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AI-driven closed-loop control of lithium plating in batteries

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Abstract

Lithium plating remains a critical bottleneck for battery safety and longevity, yet existing diagnostics lack closed-loop intervention under real operation. We introduce AI-LOMAC, an AI-driven *operando* measurement–analysis–control framework that achieves real-time, early-state-responsive regulation. Unlike threshold-based monitoring, AI-LOMAC integrates dynamic impedance spectroscopy with shape-adaptive algorithms and a vision-language model to capture interfacial signatures, enabling reliable detection of lithium plating onset >5 min before visible dendrites and autonomous mitigation. This AI-essential architecture enables proactive intervention, where early action preserves lithium reversibility by promoting re-intercalation, extending cycle life and improving capacity retention under fast charging. Validated across coin cells, Ah-scale pouch cells, and 314 Ah prismatic batteries, the framework demonstrates robust scalability to industrial-relevant formats. By establishing a closed-loop paradigm in which AI bridges transient signals to controls,

AI-LOMAC provides a generalizable strategy to enhance battery safety, extend cycle life, and adaptively manage degradation across Li-ion and other electrochemical energy systems.

Main

Lithium-ion batteries (LIBs) dominate the rechargeable energy-storage market, yet safety concerns remain a major barrier to their broader deployment, as highlighted by the continued occurrence of catastrophic failures. Although thermal runaway is the immediate cause of these events, lithium plating is widely recognized as an early initiating factor in the failure cascade¹. Lithium plating commonly arises under fast-charging, low-temperature, or overcharging conditions²⁻⁵, and its progression can drive localized dendritic growth, internal short circuits, and ultimately, severe thermal runaway hazards⁶⁻⁸. Accordingly, reliable detection of lithium plating has become a major research priority⁹⁻¹⁵. Existing diagnostic strategies generally fall into three categories: *ex situ* methods, such as scanning electron microscopy (SEM)¹¹, X-ray photoelectron spectroscopy (XPS)¹², and time-of-flight secondary ion mass spectrometry (TOF-SIMS)¹³; *in-situ/operando* physical methods, including ultrasonic imaging¹⁴, neutron diffraction¹⁶, nuclear magnetic resonance¹⁷, electron paramagnetic resonance¹⁸, and X-ray computed tomography¹⁵; and conventional electrochemical methods, such as voltage relaxation analysis¹⁹, incremental capacity analysis²⁰, and the three-electrode measurements²¹. However, current lithium-plating diagnostics still remain limited by two major challenges. The first is insufficient sensitivity at the earliest stage of lithium deposition. Most methods cannot directly quantify plated lithium, and only a few advanced techniques, including titration mass spectrometry (TMS), titration gas chromatography, and nuclear magnetic resonance, can quantitatively detect residual metallic lithium or dead lithium, typically at the level of tens to hundreds of nanomoles²²⁻²⁴. As a result, lithium plating is often recognized only after macroscopic voltage anomalies emerge, when intervention is already too late for effective prevention²⁵. Recent high-throughput quantification studies have further shown that irreversible lithium plating in graphite-based full cells depends strongly on charge rate, temperature, SOC, and electrolyte chemistry, underscoring the need for sensitive and scalable plating diagnostics²⁶. By contrast, dynamic electrochemical impedance spectroscopy (DEIS) enables

highly sensitive, real-time tracking of rapid interfacial changes under operating conditions²⁷⁻²⁹. DEIS has resolved different stages of lithium plating across charging rates³⁰⁻³², and single-frequency DEIS further improves detection efficiency and sensitivity³³⁻³⁵. We have also established an *operando* quantitative method based on dynamic surface capacitance (C_s), for tracking lithium-plating onset in working LIBs, but still lack of active control to form a closed-loop, especially for long cycling operation³³.

The second challenge is the absence of a closed-loop system integrating *operando* detection, online analysis and real-time active control for early-stage lithium plating. Early-stage plated lithium can be re-intercalated into the anode, offering a route to preserve battery safety³¹, yet such timely intervention has not been realized. This gap points to the need for next-generation battery management architectures, often conceptualized as BMS 2.0, in which battery management shifts from fixed threshold-based monitoring of external variables to *operando*-signal-guided, data-driven control of internal electrochemical states. Such intervention, however, requires an analysis layer capable of distinguishing plating-associated transitions from baseline drift and local fluctuations in *operando* signals. This remains challenging under practical operating conditions, because signal morphology can vary across cells and cycles and may not be described reliably by a single predefined analysis rule. Recent work on dynamic cycling has shown that realistic current profiles and explainable machine learning can reveal degradation behaviour that is not captured by conventional constant-current protocols, reinforcing the importance of data-driven battery management under practical operating conditions³⁶. So far, AI-enabled closed-loop battery frameworks have mainly focused on lifetime prediction and state-of-health management³⁷⁻⁴⁰, whereas real-time control informed by transient microscopic states remains largely unexplored. Promisingly, an impedance-modelling work has achieved online lithium-plating detection using a Butler–Volmer-based model combined with an extended Kalman filter recently, although the detected onset showed a delay of up to 5% state of charge (SOC)³⁴. Yu et al. recently developed a machine learning-based virtual reference electrode that predicts negative-electrode potential in LFP||Gr coin cells and enables adaptive current regulation, extending cycle life by 8.25-fold⁴¹. However, the complexity and variability of real-

world battery operation across cell formats, chemistries, ageing states and operating conditions pose substantial challenges to the transferability of such learned mappings beyond their original training domain. These studies indicate that data-driven and model-based methods can improve online battery diagnostics, but their translation into deterministic, auditable control of transient internal states still remains great challenges.

Here we introduce an AI-driven *operando* measurement-analysis-control framework (AI-LOMAC, Fig. 1) for real-time detection and suppression of early-stage lithium plating in LIBs. By coupling *operando* DEIS with online analysis and real-time active control, AI-LOMAC tracks the evolution of C_s and identifies the characteristic knee point associated with plating onset. The system then autonomously switches to a current-attenuation sequence to suppress dendrite growth and improve interfacial reversibility, which can work 24/7 without interruption. We validate this framework in commercial LCO||Gr coin cells, laboratory LFP||Gr coin cells, commercial 3.5 Ah LFP||Gr pouch cells under fast charging and 314 Ah LFP||Gr prismatic cells under low-temperature charging. These results establish C_s as both a real-time diagnostic signal and a feedback variable for current control, highlighting the advantage of AI-driven state-responsive feedback over pre-programmed charging limits and enabling battery management to shift from retrospective failure detection to proactive degradation mitigation.

