



Challenges and prospects of dead Li⁰ reactivation in lithium metal batteries

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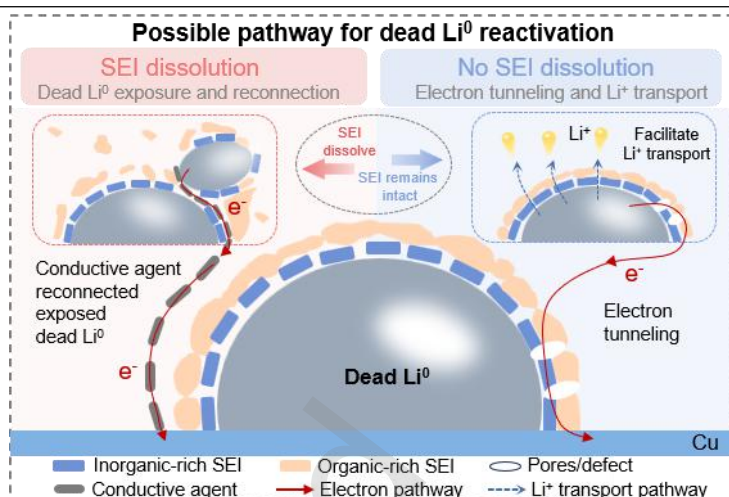
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Efficient reactivation of dead Li⁰ is crucial for achieving long-cycle-life Li metal batteries. By systematically investigating key issues of SEI dissolution, dead Li⁰ exposure, electronic reconnection for the failure of dead Li⁰ reactivation, and by combining with existing dead Li⁰ reactivation strategies, the formation and reactivation mechanism of dead Li⁰ can possibly be revealed, potentially enabling observation of high proportion of dead Li⁰ reactivation.

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ABSTRACT

High-energy-density lithium (Li) metal batteries (LMBs) are promising next-generation energy storage technologies. However, the continuous and irreversible inactive metallic Li (dead Li^0) generation severely limits battery lifespans. Notably, reactivating dead Li^0 in addition to reducing its formation is also crucial for achieving long-life LMBs. Though prototype strategies (*e.g.*, electrochemical protocols, redox chemistry) have achieved partial reactivation of dead Li^0 , their feasibility in practical cells remains limited by the large proportion of dead Li^0 that persists. Directly reconnecting exposed dead Li^0 with electrodes by introducing external electronically conductive materials is a promising solution, yet it fails to achieve the expected reactivation. To achieve efficient reactivation of dead Li^0 , key procedures including Li-stripping-related issues (*e.g.*, mass transport, ion/electron transfer), solid electrolyte interphase (SEI) evolution, and effective dead Li^0 exposure should be concurrently considered. The answer may be obtained through in-depth mechanistic studies using advanced characterization techniques. Additionally, insights into dead Li^0 reactivation can be gained from existing methods (*e.g.*, high cutoff voltage, lower current density) and from detecting environmental changes around the reactivated dead Li^0 to identify favorable conditions. Such recognition of dead Li^0 reactivation promises LMBs with prolonged lifespan, which can further be extended to other battery systems.

KEYWORDS

Li metal batteries, dead Li^0 reactivation, SEI dissolution, electronic reconnection, mass transport

1 Introduction

Lithium (Li) metal batteries (LMBs) have become promising next-generation energy storage technologies owing to the adoption of Li metal anode with a high theoretical energy density (3860 mAh g^{-1}) and low redox potential (-3.04 V versus standard hydrogen electrode) [1-3]. Presently, most LMBs use an excessive amount of Li foil as the anode, while still suffering from inferior cyclability. As for LMBs assembled according to a low capacity ratio of negative to positive electrode (N/P ratio),

they undergo even more severe capacity degradation and limited lifetimes, especially zero-excess LMBs (N/P=0) [4]. Such a dilemma of LMBs is highly related to the continuous Li loss triggered by the intrinsically high reactivity of Li.

Li metal in contact with the electrolyte suffers chemical/electrochemical corrosion to generate an unstable solid-electrolyte interphase (SEI), which undergoes repeated breakage and repair at the expense of gradual exhaustion of both Li and electrolyte. Notably, the cracked SEI exposes fresh Li to the electrolyte, inducing

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continuous Li corrosion. Moreover, the inferior SEI leads to subsequent uncontrollable dendritic Li growth and dead Li accumulation during battery operation [5]. Typically, dead Li (inactive Li) includes residual SEI debris (dead SEI) and metallic Li debris (dead Li⁰) electronically isolated by the SEI, neither of which can directly contribute to battery capacity [6,7]. Of note, dead Li⁰ is a major component of dead Li generated in various systems and conditions. Obviously, uncontrollable Li plating/stripping and Li corrosion are major culprits for producing dead Li [8]. The continuous formation and accumulation of dead Li not only reduce the battery stability but also raise safety hazards. Consequently, it is of great significance to tame the dead Li-related issues to prolong the stable and safe operation of LMBs.

In the early exploration stage of LMBs, researchers focused on reducing dead Li formation mainly by regulating SEI chemistry and controlling Li plating/stripping patterns (*e.g.*, functional skeletons, electrolyte formulations, artificial SEI) [9-12]. However, it has been found that dead Li formation cannot be totally eliminated. Consequently, emerging interest has shifted to reusing or reactivating such inactive anode materials. One such typical strategy is to introduce redox intermediates that can react with both SEI and dead Li⁰ to release Li⁺ into the electrolyte, which can be transported to the charged cathode for restorage and reuse [13]. As for dead Li⁰-dominated dead Li, rebuilding its electronic pathways to the bulk electrode can directly reuse such dead Li, which can be enabled by electrochemical protocols (*e.g.*, resting to dissolve SEI, electric pulse to drive dead Li⁰) [14,15]. In addition, heat treatment and external pressure can also facilitate the reconnection of dead Li⁰ in solid-state LMBs [16].

Though these methods have demonstrated the feasibility of dead Li⁰ reactivation, the coulombic efficiency (CE) still hardly meets the requirements of current application scenarios (at least 99.99%, 1000 stable cycles with more than 90% capacity retention) for long-life LMBs [17]. The inferiority of current dead Li reactivation strategies stems from the fact that efficient and high proportion dead Li⁰ reactivation has not yet been achieved. Next, more efficient dead Li⁰ reactivation strategies, analysis of decisive steps, and intuitive characterization methods to verify dead Li⁰ reactivation are urgently needed. In this perspective, we first elucidate the historic development of LMBs, emphasizing the importance of dead Li reactivation. Subsequently, the mechanisms of dead Li⁰ formation and reactivation strategies are classified and summarized, uncovering the failure of dead Li⁰ reactivation. A high proportion of dead Li⁰ reactivation within the observable field of view is a necessary condition for achieving practical battery reactivation. Based on the concept that electronic insulation induces isolated dead Li⁰ generation, extra Cu fibers were introduced at the interface to enable physical reconnection in Li||Cu cells under in-situ optical

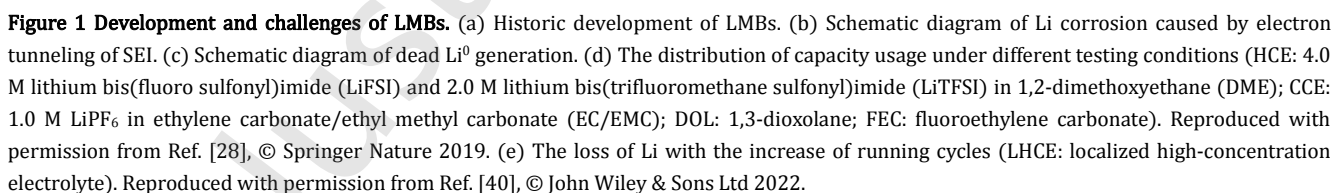
microscopy. However, no dead Li⁰ reactivation behavior was detected by in-situ optical microscopy, a simple and convenient verification technique. Notably, successful dead Li⁰ reactivation was achieved at a decreased current density. Therefore, the mechanisms of dead Li⁰ production and reactivation may need to be re-evaluated. An outlook on the key challenges is provided to address the lack of feasible, efficient dead Li reactivation methods. This work provides a new perspective and approach to reactivating dead Li⁰, ultimately aiming to achieve long-cycle-life LMBs.

