/Li). Therefore, during charging process (Li^+ intercalation) of graphite anode, metallic Li plating will not occur due to the hysteretic reduction of Li^+ .

From the viewpoint of kinetics, as many researchers reported, the solid diffusion of Li^+ in graphite interlayers dominates the reaction kinetics [29]. In details, the inferior solid diffusion of Li^+ in graphite interlayers compared to that in electrolyte and consequent transition of different staging structures cause the accumulation of Li^+ ions and a concentration gradient at the graphite/electrolyte interface. The local Li^+ concentration difference further triggers a deviation between the potential of graphite electrode (E_{Gr}) and its equilibrium potential (E_{Gr}^0) on the graphite surface, which is called electrochemical polarization. The overpotential can make the local potential of electrode surface below 0 V (vs. Li^+/Li) and cause metallic Li plating.

Nevertheless, recently, Weng et al. proposed that the Li^+ solid diffusion coefficient was estimated to be large enough to enable the fast charging at a rate of 6 C if the particle size is less than $10 \mu\text{m}$ [30]. Meanwhile, some researchers considered that the desolvation of Li^+ at the interface is the mainly limiting factor at fast charging [31,32]. That is to say, due to the requirement of faster Li^+ transport under fast-charging condition, all the transport process of Li^+ migration in the graphite anode (e.g., interfacial transport, Li^+ transport in electrolyte) can be the crucial limiting factors.

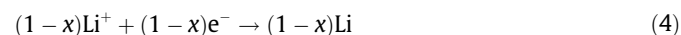
After conducting the analysis mentioned above, it is worth considering that the concentration polarization is caused by the mismatch of diffusion kinetics in the electrolyte, graphite materials,

and interphase at fast charging. According to Fick's law, the Li^+ flux indicates the amount of diffusing Li^+ ions per unit area per unit time. Thus, the Li^+ flux can provide a clearer representation for the Li^+ diffusion kinetics than solely relying on the concentration gradient. Fast charging requires a larger Li^+ flux, which puts forward exacting requirements for Li^+ transport dynamics. In detail, for typical position **A** in electrolyte and position **B** in solid electrolyte interphase (SEI), there may be a flux difference (ΔJ_1) between **A** and **B**, which refers to a mismatch in Li^+ diffusion kinetics between electrolyte and SEI, as shown in Fig. 2(b). Meanwhile, larger concentration polarization tends to occur at the interface where the flux difference is larger. Although the rate-limiting factors are still controversial and require further investigation, it can be confirmed that metallic Li plating at graphite/electrolyte interface is more likely to occur under fast charging conditions because of the electrochemical and concentration polarization. For a thick and compact graphite electrode, Li^+ diffusion throughout the entire electrode should be taken into consideration due to the overlong diffusion path, rather than just focusing on the graphite/electrolyte interface. More specific analysis is displayed in Section 3.1.

In conclusion, polarization on graphite anode always exists, causing the overpotential for either the Li^+ intercalation into graphite (η_{Gr}) or other parasitic reaction (e.g., metallic Li plating η_{Li}) [33]. In detail, under harsh fast-charging conditions, the high concentration gradient of Li^+ results in a larger overpotential (η_{Gr}) on the graphite surface, and the actual potential of graphite anode decreases to lower than 0 V (vs. Li^+/Li), reaching the thermodynamic conditions for metallic Li plating, as depicted in Eq. (2).

$$E_{\text{Gr}} - \eta_{\text{Gr}} < E_{\text{Li}} - \eta_{\text{Li}} \quad (2)$$

Therefore, during the charging process, metallic Li plating is a parasitic process that occurs alongside with the Li^+ intercalation process. These two processes have a competitive relationship for electrons and Li^+ ions. In this case, the total charging current is divided into intercalation current and plating current, as shown in Eqs. (3) and (4) [33].



Correspondingly, Eq. (3) illustrates the electrochemical intercalation of Li^+ into the graphite anode, which occurs within a potential range of 65–200 mV (vs. Li^+/Li). Eq. (4) demonstrates the electrochemical process of metallic Li plating on the surface of graphite anode. To be specific, the plated metallic Li possesses high reaction activity and causes parasitic reactions with electrolyte,

and significantly, it often could not be completely utilized during the subsequent electrochemical processes. When the current density is low, the rate of metallic Li deposition may be much slower than the rate of fresh SEI formation, and complete coverage of a robust SEI film can be achieved [34]. Deposited metallic Li may further react with the graphite and transform into reversible metallic Li during the subsequent constant voltage charging (small current density) or resting process. The irreversible capacity of batteries mainly originates from the new SEI film formation. Nevertheless, under the fast-charging conditions, the deposited metallic Li is prone to forming the dendrites [35–37], as depicted in the Sand's formula (Eq. (5)).

$$t_{\text{sand}} = \pi D \frac{(z_c c_0 F)^2}{4j^2 (1 - t_c)^2} \quad (5)$$

where D is the Li^+ diffusion coefficient, z_c is the Li^+ charge number (here $z_c = 1$), c_0 is the lithium salt concentration, F is the Faraday constant, j is the current density, t_c is the Li^+ transference number.

Sand's time (t_{sand}) describes the onset of dendritic metallic Li growth, which is inversely proportional to the current density [36]. A short Sand's time indicates the deficiency of Li^+ ions on the surface of the substrate material and a preferential dendritic growth on surface protrusions. Thus, at fast charging, the deposited metallic Li on the surface of graphite is more likely to forming dendritic Li. Besides, compared with the uniform and homogeneous deposition on the surface of Li metal foil, graphite surface as a heterogeneous and uneven substrate is more likely to form Li dendrites. Deposited dendritic metallic Li would further lose electrical contact with the graphite materials after subsequent stripping, referred to as “dead” Li, inhibiting Li^+ intercalation into graphite and causing an irreversible loss of active lithium, which are harmful for Coulombic efficiency and cycling performance. Worst of all, continuous growth of dendritic metallic Li would puncture the separator, triggering a short circuit in the batteries, which increases the risk of thermal runaway and significantly weakens the safety of LIBs, as mentioned in Section 1.

2.2. Monitoring metallic Li plating on graphite

Given the performance degradation and safety risks associated with dendritic metallic Li deposition on graphite anodes, it is vital to devise effective and precise strategies to detect the onset of lithium plating and comprehend Li plating behavior under fast-charging conditions. Broadly, the methods for monitoring Li plating on graphite anodes can be categorized into ex-situ or in-situ techniques.

Ex-situ techniques typically necessitate the disassembly of batteries to perform postmortem characterization of the graphite electrode, employing tools such as optical microscopy, atomic force microscopy (AFM), scanning electron microscopy (SEM), and transmission electron microscopy (TEM) [38]. For instance, SEM is proved to be instrumental in examining the spatial distribution of deposited Li across varying fields of view [39]. Mass spectrometry titration techniques provide exceptionally accurate measurements of the initiation of Li plating [40]. However, it is noteworthy that lithiated graphite (LiC_6) exhibits similar characteristics (e.g., high reaction activity) to metallic Li, necessitating meticulous sample preparation and additional constraints (e.g., cryogenic conditions) to minimize effects on the deposited Li and preserve the typical equilibrium/steady state in the above techniques.

In-situ techniques can accurately reflect the operational state of the batteries and monitor the occurrence and evolution of Li plating in real time. Given that Li plating is a parasitic reaction occurring concurrently with Li^+ intercalation into graphite, in-situ

electrochemical methods (e.g., differential voltage analysis, electrochemical impedance spectroscopy (EIS), and differential open-circuit voltage analysis) for detecting deposited Li have been extensively explored. To enhance the precision of voltage analysis techniques, a three-electrode cell configuration is indispensable. For instance, Xu et al. proposed that the relaxation time constant of the charge-transfer process could serve as a promising indicator of Li plating onset through pulse/relaxation tests [41]. Although electrochemical methods have been proven to be effective in monitoring the onset of Li plating, they are incapable of directly observing dendritic metallic Li. Guo et al. observed dendritic metallic Li growth behavior on the graphite anode surface in commercial LIBs using in-situ optical microscopy [42]. Additionally, in-situ microscopic techniques are frequently employed to directly observe the absence of dendritic metallic Li, with views ranging from an entire electrode surface to fractions of individual electrode particles [43].

Each method possesses a unique set of strengths and limitations, contributing to the elucidation of a segment of the complex Li plating process. The amalgamation of multiple characterization techniques holds the greatest potential in comprehensively revealing the process of Li plating. Therefore, it is crucial to diligently monitor Li plating, particularly by utilizing operando detection methods.

3. Suppressing Li dendrite growth on graphite

The primary cause of Li metal nucleation and growth is the polarization resulting from the significant resistance to Li^+ transport under various conditions [44,45]. Hence, the objective of fast-charging LIBs is to achieve improved kinetics of Li^+ transport in graphite anode, aiming to prevent safety hazards caused by the growth of Li metal dendrites.

The Li^+ migration in the graphite anode can be divided into the following four consecutive steps in Fig. 3 [29,31,46].

- The Li^+ transport in electrolyte within porous graphite electrode.
- The Li^+ desolvation at the SEI.
- The Li^+ transport through the SEI.
- The Li^+ transport within the bulk material.

In general, the Li^+ desolvation is widely acknowledged as the most energy-consuming step from thermodynamic viewpoint [31]. Inadequate Li^+ transport behavior within a thick graphite anode with high tortuosity has often emerged as a limiting factor that cannot be overlooked easily for practical fast-charging LIBs with high energy density [47]. The composition and structure of SEI, as well as the structure of bulk material, also play significant effects on the Li^+ migration behavior and fast-charging capability [48]. Therefore, all four steps impact Li^+ transport and the rate-determining step is closely related to materials/electrode structure and internal and external working conditions of the battery. Therefore, it cannot be straightforwardly predetermined. Currently, numerous studies have focused on optimizing Li^+ transport behavior in order to hinder the growth of Li dendrites during the cycling, especially under fast-charging conditions.

