

Leveraging Probabilistic Degradation States for Early-Stage Lifetime Prediction of Lithium-ion Batteries

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Abstract: While lithium-ion batteries (LIBs) are playing crucial roles in modern energy storage, the accurate and explainable prediction of their lifetimes remains challenging due to their complex and nonlinear degradation patterns. To accommodate these issues, we have investigated the electrochemical principles governing degradation to define distinct states and modeled the statistical transitions between them. In light of this, we propose the innovative probabilistic degradation states (PDSs) framework, which provides a universal architecture for enhancing the performance of any base model in early-stage LIB lifetime prediction. By leveraging probabilistic transitions between discrete degradation states, the PDS method delivers accurate and physically interpretable battery lifetime predictions, outperforming fifteen state-of-the-art benchmarks by achieving a prediction root-mean-squared error (RMSE) of 59.02 cycles. The extensive experiments across three independent datasets further demonstrate the superior performance of PDS over strong baselines in data-scarce scenarios. Through providing insights into the underlying aging mechanisms, the proposed method offers a dual solution for achieving both appropriate degradation representation and effective LIB lifetime prediction.

Keywords: lifetime prediction, nonlinear degradation, probabilistic degradation states, physically interpretable

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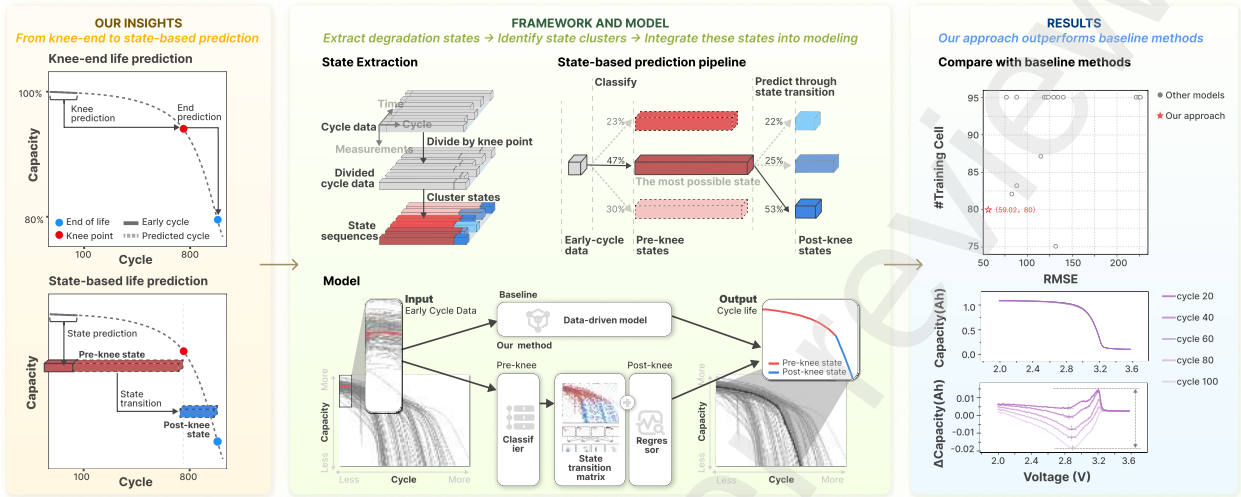
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Graphical Abstract

Leveraging probabilistic degradation states for early-stage lifetime prediction of lithium-ion batteries



Highlights

Leveraging probabilistic degradation states for early-stage lifetime prediction of lithium-ion batteries

- Interpretable PDS framework recasts LIB life prediction via pre-/post-knee states
- Probabilistic state transitions linearize the nonlinear degradation learning task
- A PDS-enhanced linear model rivals complex deep networks on scarce data
- Achieves 59.02-cycle RMSE, surpassing 15 SOTA benchmarks on three datasets
- dQ/dV -V analysis validates states against LLI and LAMdeNE mechanisms

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ABSTRACT

While lithium-ion batteries (LIBs) are playing crucial roles in modern energy storage, the accurate and explainable prediction of their lifetimes remains challenging due to their complex and nonlinear degradation patterns. To accommodate these issues, we have investigated the electrochemical principles governing degradation to define distinct states and modeled the statistical transitions between them. In light of this, we propose the innovative probabilistic degradation states (PDSs) framework, which provides a universal architecture for enhancing the performance of any base model in early-stage LIB lifetime prediction. By leveraging probabilistic transitions between discrete degradation states, the PDS method delivers accurate and physically interpretable battery lifetime predictions, outperforming fifteen state-of-the-art benchmarks by achieving a prediction root-mean-squared error (RMSE) of 59.02 cycles. The extensive experiments across three independent datasets further demonstrate the superior performance of PDS over strong baselines in data-scarce scenarios. Through providing insights into the underlying aging mechanisms, the proposed method offers a dual solution for achieving both appropriate degradation representation and effective LIB lifetime prediction.

1. Introduction

Lithium-ion batteries (LIBs) are fundamental to the current energy shift, supplying power to a range of applications from portable electronics to electric vehicles and grid-scale storage [1, 2]. Ensuring their long-term reliability is paramount, making the accurate prediction of battery lifetime from limited early-cycle data a critical objective, which is essential for strategic management [3, 4] and risk mitigation [5, 6] of energy systems. However, achieving accurate early-stage lifetime prediction is challenging as battery capacity degradation arises from a complex interplay of electrochemical and mechanical processes, which are also highly sensitive to operating conditions like current rate and temperature [7]. These underlying degradation pathways are subtly encoded within early-cycle data in a highly convoluted and non-linear manner, posing a primary barrier to developing predictive models that are both accurate and interpretable.

In response to this challenge, data-driven approaches have emerged as the dominant strategy for predicting LIB lifetime [8]. From elastic nets to deep neural networks, various models are used to learn the non-linear mapping between early-cycle measurements and ultimate cycle life [3, 9]. Pioneering work of Severson et al. [10] demonstrated the power of data-driven approaches using an elastic net model to predict cycle life from early data. This breakthrough catalyzed a wave of subsequent research applying a broader spectrum of algorithms. These include traditional machine learning models, such as decision trees [7], as well as more complex deep learning architectures like recurrent neural networks (RNNs) [11] and graph neural networks (GNNs) [12]. Despite their predictive power, the practical utility of these models is fundamentally limited by their lack of interpretability. These end-to-end models often operate as “black boxes,” rendering their internal decision-making processes opaque and inscrutable [13], raising difficulties on examining the physical rationalities on capturing meaningful degradation signatures beyond solely exploiting statistical correlations contained in the data. Consequently, such models often struggle to provide sufficient scientific insights that are necessary for gaining deep understanding on battery degradation mechanisms.

A promising strategy to move beyond “black-box” models is to anchor predictions in physically meaningful patterns with electrochemical significance. This motivates us to consider the degradation trajectory as a sequence of latent degradation states governed by distinct electrochemical mechanisms, other than a monolithic process. From this perspective, the “knee point” is a natural manifestation of state transition as it marks a point where the dominant mode of capacity loss switches rapidly [14], given the observations on the relatively slow and accelerated decay rates divided by it. This physical marker has inspired a class of methods, broadly termed knee-end life predictions (Fig. 1a₁), that leverage the knee’s timing as a predictor [7, 15], a marker for classification [16], or a boundary for piecewise

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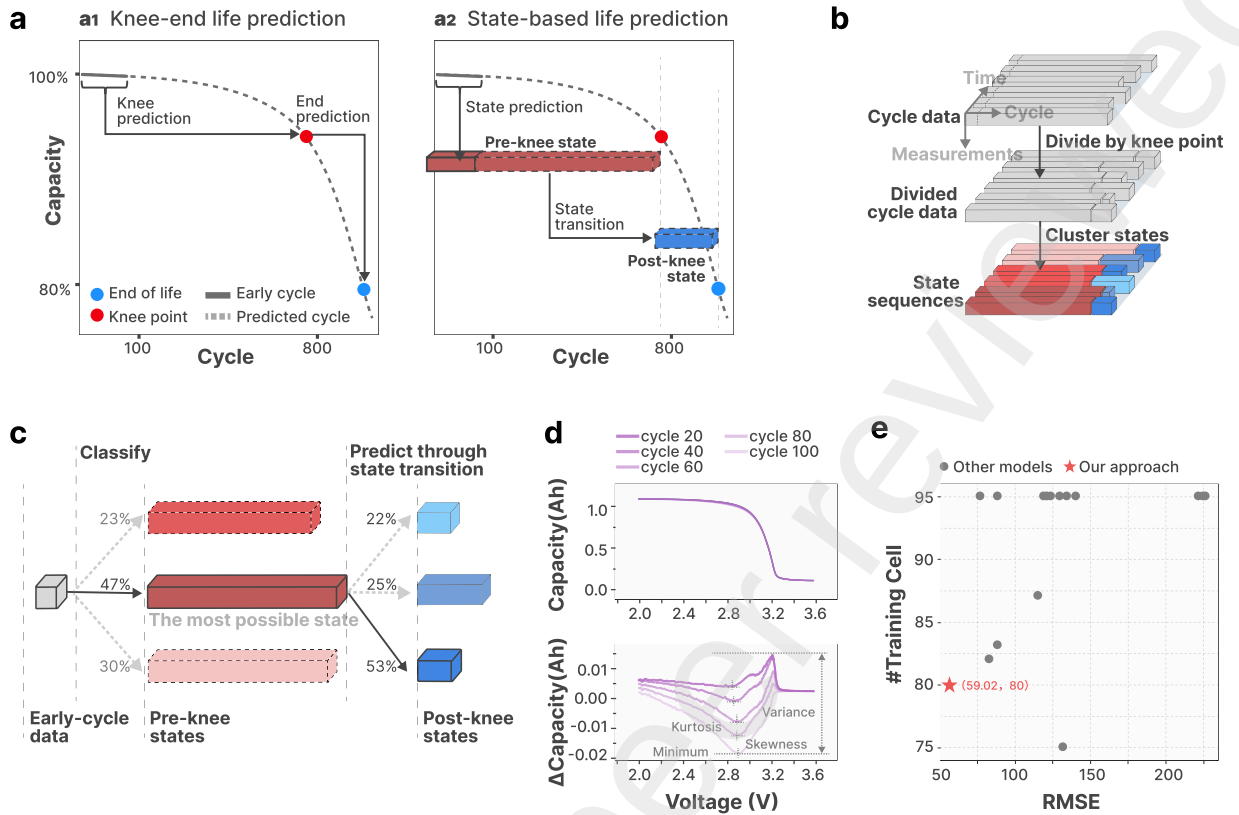


Figure 1: An overview of Probabilistic Degradation State (PDS) prediction framework. **a** Conceptual comparison: (**a**₁) conventional knee-end life prediction involved the knee point; (**a**₂) state-based life prediction that explicitly models pre- and post-knee degradation. **b** Extraction of degradation states split by the identified knee point. **c** Integration of PDS representations into the cycle life prediction pipeline. **d** A part of early-cycle features [10] derived from capacity–voltage curves adopted for prediction. **e** Root-mean-square error (RMSE) comparison between the proposed method and current SOTAs.

mathematical functions [17]. However, a fundamental limitation shared by these methods is that they fail to investigate the transitions of degradation modes, thereby missing important basis for modeling the internal dynamics of batteries.

To address this critical gap and establish effective connections between data-based observations and underlying electrochemical principles, we propose the novel probabilistic degradation states (PDSs) framework that provides a universal architecture for enhancing the performance of any base model in early-stage LIB lifetime prediction. It breaks the degradation trajectory into consecutive pieces, which are then mapped to discrete states representing physically meaningful degradation mechanisms (Fig. 1a₂). Other than projecting each degradation piece into a single state, the PDS framework innovatively considers degradation as the joint effect of multiple states, assesses their individual contributions by estimating the likelihood of each component (Fig. 1c), and investigates the transition of degradation mode through leveraging a probabilistic model (Fig. 1d). Finally, a regression module is incorporated for the complementary of the probabilistic model to enable accurate battery lifetime prediction, which can flexibly support any customized base model to allow the timely update of recent advances. By utilizing the data collected from the first 100 cycles of 80 cells for training, the PDS framework achieves a prediction root-mean-square error (RMSE) 59.02 cycles in minimum, outperforming fifteen state-of-the-art benchmarks (Fig. 1e). Further experiments have been conducted under data-scarce scenarios, where PDS reduces the prediction RMSE by 3–29% and increases the coefficient of determination (R^2) by up to 90% compared with four strong baselines across three independent datasets [6, 10, 18]. In summary, the PDS method has gained deep insights into the underlying aging mechanisms of batteries, offering an integrated solution for both appropriate degradation representation and effective lifetime prediction.

2. Results

2.1. Framework Overview

The proposed PDS framework is constructed in three stages, including 1) the discovery of degradation states, 2) the learning of probabilistic transition between degradation states, and 3) the formulation of residual regression model. Subsequently, the PDS framework can be utilized for battery lifetime prediction (Fig. 2a). This work utilizes the knee point as a partition to segment an entire degradation trajectory into pre-knee and post-knee phases (Fig. 1b), where the knee point is recognized using the Bacon-Watts model [19]. For all segmented degradation trajectories obtained from the training data, we investigate their similarities to discover degradation states through clustering method. The segments with high similarities are subsequently assigned into groups using the k -means++ algorithm with dynamic time warping (DTW) as the measurement of distance, such that each group represent a degradation state. Given a pre-knee segment, a classifier is trained to identify the specific initial degradation state it belongs to using features (Supplementary Table 1) selected via recursive feature elimination [20] (Supplementary Note 1) from early-cycle data (Fig. 1d). Furthermore, we believe each segment of degradation trajectory can be simultaneously impacted by multiple electrochemical mechanisms. Therefore, we use the classifier to quantify the contribution of each one by estimating the likelihoods. To capture the shifts of dominant degradation mechanisms, a state transition probability matrix is established by empirically tracking how frequently transitions occur between pre-knee and post-knee states. Based on this formulation, the lifetime of a battery can be preliminarily estimated using the expected lifetime of all relevant pre-knee and post-knee degradation states. To achieve more accurate prediction, a residual regression model is incorporated as a complementary of the degradation-state-based prediction through catching the non-linear, cell-specific deviation from the main degradation pattern, which can be flexibly customized based on the specific characteristics of the data.

Our experiments utilize three commonly benchmarked datasets, including MATR [10](124 cells), CLO [6](45 cells), and Stanford [18](222 cells). Results of all the following experiments are averaged over five independent repetitions to ensure statistical robustness. Detailed experimental settings (e.g., hyperparameters, metrics and dataset information) are provided in Supplementary Table 2 and Note 2-3.

2.2. Comparisons with state-of-the-art methods

To rigorously evaluate our framework's performance, we benchmark it against a comprehensive set of prior work using the public datasets. Establishing a fair comparison methodology is non-trivial, given the diverse nature of existing approaches, which primarily fall into traditional machine learning (ML) and deep learning (DL) categories. Therefore, we designed a two-stage evaluation protocol. First, for traditional ML models, we directly compare our results with the performance metrics reported in their respective publications. This allows for a direct benchmark against the established state-of-the-art (SOTA) under their authors' optimized conditions. Our systematic review identified 15 such models from 6 studies for this comparison in recent years (Fig. 1e). Detailed information of related works can be found in Supplementary Table 3. Second, for influential non-open-source DL methods where code is unavailable, we directly manipulate representative base architectures (e.g., linear regression) shared by these approaches for a controlled and unbiased assessment; for open-source DL methods (e.g. tabular prior-data fitted network [21, 22]), we compare our framework with them directly through experimental study.

Utilizing the first 100 cycles of 80 cells as training data, our PDS framework achieves a root-mean-squared error (RMSE) of 59.02 cycles, a result that outperforms the 15 benchmarked models (Fig. 1e). Beyond this high predictive accuracy, the proposed PDS framework is also characterized by its core innovation on the mechanistic interpretability. Take the dataset from MATR [10] as an example, our framework divided commercial lithium-ion cells' cycle data into distinct segments based on knee point. Then, we determined the optimal cluster count $k = 6$ for pre-knee and $k = 6$ for post-knee using the elbow method (intermediate results can be found in Supplementary Table 4), with cluster validity confirmed by high silhouette scores (> 0.65) and a low Davies-Bouldin index (< 0.8). We finally gain 9 degradation states after removing anomalous clusters whose segment counts account for less than 1% of the total, 5 for pre-knee ($S_1 - S_5$) and 4 for post-knee ($S_6 - S_9$). This state-based representation provides a powerful diagnostic lens. For instance, we can map a specific cell's degradation as a pathway through these states (e.g., $S_1 \rightarrow S_9$). This can help distinguish between different patterns of degradation even in the earliest stages, using a combination of simple foundational models.

