



Diagnosis of electrode-level degradation modes from field EV data without in-life reference cycles

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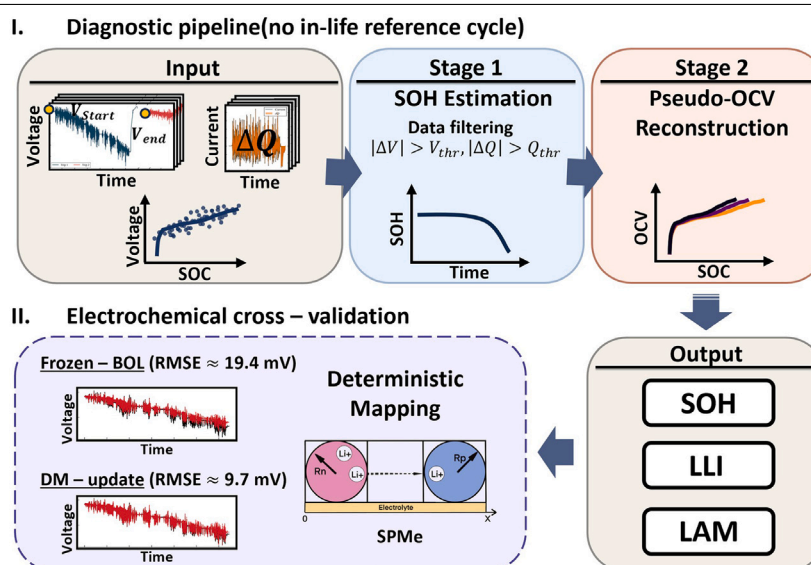
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HIGHLIGHTS

- Electrode-level degradation from EV field data without in-life reference cycles.
- Pseudo-OCV is reconstructed without any aged-state capacity reference.
- Voltage-bin weighting corrects the skewed SOC coverage of real-world driving.
- An independent SPMe cross-check predicts aged driving voltage within 9–13 mV.
- The method separates anomalous rapid fade from normal aging across three EVs.

GRAPHICAL ABSTRACT



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ABSTRACT

Mechanistic open-circuit voltage (OCV) reconstruction enables interpretable diagnosis of battery degradation, but existing methods require a reference capacity rarely available in production fleets. We propose a reference-free pipeline—requiring only a one-time beginning-of-life characterization at manufacture—that propagates a single field-data modality through state-of-health (SOH) estimation, pseudo-OCV reconstruction, and electrode-level degradation-mode identification. A preliminary SOH trajectory, obtained by mapping charge increments to a reference OCV curve with Gaussian process smoothing, anchors the ΔQ -method, whose stoichiometric parameters are fitted with a voltage-bin-weighted loss that corrects the high-SOC skew of field data. Applied to multi-year records from three production electric vehicles, the method distinguished normal aging from an anomalous rapid-fade event. Positive-electrode active-material loss and lithium-inventory loss were the primary

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degradation modes, while negative-electrode active-material loss was buffered by the surplus anode capacity. The voltage-bin weighting reduced reconstruction errors by up to 45%. Tested against an independent single-particle model with no fitting to the driving data, the reconstructed modes predicted the aged-stage driving voltage within a 9–13 mV band per vehicle. Requiring only onboard relaxed-voltage and charge logs, the framework suits fleet-wide health monitoring.

1. Introduction

With the rapid growth of the electric vehicle (EV) market, accurate estimation of the state of health (SOH) has become a prerequisite for safe operation, optimal charging control, and residual-value assessment of battery packs [1–3]. SOH is conventionally defined as the ratio of the current full-charge capacity to its beginning-of-life (BOL) value. However, such a scalar metric alone does not reveal the underlying *aging path*: two cells at the same SOH but with different operational histories can exhibit markedly different future degradation trajectories [4–6]. This path dependence originates from the interplay of distinct electrochemical degradation mechanisms, commonly grouped into three degradation modes (DMs): loss of lithium inventory (LLI), loss of active material at the negative electrode (LAM_n), and loss of active material at the positive electrode (LAM_p) [7,8]. Because these DMs alter the alignment of the electrode open-circuit potentials (OCPs) that compose the cell voltage [9,10], reconstructing the full-cell open-circuit voltage (OCV) from electrode-level parameters offers a physically interpretable route to degradation diagnosis—one that scalar SOH cannot provide.

Data-driven approaches have achieved high accuracy in SOH estimation and cycle-life prediction under laboratory conditions. Severson et al. [11] demonstrated that machine learning could predict cycle life from early-cycle discharge voltage curves, and subsequent studies extended this paradigm with deep neural networks [12], degradation-pattern recognition with feature extrapolation [13], transfer learning [14], and impedance-based feature extraction [15]. Despite these advances, purely data-driven methods face two persistent limitations: (i) they typically require large volumes of laboratory cycling data for training and generalize poorly to unseen cell chemistries or real-world operating conditions [16–18]; and (ii) the trained models are inherently *black boxes*—they estimate a scalar health indicator without identifying the electrode-level degradation mechanisms that govern future aging behavior.

Mechanistic modeling approaches overcome the interpretability limitation by describing the full-cell OCV as the difference of two half-cell OCPs whose stoichiometric windows are parameterized by alignment parameters [8,19]. Fitting these parameters to measured voltage data enables simultaneous estimation of SOH and the individual DMs. Bloom et al. [9,20] pioneered differential voltage analysis (DVA) for non-destructive degradation identification, and the framework was subsequently complemented by incremental capacity analysis (ICA) [21–23], extended to higher C-rates and varying temperatures [19,24], and applied to partial charging segments at different current rates [25,26]. Recently, Hofmann et al. [27] introduced the ΔQ -method, which uses only relaxed-voltage points and the accumulated charge between them, rendering it independent of the applied C-rate and applicable after nearly every charging or driving event. Validated on BMW i3 fleet data, the ΔQ -method achieved a mean absolute SOH error of 2.52% and an OCV reconstruction error of 7.19 mV.

Despite this progress, all existing mechanistic approaches share a common practical constraint: they require a *reference capacity* — either from a testbench measurement or a trusted onboard estimate — to anchor the stoichiometric fitting and to derive the SOH from the identified alignment parameters [25,27–30]. Moreover, mechanistic approaches typically require low C-rates and wide state-of-charge (SOC) windows: for the cell investigated in [25], reliable DM estimation demanded charging at $\leq C/15$, with a 20%–70% SOC window sufficient only at C/30, whereas capacity estimation alone remained accurate

up to $\sim C/4$ over a 10%–80% window. These operating requirements are rarely met in real-world EV operation, where charging events are partial, driving patterns are uncontrolled, and the SOC distribution is heavily skewed toward the 70%–90% range. The methods further presuppose accurate half-cell OCP reference curves; because these are temperature-dependent, a mismatch between the operating and characterization temperatures biases the identified stoichiometries. A more fundamental difficulty is the limited identifiability of the inverse problem itself: the relaxed-voltage charge increment constrains only the *difference* of the bounding stoichiometries, so the alignment parameters are under-determined, admitting a family of rigidly shifted OCV curves (Section 3.2). This ambiguity is normally resolved by the curvature of the mid-SOC region, but it becomes acute when field data concentrate in the narrow, low-curvature high-SOC range. The dependence of degradation-mode identifiability on the available SOC window has been established analytically [31], and the ill-posedness of such inverse problems is well documented [32].

Field data from production EVs present additional challenges: the absence of laboratory-grade ground-truth measurements, noise from commercial-grade sensors, and significant vehicle-to-vehicle variability in usage patterns [16,33], motivating a growing body of work on real-world EV health diagnostics [34–38]. Although several studies have addressed SOH estimation from real-world EV data using data-driven methods [17,39–41], and recent work has reconstructed the OCV–SOC curve from onboard or field data for state and capacity estimation [42–44], no prior work has applied a full mechanistic OCV reconstruction framework to unstructured field EV data without any external capacity reference. The present work bridges this gap: it retains the ΔQ -method as its mechanistic core but removes the method’s last external dependency — the reference-capacity anchor — by generating an internal capacity reference from field EV data alone, propagating it to electrode-level degradation modes, and testing each reconstructed state out-of-sample against an independent single-particle model with electrolyte (SPMe). Throughout this work, “reference-free” denotes the absence of any aged-state capacity reference during operation: no periodic reference performance test, laboratory capacity check, or trusted onboard SOH estimate is required at any point in the vehicle’s life. The only prior inputs are the one-time BOL characterization — the BOL capacity and the fresh half-cell OCP curves — obtained at the manufacturing stage and never re-measured; the sensitivity of the pipeline to tolerances in these BOL inputs is quantified in Section 4.3. The research gap addressed here is thus explicit: existing electrode-level mechanistic diagnoses require an in-life reference capacity that field-operated EVs do not provide. The contributions of this work are threefold: (i) a two-stage pipeline that generates an internal capacity reference from field data alone and propagates it to the electrode-level degradation modes; (ii) a voltage-bin-weighted loss that corrects the SOC-skewed voltage coverage of field data; and (iii) a per-vehicle, out-of-sample cross-validation of the reconstructed modes against an independent SPMe.

Specifically, we propose a reference-free, two-stage pipeline. In the first stage, an SOH trajectory is estimated directly from partial charge/discharge segments by comparing measured charge increments against the initial-cell OCV curve, with subsequent Gaussian process regression (GPR) smoothing [45] to produce a physically consistent capacity trend. In the second stage, the smoothed SOH trajectory serves as the capacity reference for the ΔQ -method, in which the stoichiometric alignment parameters are identified by minimizing a *voltage-bin-weighted* loss function that compensates for the uneven SOC

distribution inherent in field data. To constrain the otherwise ill-posed reconstruction, the pipeline further combines this weighting with a low-SOC boundary anchor and a physical regularization term, enabling electrode-level stoichiometric limits to be identified even under the narrow voltage coverage of field data—a robustness not required of laboratory OCV reconstruction with full-range coverage.

The remainder of the paper is organized as follows. Section 2 describes the field dataset and segment extraction employed in this study. Section 3 presents the proposed two-stage methodology and the electrochemical cross-validation procedure. Section 4 reports the vehicle-level results and the independent electrochemical cross-validation, interprets the estimated degradation modes, and presents an ablation and sensitivity analysis before discussing limitations and future work. Section 5 concludes the paper.

2. Field dataset and segment extraction

High-resolution, multi-year time-series data were collected from three instrumented production EVs, denoted VIN 1, VIN 2, and VIN 3, and used for both methodology development and longitudinal degradation analysis. In the absence of laboratory-grade ground-truth measurements, such as standard OCV tests or reference-capacity measurements, all battery health indicators presented in this work were inferred from raw operational records from the field. The three vehicles were of the same model, equipped with nickel–manganese–cobalt (NMC)/graphite lithium-ion cells, and operated on public roads in South Korea. Specific cell and pack specifications are withheld under a non-disclosure agreement; however, the methodology is formulated in terms of the normalized health indicator

$$\text{SOH} = \frac{Q}{Q_{\text{ref}}}, \quad (1)$$

and is therefore independent of the absolute capacity. Here, Q_{ref} denotes the BOL capacity obtained from an in-house full-charge–discharge test. All signals were recorded at a 1 Hz sampling frequency; the logged channels comprised terminal voltage and pack current, whereas neither cell nor ambient temperature was recorded.

Reference half-cell OCP curves for the positive and negative electrodes, U_p and U_n , were obtained from in-house characterization of fresh electrodes harvested from a pristine cell of the same production batch. These OCP curves served as the basis for both the SOC mapping and the pseudo-OCV reconstruction. Both the BOL capacity Q_{ref} and these OCP curves were obtained once, at the manufacturing stage, and were not re-measured at any point during the observation period.