2 Historic development and challenges of Li metal batteries

The first LMBs emerged in the 1970s, and were recognized for their high energy density [18,19]. However, their development has stalled due to the fire incidents caused by Li dendrite growth [20]. The subsequent rise of electric vehicles and large-scale energy storage, which require higher energy density, has promoted the revival of LMBs. Notably, Peled et al. proposed the formation of SEI around Li dendrites, a powerful indicator of anode interfacial stability [21,22]. In recent decades, SEI engineering has enabled the mitigation of Li dendrite-induced issues, achieving the safe operation of high-energy-density LMBs [23-25].

In addition to battery safety issues, capacity degradation of LMBs also challenges their practical applications. This problem, due to its unclear mechanism, has long plagued the realization of long-cycle-life LMBs. The emergence of advanced characterization techniques has brought a turning point by attributing capacity degradation to dead Li accumulation, thereby exposing previously neglected issues of Li corrosion and dead Li⁰ formation [26,27]. Meng's group identified dead Li as consisting of Li⁺ compounds in the SEI and electrically isolated unreacted metallic Li⁰ (*i.e.*, dead Li⁰), and quantified its composition using titration gas chromatography (TGC) [28]. The maturity of quantification techniques has deepened the understanding of dead Li formation. Grey et al. quantified dead Li by in-situ nuclear magnetic resonance spectroscopy (NMR) and revealed that Li corrosion is related to the properties of the generated SEI [29]. This finding indicates that both the SEI and dead Li⁰ are closely related to Li corrosion behavior. The problem of Li corrosion has come to the attention of researchers. Cui et al. attributed the fast galvanic corrosion of Li to electron tunneling through the SEI [30]. By cutting off the electron tunneling, Jin et al. constructed an artificial SEI to suppress Li corrosion. In addition, for the unavoidably generated dead Li⁰ as a corrosion product, reactivation strategies have been explored [13-15]. Nevertheless, the CE of LMBs is still inferior, and efficient reactivation of dead Li⁰ remains a challenge. The subsequent emergence of the Cryo-TEM technique has promoted in-depth research on Li deactivation and reactivation [31-33]. For example, Erni et al. employed operando electrochemical

to study Li dendrites. It was found that Li dendrites are strong yet brittle (Fig. 1(a)) [35], which originates from limited dislocation sources and a stiff SEI coating. This discovery reveals the failure mechanism within solid-state electrolyte systems. Therefore, with the help of advanced technologies, it is possible to gain deeper insights into the formation mechanism of dead Li and to achieve its effective reactivation.



As mentioned above, anode-free LMBs, as next-generation, high-safety, and high-energy-density energy storage technology, suffer more severe capacity loss under operating conditions, triggered by SEI evolution and dead Li^0 accumulation [36,37]. In detail, electron tunneling through the native, unstable SEI induces severe Li-copper (Cu) galvanic corrosion, leading to the rupture, regeneration, and eventual thickening of the SEI (Fig. 1(b)) [38]. The resultant thick SEI can isolate metallic Li from the bulk electrode and block its electron availability, resulting in dead Li^0 losing its electrochemical reactivity

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batteries typically contain a relatively high amount of dead Li^0 because the non-uniform, three-dimensional Li structures formed during deposition are prone to electrical disconnection during the stripping process. In solid-state battery systems, the relative proportion of dead Li^0 varies significantly among different electrolytes. Obviously, reactivating dead Li^0 , especially in anode-free battery systems with a high proportion of dead Li^0 , appears to be an effective method to extend the lifespans of LMBs.

To address the electronic disconnection problem of dead Li^0 caused by Li corrosion and SEI failure, researchers have developed several effective reactivation strategies. These reactivation strategies can be categorized into two modes: conversion reactivation and electronic reconnection reactivation. Conversion reactivation typically utilizes intermediates to carry electrons, transforming dead Li into other species capable of re-participating in the charge-discharge process of

batteries. Redox mediators as indirect electron carriers can simultaneously react with dead SEI and dead Li^0 to release Li^+ for reuse (Fig. 2(a)). For instance, Jin et al. applied the I^-/I_3^- redox mediator to reactivate dead Li^0 . In this process, I_3^- captures electrons from dead Li^0 , converting to I^- and releasing free Li^+ , which then migrates to the cathode. In the charged state, I^- releases electrons to the cathode material, where delithiated LiFePO_4 captures Li^+ via a spontaneous lithiation reaction for subsequent reuse [13]. Additionally, a series of redox mediators of SnI_4 , SrI_2 , LiBr , 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO), and 1,4-di-tert-butyl-2,5-dimethoxybenzene (DDB) have been employed to reactivate accumulated dead Li^0 [41-44]. Notably, to prevent severe battery self-discharge and cyclic reactions for reactivating dead Li, the redox potential of the redox mediators must fall precisely between the redox potential of the cathode materials and the charge cut-off voltage of batteries [45].

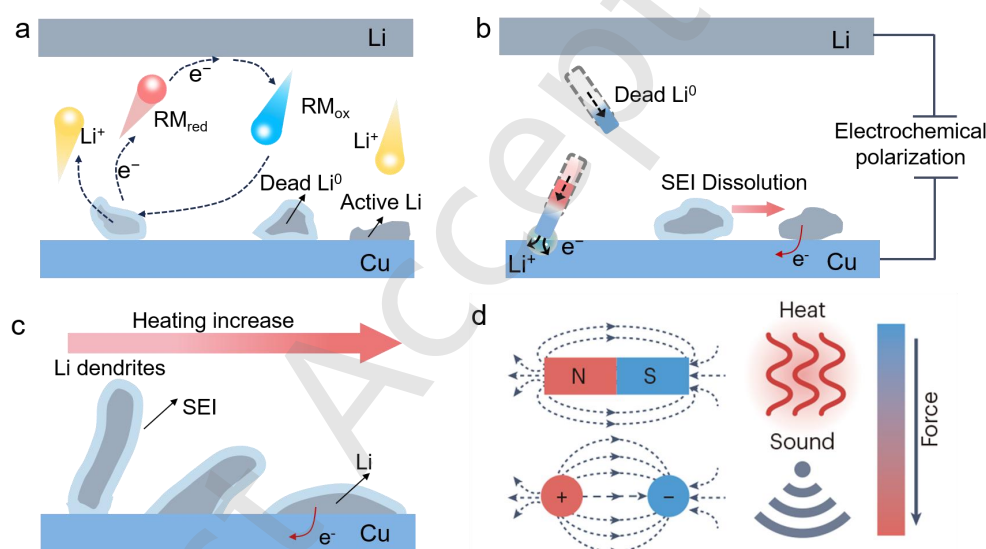


Figure 2 Dead Li^0 reactivation strategies. (a) Redox mediators, (b) Electrochemical polarization drivers, (c) Heating promotion, and (d) Physical fields. Reproduced with permission from Ref. [47], © Springer Nature Limited 2025.