3.1. Li^+ transport in electrolyte within the graphite electrode

Adequate attention should be dedicated to facilitating charge transfer within high-loading electrodes in order to pursue fast-charging LIBs with high energy density. Ideally, a uniform distribution of Li^+ throughout the thickness of the graphite electrode should be achieved for consistent intercalation (Fig. 4a). However,

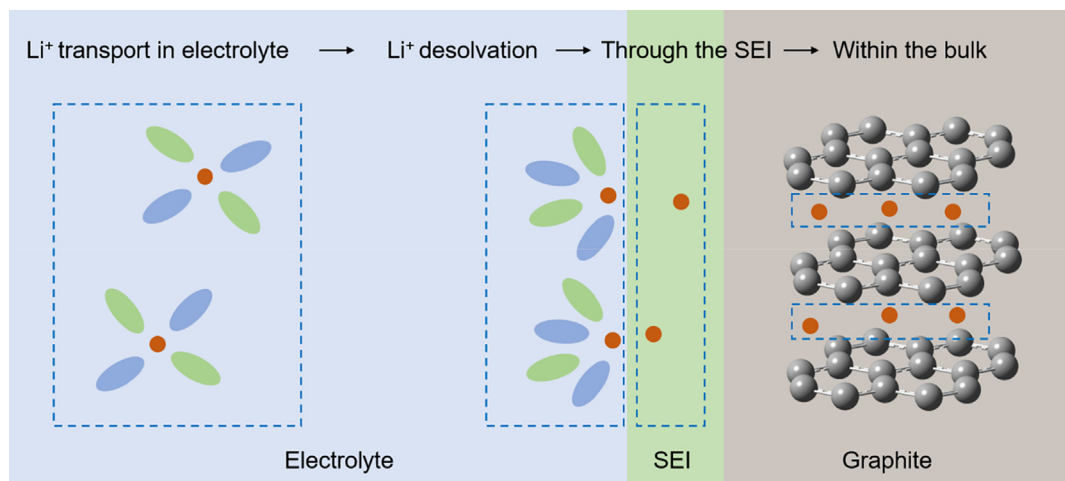


Fig. 3. Schematic of Li^+ transport at the graphite anode.

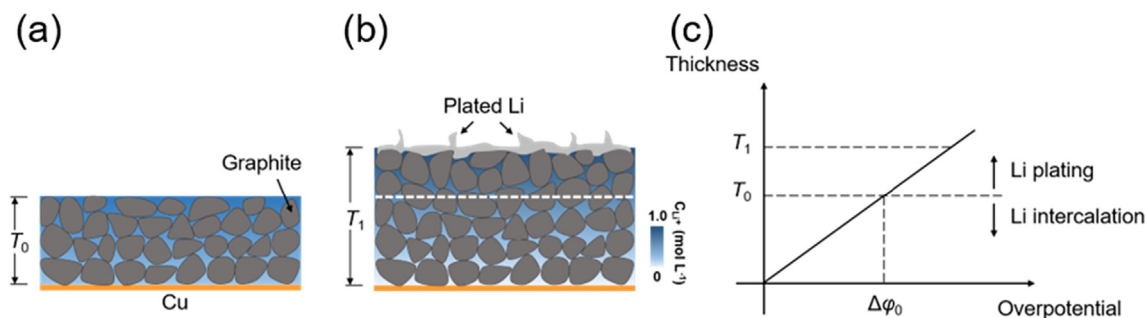


Fig. 4. Schematic of Li^+ transport in the graphite electrode with different thicknesses of (a) T_0 and (b) T_1 . (c) Relationship between electrode overpotential and electrode thickness. For an electrode with specific thickness (T_0) working under certain current density (C_0), Li plating would take place on electrode surface once the overpotential ($\Delta\phi$) surpasses the lithiation overpotential of active material ($\Delta\phi_0$). Larger thickness (T_1) than T_0 tends to encounter Li plating risk under the same current density (C_0).

the sluggish Li^+ transport behavior within the electrode leads to significant Li^+ concentration gradient from top to bottom under high current density [49,50]. Consequently, Li^+ tend to only intercalate into the graphite on the surface facing upwards. When the overpotential ($\Delta\phi$) caused by the Li^+ concentration polarization surpasses the lithiation overpotential ($\Delta\phi_0$) of active material, reduction of Li^+ results in formation of metallic Li dendrites under fast-charging condition (Fig. 4b) [50–52]. Under identical current densities, the thicker the electrode is, the higher the possibility of Li plating taking place (Fig. 4c). This phenomenon not only impacts the utilization of graphite anode capacity, but also raises concerns regarding cycling stability and safety [53,54]. To date, considerable efforts have been devoted to addressing this challenge and improving Li^+ transport capability across the electrode thickness, among which electrolyte design aimed at increasing Li^+ conductivity and electrode engineering focused on optimizing Li^+ transport pathway have been regarded as two effective strategies.

3.1.1. Electrolyte engineering

Electrolyte functions as the conduit for Li^+ transport in LIBs, consisting of solvents and lithium salts [55]. These solvents dissociate lithium salts, thereby generating Li^+ with a solvation structure within electrolyte. Consequently, the diffusion of Li^+ within the electrolyte is determined by the properties (e.g., viscosity) of solvent, as depicted in Eq. (6).

$$D = \frac{kT}{\alpha\pi\eta r_s} \quad (6)$$

Where k is the Boltzmann's constant, T is the temperature, η is the electrolyte viscosity, and r_s is the Stokes radius and α is a constant which ranges from 4 to 6 for perfect slipping conditions [56,57]. According to Eq. (6), the viscosity of electrolyte is inversely proportional to the diffusion coefficient of electrolyte.

The physical properties of common electrolyte solvents for LIBs are shown in Table S1. The selection of different solvents could regulate Li^+ transport property in electrolyte, and solvents with low viscosity often enable fast Li^+ transport kinetics. The commonly used commercial electrolyte typically consists of 1 M LiPF_6 in ethylene carbonate (EC)/diethyl carbonate (DEC), EC/ethyl methyl carbonate (EMC), EC/dimethyl carbonate (DMC), or EC/DMC/EMC hybrids, and displays Li^+ conductivity of 10–12 mS cm^{-1} [55,58–61]. Although EC serves as the primary solvent for dissolving LiPF_6 , its high viscosity (1.93 mPa s, 40 °C, Table S1) limits the Li^+ transport behavior. Additionally, its high melting point (36.4 °C) hinders the low-temperature application of LIBs.

Aliphatic esters [59,61,62] and nitrile solvents [63,64] with lower viscosity (Table S1) are commonly utilized as co-solvents or additives in electrolytes to enhance ionic conductivity. Methyl acetate (MA), which exhibits a low viscosity of 0.40 mPa s (Table S1), was proposed for dissolving lithium salt (LiPF_6) [59]. The highest conductivity of the electrolytes (different concentrations of LiPF_6 in MA) reaches 25 mS cm^{-1} , significantly surpassing all observed values in LiPF_6 -based electrolytes with different concentrations (EC/DMC, with the highest value of 11 mS cm^{-1}) at 20 °C. The $\text{LiNi}_{0.5}\text{Mn}_{0.2}\text{Co}_{0.3}\text{O}_2$ (NMC532)||graphite cells with high-areal-capacity graphite electrode ($\sim 5.6 \text{ mA h cm}^{-2}$) and

MA-based electrolyte retained approximately 65% capacity at 4 C (based on the capacity at 0.05 C). In contrast, cells with an EC/DMC-based electrolyte delivered only 20% of the discharge capacity under similar conditions (Fig. 5a). Similar to MA, methyl propionate (MP) also possesses a low viscosity of 0.43 mPa s (Table S1), and an MP-based electrolyte (1 M LiPF₆ in MP/fluoroethylene carbonate (FEC)) exhibits higher Li⁺ conductivity compared to that in 1 M LiPF₆ in EC/DMC and 1.2 M LiPF₆ in EC/EMC over a wide temperature range (Fig. 5b) [61]. The NMC622||graphite cell employing an MP-based electrolyte demonstrated the excellent first cycle capacity that was 82.5% of the capacity at 0.1 C when charged at 4 C and discharged at C/3, and retained 88.2% of the first cycle capacity after 500 cycles. Conversely, cells with 1.2 M LiPF₆ in EC/EMC maintained only 73.4% of the capacity at 0.1 C under high current density (charged at 4 C and discharged at C/3), and sharply decreased to 54.1% after 500 cycles. Acetonitrile (AN), one of the nitrile solvents, exhibits a high dielectric constant (35.95, Table S1) and low viscosity (0.34 mPa s, Table S1) [64], enabling it to effectively dissolve lithium salt and facilitate Li⁺ transport. The NMC811||graphite pouch cell (mass loading of ~2.85 mA h cm⁻² for NMC811) with an AN-based electrolyte (1 M LiPF₆ in FEC/AN) delivered exceptional rate capability and minimal polarization overpotential, retaining 85% of the original capacity when charged at 6 C and discharge at 1 C after 100 cycles, while cells with 1 M LiPF₆ in EC/DMC only retained less than 25% of the initial capacity after 50 cycles under the same condition (Fig. 5c).

Sand's formula (Eq. (5)) elucidates that Li⁺ transference number of electrolyte contributes to morphology of the deposited Li [36]. The value of Li⁺ transference number, representing its fractional contribution to overall conductivity, heavily relies on both the solvated structure of Li⁺ and anion structure of lithium salts. Efforts have been devoted to improving Li⁺ transference number of the electrolyte by optimization of lithium salts [65–67] or solvents [68]. For instance, benzimidazole-based lithium salt 5,6-dicyano-2-(trifluoromethyl) benzimidazolide (LiTDBI) was synthesized to design a novel electrolyte [66]. TDBI⁻ possessed a larger radius in comparison to conventional PF₆⁻, which hindered the diffusion of anions, resulting in a larger Li⁺ transference number. The electrolyte (~0.2 M LiTDBI in EC/DMC) achieved an impressive Li⁺ transference number of 0.74, surpassing the conventional LiPF₆-based electrolytes with a Li⁺ transference number of 0.4 (Fig. 5d). It is noteworthy that ionic liquids or polymer-based electrolytes typically exhibit higher Li⁺ transference number in comparison to liquid organic electrolytes. However, their overall ionic conductivity is comparatively lower, resulting in diminished rate performance.