2.3. Impact of PDS modeling on prediction performance

To evaluate our framework and demonstrate the broad utility of our PDS framework, we benchmark it against our reimplementations of representative machine learning and deep learning models and then conducted a comprehensive

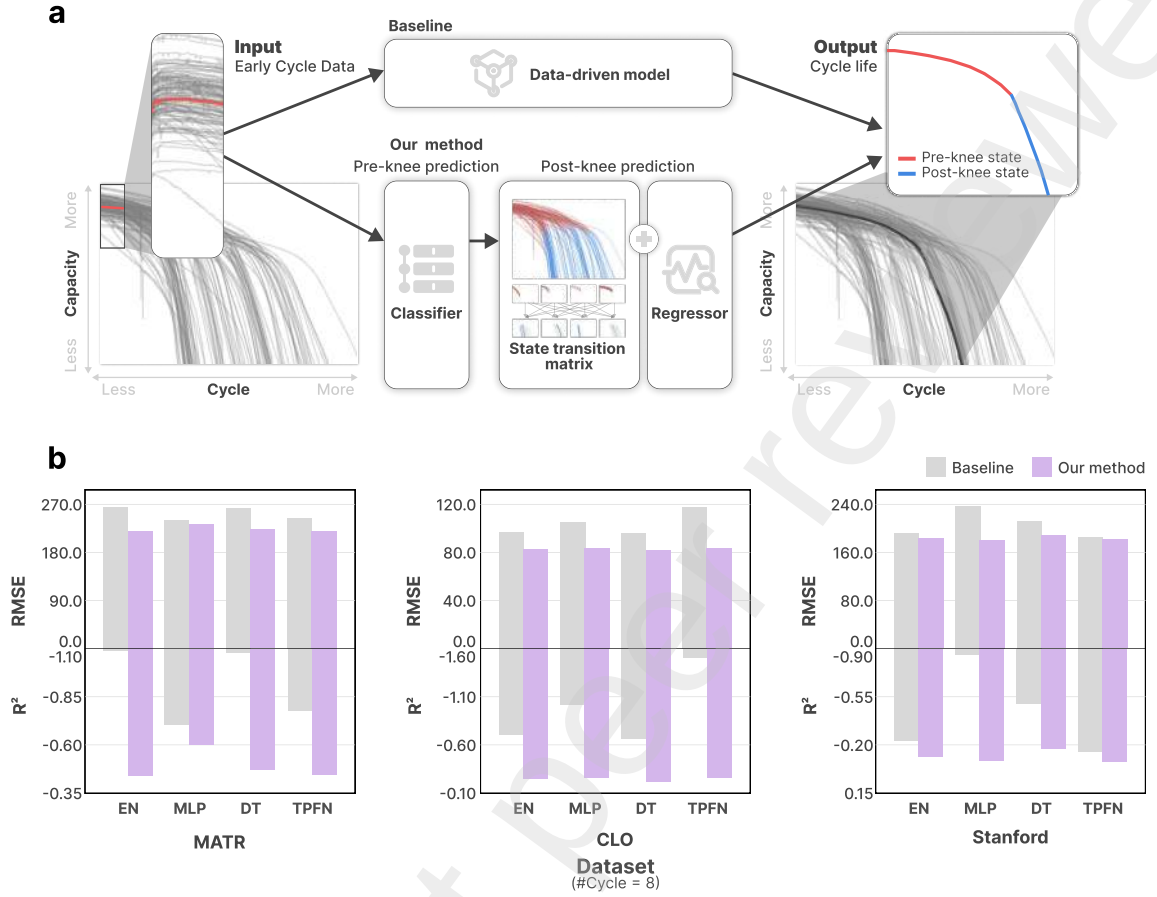


Figure 2: Comparison of the PDS-enhanced framework with baseline methods. a Architectural sketch of the baseline model (top) and the PDS-enhanced pipeline (bottom). **b** Performance comparison between PDS-enhanced models and baseline models on three datasets (MATR [10], CLO [6], and Stanford [18]) using only the first 8 cycles and 15% of the cells. PDS-enhanced models consistently outperform baselines of Elastic Net (EN) [10, 23], Decision Tree (DT) [7, 24], Multi-Layer Perceptron (MLP) [25], and Tabular Prior-data Fitted Network (TPFN) [21].

ablation study. To complete the comparison promised in the previous section, we evaluate our PDS framework against several reimplemented non-open-source models. We selected four representative regressors to span the model complexity spectrum: Elastic Net (EN) [10, 23] for linear models, Decision Tree (DT) [7, 24] for classical non-linear models, Multi-Layer Perceptron (MLP) [25] as a foundational neural network, and Tabular Prior-data Fitted Network (TPFN) [21] as a state-of-the-art deep learning method. This allows for a controlled comparison against common modeling strategies, complementing the wider body of work that employs other deep learning architectures such as CNNs [26], LSTMs [27], and Transformers [28] for this task. Comparative results across the MATR [10], CLO [6], and Stanford [18] datasets are summarized in Fig. 2. Our PDS framework achieves a salient lower RMSE and a higher coefficient of determination (R^2) than EN, DT, MLP, and TPFN in most datasets (Fig. 2b). It is worth noting that the impressive performance is achieved using only 8 cycles and 15% cells. PDS uniformly elevates every regressor, abolishing the data-hungry handicap of MLP (21% RMSE reduced) and TPFN (29% RMSE reduced) especially on CLO [6] dataset which contains only 45 cells in total. PDS also collapses the original performance spread: post-integration all models converge to the same, superior level. Notably, the performance of the MLP, as measured by the R^2 , achieved an approximately 90% improvement on the Stanford [18] dataset. Post-PDS, Post-EN marginally surpasses almost other models. Linear EN captures the simplified, near-linear state patterns unveiled by PDS. PDS achieves this

by replacing the monolithic nonlinear degradation curve with a low-dimensional, knee-divided state sequence that is intrinsically linear in probability space.

Ablation study is also conducted to quantify the performance boost gained by integrating our state modeling approach into the most common life cycle prediction workflows. The motivation for this study is two-fold. From a practical standpoint, making accurate predictions from limited data is a primary bottleneck for industrial battery management. More fundamentally, this study tests a hypothesis of our work: that by embedding mechanistic insights into the model via discrete degradation states, we can not only achieve transparency but also superior data efficiency. Specifically, instead of learning complex, non-linear degradation dynamics from raw data, a model augmented with our PDS features learns the simpler task of identifying a cell's state, thus modeling the decay process more effectively. We evaluated performance on MATR [10] dataset under two critical dimensions of data scarcity: (1) using a small training cohort of only 17-21 cells (15% of the total), and (2) making predictions based on very few early measurement cycles (the first 6-10 cycles. Results for cycle counts of five or fewer are not reported due to insufficient data density to support reliable prediction.).

We constructed three groups of tests to evaluate the PDS framework (Fig. 2a) as shown in Fig. 3. Our results reveal distinct performance patterns across the different modeling stages. First, we confirmed that simply segmenting the degradation curve at the knee point without applying state modeling yields only marginal, statistically insignificant gains (Fig. 3a). However, when introducing PDS to characterize the pre-knee phase (Fig. 3b), the models reduced prediction error by a substantial 2-12% compared to their non-state-based counterparts, confirming that state-driven characterization significantly improves early-phase prediction. For the post-knee analysis (Fig. 3c), we observed that state transition modeling primarily captured the dominant linear degradation trends. This was validated by our final model, which combined pre-knee state modeling with a state-enhanced post-knee regressor, effectively harmonizing the performance of both linear and non-linear models and confirming the complementary roles of state transitions and residual regression. Experiments with an alternative parameter set (cycle range 6–10) further corroborate this finding (Supplementary Fig. 1).

In conclusion, these ablation studies demonstrate that integrating our PDS framework consistently enhances the performance of standard regressors, enabling more accurate predictions with significantly less data than standalone methods. The framework provides not only a quantitative boost in accuracy but also qualitative, mechanistic insight by decoupling the degradation process into distinct, manageable states. This dual advantage of superior data efficiency and enhanced interpretability establishes our state-based modeling as a powerful and versatile tool for creating more trustworthy and insightful battery lifetime predictions.

2.4. Characterization of identified degradation states and interpretation of state transition

This section characterizes degradation states identified in a population of lithium-ion cells on MATR [10] dataset and provides an overview of their degradation transition patterns. We have identified totally nine degradation states, categorized into five pre-knee states ($S_1 - S_5$) and four post-knee states ($S_6 - S_9$). The overview of the degradation states is presented in Fig. 4a. The five pre-knee states ($S_1 - S_5$) represent distinct patterns of progressive but largely recoverable degradation. These states vary significantly in their characteristics. S_1 exhibits stable linear aging (capacity fade: 0.07%/cycle), while S_2 exhibits the sharpest rate of capacity decline among the pre-knee states. Conversely, the four post-knee states ($S_6 - S_9$) denote the onset of rapid, irreversible failure. These states are universally characterized by a sharp decline in capacity over a very short duration. Each terminal state serves as an “end-of-life” signature, corresponding to dominant failure mechanisms.

Finally, we investigated whether the discovered states form a coherent degradation pathway. We hypothesized that if the states are physically meaningful, then transitions should occur more frequently between physically similar states. To test this, we quantified state similarity using the Euclidean distance between their representative degradation curves (Fig. 4b) and calculated the observed transition frequencies in a state transition matrix (Fig. 4c). The results reveal an insightful relationship between these two metrics, one that is consistent with our hypothesis. We observed a notable trend where Euclidean distance between states appears to be inversely related to transition probability. For example, S_3 is closest to S_6 , and their transition is also the pathway observed the most frequently. Besides, this general trend, where shorter distances between states tend to correspond with a higher likelihood of transition, holds true across the matrices. Therefore, the state transition matrix captures the degradation patterns through probability and helps with regressor prediction. The state patterns are closely associated with the macro representation, which can motivate the regressors to achieve excellent performance.

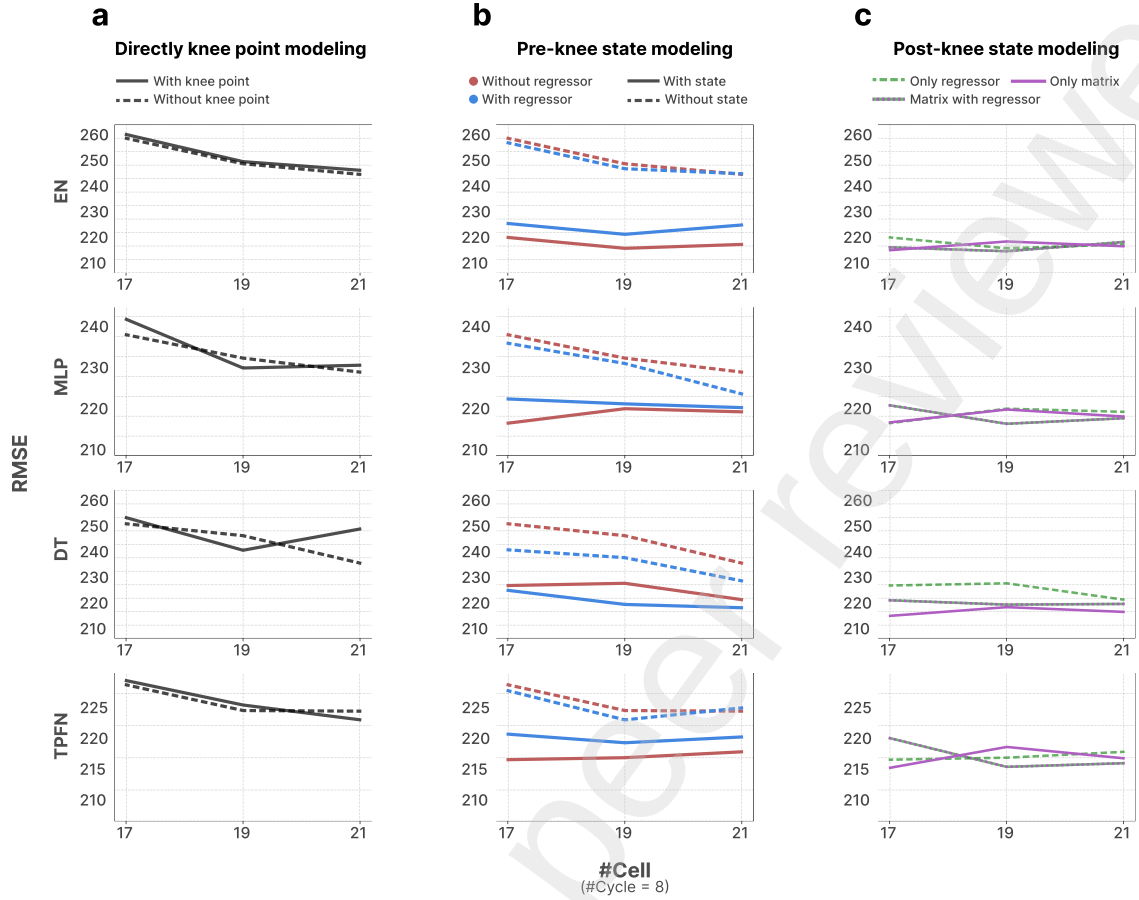


Figure 3: Ablation study on state modeling before and after the knee point across four baselines. All experiments were conducted at 8 cycles with varying cell usage. **a** Impact of explicitly detecting the knee point. Directly knee point modeling yields insignificant gains. **b** Contribution of modeling degradation pre-knee state. Pre-knee state modeling reduces RMSE significantly. **c** Contribution of modeling degradation post-state and regressor. Post-knee state modeling learns primary degradation trend.

2.5. Detailed analysis of identified degradation states

To elucidate the physical mechanisms captured by our data-driven state model, we perform a systematic interpretation using differential capacity analysis (dQ/dV-V) [30]. This analysis connects the identified degradation states to established electrochemical degradation phenomena. We first investigate the dominant degradation modes, Loss of Lithium Inventory (LLI) and Loss of Active Material (LAM_{deNE}), by contrasting the behavior of the cells in the pre-knee and post-knee regimes. Following this, we provide a granular analysis of the electrochemical signatures of each distinct state. Finally, we quantify the evolution of specific features within the dV/dQ curves to deconvolve the contributions of the underlying degradation pathways.

The evolution of the cell's electrochemical signature across the identified degradation states is detailed in Fig. 5. To capture the degradation dynamics within each state, the dQ/dV profiles were averaged across all cells. As illustrated in Fig. 5a, we define the “first cycle” as the initial cycle after a cell enters a state and the “end cycle” as the final cycle before it transitions out. Fig. 5b presents these averaged dQ/dV curves for the first and end cycles, organized into pre-knee ($S_1 - S_5$) and post-knee ($S_6 - S_9$) regimes. A clear distinction in the degradation behavior between the two regimes is evident. In the pre-knee states, the dQ/dV profiles evolve slowly, with minimal variation between the first and end cycles. In contrast, the post-knee states exhibit dramatic changes. The primary peak in the post-knee curves shows a significant decrease in intensity and a shift to lower potentials, which are characteristic signatures of

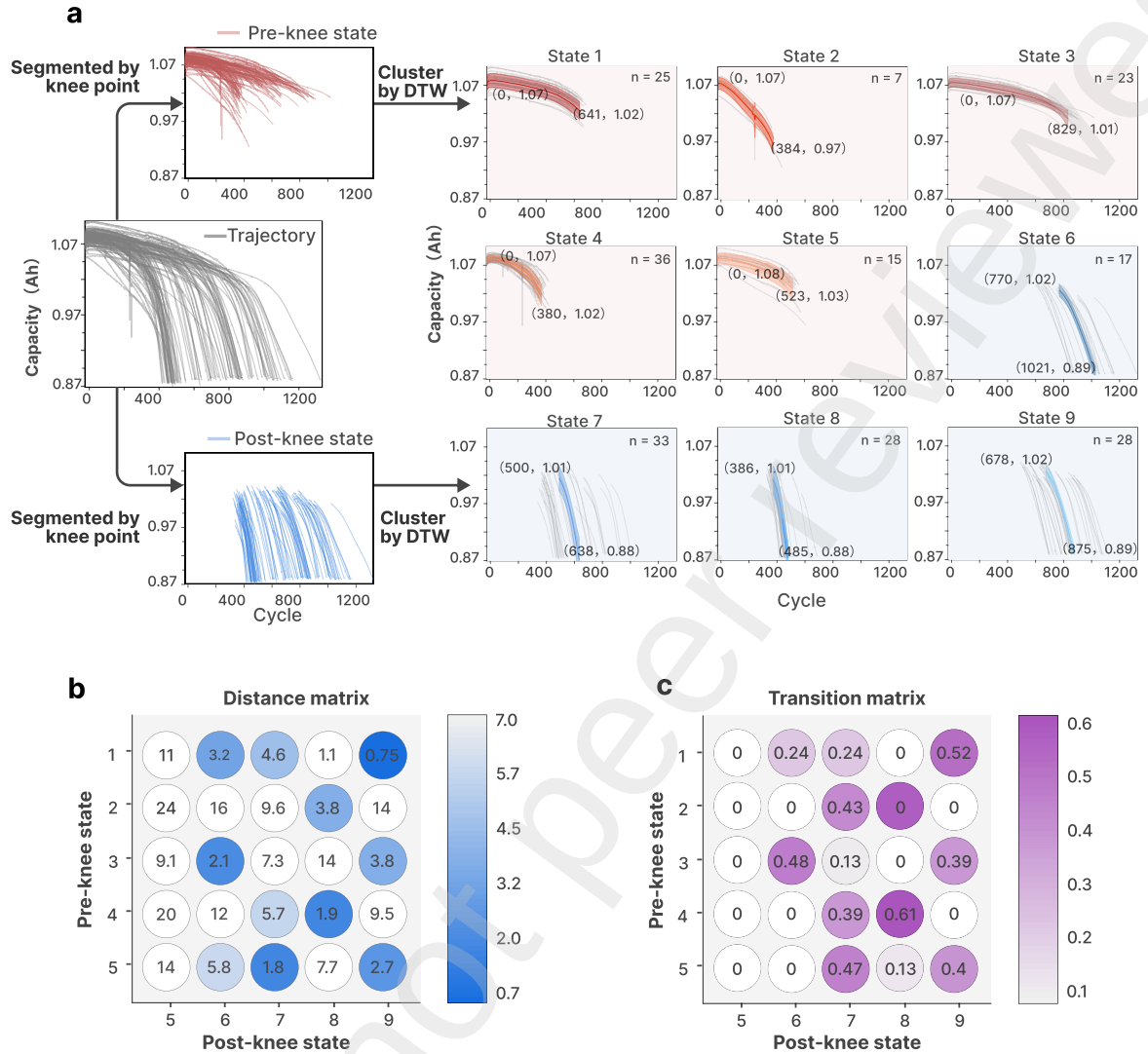


Figure 4: An overview of the degradation trajectories in each state and the analysis of state transition. **a** The trajectories of discharging capacity degradation are divided by knee points and clustered into states using Dynamic Time Warping (DTW). The mean trajectories with standard deviations are shown in each state, along with their start and end coordinates. The number of trajectories is indicated in the upper right corner. **b** A state distance matrix. The distance of each state is computed with the Euclidean distance of the end cycle dQ/dV-V curve in a state and the first cycle dQ/dV-V curve in another state. **c** A state transition matrix. The matrix reflects the probability of state transition. A closer distance of states reflects the larger probability of transition. The distance matrix at the micro level is in accord with the transition matrix at the macro level.

LAM_{deNE}[31, 32]. Concurrently, the horizontal shift of the entire profile across all states reflects a continuous LLI. These combined observations indicate that while LLI is a persistent degradation mode throughout the battery's life, substantial LAM_{deNE} is predominantly activated during the post-knee phase. An analogous analysis based on dV/dQ-Q curves, which confirms these findings, is provided in Supplementary Fig.2.