VIN 1 exhibited anomalous rapid capacity fade, with an observation period of approximately 1.5 years, corresponding to 123 equivalent full cycles (EFC). VIN 2 and VIN 3 served as normal-aging references, each spanning approximately 4 years of operation (366 and 296 EFC, respectively). Although VIN 2 and VIN 3 shared comparable observation durations, VIN 3 accumulated fewer EFC owing to lower driving intensity. VIN 3 also exhibited the lowest SOH at the start of the observation period among the three vehicles, providing a complementary baseline for evaluating the method under more advanced degradation conditions. Mileage records were not available for the three instrumented vehicles.

Given that direct measurement of the OCV is infeasible in uncontrolled field environments, relaxed-voltage points were extracted following periods of vehicle inactivity. A relaxed-voltage point is operationally defined as the terminal voltage measured at the onset of an operating event (e.g., trip commencement or charging start) following a rest period. Each pair of temporally consecutive relaxed-voltage points, together with the net charge accumulated between them, constitutes a *segment* (v_{start} , v_{end} , ΔQ). These segments serve as the fundamental input units for both the SOH estimation and the pseudo-OCV reconstruction.

Because the raw extraction yielded a mixture of near-equilibrium measurements and residual non-equilibrium points—particularly those recorded during or shortly after active charging—a point-level data-quality screening was applied as the first filtering step. The minimum orthogonal distance from each point to the reference OCV curve was

Table 1

Counts of relaxed-voltage events as a function of the minimum rest-duration threshold.

Vehicle	Total	≥ 0.5 h	≥ 1.0 h	≥ 1.5 h	≥ 2.0 h	≥ 4.0 h
VIN 1	1233	420	314	250	207	106
VIN 2	3432	2270	1169	973	905	786
VIN 3	3813	2261	1783	1559	1316	733

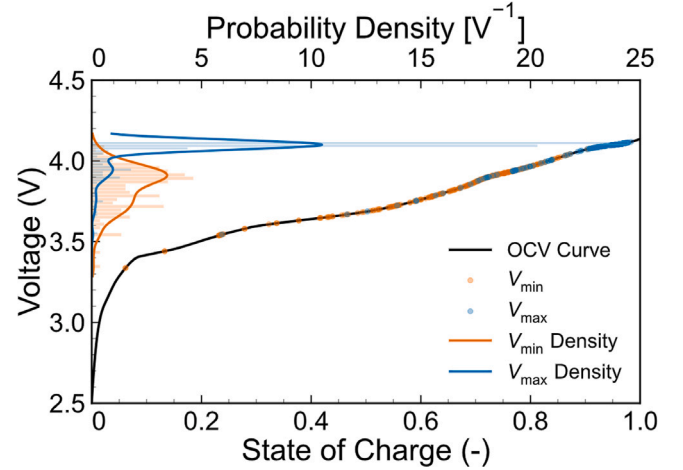


Fig. 1. Distributions of the segment endpoint voltages v_{max} (upper) and v_{min} (lower) mapped to the SOC domain via the reference OCV curve.

computed in a normalized SOC–voltage space (both axes scaled to $[0, 1]$), and points whose distance exceeded $d_{\text{tol}} = 0.05$ were discarded. The threshold was applied uniformly across all vehicles and analysis windows. The majority of discarded points were concentrated near the upper voltage limit (~ 4.2 V); the underlying causes and the effect of the screening on the data distribution are discussed in [Appendix A](#).

A minimum rest-duration threshold of $t_{\text{rest}} \geq 0.5$ h was then imposed on the retained points to ensure sufficient voltage relaxation toward thermodynamic equilibrium. [Table 1](#) summarizes the number of valid events as a function of the rest-duration threshold. Although increasing the threshold (e.g., $t_{\text{rest}} \geq 1.0$ h) brought the measured voltages closer to the true equilibrium, it sharply reduced the number of available data points, which could compromise the continuity required for longitudinal performance analysis. Therefore, $t_{\text{rest}} \geq 0.5$ h was adopted as a practical trade-off between relaxation fidelity and data density.

To assess the operational coverage of the extracted data, the relaxed voltages were mapped to the SOC domain using the reference OCV curve. For each segment, the higher and lower endpoint voltages are denoted v_{max} and v_{min} , respectively. [Fig. 1](#) shows the distributions of v_{max} and v_{min} separately. The v_{max} values were concentrated near ~ 4.2 V (corresponding to $\sim 100\%$ SOC), because these points were predominantly recorded after a charging event—either at the onset of a subsequent trip or during a rest period following charge termination. In contrast, the v_{min} values were distributed primarily between 70% and 90% SOC (approximately 3.6–4.0 V), reflecting voltages captured before charging or after driving. This asymmetry is a direct consequence of typical EV user behavior: frequent top-up charging and avoidance of deep discharges. A data distribution heavily skewed toward specific SOC intervals can introduce bias into the reconstruction and compromise the identification of degradation behavior across the entire operational range. To address this sparsity in the low-voltage region and to ensure robust inference, we incorporated a voltage-bin-weighted loss function into the optimization, as detailed in the following section.

3. Methodology

The proposed framework builds upon the mechanistic modeling approach introduced by Hofmann et al. [27], referred to as the ΔQ -

method. In that method, the full-cell OCV is described as the difference of two half-cell OCPs, U_p and U_n , whose stoichiometric ranges are parameterized by four alignment parameters. Given a set of relaxed-voltage points $\{V_1, V_2, \dots, V_N\}$ and the accumulated charge between consecutive points $\{\Delta Q_1, \Delta Q_2, \dots, \Delta Q_{N-1}\}$, the alignment parameters are identified by minimizing the discrepancy between the measured and reconstructed ΔQ values. The method is independent of C-rate and applicable after almost every charging or driving event, making it well suited for field deployment.

A key requirement of the original ΔQ -method is access to a reference capacity (Section 1) to constrain the optimization search space and to derive the SOH from the fitted alignment parameters. Here, “reference-free” denotes the absence of any aged-state capacity reference during operation; the pipeline still relies on beginning-of-life electrode characterization (the fresh-cell OCP curves and Q_{ref}), which is available at the manufacturing stage and requires no per-vehicle field measurement. In many real-world fleet scenarios, however, such reference measurements are unavailable or infrequent, and the onboard SOH estimate itself may lack the accuracy required for electrode-level diagnosis. To address this limitation, the present work introduces a two-stage pipeline: the first stage estimates an SOH trajectory directly from the field data — by comparing measured charge increments against the initial-cell OCV curve and smoothing the resulting estimates with GPR — and the second stage uses this trajectory as the capacity reference for the mechanistic OCV reconstruction in each optimization window. This decoupled design removes the dependence on external capacity measurements and enables degradation diagnosis from field data alone.

Fig. 2 provides a schematic overview of the pipeline. The inputs — relaxed-voltage pairs $(v_{\text{start}}, v_{\text{end}})$, current profiles, and the accumulated charge ΔQ — are first filtered by thresholds on the minimum voltage swing and charge throughput. Stage1 estimates an SOH trajectory from these filtered segments and smooths it via GPR. Stage2 takes the smoothed SOH as its capacity reference and reconstructs the pseudo-OCV curve by fitting the stoichiometric alignment parameters, yielding the SOH together with the electrode-level degradation-mode indicators (LLI, LAM_p, LAM_n). Information flows strictly from Stage 1 to Stage 2; no feedback path exists. The remainder of this section details the two stages of the pipeline: preliminary SOH estimation (Section 3.1) and pseudo-OCV reconstruction (Section 3.2).

3.1. Preliminary SOH estimation from field data

The fundamental inputs are partial charge/discharge segments $\{v_{\text{start},i}, v_{\text{end},i}, \Delta Q_i, t_i\}$ extracted from field driving data. Since segments with negligible SOC variation provide insufficient information for capacity estimation and are highly susceptible to measurement noise, a two-step filtering process was applied:

$$|v_{\text{start}} - v_{\text{end}}| > V_{\text{thr}}, \quad (2)$$

$$|\Delta Q| > Q_{\text{thr}}, \quad (3)$$

where $V_{\text{thr}} = 350$ mV is the minimum voltage swing requirement and $Q_{\text{thr}} = 0.1 Q_{\text{ref}}$ is a charge throughput threshold defined as a fraction of the reference capacity. This restricted the estimation to segments with sufficient SOC spans.

For each valid segment, a local SOH value was estimated by comparing the observed ΔQ_i with the theoretical capacity difference $Q_{\text{ref}} \Delta z_{\text{ref},i}$ derived from the initial-cell OCV curve, recalling the SOH definition in Eq. (1). The estimation was performed sequentially over the filtered segments. Let $z_{\text{ref}}(v) = \text{OCV}_{\text{ref}}^{-1}(v)$ denote the inverse of the initial-cell OCV curve. For a given voltage pair $(v_{\text{start},i}, v_{\text{end},i})$, the reference SOC difference is

$$\Delta z_{\text{ref},i} = z_{\text{ref}}(v_{\text{end},i}) - z_{\text{ref}}(v_{\text{start},i}). \quad (4)$$

The local SOH estimate for segment i is then obtained as

$$\widehat{\text{SOH}}_i = \frac{\Delta Q_i}{Q_{\text{ref}} \Delta z_{\text{ref},i}}. \quad (5)$$

Because the initial-cell OCV is monotonic over the relevant voltage range, the inverse z_{ref} is single-valued and Eq. (5) is evaluated directly for each segment. To ensure temporal consistency, each local SOH estimate was checked against that of the preceding segment, and physically implausible jumps were rejected before GPR smoothing. The local SOH estimates were then processed using GPR with a radial basis function (RBF) kernel combined with a white noise kernel. The composite kernel is defined as

$$\mathcal{K}(x_i, x_j) = \sigma_f^2 \exp\left(-\frac{(x_i - x_j)^2}{2l^2}\right) + \sigma_n^2 \delta_{ij}, \quad (6)$$

where x_i denotes the cumulative charge throughput at segment i , l is the length scale, σ_f^2 is the signal variance, σ_n^2 is the noise variance, and δ_{ij} is the Kronecker delta. The length-scale bounds were set adaptively: the lower bound was defined as $\max(1, \Delta x_{\text{span}}/5)$ and the upper bound as $\max(10, 2\Delta x_{\text{span}})$, where Δx_{span} is the span of the input domain. All hyperparameters were optimized by maximizing the log marginal likelihood with ten random restarts. This procedure yielded a physically consistent and continuous SOH trajectory (SOH_{GPR}) that served as the capacity input for the subsequent pseudo-OCV reconstruction.

3.2. Pseudo-OCV reconstruction

Unlike the preliminary SOH estimation stage, the pseudo-OCV reconstruction used all available relaxed-voltage segments, without applying the filtering criteria of Eqs. (2)–(3). Because the identification of stoichiometric limits relies on subtle curvature features of the OCV curve, a dense and broadly distributed set of voltage–charge pairs was essential to constrain the inverse problem. However, as illustrated in Fig. 1, real-world segments were heavily concentrated in the high-SOC region (approximately 70%–90% SOC), reflecting typical EV charging behavior. This distributional imbalance was mitigated by a weighting scheme in the loss function, detailed below.