During charging, the solvated Li⁺ migrates from the electrolyte towards the anode, and the Li⁺ conductivity serves as a characterization for this behavior, which is a measure of the tendency towards Li⁺ conduction, the movement of Li⁺ [69]. The full dissolution of lithium salt provides a basis for sufficient Li⁺ conductivity. Thus, the role of dielectric constant should be taken into consideration when selecting the electrolyte solvent. Moreover, while faster Li⁺ transport is often realized, aliphatic ester and nitrile solvents fail to form a dense protective layer on the graphite surface, leading to persistent parasitic reactions on the anode surface with these solvents [70,71]. A variety of electrolyte components have been investigated to stabilize the anode surface by electrochemically driving the formation of a passivation layer. It is well-known that FEC, lithium difluoro(oxalato)borate (LiDFOB), and vinylene carbonate (VC), all of which feature high reduction potential, have been extensively utilized as film-forming additives in fast-charging electrolytes [72]. For example, FEC is commonly employed to construct a dense protective LiF-rich SEI through reduction reactions at the graphite anode interface. However, the high viscosity of FEC complicates Li⁺ desolvation, a topic that will

be discussed in detail in Section 3.2.1. With regard to practical applications for fast-charging LIBs, it is necessary to explore an electrolyte that possesses high Li⁺ conductivity, a wide electrochemical window, and good electrode compatibility, a task that continues to pose a significant challenge.

3.1.2. Design of electrode structures

During the fabrication of a regular electrode, active materials, carbon black, and binder are mixed together, followed by coating the mixture on the current collector and drying at a high temperature under vacuum. An interconnected pore network is formed during the drying process after the removal of the solvents. In a practical cell, the electrolyte infiltrates into the pore structure within the electrode to build Li⁺ transport channels. However, the high tortuosity of the pore structure in regular electrode often causes long Li⁺ transport distance due to the randomly packed structure of solid components in the electrode [73]. The effective Li⁺ conductivity (D_{eff}) in an electrode can be calculated by Eq. (7), where τ is the electrode tortuosity, ε is the electrode porosity, and D is the Li⁺ conductivity of electrolyte [74,75]. A low value for τ and a high value for ε could enable high D_{eff} , therefore improving rate capability of an electrode. Hence, abundant attempts have focused on the construction of directionally-ordered pore structures within electrodes to decrease the electrode tortuosity, such as channel fabrication [76–78] and gradient structure [79–81].

$$D_{\text{eff}} = \frac{\varepsilon}{\tau} D \quad (7)$$

The electrode tortuosity can be effectively reduced through the introduction of supplementary channels. Polytetrafluoroethylene (PTFE) can be employed as a pore-formation agent, which undergoes depolymerization to form micro-sized pores after heat treatment, in order to synthesize a graphite anode with abundant inner pores for Li⁺ migration [76]. Li metal deposition was observed on the surface of the cycled regular graphite anode, which explained the decrease in capacity during cycling (92 mA h g⁻¹ after 100 cycles at 2 C). In sharp contrast, the graphite electrode fabricated with pore formation agent maintained its pristine morphology without any observable deposition of Li metal and delivered a much higher capacity of 233 mA h g⁻¹ under the same test condition. Such pore structure design reduced electrode tortuosity, facilitated Li⁺ transport within the electrode, and suppressed undesired Li metal plating. However, blindly increasing the porosity of the electrode would sacrifice the energy density of LIBs. In this respect, regulating ordered pore structure without excessively decreasing active materials density is a promising approach to achieve practical fast-charging LIBs. Modified by superparamagnetic iron oxide nanoparticles, vertical arrangement of graphite particles was achieved under a low magnetic field, greatly reducing the tortuosity of the graphite anode (Fig. 6a) [77]. A more ordered Li⁺ transport path enabled a high-loading aligned electrode (~10 mg cm⁻²) and showed 1.6 times higher capacity compared to a regular graphite electrode after 50 cycles at 1 C (Fig. 6b). A gradient electrode structure design could be another superior approach to achieving the balance between fast-charging performance and energy density. A three-layered gradient porosity graphite electrode was fabricated by a repeated slurry coating method (Fig. 6c) [80]. The gradient pore structure not only facilitated fast Li⁺ transport over the entire electrode but also ensured high energy density of batteries. Such a three-layered graphite electrode with a gradient pore structure (~9 mg cm⁻²) enabled a graphite||Li cell with a high capacity of 125 mA h g⁻¹ at 2 C after 150 cycles. In sharp contrast, the cell with regular (single-layered) graphite electrode with similar mass loading displayed low capacity of 50 mA h g⁻¹ after 75 cycles (Fig. 6d). Apart from

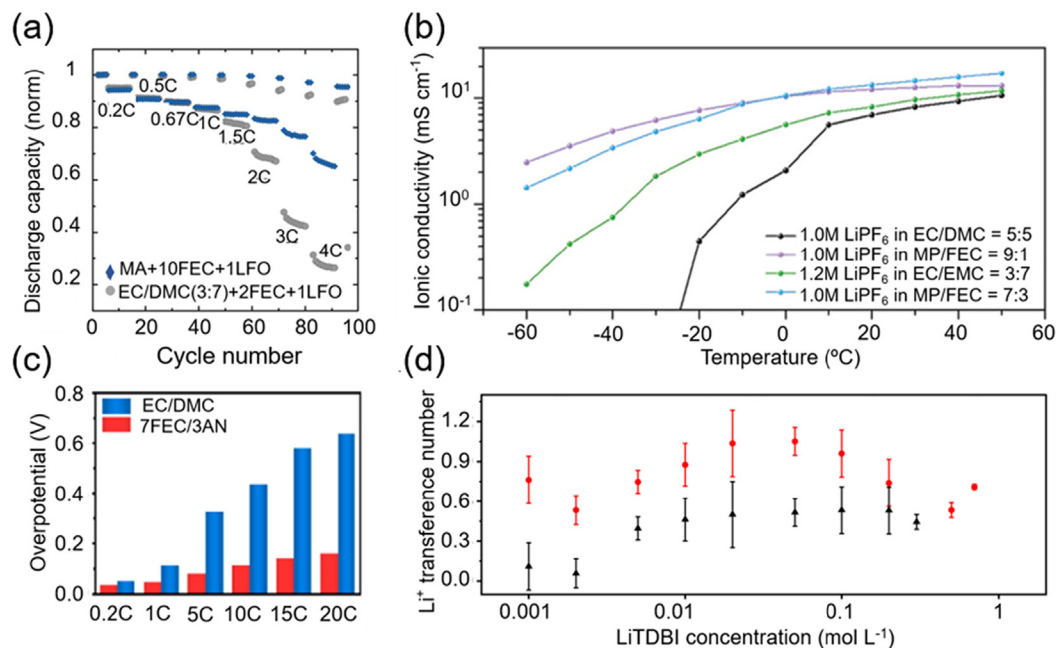


Fig. 5. Electrolyte engineering for facilitating Li^+ transport. (a) Fast-charging performance of MA-based electrolyte. Reproduced with permission from ref. [59]. Copyright 2020, American Chemical Society. (b) Ionic conductivity of MP-based electrolyte. Reproduced with permission from ref. [61]. Copyright 2022, Wiley-VCH GmbH. (c) Overpotential in acetonitrile-based electrolyte at different rates. Reproduced with permission from ref. [64]. Copyright 2022, American Chemical Society. (d) Li^+ transference number of LiTDBI-based electrolyte. Reproduced with permission from ref. [66]. Copyright 2015, Elsevier.

the gradient porosity design, a dual-gradient (electrode porosity and particle size) structure design for the graphite electrode has the potential to mitigate Li^+ concentration polarization (Fig. 6e) [81]. The dual-gradient anode possesses smaller particles and higher porosity on top, while larger particles and lower porosity were observed at the bottom, facilitating efficient Li^+ diffusion from top to bottom. The full cells with such graphite anodes and LiCoO_2 cathodes achieved 60% capacity in just 5.6 min and 80% capacity in 11.4 min at 6 C, much shorter than those using randomly structured graphite anode (12.4 and 23.8 min), exhibiting both high volumetric energy density (701 and 550 Wh L^{-1} of the random graphite anode) and fast-charging capability (Fig. 6f).

It is noted that the thickness of a regular electrode (T) determines the Li^+ diffusion distance (A). In a regular porous electrode, even if the porosity is sufficient to ensure fast transport of Li^+ , they are equal ($T = A$). Constructing a thinner electrode is a straight way to improve the fast-charging performance of LIBs; however, it significantly sacrifices the energy density. The tradeoff between energy density and fast-charging capability largely lies in the regulation of the electrode thickness in industry. Impressively, the introduction of additional aligned vertical pores could decouple T and A (Fig. 7a). With the same electrode thickness, A could be significantly reduced, enabling much improved fast-charging capability of batteries. Hence, the construction of aligned vertical pores, achieved by introduction of sacrificial magnetic phase [82,83], freeze tape casting [84,85], and laser-patterning process [10,86], could greatly facilitate Li^+ diffusion within graphite anode, due to the increased electrode porosity.