Commercial LCO||Gr coin cells with AI-LOMAC

As illustrated in Fig. 1a, AI-LOMAC is built on an *operando* measurement–analysis–control workflow comprising four coordinated modules, for the batteries under operation (module I). In this architecture, *operando* DEIS acquisition (module II), online analysis and feature extraction (module III), and real-time active control (module IV) are performed continuously within the same uninterrupted electrochemical cycle. The workflow first starts from the *operando* measurement module. After initialization, the battery enters the primary fast-charge step, during which the measurement module periodically acquires DEIS data (Fig. 1b, step i). These measurements provide minimally perturbative *operando* access to the evolving interfacial state of the graphite anode. The characteristic frequency used for C_s extraction was identified from impedance

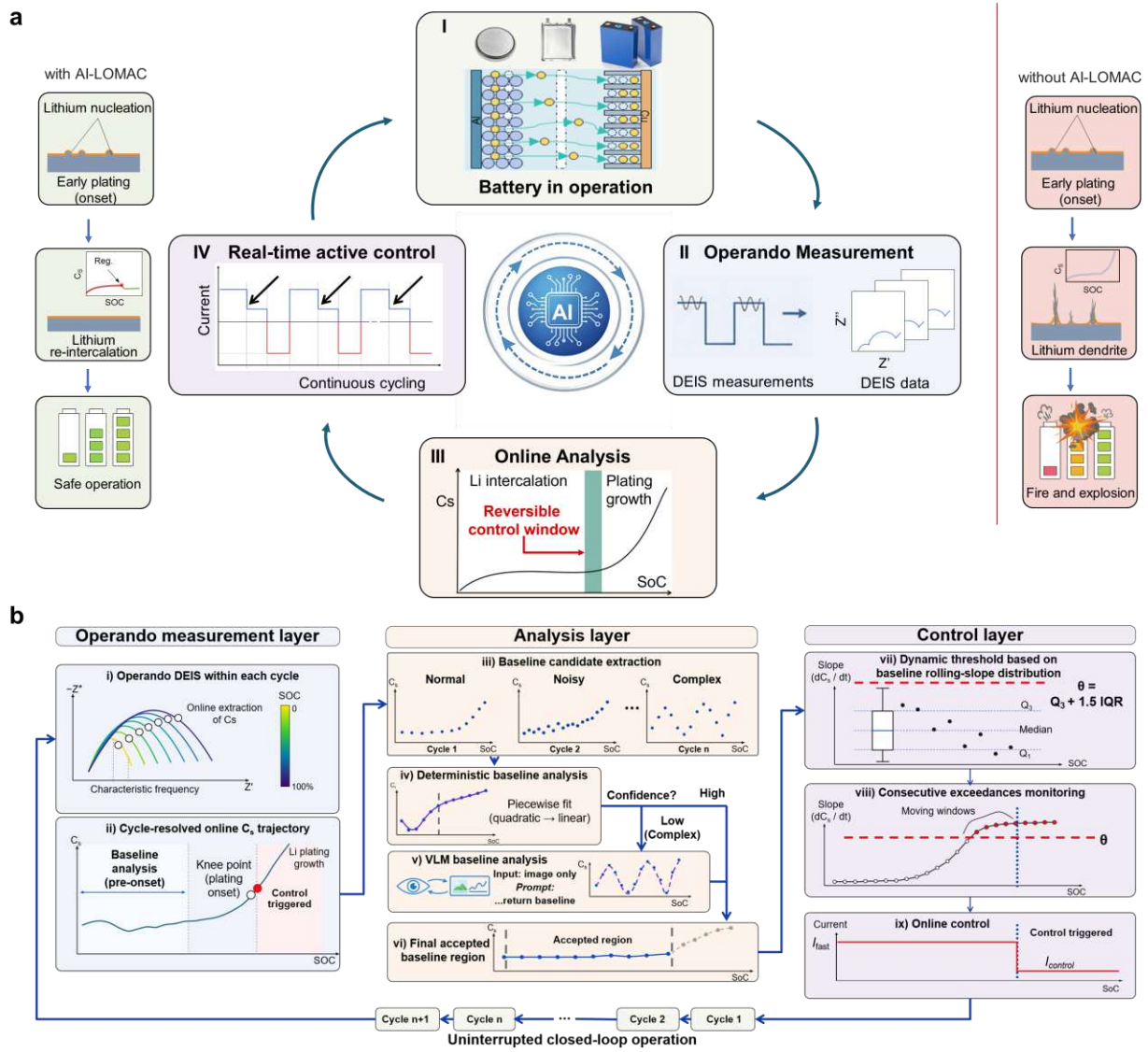


Fig. 1 | AI-driven operando measurement–analysis–control (AI-LOMAC) framework. **a**, Continuous closed-loop operation linking the four coordinated modules, including battery in operation (module I), operando measurement (module II), online analysis (module III) and real-time active control (module IV). With AI-LOMAC, the lithium nucleation on the anode will be suppressed at early-stage actively and the deposited lithium will re-intercalate into the anode, ensuring the long-term safe operation of the batteries (left side of Fig. 1a). In contrast, if without AI-LOMAC, after the onset of early lithium plating, lithium dendrites will continue to grow, pierce the separator and cause a short circuit, ultimately leading to fire hazards or even explosions (the right side of Fig. 1a). **b**, Cycle-resolved measurement (i, ii), analysis (iii–vi) and control (vii–ix) layers. High-confidence trajectories retain the deterministic baseline, whereas atypical low-confidence trajectories receive VLM-assisted baseline localization. The accepted baseline then enters the common deterministic knee-point detection and current-regulation workflow.

analysis of the graphite-anode charge-transfer response (see Methods and Supplementary Fig. 1 for details). The extracted impedance signals are then transferred to the analysis module, where DEIS-derived C_s are analyzed online to identify lithium-plating relevant changes (Fig. 1b, operando measurement layer, step ii). The C_s trajectory is then processed by confidence-aware routing: the shape-adaptive deterministic baseline is retained for high-confidence trajectories, whereas atypical low-confidence trajectories are rendered as images for vision-language model (VLM)-assisted baseline localization (Fig. 1b, analysis layer, steps iii-v; see Methods and Supplementary Fig. 2). The accepted baseline from either route enters the same deterministic detection procedure, in which a rolling-slope threshold and consecutive exceedances identify the C_s knee point, indicating plating onset (Fig. 1b, analysis layer, step vi; see Methods and Supplementary Fig. 3). Scheduled fast charging continues in the absence of a warning; once the onset criterion is met, the current is reduced to the predefined regulation level (Fig. 1a, module IV; Fig. 1b, control layer). This sequence is repeated across successive cycles until the voltage or capacity cut-off is reached. Ultimately, we confirmed that, the lithium nucleation on the anode will be suppressed at early-stage actively and the deposited lithium will re-intercalation into the anode, thus ensuring the long-term safe operation of the batteries with AI-LOMAC (left side of Fig. 1a). In contrast, if without AI-LOMAC, after the onset of early lithium plating, lithium dendrites will continue to grow, pierce the separator and cause a short circuit, ultimately leading to fire hazards or even explosions (the right side of Fig. 1a). A detailed description of the AI-LOMAC workflow is also provided in the Methods.