A granular analysis of the degradation trajectory within each individual state is presented in Fig. 5c. We systematically evaluate the evolution of the dQ/dV profiles by tracking two key metrics: the shift in peak voltage (an indicator of polarization) and the attenuation of peak intensity (an indicator of capacity loss). First, we observe a

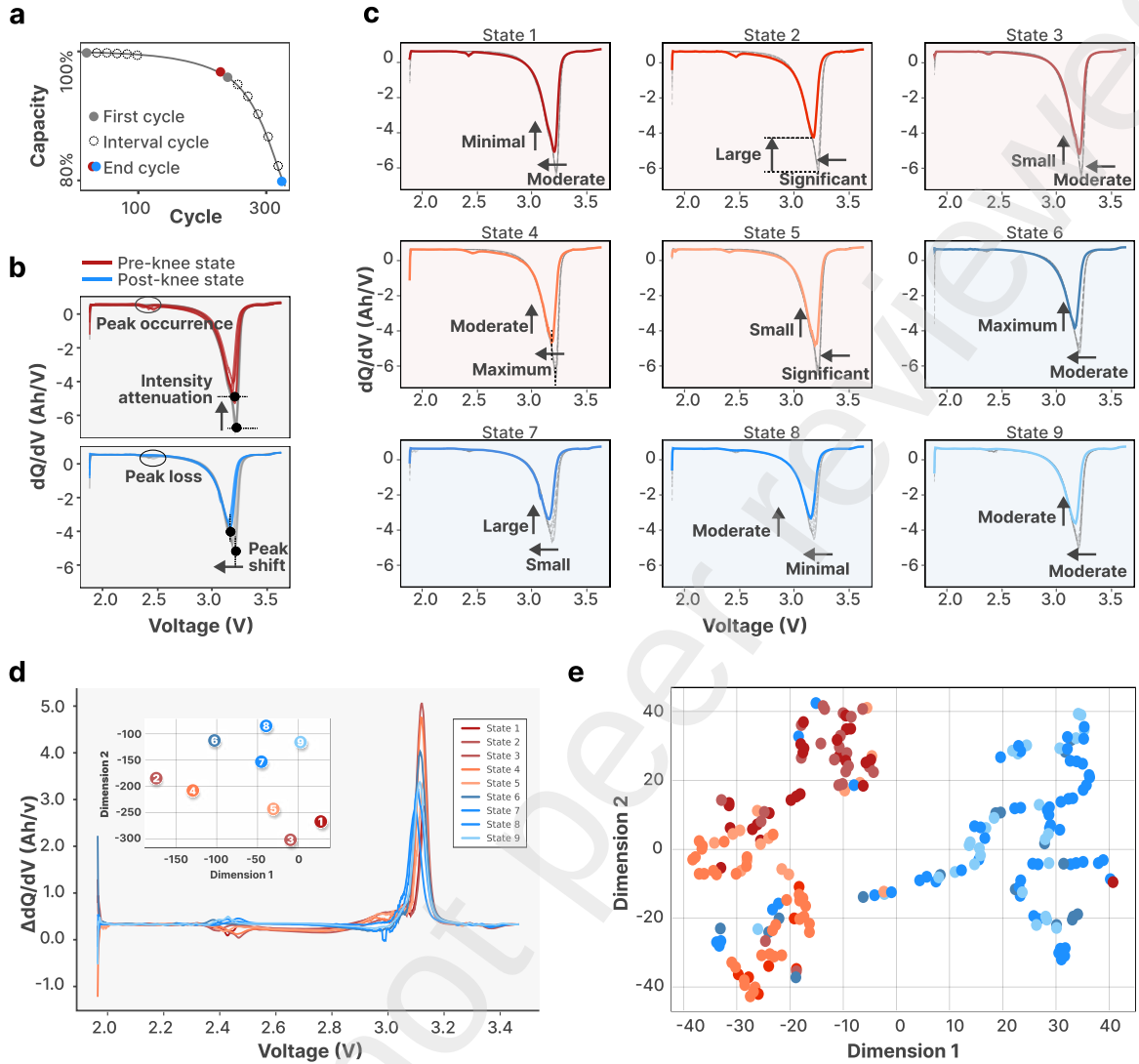


Figure 5: An illustration of the dQ/dV - V curves of states. **a** The data acquisition from degradation of LIB. Data acquisition involves sampling the first 100 cycles at 20-cycle intervals, along with the end cycle in each state. **b** Comparative analysis of pre-knee and post-knee states in the context of dQ/dV - V curves. The state patterns show LAM_{deNE} and LLI to varying degrees. **c** The dQ/dV - V curves in each state. The solid gray curve is the first cycle of the fragment in the state, a thin dotted gray curve is added every 20 cycles until 100, and the colored curve is the end cycle of the fragment. The degree of voltage shift and intensity attenuation is noted in each state. **d** Difference of the dQ/dV - V curves in the first cycle and the end cycle of each state. A scatter plot shows the similarity of the curve patterns. The state curves are mapped to two-dimensional space with t-SNE [29]. A closer distance of state dots reflects the larger similarity of PDSs. The states are more similar in the pre-knee states or the post-knee states. **e** A scatter plot showing the similarity of all samples in the PDSs. There is a large gap between pre-knee states and post-knee states.

frequent shift of the primary peak towards higher potentials with increased cycling, indicating a progressive rise in cell impedance. The magnitude of this voltage shift varies by state, with S_2 , S_6 , and S_7 exhibiting the most pronounced increases. This effect is likely attributable to kinetic hindrances, such as impedance growth at the solid-electrolyte interphase (SEI) or changes in the active material's bulk properties that impede ion diffusion. Second, the peak intensity diminishes over time in all states, reflecting the gradual loss of electrochemically active material. This decay is particularly accelerated in S_2 , S_4 , and S_5 , suggesting these phases are governed by more aggressive degradation mechanisms. Furthermore, the general flattening of the curve shape in the high-voltage region suggests increasing

limitations in charge transfer. In contrast, the relatively subtle changes within S_7 and S_8 signify a period of greater electrochemical stability. A quantitative summary of the peak shift and intensity decay rates for each state is detailed in Supplementary Tables 5 and 6 and discussed further in Supplementary Note 4.

To visually validate that the identified states represent distinct physical regimes, we conducted a feature-based analysis of the dQ/dV-V curves. We performed two complementary analyses using t-SNE [29] for dimensionality reduction. First, to quantify the unique degradation pattern within each state, we created a characteristic “difference profile” by subtracting the first-cycle dQ/dV-V curve from that of the end-cycle. We then extracted a feature vector from each of these difference profiles (feature details can be found in Supplementary Table 7). The t-SNE visualization of these state-level features is shown in Fig. 5d. The visualization confirms that the states cluster into two distinct supergroups, corresponding to the pre-knee and post-knee regimes. Within these groups, the proximity between individual states indicates a high degree of similarity in their underlying degradation mechanisms, for example, S_2 and S_4 appear to be closer than others. Second, to confirm that our states effectively partition the entire degradation trajectory, we extracted features from the dQ/dV-V curve of every state. As shown in the t-SNE plot in Fig. 5e, cycles belonging to the same state form tight, localized clusters. The clear boundaries between adjacent clusters validate that our method successfully segments the battery’s lifetime into discrete periods, each characterized by a self-similar electrochemical signature.

3. Discussion

Cycle life prediction using early cycle data is crucial for advancing research on battery degradation. However, the complex, nonlinear degradation behavior of LIBs (lithium-ion batteries) limits traditional data-driven methods in terms of interpretability. In recent years, knee-based predictions have emerged as a promising approach to address these limitations. The knee point, a key feature in degradation trajectories, can reveal underlying patterns and provide an explainable aging process. Most existing methods focus on correlating the knee point with cycle life rather than modeling the degradation patterns themselves. In this study, we systematically investigate the degradation patterns before and after the knee point. We introduce the probabilistic degradation state (PDS) to quantitatively capture these patterns and explore their predictive relevance to cycle life via statistical learning within our proposed framework. The state patterns are validated against micro-level degradation patterns to confirm their generalizability.

Three datasets (MATR [10], CLO [6], and Stanford [18]) are used to analyze degradation patterns. The cells undergo charging and discharging under diverse conditions, such as charging protocols, temperature variations, and calendar aging [33]. Despite identical usage conditions, individual cells exhibit distinct degradation behaviors [17], which cannot be consistently partitioned using conventional metrics (e.g., current rate) [7]. However, degradation trajectories show inherent similarities, with a knee point present in nearly all cases. The fragments before and after this knee point exhibit distinct features and degradation dynamics. The PDS provides a novel approach to quantifying and distinguishing these patterns. Using this method, degradation data are partitioned into 212 fragments and clustered into nine distinct states. These states differ in trajectory shape and duration, while fragments within the same state share similar degradation characteristics, reflecting underlying, consistent patterns. Compared to existing methods that rely on feature extraction [7], state partitioning directly separates degradation patterns using capacity data. This approach reduces the need for extensive measurements, lowering data acquisition costs while maintaining cost-effectiveness. Unlike methods requiring specialized equipment (e.g., electrochemical impedance spectroscopy, EIS [34]), our method only involves capacity measurements. Additionally, the relevance of degradation states is quantified via the state transition matrix, which captures transition probabilities and enhances interpretability in cycle life prediction.

The partition and transition of PDS are validated through degradation mode analysis, leveraging mechanism data such as dQ/dV-V and dV/dQ-Q curves. These data confirm that PDS effectively captures degradation patterns associated with varying degrees of Loss of Lithium Inventory (LLI) and Loss of Active Material (LAM_{deNE}). Quantitative analysis in a two-dimensional space reveals significant differences between states, with samples within the same PDS exhibiting similar characteristics and spatial proximity. The state distance matrix further illustrates aging trends within states, aligning with the state transition matrix to demonstrate state relevance. The analysis confirms the interpretability of PDS, enabling low-cost, quantitative representation of degradation patterns. By recognizing and analyzing these states and their transitions, LIB degradation can be divided into manageable phases, each reflecting distinct battery life stages and underlying mechanisms.

A central challenge in battery prognostics is that data-driven models often require large datasets to learn complex, non-linear degradation dynamics from scratch. Our PDS framework addresses this limitation by fundamentally

reframing the prediction task. Instead of relying on a single, complex model to capture the entire battery lifetime, we embed electrochemical insight by segmenting the degradation pathway into a series of quasi-linear, physically distinct states. This approach hypothesizes that the degradation dynamics within a state are far simpler to model than the dynamics between states. Our experimental results provide strong evidence for this hypothesis. Conventional regressors struggle with predictions based on very early-cycle data, as these initial features often lack a clear correlation with total lifespan (Fig. 2a). However, by incorporating the PDS and its transition probabilities, our PDS-integrated models achieve substantially higher accuracy. Furthermore, this state-based approach demonstrates remarkable data efficiency. While reducing the training data size degrades the performance of all models, PDS integration significantly mitigates this effect (Fig. 2b). Most notably, the state-integrated Elastic Net, a simple linear model, ultimately outperforms more complex non-linear models at more cells. This key finding suggests that by correctly partitioning the problem, we linearize the learning task, enabling simpler, more interpretable, and data-efficient models to achieve superior performance. This marks a pivotal shift from brute-force data correlation to a more practical, physically-grounded prognostic strategy. Ablation results in Fig. 3 further validate PDS's role in capturing pre- and post-knee degradation phases. Unlike methods relying solely on the knee point, PDS focuses on state transitions, enabling efficient use of limited data. Post-knee fragments, characterized by sharp capacity declines, require distinct prediction strategies compared to pre-knee phases. The ability of the framework to handle these differences highlights PDS's feasibility for accurate, low-cost predictions. By granularly examining aging processes, PDS identifies subtle patterns missed in whole-dataset analyses, enabling more nuanced predictive models. This approach aligns with recent findings on critical transitions in battery degradation [9, 12], where state shifts mark significant behavioral changes.

This study represents a foundational step toward quantitatively representing degradation patterns in lithium-ion batteries. The PDS effectively captures pre- and post-knee degradation dynamics, enhancing both interpretability and prediction accuracy in cycle life estimation. Mechanistic analysis validates the rationality of PDS, while the proposed framework provides a low-cost, pattern-driven approach to cycle life prediction. Additionally, PDS demonstrates utility in identifying degradation mechanism modes. However, two limitations exist. First, PDS extraction relies on knee-point detection and trajectory clustering algorithms, which may introduce errors or outliers, potentially affecting degradation analysis and prediction accuracy. Second, the framework currently computes the state transition matrix using an empirical, count-based approach. This direct method may have limited generalizability, particularly in scenarios with sparse data or novel degradation trajectories. Future work will address these limitations. For PDS extraction, we will explore advanced machine learning techniques to improve knee-point detection and clustering robustness. Additionally, we will investigate the relationships between PDSs, including pre-knee/post-knee states and state transitions. While this study observed overlaps between states, future research will quantitatively analyze these overlaps to refine state boundaries and improve prediction reliability. To enhance the transition model, we plan to move beyond static empirical matrices and investigate dynamic methods, such as Bayesian inference. These approaches will allow the framework to capture uncertainty and adapt its predictions as new data becomes available, paving the way for a more resilient and universally applicable prognostic tool.

4. Conclusions

To conclude, this study introduces PDS as a novel method to enhance the interpretability and accuracy of cycle life prediction for lithium-ion batteries. By analyzing degradation patterns before and after the knee point, PDS provides a quantitative representation of battery states, facilitating the analysis and prediction of battery life. This approach requires fewer measurements, reducing costs, and significantly improves the predictive power of various regressors, especially when early-cycle data is limited.

5. Methods

5.1. Probabilistic degradation state identification

The degradation state exhibits distinct patterns before and after the knee point [14], a critical transition where capacity fade accelerates and significantly affects the prediction of life cycle. We introduce a Probabilistic Degradation State (PDS) identification framework to systematically identify state-specific degradation modes, overcoming the interpretational ambiguities inherent to conventional single-state aging models. This approach enhances the accuracy of cycle life predictions while providing mechanistic clarity to aging trajectories. This framework operates in two stages: cycle data partitioning and state clustering.

First, our method partitions the cycling data of each cell into pre- and post-knee fragments. To achieve this, we detect the knee point using the Bacon-Watts model [19], selected from multiple candidate algorithms for its ability to capture the transition from gradual to accelerated degradation—a critical feature aligning with our focus on state-specific aging dynamics. Formally, let $\mathbf{X}_i = \{\mathbf{X}_i^1, \mathbf{X}_i^2, \dots, \mathbf{X}_i^{N_i}\}$ denote the cycle data of cell i , where $\mathbf{X}_i^j$ represents the j -th charge/discharge cycle. The knee point for cell i , identified at cycle t_i , divides the data into two fragments: $\mathbf{f}_{i,1} = \{\mathbf{X}_i^1, \dots, \mathbf{X}_i^{t_i-1}\}$ (pre-knee) and $\mathbf{f}_{i,2} = \{\mathbf{X}_i^{t_i}, \dots, \mathbf{X}_i^{N_i}\}$ (post-knee). These fragments are then analyzed independently to extract features that capture the distinct degradation dynamics.

Second, pre- and post-knee degradation fragments from all cells are aggregated to identify PDSs through k -means++ clustering [35] with Dynamic Time Warping (DTW). k -means++ is selected due to its superior initialization scheme, which strategically places initial cluster centroids to accelerate convergence and reduce sensitivity to random initialization, thereby improving robustness for high-dimensional temporal degradation data. The optimal number of clusters k is determined via the elbow method, ensuring distinct degradation patterns are captured while avoiding overfitting. DTW is used to measure the similarity of the degradation fragments. Each cluster is assigned a PDS label s_x , with all fragments within a cluster inheriting its label. All PDS form a state set $S = \{s_1, s_2, \dots, s_k\}$. For every state s_k , our method calculates its average fragment length D_k .

5.2. Probabilistic degradation state dynamics modeling

The probabilistic degradation states represent sequential phases of aging, yet cycle life variability arises not from isolated states but from their transition dynamics. Batteries evolve through these states probabilistically, akin to Markovian systems [36] where future aging depends only on the current degradation state. We quantify the transition dynamics by modeling them as stochastic jumps between identified states S . Inspired by the hidden Markov model [37], we use a state transition matrix $\mathbf{A} = \{a_{ij}\}$, where $a_{ij} = P(s_j|s_i)$ describes the probability to transit from s_i to s_j . The pre-knee states appear before the knee point, while the post-knee state appears after the knee point.

Our method constructs the state transition matrix $\mathbf{A}$ based on the identified state set S and every cell's cycle data. Every cell's cycle data corresponds to two consecutive state s_p (pre-knee) and s_q (post-knee). For every pair of states (s_p, s_q) , we count up a frequency item $\tilde{a}_{pq}$. Then, every item a_{ij} in matrix $\mathbf{A}$ can be produced as:

$$a_{ij} = \tilde{a}_{ij} / \sum_{j=1}^k \tilde{a}_{ij} \quad (1)$$

The pre-knee states S^0 consist of all states that appear pre-knee point, while the post-knee state S^* are those that do not have subsequent states. Here, they can be defined as $S^* = \{s | \sum_{j=1}^k a_{sj} = 0\}$ where k is the number of labeled states.

5.3. State-transition-driven cycle life prediction

Our framework predicts a cell's total cycle life (pre-knee + post-knee) by modeling state transitions between pre-knee and post-knee degradation regimes (Fig. 2a), inferred from early-cycle data. For a cell i observed up to cycle t^* (e.g., the first 3 cycles), the prediction involves two sequential steps: (1) pre-knee phase estimation and (2) post-knee phase estimation.

Prior to cycle life estimation, we perform key preparatory steps including feature engineering and dataset partitioning. We extract regression features $\mathbf{V}^r$ from early cycles (cutoff t^*) using domain-established methods [10] (Supplementary Table 1), and select classification features $\mathbf{V}^c$ via recursive feature elimination [20] (Supplementary Note 1). The dataset is split 80%/20% (training/validation), stratified by usage conditions and initial health.

To predict the duration of the pre-knee phase (L_{pre}), we employ a classification approach based on the cell's early-cycle data. A classifier processes initial cycling data to output a probability vector $\mathbf{p} = [p_1, p_2, \dots, p_k]$, where p_i represents the likelihood of the cell following state s_i . The process can be represented as $\mathbf{p} = h(\mathbf{V}^c)$. The tabular prior-data fitted network [21] is chosen as the classifier in the experiment. Each state s_i is associated with a precomputed average duration D_i , derived from the training dataset. The expected pre-knee life is then calculated as the weighted sum:

$$L_{pre} = \sum_{i=1}^k p_i D_i \quad (2)$$

The post-knee lifespan (L_{post}) is predicted through a hybrid model combining state transition dynamics and data-driven regression. The transition-driven term $L_{trans} = \mathbf{p} \cdot \mathbf{A} \cdot \mathbf{D}$ integrates the cell's state probability vector ($\mathbf{p}$), the state transition matrix ($\mathbf{A}$), and the state duration ($\mathbf{D}$). The data-driven regression part is to model residual nonlinear effects. The data-driven regression term $L_{reg} = g(\mathbf{V}^r)$, where the input is early-cycle features and the output is the post-knee state duration. The final prediction blends both terms:

$$L_{post} = \alpha \cdot L_{reg} + (1 - \alpha) \cdot L_{trans}, \alpha < 0.5 \quad (3)$$

, where $\alpha < 0.5$ reflects the dominance of state transition dynamics in degradation. Thus, the final predicted cycle life is:

$$L = L_{pre} + L_{post} \quad (4)$$

The framework includes the training stage and the predicting stage. In the training stage, the state transition matrix $\mathbf{A}$, the pre-knee states S^0 , the post-knee states S^* , and the state durations $\mathbf{D}$ are computed with the training sets. First, the classifier is trained on early-cycle features using cross-entropy loss to accurately map input data to pre-knee state probabilities ($\mathbf{p}$). Once optimized, the classifier's weights are frozen to prevent co-adaptation. Next, the regressor is trained using both the classifier's probability outputs and early-cycle features. We use root-mean-square error (RMSE) [10] as the loss function. In the predicting stage, given early cycle data as input, regression features $\mathbf{V}^r$ and classification features $\mathbf{V}^c$ are extracted and fed in the regressor g and the classifier h , respectively. The state in one of the pre-knee states and its duration can be determined. The following post-knee states and their durations are produced with the Monte Carlo algorithm. Eventually, after a linear summation of every state duration, the cycle life is predicted.

