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## Förord

This project has been funded by the Swedish Energy Agency through the Vehicle Strategic Research and Innovation Program. The first period was funded by Grant No. 50187-1, and the two-month extension from July to September in 2023 was funded by Grant No. P2019-90083. In addition to the project participants mentioned above, Shafiq Urréhman (Main architect of AI technologies and lead strategic expert) and Lars Nilsson (Head of vehicle integration) at CEVT AB contributed to this project through discussions and providing data and experiment resources. Erik Preihs (Module team director at CEVT) and Sven Sjöberg (former head of battery management and high voltage systems at CEVT) contributed to the conception and planning during the grant application and were involved in the recruitment, management, and discussion in about the first 10 months of this project. Chengqi She (a visiting PhD student at Chalmers) and Xiaopeng Tang (a postdoc at Hong Kong University of Science and Technology) were working on parallel research projects and have partly contributed to this project without any funding from the Swedish Energy Agency.

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## Sammanfattning

Noggrann förutsägelse av batteriets åldringsbana och återstående livslängd krävs inte bara för att säkerställa säker och tillförlitlig drift av elfordon utan är också det grundläggande steget mot hälsomedveten användning, effektivt och tidigt underhåll och bedömning av batteriets restvärde. Icke-linjäritet, ett brett spektrum av driftsförhållanden och cell-till-cell-variationer gör dock förutsägelse av batteriets hälsa till en mycket utmanande uppgift.

Teamet för batterihanteringssystem vid Chalmers tekniska högskola och gruppen för högspänningsbatterier vid CEVT har gått samman för att ta itu med problemet i det här projektet. Vi har särskilt utnyttjat maskininlärning och big data för att utveckla exakta, tillförlitliga och praktiska metoder för att diagnostisera hälsotillståndet, prognostisera den framtida åldringsbanan och förutsäga den återstående livslängden för litiumjonbatterier. Den förbättrade uppskattningen och förutsägelsen av batteriets åldrande kan leda till betydande fördelar och kan användas direkt av batteritillverkare, BMS-programvaruleverantörer, elbilsföretag, bilförsäkringsleverantörer samt lagstiftare och beslutsfattare. Dessutom föreslog vi en maskininlärningsbaserad livslång uppskattningsmetod för litiumpläteringspotential och använde den för hälsomedveten snabb batteriladdning. Genom omfattande simuleringar och skräddarsydda tester av myntceller i labbskala har vi visat att vi kan uppnå både snabbare laddning och en 100 % längre batterilivslängd. Om detta framsteg implementeras kommersiellt kan det avsevärt öka bekvämligheten och acceptansen för elbilar och därmed påskynda övergången till ett hållbart transportsystem.

Nästa fas innebär att använda de utvecklade modellerna för åldringsprognoser för optimerad batterilivslängd och utöka snabbladdningsresultaten från simulering och skräddarsydda myntceller i labbskala till verkliga kommersiella batterier.

#### Summary

Accurately predicting battery ageing trajectory and remaining useful life is not only required to ensure safe and reliable operation of electric vehicles (EVs) but is also the fundamental step towards health-conscious use, effective and timely maintenance, residual value assessment of the battery. However, the non-linearity, wide range of operating conditions, and cell to cell variations make battery health prediction a very challenging task.

The battery management system (BMS) team at Chalmers University of Technology and the High Voltage Battery (HVB) group in CEVT joined the effort to tackle the problem in this project. Specifically, we have leveraged machine learning and big data to develop accurate, reliable, and practical methods for diagnosing the state of health, prognosing the future aging trajectory, and predicting the remaining useful life of lithium-ion batteries. The improved estimation and prediction of battery aging can result in significant benefits and can be used directly by battery manufacturers, BMS software providers, EV companies, automotive insurance providers, as well as regulatory and policy makers. Furthermore, we proposed a machine learning-based lifelong estimation method for lithium plating potential and used it for health-aware fast battery charging. Through extensive simulations and tailored lab-scale coin cell tests, we have demonstrated the capability to achieve both faster charging and a 100% longer battery lifetime. If commercially implemented, this advancement is poised to significantly enhance the convenience and acceptance of EVs, thereby expediting the transition to a sustainable transport system.

The next phase involves employing the developed ageing prediction models for optimised battery lifetime and expanding the fast charging results from simulation and tailored lab-scale coin cells to real-world commercial batteries.

## Inledning/Bakgrund

Within the transportation sector, the anticipated battery needs and costs for the transition to partial or full electrification of the vehicle fleet are substantial. This is exemplified by Volkswagen's announcement of securing €20 billion worth of battery supplies by 2025. According to a 2019 Statista study, the global market demand for lithium (Li)-ion batteries used in EVs is expected to surge over 20 times, from 74 GWh in 2017 to over 1,500 GWh in 2030. This growth presents a significant opportunity for the battery market, but uncertainties persist in optimizing battery usage for long lifetimes without compromising performance.

Battery degradation begins upon production and can accelerate if not managed appropriately. Extensive research has focused on determining the State of Health (SoH) of batteries to enable health-conscious usage and early detection of aging and potential thermal issues [1]–[4]. SoH is often quantified by capacity fade and internal resistance increase, affecting energy and power density. Current methods treat SoH observation as an online parameter identification problem, employing equivalent circuit models or data-driven models [5].