The cell’s thermodynamic state was described by reconstructing the pseudo-OCV from reference half-cell open-circuit potentials, U_p and U_n , obtained from characterization of fresh electrodes. Each electrode potential is expressed as a function of its normalized stoichiometry $\theta_m \in [0, 1]$, defined as the degree of delithiation of electrode m . With increasing cell SOC z , the positive electrode is delithiated and the negative electrode is lithiated; accordingly θ_p increases and θ_n decreases with z , as expressed in Eqs. (7)–(8).

$$\theta_p(z) = \theta_{p,\min} + W_p z, \quad (7)$$

$$\theta_n(z) = \theta_{n,\min} + W_n (1 - z), \quad (8)$$

where $W_m = \theta_{m,\max} - \theta_{m,\min}$ is the stoichiometric window width of electrode m . The full-cell pseudo-OCV is then reconstructed as

$$\text{OCV}(z; \Theta) = U_p(\theta_p(z)) - U_n(\theta_n(z)), \quad (9)$$

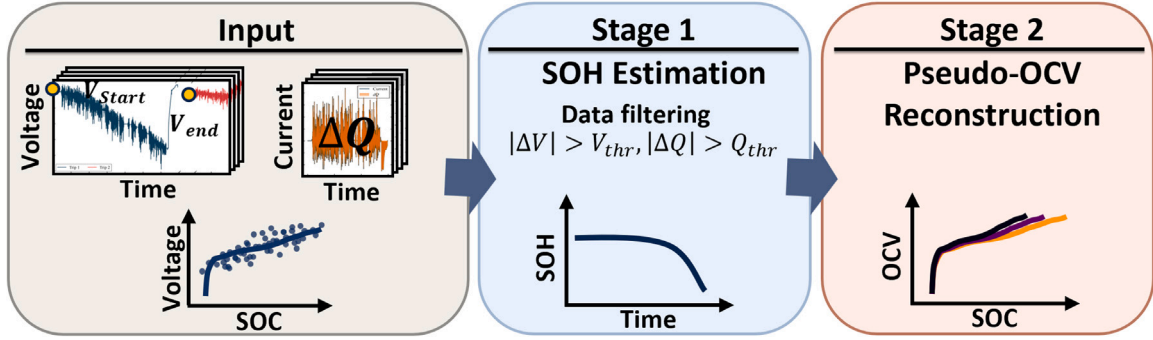
where $\Theta = (\theta_{p,\min}, \theta_{p,\max}, \theta_{n,\min}, \theta_{n,\max})$ collects the four stoichiometric limits to be identified. At the beginning of life, Θ was initialized from the fresh-electrode characterization data.

To track electrode-level degradation over time, data points were grouped into overlapping windows based on the EFC metric. A window spanning two equivalent full cycles ($\text{EFC}_{\text{window}} = 2$) and a stride of one cycle ($\text{EFC}_{\text{stride}} = 1$) were employed, where one EFC corresponds to a cumulative charge throughput of Q_{ref} . To ensure statistical reliability, each window was required to contain at least 30 data points ($N \geq 30$); the window width was dynamically expanded if this criterion was not met. The effective window capacity Q_{window} was determined by the mean of the SOH_{GPR} values within the window:

$$Q_{\text{window}} = Q_{\text{ref}} \cdot \frac{1}{N} \sum_{i=1}^N \text{SOH}_{\text{GPR},i}, \quad (10)$$

where N is the number of data points in the current window. Because Q_{window} is derived directly from the Stage1 output SOH_{GPR} , any systematic bias in the preliminary capacity estimate propagates into the

I. Diagnostic pipeline(no in-life reference cycle)



II. Electrochemical cross – validation

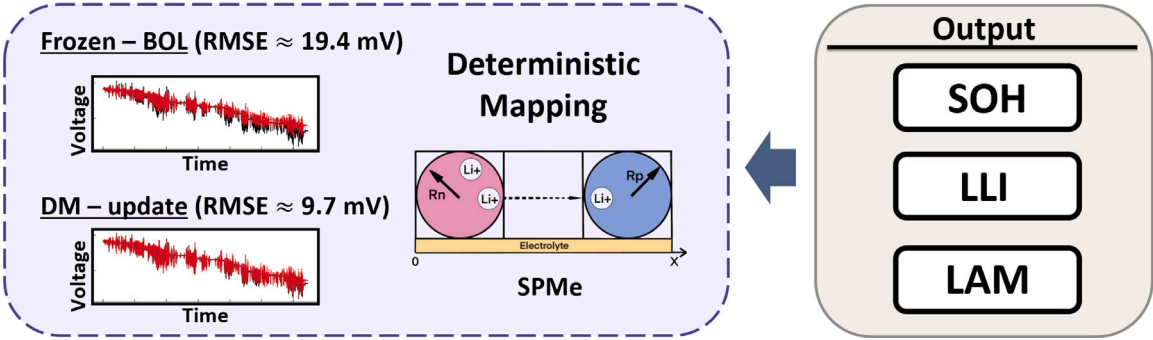


Fig. 2. Overview of the proposed framework. (I) Diagnostic pipeline: field voltage, current, and accumulated-charge (ΔQ) records pass through Stage 1 (SOH estimation) and Stage 2 (pseudo-OCV reconstruction) to yield the SOH and the degradation modes (LLI, LAM), requiring no laboratory reference. (II) Electrochemical cross-validation (offline, optional): the degradation modes are mapped deterministically onto an SPM, without re-identification from the driving data, to predict the measured driving voltage and support their physical validity.

Stage2 stoichiometric identification. The quantitative impact of this coupling is analyzed in Section 4.3.

The stoichiometric limits θ were identified by minimizing the total loss function

$$\mathcal{L} = \mathcal{L}_{\text{data}} + \lambda \mathcal{L}_{\text{phys}}, \quad (11)$$

where $\lambda = 100$ is a fixed regularization weight. $\mathcal{L}_{\text{data}}$ is typically of order 10^0 Ah, whereas $\mathcal{L}_{\text{phys}}$, formed from squared stoichiometric increments with $\theta \in [0, 1]$, is of order 10^{-2} ; λ was chosen to bring the two terms to comparable magnitude.

The data fidelity term $\mathcal{L}_{\text{data}}$ was defined as the weighted root-mean-square error between the measured and predicted charge increments. Because the relaxed-voltage segments are unevenly distributed across the voltage range (see Section 3.2), a voltage-bin weighting scheme was applied. For each segment, the reference voltage was taken as $v_{\min,i} = \min(v_{\text{start},i}, v_{\text{end},i})$. The range of $v_{\min}$ values within the current window was partitioned into five equal-width bins in $v_{\min}$ (equivalently, in SOC), and each segment was assigned a raw weight $\tilde{w}_i = 1/\sqrt{n_{\text{bin},i}}$, where $n_{\text{bin},i}$ is the count of segments in the bin to which the i th segment belongs. The weights were subsequently normalized so that their mean equals unity: $w_i = \tilde{w}_i / \bar{\tilde{w}}$. This reduced the influence of densely populated high-SOC bins and prevented the optimizer from fitting only the dominant voltage cluster. The resulting data-fidelity term is

$$\mathcal{L}_{\text{data}} = \sqrt{\frac{\sum_{i=1}^N w_i (\Delta Q_i - \Delta \hat{Q}_i)^2}{\sum_{i=1}^N w_i}}, \quad (12)$$

where ΔQ_i is the measured charge throughput of the i th segment, $\Delta \hat{Q}_i$ is the corresponding prediction from the reconstructed pseudo-OCV, and N is the number of valid segments within the current window.

The predicted charge increment $\Delta \hat{Q}_i$ was obtained as follows. Given the reconstructed pseudo-OCV curve $\text{OCV}(z; \theta)$ from Eq. (9), the inverse mapping $\hat{z} = \text{OCV}^{-1}(v; \theta)$ was used to obtain the corresponding state-of-charge values $\hat{z}_{\text{start},i}$ and $\hat{z}_{\text{end},i}$ for each segment with observed terminal voltages $(v_{\text{start},i}, v_{\text{end},i})$. The predicted charge increment was then calculated as

$$\Delta \hat{Q}_i = Q_{\text{window}} (\hat{z}_{\text{end},i} - \hat{z}_{\text{start},i}), \quad (13)$$

where Q_{window} is the effective window capacity defined in Eq. (10).

The physical regularization term $\mathcal{L}_{\text{phys}}$ penalizes abrupt inter-window changes in the stoichiometric window width W_m and the window center C_m , defined as

$$W_m = \theta_{m,\max} - \theta_{m,\min}, \quad C_m = \frac{\theta_{m,\max} + \theta_{m,\min}}{2}, \quad m \in \{p, n\}. \quad (14)$$

Because field data contain substantial measurement noise and non-uniform voltage coverage, unconstrained window-by-window optimization of θ can produce spurious jumps in W_m and C_m . To suppress such artifacts, the penalty acts on the change in these quantities between consecutive windows:

$$\mathcal{L}_{\text{phys}} = \sum_{m \in \{p, n\}} \left[(C_m^{(k)} - C_m^{(k-1)})^2 + (W_m^{(k)} - W_m^{(k-1)})^2 \right], \quad (15)$$

where superscript (k) denotes the current window and $(k-1)$ the preceding window. The penalty is symmetric with respect to the sign of the change, i.e., no directional constraint is imposed, so that the regularization does not bias the estimated degradation trajectory toward any particular mode. For the initial window ($k = 1$), $C_m^{(0)}$ and $W_m^{(0)}$ were set to the fresh-cell values, and optimization then proceeded sequentially across windows, each initialized from the optimal parameters of the preceding window.

A structural challenge of the inverse problem is that the predicted ΔQ between two relaxed-voltage points depends only on the difference

of the corresponding SOC values, not on their absolute positions. Consequently, the four stoichiometric limits admit a family of OCV curves that can shift rigidly along the SOC axis without substantially changing the local ΔQ predictions (translational degeneracy). In principle, the curvature features of the OCV — in particular the transitions between graphite staging plateaus in the mid-SOC range — can resolve this ambiguity. However, as noted above, the field data concentration in the 70%–90% SOC range provides limited curvature information, and the mid-SOC transition region that carries the most discriminative features is precisely the range with the fewest observations.

To mitigate this ill-posedness, we adopted three complementary measures. First, a boundary constraint ($OCV = 2.5$ V at $z = 0$) was enforced to anchor the low-SOC end of the reconstructed curve to the physical discharge cutoff, thereby removing the translational degeneracy. Second, the physical regularization term $\mathcal{L}_{\text{phys}}$ in Eq. (15) penalized abrupt inter-window parameter changes. Third, sequential initialization from the preceding window provided a warm start that biased the optimizer toward physically continuous trajectories. In addition, a per-window acceptance criterion was applied: if the weighted R^2 between the measured and reconstructed charge increments fell below a threshold ($R_{\text{thr}}^2 = 0.95$), the stoichiometric parameters of that window were rejected and replaced by those of the most recent accepted window, preventing poorly constrained fits from propagating into the degradation-mode trajectory; the per-window statistics in Table 3 are reported prior to this gating. For VIN 1, all 28 windows attained $R^2 > 0.8$ (median 0.93, none below 0.5), so its degradation-mode trajectory rests on a continuously well-constrained set of windows despite the small window count. Although these measures provided sufficient constraint for the majority of EFC windows, residual non-uniqueness under sparse or narrowly distributed voltage coverage could not be fully excluded.

The optimization was solved with the Sequential Least-Squares Programming (SLSQP) algorithm implemented in `scipy.optimize.minimize`, with a maximum of 400 iterations and a function tolerance of 10^{-9} . Each stoichiometric limit was box-constrained to $\theta_{m,\min} \in [0, 0.3]$ and $\theta_{m,\max} \in [0, 1]$ for $m \in \{p, n\}$. The boundary constraint $OCV(0; \theta) = 2.5$ V was imposed as an equality constraint within the SLSQP formulation.