To evaluate the AI-LOMAC closed-loop system, we first established a reproducible plating condition in commercial LiCoO₂/Graphite (LCO||Gr) coin cells with a nominal capacity of 70 mAh. Figure 2a illustrates the entire experimental procedure, which includes simultaneously acquiring charge-discharge profiles and DEIS data with *operando* measurement, disassembling the cells for *ex situ* characterizations such as SEM and TMS, and coupling optical microscopy with charge-discharge and DEIS measurements using a custom-made perforated cell. The optical image inset in Fig. 2b shows the disassembled 14 layers of the graphite anodes, each with a diameter of 16.5 mm. Stacked commercial cells introduce

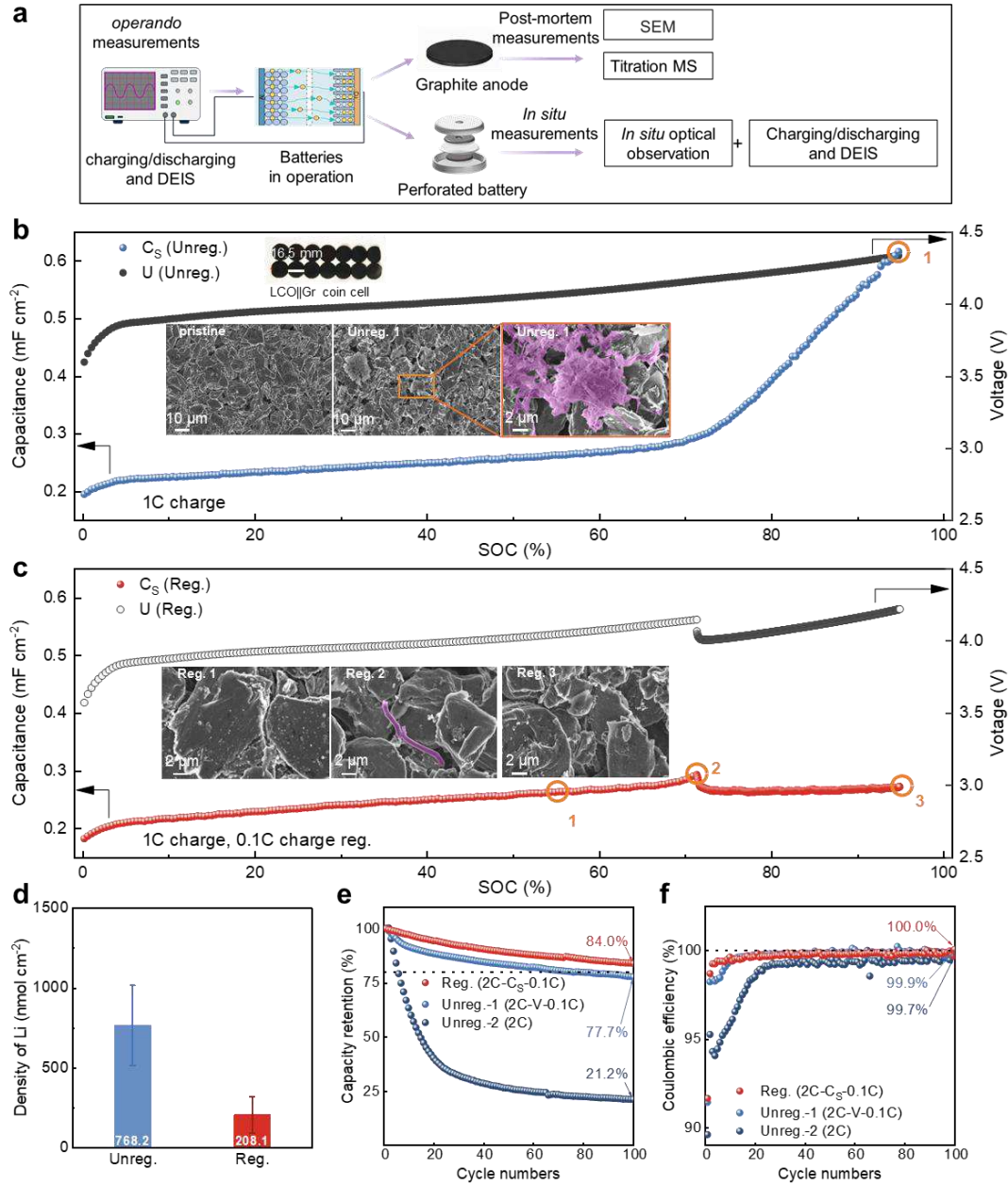


Fig. 2 | Implementation of active closed-loop on LCO||Gr commercial coin cells. **a**, Illustration of the entire experimental procedure, including *operando*, *in situ* and *ex situ* measurements. **b**, C_s and voltage profiles during charging under open-loop operation (Unreg.). SEM insets show the pristine graphite anode and the graphite anode at point 1. The optical inset shows the disassembled 14 layers of the graphite anodes. **c**, C_s and voltage profiles during charging under active closed-loop regulation. SEM insets show the graphite anodes at the labelled electrochemical states. **d**, Areal density of residual inactive metallic lithium in the unregulated (blue) and regulated (red) groups measured with TMS. **e**, **f**, Capacity retention (**e**) and Coulombic efficiency (**f**) over 100 cycles under closed-loop regulation (red, Reg.) and two open-loop protocols (blue, Unreg.-1 and Unreg.-2) at a primary charging rate of 2C.

greater through-plane heterogeneity in pressure, heat dissipation, and electrolyte distribution, which may promote non-uniform lithium plating during fast charging^{3,42,43}. As shown in Fig. 2b, under open-loop 1C charging (Unreg.), C_s continued to increase after the knee point, consistent with the overcharging condition and continued lithium deposition. The pristine graphite anode showed a clean surface (the SEM inset in Fig. 2b, pristine), whereas mossy lithium deposits were observed after overcharging (the SEM inset in Fig. 2b, Unreg. 1, point 1). After subsequent discharge to 2.5 V, fine filamentous residual lithium remained on the graphite surface (Supplementary Fig. 4a-c), suggesting incomplete stripping and the presence of residual metallic lithium that is likely electrochemically inactive under the applied conditions. This residual lithium is consistent with root-fracture during stripping, in which dendritic lithium becomes electrically isolated from the graphite matrix⁴⁴⁻⁴⁶. In the controlled cells, AI-LOMAC intervened when the C_s knee point was detected at ~71.4% SOC (Fig. 2c, point 2) by switching the charge current from 1C to 0.1C until the same SOC was reached followed by discharge at 0.2C to 2.5 V (Supplementary Fig. 4d). This intervention instantly stopped the rising of C_s , which subsequently decreased to a stabilized value of 0.27 mF cm⁻² at the end of charge (Fig. 2c). SEM images showed a clean graphite surface before the C_s knee point (the SEM inset in Fig. 2c, Reg. 1, point 1), only sparse filamentous lithium has been found at the knee point (the SEM inset in Fig. 2c, Reg. 2, point 2), and a restored and uniform surface after the 0.1C regulation step (the SEM inset in Fig. 2c, Reg. 3, point 3).

After discharge, the regulated anode remained free of residual lithium dendrites (Supplementary Fig. 4d-f), in contrast to the inactive lithium retained in the open-loop cell. These results show that AI-LOMAC suppresses the progression from early lithium deposition to persistent inactive lithium in commercial LCO||Gr coin cells. Two synergistic mechanisms are hypothesized to drive this morphological change. First, reducing the current density weakens tip-enhanced polarization and suppresses vertical dendrite growth⁴⁷. Second, the low-rate window may promote the dissolution, migration, and re-intercalation of weakly connected metallic lithium. Previous studies have shown that isolated lithium can undergo dynamic polarization under an electric field, dissolving at one end and

1 redepositing at the other, effectively reconnecting to the anode for re-intercalation^{8,48}. Thus, AI-LOMAC enables
2 early detection of lithium plating and drives more reversible interfacial lithium utilization.