Equivalent circuit models, though conceptually simple, face challenges as many parameters vary with temperature, State of Charge (SoC), and SoH. At Chalmers, effective estimation tools have previously been developed under specific conditions, including multi-timescale SoC and SoH estimators [6]. Data-driven health models, using machine learning (ML) algorithms like random forest regression [7] and support vector machines [8], have shown promising results in accuracy and computation.

The overarching challenge is to develop robust algorithms for SoH estimation and Remaining Useful Life (RUL) prediction. RUL prediction is crucial for cell selection, grouping, and cost-efficient aftermarket processes, guaranteeing the



lifespan of vehicle battery systems, and optimizing battery control for extended life [9], [10].

The project addresses the critical need to extend the lifespan of vehicle battery systems amid the increasing shift towards electrification. Achieving a fossil-fuel-free future depends on advancements in battery technology, particularly for lithium-ion batteries, recognized as the leading commercial energy storage source for vehicles. Despite improvements in quality and decreased prices, lithium-ion batteries remain a significant cost and environmental concern. Prolonging battery lifespan is vital for resource efficiency, environmental sustainability, and vehicle economy and reliability.

The project tackles these challenges through the development of ML algorithms for SoH estimation and RUL prediction. It is funded by Swedish Energy Agency, enabling research and development in this critical area. The timeline for the project spans from August 1<sup>st</sup>, 2020 to September 30<sup>th</sup>, 2023. The project is an industrial PhD project and mainly to fund PhD student Yizhou Zhang for his PhD study. He started to work on this project since September 2020.

## Genomförande

The initial phase of the PhD student's research involved an exhaustive literature review to gain a comprehensive understanding of the state of the art in battery ageing diagnostics and prognostics methods. Subsequently, an in-depth exploration of available battery datasets was undertaken, encompassing both laboratory cycling data and commercial vehicle fleet data. The laboratory cycling dataset, characterized by meticulously recorded reference performance tests conducted in a controlled environment, served as an ideal testbed for validating the algorithms developed in this project. Conversely, the customer fleet dataset was instrumental in representing real-world scenarios, allowing for the verification of the practicality and efficacy of the proposed methods throughout the project. To streamline the utilization of these datasets, a systematic data processing pipeline was implemented, ensuring the methodical filtration and storage of relevant data for subsequent algorithm development.

#### Online battery aging trajectory prediction using histogram data

Precisely forecasting the aging trajectory and remaining useful life of batteries is crucial, not only for ensuring the safe and reliable operation of electric vehicles (EVs) but also as a fundamental step towards health-conscious battery usage and the assessment of residual value. The inherent non-linearity, diverse operating conditions, and variations between individual cells present formidable challenges in accurately predicting battery health. These challenges are particularly pronounced when dealing with batteries operating under real-world conditions. A method that systematically addresses these complexities and is applicable to the operational data of batteries in the field is of great importance.

The paramount significance lies in the application of data-driven methods to process the dataset and formulate pertinent and efficient input features. Figure 1

provides a visual representation of the three raw datasets employed in this study. Subfigures (a)-(c) depict the laboratory cycling data from over 100 lithium iron phosphate (LFP) batteries, while (d)-(f) showcase the laboratory cycling data from approximately 20 Lithium cobalt oxide batteries. Additionally, (g)-(l) represent datasets derived from real-world vehicle fleets, encompassing data from over 7000 in-service plug-in hybrid vehicles.

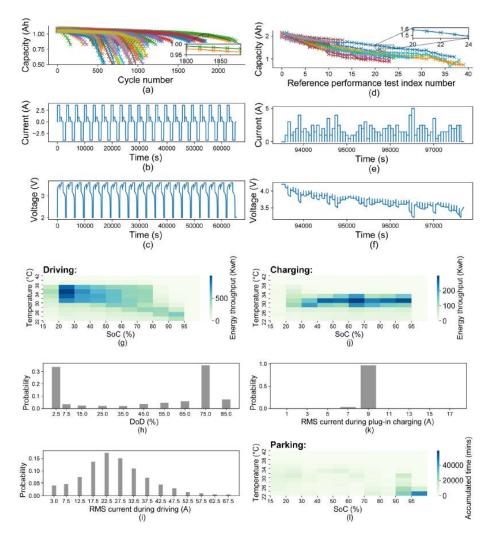


Figure 1. Illustration of three battery datasets used for algorithm development, validation, and tests.

While the battery aging process is intricate, the stress factors contributing to capacity fade remain consistent [11]. According to [12]–[16], several factors significantly influence battery capacity degradation, including Depth of Discharge (DoD), charge and discharge current rates, temperature, voltage, accumulated cycling/calendar time, accumulated ampere-hour (Ah) throughput, and SoC. These widely acknowledged stress factors are utilized in a two-step process to construct an initial feature pool.