Alternatively, electronic reconnection of dead Li^0 can be achieved through its direct, directional migration toward the current collector, which can be achieved by local electric field modulation, SEI dissolution, and high-temperature melting strategies. Current pulse is a typical local electric field modulation method, in which electrochemical polarization drives the spatial migration of dead Li^0 , allowing internal dead Li^0 to contact the bulk electrode for reactivation (Fig. 2(b)). For example, Cui's group discovered the polarization behavior of dead Li^0 under the built-in electric field of a battery. The dead Li^0 regains electrons by moving and reconnecting with the anode after applying a fast current pulse [14]. Moreover, the voltage pulse achieved capacity recovery when applied to silicon anode batteries [46]. Notably, the pulse current, voltage, and time need to be precisely controlled to

achieve effective reactivation. Furthermore, dissolving the non-conductive SEI surrounding dead Li^0 facilitates electronic connection between the exposed dead Li^0 and the current collector, thereby increasing the probability of reactivating the dead Li^0 . Accordingly, with a battery resting protocol at Li stripping state, the successful SEI dissolution promoted dead Li reactivation to 31.7% at a stripping current density of 5 mA cm^{-2} [15]. The duration of storage and the configuration of batteries have a significant impact on their performance. Last but not least, by leveraging the low melting point of metallic Li, electronic reconnection to the current collector can be achieved through the fluidity of the heated Li. Heating treatment promotes Li melting and creeping, freeing dead Li^0 from the encapsulating electronically insulating SEI, and restoring its electrochemical activity (Fig. 2(c)). Li et

al. discovered that a high current density of 9 mA cm^{-2} causes significant self-heating of the Li dendrites, thereby inducing extensive surface migration and reuse of Li [16]. The implementation of the thermal melting strategies for dead-Li⁰ reactivation requires precise control over key parameters, including the melting temperature, heating duration, temperature uniformity, and the thermal stability of the SEI and electrolyte.

Conclusively, the above dead Li⁰ reactivation strategies have demonstrated efficacy in dead Li⁰ reactivation. However, their limitations cannot be overlooked. Redox mediators not only react with isolated dead Li⁰ but also with the active Li metal anode, leading to serious side effects. Electrochemical protocol reactivation strategies impose stringent implementation conditions, rendering it difficult to strike a balance between reactivation and side reactions of the battery. The Li thermal melting methods involve complex thermal management and are applicable only to certain battery systems. Therefore, developing simpler and more effective

reactivation strategies to reconnect electrons between dead Li⁰ and current collectors has become imperative. Based on these considerations, Jin et al. proposed using potentially feasible physical fields, such as magnetic fields, thermal fields, and frequency modulation, to reactivate dead Li⁰ by re-establishing electronic reconnection (Fig. 2(d)) [47]. Notably, how to achieve and verify dead Li⁰ reactivation using these proposed methods has become an important topic.

4 Methods and limitations of verification methods for dead Li⁰ reactivation

Currently, quantification methods primarily fall into three categories: electrochemical diagnostic techniques (relying on electrical responses), chemical titration and advanced spectroscopic techniques (relying on the signal from dead Li⁰), and imaging techniques (relying on the direct observation of morphology). Generally, successfully reactivated dead Li⁰ can supplement the capacity loss, achieving a CE higher than 100% in a Li || Cu half-cell (Fig. 3(a) and 3(b)) [15]. This electrochemical approach is

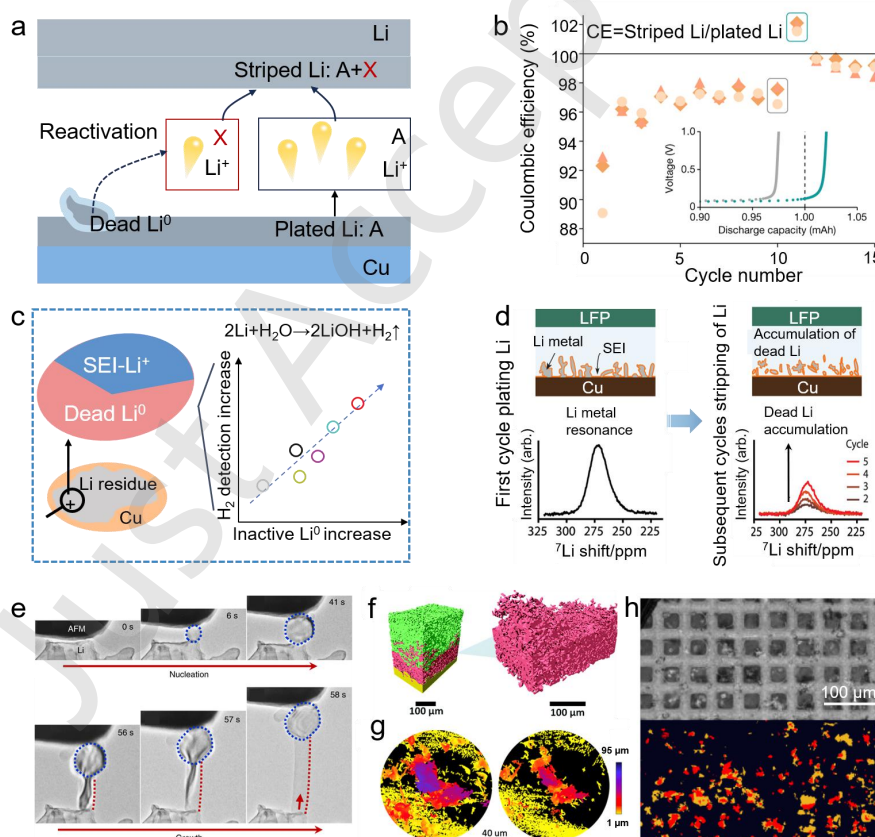


Figure 3 Dead Li reactivation verification methods and techniques. (a) The Li⁺ contribution from dead Li⁰ reactivation. (b) CE results of cells with dead Li⁰ being reactivated. Reproduced with permission from Ref. [15], © Springer Nature Limited 2024. (c) Quantification of dead Li by TGC or T-DEMS. (d) In-situ Li quantification by NMR. Reproduced with permission from Ref. [29], © American Chemical Society 2020. (e) Sequential TEM snapshots of Li nucleation and growth process. Reproduced with permission from Ref. [52], © Springer Nature Limited 2019. (f) 3D renderings after cycling. Green for separator, pink for mossy Li, and yellow for residual Li. Reproduced with permission from Ref. [53], © Royal Society of Chemistry 2017. (g) Close up of dead Li microstructures marked in maps after stripping at 0.5 mA cm^{-2} . The height of Li projected on one pixel ($0.325 \mu\text{m} \times 0.325 \mu\text{m}$) on the Cu surface is visualized by the color scale bar, and the black pixels are bare Cu substrate. Reproduced with permission from Ref. [54], © Springer Nature Limited 2023. (h) The observation of dead Li⁰ evaluation by in-situ optical microscopy. Red for dead Li⁰ and yellow for recovered Li⁰. Reproduced with permission from Ref. [15], © Springer Nature Limited 2024.

applicable to battery systems where a large amount of dead Li^0 is reactivated. However, CE may be less than 100% when the quantity of reactivated dead Li cannot compensate for the Li loss caused by side reactions. Typically, the reactivation of dead Li^0 is accompanied by an increase in capacity, which can be used to verify its reactivation. Inevitably, part of the capacity improvement may originate from SEI evolution or Li corrosion.