The sacrificial magnetic phase can be utilized to generate directional pore arrays within the electrode under an external magnetic field, with an example of high-area-capacity LiCoO_2 [82]. The sacrificial phase was added in the slurry for electrode coating and would be eliminated at high temperatures during sintering, resulting in the formation of directional channels (Fig. 7b and c). These directional pores facilitated Li^+ transport kinetics and yielded

excellent capacity release even under very high mass loading ($>12 \text{ mA h cm}^{-2}$) at 2 C. Recently, freeze tape casting has become an effective approach to optimizing electrode structure. Through freeze tape casting, a bilayer structured graphite electrode with directionally aligned channels was fabricated to facilitate Li^+ transport behavior [84]. In comparison with regular graphite anodes that possess randomly distributed particles, the graphite anode via this freeze tape casting (FTC) boasts numerous well-aligned channels, leading to decreased Li^+ diffusion distance and thus enhancing charge/discharge performance. Full cells with the bilayer hybrid FTC electrode exhibited a capacity retention of 55% after 1000 cycles at 5 C in comparison to those with the regular porous electrode (45%) (Fig. 7d). Apart from freeze tape casting, highly ordered graphite electrode arrays of vertical pore channels could also be produced by a controllable laser-patterning process (Fig. 7e) [10].

For electrodes with additional aligned vertical pores, further investigation is necessary to determine the effect of pore size and shape on the electrode mechanical properties, aiming for improving fast-charging performance and extending cycle life. In addition to considering tortuosity and porosity as discussed above, factors such as active material morphology and particle size will affect Li^+ diffusion within the graphite anode, which deserve to be carefully regulated in practical applications. Furthermore, simplicity and cost would also be taken into consideration for the practical implantation of electrode pore structure and tortuosity regulation for fast-charging LIBs in industry.

3.2. Li^+ desolvation and transport across SEI

The working potential of graphite anodes ($\sim 0.1 \text{ V vs. Li}^+/\text{Li}$) closely approximates the potential of metallic Li plating ($\sim 0 \text{ V vs. Li}^+/\text{Li}$) [87]. Electrolytes tend to decompose below 1.5 V before the first lithiation of the graphite anode, and the products of the reaction between electrolyte and electrode participate in the formation of

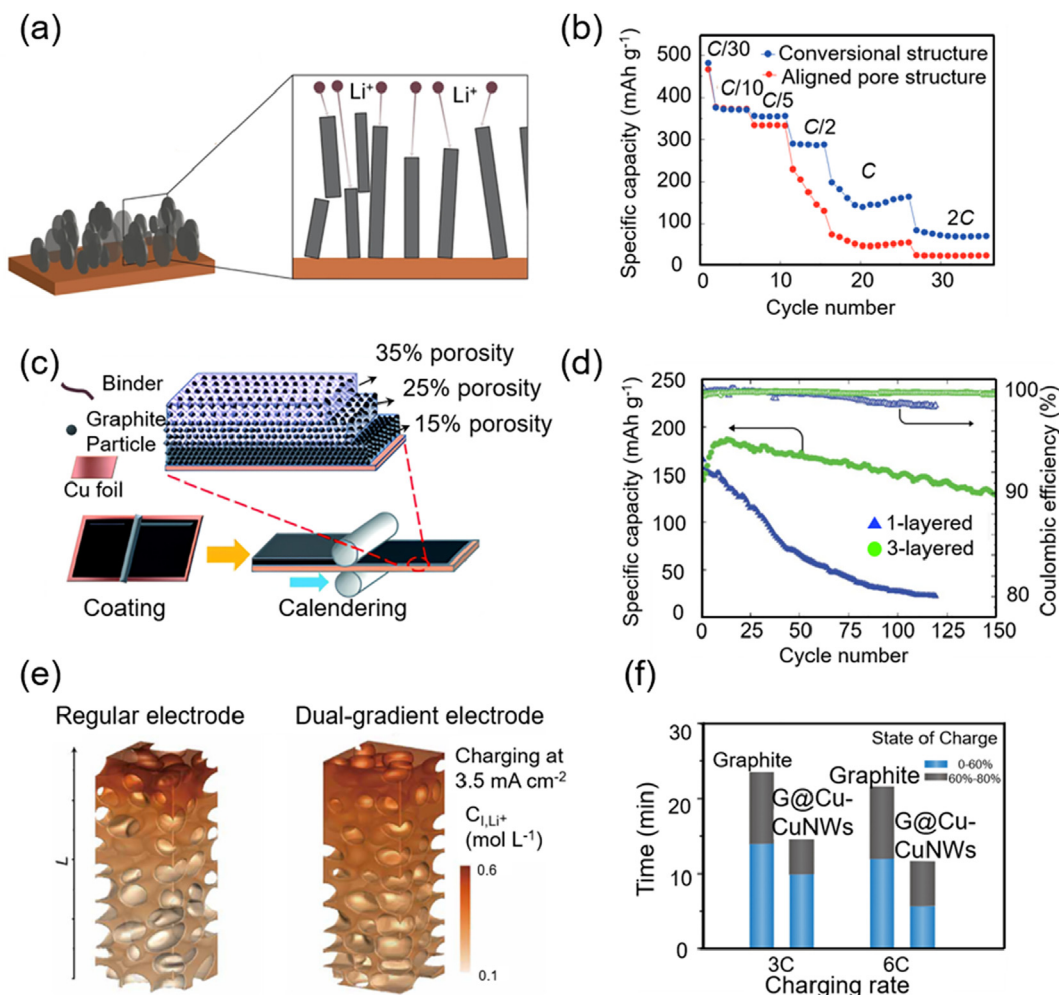


Fig. 6. Electrode structure designs for low-tortuosity electrode. (a) Schematics of graphite electrode with vertically aligned pore structure fabricated by superparamagnetic iron oxide nanoparticle modification and (b) its rate performance comparison to graphite electrode with conversational electrode structure under the same test condition. Reproduced with permission from ref. [77]. Copyright 2015, Springer Nature Limited. (c) Schematics of 3-layered gradient porosity graphite electrode and (d) its rate performance comparison to graphite electrode with conversational electrode structure under the same test condition. Reproduced with permission from ref. [80]. Copyright 2022, Royal Society of Chemistry. (e) Cross-section illustration of the dual-gradient (electrode porosity and particle size) structure electrode and regular graphite electrodes and (f) their fast-charging performance comparison. Reproduced with permission from ref. [81]. Copyright 2022, American Association for the Advancement of Science.

the SEI, which plays a role in passivating the graphite anode interface [88]. The mechanism for SEI formation has been explained. The potential for lithium intercalation into graphite is lower than the lowest unoccupied molecular orbital (LUMO) of solvent molecules (Fig. 8a) [89–91]. A widely accepted SEI structural model is Peled's mosaic model proposed in 1997, where SEI consists of different components in the form of heteropoly microphases and its inner layer consists mainly of inorganic components (e.g., Li_2O , LiF , and Li_2CO_3) while its outer layer consists primarily of organic components (e.g., lithium ethylene dicarbonate) (Fig. 8b) [92,93]. Li^+ can transport through the bulk and interface of the heteropoly-microphase. The in-situ liquid secondary ion mass spectrometry (liquid-SIMS) technique further refined the double-layer SEI structure [94]. The inner inorganic layer is dense, where Li^+ diffusion occurs through interstitial knock-offs, whereas the outer organic layer has a porous structure that allows electrolyte penetration to transport Li^+ (Fig. 8c) [95,96]. Numerous efforts have been devoted to exploring strategies for accelerating Li^+ migration in LIBs. Electrolyte engineering could not only affect the Li^+ transport within the bulk, but also regulate Li^+ desolvation, SEI component, and structure, all of which play significant roles in the electrochemical performance of LIBs.

3.2.1. Desolvation of Li^+ solvated structure

Li^+ interacts with solvents and anions to form solvated Li^+ in regular electrolytes (Fig. 9a) [55,97]. The desolvation process occurs on the electrode surface before Li^+ transports into SEI and intercalates into graphite. Solvents with a strong affinity for Li^+ are often used to sufficiently dissolve lithium salts, which however leads to difficulties in Li^+ desolvation. Solvents with low Li^+ binding energy are beneficial for easy Li^+ desolvation, which mainly depends on the composition of the electrolyte [31,98]. Introducing competitive anions or non-polar solvents can effectively weaken the interaction between Li^+ and regular solvents and reduce the Li^+ desolvation energy in the electrolyte (Fig. 9b) [99]. Through rational electrolyte design, a dual-salt electrolyte consisting of lithium bis(fluorosulfonyl)imide (LiFSI) and lithium difluoro(oxalato)borate (LiDFOB) in methyl sulfite (DMS) achieved fast Li^+ desolvation within a wide temperature range [100]. Calculation results revealed that bond length between Li^+ and DMS was shortened, implying weaker interaction between Li^+ and solvents, when an FSI^- or DFOB^- anion entered the Li^+ coordination structure (Fig. 9c). The LiCoO_2 ||graphite full cell (mass loading of 10 mg cm^{-2} for LiCoO_2) exhibited a capacity retention of 80% after 4500 cycles at 6 C and 30 °C, showing excellent long-cycling performance.