The SOH reported by the pipeline was the Stage 1 estimate SOH_{GPR} ; Stage 2 did not independently re-derive the capacity but instead decomposed the degradation into electrode-level degradation-mode indicators, defined as follows. Loss of active material at each electrode was quantified by the relative change in stoichiometric window width with respect to the fresh-cell value:

$$LAM_m = \frac{W_m^{(0)} - W_m^{(k)}}{W_m^{(0)}}, \quad m \in \{p, n\}. \quad (16)$$

LLI was defined as the relative decrease in the cyclable lithium capacity C_{lit} . For NMC/graphite cells with an oversized anode (negative-to-positive capacity ratio $N/P > 1$), the cathode limits the charge direction and the anode limits the discharge direction throughout most of the usable lifetime [46]. Under this primary overhang configuration, C_{lit} reduces to [27]

$$C_{\text{lit}} = (\theta_{p,\max} - \theta_{n,\min}) Q_{\text{ref}}, \quad (17)$$

and the LLI is given by

$$LLI = \frac{C_{\text{lit}}^{(0)} - C_{\text{lit}}^{(k)}}{C_{\text{lit}}^{(0)}}. \quad (18)$$

The general definition covering alternative overhang configurations is detailed in Eq. (15) of [27] and was adopted in this work. A graphical illustration of the three indicators is provided in Appendix B.

3.3. Cross-validation via the single-particle model with electrolyte

The SPMe serves purely as an independent, offline verification module; it is not part of the diagnostic pipeline and plays no role in estimating SOH or the degradation modes. The reference-free property of the diagnosis (Sections 3.1–3.2) is thus preserved—the model only tests, after the fact, whether the field-derived indicators are electrochemically self-consistent. To assess the physical validity of the reconstructed degradation modes independently of the relaxed-voltage data from which they were derived, the Stage 2 indicators were mapped deterministically onto the SPMe and used to predict the measured driving voltage. The standard SPMe formulation — solid- and electrolyte-phase diffusion, Butler–Volmer kinetics, and the terminal-voltage expression — was implemented in an in-house code; the governing equations are summarized in Appendix C. Because the degradation-mode indicators were extracted solely from relaxed-voltage and charge-increment records, never from the driving voltage, the comparison constitutes a cross-modality test of the reconstruction. The SPMe and the Stage 2 reconstruction share the same fresh-electrode OCP curves; the test is therefore out-of-sample — independent of the driving data and of the relaxed-voltage records — but rests on the same thermodynamic reference. What the test examines is whether the electrode-level shifts identified in Stage 2, applied without re-fitting, reproduce the dynamic driving voltage through the full kinetic and transport model—a relationship that the bare OCP curves alone cannot probe.

The BOL parameter set combined manufacturer-supplied values with eight parameters identified from BOL driving data: the solid-phase diffusivity, reaction-rate constant, active-material volume fraction, and porosity of each electrode. These eight parameters were fitted by a genetic algorithm (GA) that minimized the root-mean-square error between the SPMe-predicted and measured terminal voltage over BOL driving segments [47], with each parameter constrained to a physically motivated range to limit non-uniqueness; this is a one-time calibration of the validation model, and the aged-state evaluation introduces no further fitting. Table 2 lists the BOL parameters and their provenance. Absolute parameter values are withheld under the non-disclosure agreement governing the cell specifications.

The aged-state model was constructed by perturbing the BOL parameters according to the Stage 2 degradation-mode indicators, without any re-identification from the driving data. Loss of active material was applied as a proportional reduction of the active-material volume fraction at each electrode,

$$\varepsilon_{s,m}^{(k)} = (1 - LAM_m^{(k)}) \varepsilon_{s,m}^{\text{BOL}}, \quad m \in \{p, n\}, \quad (19)$$

consistent with the geometric interpretation of LAM_m as a contraction of the electrode stoichiometric window defined in Eq. (16) [48]. The negative-electrode porosity was scaled by the lithium-inventory loss,

$$\varepsilon_{e,n}^{(k)} = (1 - LLI^{(k)}) \varepsilon_{e,n}^{\text{BOL}}, \quad (20)$$

under the simplifying assumption that the solid–electrolyte interphase (SEI) growth responsible for the LLI fills the negative-electrode pore space in equal proportion [49]. The loss of cyclable lithium and the pore-volume reduction are not, in general, identical: the solid volume produced per mole of lithium consumed is governed by the molar volumes of the specific SEI decomposition products, which differ by nearly an order of magnitude between inorganic and organic species [50,51]. A linear closure with a single effective coefficient is nonetheless standard practice in physics-based degradation models [48]. In the absence of an independent measurement of the SEI composition from field data, the parsimonious one-to-one scaling was adopted. Its effect on the validation is quantified in Section 4.2. The positive-electrode porosity was held at its BOL value, and the electrode stoichiometric windows were set directly from the limits θ identified in Stage 2. In this baseline DM-updated configuration, the solid-phase diffusivity, the reaction-rate constant, and the lumped contact resistance were held at their BOL values, so that kinetic and resistive aging were not represented; their omission is reflected in the residual prediction error analyzed in Section 4.2.

Table 2

BOL parameters of the SPMe and their provenance, with electrode index $m \in \{p, n\}$. Absolute values are withheld under a non-disclosure agreement.

Parameter	Source
Solid-phase diffusivity $D_{s,m}$	Fitted (GA)
Reaction-rate constant k_m	Fitted (GA)
Active-material volume fraction $\varepsilon_{s,m}$	Fitted (GA)
Porosity $\varepsilon_{p,m}$	Fitted (GA)
Electrode thickness, particle radius	Manufacturer
Electrolyte transport properties	Manufacturer
Stoichiometric window θ	Stage 2 output (no re-fitting)

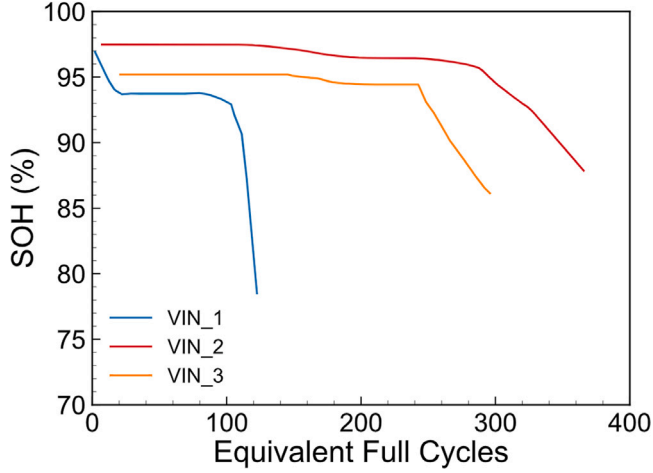


Fig. 3. Estimated SOH_{GPR} trajectories for VIN1, 2, and 3 as a function of equivalent full cycles.

4. Results and discussion

The proposed two-stage pipeline was applied to the longitudinal vehicle-level dataset (VIN1, 2, and 3) described in Section 2. VIN1 exhibited anomalous rapid capacity fade, whereas VIN2 and 3 followed typical aging patterns.

4.1. Vehicle-level analysis

Fig. 3 presents the estimated SOH_{GPR} trajectories for VIN1, 2, and 3. After outlier rejection and GPR smoothing [45], the resulting trajectories were physically consistent. VIN2 exhibited the highest SOH at the start of observation (97.47%) and the slowest degradation rate, remaining above 96% for the first 36 months before declining to 87.85% by the end of the 48-month observation period (366 EFC). VIN3, which started at the lowest such value (95.20%), showed a gradual decline over 296 EFC followed by an accelerated drop to 86.14%. In contrast, VIN1 displayed a markedly different trajectory: from a starting SOH of 97.31%, the capacity faded steeply at approximately 6 months (~ 100 EFC), and the SOH dropped to 78.51% by 123 EFC.

The smoothed SOH trajectories were then used as the capacity reference for the pseudo-OCV reconstruction (Stage 2). **Fig. 4** illustrates the evolution of the reconstructed pseudo-OCV curves for VIN3, the vehicle with the broadest voltage coverage. **Fig. 4(a)** shows the reference half-cell OCPs (U_p , U_n) with the identified stoichiometric endpoints migrating over successive EFC windows, and **Fig. 4(b)** displays the corresponding full-cell pseudo-OCV curves, where each curve represents one EFC window. As the battery aged, the reconstructed OCV progressively shifted, reflecting the gradual narrowing of the electrode stoichiometric windows. To quantify the reconstruction fidelity, the coefficient of determination (R^2) between the measured and predicted charge increments was evaluated for each EFC window. **Table 3** summarizes the per-vehicle statistics. All three vehicles achieved median R^2

Table 3

Pseudo-OCV reconstruction quality per vehicle. Statistics are computed over all EFC windows.

Vehicle	N_{window}	R^2 (mean $\pm$ std)	R^2 (median)
VIN 1	28	0.918 ± 0.055	0.932
VIN 2	201	0.906 ± 0.233	0.992
VIN 3	44	0.943 ± 0.068	0.965

values above 0.93, confirming that the reconstructed pseudo-OCV consistently captured the dominant charge–voltage relationship across the majority of EFC windows. The mean R^2 exceeded 0.9 for all vehicles; however, VIN2 exhibited a notably larger standard deviation (0.233) than those of VIN1 (0.055) and VIN3 (0.068). For VIN2, 80.6% of the 201 windows attained $R^2 \geq 0.9$ ($86.1\% \geq 0.8$); the elevated standard deviation originated from a small subset of windows ($R^2 < 0.5$ in 14 windows) concentrated at low EFC (mean EFC ≈ 64 versus ≈ 128 for the well-fitted windows), where each window contained only a few accumulated segments sparsely scattered over a comparatively wide voltage range and thus provided insufficient curvature constraint for the stoichiometric fit. Because such low- R^2 windows fell below the acceptance threshold (Section 3.2), their parameters were replaced by those of the most recent accepted window before the degradation-mode trajectory was computed; the variance in **Table 3** thus reflects the raw per-window fit quality prior to this gating. The median R^2 of 0.992 confirmed that the reconstruction was robust once sufficient data had accumulated. The identified stoichiometric limits were converted into degradation-mode indicators via Eqs. (16)–(18).

Fig. 5 presents the temporal evolution of the four stoichiometric limits. $\theta_{p,\min}$ remained relatively stable for VIN2 and VIN3, whereas VIN1 showed a gradual increase toward the end of the observation period, coinciding with the rapid SOH decline observed in **Fig. 3**. $\theta_{p,\max}$ decreased across all vehicles, reflecting a progressive increase in LAM_p . In NMC cathodes, this degradation pathway is driven by structural transitions from the layered to the spinel and rock-salt phases at high states of delithiation, often accompanied by transition-metal dissolution into the electrolyte [3,7]. $\theta_{n,\min}$ remained near zero across all vehicles and aging stages, indicating negligible sensitivity to degradation. $\theta_{n,\max}$ exhibited the largest and most consistent change: it decreased monotonically from its fresh-cell value in VIN1 and VIN3, with VIN2 showing a late-stage recovery, corresponding directly to an increase in LAM_n . In graphite anodes, this loss of active material is primarily attributed to repeated volumetric expansion and contraction during lithiation cycles, which induce particle cracking and electrical contact loss [52,53].

VIN2 exhibited an anomalous recovery in $\theta_{n,\max}$ in the late stage; however, the notably large R^2 standard deviation for VIN2 (**Table 3**) indicated that the reconstruction fidelity deteriorated in the affected windows, suggesting that this late-stage anomaly is an artifact rather than a genuine electrochemical change. Unlike the low- R^2 windows discussed above ($R^2 < 0.5$, concentrated at low EFC and rejected by the acceptance gate), these high-EFC windows passed the $R^2 \geq 0.95$ gate yet were constrained only weakly — their accepted fits clustered at $R^2 \approx 0.95$ – 0.97 over a comparatively narrow voltage span — so a residual recovery artifact persisted in the degradation-mode trajectory. Analogous fluctuations in the estimated degradation modes were reported by Hofmann et al. [27], who found that DM anomalies in the BMW i3 vehicle dataset were concentrated at checkpoints with elevated SOH- and OCV-reconstruction errors, confirming that reconstruction fidelity is a prerequisite for reliable DM tracking. A seasonal oscillation with a period of approximately 12 months was observed in $\theta_{p,\max}$ and LAM_p . This seasonal variation was attributed to the temperature dependence of electrode open-circuit potentials [54]: the OCP reference curves were obtained at a fixed temperature, so seasonal ambient fluctuations were absorbed into the identified stoichiometric limits.