Given the limitations of the above electrochemical methods in defining and quantifying dead Li^0 reactivation, quantitative analysis of dead Li^0 becomes essential. Titration gas chromatography (TGC)/titration differential electrochemical mass spectrometry (T-DEMS) can quantify dead Li^0 by measuring H_2 released from the reaction between dead Li^0 and H_2O (Fig. 3(c)) [48-50]. The extent of reactivation is reflected by the decrease in dead Li^0 within the electrode. However, some studies have

shown that LiH in the electrode also participates in the reaction [51]. Alternatively, in-situ NMR can detect dead Li^0 in a specific cycle as well as its accumulation, using ^7Li signal intensity (Fig. 3(d)) [29]. These quantitative methods can effectively confirm the reduction of dead Li^0 during cycling, while it cannot prove that the reduction of dead Li originates from its reactivation. Currently, none of the above verification methods alone can conclusively distinguish whether the observed reduction of dead Li^0 and the increased capacity or CE actually stem from dead Li^0 reactivation, rather than merely from suppressed dead Li^0 formation or Li corrosion. Therefore, different characterization techniques require cross-complementary verification. For instance, by distinguishing the Li^0 content between reactivated and non-reactivated samples and correlating the results with the electrochemical data, the successful reactivation of dead Li^0 can be confirmed.

Table1. Comparison of various techniques for dead Li^0 characterization.

Technique	Target of detection	Evidence for Li^0	Resolution	Quantitative	Application scenario	Major limitations	Evidence for dead Li^0 reactivation
Electrochemical diagnostic techniques	Capacity, CE	No	cell	Qualitative	Non-destructive cell testing	Parasitic reaction contribution	Need TGC/T-DEMS, NMR and optical microscopy
TGC/T-DEMS	Dead Li^0 by detecting H_2	Yes	Macroscopic (bulk)	Quantitative	Final state Li^0 quantification	LiH interference	Need Cryo-TEM/STEM and optical microscopy
^7Li NMR	Li^0 signal intensity	Yes	Macroscopic (bulk)	Semi-quantitative	Dynamic tracking of Li accumulation	Complex signal/quantity calibration	Need TGC/T-DEMS and optical microscopy
Cryo-TEM/STEM	Local morphology	Yes	nm	Qualitative	Microstructural analysis	Selection bias and beam damage	Need TGC/T-DEMS and optical microscopy
XTM/X-ray CT	3D morphology	No	μm -tens of μm	Semi-quantitative	3D structural analysis	Low contrast and limited resolution	Need NMR and TGC/T-DEMS
Optical microscopy	Morphology	No	μm	Qualitative	Dynamic morphological tracking	No chemical information, contrast-dependent	Need electrochemical diagnostic techniques or NMR

In addition, in-situ observation has become an intuitive method for directly verifying dead Li^0 reactivation. In-situ transmission electron microscopy (in-situ TEM) enables the observation of Li dendrite growth and evolution, which is an efficient technique for monitoring dead Li^0 reactivation (Fig. 3(e)) [52]. However, this technique is limited to observing local micron-sized regions. Furthermore, the difficulty in achieving multiple conditions for dead Li^0 reactivation restricts its application. X-ray tomographic microscopy (XTM) is a powerful analytical tool capable of non-destructive three-dimensional imaging of local material morphology and composition over large volumes with sub-micron spatial resolution (Fig. 3(f)) [53]. Nevertheless, due to the

poor contrast of Li metal, its microstructural details are difficult to distinguish from the surrounding environments. Operando synchrotron XTM technology significantly improves the contrast between Li and its surrounding environments (Fig. 3(g)) [54]. The application of these techniques is limited by the high cost of the required equipment. In-situ optical microscopy is an intuitive method for tracking the evolution of dead Li^0 , and has recently been adopted in the field of dead Li^0 reactivation owing to its macro-level online monitoring capability. The in-situ observation, accompanied by electrochemical changes (such as a CE higher than 100% and battery capacity increase—which are reproducible, whereas side reactions do not necessarily follow a regular pattern) or

NMR intensity variation (the quantification of dead Li^0), could be an efficient method to verify dead Li^0 reactivation. For example, Cui et al. demonstrated the dead Li^0 reactivation behavior by tracking and comparing the distribution of dead Li^0 before and after reactivation, and the CE exceeding 100% during the additional resting period (Fig. 3(h)) [15]. The advantages and limitations of different testing methods for dead Li^0 characterization are summarized in Table 1. Given the advantages in morphological tracking over a large field of view, in-situ optical microscopy is employed in our visualization study of dead Li^0 reactivation.

5. Visualization observation of dead Li^0 reactivation by electronic reconnection

The exploration of dead Li^0 reactivation was designed to be visualized using in-situ optical microscopy. Firstly, a Li || Cu mesh cell composed of a Li metal anode, a Cu mesh cathode and LiPF_6 in DMC/FEC (v:v=4:1) electrolyte was assembled in a specific cell setup. After Li || Cu cells underwent plating and subsequent stripping, dead Li^0

(Region I : the silver parts) and SEI (Region II : the black parts) were observed on the Cu mesh (Fig. 4(a)). The residual dead Li^0 was composed of withered, dendritic metallic Li and was covered with SEI on its surface (Fig. 4(b)). In contrast, the residual SEI consisted primarily of thick corrosion products (nanocrystalline LiF and Li_2O in an amorphous polymeric matrix) and tiny dead Li^0 fragments (Fig. 4(c)) [13,55]. To eliminate the influence of insulating SEI on dead Li^0 reactivation, the Li || Cu mesh cell was rested for 24 hours in the Li stripped state (*i.e.*, charge state) to eliminate or at least partially remove the thick SEI. After this resting period, the black SEI aggregates disappeared due to dissolution, exposing dead Li^0 (Fig. 4(d)). It is generally believed that organic components in SEI have high solubility, while inorganic components are retained due to their lower solubility [13,15,56,57]. Consequently, the dead Li^0 became covered with a thin inorganic-rich SEI (Fig. 4(e)), while the SEI aggregates left behind free-moving tiny dead Li^0 fragments covered with a thin SEI (Fig. 4(f)). The exposure of dead Li^0 provided the necessary conditions for its subsequent reactivation.