In the first step, raw data, presented in either time series or histograms of various dimensions, undergoes transformation into 1D histograms. Figure 2(a)–(c) illustrates this transformation process and its outcomes for time series laboratory

data from the NASA dataset, utilizing a current interval of 0.5 A. Similarly, (f)–(h) depict the transformation process and results for a 2D histogram derived from the fleet dataset. The second step involves the extraction and calculation of statistical properties from the constructed 1D histograms produced in the first step. Figure 2(d), (e), (i), and (j) showcase a selection of these statistical properties and their corresponding calculated values. This comprehensive approach ensures that the essential stress factors influencing battery capacity degradation are captured and integrated into the subsequent analysis.

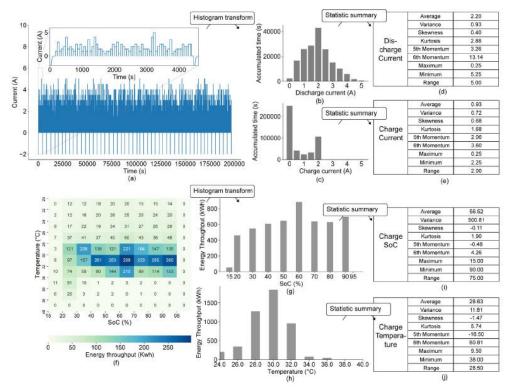


Figure 2. Illustration of the feature engineering process.

The battery aging prognosis task is cast as a regression problem within the framework of supervised ML. The comprehensive pipeline for executing this task encompassing both an offline path for global model development and an online path for model adaptation, incorporating streaming data.

The global models are exclusively developed from the offline training dataset, comprising multiple battery cells of the same type. Each model strives to comprehend the averaged aging behavior based on the selected features from these cells. The model development process involves hyperparameter tuning, method selection, model evaluation, and online deployment. Among the diverse set of ML methods available for nonlinear model regression, namely Support Vector Regression (SVR), Random Forest Regression (RFR), Gaussian Process Regression (GPR), and Artificial Neural Network (ANN).

When forecasting capacity changes using a global model, only the future model inputs (features) of the specific cell in question are utilized. The resulting prediction is essentially an open-loop model-based simulation. To clarify, the predictor is unaware of any cell-specific aging behavior, even in cases where a cell deviates significantly from others. The historical capacity profile of a cell can potentially contain valuable information for understanding its future aging characteristics. With this insight, we aim to develop an individualized prediction model for each battery cell by directly adjusting the outcomes of the global model.

The adjustment factor, which varies over time, is determined online based on the disparity between the historically predicted output trajectory by the global model and the measured trajectory from the cell under consideration. This adaptive approach allows the prediction model to dynamically account for and respond to the unique aging patterns exhibited by each battery cell, offering a more personalized and accurate prognosis over time. The overall battery aging trajectory prediction using histogram data is illustrated in Figure 3.

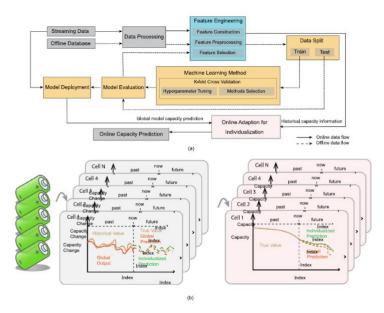


Figure 3. Pipeline to develop data-driven algorithms for battery ageing prognosis. (a) summarises all the required modules and their connections. (b) zooms in on the online adaptation module, where both the global model-based predictions and the individual cell's historical information are utilized.

#### Battery state of health (SoH) estimation under arbitrary usage conditions

Accurately estimating the State of Health (SoH) of batteries is essential for ensuring the safety, reliability, and optimal energy and power management of electric vehicles. However, from a data-driven perspective, challenges arise due to dynamic vehicle operating conditions, stochastic user behaviors, and cell-to-cell variations, making the estimation task inherently challenging. Our goal is to develop a method that can be applied under arbitrary customer usage conditions while maintaining high accuracy and robustness.

To achieve this, the dataset employed for model development and validation aims to closely represent real-world battery usage scenarios. The dataset utilized in this study was obtained from Sandia National Laboratories (SNL) [17]. The experiments conducted initially aimed to investigate the impact of various stress factors, such as discharge rate, depth of discharge, and environmental temperature,

on the degradation performance of commercial cells with different chemistry types. Specifically, cells with Nickel Manganese Cobalt (NMC) and Nickel Cobalt Aluminum (NCA) as the positive electrode were selected for analysis. Figure 4(a) and (b) depict the capacity retention trends of the NMC and NCA battery cells, respectively. During the reference performance tests, where all cells were charged under 0.5C, we accumulated the capacity during the charging phase and utilized the calculated result as the ground truth for battery capacity. The objective of this work is to estimate this capacity value in real-time, concurrent with battery usage. Figure 4(c), (d), and (e) provide examples of the current, voltage, and temperature curves, respectively, during a typical charge and discharge cycle.

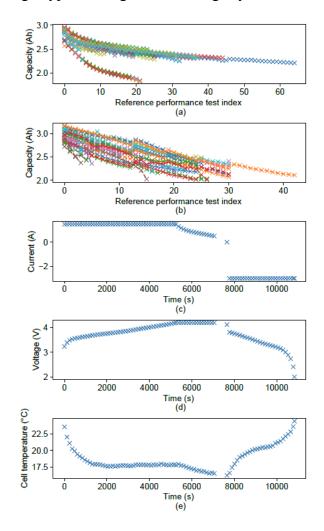
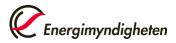


Figure 4. Illustration of the battery dataset used in the work.