The following post-knee states $s^* \in S^*$ are produced after the pre-knee state s^0 by the Monte Carlo algorithm according to a cumulative distribution function

$$\Phi(s) = \sum_{s_j \leq s} \mathbf{A}_{s_j}^{s^0} \quad (5)$$

in the transition matrix $\mathbf{A}$. A random number $p^* \in [0, 1]$ is generated. The post-knee state can be determined by the first state in matrix $\mathbf{A}$ that exceeds the random value, i.e.,

$$\text{argmin } s \quad \text{s.t.} \quad \Phi(s) \geq p^* \quad (6)$$

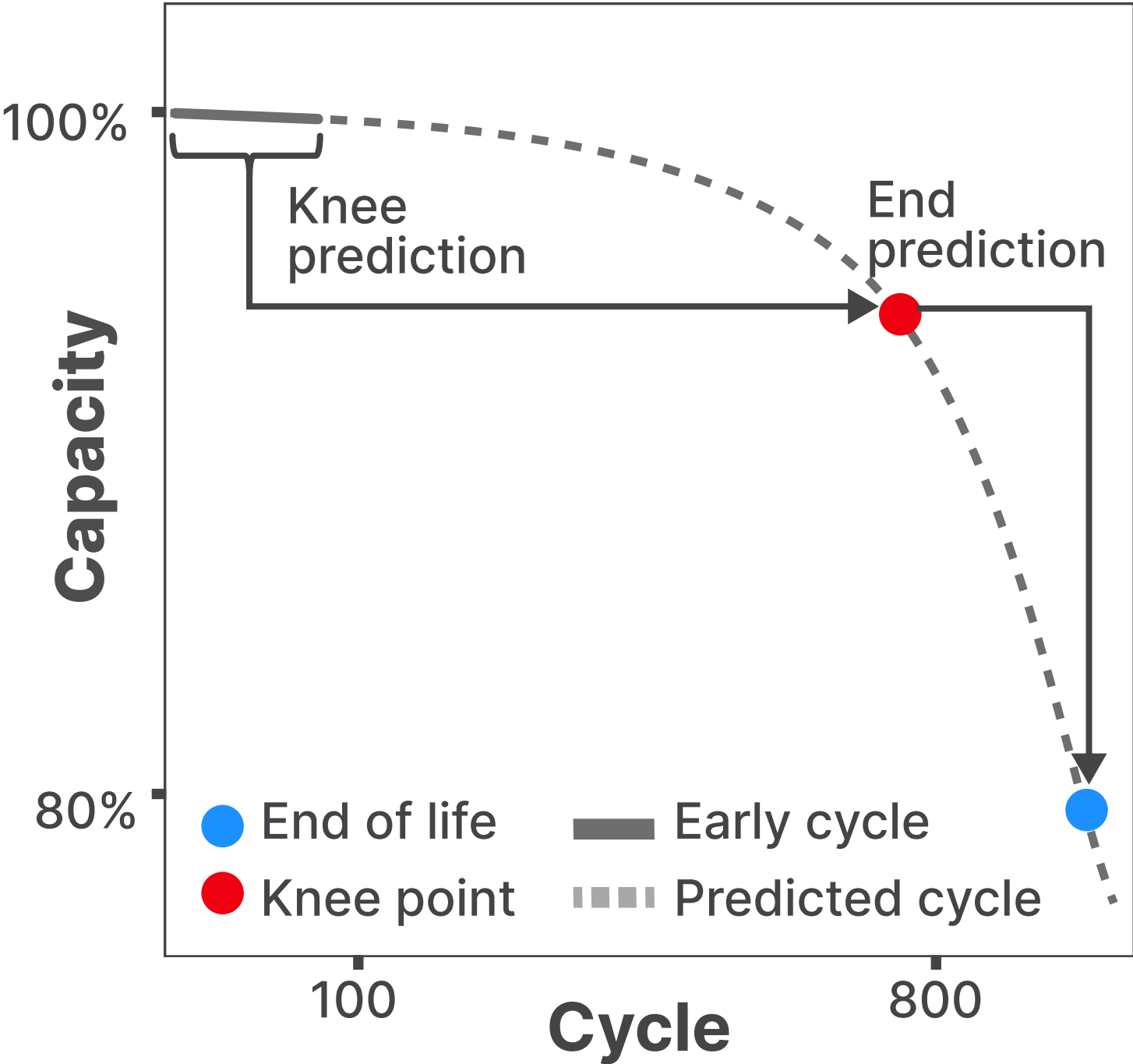
effectively choosing the state with the maximal probability.

References

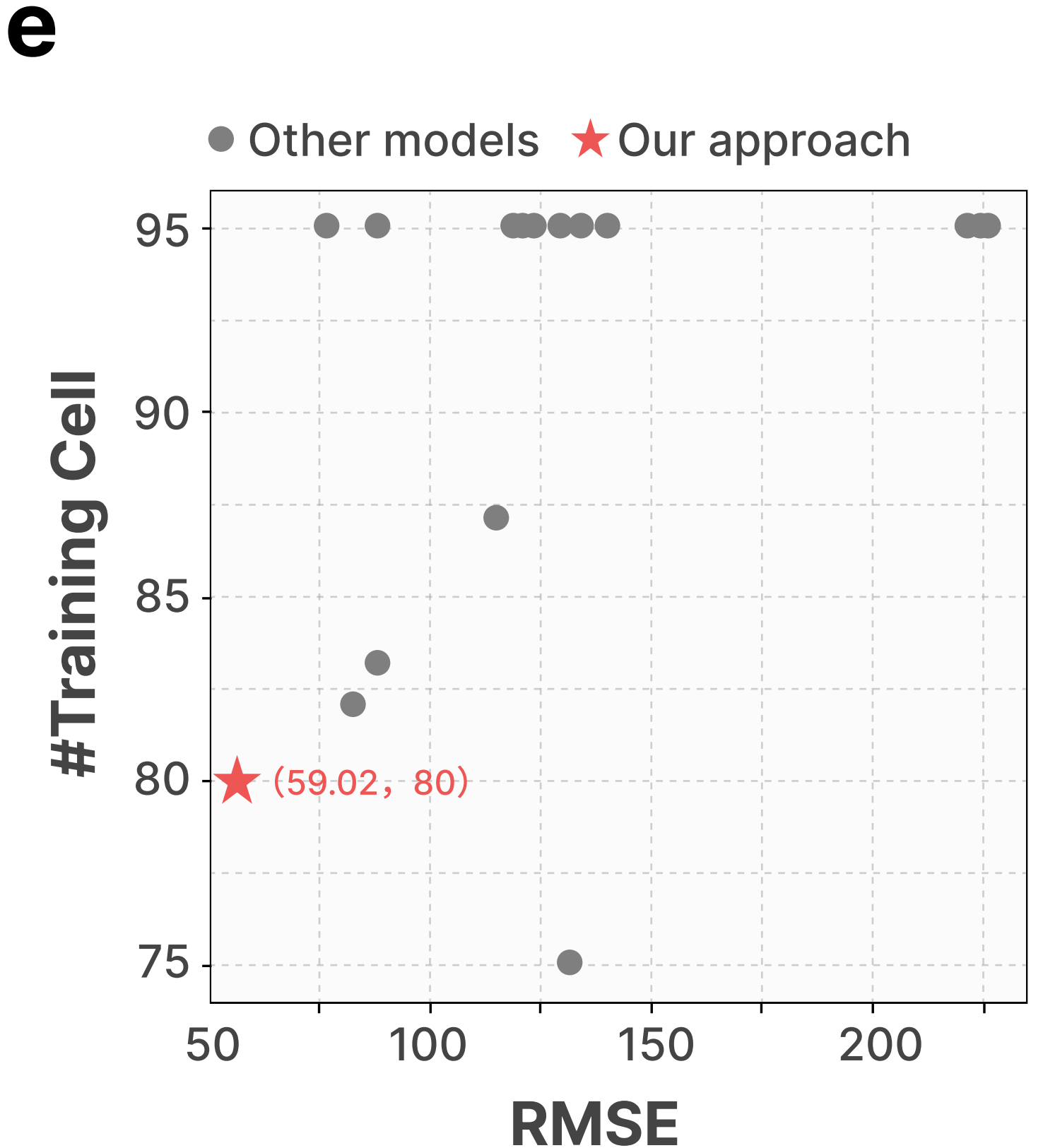
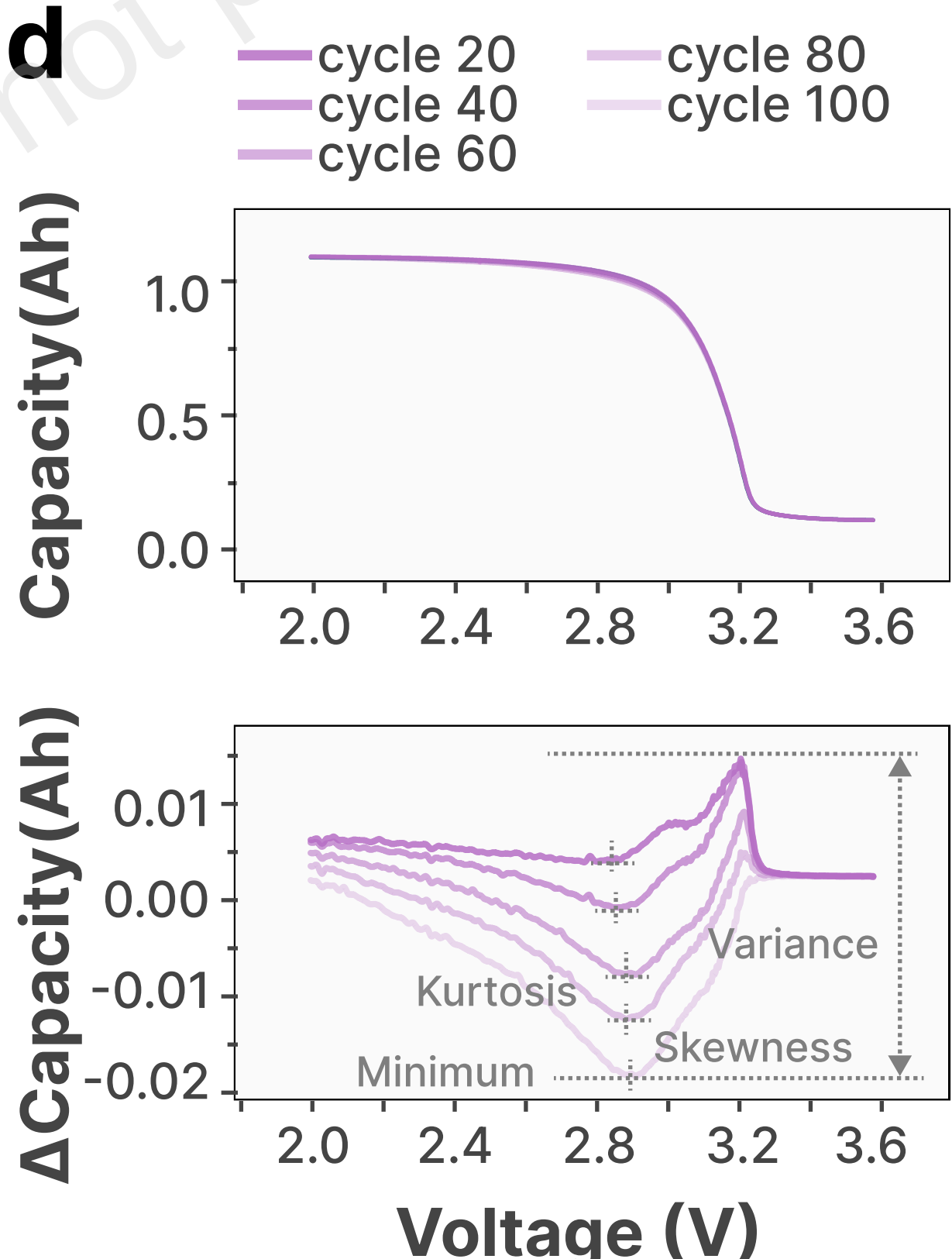
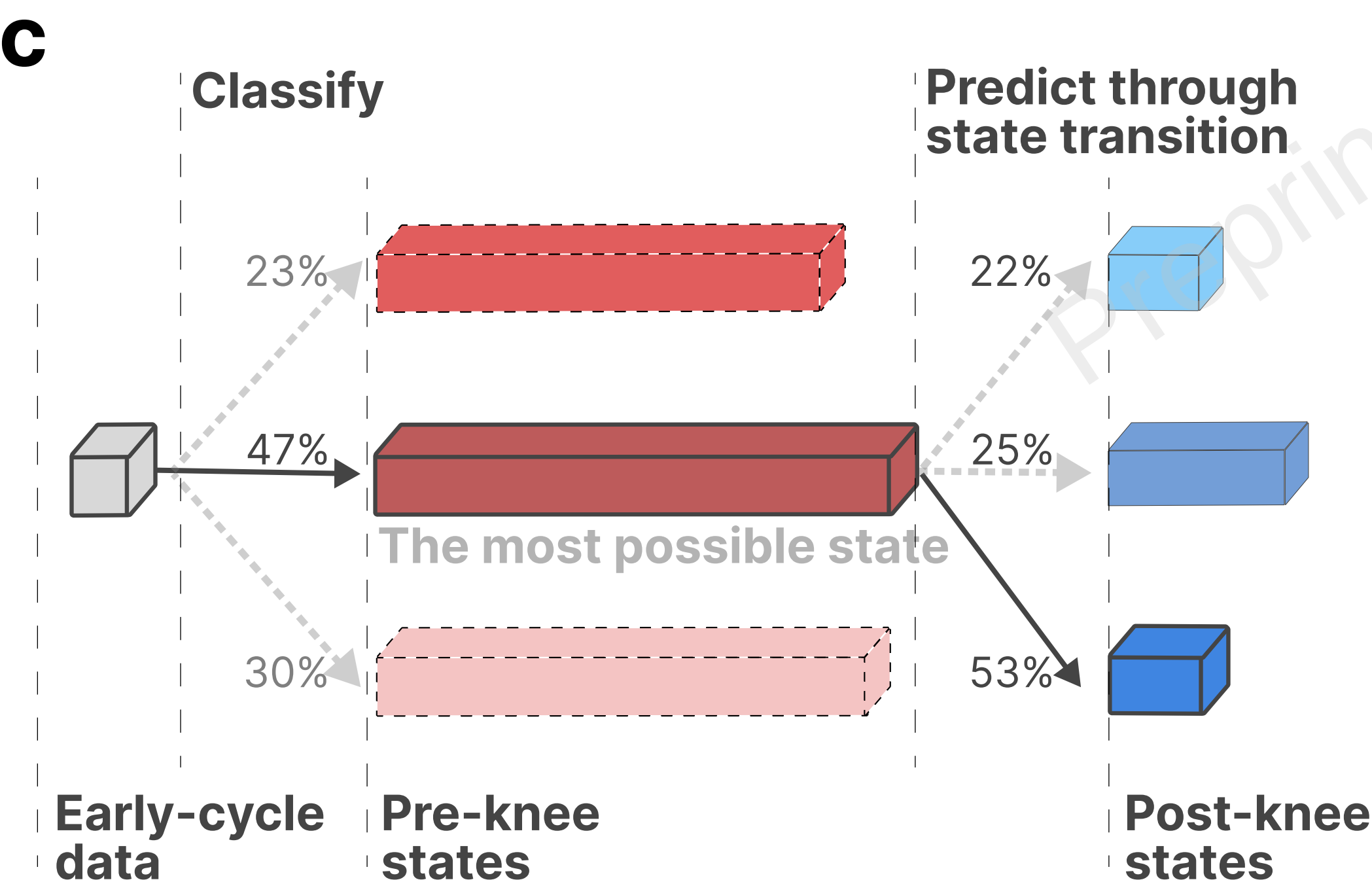
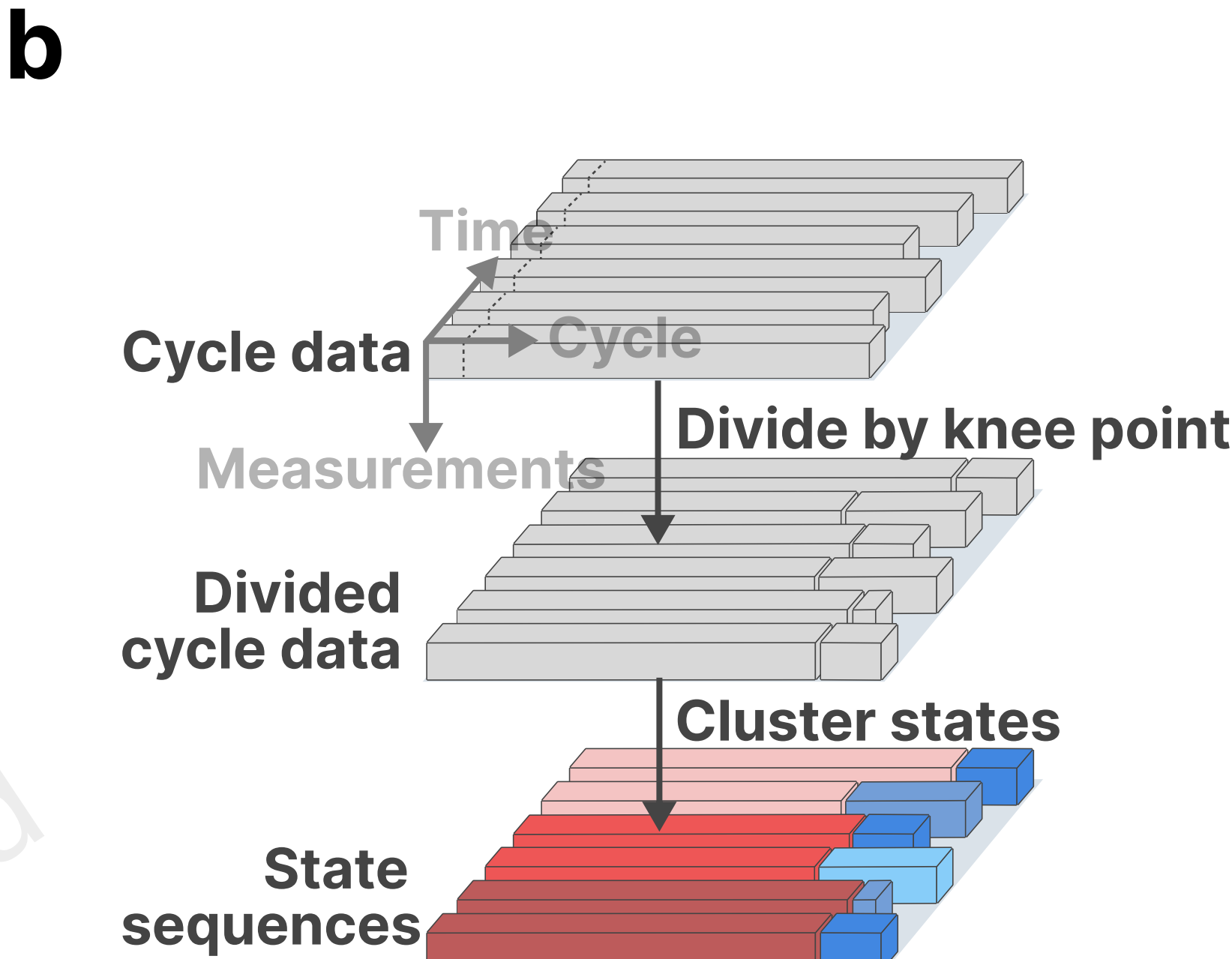
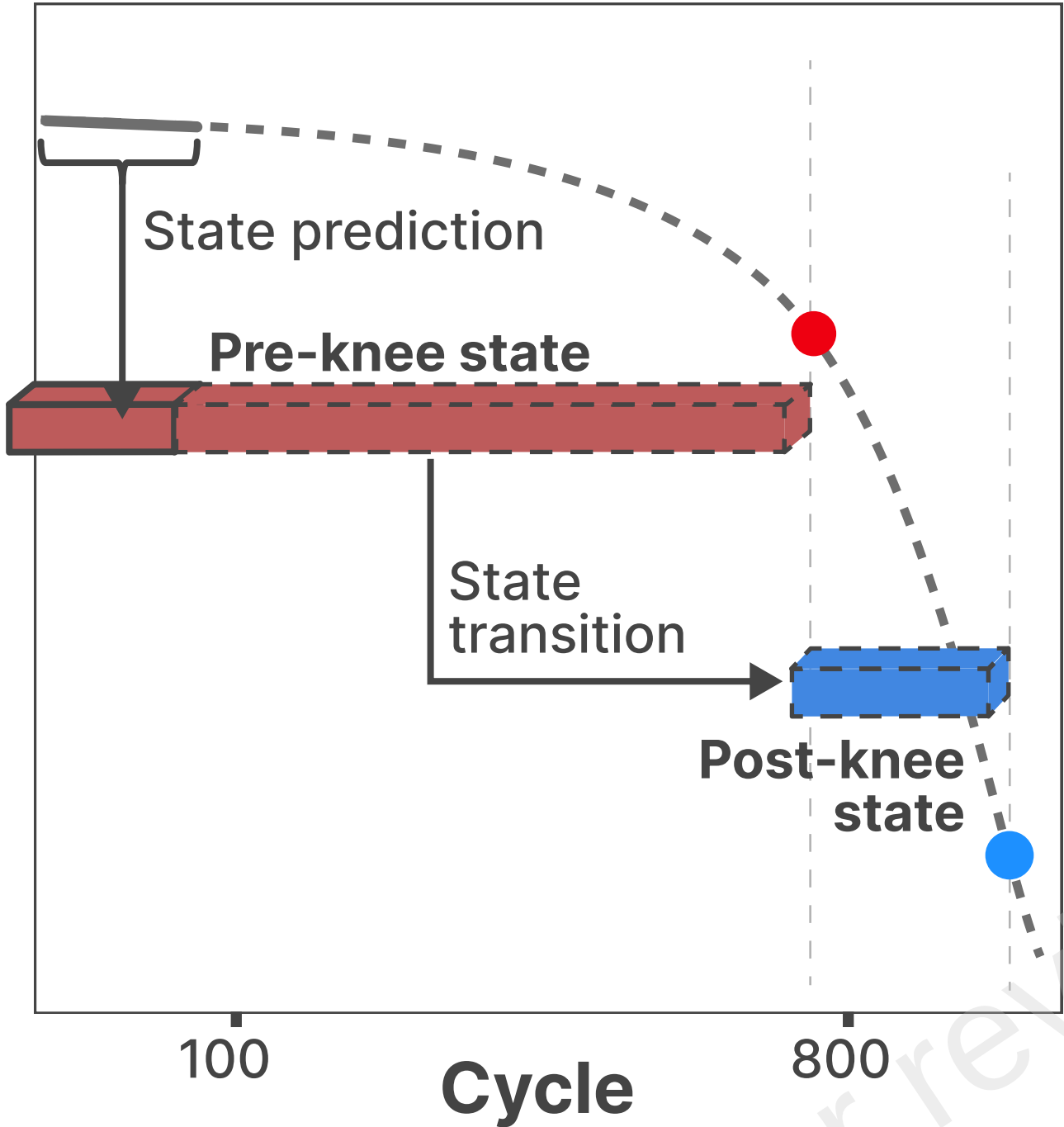
- [1] Statista, Lithium-ion batteries - statistics & facts, Website, <https://www.statista.com/topics/2049/lithium-ion-battery-industry/>, accessed on 2026-06-18 (2023).
- [2] A. Geslin, B. van Vlijmen, X. Cui, A. Bhargava, P. A. Asinger, R. D. Braatz, W. C. Chueh, Selecting the appropriate features in battery lifetime predictions, *Joule* 7 (9) (2023) 1956–1965. doi:10.1016/j.joule.2023.07.021.
- [3] X. Hu, L. Xu, X. Lin, M. Pecht, Battery lifetime prognostics, *Joule* 4 (2) (2020) 310–346. doi:10.1016/j.joule.2019.11.018.
- [4] Y. Lu, C.-Z. Zhao, J.-Q. Huang, Q. Zhang, The timescale identification decoupling complicated kinetic processes in lithium batteries, *Joule* 6 (6) (2022) 1172–1198. doi:10.1016/j.joule.2022.05.005.
- [5] D. P. Finegan, J. Zhu, X. Feng, M. Keyser, M. Ulmefors, W. Li, M. Z. Bazant, S. J. Cooper, The application of data-driven methods and physics-based learning for improving battery safety, *Joule* 5 (2) (2021) 316–329. doi:10.1016/j.joule.2020.11.018.
- [6] P. M. Attia, A. Grover, N. Jin, K. A. Severson, T. Markov, Y.-H. Liao, M. H. Chen, B. Cheong, N. Perkins, Z. Yang, P. K. Herring, M. Aykol, S. J. Harris, R. D. Braatz, S. Ermon, W. C. Chueh, Closed-loop optimization of fast-charging protocols for batteries with machine learning, *Nature* 578 (2020) 397–402. doi:10.1038/s41586-020-1994-5.
- [7] X. Jia, C. Zhang, Y. Li, C. Zou, L. Y. Wang, X. Cai, Knee-point-conscious battery aging trajectory prediction based on physics-guided machine learning, *IEEE Trans. Transport. Electrification* 10 (1) (2024) 1056–1069. doi:10.1109/TTE.2023.3266386.
- [8] F. von Bülow, T. Meisen, A review on methods for state of health forecasting of lithium-ion batteries applicable in real-world operational conditions, *J. Energy Storage* 57 (2023) 105978. doi:10.1016/j.est.2022.105978.
- [9] N. Guo, S. Chen, J. Tao, Y. Liu, J. Wan, X. Li, Semi-supervised learning for explainable few-shot battery lifetime prediction, *Joule* 8 (6) (2024) 1820–1836. doi:10.1016/j.joule.2024.02.020.
- [10] K. A. Severson, P. M. Attia, N. Jin, N. Perkins, B. Jiang, Z. Yang, M. H. Chen, M. Aykol, P. K. Herring, D. Fraggadakis, M. Z. Bazant, S. J. Harris, W. C. Chueh, R. D. Braatz, Data-driven prediction of battery cycle life before capacity degradation, *Nat. Energy* 4 (2019) 383–391. doi:10.1038/s41560-019-0356-8.