Fig. 6 presents the resulting degradation-mode trajectories. LLi increased in all vehicles—monotonically in VIN1 and VIN3, with a late-stage decline in VIN2, consistent with the continuous growth of

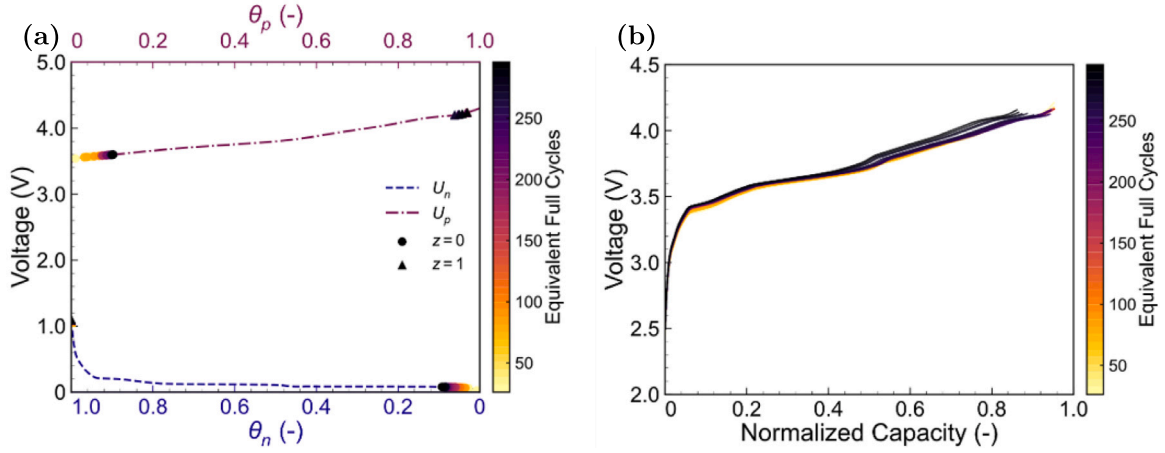


Fig. 4. Pseudo-OCV reconstruction for VIN 3. (a) Half-cell OCPs with identified stoichiometric endpoints evolving over time (color-coded by equivalent full cycles). (b) Reconstructed full-cell pseudo-OCV curves for successive EFC windows.

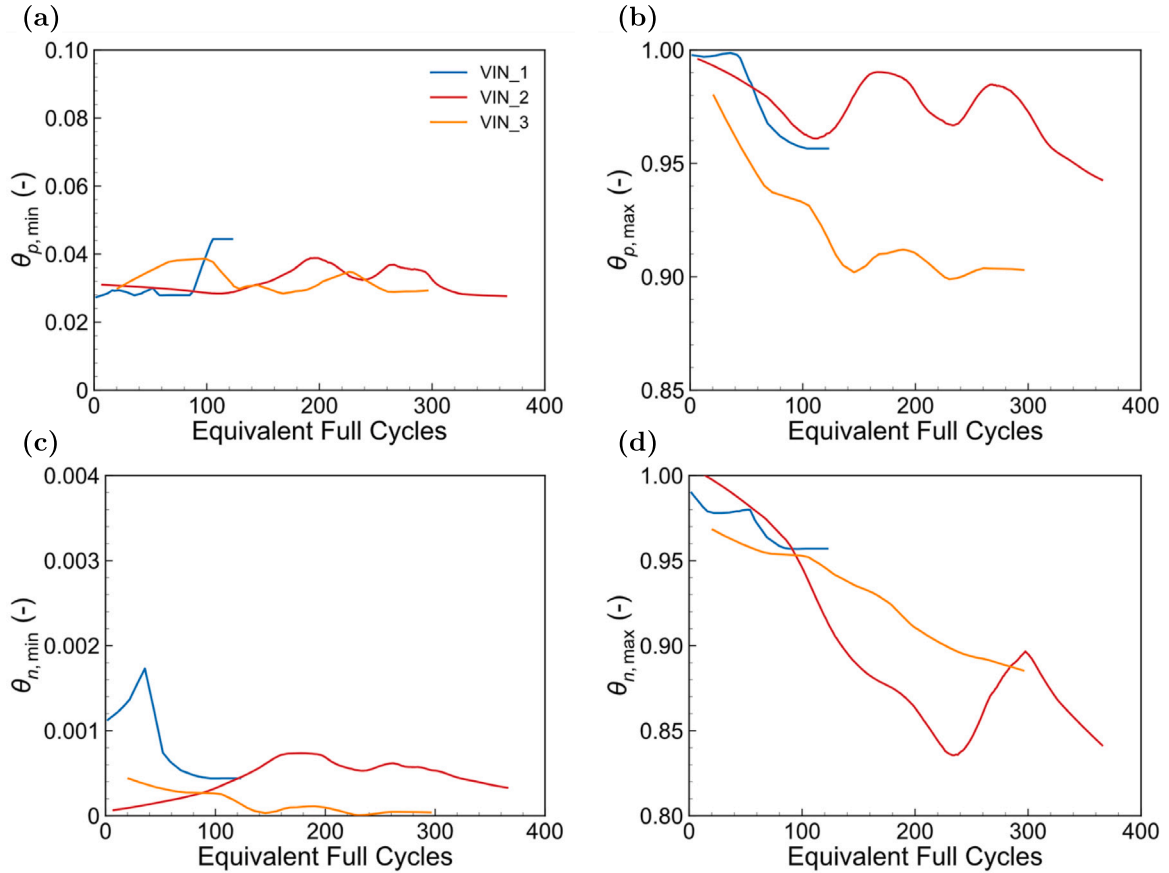


Fig. 5. Temporal evolution of the stoichiometric limits for VIN 1, 2, and 3 as a function of equivalent full cycles: (a) $\theta_{p,min}$, (b) $\theta_{p,max}$, (c) $\theta_{n,min}$, and (d) $\theta_{n,max}$.

the SEI on the graphite anode surface, which irreversibly consumes cyclable lithium [1,3]. The VIN 2 decline reflects the same late-stage fidelity artifact discussed for $\theta_{n,max}$ in Section 4.1 rather than a genuine trend. VIN 1 exhibited a sharp rise in LLI at the onset of the accelerated capacity fade (~ 100 EFC), coinciding with the SOH drop in Fig. 3. LAM_p followed a similar pattern: VIN 1 showed a sudden increase at the same point, whereas VIN 2 and VIN 3 rose more gradually. LAM_n increased in all vehicles, consistent with the $\theta_{n,max}$ trends in Fig. 5(d); VIN 2 continued to rise despite its SOH remaining above 96% for an extended period, apart from the late-stage non-monotonicity noted

above. This observation is consistent with the anode capacity overhang effect ($N/P > 1$) [46,55,56]. VIN 3, which had the lowest SOH at the start of observation among the three vehicles, exhibited higher initial values of all three degradation indicators than VIN 1 and VIN 2, consistent with its more advanced degradation state at that point.

For VIN 1, the degradation-mode composition was qualitatively similar to that of the other vehicles, suggesting that the abnormality lay in the *rate* rather than in the mechanism of degradation [57,58]. The concurrent rise in both LLI and LAM_p at ~ 100 EFC is consistent

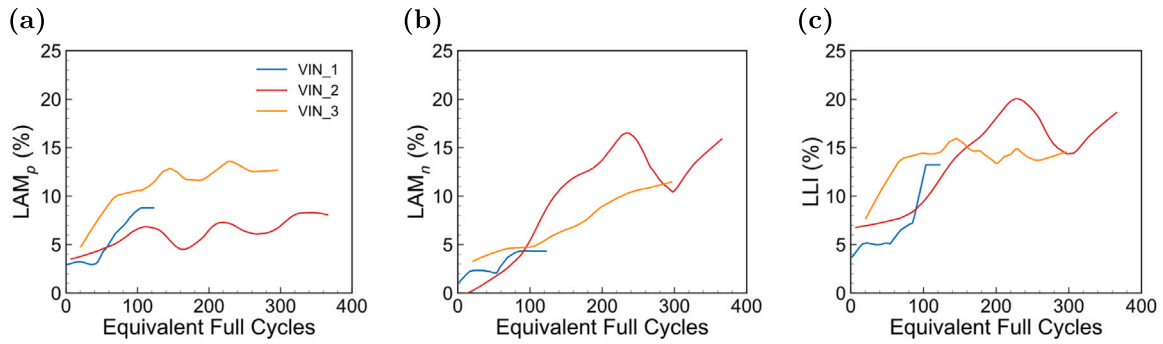


Fig. 6. Evolution of degradation-mode indicators for VIN 1, 2, and 3 as a function of equivalent full cycles: (a) LAM_p , (b) LAM_n , and (c) LLI.

with a self-reinforcing feedback loop in which accelerated lithium consumption exposes fresh electrode surface, promoting further parasitic reactions. However, the present field data did not permit identification of the specific trigger — plausible candidates included sustained high-temperature exposure, a manufacturing defect in the cell or module, or an extended period of storage at elevated SOC — and discriminating among these hypotheses would require cell-level post-mortem analysis, which was not available for this vehicle.

In summary, LLI and LAM_p tracked the estimated capacity fade closely across all three vehicles. In contrast, LAM_n increased but appeared comparatively less coupled to the SOH trajectory: VIN 2, which degraded the most slowly, accumulated the largest LAM_n among the three vehicles, consistent with the anode overhang being progressively consumed before LAM_n manifests as measurable capacity loss. Electrochemically, this hierarchy is consistent with an NMC/graphite couple operated under an oversized-anode ($N/P > 1$) design. The joint dominance of LLI and LAM_p indicates that usable-capacity fade is governed by the depletion of cyclable lithium — typically driven by parasitic lithium consumption at the negative-electrode solid-electrolyte interphase — and by the loss of positive-electrode active material, which jointly limit the accessible capacity. By contrast, the anode overhang acts as a lithium reservoir: negative-electrode active-material loss is buffered by the surplus anode capacity and therefore does not translate into measurable capacity fade until the overhang is consumed. A practical corollary is that a capacity-only SOH metric is structurally blind to LAM_n during this buffered regime, whereas the electrode-resolved indicators recovered here expose it directly, highlighting the diagnostic value of mechanistic over scalar health metrics.

4.2. Cross-validation of the reconstructed degradation modes

The reconstructed degradation modes were cross-validated against the measured driving voltage using the SPMe of Section 3.3. The test addressed a single question: whether the electrode-level shifts identified in Stage 2, when mapped onto the SPMe without any re-fitting, reproduced the dynamic driving voltage of the aged cell. For each vehicle, two configurations were compared on the same aged-stage driving segment: a frozen-BOL configuration, with all parameters held at their BOL values, and a DM-updated configuration, in which the BOL parameters were perturbed according to the Stage 2 indicators, following Eqs. (19)–(20). The only quantity that differed between the two configurations was the Stage 2 degradation-mode set, itself derived solely from relaxed-voltage and charge-increment records; no kinetic, transport, or resistance parameter was re-identified, and the driving voltage was never used in the update. Any reduction in prediction error could therefore enter through only one channel — the reconstructed OCV — which rendered the comparison an out-of-sample prediction rather than an in-sample fit.