3 TMS was further used to quantify residual metallic lithium in LCO||Gr commercial coin cells with and without closed-
4 loop regulation. TMS detects D₂ generated from the reaction between metallic lithium and D₂O with nanomolar sensitivity,
5 the apparatus and protocol are shown in Supplementary Fig. 5^{24,49}. Standard calibration curve showed excellent linearity
6 ($R^2 = 0.999$, Supplementary Fig. 6). Before disassembly, all cells were discharged at 0.2C to 2.5 V to minimize the
7 contribution of reversible intercalated lithium in graphite, so that the measured D₂ signal mainly reflected residual metallic
8 lithium rather than Li_xC₆²³. Although the charge/discharge electrochemical profiles were consistent across the experimental
9 groups (Supplementary Fig. 7), TMS quantification revealed a clear difference in residual metallic lithium. The open-loop
10 cells retained 768.2 ± 249.6 nmol cm⁻² metallic lithium after discharge, whereas the cells with AI-LOMAC retained 208.1
11 ± 114.4 nmol cm⁻² (Fig. 2d), corresponding to a 73% reduction. These TMS results support the SEM observations and
12 confirm that closed-loop regulation reduces residual metallic lithium in commercial LCO||Gr coin cells.

13 The cycling stability of this closed-loop strategy was then evaluated in the same LCO||Gr system over 100 cycles under
14 three harsh charging/discharging protocols. In the closed loop protocol, Reg. (2C–C_s–0.1C), cells were charged at 2C to
15 the online-detected C_s knee point, switched to 0.1C charging until 4.2 V, and then discharged at 2C to 2.5 V. In the voltage-
16 defined multistage protocol, Unreg.-1 (2C–V–0.1C), cells were charged at 2C to 4.2 V, switched to 0.1C charging until 4.2
17 V, and then discharged at 2C to 2.5 V. In the constant-current protocol, Unreg.-2 (2C), cells were charged at 2C to 4.2 V
18 and discharged at 2C to 2.5 V. As displayed in Fig. 2e-f, Unreg.-2 showed rapid capacity decay and fell below 80% retention
19 within 5 cycles under such harsh fast charging/discharging condition, whereas Unreg.-1 fell below 80% retention at cycle
20 76 and reached 77.7% retention at the end of testing. By contrast, the AI-LOMAC regulated cells retained 84.0% of their
21 capacity after 100 cycles. All groups maintained Coulombic efficiencies above 99.7% at the 100th cycle, with the regulated
22 group reaching 100%. These results indicate that replacing fixed voltage-defined transitions with state-responsive C_s

feedback improves fast-charge durability significantly. Extended cycle-resolved traces further confirmed repeated onset detection and current regulation across successive cycles (Supplementary Fig. 8). Compared with constant 0.5C charging, the onset-triggered 2C-C_s-0.5C and 1C-C_s-0.5C protocols shortened cumulative charging time by 38.3% and 44.1%, with capacity retention after 100 cycles lower by only 3.2 and 3.9 percentage points, respectively (Supplementary Fig. 9). We further validate the generality of the AI-LOMAC system using laboratory-assembled 1.8 mAh single-layer LFP||Gr coin cells, which allow systematic *in situ* and post-mortem characterization. The C_s and voltage profiles coupled with graphite anode SEM morphologies at different cycling stages (Supplementary Fig. 10) verify the reliable regulation performance of AI-LOMAC on these custom-built cells. TMS quantitative analysis (Supplementary Fig. 11) further confirms the effective suppression of residual metallic lithium enabled by closed-loop regulation. Specifically, unregulated cells retain 8.1 ± 5.0 nmol cm⁻² metallic lithium after discharge, while AI-LOMAC regulated cells exhibit a much lower residual lithium content of 3.0 ± 1.1 nmol cm⁻², demonstrating that the developed strategy can substantially mitigate residual metallic lithium formation in LFP||Gr configurations.

To directly visualize the timing of the closed-loop intervention, we coupled *in situ* optical microscopy (see Methods for details) with DEIS in custom-made perforated laboratory LFP||Gr coin cells. To avoid optical interference from lithium deposition on copper, a copper-free graphite electrode was prepared by coating the anode slurry directly onto a ceramic separator. In these specialized cells, geometrical constraints reduce the effective anode area, inherently elevating the local current density and accelerating localized polarization even at a nominal 1C rate. In the open-loop LFP||Gr cell (Fig. 3a-b), retrospective analysis of the recorded C_s trajectory identified a knee point at 82.7% SOC and 3.69 V (Fig. 3a, point 3), while the graphite anode surface remained optically dendrite-free (Fig. 3c, points 1-3). Visible lithium deposition appeared later at 91.5% SOC (Supplementary Fig. 12, point 1) and developed into cluster-like deposits by 95% SOC (Fig. 3c, point 4). After subsequent discharge at 0.1C, part of the deposited lithium remained inactive (Fig. 3c, points 5-6). In the closed-loop cell (Fig. 3d-e), AI-LOMAC actively identified the C_s knee point at 82.7% SOC and 3.64 V (Fig. 3d, point 3),