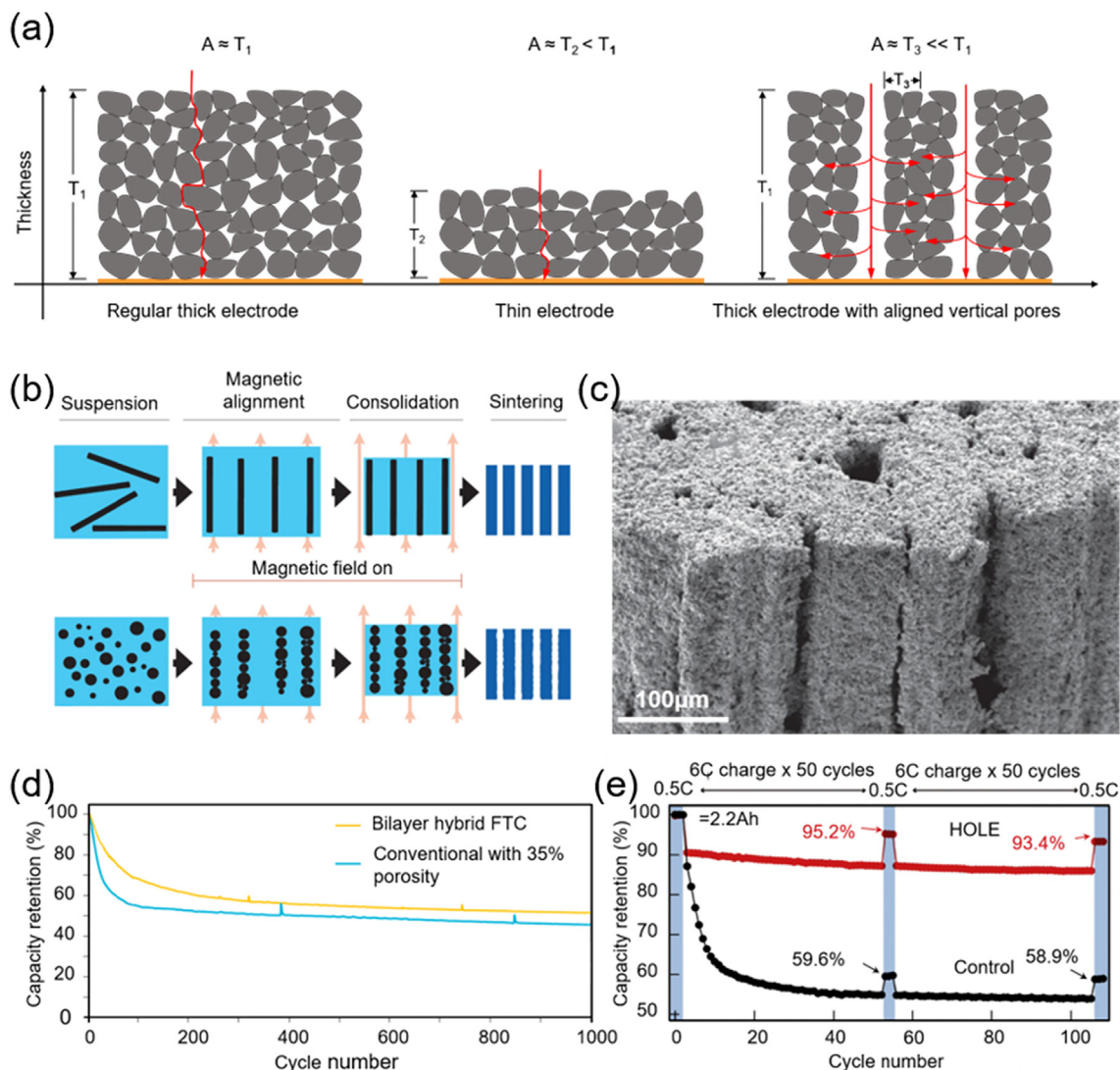


Fig. 7. Electrode structure designs for aligned vertical pores. (a) Schematic of Li^+ transport pathways in regular thick electrode, thin electrode, and thick electrode with aligned vertical pores. T is electrode thickness and A is Li^+ diffusion distance. (b) Fabrication of LiCoO_2 electrode using magnetic alignment of two different sacrificial phases and (c) the cross-section of the fabricated LiCoO_2 electrodes with aligned pore channels templated by magnetically chained emulsions (b, bottom). The sacrificial phase could be removed via sintering. Reproduced with permission from ref. [82]. Copyright 2016, Springer Nature Limited. (d) Capacity retention of bilayer hybrid FTC electrode and regular electrode. Reproduced with permission from ref. [84]. Copyright 2022, Elsevier. (e) Normalized discharge capacity of highly ordered laser-patterned electrode (HOLE) and regular graphite anode at 6 C. Reproduced with permission from ref. [10]. Copyright 2020, Elsevier.

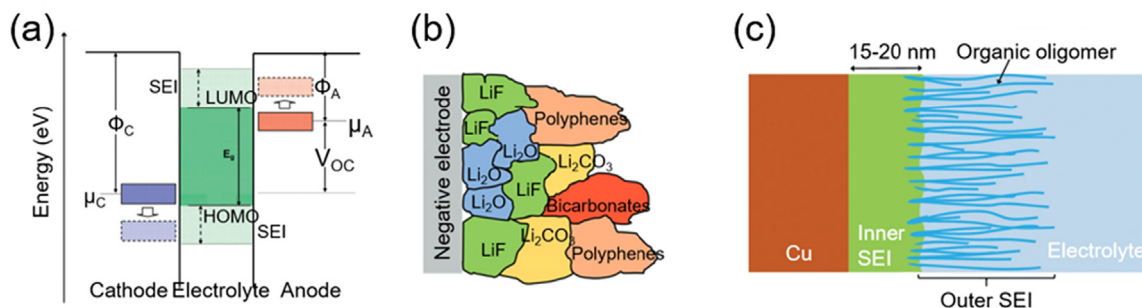


Fig. 8. (a) Schematic of electrochemical window of electrolyte. Reproduced with permission from ref. [89]. Copyright 2009, American Chemical Society. (b) Schematic of the mosaic model SEI structure. Reproduced with permission from ref. [29]. Copyright 2020, Royal Society of chemistry. (c) Schematic of Li^+ transport across SEI. Reproduced with permission from ref. [94]. Copyright 2020, Springer Nature.

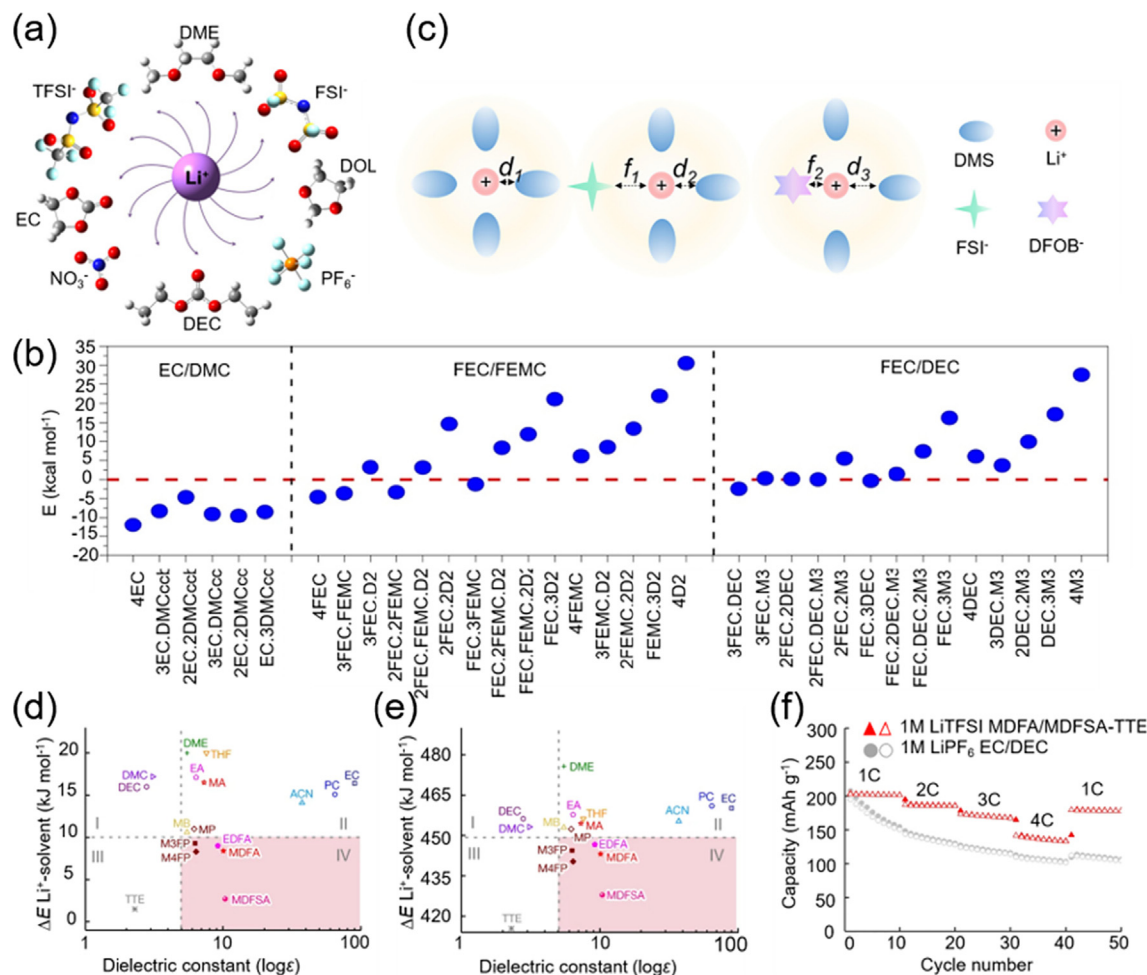


Fig. 9. (a) Schematic of interaction between Li⁺ and anions/solvents. Reproduced with permission from ref. [97]. Copyright 2019, Wiley-VCH. (b) The calculated Li⁺ solvation/desolvation energy in different electrolytes. Reproduced with permission from ref. [99]. Copyright 2019, Springer Nature. (c) Schematic of bond length between Li⁺ and anions (*f*₁, *f*₂) / solvent (*d*₁, *d*₂, *d*₃). Reproduced with permission from ref. [100]. Copyright 2023, American Chemical Society. The solvent diagram of (d) donor number versus dielectric constant and (e) binding energy between Li⁺ and solvents versus dielectric constant. (f) Rate performance of fluorinated electrolyte and 1 M LiPF₆ in EC/DEC at 4 C. Reproduced with permission from ref. [32]. Copyright 2023, Springer Nature.

Besides, solvents with weak binding energy to Li⁺ are used for designing weakly solvated electrolytes (WSE) in order to accelerate the Li⁺ desolvation process [8,101,102]. Through the competitive interaction between AN and fluorobenzene (FB), a WSE consisting of 2 M LiFSI in AN/FB succeeded in accelerating the Li⁺ desolvation [8]. The binding energy between Li⁺ and AN was significantly lower than that between Li⁺ and EC, leading to enhanced kinetics of Li⁺ desolvation. The NMC811||graphite cell (mass loading of 2 mg cm⁻² for NMC811) using the AN-based WSE showed a high specific capacity retention of 83% at 5 C (167 mA h g⁻¹) compared to the capacity at 0.5 C (201 mA h g⁻¹).