Fig. 7 shows the predicted versus measured driving voltage for VIN 2. At the aged stage, the frozen-BOL configuration deviated from the measurement by 19.38 mV, reflecting the mismatch between the

Table 4

Driving-voltage prediction RMSE (mV) evaluated on the same aged-stage driving segment. “Frozen-BOL” uses the GA-identified BOL parameters held fixed at the aged stage; “DM-updated” perturbs them according to the Stage 2 DM indicators. Reduction is the relative decrease from the frozen-BOL case to the DM-updated case.

Vehicle	Frozen-BOL (mV)	DM-updated (mV)	Reduction (%)
VIN 1 ^a	14.09	12.15	13.8
VIN 2	19.38	9.67	50.1
VIN 3	23.33	12.96	44.4

^a Evaluated after the rapid-fade event at ~100 EFC.

fresh-cell parameters and the state of the aged cell; the DM-updated configuration reduced this to 9.67 mV. The error time series (Fig. 7(b,d)) indicates that the improvement was driven primarily by the removal of a systematic bias: the frozen-BOL residual was centered at +14.87 mV, indicating that the BOL parameters underpredicted the voltage drop of the aged cell, whereas the DM update reduced this mean offset to +4.26 mV. The residual standard deviation also decreased (12.43 → 8.67 mV), with the remaining scatter dominated by high-rate transients during dynamic driving, consistent with the unmodeled kinetic and resistive terms (Section 3.3) rather than with the degradation-mode reconstruction.

Table 4 reports the RMSE across the three vehicles. The DM update reduced the error in every case, with the aged-stage prediction errors falling within a narrow 9–13 mV band (12.15, 9.67, and 12.96 mV for VIN 1, 2, and 3), even though the three vehicles differed markedly in aging severity and started from frozen-BOL errors spanning 14–23 mV. This convergence to a common floor, obtained without any per-vehicle tuning, was the central result: a single deterministic mapping brought three differently aged cells to comparable agreement with their measured driving voltage. The relative improvement was largest for VIN 2 (50.1%) and smallest for VIN 1 (13.8%); the small VIN 1 value reflected its short post-fade driving segments. Because the prediction error accumulated over the segment, a shorter segment left less room for error growth and hence a lower frozen-BOL baseline. The small reduction was therefore a consequence of segment length rather than of the Stage 2 reconstruction.

Because the solid-phase diffusivity, the reaction-rate constant, and the lumped contact resistance were held at their BOL values, the residual error at the aged stage (9–13 mV) is attributed largely to unmodeled kinetic and resistive aging and to unrecorded temperature effects. The deterministic DM update nonetheless reduced the frozen-BOL error in every case, by up to 50% (Table 4). The sensitivity of this result to the one-to-one porosity closure of Eq. (20) was quantified by scaling the LLI-to-porosity conversion coefficient λ over {0, 0.5, 1, 2} — a factor-of-four span covering the reported spread of SEI-product molar volumes [50] — with all other parameters held fixed (Table C.2, Appendix C). For VIN 2, the aged-stage RMSE varied by less than 1.8 mV across the full range, and by only 0.13 mV between $\lambda = 0$

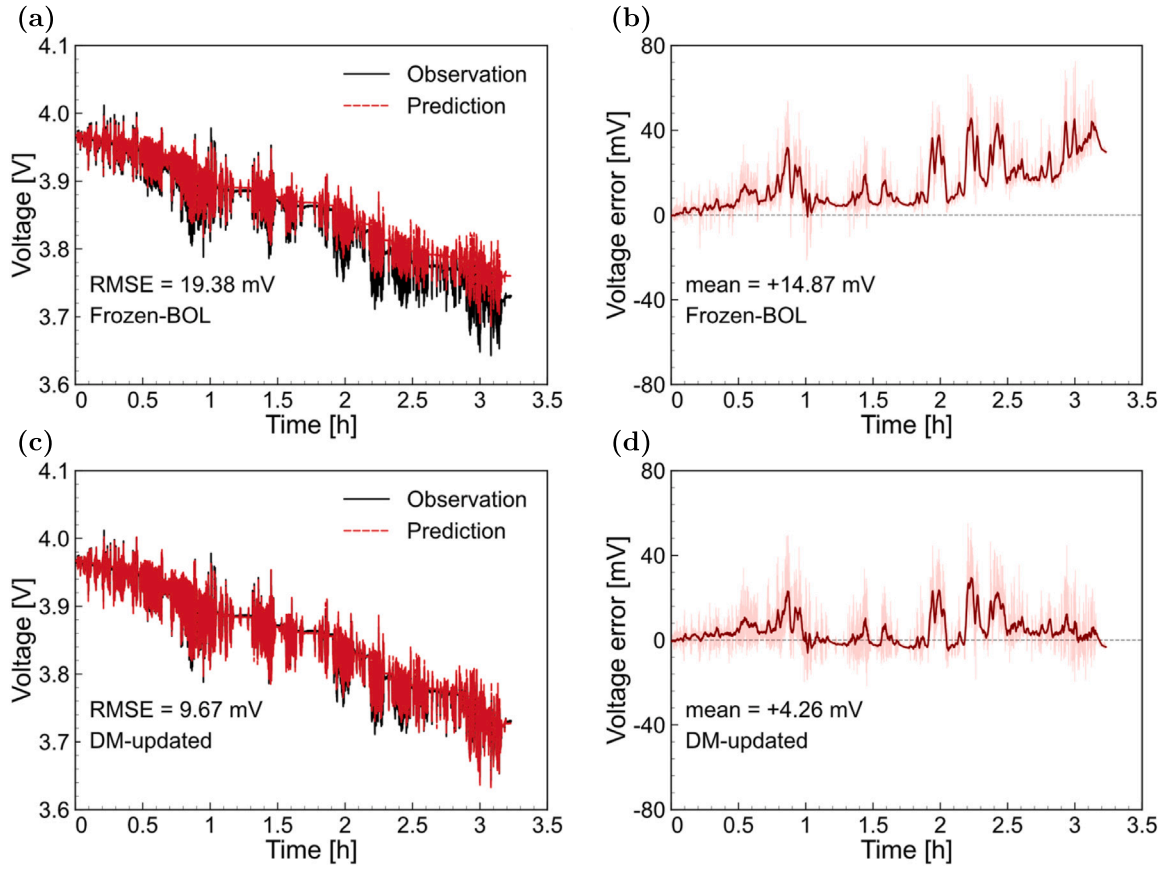


Fig. 7. SPMe-predicted versus measured driving voltage for VIN 2 at the aged life stage; (a, c) the predicted-versus-measured voltage and (b, d) the corresponding prediction-error time series, for (a, b) the frozen-BOL parameters and (c, d) the proposed degradation-mode (DM)-updated configuration. The RMSE is annotated in each panel.

(porosity update omitted entirely) and the baseline $\lambda = 1$, remaining well below the frozen-BOL error throughout; the cross-validation conclusion therefore places no weight on this closure. Re-identifying these kinetic parameters on the driving data would lower the residual further but would compromise the independence of the test. The deterministic mapping also carries a practical advantage: by requiring no per-stage re-identification, it avoids the repeated, computationally intensive parameter fitting that a conventional aging-aware electrochemical model would require at each life stage—an attractive property for fleet-scale deployment, where per-vehicle re-identification is impractical.

4.3. Effect of voltage-bin weighting and capacity-input bias

To isolate the effect of the proposed voltage-bin weighting, a controlled experiment was conducted using synthetic data with known ground truth. Because the true stoichiometric limits are prescribed, this setup also serves as an absolute benchmark of reconstruction accuracy against a reference that is otherwise unavailable for in-service production cells. Three degradation levels were imposed on the fresh-cell stoichiometry: mild ($LAM_p = 3\%$, $LAM_n = 4\%$, $SOH = 97\%$), moderate (7%, 9%, 93%), and severe (10%, 12%, 90%). The anode stoichiometry was adjusted at each level to satisfy $OCV(z=0) = 2.5$ V. For each level, 200 relaxed-voltage segments were generated by sampling from a kernel density estimate fitted to the VIN3 field-data voltage distribution (Fig. 1), thereby reproducing the characteristic concentration of observations in the high-SOC region. Gaussian noise ($\sigma_Q = 0.2$ Ah, $\sigma_V = 2$ mV) was added, and the reconstruction was repeated under uniform weights ($w_i = 1$) and the proposed bin weights ($w_i = 1/\sqrt{n_{bin,i}}$, five bins) over 50 Monte Carlo trials. All optimizer settings were identical to those in Section 3.2.

Table 5

OCV reconstruction RMSE (mV) for uniform and voltage-bin-weighted loss functions under field-like voltage distributions. Values are averaged over 50 Monte Carlo repetitions.

Degradation	SOC region	Uniform (mV)	Weighted (mV)	Reduction (%)
Mild	Low (0–0.3)	3.9	3.2	–18
	Mid (0.3–0.6)	3.9	3.0	–23
	High (0.6–1.0)	5.5	4.0	–27
Moderate	Low (0–0.3)	3.4	2.2	–35
	Mid (0.3–0.6)	3.2	2.0	–38
	High (0.6–1.0)	4.4	2.4	–45
Severe	Low (0–0.3)	2.7	2.2	–19
	Mid (0.3–0.6)	2.2	1.8	–18
	High (0.6–1.0)	3.6	2.8	–22

Table 5 reports the OCV RMSE evaluated in three SOC regions: low ($z \in [0, 0.3]$), mid ($z \in [0.3, 0.6]$), and high ($z \in [0.6, 1.0]$). The proposed weighting reduced the RMSE by 18%–45% across all regions and degradation levels (Fig. 8). The improvement was most pronounced at the moderate level, where the high-SOC RMSE decreased from 4.4 to 2.4 mV (–45%) and the mid-SOC RMSE from 3.2 to 2.0 mV (–38%). The global ΔQ fit ($R^2 > 0.999$) was virtually identical under both conditions. The difference appeared not in the aggregate fit but in the shape fidelity of the reconstructed OCV, particularly in the sparsely sampled low-SOC region.

The weighting scheme addresses the shape of the loss within a window; a separate concern is how the window's capacity input itself enters the reconstruction. The two-stage design introduces a directional coupling: a bias in the Stage 1 capacity estimate SOH_{GPR} scales Q_{window}

Table 6

Sensitivity of pseudo-OCV reconstruction to systematic bias in the capacity input Q_{window} . $\bar{\theta}_{\text{err}}$ denotes the mean absolute error of the four identified stoichiometric limits with respect to their ground-truth values. Values are averaged over three degradation levels and 50 Monte Carlo repetitions per level.

Q_{window} bias	$\bar{\theta}_{\text{err}}$	OCV RMSE (mV)	Ratio to exact
−5%	0.017	11.6	×4.5
−2%	0.008	5.2	×2.0
exact	0.003	2.6	×1.0
+2%	0.011	8.9	×3.4
+5%	0.024	21.0	×8.1

by the same factor in Eq. (10), which in turn uniformly scales all predicted charge increments ΔQ , following Eq. (13). To compensate for this mismatch, the Stage2 optimizer distorts the stoichiometric limits θ , and the resulting bias propagates into the derived degradation-mode indicators. The two stages nonetheless address distinct estimation targets: Stage 1 recovers a scalar capacity trend, whereas Stage 2 identifies the electrode-level shape parameters of the OCV curve. Because no information flows from Stage 2 back to Stage 1, the pipeline is strictly feedforward.