Practical battery usage is dynamic and subject to frequent changes in operating conditions, especially in shared EVs. Temperature variations also impact battery performance. Unlike laboratory cycling, where conditions are controlled, real-world usage is unpredictable. Charging profiles are more controllable than discharging profiles. This study focuses on charging curves to estimate capacity, eliminating assumptions from lab tests. It considers various charging scenarios,



categorizing them into six distinct cases (S1–S6), including both CC-CV charging and other strategies. The charging scenarios are categorized as follows:

S1: Complete CC-CV charging from 0% to 100% State of Charge (SoC).

S2: Partial CC-CV charging starting before the Inflection Point (IC) peak value, ending with the complete CV phase.

S3: Partial CC charging starting before the IC peak value, ending without the CV phase.

S4: Partial CC-CV charging starting after the IC peak value, ending with the complete CV phase.

S5: CC charging starting after the IC peak value, ending without the CV phase. S6: All other scenarios not covered in S1–S5.

Two sets of features are chosen from the measured charging signals: one derived from a specific voltage window and the other from the incremental capacity curve. The evolution of the selected features over the battery's lifetime is illustrated in Figure 5.

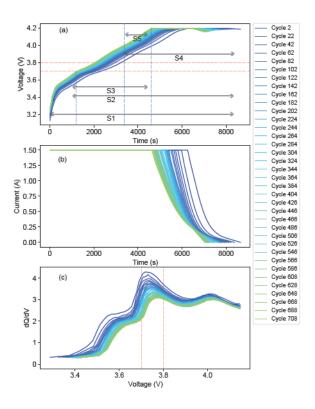


Figure 5. Illustration of the varying health indicator as the battery gradually aged. (a) shows the change of the voltage curve along with an illustration of different charging scenarios. (b) represents the change in the current curve. (c) shows the change in the IC curve.

Given that the estimation result may influence decisions, corrective actions, or proceed to the prognostic step, it is crucial to evaluate estimation uncertainty. Four ML algorithms, two probabilistic (GPR and Bayesian ridge regression) and two frequentist-based (RFR and ANN), are chosen to develop models for capacity estimation. All these algorithms can quantitatively propagate their estimation uncertainty, providing a confidence interval for their results. Henceforth, the ML algorithm output, denoted as battery capacity (Q), will be referred to as y, and the corresponding features as x. Random-search hyperparameter tuning, along with 5-fold cross-validation, is applied to identify the optimal hyperparameters for each ML algorithm. Due to different characteristics, the optimal algorithm may differ based on the datasets and the operating conditions of the battery. To obtain a more accurate and reliable estimation of battery capacity, we employ a Kalman Filter (KF) to fuse the results of all algorithms. The overall battery SoH estimation pipeline is illustrated in Figure 6.

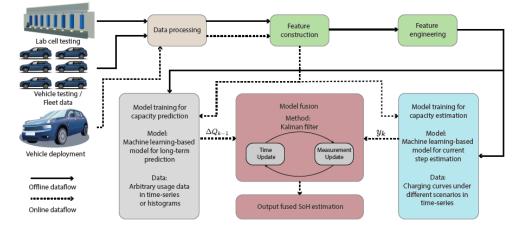


Figure 6. The overall battery SoH estimation pipeline.

# Early prediction of battery lifetime using both time-series and histogram usage data

Forecasting the lifespan of a battery proves challenging due to the inherent complexity of the battery aging process and external factors such as manufacturing variations and diverse usage patterns. The intricacies are heightened when attempting early predictions, given the non-linear nature of aging and the subtlety of capacity retention issues. Nevertheless, the significance of accurate lifetime prediction is immense, offering the potential to minimize lengthy and exhaustive testing periods, strategically plan for predictive maintenance, and substantially enhance overall battery efficiency. The majority of current research emphasizes the utilization of measured time-series data for predicting battery lifespan. However, it is crucial to recognize that the operating conditions of the battery significantly influence its longevity. To my knowledge, there is a notable absence of studies incorporating such usage information for early forecasting of battery lifespan and exploring its correlation with methodologies relying on time-series measurements.

In this study, Stanford dataset (introduced in the first part of the work) and SNL (introduced in the second part of the work) dataset are used to verify the proposed algorithm.

Battery lifespan is significantly influenced by usage patterns. Combining historical usage data and predicted future patterns improves prediction accuracy. Stress factors, like depth of discharge, charge/discharge rates, and temperatures, are crucial [11]. In the Stanford dataset, features are based on unique charging policies. Statistical properties of charge current intervals are calculated to reduce

Fixed charge/discharge policies cause gradual changes in current and voltage curves as batteries degrade. Intentional feature construction from time-series measurements aids battery lifespan prediction. Investigating dQ(V) and incremental capacity (IC) curves, which reflect hidden aging mechanisms, proves useful. We adopt dQ(V) and IC curves during discharge as baseline features for the Stanford dataset, and for the SNL dataset, we use the charge part of the profile as the baseline features. The overall battery early life prediction pipeline is illustrated in Figure 7.