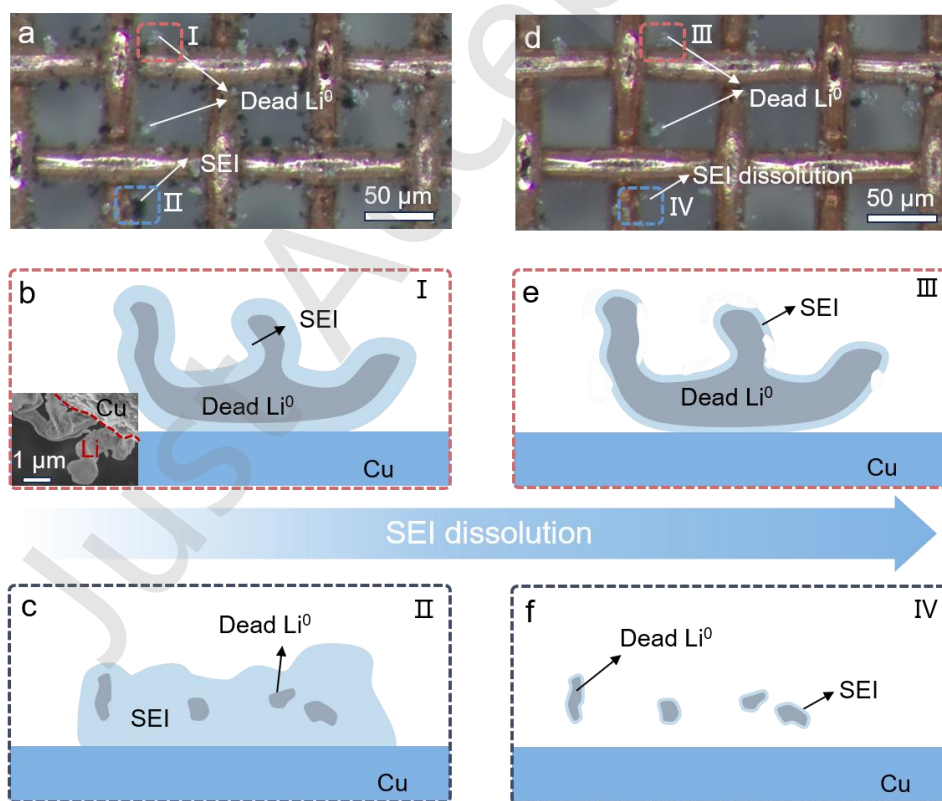


Figure 4 SEI and dead Li^0 evolution during battery resting. (a) The dead Li^0 (I) and SEI (II) distribution after 1 cycle of Li || Cu cell, and the corresponding structural diagram of (b) dead Li^0 (the inset is the corresponding SEM image) and (c) SEI. (d) The dead Li^0 (III) and SEI (IV) evolution after SEI dissolution during Li || Cu cell rest at charge end state, and the corresponding structural diagram of dissolved (e) dead Li^0 and (f) SEI.

Afterwards, an in-situ cell was assembled using the Cu mesh with exposed dead Li^0 and a Li metal anode. Taking into account the uncontrollable effects of external fields, such as magnetic, electric, and thermal fields, on the

battery, a more straightforward and effective electronic connection between the dead Li^0 and the current collector was achieved by embedding several extra Cu fibers on the surface of the Cu mesh (Fig. 5(a)). The cell was charged at

a constant current density with a cutoff voltage of 3.0 V. It was expected that the extra Cu fibers would connect dead Li^0 to the current collector (Fig. 5(b)), establishing electrical reconnection to enable dead Li^0 reactivation. However, no dead Li^0 reactivation was observed at the positions connected to the Cu fibers (Fig. 5(c)). Unexpectedly, dead Li^0 was reactivated at positions without the extra Cu fibers (Fig. 5(d)). According to the theory of dead Li^0 formation, this successful reactivation

indicates reconnection of the electronic pathway under high overpotential (Fig. 5(e)). This phenomenon prompts us to consider why direct physical reconnection cannot establish an electronic pathway, whether SEI dissolution is a necessary prerequisite for dead Li^0 reactivation, what the characteristics of the SEI are after its dissolution, and what the features of the electronic reconnection locations are.

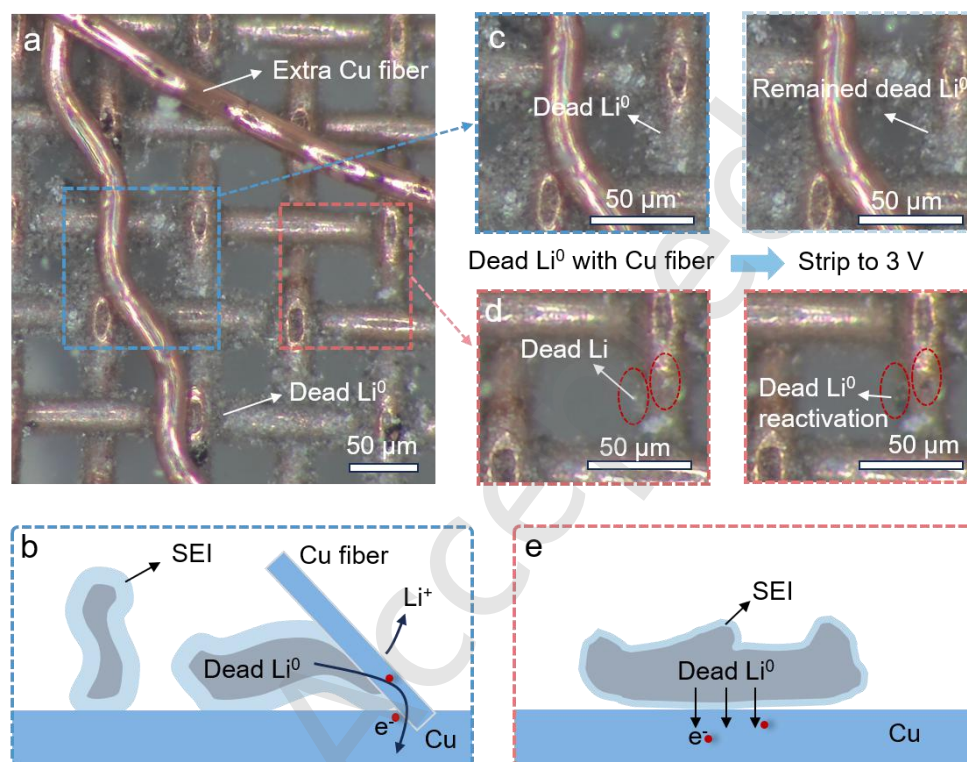


Figure 5 Reactivation of exposed dead Li^0 with externally introduced Cu fibers. (a) Dead Li^0 distribution and the introduced extra Cu fibers on the surface of Cu mesh in the $\text{Li} \parallel \text{Cu}$ mesh cell with exposed dead Li^0 . (b) Schematic diagram for the failure of dead Li^0 reactivation with the extra Cu fiber. Dead Li^0 evolution at the positions of (c) beside the extra Cu fiber and (d) without extra Cu fiber after applying the reactivation condition. (e) The success of dead Li^0 reactivation without extra Cu fiber.