To assess the sensitivity of the stoichiometric identification to errors in the capacity input, the optimization was repeated with Q_{window} biased by −5%, −2%, +2%, and +5% relative to the true capacity, using the same synthetic data and weighted-loss configuration as in the ablation study above. Table 6 summarizes the results averaged over three degradation levels and 50 Monte Carlo repetitions per level. A $\pm 2\%$ capacity error increased the OCV reconstruction RMSE by a factor of 2.0–3.4 relative to the exact-capacity baseline. A $\pm 5\%$ error, which represents an upper-bound scenario encountered under sparse data conditions, amplified the RMSE by a factor of 4.5–8.1, reaching approximately 21 mV for the overestimation case. Notably, the sensitivity was *asymmetric*: overestimation of the input capacity consistently produced larger errors than underestimation of the same magnitude (e.g., 21.0 vs. 11.6 mV at $\pm 5\%$, and 8.9 vs. 5.2 mV at $\pm 2\%$). This asymmetry arises because capacity overestimation causes the optimizer to assign wider stoichiometric windows than the true ones, potentially displacing the stoichiometric limits beyond the well-constrained region of the reference OCP curves, into ranges where interpolation accuracy degrades. These results show that errors in the Stage 1 SOH trajectory (SOH_{GPR}) propagated directly into the pseudo-OCV reconstruction, highlighting the importance of the GPR smoothing and outlier rejection employed in Stage 1. This sensitivity is to a systematic bias, whereas the Stage 1 GPR uncertainty is largely random scatter that smoothing averages out. The steep response thus reflects the value of the Stage 1 safeguards, not a fragility of the method.

The sensitivity to errors in the BOL OCP reference was quantified within the same synthetic framework. The OCP curves supplied to the reconstruction were perturbed relative to the true curves that generated the data, by per-electrode voltage offsets (± 2 , ± 5 mV) and stoichiometric stretches (± 1 , $\pm 2\%$, several times the lot-level capacity tolerance measured across 1100 commercial lithium-ion cells [59]), individually and in a worst-case combination. Table 7 reports the resulting sensitivity. Stretch errors were absorbed almost one-to-one into the LAM estimate of the corresponding electrode, leaving the other indicators unaffected. Across all perturbations, including the worst-case combination, LLI and LAM_p biases remained within 0.6 and 2 percentage points, respectively. The residual error concentrated in LAM_n , reflecting the translational degeneracy discussed in Section 3.2. Under a negative cathode-potential offset, this degeneracy admits a data-consistent shift of the positive-electrode window into the flat high-stoichiometry region of the NMC OCP. Stage 1 propagation of the same perturbations remained within the $\pm 2\%$ capacity-bias range of Table 6.

The fixed low-SOC anchor ($\text{OCV}(0) = 2.5$ V) was examined likewise. Ground-truth curves were generated with the true voltage at

Table 7

Sensitivity of the reconstruction to perturbations of the BOL OCP reference. Worst-case values over electrodes, perturbation signs, and the three degradation levels; mean over 50 Monte Carlo repetitions (median in parentheses where the distribution is bimodal).

Perturbation	OCV RMSE (mV)	$ \Delta \text{LAM}_p $ (%p)	$ \Delta \text{LAM}_n $ (%p)	$ \Delta \text{LLI} $ (%p)
None (baseline)	3.5	0.02	0.40	0.17
Offset ± 2 mV	5.6	0.31	0.66	0.25
Offset ± 5 mV ^a	11.6 (13.4)	0.70	1.19 (2.57)	0.51
Stretch $\pm 1\%$	4.1	0.93	1.36	0.21
Stretch $\pm 2\%$	6.9	1.92	2.27	0.20
Worst-case corner ^a	14.0	1.01	5.03 (8.33)	0.56

^a Beyond-tolerance stress bound.

$z = 0$ displaced by 10–50 mV, emulating the aging-induced drift of the boundary voltage that accompanies changes in stoichiometric balancing. The anchor-attributable distortion was isolated as the paired difference between fixed-anchor and true-anchor reconstructions **D**. For displacements of 10–20 mV, the distortion remained below 0.6 mV over the full SOC range and 1.5 mV in the low-SOC region, with anchor-attributable LLI biases below 0.2 percentage points; this held also for a degradation composition taken from the final Stage-2 window of VIN 1 (SOH 78.5%). At a 50 mV stress displacement the distortion remained confined to the low-SOC region, preserving the mid-SOC curvature on which the degradation-mode identification relies.

4.4. Limitations and future work

Several systematic limitations must be acknowledged. First, as noted in Section 4.1, the ~ 12 -month oscillation in $\theta_{p,\text{max}}$ and LAM_p reflected the temperature dependence of the electrode OCPs; because the reference curves were obtained at a fixed temperature, seasonal ambient fluctuations were absorbed into the identified stoichiometric limits and should be interpreted as measurement artifacts rather than true electrochemical changes. Temperature-corrected reference curves would mitigate this artifact but would require extensive OCP characterization across the operating range. The quantitative consequence of such effective OCP mismatches on the reconstruction is bounded by the sensitivity analysis of Section 4.3.

Second, the reconstruction fidelity degraded in windows containing few accumulated segments, predominantly the early-EFC windows associated with the elevated R^2 variance of VIN 2 (Table 3); such windows were rejected by the per-window acceptance gate (Section 3.2) and were not propagated into the degradation-mode trajectory.

Third, the absence of ground-truth SOH measurements prevented independent validation of the absolute estimation accuracy. The analysis relied instead on physical plausibility checks (monotonic SOH decline, a consistent degradation-mode hierarchy) and the out-of-sample cross-validation of Section 4.2, whose support derives from per-vehicle consistency rather than from sample size; absolute quantification accuracy nonetheless remained unverified, and population-level claims are reserved for the future work below.

Fourth, the sensitivity analysis in Section 4.3 demonstrated that a systematic bias in the Stage 1 capacity estimate propagated into the Stage 2 stoichiometric identification in a nonlinear and asymmetric manner. Although the GPR smoothing mitigated random scatter, systematic biases — arising, for example, from inaccurate reference OCV curves or persistent sensor offsets — would propagate directly into the degradation-mode estimates; an independent consistency check on the Stage 1 SOH trajectory, such as cross-validation against onboard coulomb-counting estimates, would reduce this risk.

Fifth, the absence of a shared public benchmark and a ground-truth capacity for production field data precluded a direct quantitative comparison against prior OCV reconstruction methods; the evaluation therefore rested on internal baselines — the uniform-weight ablation

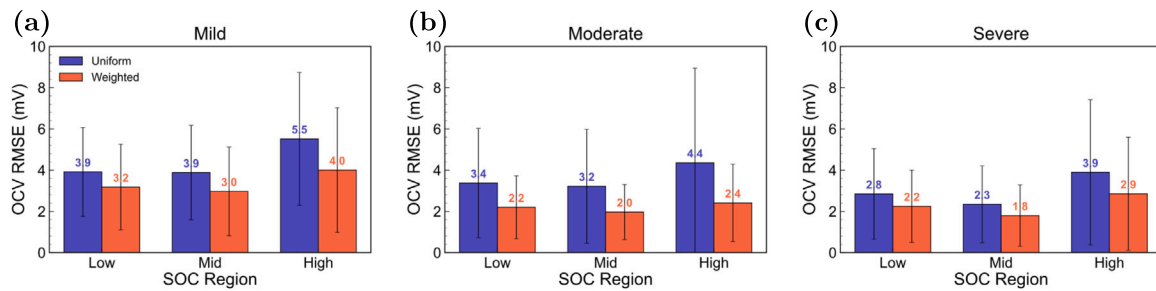


Fig. 8. OCV reconstruction RMSE by SOC region (Low: 0–0.3, Mid: 0.3–0.6, High: 0.6–1.0) for uniform and voltage-bin-weighted loss functions. Results are averaged over 50 Monte Carlo repetitions at three degradation levels. Error bars denote ± 1 standard deviation.

and the frozen-BOL configuration — and the independent electrochemical cross-validation rather than on head-to-head accuracy figures against other methods; absolute reconstruction accuracy itself is quantified against synthetic ground truth in Section 4.3.

Beyond these limitations, four further directions are envisioned: application to additional cell chemistries (e.g., LFP, NCMA) to assess whether the degradation-mode hierarchy generalizes beyond the NMC/graphite baseline, replacement of the fixed low-SOC anchor by a state-dependent boundary condition that tracks the aging-induced drift of the boundary voltage, incorporation of onboard or inferred cell temperature (through temperature-dependent OCP reference curves or an entropic-coefficient, dU/dT , correction) to remove the seasonal artifact identified above, and extension to a larger, mileage-resolved vehicle population, which would enable testing whether the degradation-mode indicators separate calendar-driven from cycle-driven degradation as established electrode mechanisms predict [7,52]. Among these, chemistries with flat OCP plateaus such as LFP are the most challenging, as the curvature features that resolve the stoichiometric alignment are weaker.

5. Conclusion

This study presented a reference-free, two-stage pipeline that diagnoses electrode-level battery degradation from unstructured EV field data—without the reference capacity that existing mechanistic methods require. The field-derived degradation modes were subsequently verified against the measured driving voltage through an independent electrochemical cross-validation based on an SPMe, with no re-identification from the driving data. The first stage estimates an SOH trajectory from partial charge/discharge segments, smoothed via GPR, and the second reconstructs pseudo-OCV curves from relaxed-voltage endpoints via the reference-free ΔQ -method to extract electrode-level stoichiometric limits and the dominant degradation mechanisms. A voltage-bin-weighted loss function was introduced to account for the non-uniform voltage coverage inherent in real-world driving data, and a physical regularization term was employed to suppress optimization artifacts. The proposed weighting reduced OCV reconstruction errors by up to 45% relative to uniform weighting, demonstrating the value of distribution-aware weighting under the skewed voltage coverage of field data.

Vehicle-level analysis demonstrated that the pipeline captured distinct degradation trajectories. VIN 1 exhibited an anomalous rapid-fade event, marked by a concurrent rise in LLI and LAM_p , whereas VIN 2 and VIN 3 followed normal-aging baselines in which LAM_n accumulated without contributing to capacity fade, buffered by the surplus anode capacity. Across all three vehicles, LLI and LAM_p were the primary contributors to capacity fade. Mapping the reconstructed degradation modes deterministically onto an SPMe brought the aged-stage driving-voltage prediction error to within a narrow 9–13 mV band, with no per-vehicle parameter identification, supporting the physical validity of the estimated indicators. A sensitivity analysis revealed that errors

in the Stage1 capacity estimate propagated directly and asymmetrically into the Stage2 reconstruction, highlighting the importance of robust Stage1 SOH estimation. The validation emphasized depth over breadth. Each of the three vehicles was subjected to an independent electrochemical out-of-sample test, providing per-vehicle mechanistic evidence that population-scale correlation alone cannot supply. Quantitatively, the pipeline spanned an SOH range of 97.5%–78.5% with per-window reconstruction attaining median R^2 of 0.93–0.99 and, against synthetic ground truth, an absolute OCV error below 4 mV. Controlled analyses showed a ± 2 –5% capacity-input bias amplifying this error by factors of 2.0–8.1, while perturbations of the OCP reference, the SEI-porosity coefficient, and the low-SOC anchor left LLI and LAM_p robust to within about 2 percentage points.

From a practical perspective, the framework requires only relaxed-voltage points and accumulated-charge records routinely available from onboard logging, and all computations are performed offline. These characteristics suggest that it could serve as a field-deployable diagnostic tool for predictive maintenance scheduling, degradation-aware charging protocols, and early detection of anomalous aging events. Because these stages require no per-vehicle calibration or laboratory reference, they apply to any vehicle with onboard voltage and charge logs. The electrochemical check, which is computationally more involved, remains an optional extension for units that need closer examination. The method is thus suited to large-scale screening, with mechanistic validation reserved for flagged or high-value cases.