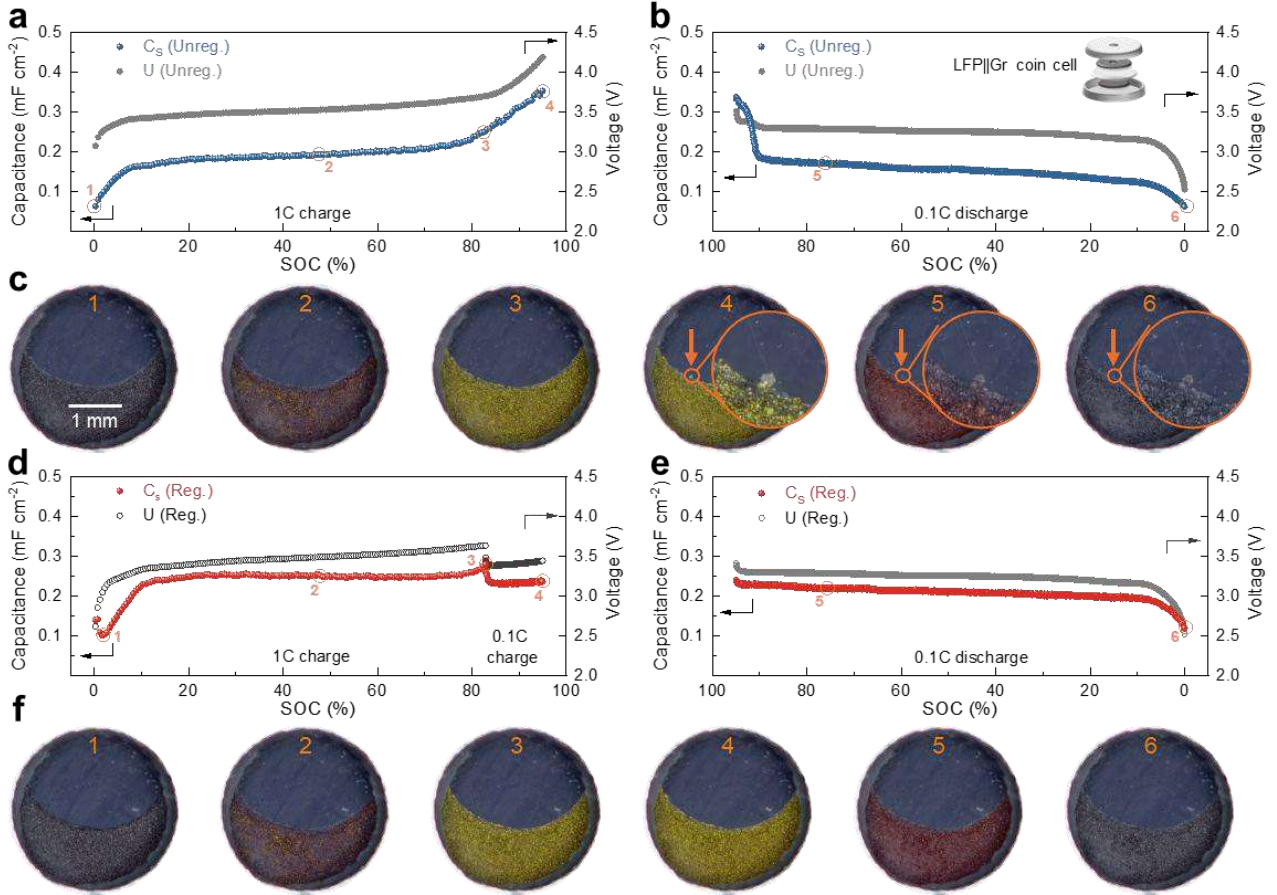


Fig. 3 | Validation of the AI-LOMAC system by *in situ* optical microscopy with custom-made laboratory LFP||Gr coin cells. **a, b,** C_s and voltage profiles of the open-loop cell during charging (**a**) and discharging (**b**). The inset in Fig.3b shows the structure of the perforated laboratory LFP||Gr coin cell, in which the copper-free graphite anode was punched with a hole and coated on the top of the separator. **c,** *In situ* optical images of the graphite anode in the open-loop unregulated cell at the labelled states. **d, e,** C_s and voltage profiles under active closed-loop during charging (**d**) and discharging (**e**). **f,** *In situ* optical images of the graphite anode in the regulated cell at the labelled states. The labelled optical images correspond to the numbered points on the C_s profiles, and the selected states correspond to matched SOC values between the open-loop and closed-loop cells.

immediately reduced the current to 0.1C, and maintained a macroscopically dendrite-free graphite surface despite the same total charge capacity (Fig. 3f, points 1-4). No lithium dendrites were observed before the C_s knee point (Fig. 3f, points 1-2) or during subsequent discharge at 0.1C (Fig. 3f, points 5-6). Retrospective alignment of the open-loop C_s trace with the optical images showed that the C_s knee point preceded the first visible lithium deposition by approximately 316 s (Supplementary Fig. 12). Thus, in this optical cell, the C_s marker provided an earlier warning than direct optical observation.

Application of AI-LOMAC to high-capacity LFP||Gr cells

To further validate the practicability, AI-LOMAC was applied to commercial 3.5 Ah LFP||Gr pouch cells. The open-loop cells were charged continuously at 3C to the prescribed charge capacity, whereas the regulated cells switched to 0.1C at the online-detected C_s knee point and then charged to the same capacity to ensure comparability (Fig. 4a). After current reduction, the C_s trajectory decreased after the knee point, consistent with the response observed in coin cells (red curve). In contrast, the C_s increased continuously after passing the knee point for the unregulated one (blue curve). These results indicate that the C_s -triggered mitigation protocol is transferable from coin cells to Ah-scale pouch cells. The AI-LOMAC strategy was finally evaluated in commercial 314 Ah LFP||Gr prismatic cells in an environmental chamber at 0°C with an initial charging rate of 0.5C. As shown in Fig. 4b, the open-loop cell was charged continuously to a 3.65 V cut-off, whereas the regulated cell switched to 0.1C at the online-detected C_s knee point and was then charged to the same voltage cut-off. The curves of C_s show different trajectories for the unregulated (blue) and regulated (red) cells. After discharge to 2.5 V, post-mortem optical inspection showed multi-regional macroscopic lithium deposition on the graphite anode of the unregulated cell (Fig. 4c-d, corresponding to point 1 in Fig. 4b), whereas the graphite anode of the closed-loop regulated cell showed only one localized deposition region (Fig. 4e-f, corresponding to point 2 in Fig. 4b). SEM analysis of the selected regions showed that, relative to the pristine graphite anode (Supplementary Fig. 13), the graphite anode of the open-loop cell was covered with dense dendritic lithium after discharge (Fig. 4g-h), whereas only sparse residual lithium deposits were observed on the graphite anode of the closed-loop cell (Fig. 4i-j). These results indicate that C_s -triggered regulation reduced both the spatial extent and severity of lithium deposition in 314 Ah prismatic cells. The remaining localized plating indicates that large-format implementation may require further optimization of the detection threshold and confirmation window to enhance the detection sensitivity and advance the lithium plating detection and regulation to an earlier stage.

