Machine Learning-Based Optimization of SOC Windows for High-Performance μ-Si Anodes

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1. Introduction

High energy density and extreme fast charging (XFC) are essential for electric vehicles, necessitating the use of silicon (Si) anodes.[1][2] However, the continuous loss of lithium inventory from severe volume expansion remains a critical bottleneck.[1] While Anode-Tailored Full-cell Design (ATFD) provides a strategy to mitigate this by restricting the operating SOC, the interdependent nature of pre-lithiation levels and SOC limits creates a complex, multi-dimensional design space.[3] Conventional trial-and-error approaches are insufficient to navigate these non-linear correlations efficiently. In this work, we employ machine learning (ML) to precisely identify the global optimum for both pre-lithiation degree and SOC windows, establishing a framework that maximizes XFC performance while suppressing electrode degradation.

2. Electrochemical Phase Transitions and Degradation in micro-silicon (m-Si) Anodes

Lithiation of micro-silicon (m-Si) involves a phase transition from amorphous Li_xSi (K_2) to crystalline c-$Li_{15}Si_4$ (K_3) at low potentials. While the K_2 phase enables rapid lithium diffusion, K_3 crystallization triggers severe localized stress and particle pulverization, leading to irreversible lithium inventory depletion. The Anode-Tailored Full-cell Design (ATFD) strategy mitigates these issues by confining operation to the K_2 regime to maintain structural integrity. However, because phase stability is non-linearly coupled with pre-lithiation levels, precise calibration of the starting SOC and window width is essential to optimize the trade-off between energy density and cycle life.

3. Methodology and Optimization Framework

3.1 Machine Learning Framework for Precision Pre-lithiation and SOC Mapping

To resolve the inherent volume expansion issues in micro-silicon (*m*-Si) anodes, this study employs a data-driven closed-loop optimization methodology. We utilize Bayesian optimization (BO) with Gaussian process regression (GPR) as a surrogate model to optimize the pre-lithiation level (Q_{pre}) and operating SOC window (ΔSOC), which collectively dictate the kinetic state of the electrode. The input features, specifically voltage relaxation ΔV_{relax}) and overpotential extracted from galvanostatic intermittent titration technique (GITT) measurements, are directly correlated with the lithium diffusion coefficient (D_{Li}) and serve as a physical basis for predicting internal phase transition behavior. To ensure robust predictive performance despite the limited *m*-Si experimental data, we incorporate transfer learning by leveraging degradation patterns learned from graphite anode datasets, thereby significantly accelerating model convergence.

3.2 Transitioning from Discrete SOC Sampling to ML-Driven Continuous Parameter Discovery

The primary contribution of this work lies in transitioning from the passive exploration of seven discrete SOC intervals (R1–R7), as reported in previous literature (Lee et al., *PNAS* 2025, 122, 1, e2417053121), to intelligent optimization within a continuous parameter space. While conventional approaches were limited to searching for optima within a fixed 40% window width, the proposed ML framework dynamically identifies the optimal starting SOC and window width through an acquisition function.

Specifically, the model is designed to maximize the K_2 phase ($Li_xSi \rightarrow Li_{x+y}Si$), characterized by rapid lithium diffusion, while actively avoiding the K_3 crystallization stage (c-$Li_{15}Si_4$) that triggers mechanical failure. This approach numerically resolves the inherent trade-off between energy density and cycle life in Si-anode design. Furthermore, by reducing experimental trial-and-error by more than 90%, this framework demonstrates the viability of autonomous research in battery materials development.

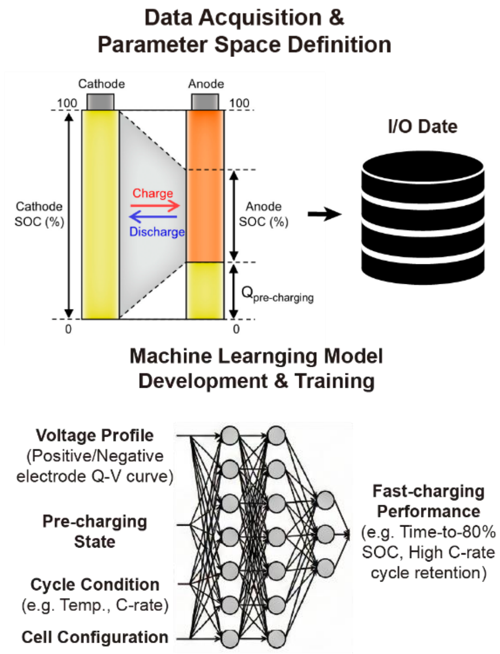

Fig. 1 : Overall workflow

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