Fluorine atoms with high electronegativity can achieve a more uniform electron distribution in solvent molecules, thereby weakening the interaction between Li⁺ and solvents [103]. As a result, designing fluorinated solvents is also an effective strategy to accelerate Li⁺ desolvation [32,104]. To minimize the Li⁺-solvent binding energy while ensuring sufficiently lithium salt dissociation, solvents with a relatively low donor number (less than 10) and high dielectric constant (more than 5) values were studied (Fig. 9d and e) [32]. Through comprehensive screening, the fluorinated electrolyte consisting of 1 M lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) in methyl difluoro acetate (MDFA)/methyl 2,2-difluoro-2-(fluorosulfonyl) acetate (MDFSA)/1,1,2,2-tetrafluoroethyl-2,2,3,3-tetrafluoropropylether (TTE) delivered a high specific

capacity of 140 mA h g⁻¹ at 4 C in NMC811||graphite full cells, much higher than 106 mA h g⁻¹ at 4 C in 1 M LiPF₆ in EC/DEC (Fig. 9f).

Despite the positive effect on Li⁺ desolvation of solvents with weak interaction with Li⁺, they often face difficulties in dissociating lithium salts. Subsequently, the low ionic conductivity inherent to WSEs emerges as a principal limitation, thereby posing substantial challenges for their practical implementation [8]. It should be noted that solvents in Li⁺ solvated structure tend to be reduced on the electrode surface during SEI formation. The co-intercalation of Li⁺ solvation structure into graphite particles would irreversibly destroy the graphite lattice, due to electrolyte decomposition within the graphite particle, resulting in shedding of the active material [105,106]. SEI could provide locations for Li⁺ desolvation on the electrode surface and function as a shield for co-intercalation of Li⁺ solvated structure. Accelerating Li⁺ desolvation could facilitate electrochemical reaction kinetics and weakening the interaction between Li⁺ and solvents could be an effective approach.

3.2.2. SEI engineering

Ideally, the SEI should uniformly cover the graphite surface to inhibit parasitic reactions between the electrolyte and active material, as well as facilitating Li⁺ transport [88,107]. Sufficient chemi-

cal and mechanical stability of the SEI is highly desirable for achieving long-term cycling stability of the anode. The component and structure of the SEI play vital roles in its properties, including Li^+ transport and mechanical stability. However, SEI always undergoes a dynamic process of “dissolution/regeneration” during charge/discharge cycling [108,109]. Due to cracks of the SEI caused by anode volume changes during cycling, electrolytes tend to be continuously consumed on freshly exposed active surfaces, resulting in sustained growth of SEI [110]. Cryogenic transmission electron microscopy (cryo-TEM) was recently explored to track the evolution of SEI on a carbon black anode [111]. The results showed that when dominated by inorganic components, SEI maintained a compact structure within thin thickness (~ 5 nm) and passivated the anode effectively; however, when dominated by alkyl carbonate (organic species produced via the electrolyte decomposition), SEI was relatively loose (~ 100 nm) after 20 cycles (Fig. 10a). The continuous growth of the SEI would exacerbate heterogeneity at electrode interfaces and impede Li^+ intercalation into graphite. In this regard, an inorganic component-dominated SEI is more preferable for long-term cycling stability.

SEI often contains numerous organic species arising from the decomposition of free organic solvents. In order to achieve an inorganic-rich SEI, certain fluorine-containing solvents/additives and anions have been introduced into the electrolyte to induce the formation of a LiF-rich SEI [112,113]. Fluorosulfonyl isocyanate (FI), which has a high reduction potential (above 2.8 V vs. Li^+/Li), was employed as a novel electrolyte additive for forming the SEI (Fig. 10b) [112]. FI is reduced prior to the carbonate solvents, yielding a thick and protective inorganic SEI on the graphite surface. The graphite||Li cell (mass loading of 3.31 mg cm^{-2} for graphite) with 1 M LiPF_6 in EC/DMC-2 wt% FI exhibited doubled charge capacity at 5 C in comparison to those with 1 M LiPF_6 in EC/DMC.

With the focus directed towards anions, the preferential decomposition of anions has emerged as a novel strategy for constructing an inorganic-rich SEI. Due to the weak interaction between Li^+ and solvents in WSE, anions are capable of entering the Li^+ solvated sheath and preferentially decompose at the interface (Fig. 10c) [114–116]. It is reported that a WSE composed of 1 M LiFSI in 1,4-dioxane (1,4-DX), a cyclic ether solvent, could enable an anion-dominated Li^+ solvated structure [114]. The binding energy between Li^+ and FSI^- (-3.15 eV) is twice more than that between Li^+ and 1,4-DX (-1.13 eV), ensuring that anions enter the Li^+ solvated shell and decompose preferentially at the interface to form an inorganic SEI. Temperature-dependent EIS revealed that the activation energy for Li^+ transport through SEI in such WSE (26.6 kJ mol^{-1}) was significantly lower than that in 1 M LiFSI in EC/EMC (44.7 kJ mol^{-1}). WSE with 2% EC exhibited capacity retention of 92% of its initial capacity in graphite||Li half cells after 500 cycles at 1 C, while the capacity in 1 M LiFSI in EC/EMC decreased to only 34% of its initial value after 300 cycles at 1 C.

In regular electrolytes, Li^+ and the anions of the lithium salt primarily exist as solvent separated ion pairs (SSIP), with a significant number of free solvent molecules dissociated in the electrolyte. This high concentration of free solvent molecules increases their likelihood of decomposition [70]. However, when the salt concentration reaches a certain high degree (>2 M), contact ion pairs (CIP) and higher-order agglomerates (AGG) start to form. The coordination structure of Li^+ -anions reduces the LUMO level of anions, facilitating their approach to the anode surface and enhancing their tendency for reduction on the graphite anode surface [117–119]. At the same time, numerous coordinated Li^+ -solvent molecules significantly reduce the presence of free solvent molecules in the electrolyte, thereby impeding the decomposition of organic solvents. Therefore, increasing lithium salt concentration is considered as an effective strategy for building advanced SEI [120–122]. It is reported that a localized high-concentration electrolyte, consisting

of 1.6 M LiFSI in DMC/EC with TTE as the diluent, enabled the formation of a uniform and robust SEI on the graphite surface through the preferential decomposition of anions [121]. A LiF-rich SEI with high Li^+ conductivity and high Li^+ flux achieved uniform Li distribution on graphite anode. When using such electrolyte in LiFePO_4 ||graphite pouch cells (1.2 Ah), fast-charging cycling stability is observed with 84.4% of initial capacity retention for 150 cycles at 6 C. In sharp contrast, cells using 1 M LiPF_6 in EC/DMC experienced significant capacity fade to nearly zero after only 50 cycles. It should be noted that an SEI containing excessive inorganic components could become very brittle and less adaptable to volume changes in the graphite anode during cycling. This brittleness makes it susceptible to high-stress-induced destruction, leading to repeating crushing and reconstruction of the SEI [114,123]. Consequently, this could lead to a continuous increase in SEI thickness, posing a risk for nucleation of metallic Li and growth of dendritic metallic Li on exposed fresh graphite surfaces. Therefore, it is crucial to emphasize the tradeoff between organic and inorganic components within SEI for safe fast-charging LIBs.

Besides, the construction of an “artificial SEI”, a preformed coating layer, has been proved as an effective strategy to promoting Li^+ transport on the anode surface [6,124,125]. The artificial SEI separates the electrode from the electrolyte and prevents excessive parasitic reactions of the electrolyte that lead to the formation of a thicker native SEI. The surface coating could effectively address the continuous growth of SEI during cycling by applying a layer with low electron conductivity at the interface, which inhibits continuous electrolyte decomposition. A single-ion conducting solid electrolyte ($\text{Li}_3\text{BO}_3\text{-Li}_2\text{CO}_3$, LBCO) was deposited onto graphite anodes as a stable artificial SEI using atomic layer deposition [124]. X-ray photoelectron spectroscopy (XPS) results demonstrated that this specific coating layer effectively suppressed the continuous electrolyte decomposition. The content of oxygen (O), fluorine (F), and boron (B) on the LBCO-coated electrode surface was much higher than that on a regular graphite electrode surface after cycling, implying an increased content of inorganic components in the SEI (Fig. 10d). The SEI resistance of the LBCO-coated electrode exhibited a fourfold reduction compared to that of naturally formed SEI on the regular electrode. The NMC532||graphite pouch cell with an LBCO-coated graphite anode retained 80% of its initial capacity at 4 C after 500 cycles while the capacity of the cell with an uncoated graphite anode faded to only 80% after just 12 cycles. At the same time, the presence of a reasonable coating layer is beneficial for promoting the dynamics of Li^+ intercalation. It is reported that the Li^+ diffusion rate could be significantly enhanced by coating graphite with a nanoscale turbostratic carbon layer (G@TC) [6]. Those additional fast Li^+ diffusion pathways greatly facilitated Li^+ transport (Fig. 10e). As a result, the coated graphite anode exhibited an improved Li^+ diffusion coefficient (D_{Li^+}) (6.6×10^{-10} and $0.7 \times 10^{-10} \text{ cm}^2 \text{ s}^{-1}$ at $x = 0.4$ and 0.7 in $\text{Li}_x\text{-C}_6$, respectively), calculated from the galvanostatic intermittent titration technique (GITT), which were both higher than that of the regular graphite electrode (3.5×10^{-10} and $0.4 \times 10^{-10} \text{ cm}^2 \text{ s}^{-1}$, respectively). The LiFePO_4 ||G@TC cell could achieve an impressive capacity retention of 87% after 300 cycles at 1 C, outperforming the regular graphite anode (70% after 250 cycles).