In order to explore the above confusions, dead Li^0 reactivation was investigated under conditions without additional SEI dissolution. When the Li stripping current density decreased from 2.0 mA cm^{-2} to 0.1 mA cm^{-2} , an additional capacity of $10.6 \mu\text{Ah}$ was achieved, and some dead Li^0 (inside the red circles) was observed to be reactivated (Fig. 6(a, b)). Under these low-current-density reactivation conditions, the SEI can be considered unchanged, while the polarization was lower. Therefore, the additional Li stripping reaction was attributed to sufficient time for ion transport (Fig. 6(c)). The result indicates that dead Li^0 reactivation may not be limited solely by electronic disconnection, but is also influenced by the kinetics of ion transport.

Similarly, when the cutoff voltage was increased from 1.2 V to 3.0 V, another $6.9 \mu\text{Ah}$ of Li was stripped from the Cu mesh electrode (Fig. 6(d)), and partial reactivation of dead Li^0 was observed (Fig. 6(e)). The high reaction overpotential accelerated ion transport through an

increased concentration difference and fast electron transfer kinetics (Fig. 6(f)). Using the above electrochemical protocols, partial dead Li^0 reactivation was achieved not by modifying the native SEI, but solely by controlling the battery operating conditions (*e.g.*, current density and cutoff voltage) to regulate the reaction balance. Notably, high proportion of dead Li^0 reactivation has not been observed using in-situ optical microscopy in our research, even when the above conditions for dead Li^0 reactivation were applied. The evolution of SEI and interfacial electron/ion transport may lead to different dead Li^0 reactivation mechanisms under various conditions. Achieving dead Li^0 reactivation through physical reconnection may not be as straightforward as we initially proposed. Exploring dead Li^0 formation, evaluating the conditions for its reactivation, and establishing the underlying mechanisms are essential for the success of effective dead Li^0 reactivation.

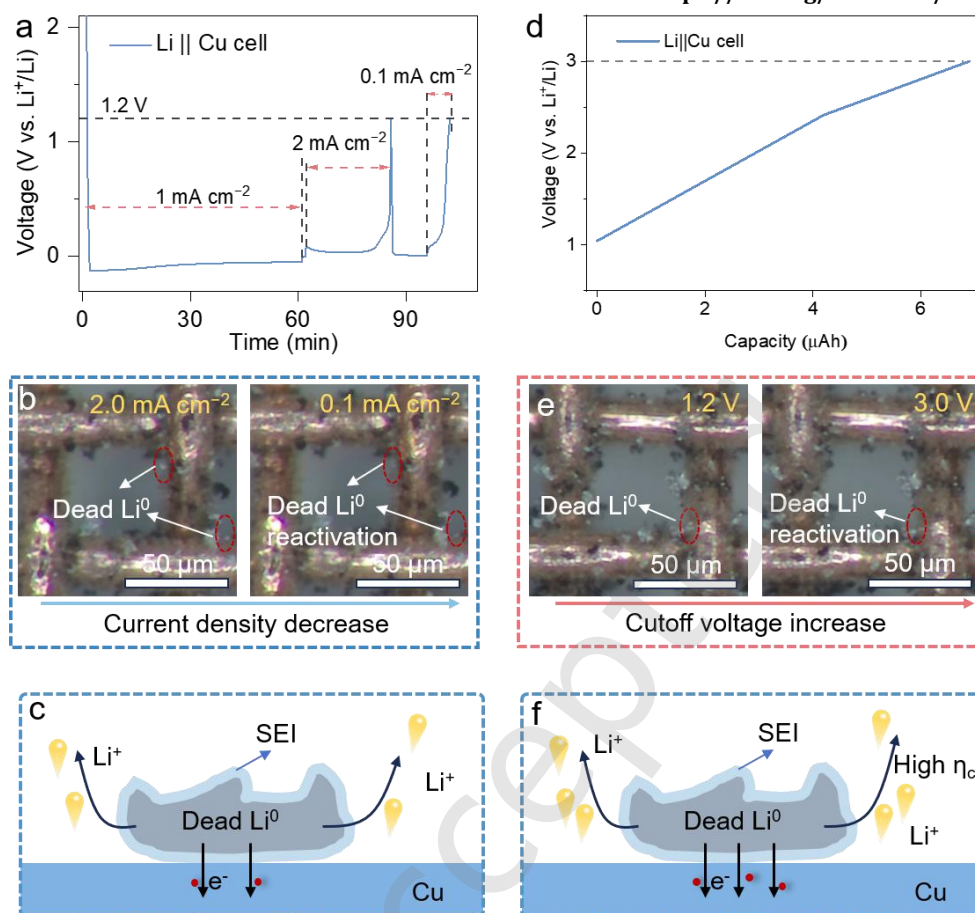


Figure 6 Reactivation of dead Li^0 under charge/discharge protocol regulation. (a) The discharge/charge plots of Li || Cu cells at decreased stripping current densities. (b) The dead Li^0 reactivation observed by in-situ optical microscopy with the current density decreasing from 2.0 mA cm^{-2} to 0.1 mA cm^{-2} , and (c) its corresponding dead Li^0 reactivation schematic diagram. (d) The charging voltage profile of Li || Cu cells. (e) The dead Li^0 reactivation observed by in-situ optical microscopy with the cutoff voltage increasing from 1.2 V to 3.0 V, and (f) its corresponding dead Li^0 reactivation schematic diagram.

6. Issues to be explored for dead Li^0 reactivation

The production and accumulation of dead Li^0 severely degrade the cycle life of Li metal batteries, especially under extreme working conditions (*e.g.*, high rate and low temperature). Several dead Li^0 reactivation strategies have been developed and validated by various characterization techniques. However, a large proportion of dead Li^0 was observed even after applying these reactivation methods. In particular, dead Li^0 reactivation through physical reconnection still lacks a practically feasible method. To achieve efficient dead Li^0 reactivation, the following issues should be considered (Fig. 7).

(1) Whether dead Li^0 becomes exposed after SEI dissolution remains to be clarified. The initially formed dead Li^0 is covered with SEI (Fig. 7(a)). For SEI-dissolved dead Li^0 , we added Cu fibers onto its surface to construct electronic pathways. However, after applying a reactivation command, the dead Li^0 around Cu fibers did not exhibit reactivation behavior (*i.e.*, the dead Li^0 particles did not become smaller or disappear). The

reason may be that the dead Li^0 , despite SEI dissolution, is not fully exposed but remains surrounded by invisible components (Fig. 7(b), left column). To verify this hypothesis, the dissolved and remaining components of the SEI should be identified. First, the chemical changes induced by the SEI dissolution can be detected using X-ray photoelectron spectroscopy (XPS), energy dispersive X-ray spectroscopy (EDS) mapping to confirm the elemental composition of the interface after the removal of soluble SEI components, or Fourier transform infrared (FTIR) spectroscopy to verify changes in the elemental content of the dissolution medium. The primary potential components undergoing dissolution (*e.g.*, organic or inorganic species) can be inferred by analyzing the elemental content changes before and after SEI dissolution. In addition, analyzing the chemical environment surrounding dead Li^0 with other visualization techniques is another feasible approach for investigating this issue. For example, Raman imaging could provide information on dissolved components by monitoring specific functional groups around the dead Li^0 .