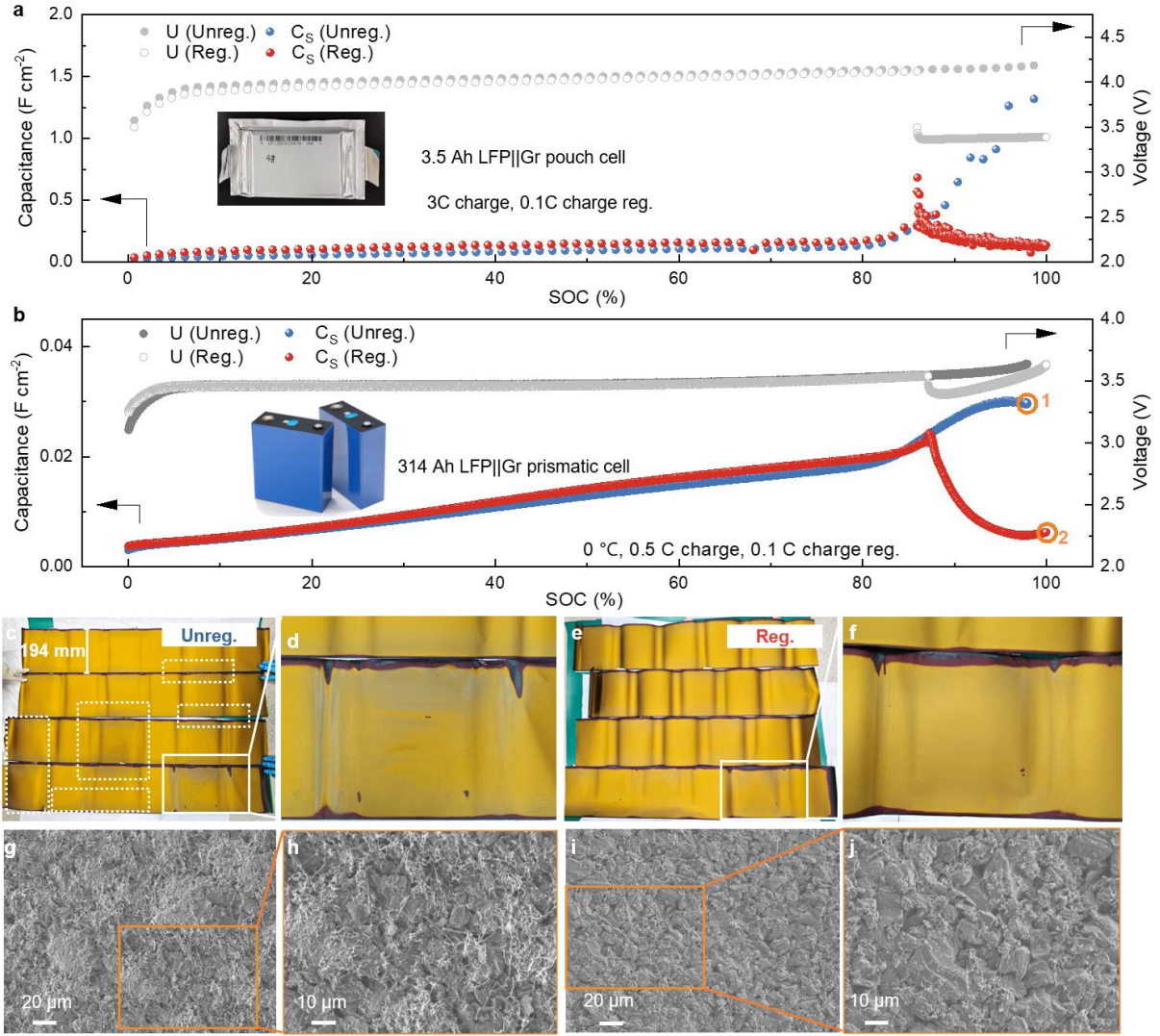


Fig. 4 | Practicability of active closed-loop regulation in commercial high-capacity batteries. **a**, C_s and voltage profiles of unregulated open-loop and regulated closed-loop 3.5 Ah LFP||Gr pouch cells during charging. **b**, C_s and voltage profiles of unregulated open-loop and regulated closed-loop 314 Ah LFP||Gr prismatic cells during charging. **c**, Optical image of the graphite anode of the open-loop unregulated 314 Ah LFP||Gr cell after discharge to 2.5 V. **d**, Magnified optical image of the solid-boxed region in (c). **e**, Optical image of the graphite anode of the closed-loop regulated 314 Ah LFP||Gr cell after discharge to 2.5 V. **f**, Magnified optical image of the solid-boxed region in (e). **g**, SEM image of a selected region in (d). **h**, Magnified view of the orange-boxed region in (g). **i**, SEM image of a selected region in (f). **j**, Magnified view of the orange-boxed region in (i). The 194 mm annotation indicates the lateral dimension of the anode shown in the optical images. In the voltage profiles, filled and open circles denote open-loop and closed-loop cells, respectively. In the C_s profiles, blue and red curves represent the unregulated open-loop and regulated closed-loop cells, respectively.

Discussion and conclusions

This work presents AI-LOMAC, an AI-driven closed-loop framework that shifts lithium-plating management from post-failure diagnosis to real-time early intervention. C_s provides a surface capacitance signature that marks early plating onset, verified by *operando* method to offer more than 5 minutes of advance warning. This enables targeted regulation that lifts capacity retention fourfold and cuts inactive lithium by 73%, substantially improving both safety and cycle life, especially under harsh conditions such as fast charging or overcharging operation. Validated across coin, pouch and large-format prismatic cells, the platform exhibits robust industrial scalability. Future upgrades will incorporate acoustic, mechanical and thermal multi-physics signals alongside high-fidelity digital twins to rapidly screen charging, pulse and thermal/pressure tuning strategies for dendrite suppression via adaptive intelligent control.

Critically, AI-LOMAC is more than a safety BMS 2.0. Operating 24/7 across diverse real-world scenarios, including fast-charging EVs, low-temperature maritime operations, and grid frequency regulation, it generates an unprecedented volume of correlated, multi-modal data. This positions AI not merely as an interpreter of existing signals, but as a potential creator of new knowledge. The resulting data ecosystem lays the foundation for AI to propose novel materials, electrolyte formulations, electrode architectures, and operational strategies that have never been conceived by human intuition, a transition from AI as assistant to AI as inventor. This closed-loop architecture, where AI bridges transient interfacial signals to autonomous controls, transcends a mere technological advance; it embodies a new philosophy of system operation. By generating and continuously learning from multi-modal operational data, AI-LOMAC positions artificial intelligence not merely as a guardian of battery safety, but as an active partner in discovery, capable of conceiving materials, protocols, and strategies beyond human intuition. In doing so, it charts a course towards a future where intelligent systems not only manage energy, but continuously reinvent the very rules by which energy is stored and delivered.

1 **Methods**

2 **Materials and reagents**

3 Graphite (Gr) and lithium iron phosphate (LiFePO_4 , LFP) powders were procured from Shanghai Shanshan Technology
4 Co., Ltd. and Shenzhen Dynanonic Co., Ltd., respectively. Conductive carbon black (Super P) and carbon nanotubes (CNTs)
5 were obtained from Hefei Kejing Materials Technology Co., Ltd. and Jiangsu Cnano Technology Co., Ltd., respectively.
6 Polymeric binders, including sodium carboxymethyl cellulose (CMC) and styrene-butadiene rubber (SBR), were supplied
7 by Daicel Corporation (Japan) and Zeon Corporation (Japan), respectively. Polyvinylidene fluoride (PVDF) was purchased
8 from Solvay (Belgium). Dimethyl carbonate (DMC) was purchased from Shanghai Jizhi Biochemical Technology Co., Ltd.
9 Lithium metal foil (99.95%), ceramic-coated separators (comprising a 12 μm base with 2 μm coatings on both sides), and
10 commercial LFP01 electrolyte were all sourced from Dongguan Kelude Experiment Equipment Co., Ltd. High-purity argon
11 gas (Ar, 99.999%) and deuterium oxide (D_2O , 99.9 atom% D) were provided by Linde Gas and Energy Chemical,
12 respectively.

13 **Battery configurations and electrochemical testing protocols**

14 Four cell configurations were used to evaluate the applicability of the closed-loop strategy across different cell formats.
15 First, laboratory-assembled CR2016 LFP||Gr full cells were prepared with a negative-to-positive electrode capacity ratio
16 (N/P ratio) of approximately 1.1 and a nominal capacity of approximately 1.8 mAh. The LFP cathode consisted of 95 wt%
17 LFP, 1 wt% Super P, 1 wt% CNTs, and 3 wt% PVDF coated on aluminum foil. The Gr anode comprised 95.5 wt% Gr, 1.4
18 wt% Super P, 0.1 wt% CNTs, 1.2 wt% CMC, and 1.8 wt% SBR coated on copper foil. The fabricated electrodes exhibited
19 areal mass loadings of 5.7 mg cm^{-2} (95.5 wt% active material) for the Gr anode and 12.5 mg cm^{-2} (95.0 wt% active material)
20 for the LFP cathodes. Commercial LFP01 electrolyte was used. Cell assembly was performed in a high-purity Ar-filled
21 glovebox (99.999% Ar; O_2 and H_2O < 0.01 ppm). In addition, a laboratory-assembled three-electrode LFP||Gr pouch cell
22 with a capacity of 43.8 mAh was prepared for electrode-resolved impedance measurements and characteristic-frequency

identification. This three-electrode cell used the same LFP cathode, Gr anode, and LFP01 electrolyte as the laboratory LFP||Gr coin cells, with a separator-isolated Li reference electrode introduced to decouple the impedance contributions from the Gr anode and LFP cathode. This cell was used only for mechanistic impedance analysis and was not included in the closed-loop scalability tests. Second, commercial LIR2032H-type LiCoO₂||graphite (LCO||Gr) coin cells with a nominal capacity of 70 mAh were purchased from CT-Energy Technology Co., Ltd. Third, commercial 3.5 Ah LFP||Gr pouch cells were sourced from Hithium Energy Storage Technology Co., Ltd. Finally, commercial 314 Ah LFP||Gr energy storage prismatic cells were provided by EVE Energy Co., Ltd. Electrochemical tests were performed using a battery testing system (Wuhan LANHE) and a potentiostat (Biologic VMP-300). Unless otherwise specified, tests were conducted in a temperature-controlled environmental chamber maintained at 25 ± 0.5 °C. Before electrochemical evaluation, cells were preconditioned by three charge–discharge cycles at 0.1C for LFP||Gr cells and 0.2C for LCO||Gr cells. The reference capacity and cell-to-cell consistency were then evaluated by cycling the cells at charge rates of 0.5C, 1C, and 2C, with discharge at 0.2C.