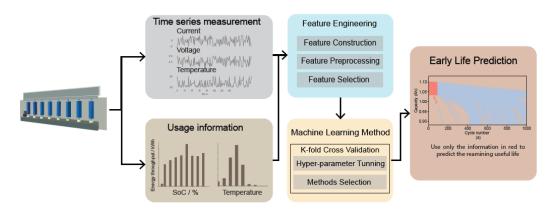


Figure 7. The overall battery early life prediction pipeline.

The early prediction of battery lifespan is approached as a regression problem, aiming to minimize the disparity between measured lifetime and model predictions across all test set batteries. Four ML algorithms are examined for their performance in this context. Two linear models, elastic net (EN) and Bayesian ridge regression (BRR), are considered, alongside two nonlinear models—support vector regression (SVR) and random forest regression (RFR).

#### Lifelong estimation of lithium plating potential

Navigating the trade-off between fast charging and prolonged battery lifespan poses a persistent challenge. We first experimentally dive into resolving this dilemma by showcasing the effectiveness of employing a controlled anode potential fast charging strategy. This strategy is designed to concurrently achieve rapid charging and an extended battery lifetime. To further enhance our approach, we introduce a battery aging mode quantification algorithm. This algorithm allows us to estimate critical parameters such as capacity, loss of lithium inventory (LLI), and loss of active materials (LAM) in the electrodes. Remarkably, these estimations are derived solely from practical and accessible partial slow charging voltage curves. Subsequently, we propose a framework for estimating the lifelong battery anode potential. This framework leverages both the quantified aging mode and real-time measurements of current, voltage, and temperature. The ultimate goal is to facilitate health-conscious fast charging for batteries. Our comprehensive framework undergoes training and testing using an extensive synthetic dataset covering over 1,500 aging trajectories. The results demonstrate its robustness and highlight its potential for implementation in Battery Management Systems (BMS). Noteworthy, the framework exhibits high accuracy even under realistic measurement uncertainties and biased aging mode estimates, emphasizing its suitability for practical applications. The detailed framework is illustrated in Figure 8.

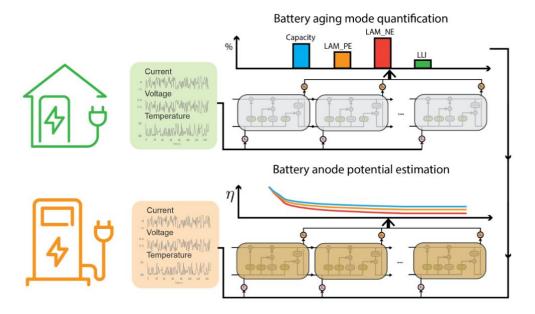


Figure 8. The schematic of workflow for lifelong battery anode potential estimation.

## Resulted

#### Online battery aging trajectory prediction using histogram data

The global model aging trajectory prediction results using the Stanford dataset is shown in Figure 9 and the numerical results is shown in Table 1. The mean absolute percentage error (MAPE) of less than 1.7% and a root mean squared percentage error (RMSPE) of less than 3.3% is achieved with the best performed global model. Additionally, the majority of predicted capacity values for the 40 LFP cells in the test set fall within  $\pm 5\%$  error bounds, as depicted in Figure 9(a)–(e). These findings affirm the reliability of the proposed global models in predicting the lifelong capacity profile for unseen battery cells. Both the constructed histogram-based features and the proposed feature engineering method prove effective across various ML methods, including SVR, RFR, GPR, and ANN. Notably, RFR and ANN outperform alternatives, achieving MAPE errors of 0.93% and 1.13%, respectively. The small prediction errors and robust performance across the entire capacity range position the proposed models competitively against prevalent models developed directly from time series data.

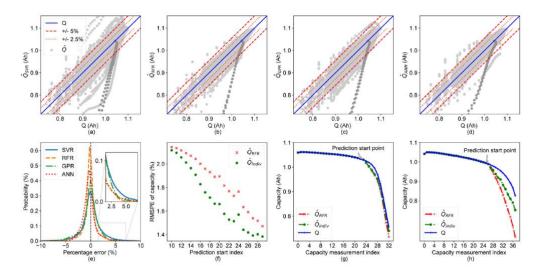


Figure 9. Validation of the developed global models and online adaptation algorithm using the Stanford dataset. (a)–(d) show the predicted capacity by SVR, RFR, GPR, and ANN, respectively, versus the measured capacity Q. One specific cell, 'b2c47', shows an abnormal long lifetime compared to others with similar cycling conditions and is highlighted in dark grey. (e) presents the percentage error histogram of the four ML methods to predict the capacity trajectory, in which the predictors stand at the first data sample. (f) shows root mean squared percentage error using the RFR-based global model and the individualised model. (g) represents the prediction results for a randomly selected cell, 'b4c36', and (h) for the abnormal cell 'b2c47'.