(2) Li corrosion has a non-negligible effect on the

reactivation of dead Li^0 . When dead Li^0 is exposed by SEI dissolution, the exposed thermodynamically unstable dead Li^0 corrodes with the electrolyte to generate new SEI. The SEI dissolution and regeneration occur simultaneously. Consequently, the relative rates of SEI dissolution and regeneration are crucial to achieving dead Li^0 exposure. Dead Li^0 will be difficult to expose under conditions favoring faster SEI regeneration (Fig. 7(b), right column). Moreover, the effects of the applied dead Li^0 reactivation conditions on Li corrosion and battery cycle life should also be given sufficient attention. The implementation of certain reactivation conditions enables the effective release of inactive Li sources. However, this may be achieved at the cost of damaging the stable SEI or increasing Li corrosion (*e.g.*, the construction of electronic pathways accelerates galvanic corrosion), which is detrimental to the long-term cycle stability of the battery. Notably, the implementation of dead Li^0 reactivation strategies must be accompanied by an evaluation of the battery's corrosion behavior.

(3) Whether the electronic pathway has truly been established at the interphase of dead Li^0 remains a question. Theoretically, the construction of an electronic pathway on exposed dead Li^0 can be achieved by

introducing a conductive substrate or additives (*e.g.*, extra Cu fibers), or by re-plating Li. However, no method has yet been found to reactivate dead Li^0 in large proportions, leading us to consider whether these methods actually construct a conductive electronic pathway or merely create a visual illusion (Fig. 7(c)). This necessitates in-depth exploration and the development of targeted, advanced characterization methods.

Electrochemical response may be a simple and effective method to verify whether an electronic pathway has been constructed. First, symmetric cells are assembled using the target electrodes. Once the electronic pathway is constructed, the current density measured by zero resistance amperometry (ZRA) is expected to increase, while the impedance obtained from electrochemical impedance spectroscopy (EIS) correspondingly decreases. Moreover, conductive atomic force microscopy (C-AFM) can acquire current distribution maps at the nanometer or even atomic level on the sample surface, visually displaying the surface conductivity of the materials and their spatial variation [58]. Therefore, C-AFM should be a powerful technique for further detecting the electronic reconstruction of dead Li^0 .

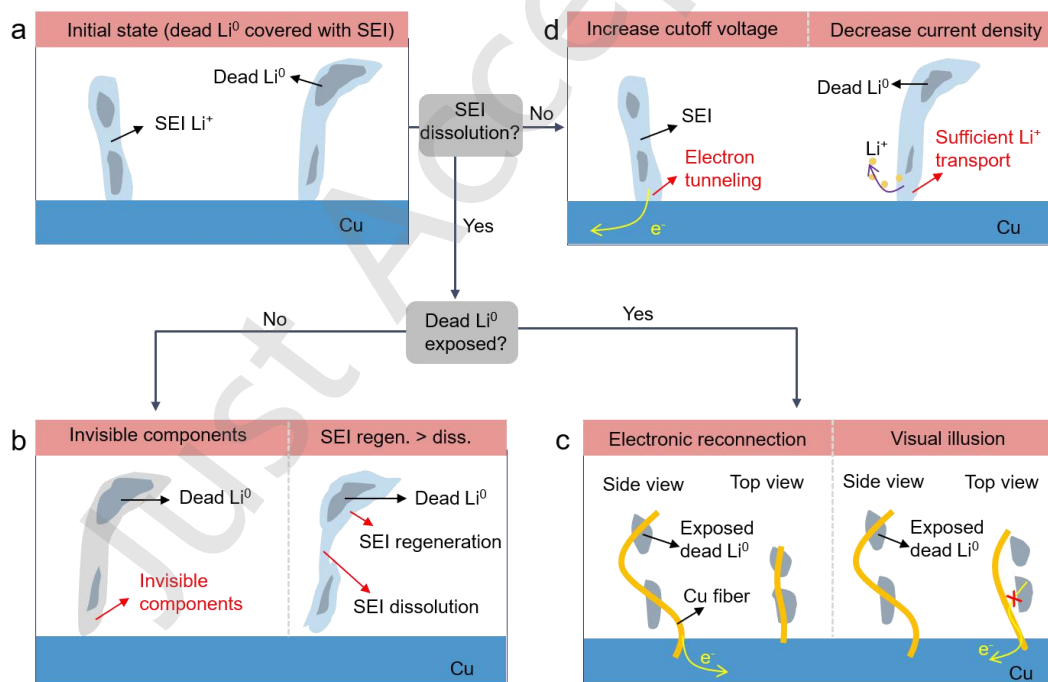


Figure 7 Concerns regarding dead Li^0 reactivation. (a) The initial structural composition of dead Li^0 and SEI. (b) Possible factors for unexposed dead Li^0 . (c) Visual illusion of electronic reconnection for the failure of dead Li^0 reactivation. (d) Dead Li^0 reactivation under the absence of SEI dissolution conditions.

(4) Whether dead Li^0 is generated only by electronic disconnection also remains to be uncovered. Existing reactivation methods serve as a starting point for dead Li^0 reactivation research. Investigating the characteristics of successful dead Li^0 reactivation sites may provide a breakthrough in identifying the key factors governing dead Li^0 reactivation. As described in our study, when the

battery cutoff voltage increases or the current density decreases, the amount of dead Li^0 remaining on the electrode further diminishes, which can effectively be considered a method for achieving dead Li^0 reactivation. This phenomenon raises several points for consideration. Dead Li^0 reactivation still occurs even without SEI dissolution. Certain sites are more favorable for

reactivation, and these sites share common characteristics that need to be identified. Considering these issues, we hypothesize that, in addition to electronic disconnection, mass transport limitations at the interface also play an important role in dead Li^0 formation (Fig. 7(d)). To address these questions, exploring the reactivation mechanism of dead Li^0 and identifying the common characteristics under different conditions may represent an effective strategy to overcome the barrier to dead Li^0 reactivation.

(5) All-solid-state electrolyte battery systems present a more complex challenge in terms of dead Li^0 production and reactivation. In contrast to liquid battery systems, the non-flowable solid electrolyte causes Li to detach from the anode/electrolyte interphase during stripping. This gradually creates nanometer- to submicron-sized holes, leading to complete loss of physical contact with the current collector and conversion to inactive dead Li^0 [59,60]. Furthermore, under internal battery pressure, Li undergoes mechanical breakage because of stress concentration at grain boundaries and defects [61]. In addition, side reactions at the interphase form a passivation layer that encapsulates the remaining metallic Li [62]. Therefore, the dead Li^0 in all-solid-state Li batteries is mainly located at the interphase and within the electrolyte. Because some dead Li^0 in solid-state battery systems is dispersed within the solid electrolyte, controlling the dead Li^0 interphase for reactivation is more difficult. Moreover, in-situ observation and quantification are challenging in solid-state systems, making it difficult to investigate the reactivation mechanism of dead Li^0 . Restoring electron/ion transport, such as designing self-healing reactivation strategies that incorporate dead Li^0 as a dynamic component into cycle management, holds promise for addressing dead Li^0 trapped within the solid electrolytes.