Identification of the characteristic frequency for the graphite anode charge-transfer process

The characteristic frequency associated with the graphite-anode charge-transfer process was identified from impedance spectra collected at different states of charge (SOC). In contrast to the real part of impedance (Z'), which contains overlapping resistive contributions from multiple electrochemical and transport processes, the imaginary part of impedance (Z'') is more sensitive to capacitive responses near their characteristic frequencies³³. Within the charge-transfer region, Z'' is therefore mainly governed by the charge-transfer resistance (R_{ct}) and the electrical double-layer capacitance (C_d). The imaginary components of the impedance spectra collected from two batteries at different SOC are shown in Supplementary Fig. 1. Because capacitive elements, including the SEI capacitance (C_{SEI}) and C_d , respond at different characteristic frequencies, Z'' varies with frequency. In the mid-frequency region, the absolute value of Z'' decreases with increasing SOC. Because this region was sufficiently separated from the dominant frequency ranges of other processes, the Z'' response in

this region was assigned primarily to the graphite-anode charge-transfer process. For commercial LCO||Gr coin cells in a two-electrode configuration, the cells were charged at 1C, and DEIS were collected at different SOC levels over a frequency range of 1 Hz to 100 kHz using an AC current amplitude equivalent to 0.1C (Supplementary Fig. 1a). For the laboratory-assembled LFP||Gr pouch cell with a three-electrode configuration and a capacity of 43.8 mAh, the cell was charged at 0.5C and rested for 3 h at every 20% SOC increment before static EIS measurements. Static EIS measurements were then carried out over a frequency range of 0.1 Hz to 100 kHz with a voltage amplitude of 10 mV (Supplementary Fig. 1b). The three-electrode configuration enabled separation of the impedance contributions from Gr anode and LFP cathode. The optimal SOC-dependent trend was found near 100 Hz, characterized from both different batteries. Therefore, $f = 100$ Hz was selected as the characteristic frequency for monitoring the graphite-anode charge-transfer response.

Implementation of the confidence-aware AI-LOMAC analysis and control workflow

The central requirement for closed-loop operation is to transform the continuous DEIS stream into an executable control variable. Each DEIS window provided Z' , Z'' , magnitude ($|Z|$), and phase components of the impedance on a logarithmic frequency grid. Based on the characteristic-frequency analysis described above, the graphite-anode interfacial capacitance, C_s , was calculated at $f = 100$ Hz and used as the plating-sensitive descriptor. For a predominantly parallel R_{ct} - C_d response, C_s was calculated from the imaginary component of impedance according to the impedance-capacitance relationship:

$$C_s = -\frac{1}{\omega Z''} = \frac{1}{\omega^2 R_{ct}^2 C_d} + C_d \quad (1)$$

where $\omega = 2\pi f$ and $f = 100$ Hz. Changes in C_s reflect the evolution of surface electrochemical processes on graphite, including the transition from lithium intercalation to plating-prone interfacial states. During fast charging before lithium plating, C_s evolves gradually with SOC. When lithium plating begins, the C_s trajectory deviates from the pre-onset baseline and increases rapidly, accompanied by an increase in local curvature. This transition produces a knee point in the C_s -SOC curve³³. Therefore, AI-LOMAC uses the C_s knee point, rather than the absolute C_s value, as the operational early-warning marker (Supplementary Fig. 3a). This strategy reduces sensitivity to cell-to-cell baseline variations while retaining a

physically interpretable signature of the transition from intercalation-dominated behaviour to plating-prone interfacial behaviour.

A key analytical challenge was the reliable determination of the pre-onset baseline as the morphology of the C_s trajectory changed during cycling. Each cycle-resolved trajectory was therefore processed by confidence-aware routing (Fig. 1b and Supplementary Fig. 2). After smoothing, the deterministic route applied shape-adaptive piecewise quadratic-to-linear regression to estimate a candidate baseline. The goodness of fit, R^2 , was used as the routing confidence, and trajectories with $R^2 \geq 0.97$ were accepted directly. For the 1C implementation shown in Supplementary Fig. 2, trajectories with $R^2 < 0.97$ were rendered over 0 – 1,800 s and submitted once per cycle to the VLM (Gemini-3-pro-image-preview). A fixed prompt asked the model to identify the start time of the near-linear terminal segment from the overall curve morphology, ignore small local fluctuations and return a single numerical time. A preset response timeout prevented the VLM request from interrupting cycling; if no response was received, the locally estimated boundary was retained. The baseline selected through either route was passed to the same deterministic slope-thresholding and five-consecutive-point confirmation procedure. The VLM was restricted to baseline localization and did not calculate the threshold, confirm the knee point or issue a current-control command. Independent upper and lower voltage cut-offs remained active throughout charging and discharging.

Using the accepted baseline, the local slope of C_s was calculated with an n-point rolling least-squares regression window rather than a two-point difference. Unless otherwise stated, $n = 5$ was used to estimate each local slope. This five-point regression window is distinct from the five consecutive suprathreshold slope estimates required to confirm the knee point. The trigger threshold was calculated from the selected pre-onset baseline region as:

$$\theta = Q_3(\dot{C}_s^{base}) + 1.5IQR(\dot{C}_s^{base}) \quad (2)$$

where $\dot{C}_s^{base}$ denotes the local-slope values within the accepted baseline region, Q_3 is their 75th percentile, and $IQR(\dot{C}_s^{base})$ denotes the interquartile range. A lithium-plating warning was issued only when:

$$\dot{C}_{s,t} \geq \theta \quad (3)$$

for m consecutive DEIS evaluations. Unless otherwise stated, $m = 5$ was used. This consecutive-point confirmation rule suppressed isolated noise-induced excursions and converted the continuously evolving DEIS response into a binary control command. A smaller m increases sensitivity but may increase false-positive triggering, whereas a larger m improves specificity at the cost of response speed.

The detection algorithm was evaluated before closed-loop regulation. 16 laboratory-assembled LFP||Gr coin cells were charged under open-loop operation while candidate knee points were logged online. The online assignments were compared with offline review of the C_s trajectories, and the plating-relevant knee point was captured in all cases (Supplementary Fig. 3b). Closed-loop regulation was then performed on 40 fresh cells, in which the controller switched the fast-charge segment to the low-current mitigation step at the online-detected C_s knee point (Supplementary Fig. 3c). Online and offline assignments were consistent in 39 of 40 cells; the remaining discrepant cell was excluded from subsequent statistical analyses. The knee-point distributions obtained from open-loop and closed-loop tests were similar, indicating that the spread mainly arose from cell-to-cell variability rather than measurement instability (Supplementary Fig. 3d). The robustness of the warning rule was further examined by analyzing the maximum number of consecutive suprathreshold points outside the true knee point region (Supplementary Fig. 3e). Short suprathreshold sequences occasionally appeared because of local derivative fluctuations, but they rarely persisted long enough to satisfy the five-point confirmation rule. This analysis identifies the consecutive-point requirement as a tunable parameter for balancing sensitivity and robustness in real-time operation.