CRedit authorship contribution statement

Kyeongun Cho: Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Wooju Lee:** Resources, Investigation. **Jung-II Choi:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work, the authors used Claude (Anthropic) to polish the writing. After using this tool, all authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

Declaration of competing interest

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

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Appendix A. OCV-based data-quality screening

Fig. A.1 illustrates the point-level screening procedure described in Section 2. For a representative vehicle and time window, Fig. A.1(a) displays the extracted voltage–charge points overlaid on the reference OCV curve with the orthogonal tolerance band ($d_{tol} = 0.05$ in the normalized SOC–voltage space). Points within the band (blue) were retained, whereas points outside (red) were discarded.

The discarded points were predominantly concentrated near the upper voltage limit (~ 4.2 V). Three instrumentation and operational factors contributed to this pattern:

1. *Constant-voltage charging phase.* Data points captured during the constant-voltage (CV) stage of constant-current–constant-voltage (CC–CV) charging exhibited a terminal voltage clamped near the charge cutoff while the accumulated charge continued to increase. These points appeared as a horizontal streak at the cutoff voltage with varying capacity offsets, deviating from the OCV curve along the SOC axis.
2. *Current-sensor drift during prolonged parking.* When a vehicle remained parked at a high SOC for an extended period, two sources of spurious charge accumulation arose: the zero-offset error of the current sensor, which produced a small phantom current that was integrated by the coulomb counter, and the BMS quiescent current, which drew a real but untracked load from the pack. Both effects inflated the recorded ΔQ without a corresponding change in terminal voltage, displacing the mapped data point along the capacity axis.
3. *Charging overpotential.* Some points were recorded during active charging rather than at rest. Under load, the terminal voltage (closed-circuit voltage) exceeded the equilibrium OCV by the internal overpotential, causing the SOC-mapping algorithm to overestimate the SOC. These points lay systematically above the reference OCV curve in the parity plot of Fig. A.1(b).

Fig. A.1(b) presents a parity plot comparing the measured end voltage against the value expected from the reference OCV at the mapped SOC. Retained points clustered tightly along the diagonal, confirming consistency with the thermodynamic reference, whereas discarded points exhibited systematic positive deviations at high voltages.

Appendix B. Degradation-mode derivation from reconstructed OCV

Fig. B.1 illustrates the geometric interpretation of the three degradation-mode indicators using the pristine (dashed) and degraded (solid) half-cell OCP curves for a representative vehicle [27]. Loss of active material at each electrode (LAM_m) corresponds to a contraction of the stoichiometric window $W_m = \theta_{m,max} - \theta_{m,min}$ along the normalized capacity axis, as defined in Eq. (16). In the figure, the cathode OCP (U_p , green) narrows from its fresh-cell range, yielding LAM_p , and the anode OCP (U_n , blue) narrows analogously, yielding LAM_n . Both contractions are indicated by inward-pointing arrows labeled “SHRINK”.

LLI manifests as a rightward shift of the anode OCP relative to the cathode OCP along the capacity axis, reducing the cyclable lithium

Table C.1

Governing equations of the single-particle model with electrolyte (SPMe), after Marquis et al. [60]. Here $m \in \{p, n\}$ denotes the electrode.

Process	Governing equation
Solid-phase diffusion	$\frac{\partial c_{s,m}}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(D_{s,m} r^2 \frac{\partial c_{s,m}}{\partial r} \right)$
Boundary conditions	$-D_{s,m} \frac{\partial c_{s,m}}{\partial r} \Big _{r=R_m} = \frac{j_m}{F}, \quad \frac{\partial c_{s,m}}{\partial r} \Big _{r=0} = 0$
Electrolyte-phase diffusion	$\epsilon_{e,m} \frac{\partial c_e}{\partial t} = \frac{\partial}{\partial x} \left(D_e^{\text{eff}} \frac{\partial c_e}{\partial x} \right) + \frac{1-\beta}{F} a_m j_m$
Reaction kinetics (Butler–Volmer)	$j_m = 2 i_{0,m} \sinh \left(\frac{F \eta_m}{2RT} \right)$
Terminal voltage	$V(t) = U_p(\theta_p) - U_n(\theta_n) + (\eta_p - \eta_n) - \eta_e - I R_c$

capacity C_{lit} , as defined in Eqs. (17)–(18). This shift is indicated by the arrow labeled “SHIFT” at the low-capacity end of the anode curve. Physically, it reflects the irreversible consumption of lithium by side reactions such as SEI growth, which immobilizes lithium that previously shuttled between the two electrodes.

Appendix C. Single-particle model with electrolyte

The electrochemical cross-validation in Section 3.3 used an in-house implementation of the SPMe, following the asymptotic formulation of Marquis et al. [60]; the reader is referred to that work for the full derivation. Each electrode is represented by a single spherical particle, lithium transport in the solid and electrolyte phases follows Fickian diffusion, and the intercalation current obeys Butler–Volmer kinetics. The governing relations are collected in Table C.1. All simulations assumed a constant temperature of 25 °C, because cell temperature was not recorded in the field dataset (Section 2); the half-cell open-circuit potentials U_p and U_n were the same fresh-electrode characterization curves used in Stages 1 and 2.

The surface stoichiometry is $\theta_m = 1 - c_{s,m}(R_m)/c_{s,m}^{\text{max}}$, where $c_{s,m}$ denotes the surface lithium concentration, consistent with the stoichiometry convention of Section 3.2 (Eqs. (7)–(8)), and j_m is the interfacial current density per unit particle surface area (A m^{-2}), whose electrode-averaged values are $j_n = I/(a_n L_n A)$ and $j_p = -I/(a_p L_p A)$, with specific surface area $a_m = 3 \epsilon_{s,m}/R_m$ and electrode cross-sectional area A . In the electrolyte equation, the effective diffusivity follows a Bruggeman relation, $D_e^{\text{eff}} = \epsilon_{e,m}^b D_e$, so that the porosity $\epsilon_{e,m}$ perturbed in Eq. (20) enters the electrolyte transport directly; the volumetric reaction source $a_m j_m$ is positive in the negative electrode, negative in the positive electrode, and zero in the separator. The Butler–Volmer expression uses the symmetric form ($\alpha_a = \alpha_c = 1/2$) of [60], with exchange-current density

$$i_{0,m} = k_m F (c_e)^{1/2} (c_{s,m}^{\text{max}} - c_{s,m}^{\text{surf}})^{1/2} (c_{s,m}^{\text{surf}})^{1/2}, \quad m \in \{p, n\}, \quad (\text{C.1})$$

and overpotential

$$\eta_m = \phi_{s,m} - \phi_e - U_m(\theta_m), \quad m \in \{p, n\}. \quad (\text{C.2})$$

In the terminal-voltage expression, η_e is the combined electrolyte concentration and ohmic overpotential and R_c is the lumped contact resistance, held at its BOL value in the aged-state simulations together with $D_{s,m}$ and k_m .

Table C.2 reports the sensitivity of the aged-stage driving-voltage prediction to the LLI-to-porosity conversion coefficient λ of Eq. (20), discussed in Section 4.2.

Appendix D. Anchor–displacement sensitivity

Table D.1 details the anchor–displacement analysis summarized in Section 4.3. Ground-truth OCV curves were generated with the true voltage at $z = 0$ displaced to 2.5 V + δ , and each synthetic dataset was fitted with the anchor fixed at 2.5 V and with an oracle anchor at the true 2.5 + δ V; the paired difference isolates the distortion attributable to the anchor alone.

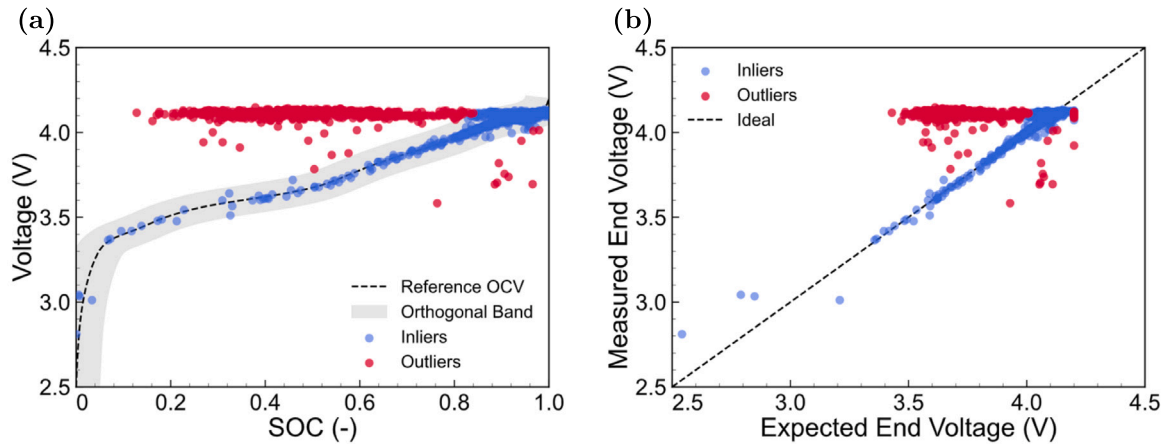


Fig. A.1. OCV-based data-quality screening for a representative vehicle. (a) Voltage–charge points with the orthogonal tolerance band around the reference OCV curve. (b) Parity plot of measured versus expected end voltage. Blue: retained points; red: discarded points.

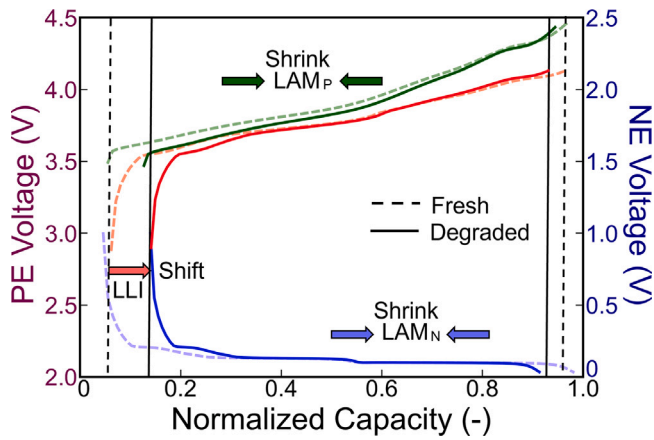


Fig. B.1. Geometric interpretation of degradation-mode indicators for a representative vehicle. Dashed curves: pristine half-cell OCPs; solid curves: degraded OCPs. LAM_P and LAM_N correspond to the contraction (“SHRINK”) of the cathode and anode stoichiometric windows, respectively. LLI corresponds to the rightward shift (“SHIFT”) of the anode OCP, which reduces the cyclable lithium capacity.

Table C.2

Sensitivity of the VIN 2 aged-stage driving-voltage prediction to the LLI-toporosity conversion coefficient λ in Eq. (20). $\lambda = 0$ omits the porosity update and $\lambda = 1$ is the adopted (baseline) configuration; all other parameters are held at their baseline values.

λ	$\varepsilon_{e,n}/\varepsilon_{e,n}^{\text{BOL}}$	Driving-voltage RMSE (mV)
0	1.000	9.80
0.5	0.910	9.63
1	0.820	9.67
2	0.639	11.43
Frozen-BOL	–	19.38

Table D.1

Anchor-attributable distortion (paired difference between fixed-anchor and oracle-anchor reconstructions; median over 50 Monte Carlo repetitions, range over degradation levels from SOH 97% to the VIN-1-derived 78.5% composition). The 50 mV displacement is a beyond-tolerance stress bound.

δ (mV)	OCV, full range (mV)	OCV, low SOC (mV)	$ \Delta\text{LLI} $ (%)
10	≤ 0.2	≤ 0.6	≤ 0.09
20	≤ 0.6	≤ 1.5	≤ 0.18
50	≤ 2.8	≤ 5.7	≤ 0.44

Data availability

The data that has been used is confidential.

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