- [11] J. Lu, R. Xiong, J. Tian, C. Wang, C.-W. Hsu, N.-T. Tsou, F. Sun, J. Li, Battery degradation prediction against uncertain future conditions with recurrent neural network enabled deep learning, *Energy Storage Mater.* 50 (2022) 139–151. doi:10.1016/j.ensm.2022.05.007.
- [12] Z. Wang, F. Yang, Q. Xu, Y. Wang, H. Yan, M. Xie, Capacity estimation of lithium-ion batteries based on data aggregation and feature fusion via graph neural network, *Appl. Energ.* 336 (2023) 120808–120819. doi:10.1016/j.apenergy.2023.120808.
- [13] W. Li, N. Sengupta, P. Dechent, D. Howey, A. Annaswamy, D. U. Sauer, One-shot battery degradation trajectory prediction with deep learning, *J. Power Sources* 506 (2021) 230024–230034. doi:10.1016/j.jpowsour.2021.230024.
- [14] P. M. Attia, A. Bills, F. B. Planella, P. Dechent, G. dos Reis, M. Dubarry, P. Gasper, R. Gilchrist, S. Greenbank, D. Howey, O. Liu, E. Khoo, Y. Preger, A. Soni, S. Sripad, A. G. Stefanopoulou, V. Sulzer, Review—“knees” in lithium-ion battery aging trajectories, *J. Electrochem. Soc.* 169 (6) (2022) 60517–60545. doi:10.1149/1945-7111/ac6d13.
- [15] M. Haris, M. N. Hasan, S. Qin, Degradation curve prediction of lithium-ion batteries based on knee point detection algorithm and convolutional neural network, *IEEE Trans. Instrum. Meas.* 71 (2022) 1–10. doi:10.1109/TIM.2022.3181307.
- [16] K. Q. Zhou, Y. Qin, C. Yuen, Lithium-ion battery state of health estimation by matrix profile empowered online knee onset identification, *IEEE Trans. Transport. Electrific.* 10 (1) (2024) 1935–1946. doi:10.1109/TTE.2023.3265981.
- [17] J. Zhang, Y. Zhang, J. Fu, D. Zhao, P. Liu, Z. Zhang, Capacity fading knee-point recognition method and life prediction for lithium-ion batteries using segmented capacity degradation model, *Reliab. Eng. Syst. Safe.* 251 (2024) 110395–110405. doi:10.1016/j.ress.2024.110395.
- [18] X. Cui, S. D. Kang, S. Wang, J. A. Rose, H. Lian, A. Geslin, S. B. Torrisi, M. Z. Bazant, S. Sun, W. C. Chueh, Data-driven analysis of battery formation reveals the role of electrode utilization in extending cycle life, *Joule* 8 (11) (2024) 3072–3087. doi:10.1016/j.joule.2024.07.024.
- [19] P. Fermín-Cueto, E. McTurk, M. Allerhand, E. Medina-Lopez, M. F. Anjos, J. Sylvester, G. dos Reis, Identification and machine learning prediction of knee-point and knee-onset in capacity degradation curves of lithium-ion cells, *Energy and AI* 1 (2020) 100006–100015. doi:10.1016/j.egyai.2020.100006.
- [20] I. Guyon, J. Weston, S. Barnhill, V. Vapnik, Gene selection for cancer classification using support vector machines, *Mach. Learn.* 46 (1) (2002) 389–422. doi:10.1023/A:1012487302797.
- [21] N. Hollmann, S. Müller, L. Purucker, A. Krishnakumar, M. Körfer, S. B. Hoo, R. T. Schirmer, F. Hutter, Accurate predictions on small data with a tabular foundation model, *Nature* 637 (8045) (2025) 319–326. doi:10.1038/s41586-024-08328-6.
- [22] H. Zhang, Y. Li, S. Zheng, Z. Lu, X. Gui, W. Xu, J. Bian, Battery lifetime prediction across diverse ageing conditions with inter-cell deep learning, *Nat. Mach. Intell.* 7 (2) (2025) 270–277. doi:10.1038/s42256-024-00972-x.
- [23] H. Zou, T. Hastie, Regularization and variable selection via the elastic net, *J. R. Stat. Soc. Ser. B Stat. Methodol.* 67 (2) (2005) 301–320. doi:10.1111/j.1467-9868.2005.00503.x.
- [24] J. R. Quinlan, Induction of decision trees, *Mach. Learn.* 1 (1) (1986) 81–106. doi:10.1007/BF00116251.
- [25] F. Rosenblatt, The perceptron: a probabilistic model for information storage and organization in the brain, *Psychol. Rev.* 65 (6) (1958) 386. doi:10.1037/h0042519.
- [26] J. Hou, T. Su, T. Gao, Y. Yang, W. Xue, Early prediction of battery lifetime for lithium-ion batteries based on a hybrid clustered CNN model, *Energy* 319 (2025) 134992. doi:10.1016/j.energy.2025.134992.
- [27] Z. Chen, L. Chen, Z. Ma, K. Xu, Y. Zhou, W. Shen, Joint modeling for early predictions of li-ion battery cycle life and degradation trajectory, *Energy* 277 (2023) 127633. doi:10.1016/j.energy.2023.127633.
- [28] B. Zhao, W. Zhang, Y. Zhang, C. Zhang, C. Zhang, J. Zhang, Research on the remaining useful life prediction method for lithium-ion batteries by fusion of feature engineering and deep learning, *Appl. Energ.* 358 (2024) 122325. doi:10.1016/j.apenergy.2023.122325.
- [29] A. C. Belkina, C. Ciccolella, R. Anno, R. L. Halpert, J. Spidlen, J. E. Snyder-Cappione, Automated optimized parameters for T-distributed stochastic neighbor embedding improve visualization and analysis of large datasets, *Nat. Commun.* 10 (1) (2019) 5415–5426. doi:10.1038/s41467-019-13055-y.
- [30] M. Kim, I. Kim, J. Kim, J. W. Choi, Lifetime prediction of lithium ion batteries by using the heterogeneity of graphite anodes, *ACS Energy Lett.* 8 (7) (2023) 2946–2953. doi:10.1021/acsenenergylett.3c00695.
- [31] I. Bloom, A. N. Jansen, D. P. Abraham, J. Knuth, S. A. Jones, V. S. Battaglia, G. L. Henriksen, Differential voltage analyses of high-power, lithium-ion cells: 1. Technique and application, *J. Power Sources* 139 (1) (2005) 295–303. doi:10.1016/j.jpowsour.2004.07.021.
- [32] K. Ando, T. Matsuda, D. Imamura, Degradation diagnosis of lithium-ion batteries with a $\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2$ and LiMn_2O_4 blended cathode using dV/dQ curve analysis, *J. Power Sources* 390 (1) (2018) 278–285. doi:10.1016/j.jpowsour.2018.04.043.
- [33] T. Lin, S. Chen, S. J. Harris, T. Zhao, Y. Liu, J. Wan, Investigating explainable transfer learning for battery lifetime prediction under state transitions, *eScience* (2024) 100280–100304. doi:10.1016/j.esci.2024.100280.
- [34] Y. Zhang, Q. Tang, Y. Zhang, J. Wang, U. Stimming, A. A. Lee, Identifying degradation patterns of lithium ion batteries from impedance spectroscopy using machine learning, *Nat. Commun.* 11 (2020) 1706–1711. doi:10.1038/s41467-020-15235-7.
- [35] D. Arthur, S. Vassilvitskii, K-means++: The advantages of careful seeding, in: *Proc. Annu. ACM-SIAM Symp. Discret. Algorithms*, Society for Industrial and Applied Mathematics, Philadelphia, PA, USA, 2007, pp. 1027–1035. doi:10.5555/1283383.1283494.
- [36] T. Ouyang, C. Wang, P. Xu, J. Ye, B. Liu, Prognostics and health management of lithium-ion batteries based on modeling techniques and bayesian approaches: A review, *Sustainable Energy Technol. Assess.* 55 (2023) 102915. doi:10.1016/j.seta.2022.102915.
- [37] L. Rabiner, A tutorial on hidden Markov models and selected applications in speech recognition, *Proc. IEEE* 77 (2) (1989) 257–286. doi:10.1109/5.18626.