The VLM fallback was motivated by retrospective analysis of an atypical trajectory from the initial long-cycle experiment (Supplementary Fig. 2). The fixed 200-600 s baseline missed the onset, and the initial piecewise regression did not provide a reliable boundary; using the VLM-localized baseline allowed the unchanged deterministic detector to recover the onset. In the subsequent 100-cycle online experiment, the updated piecewise regression satisfied $R^2 \geq 0.97$ for all

trajectories, so the deterministic route was retained and the VLM was not invoked. The VLM therefore served as an on-demand fallback for atypical morphologies rather than a continuously active controller.

***Operando* detection of lithium plating and closed-loop regulation protocols**

Lithium plating was induced by galvanostatic charging at elevated C-rates or low temperature. Unless otherwise stated, the charge endpoint was set to 95% of the reference discharge capacity measured at 0.1C for LFP||Gr cells and 0.2C for LCO||Gr cells, followed by low-rate discharge to the cut-off voltage. DEIS spectra were collected during the charge and discharge process using a Biologic VMP-300 potentiostat. The frequency range spanned from 100 kHz to 1 Hz. The AC perturbation amplitude was set to a current equivalent of 0.1C for applied currents below 1C and to 10% of the applied DC current for currents rates exceeding 1C. The onset of lithium plating was identified by monitoring the variation trend of C_s at characteristic frequencies. The experiments were categorized into two groups. For the unregulated (open-loop) group, cells were charged at the prescribed high rate to the target capacity or voltage cut-off and then discharged at low rate. Conversely, for the AI-LOMAC regulated (closed-loop) group, cells were initially charged at an identical high-rate condition. Once the C_s knee point was identified by the online algorithm, the current was automatically reduced to the predefined mitigation current, and charging continued to the same termination criterion used for the open-loop group.

Evaluation of onset-triggered charging protocols

To evaluate the balance between charging time and cycling performance, LCO||Gr coin cells were subjected to three cycling protocols: constant 0.5C charging, onset-triggered 2C- C_s -0.5C charging and onset-triggered 1C- C_s -0.5C charging. For the onset-triggered protocols, charging began at 2C or 1C. If the online analysis detected a C_s knee point, a lithium-plating warning was issued and the charging current was reduced to 0.5C until the upper cut-off voltage of 4.2 V was reached. If no warning was issued, charging continued at the initial rate until the same voltage cut-off. All cells were discharged at 0.5C to 2.75 V. For better comparison, 2C or 1C charging and 0.5C discharging have also been tested. Each protocol was repeated for 100 cycles, after which cumulative charging time and capacity retention were compared with those obtained

under constant 0.5C charging.

***In situ* optical microscopy and specialized cell architecture**

In situ optical microscopy was conducted using an industrial camera (GP-660V, Gaopin, Kunshan, China), equipped with a 4.5× objective lens and operating at a capture rate of 60 frames per second (fps). Custom-made perforated optical coin cells were assembled to visualize lithium plating on the Gr anode. The LFP cathode slurry consisted of LFP, Super P, CNTs and PVDF at a mass ratio of 95:1:1:3 and was coated on aluminum foil. To avoid optical interference from lithium deposition on copper, a copper-free graphite electrode was prepared by coating the anode slurry directly onto a ceramic separator. The graphite anode slurry consisted of graphite, Super P, CNTs, CMC and SBR at a mass ratio of 95.5:1.4:0.1:1.2:1.8. The LFP cathode was punched into discs (11 mm in diameter). The separator-supported graphite anode had an outer diameter of 12 mm and a 3 mm central aperture, forming an annular electrode. This geometry reduced the N/P ratio to approximately 1.0, compared with 1.1-1.2 in intact reference cells. The optical cell stack consisted of the LFP cathode, separator and separator-supported Gr anode. The electrochemically active graphite surface faced the optical window to allow direct visualization (Fig. 3b inset).

Morphological characterization

Anode morphology was characterized by field-emission scanning electron microscopy (SEM, Zeiss GeminiSEM 500). To minimize oxidation of lithium deposits and SEI components, all *ex situ* samples were prepared under inert atmosphere. Cells were disassembled in an Ar-filled glovebox (99.999% Ar; O₂ and H₂O < 0.01 ppm). The anodes were removed, rinsed three times with DMC to remove residual electrolyte, vacuum dried, and stored in sealed vials before characterization. For SEM imaging, samples were mounted on SEM holders in the glovebox and transferred rapidly to the SEM chamber, with air exposure limited to less than 10 s. To capture specific electrochemical states, cells were stopped at predefined charge or discharge points and immediately disassembled in the glovebox.

Quantitative analysis *via* isotope-labeled titration mass spectrometry

Isotope-labelled titration mass spectrometry (TMS) was used to quantify residual metallic lithium and evaluate the effect of closed-loop regulation. The method is based on the reaction between metallic lithium and deuterium oxide ($2\text{Li} + 2\text{D}_2\text{O} \rightarrow 2\text{LiOD} + \text{D}_2\uparrow$). The generated D_2 signal was assigned to metallic lithium, whereas proton-containing species such as LiH or organic residues mainly generate HD , enabling isotopic discrimination. Before disassembly, $\text{LFP}||\text{Gr}$ and $\text{LCO}||\text{Gr}$ cells were discharged at 0.2C to 2.5 V , to minimize the contribution of reversible intercalated lithium in graphite. The cells were then disassembled in an Ar-filled glovebox, and the anodes were rinsed with DMC and vacuum dried as described above.

The prepared anodes were sealed in 8 mL headspace vials inside the glovebox. Each vial was connected to a mass spectrometer (Hiden HPR-20) and a high-purity Ar carrier gas (Ar, 99.999%) line through a double-path capillary system. The vials were sealed with gas-tight elastomeric septa. Before measurement, the gas lines were purged with high-purity Ar at 1 mL min^{-1} to remove atmospheric contaminants. The headspace vial was then connected to the gas circuit using a four-way valve. After baseline stabilization, a fixed excess volume (approximately 1.5 mL) of D_2O was injected into the vial *via* a syringe to react with metallic lithium. The evolved D_2 gas ($m/z = 4$) was swept by the Ar flow into the mass spectrometer for real-time detection. The D_2 peak area was integrated and converted to the mass of metallic lithium using an external calibration curve.

For calibration, lithium metal standards with masses of 0.10-0.95 mg were weighed in the glovebox using a semi-micro analytical balance with 0.01 mg readability. The standards were reacted with excess D_2O in sealed vessels under identical analytical conditions. The integrated D_2 peak area was plotted against lithium mass to establish the calibration curve (Supplementary Fig. 6). Unless otherwise stated, TMS quantification was performed using three independent cells for each group.

Data availability

All the research data has been uploaded to the figshare repository.

Code availability

The platform-independent source code implementing the AI-LOMAC analysis and decision logic is provided as Supplementary Software. It includes shape-adaptive baseline estimation, confidence-aware VLM routing, slope-threshold calculation, consecutive-point confirmation and abstract control actions.

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Competing interests

The authors declare no competing interests.

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