Table 1. Prediction error comparisons of four different ML algorithms on three distinct datasets.

methods.						
Algorithms	orithms MAPE (%)		RMSPE (%)			
	Stanford	NASA	Vehicle fleet	Stanford	NASA	Vehicle fleet
SVR	1.61	4.41	1.46	3.22	6.33	2.18
RFR	0.93	4.21	1.43	2.12	5.76	2.16
GPR	1.35	3.23	1.44	2.58	4.45	2.11
ANN	1.13	3.64	1.41	1.92	5.08	2.12

MAPE and RMSPE of the predicted capacity change by four machine learning methods.

The prediction performance of individualized models, using RFR as an example, is illustrated in Figure 9(f). In the best case, the individualized model reduces the prediction error by 13.7%, with an average improvement of 8.6%. These enhancements are noteworthy for battery aging and lifespan prognosis, particularly in applications related to health optimization and extending the lifespan of numerous battery cells. Beyond handling all testing cells in a batch, we explore the performance of online adaptation for individual cells. As shown in Figure 9(f)–(h), for both the abnormal cell 'b2c47' and the normal cell 'b2c35', the individualized model effectively learns from historical aging information, continuously adjusting global predictions along aging trajectories to approach the ground truth. This results in more accurate and robust predictions.

After demonstrating effectiveness on the Stanford dataset, we extend the evaluation of the designed algorithms to battery cells within the NASA and vehicle fleet datasets. The calibrated capacity profiles in the NASA dataset occasionally exhibit local peaks, potentially resulting from drifted measurements. In the fleet data, capacity measurements are generated by onboard ECUs, with unknown accuracy. In such cases, we treat all measurements as ground truth, acknowledging this might slightly impact numerical results—a common consideration in real-world battery data studies. Results for these datasets are presented in Table 1 and Figure 10.

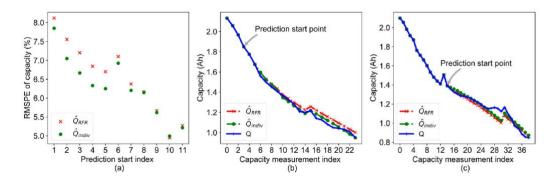


Figure 10. Comparison of the RFR-based global model and the individual model for predicting battery capacity of the NASA dataset.

In the fleet dataset, global models based on various ML methods exhibit nearly identical prediction errors, approximately 1.45% in MAPE and 2.15% in RMSPE. Despite slightly lower performance in the NASA dataset, with a MAPE of 3.23%, this error level remains suitable for many industrial applications that often require errors to be below 5%. These findings underscore the effectiveness and practicality of histogram-based models for battery aging prognosis.

The online adaptation algorithm is tested on the NASA dataset, confirming its effectiveness in improving model accuracy and robustness in the presence of cell variations. As illustrated in Figure 10, the individualized model consistently reduces prediction errors at nearly every prediction start index, achieving a maximum reduction of 7.5% in RMSPE. Similar to the results in Figure 9(g)–(h), the individualized model closely tracks the measured capacity profile compared to the global model. Importantly, this holds true regardless of whether the global model overpredicts or underpredicts measured values, and the performance is sustained even when battery degradation exceeds 50% of the nominal capacity.

#### Battery state of health (SoH) estimation under arbitrary usage conditions

The estimation errors of various SoH estimation algorithms are quantitatively studied for NMC-type batteries, with the results detailed in Table 2. Across scenarios S1 to S5, all the ML models derived, including the Kalman Filter (KF), achieve reasonable estimates, with a MAPE ranging from 0.629% for the best case to 2.27% for the worst case. In the uncommon scenario S6, where batteries operate under a full lifespan, the proposed model fusion method estimates the capacity trajectory at a MAPE of 3.899%.

Algorithms	5	S1 S2		\$3		S4		S5		S6		
Algorithms	MAPE	RMSPE										
GPR	0.693	1.073	0.887	1.323	0.915	1.491	1.532	1.999	2.110	2.757	-	-
BRR	0.772	1.182	0.799	1.009	1.025	1.527	1.984	2.598	2.270	2.910	-	-
RFR	0.632	0.857	0.874	1.202	0.877	1.231	1.817	2.312	1.725	2.262	-	-
DeNN	0.919	1.418	0.816	1.114	1.027	1.471	1.576	2.132	1.860	2.334	-	-
KF	0.629	0.861	0.714	0.880	0.751	1.178	1.662	2.110	1.731	2.229	3.899	5.611

Table 2. Results of different SoH estimation algorithms for NMC batteries under Scenarios 1-6.

Results in Table 2 also confirm the superiority of the proposed KF-based fusion method, generally outperforming or matching the best-performing individual model. Figure 11 provides a closer look, showing that under the first five scenarios, the KF rarely exceeds  $\pm 5\%$  error, with results within  $\pm 2.5\%$  error for scenarios S1–2. Figure 12 demonstrates that under S1, KF follows the measured capacity better than the best individual ML model, namely RFR.

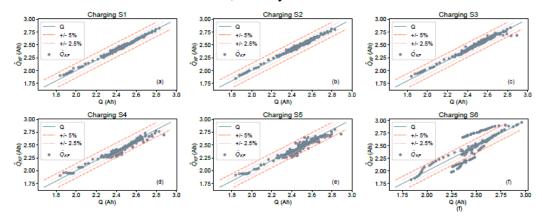


Figure 11. The estimation results for the NMC-type battery under different charging scenarios.