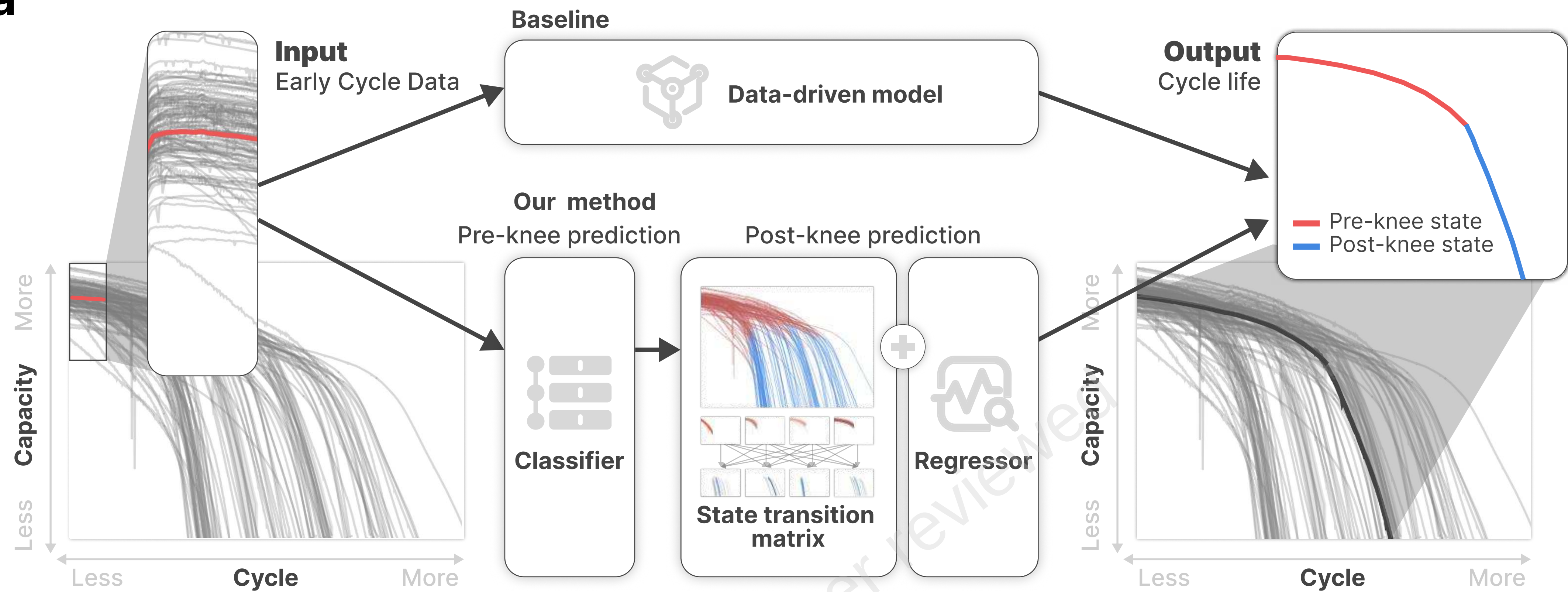
a **a1** Knee-end life prediction



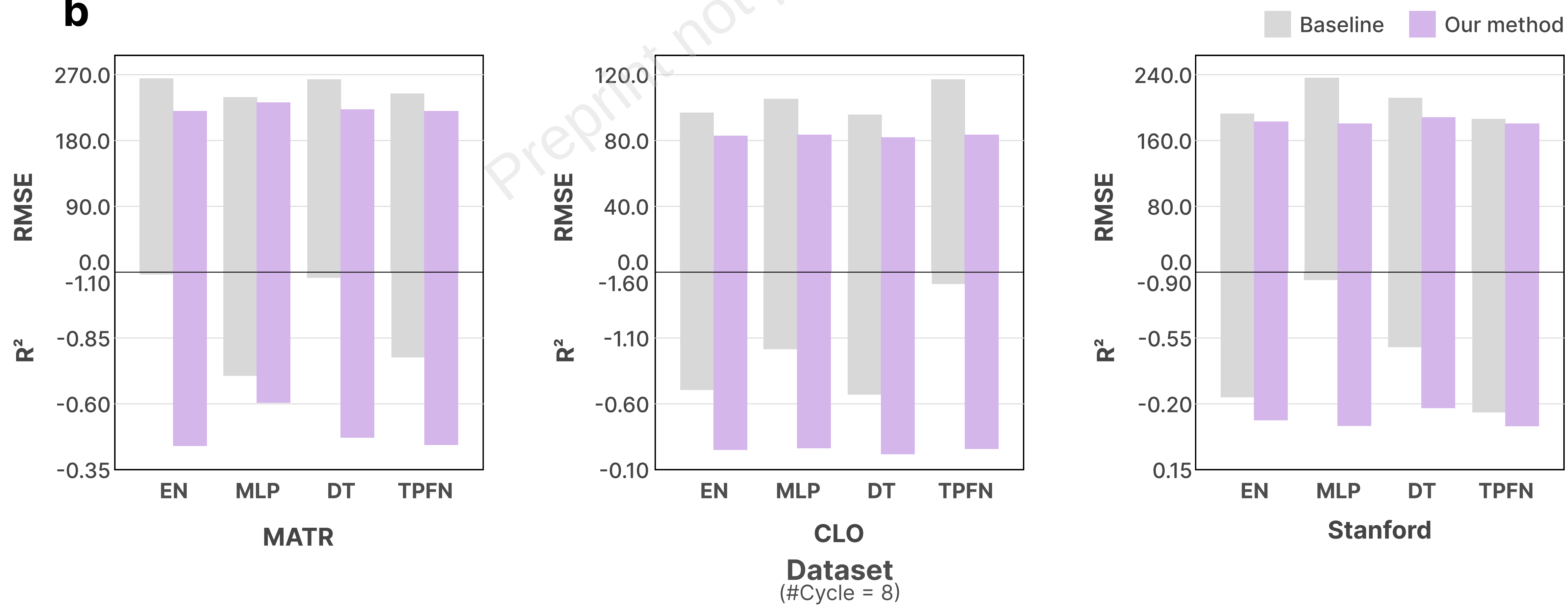
a2 State-based life prediction

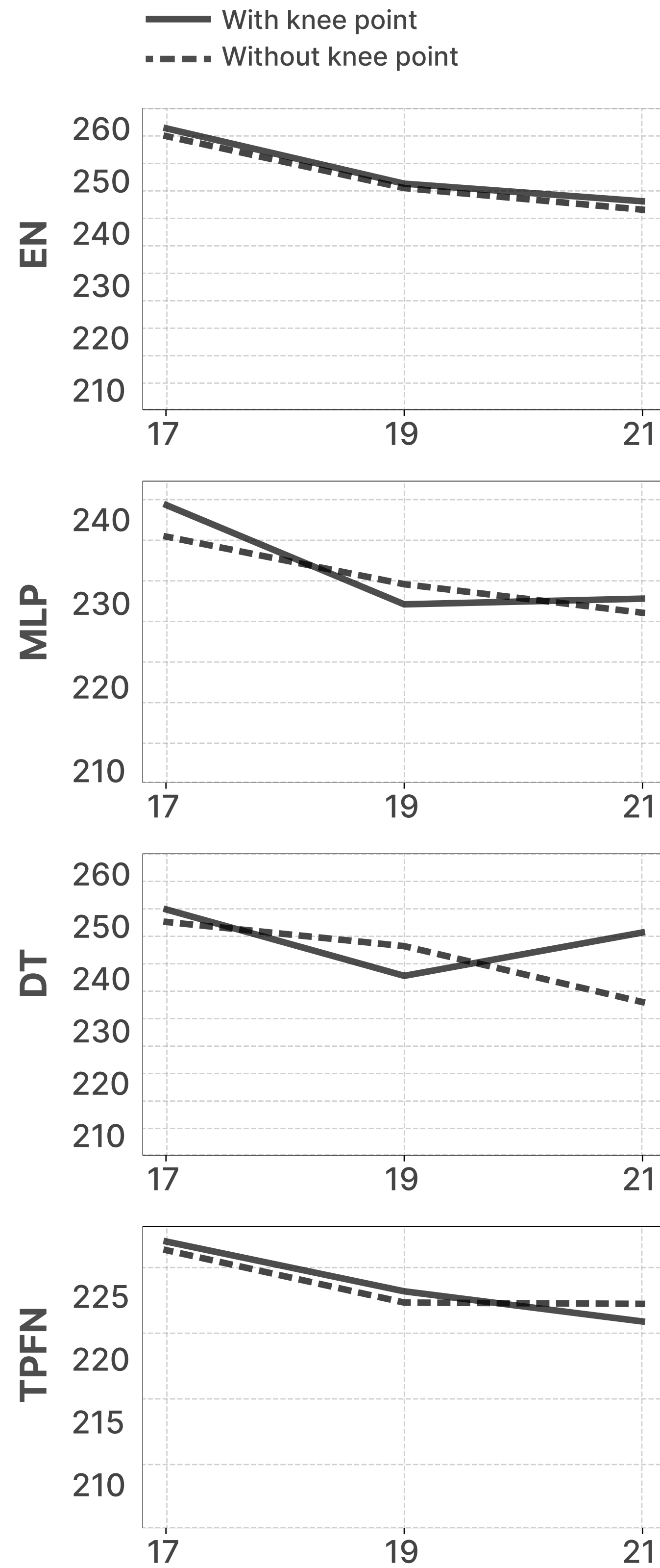
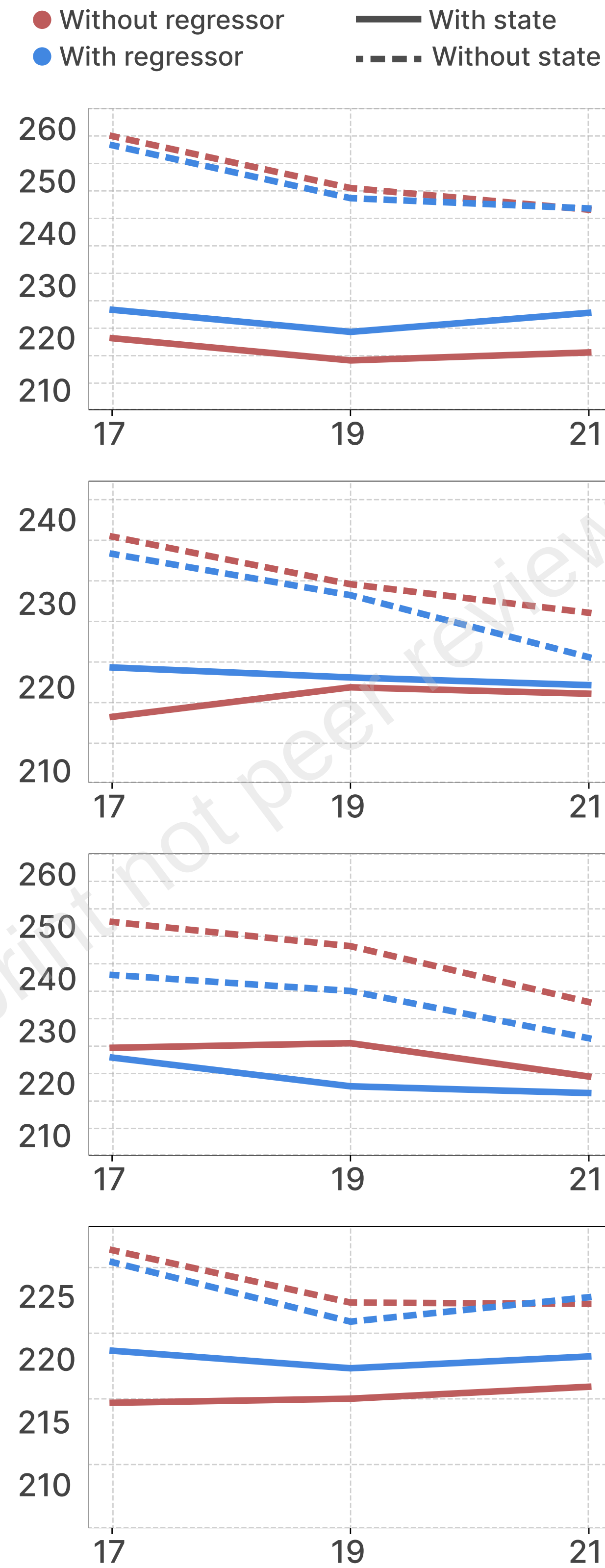
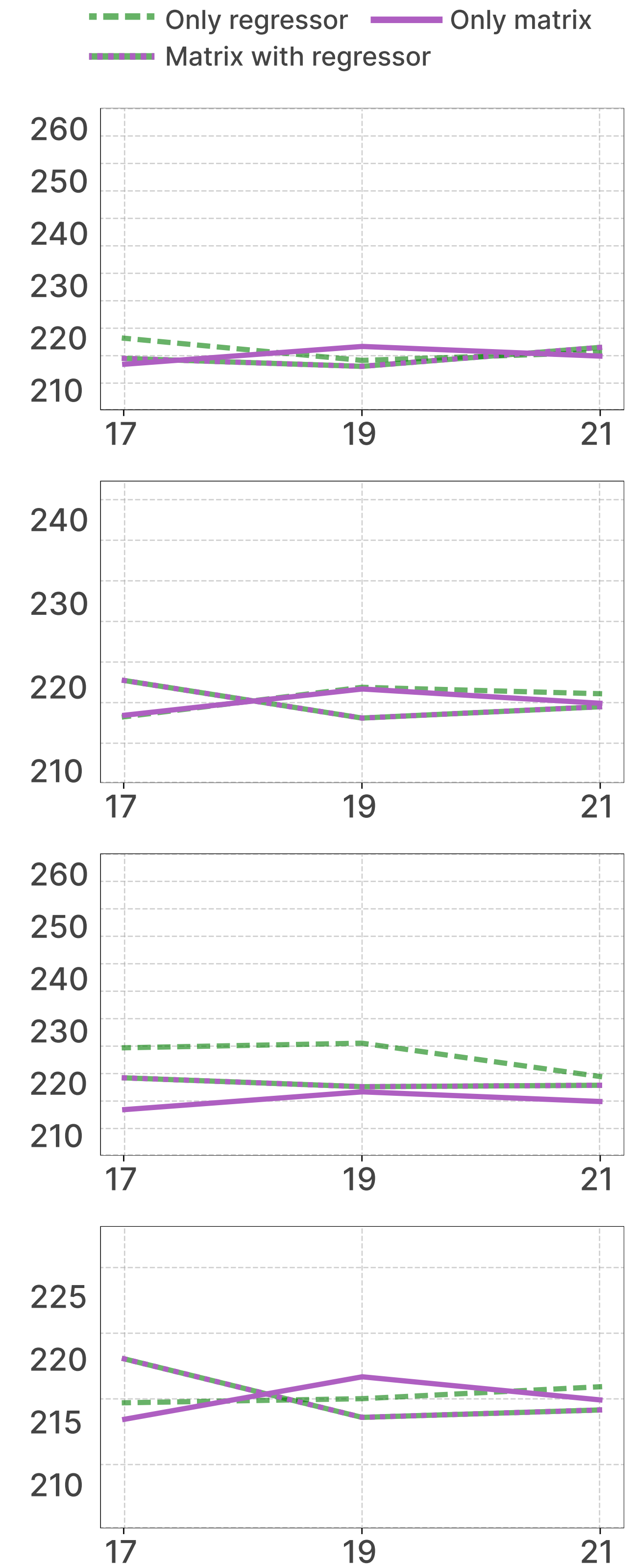


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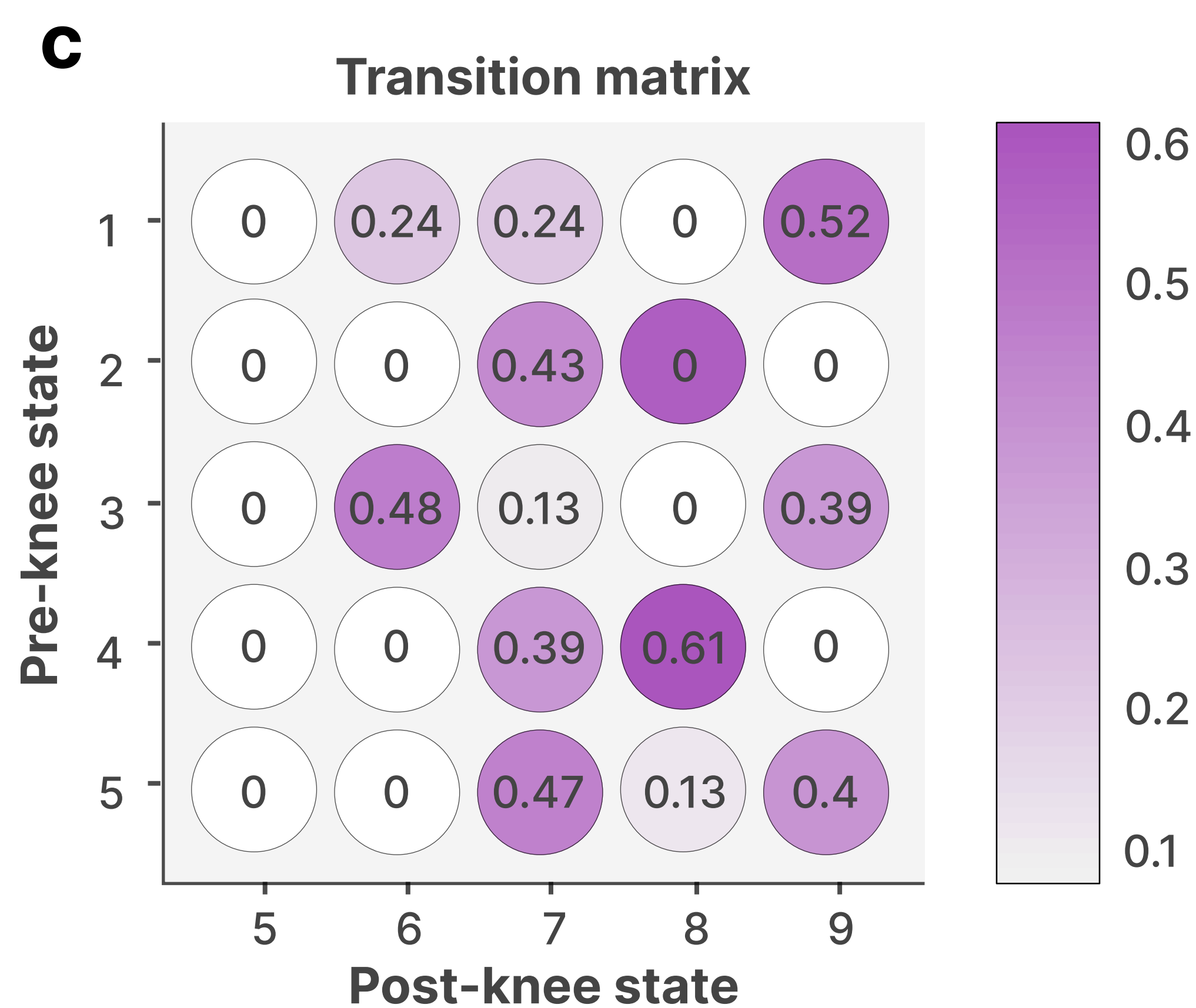
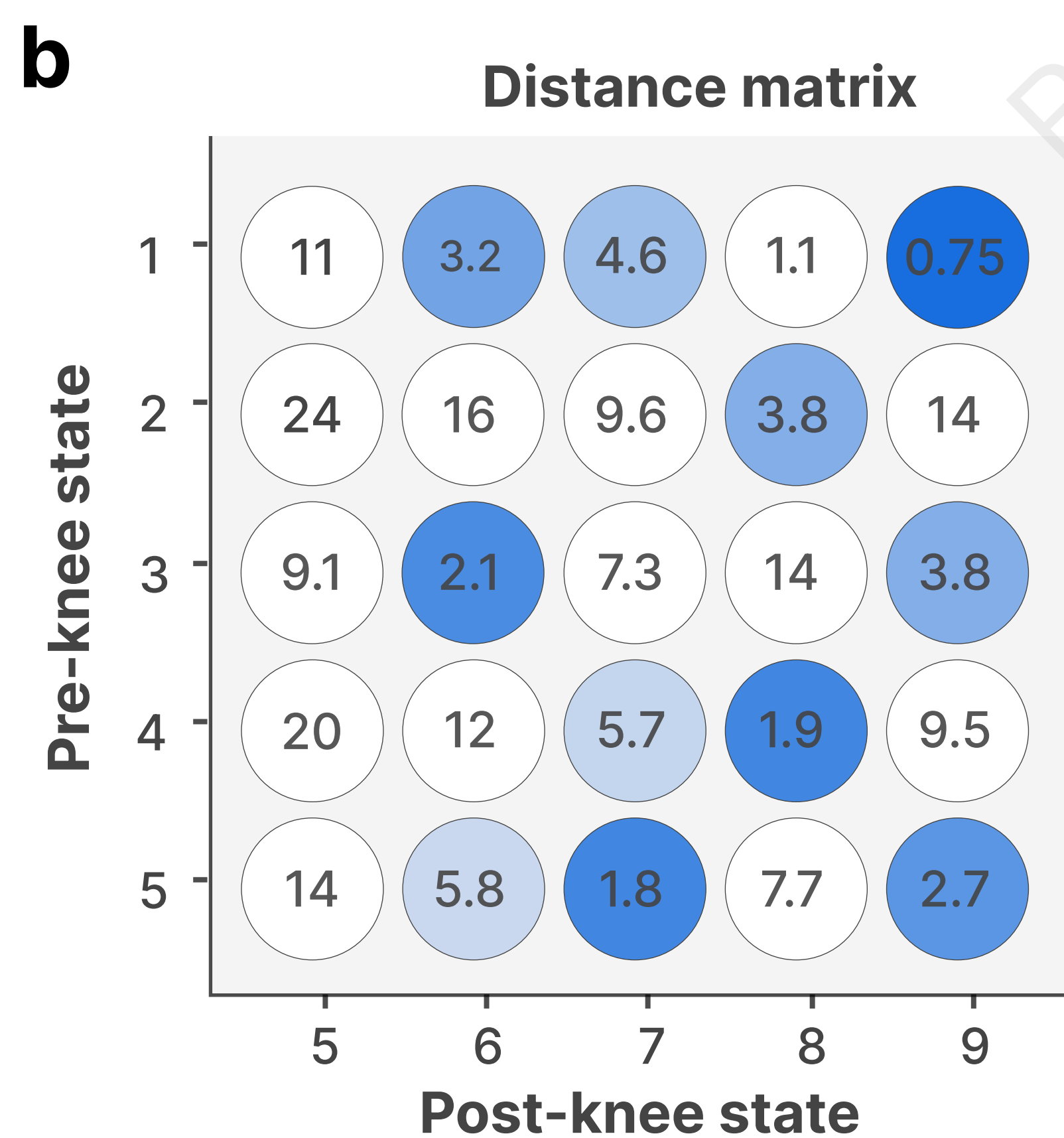
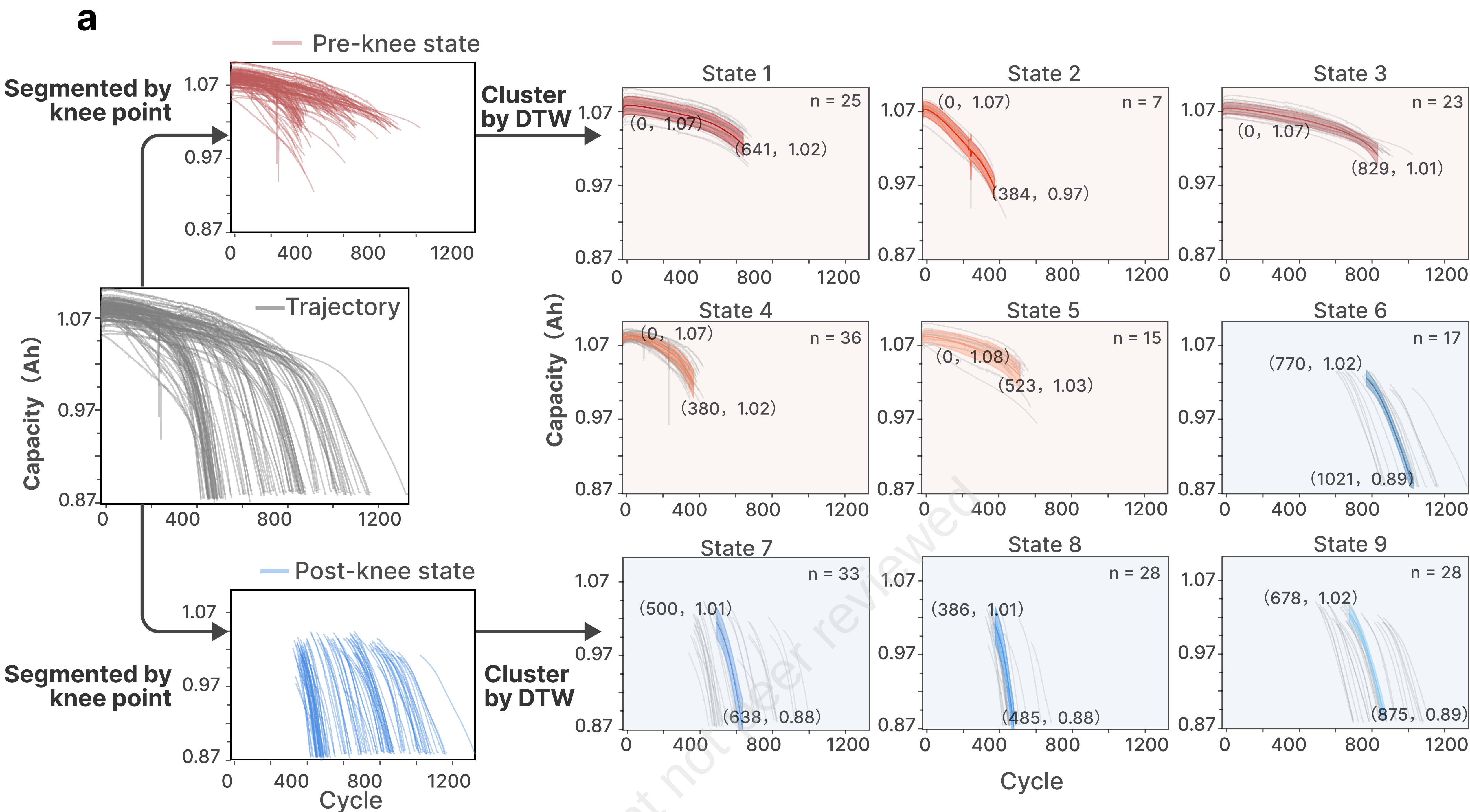