(6) The behavior of dead Li in practical batteries is more complex than that of in-situ optical cells. Extrapolating findings from optical cells to multilayer batteries presents significant challenges due to the coupling of various scale-dependent variables, such as electrode area, areal capacity, stack thickness and layer count, N/P ratio, electrolyte/capacity ratio, cell pressure, configuration, temperature, and the spatial distribution of inactive Li. Consequently, extrapolation of research findings must account for the spatial distribution of these variables and their evolution relative to cell structure. Furthermore, observations based on optical Li||Cu-mesh cells cannot accurately reflect the reactivation of dead Li^0 in intact, lean-electrolyte, sealed full cells. This is because the low electrolyte volume, constrained N/P ratio, and pressure gradients within the multi-layer stacked structure of practical full cells impose fundamental limitations on mediator transport, ion accessibility, and the distribution of inactive Li.

Implementing dead Li^0 reactivation strategies in practical batteries imposes more stringent requirements.

For instance, redox mediator strategies need to ensure effective intermediate transport and interfacial compatibility while minimizing parasitic reactions and spatial non-uniformity. For electron reconnection strategies, it is essential to establish durable electron transport pathways between dead Li^0 and the current collector or conductive framework, while ensuring good ionic accessibility and avoiding further interfacial damage. Transitioning dead Li^0 reactivation technology from proof-of-concept in model systems to practical battery applications requires both implementable reactivation schemes and reliable operando validation methods. Bridging this gap will require the development of in-situ characterization techniques compatible with practical cell formats (*e.g.*, operando NMR for sealed pouch cells, operando X-ray tomography for multilayer stacks) and validation of reactivation strategies in full cells under industry-relevant parameters.

7. Outlook of possible factors influencing the dead Li^0 reactivation

The production and accumulation of dead Li^0 during the degradation of Li metal batteries have attracted increasing research attention. Several dead Li^0 reactivation strategies have been developed. However, the CE still does not reach a sufficiently high level (above 99.99%) to enable long battery lifespan, especially for anode-free batteries. From the observation of the Li metal anode interphase, a large proportion of dead Li^0 remains even after applying reactivation strategies. Based on the current understanding of dead Li^0 formation (*i.e.*, electronic disconnection), a physical reconnection strategy for dead Li^0 reactivation was designed. To address the challenges of dead Li^0 reactivation, the mechanism of dead Li^0 generation and reactivation may need to be reconsidered, and the following factors could serve as entry points for future investigations.

(1) Understanding the underlying mechanisms of dead Li^0 formation. The essence of dead Li^0 production lies in the residual reactants resulting from incomplete reactions (*i.e.*, low conversion rate). Therefore, returning to fundamental research on basic chemical reactions and exploring the thermodynamic and kinetic origins are key to identifying the root causes of dead Li^0 formation, with a particular focus on ion/electron transport behavior at the interface. The observation that dead Li^0 reactivation occurs at decreased current density suggests that dead Li^0 production may not be limited solely by electronic connection, but may also be significantly affected by mass (ions) transport. Identifying the rate-limiting step in the dead Li^0 remaining state and decoupling electron and ion transport in its formation are essential for achieving efficient dead Li^0 reactivation in the future.

(2) Monitoring the relationship between SEI evolution and dead Li^0 . Regardless of whether dead Li^0 production originates from mass transport or electron transfer, both ions and electrons must pass through the

SEI. Consequently, dead Li^0 production and reactivation are tightly linked to SEI behavior. Batteries are most often in a resting state, during which SEI dissolution, reconstruction, and corrosion occur. Understanding how ion and electron transport during resting periods affects CE can guide the construction of an appropriate SEI. Notably, the requirements for dead Li^0 reactivation and for suppressing Li corrosion are contradictory. Specifically, dead Li^0 reactivation requires ion and electron transport, whereas inhibiting Li corrosion requires interruption of the circuit. Therefore, constructing a precisely regulated SEI may offer a potential solution.

(3) Developing effective strategies to distinguish between dead Li^0 reactivation and parasitic reactions is crucial. Parasitic reactions are inherently present in the complex battery environment, and currently, no single characterization technique can unambiguously confirm dead Li^0 reactivation. Therefore, a combined approach involving multiple characterization methods is essential to establish that the amount of dead Li^0 has genuinely decreased, the observed capacity increase originates from the reduction of Li^0 , and this additional capacity is truly reversible. Only through such comprehensive validation can the effective reactivation of dead Li^0 be conclusively demonstrated. Notably, isotopic labeling of dead Li^0 , combined with the detection of isotopically labeled newly formed Li deposits, represents a particularly powerful strategy. This approach can serve as a robust means to verify that dead Li^0 has genuinely re-entered the electrochemical cycle, and may offer a promising route to differentiate reactivation from parasitic side reactions.

(4) Artificial intelligence (AI) and machine learning may provide complementary tools for Li^0 reactivation. For instance, density functional theory (DFT) calculations combined with high-throughput virtual screening can narrow the range of candidate molecules by evaluating parameters of reaction window, side reactions, decomposition products, solubility, and stability [63]. Similarly, AI may be used for screening suitable redox mediators to minimize side reactions during dead Li reactivation. In addition, reinforcement learning may be employed to optimize electrochemical pulse sequences for establishing effective electrical reconnection of dead Li^0 . Notably, its effectiveness critically depends on the definition and reliability of the reward function. Therefore, the successful application of AI to dead Li^0 reactivation ultimately relies on a well-established understanding of the underlying reactivation mechanism and, more importantly, on reliable criteria for quantitatively distinguishing genuine Li^0 recovery from parasitic Li consumption. Once these fundamental issues are resolved, AI could serve as a powerful tool for accelerating the discovery of reactivation agents and optimizing operating protocols, thereby facilitating the development of practical dead Li^0 reactivation strategies.

It is anticipated that, with deeper understanding and further investigation into the formation and reactivation

mechanisms of dead Li^0 , its generation and accumulation will be significantly mitigated. However, these reactivation technologies are still at the laboratory stage, and their feasibility and effectiveness in practical battery applications require further verification. Furthermore, reactivating dead Li (Li^+) within the SEI remains a major challenge, primarily due to the chemically stable state of Li^+ in the SEI and the severe side reactions triggered by disruption of interfacial stability during the reactivation process. Notably, for battery systems in which the SEI constitutes the primary component of dead Li, eliminating the unstable dynamic evolution and accumulation of the SEI represents an effective strategy for achieving a sufficiently high CE for long-lifespan LMBs.

8. Conclusion

In conclusion, the Li metal anode interface after prolonged battery operation generates a large amount of inactive dead Li^0 , which greatly deteriorates the battery capacity, lifespan, and safety. Therefore, achieving dead Li^0 reactivation is crucial for the commercial applications of these batteries. Identifying the necessary conditions for dead Li^0 reactivation has become a key research focus. Experimental results show that direct physical electronic reconnection does not reactivate dead Li^0 . The formation of dead Li^0 is not only related to electron transport, but is also closely associated with mass transport. Studying the compositional evolution of SEI and dead Li^0 during SEI dissolution, exploring the characteristics of dead Li^0 reactivation under existing reactivation conditions, and leveraging AI assistance are expected to become effective strategies for achieving dead Li^0 reactivation. By investigating the production mechanisms and feasible reactivation strategies for dead Li^0 , a more stable and safer Li metal battery is expected to be realized. Furthermore, the exploration of dead Li^0 reactivation is also of great significance for Li-ion batteries and other alkali metal battery families.

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Declaration of conflicting interests

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

Data availability

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

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