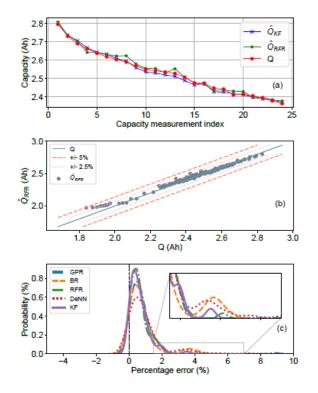


Figure 12. Estimation results of a randomly selected NMC-type cell under Scenario 1.



All developed models provide a 95th percentile estimation confidence interval valuable for predictive battery maintenance and usage optimization. Fusion of estimation results using KF significantly tightens the confidence interval, reducing uncertainty (Figure 13). After the seventh index, the standard deviations of the best and worst individual models are 2 and 5 times larger than one KF has, respectively.

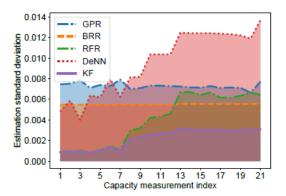


Figure 13. The standard deviation of the results from the individual ML model and the model fusion for a randomly selected cell.

In practical scenarios, it's uncommon for a battery to undergo only one charging profile throughout its entire lifespan. Hence, it's valuable to explore estimation performance when various charging profiles are applied to a specific battery. As no cell in the database underwent multiple charging profiles, cells experiencing full CC-CV charging were selected, and their profiles were manually truncated to mimic partial charging. While these results may not precisely reflect real-world usage, the focus here is on qualitative outcomes. Charging profiles for each cell are assumed to rotate periodically among six scenarios during its lifetime, with rotating protocols and results detailed in Table 3. Features are extracted based on each scenario's availability. It can be seen that, under S6, where no time-series feature is available, individual estimation models cannot estimate the capacity value. In such cases, when S6 is activated, the capacity remains unchanged from the previous time-step estimate. Table 3 illustrates that within each protocol, the KF consistently outperforms any individual ML model in providing better estimation results. When S6 is activated more frequently, the estimation results of the individual models generally become worse. On the contrary, the KF is still very reliable and continuously follows the ground truth at around 1 % RMSPE.



Algorithms	Protocol 1: Periodically	repeat $\{S1, \dots, S5, S6\}$			
	MAPE	RMSPE			
GPR	1.337	2.408			
BR	1.373	1.847			
RFR	1.314	1.706			
DeNN	1.092	1.58			
KF	0.631	0.813			
	Protocol 2: Pe	riodically repeat			
		$S5, S6, S6\}$			
GPR	1.486	2.278			
BR	1.723	2.245			
RFR	1.710	2.224			
DeNN	1.327	1.820			
KF	0.648	0.914			
	Protocol 3: Periodically repeat				
	$\{S1, \cdots, S5, S6, S6, S6\}$				
GPR	1.678	2.598			
BR	1.843	2.366			
RFR	1.810	2.297			
DeNN	1.310	1.806			
KF	0.667	0.846			
	Protocol 4: Periodically repeat				
	$\{S1, \cdots, S5, S6, S6, S6, S6, S6\}$				
GPR	2.043	2.999			
BR	2.271	3.045			
RFR	2.199	2.820			
DeNN	1.706	2.387			
KF	0.797	1.031			

Table 3. SoH estimation results under various practical charging scenarios.

## Early prediction of battery lifetime using both time-series and histogram usage data

Initially, only cycling information from the first 100 cycles is used for prediction. Subsequently, we change the number of early life cycles to examine its sensitivity to prediction results. To compare prediction performance using different feature sources, we apply the same machine learning algorithm, RFR in this case, trained with different feature inputs. Results are summarized in Table 4.

Table 4. Results of different feature inputs and two combination methods for early prediction of battery lifetime using Stanford dataset.

Feature input	$R^2$	RMSE (cycles)	MAPE (%)
Time-series	0.78	197	15.41
Histogram	0.85	162	14.31
Combined features	0.87	149	10.51
Combined models	0.87	151	10.11

Observing the table, it's apparent that the prediction performance using either timeseries features or usage-related histogram features alone is similar in terms of MAPE, with the histogram feature-based algorithm slightly outperforming the time-series features. This suggests that the two feature sources are effectively interchangeable for battery life prediction. However, when both feature sources are combined, a significant performance improvement is achieved, indicating that the two feature sources are complementary and should be used together when possible. Examining detailed prediction results in Figure 14, the model trained with combined features tends to violate the  $\pm 100$  cycles prediction boundary of the measured lifetime less than when using individual feature sources. Zoom-in figures display the error histogram of prediction results using different feature sources, again showing the superiority of combining both feature sources, including less extreme predictions and a narrower error distribution. This superiority arises from the fact that usage-related features can indicate how cycling profiles affect battery life on average, while time-series features identify cell-to-cell variations.



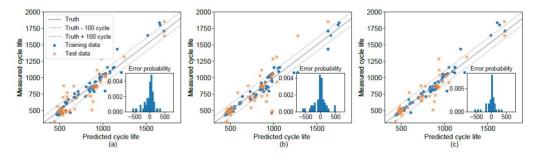


Figure 14. Prediction results using different feature inputs extracted from the Stanford dataset. (a) Using time-series features only. (b) Using usage-related histogram features only. (c) Using both feature sources.