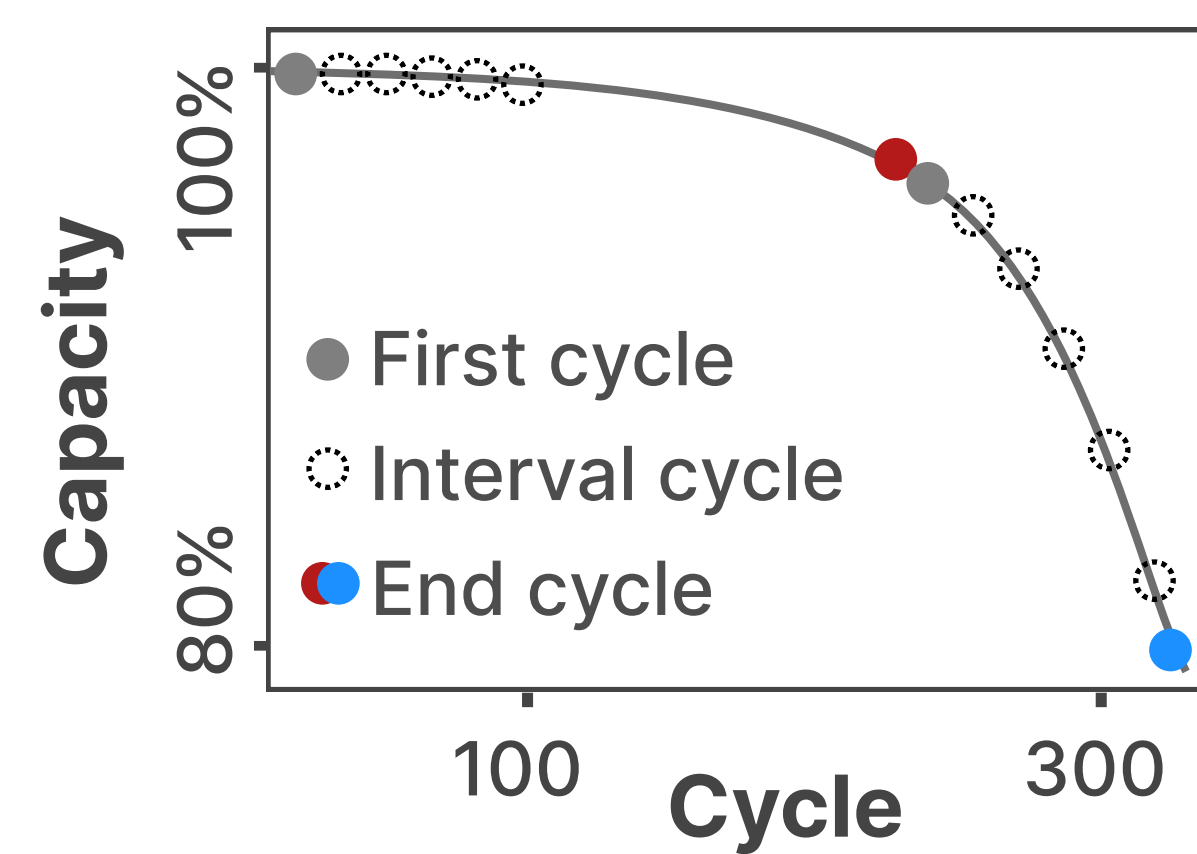
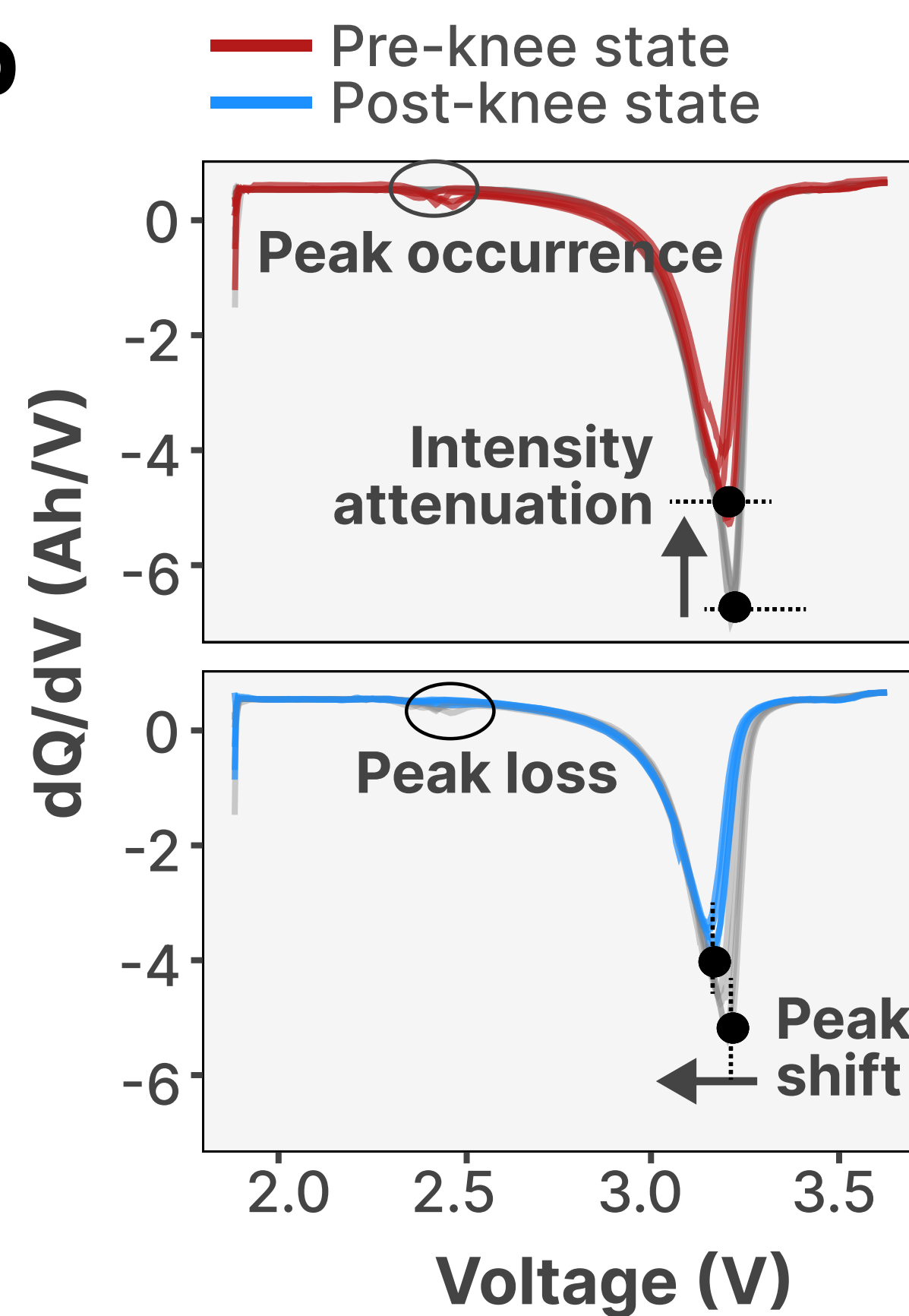
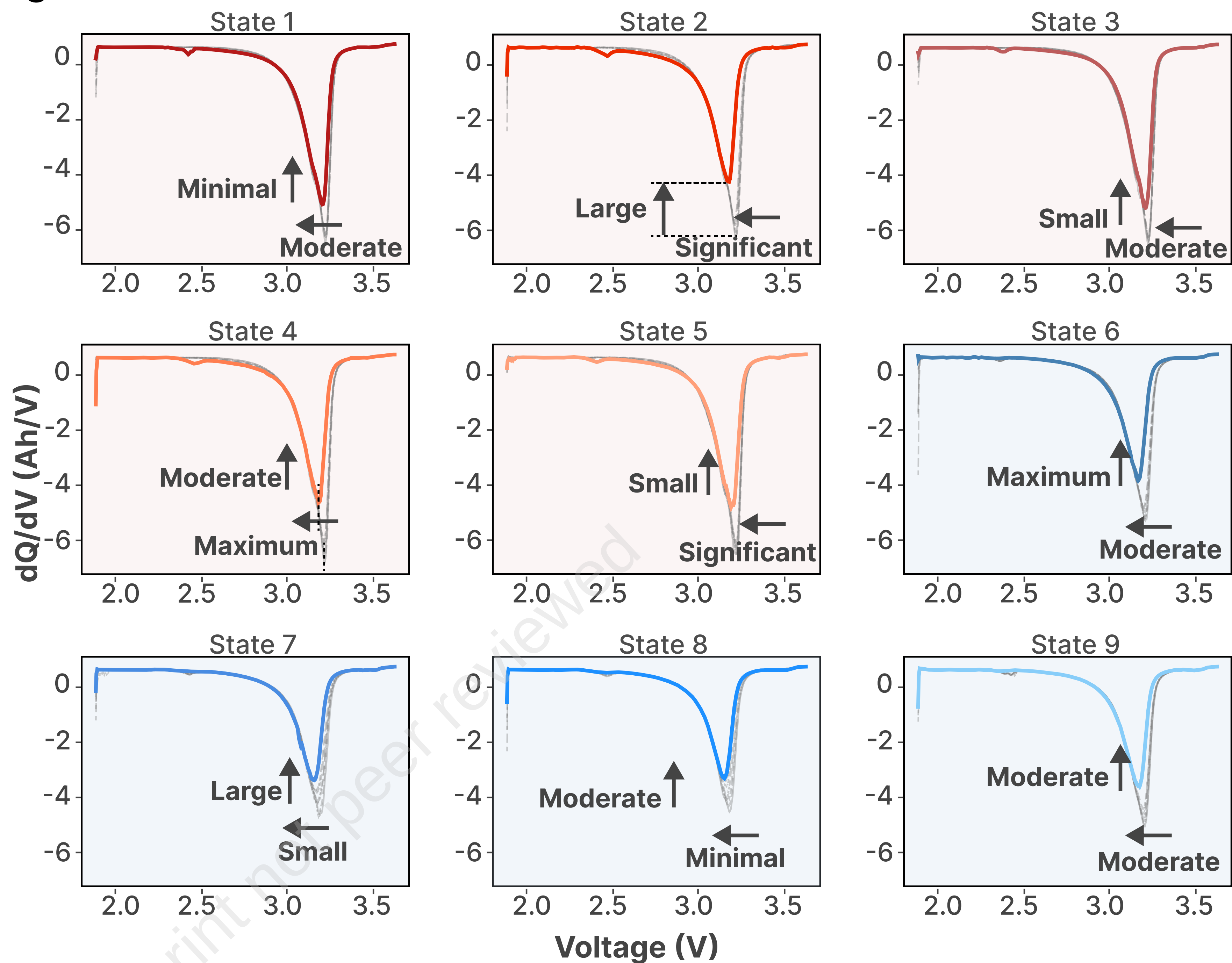
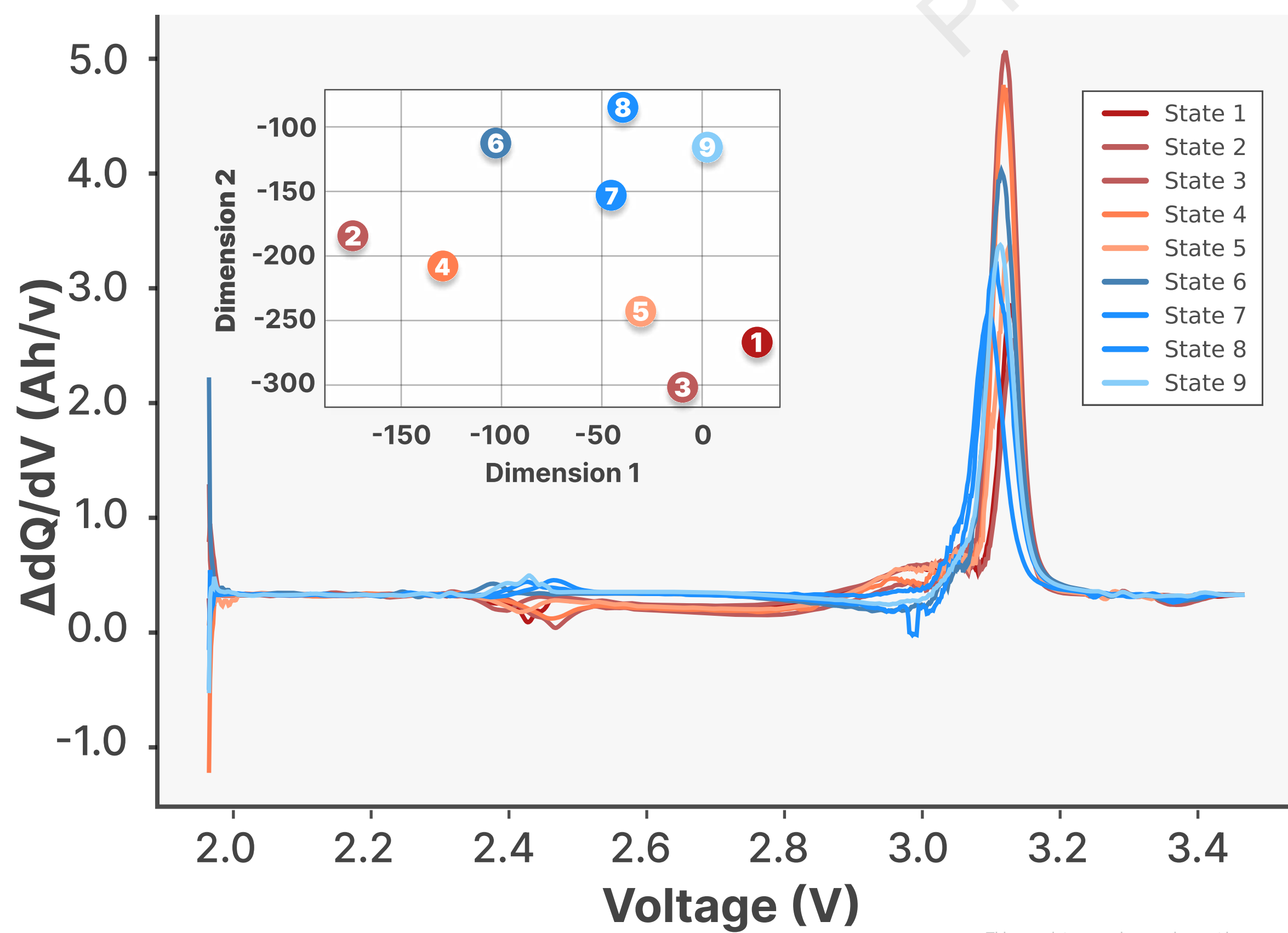
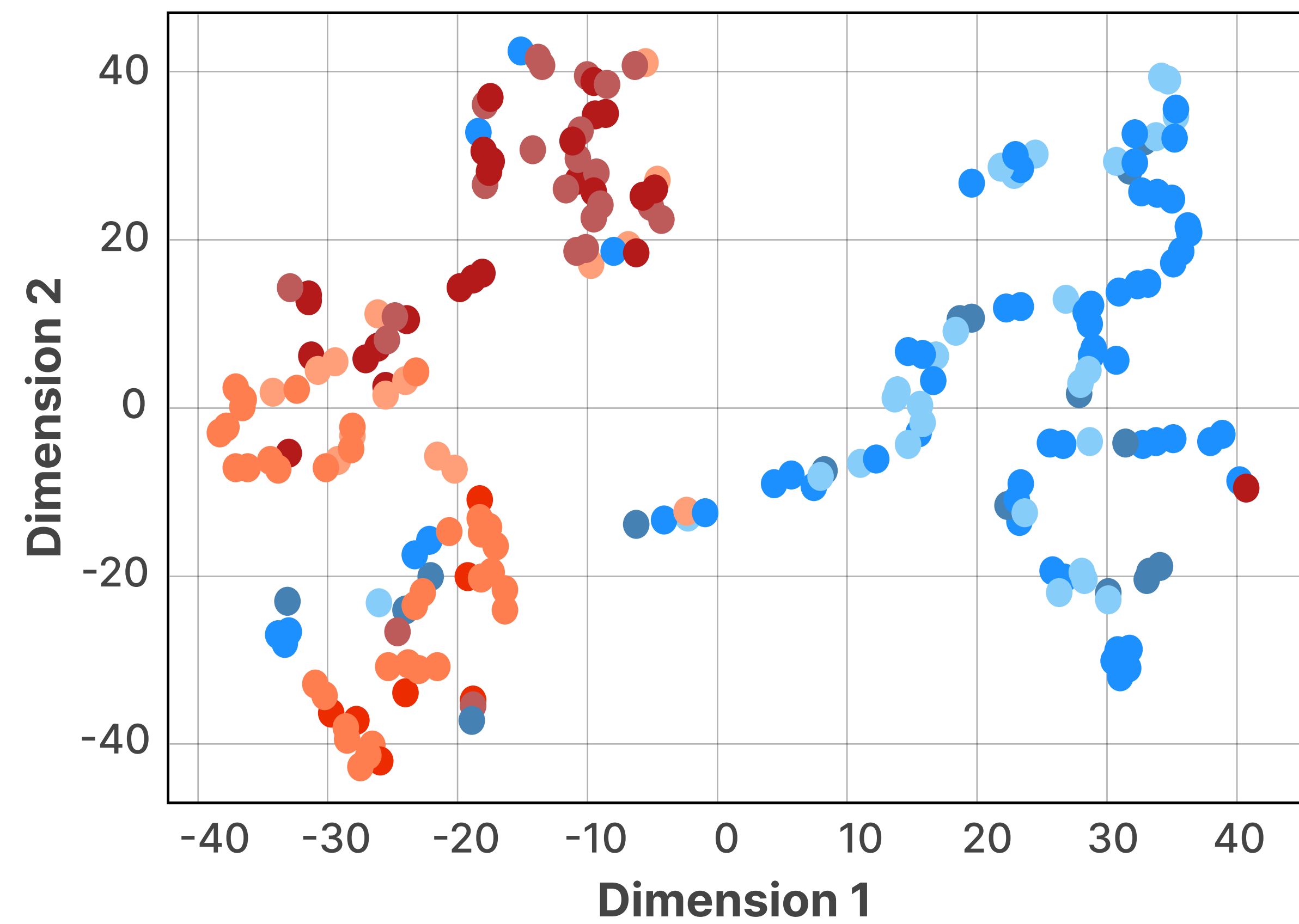
b



a**Directly knee point modeling****b****Pre-knee state modeling****c****Post-knee state modeling**