It is worth noting that constructing a perfect artificial SEI on the graphite anode remains a significant challenge, and therefore complete suppression of electrolyte decomposition cannot be achieved. A new SEI (native SEI) would still grow during the charge/discharge processes [124]. Currently, studies primarily focus on the construction of efficient and stable SEI, with little attention paid to the interphase evolution upon the artificial SEI, which plays a crucial role in the electrochemical performance of anodes. The SEI remains unstable and dynamically evolves during electrochemical cycling, which makes it still one of the most mysterious parts in battery

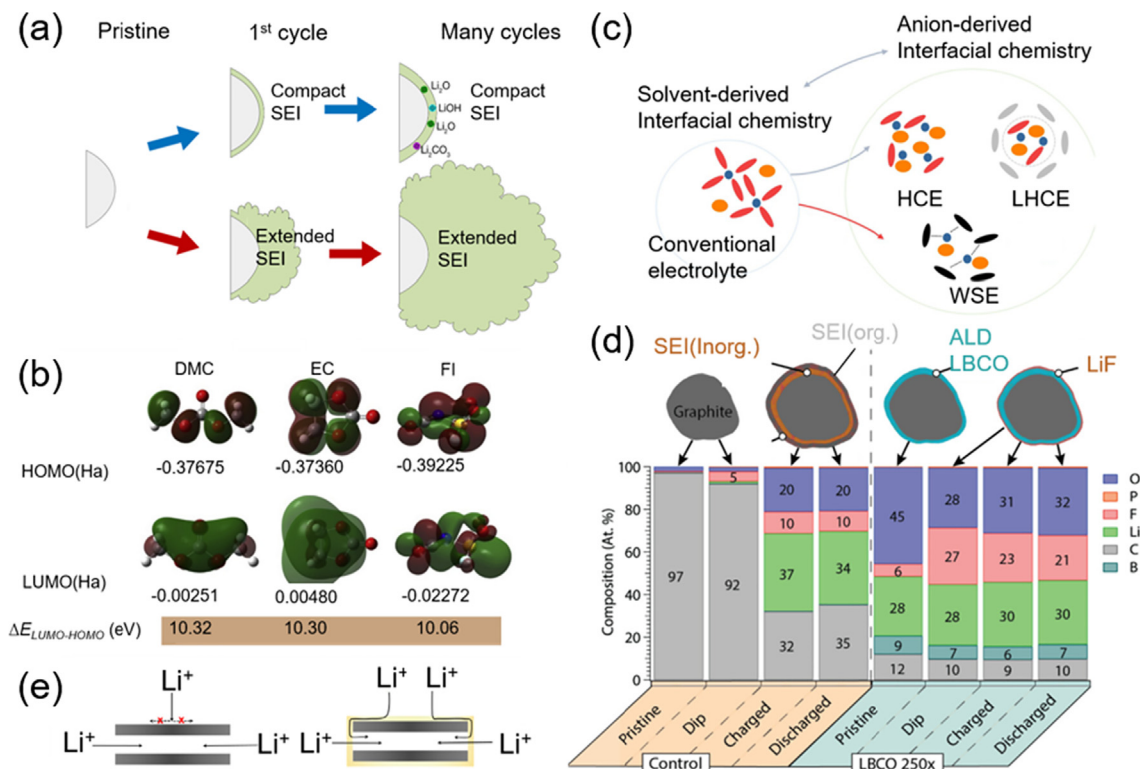


Fig. 10. (a) Schematic of two different pathways of SEI evolution on carbonaceous anodes. Reproduced with permission from ref. [111]. Copyright 2019, American Chemical Society. (b) Schematic of highest occupied molecular orbital (HOMO) and LUMO of DMC, EC, and FI molecules. Reproduced with permission from ref. [112]. Copyright 2019, Elsevier. (c) Schematic of solvation structures in conventional electrolyte, high concentrated electrolyte (HCE), localized high concentrated electrolyte (LHCE), and weakly solvating electrolyte (WSE). Reproduced with permission from ref. [114]. Copyright 2020, Wiley-VCH GmbH. (d) Schematic evolution of the surface SEI film for regular and LBCO-coated electrodes. Reproduced with permission from ref. [124]. Copyright 2022, Wiley-VCH GmbH. (e) Scheme of Li⁺ diffusion into regular graphite anode and turbostratic carbon-coated graphite anode. Reproduced with permission from ref. [6]. Copyright 2020, Wiley-VCH GmbH.

studies. Additionally, the chemical and environmental instability of the SEI poses difficulties for its investigation. Innovative experimental ideas and characterization methods are highly desirable to deeply understand the SEI.

3.3. Li⁺ transport in bulk material

After passing through the SEI, Li⁺ intercalate into the graphite particles to form Li-graphite intercalation compounds (Li_xC_6). However, graphite serves as a very typical two-dimensional electrode material, where Li⁺ can only fast enter the graphite layer along its terminal direction. By accurately quantifying the surface heterogeneity, high proportion of exposed non-basal (edge and defect) planes is advantageous in terms of fast-charging kinetic performance of graphite anode, on which the Li⁺ diffusion is 4 orders of magnitude faster than that across the basal plane [126]. This property of the graphite anode increases the Li⁺ transport distance, since Li⁺ contacting the basal plane cannot intercalate into graphite particles directly and will be trapped in a slow transport process towards the edge plane [127].

Defects on the surface of graphite particles can accelerate Li⁺ intercalation kinetics [128]. KOH was applied to etch graphite particles under high temperature to form a multi-channel structured graphite anode (Fig. 11a) [129]. Holes on the surface could increase the active sites for Li⁺ intercalation and de-intercalation, inducing a shorter Li⁺ diffusion distance. As a result, the full cell with the KOH etched graphite anode exhibited good capacity retention of 93% after 100 cycles at 3 C, outperforming the full cell with the regular graphite anode (85%). The air oxidation method as an optimized method can also help to produce defects on the graphite surface

to design multi-channel structured graphite electrodes, which is promising for practical applications (Fig. 11b) [130]. As for the regulation of particle morphology, spheroidization processing is a common method to improve the electrochemical performance of graphite anodes. Spheroidized graphite particles possess reduced anisotropy, homogeneous particle size, and an increased amount of non-basal planes, facilitating fast Li⁺ intercalation [131,132].

The chemical diffusion coefficient of Li⁺ in graphite can be measured through potential relaxation technique (PRT) [30]. With the continuous Li⁺ intercalation, the Li⁺ diffusion coefficient tends to be stable (Fig. 11c). The lowest diffusion coefficient was selected to estimate the time of Li⁺ diffusion to the center of graphite particle. The results showed that it only took 7.89 min (7.60 C) for Li⁺ diffusion from the surface to the bulk center of graphite when the diameter of graphite particle was 10 μm . Hence, with diameter less than 10 μm , graphite particles could enable Li⁺ transport within graphite particles at fast-charging rate of 6 C. Besides reducing particle size, increasing interlayer spacing of graphite can enhance Li⁺ diffusion capability within bulk graphite particles [133,134]. Oxidation under a mild condition endowed expanded graphite particles with a larger layer spacing of 0.339 nm compared to pristine graphite particles (0.336 nm) (Fig. 11d).

Graphite is long-term the dominative anode material in LIBs. Strategies with economy and industry compatibility for improving graphite are highly desirable. It is worth noting that although graphite particles with smaller particle size can reduce the Li⁺ diffusion distance and thus increase the Li⁺ diffusion coefficient, their relatively larger specific surface areas can lead to serious side reactions between the active particles and electrolytes [135]. The increased interlayer spacing may also reduce the structural stabil-

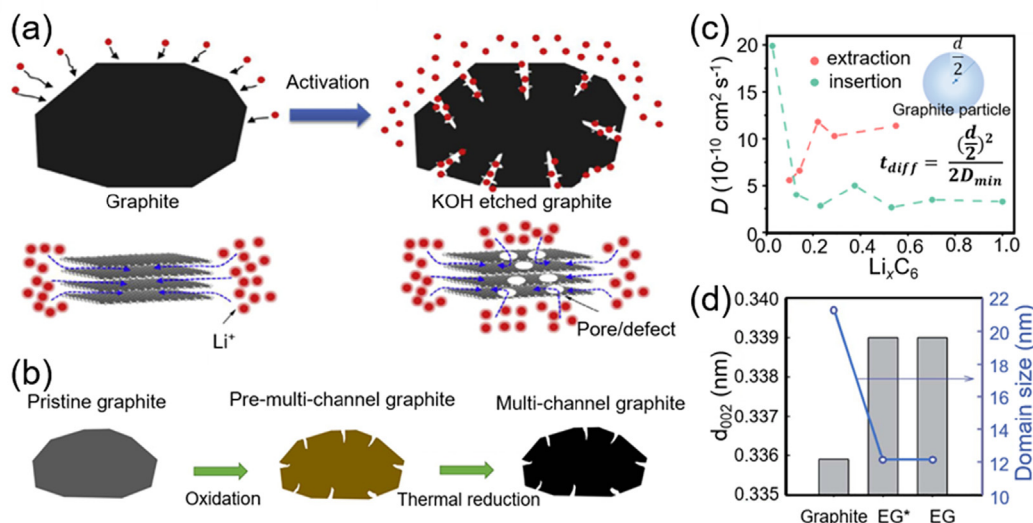


Fig. 11. (a) Schematic of pristine graphite and KOH etched graphite. Reproduced with permission from ref. [129]. Copyright 2015, Elsevier. (b) Schematic of oxidized graphite. Reproduced with permission from ref. [130]. Copyright 2018, IOP Publishing. (c) Calculated Li^+ chemical diffusion coefficient D in the lithiated graphite (Li_xC_6 , $0 \leq x \leq 1$). Reproduced with permission from ref. [30]. Copyright 2023, SpringerLink. (d) Comparison of layer spacing values and domain sizes. Reproduced with permission from ref. [133]. Copyright 2014, Royal Society of Chemistry.

ity of graphite particles during electrochemical cycling. In addition, the interaction between the material and the interface needs to be studied systematically for these specifically designed graphite materials. For example, it remains unclear that how defect engineering of graphite would influence the surficial physicochemical properties and the generated SEI during electrochemical process.