#Cell
(#Cycle = 8)

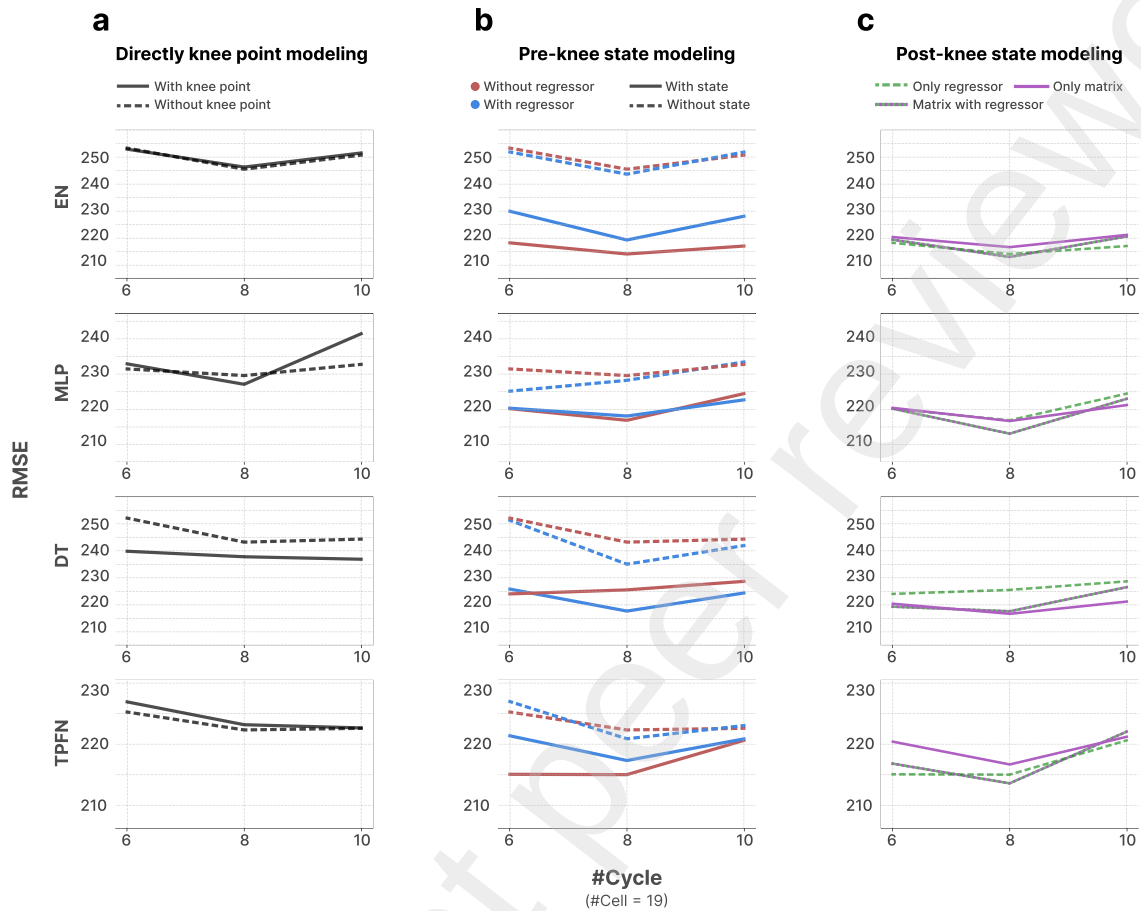


a**b****c****d****e**

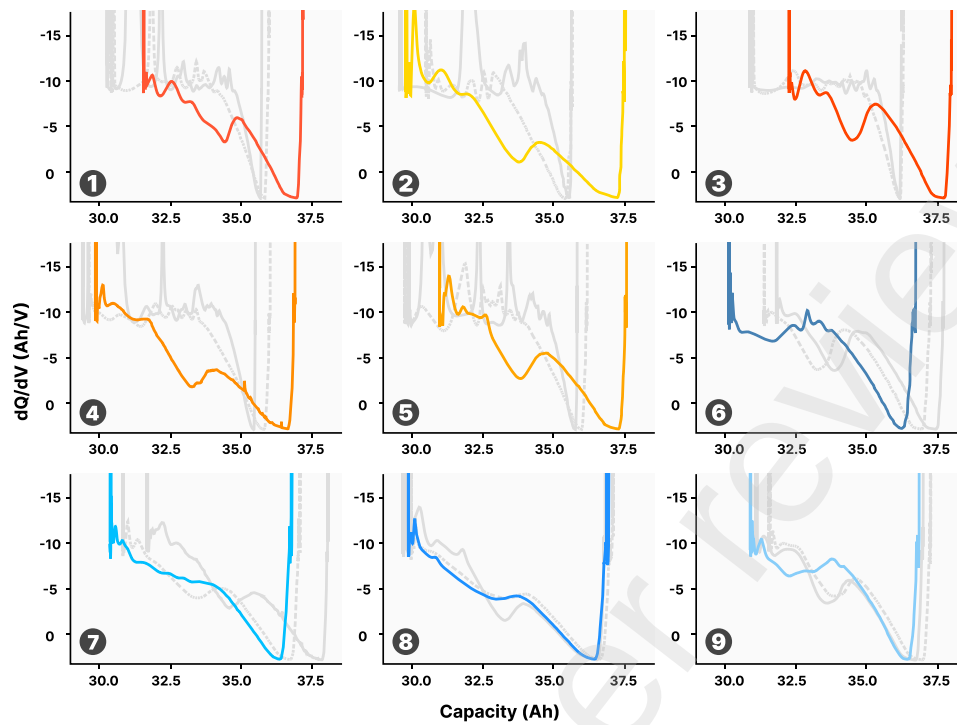
Leveraging probabilistic degradation states for early-stage lifetime prediction of lithium-ion batteries

SUPPLEMENTARY MATERIAL

SUPPLEMENTARY FIGURES



Supplementary Figure 1 Ablation study on state modeling before and after the knee point across four baselines. All experiments were conducted at 19 cells with varying cycle usage. **a** Impact of explicitly detecting the knee point. **b** Contribution of modeling degradation pre-knee state. **c** Contribution of modeling degradation post-state and regressor.



Supplementary Figure 2 dV/dQ-Q curves of Probabilistic Degradation States (PDSs). The solid gray curve is the first cycle of the fragment in the state, a thin dotted gray curve is added after 100 cycles, and the colored curve is the end cycle of the fragment. The curves show different patterns of degradation modes to varying degrees.

SUPPLEMENTARY TABLES

Supplementary Table 1. The details of prediction features [1].

Index	Definition
1	Minimum value of the difference in discharge capacity-voltage curve, between cycle x and cycle 2
2	Variance of difference in discharge capacity-voltage curve, between cycle x and cycle 2
3	Skewness of difference in discharge capacity-voltage curve, between cycle x and cycle 2
4	Kurtosis of difference in discharge capacity-voltage curve, between cycle x and cycle 2
5	Slope of the linear fit to the capacity fade curve, from cycle 2 to cycle x
6	Intercept of the linear fit to capacity fade curve, from cycle 2 to cycle x
7	Discharge capacity at cycle 2
8	Difference between the maximum discharge capacity and the discharge capacity at cycle 2
9	Average charge time from cycle 2 to cycle $x/2$
10	Integral of temperature over time, from cycle 2 to cycle x
11	Minimum internal resistance, from cycle 2 to cycle x
12	Internal resistance difference between cycle x and cycle 2

x is the early cycle, e.g. 20, 40.

Supplementary Table 2. The initial hyper-parameters in the grid search.

Regressor	Parameter	Values
Elastic Net	L1 ratio	0, 0.25, 0.5, 0.75, 1.0
Decision Tree	Max depth	None, 10, 50
Multi-layer Perceptron	Hidden layer sizes	(25,), (16,), (8,)
TabPFN	Number of estimators	4, 8, 16

Supplementary Table 3. Related works and their performance on MATR dataset.

Ref	Model	#Training cells	#Early cycle	RMSE
Yang et al. [2]	GBRT	82	100	82.80
Fei et al. [3]	SVM	87	100	115.00
Zhang and Zhao [4]	Proposed	75	100	132.00
Cao et al. [5]	Proposed	83	100	88.00
Yang and Yang [6]	Proposed	95	2	121.02
Yang and Yang [6]	Proposed	95	10	123.73
Yang and Yang [6]	Proposed	95	50	134.17
Yang and Yang [6]	Proposed	95	100	140.82
Yang and Yang [6]	LightGBM	95	2	221.31
Yang and Yang [6]	ElasticNet	95	2	224.75
Yang and Yang [6]	XGBoost	95	2	226.24
Yang et al. [7]	Full model	95	2	76.81
Yang et al. [7]	Voltage mode	95	2	88.57
Yang et al. [7]	Capacity model	95	2	119.21
Yang et al. [7]	Current model	95	2	130.08

Supplementary Table 4. Elbow-method intermediate results (Sum of Squared Errors, SSE) for pre- and post-knee data.

Number of Clusters	SSE of Pre-knee	SSE of Post-knee
1	190.08	44.72
2	73.40	20.66
3	44.74	10.48
4	27.81	6.85
5	17.86	4.86
6	13.15	3.63
7	11.21	3.12
8	9.53	2.47
9	8.05	1.82
10	6.83	1.59

Supplementary Table 5. The voltage shifting and intensity attenuation of dQ/dV-V curve in each state.

State	Initial peak voltage (V)	Initial peak intensity	Final peak voltage (V)	Final peak intensity	Voltage shifting (V)	Intensity attenuation
1	3.1486	-6.6671	3.1366	-5.4144	-0.012	1.2527
2	3.1471	-6.4996	3.1126	-4.6362	-0.0345	1.8634
3	3.1547	-6.7346	3.1411	-5.5143	-0.0135	1.2203
4	3.1456	-7.0815	3.1201	-4.9877	-0.0255	2.0938
5	3.1471	-6.7611	3.1336	-5.1351	-0.0135	1.626
6	3.1396	-5.6137	3.1036	-4.2666	-0.036	1.3471
7	3.1231	-5.0214	3.0931	-3.8282	-0.03	1.1932
8	3.1156	-4.9044	3.0871	-3.766	-0.0285	1.1385
9	3.1366	-5.3996	3.1111	-4.0466	-0.0255	1.353

Supplementary Table 6. Individual analysis for each PDS.

State	Phenomenon	Possible Mechanism
1	Minimal voltage shift (-0.0120 V), moderate intensity attenuation (+1.2527).	Surface passivation of active substances, such as the growth of SEI film, leads to an increase in charge transfer impedance, but the structural degradation is relatively mild.
2	Large voltage shift (-0.0345 V) and significant intensity attenuation (+1.8634).	Due to enhanced electrochemical polarization and loss of active substances, mass transfer or reaction kinetics are limited. This may be related to particle cracking or irreversible loss of active materials.
3	Small voltage shift (-0.0135 V), moderate intensity attenuation (+1.2203).	Aging is mainly driven by a slow increase in charge transfer impedance, which may be due to the accumulation of SEI film or side reaction products.
4	Moderate voltage shift (-0.0255 V), maximum intensity attenuation (+2.0938).	Severe loss of active substances, such as particle detachment or irreversible capacity loss. Side reactions, such as electrolyte decomposition or increased polarization, may be the main cause.
5	Small voltage shift (-0.0135 V), significant intensity attenuation (+1.6260).	Similar to State 2, it may be due to a decrease in inter-particle conductivity or gradual loss of active substances, but to a lesser extent.
6	Maximum voltage shift (-0.0360 V), moderate intensity attenuation (+1.3471).	The rapid increase in polarization may be due to an increase in SEI film thickness or electrolyte loss. Structural degradation caused by mechanical stress may also be involved.
7	Large voltage shift (-0.0300 V), small intensity attenuation (+1.1932).	Mainly manifested as slow surface passivation or uniform aging, indicating that the material may be more stable.
8	Moderate voltage shift (-0.0285 V), minimal intensity attenuation (+1.1385).	The stable charge transfer and minimal mechanical damage may be related to the good stability or structural design of the material.
9	Moderate voltage shift (-0.0255 V), with moderate intensity attenuation (+1.3530).	Stable surface passivation process, uniform growth of SEI film, and overall durability of the material.

Supplementary Table 7. The details of dQ/dV-V curve dimensionality reduction features.

Index	Definition
1	Variance value of the difference in dQ/dV-V curve, between end cycle and first cycle
2	Maximum value of the difference in dQ/dV-V curve, between end cycle and first cycle
3	Mean value of the difference in dQ/dV-V curve, between end cycle and first cycle
4	Median value of the difference in dQ/dV-V curve, between end cycle and first cycle
5	Sum value of the difference in dQ/dV-V curve, between end cycle and first cycle

SUPPLEMENTARY NOTES

Supplementary Note 1 Filter features via recursive feature elimination with cross-validation.

Input: data matrix $\mathbf{X} \in \mathbb{R}^{n \times p}$, target $\mathbf{y} \in \mathbb{R}^n$, estimator $\mathcal{E}$, desired number of features k , elimination step size s

Output: Boolean mask $\mathbf{m} \in \{0, 1\}^p$ indicating selected features, ranking vector $\mathbf{r} \in \mathbb{N}^p$

```
1 Initialize  $\mathbf{m} \leftarrow \mathbf{1}_p$ ,  $\mathbf{r} \leftarrow \mathbf{1}_p$ 
2 while  $\sum_{j=1}^p m_j > k$  do
3   remaining  $\leftarrow \{j : m_j = 1\}$ 
4   Train estimator  $\mathcal{E}$  on  $(\mathbf{X}_{\text{remaining}}, \mathbf{y})$ 
5   Obtain feature importances  $\eta \in \mathbb{R}^{\text{remaining}}$ 
6   Rank features in remaining by  $\eta$  in ascending order
7   Drop the worst  $\min(s, |\text{remaining}| - k)$  features from remaining
8   Update  $\mathbf{m}$  (set dropped features to 0) and  $\mathbf{r}$  (assign higher ranks to dropped features) accordingly
end
9 Retrain estimator  $\mathcal{E}$  on  $(\mathbf{X}_{\mathbf{m}=1}, \mathbf{y})$ 
return  $\mathbf{m}, \mathbf{r}$ 
```

Supplementary Note 2 Three datasets used in this study.

Our experiments utilize three commonly benchmarked datasets, including MATR [1], CLO [8], and Stanford [9].

- **MATR [1]** comprises 124 commercial LFP/graphite cylindrical cells (A123 APR18650M1A, 1.1 Ah, 3.3 V) cycled to failure at 30 °C on a 48-channel Arbin potentiostat. All cells were discharged at 4 C and charged with a one- or two-step fast-charging policy of the form " $C_1(Q_1)$ – C_2 " until 80 % SOC, followed by 1 C CC-CV to 3.6 V (lower cut-off 2.0 V).
- **CLO [8]** contains 45 cells (9 protocols $\times$ 5 repeats) of the same chemistry and form factor, tested under identical temperature and equipment settings. After discarding the initial cycle owing to a schedule-file error and re-launching two replacement units, every protocol retained five valid cells cycled to failure.
- **Stanford [9]** includes 222 NCM523/artificial-graphite pouch cells (nominal 0.24 Ah) cycled at 30 °C between 3.0 V and 4.2 V. A full-depth Format-2 protocol—recording only charge capacity during charging and only discharge capacity during discharging—was applied until end-of-life was reached.

Supplementary Note 3 Metrics used in this study.

Root-mean-square error (RMSE) and coefficient of determination (R^2) are chosen to evaluate framework performance. The metrics are defined as follows:

$$\text{RMSE} = \sqrt{\frac{1}{n} \sum_{i=1}^n (y_i - \hat{y}_i)^2} \quad (1)$$

$$R^2 = 1 - \frac{\sum_{i=1}^n (y_i - \hat{y}_i)^2}{\sum_{i=1}^n (y_i - \bar{y})^2} \quad (2)$$

where y_i is the observed cycle life, $\hat{y}_i$ is the predicted cycle life, $\bar{y}$ is the average of the observed cycle life and n is the total number of samples.

Supplementary Note 4 Quantitative analysis for dQ/dV-V curve of each PDS.

According to Supplementary Table 5, there are voltage shifting, and peak intensity attenuation in each PDS.

- Voltage shifting: The peak voltage of each state exhibits a phenomenon of rightward shift (high voltage direction). The most significant voltage shift is observed in State 6 and State 2 (-0.0360 V and -0.0345 V, respectively), indicating that material aging in these clusters leads to more pronounced changes in electrochemical properties.
- Peak intensity attenuation: The peak intensities (negative dQ/dV values) of all states decreased, indicating a decrease in electrochemical activity during the aging process. State 4 has the highest intensity attenuation (+2.0938), while State 8 has the lowest attenuation (+1.1385), indicating significant differences in aging rates among different clusters.
- Durability comparison: State 7 and State 8 have smaller aging amplitudes and may have better electrochemical stability.

REFERENCES

1. K. A. Severson, P. M. Attia, N. Jin, *et al.*, "Data-driven prediction of battery cycle life before capacity degradation," *Nat. Energy* **4**, 383–391 (2019).
2. F. Yang, D. Wang, F. Xu, *et al.*, "Lifespan prediction of lithium-ion batteries based on various extracted features and gradient boosting regression tree model," *J. Power Sources* **476**, 228654–228663 (2020).
3. Z. Fei, F. Yang, K.-L. Tsui, *et al.*, "Early prediction of battery lifetime via a machine learning based framework," *Energy* **225**, 120205–120218 (2021).
4. Y. Zhang and M. Zhao, "Cloud-based in-situ battery life prediction and classification using machine learning," *Energy Storage Mater.* **57**, 346–359 (2023).
5. G. Cao, Y. Jia, S. Chen, *et al.*, "Lithium-ion battery future degradation trajectory early description amid data-driven end-of-life point and knee point co-prediction," *J. Clean. Prod.* **477**, 143900–143911 (2024).
6. W. Yang and H. Yang, "Ultra-early prediction of lithium-ion battery cycle life based on assembled capacity curve extracted from a single cycle," *J. Power Sources* **640**, 236620–236631 (2025).
7. W. Yang, F. Min, J. Xie, and H. Yang, "Ultra-early prediction of lithium-ion battery cycle life based on visualized single-cycle data," *IEEE Transactions on Power Electron.* **40**, 7342–7353 (2025).
8. P. M. Attia, A. Grover, N. Jin, *et al.*, "Closed-loop optimization of fast-charging protocols for batteries with machine learning," *Nature* **578**, 397–402 (2020).
9. X. Cui, S. D. Kang, S. Wang, *et al.*, "Data-driven analysis of battery formation reveals the role of electrode utilization in extending cycle life," *Joule* **8**, 3072–3087 (2024).