3.4. The others

In the context of graphite-based fast-charging LIBs, it is imperative for these batteries to acclimate to a diverse range of external operational conditions. However, these varying conditions invariably affect the intercalation behavior of Li^+ ions [38]. Each stage of Li^+ transport during the charging process exhibits a temperature dependency. The capacity degradation and Li dendrite formation at lower temperatures can be ascribed to the pronounced influence of low temperature on Li^+ transport kinetics, in line with the Arrhenius law. Beyond the optimization of electrolytes or electrodes at the material level, internal preheating methods are being earnestly considered due to their superior efficiency and uniformity. These include self-heating by battery discharge, convective heating using a fan and a resistance heater powered by the battery, mutual pulse heating, and alternating current heating. For instance, a thermal stimulation method was present to reduce the impact of low temperatures on fast-charging performance (e.g., graphite intercalation kinetics was improved by 13 times) through charging at 60 °C and discharging at room temperature [18].

The refinement of charging protocols could potentially augment battery performance by modulating electron flux across varying states of charge (SOC). An accumulation of numerous Li^+ ions at a high SOC on the graphite surface leads to increased polarization, which in turn promotes the nucleation of metallic Li. A conventional charging protocol typically encompasses a constant current (CC) phase until a predetermined cut-off voltage is reached, succeeded by a constant voltage (CV) phase until the current decreases to zero. The inclusion of an additional CV step, where the current is gradually diminished to zero, facilitates the homogenization of concentration gradients within the electrode [136]. Although escalating the charging current shortens the time needed for attaining the cut-off voltage, it necessitates an extended duration for restor-

ing electrode capacity during the CV phase. Innovative charging protocols, such as pulsed current and tapered current, have been suggested to curtail charging time while conserving battery lifespan [41].

4. Regulating uniform lithium plating on graphite

Lithium metal is ubiquitously considered the 'holy grail' anode for rechargeable lithium-based batteries, owing to its unparalleled energy density. Nevertheless, the high reactivity, Li dendrite formation, and large volume change during the charge/discharge process pose substantial challenges to the longevity of lithium metal batteries (LMBs). To this point, a multitude of studies have suggested enhancements to LMBs through strategic electrode structure design, meticulous interface engineering, and meticulous electrolyte optimization, where the regulation of Li plating is a critical step [137,138].

It is noteworthy that achieving controlled metallic Li plating on graphite to realize a highly reversible "graphite-Li hybrid" anode, rather than the formation of metallic Li dendrites, represents a promising avenue to meet the high energy demands and rapid charging requirements of LIBs [139]. Uniform and reversible deposition of metallic Li with a low accessible surface area and excellent connection to the graphite electrode under fast-charging conditions not only mitigates potential safety hazards associated with short circuits but also minimizes irreversible Li loss and enhances electrochemical cycling stability.

To construct a "graphite-Li hybrid" anode through regulating uniform Li plating, it is essential to have a lithiophilic graphite surface and reserved space for metallic Li deposition [140–143]. This can be achieved through appropriate electrolyte design and pre-treatment of the anode surface/structure. Compared to the Li metal anode, the graphite material in the "graphite-Li hybrid" electrode not only serves as active material for Li^+ storage but also functions as a collector/host for the uniform Li-metal layer under high current density. Consequently, various modification strategies for the Li metal anode could be effective in enhancing fast-charging LIBs based on "graphite-Li hybrid" anode. One such strategy involves incorporating LiNO_3 , a widely used additive for Li metal anodes that contributes to the formation of a Li_3N -rich SEI facilitat-

ing uniform Li plating [142]. Due to NO_3^- preferentially being adsorbed onto the Helmholtz layer, the resulting reduced Li_3N is closer to the graphite surface compared to well-known LiF . Hence, Li^+ ion diffusion on the graphite surface is accelerated by the presence of Li_3N in the SEI, prompting Li nucleation to follow a two-dimensional progressive/instantaneous (2DP/2DI) mode instead of forming lithium dendrites during fast-charging conditions. A 1.2-Ah LiFePO_4 ||graphite pouch cell using the LiNO_3 -based electrolyte demonstrated stable capacity after 150 cycles at 3 C (cell using the EC/DMC/FEC electrolyte retained only 78.1% capacity), thus illustrating the inhibitory effect of the regulation of uniform lithium plating on the capacity fade induced by lithium dendrites. Although several studies have focused on the uniform metallic Li plating on graphite, there are still several mysteries should be further investigated: (1) the plating/stripping behavior of metallic lithium on graphite anode surface in view of the inherent heterogeneity between graphite anode and metallic Li; (2) the real utilization of deposited metallic Li on the graphite substrate.

5. Conclusions and perspectives

Graphite has long-term been the dominant anode material for LIBs due to its low cost, abundant resources, stable electrochemical performance, and superior industrial compatibility. To meet the increasing demand for fast charging in the current energy storage market, it is crucial to address the problem of dendritic metallic lithium formation on the graphite surface under fast charging conditions. This review provides a fundamental understanding of metallic Li dendrites formation on the graphite anode during fast charging and offers insights into strategies for inhibiting the growth of metallic Li dendrites by showcasing typical current research progress (e.g., electrolyte engineering, electrode structures designing, Li^+ desolvation regulating, SEI constructing, and bulk materials modifying). It should be noted that fast-charging design should be conducted on the basis of maintaining high energy density in state-of-the-art graphite-based LIBs.

Metallic Li plating is a competitive process for Li^+ intercalation into graphite anode under fast-charging conditions. The key to inhibiting the process of dendritic metallic Li plating lies in improving Li^+ migration. In theory, regular graphite particles can facilitate Li^+ transport within them at fast-charging rate of 6 C [30]. Besides designing materials that promote Li^+ transport within bulk particles, numerous investigations focus on improving Li^+ transport in electrolyte within porous graphite electrodes and facilitating Li^+ desolvation and transport at the SEI. Electrolyte design and electrode structural engineering have been regarded as two effective strategies for accelerating Li^+ transport within graphite electrode. For electrolyte designing, the initial Li^+ concentration, Li^+ transference number, and solvent viscosity all contribute to concentration polarization in electrodes. Co-solvents or additives in electrolytes have been identified as effective approaches to reduce polarization and promote transport properties of Li^+ , such as using aliphatic esters and nitrile solvents with lower viscosity to enhance ionic conductivity and employing novel lithium salts to improve the Li^+ transference number. A porous electrode with high porosity and low tortuosity might be highly conducive to the penetration of the electrolyte into the electrode, thereby reducing Li^+ concentration gradient and consequently minimizing electrode overpotential. Regarding Li^+ desolvation, nonpolar solvents with low Li^+ binding energy are beneficial for facilitating Li^+ desolvation; however, they possess low dissociation of lithium salts. Solid-state lithium-based batteries (SSLBs), which utilize solid-state electrolytes, present a promising avenue to bypass the kinetic limitations induced by Li^+ desolvation in liquid electrolytes [144–147]. Nonetheless, the restricted ionic conductivity of many

solid-state electrolytes at ambient temperatures could be a critical impediment, hindering the prevention of lithium dendrite growth under high current density conditions. Once the challenge of low ionic conductivity is effectively tackled, the actualization of rapid-charging solid-state lithium-ion batteries may transition from theory to feasibility.

Despite significant progress in improving the fast-charging property of graphite anodes and suppressing dendritic metallic Li formation, current studies often focus on enhancing certain aspects of material/electrolyte/electrode properties to facilitate electrochemical reactions rather than considering overall electrochemical performance. Regarding the complexity of charge transport and electrochemical reactions of LIBs, comprehensive and in-depth studies should be conducted to realize fast-charging LIBs with high energy density and safety. It is imperative to note that the effects of individual battery components are not isolated. Rather, they exhibit intricate interactions. For instance, there still lacks of complete understanding regarding how the components and structures of SEI affect Li^+ transport properties. The SEI functions as a “ Li^+ conversion station” in terms of its chemical environment, where Li^+ interacts with solvent in electrolyte before entering into SEI and Li^+ migrates within bulk active materials based on the material properties (e.g., ionic conductivity and surface structure). From the aspect of SEI generation, attentions should be paid to the effect of substrates on both nucleation and growth of SEI, as well as its properties. Besides, while numerous studies have been devoted to the effect of the electrolyte on Li^+ desolvation, the impact of SEI is overlooked. Therefore, the constituents of the electrolyte can influence both the composition and structure of the SEI, while the preformed SEI can significantly impact the Li^+ desolvation process occurring at the interface [148]. It is believed that comprehending the above aspects will undoubtedly contribute to advancing SEI design for achieving safer fast-charging LIBs. A synergistic amalgamation of these strategies could potentially yield superior outcomes.

In this review, it is worth pointing out that controllable metallic Li plating, instead of metallic Li dendrites formation, emerges as a promising direction to realize the demands of fast charging and high energy of LIBs. Highly reversible Li plating on graphite anode can assist in constructing a “graphite-Li hybrid” anode and significantly increase the overall specific capacity of the anode while improving the energy density of LIBs during fast charging. Uniform and reversible metallic Li plating with a low accessible surface area and good connection to graphite electrode under fast-charging conditions not only avoids the potential safety hazards caused by short circuits but also reduces the irreversible Li loss and enhances the electrochemical cycling stability. The remaining challenge lies in regulating Li plating on graphite anode. A lithiophilic graphite surface and reserved space for metallic Li deposition are essential, which can be achieved through proper electrolyte design and pre-treatment of anode surface/structure. The use of graphite-Li hybrid electrode could potentially provide an effective solution for fast-charging battery design in the future.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

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