Instead of combining the feature sources and training one ML algorithm, an alternative option is to train two ML algorithms using all the feature inputs and fuse their prediction results. The last row in Table 4 shows the numerical result of training two RFR prediction models and then fusing the prediction results using an appropriate method, such as an ensemble approach. Not surprisingly, the obtained results are similar to those achieved by using combined features to train a single ML algorithm.

In addition to the prediction results obtained using the first 100 cycles, we assess the robustness of the developed methods by varying the early life data from 20 to 300 cycles, as shown in Figure 15. It is noteworthy that incorporating usage-related features can significantly enhance prediction accuracy compared to the case of using only time-series features, especially in early prediction scenarios. For instance, when utilizing the first 20 cycles, prediction accuracy can be improved by around 45%. Furthermore, the consistent superiority of using combined features is observed across the entire examined range of [20, 300] cycles, underscoring the importance of including such information in the feature construction step.

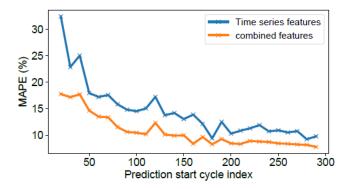


Figure 15. Prediction error as a function of prediction start cycle number.

#### Lifelong estimation of lithium plating potential

Figure 16(a) provides an overview of the results from the three-electrode battery cell aging test. The benefits of employing an anode potential control (CC-C $\eta$ ) charging strategy become evident when compared to the conventional CC-CV charging strategy. Notably, the overall lifetime of the CC-C $\eta$  group shows a remarkable



doubling compared to the group utilizing the CC-CV charging strategy. Furthermore, the charging speed of the CC-C $\eta$  group is observed to be 30% faster at the initiation of the battery's life and increases to 40% after 200 cycles, as depicted in Figures 6(b)–(e). This underscores the dual advantage of the anode potential control charging strategy: not only does it extend the battery's lifetime, but it also reduces charging time. It is crucial to highlight a notable observation in the aging curve of the CC-CV group. There is a sharp reduction in capacity at the early stages, followed by a stabilization of the degradation rate. We posit that this phenomenon may be attributed to the onset of lithium plating, even occurring at the conclusion of the battery's very early life. This is evident in Figure16(c) and (e), where the anode potential turns negative during the initial cycle.

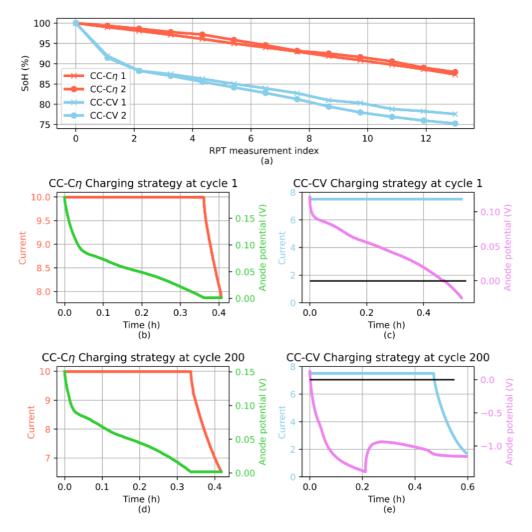


Figure 16. Three-electrode cells test results.

Overall, the estimation results using the proposed framework is really promising and a random selected cell are chosen to demonstrate the effectiveness of the algorithm as shown in Figure 17.



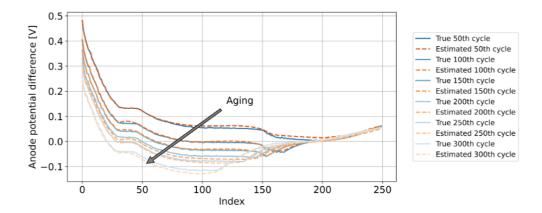


Figure 17. Anode potential estimation results of a randomly selected cell over its entire lifetime.

#### Diskussion

In this project, we have developed accurate, reliable, and practical methods for diagnosing the state of health, prognosing the future aging trajectory, and predicting the remaining useful life of lithium-ion batteries. These innovative methods are sufficiently generic and can be applied equally to various types of lithium-ion batteries and other battery chemistries. The improved estimation and prediction of battery aging can result in significant benefits, including: increased battery reliability, reduced costs for battery inspection, maintenance, replacement, and associated warranty and insurance services, facilitation of second-life battery usage and after-market treatment, and provision of a solid foundation for enhancing battery health. These results can be leveraged by battery design and manufacturing industries, battery management services, as well as regulatory and policy makers.

Furthermore, we proposed a machine learning-based lifelong estimation method for lithium plating potential and used it for health-aware fast battery charging. Through extensive simulations and lab-scale coin cell tests with specific sensors, we have demonstrated the capability to achieve both faster charging and a 100% longer battery lifetime. This advancement is poised to significantly enhance the convenience and acceptance of EVs, thereby expediting the transition to a sustainable transport system. The substantially extended lifetime will directly contribute to significantly enhanced sustainability. If successfully implemented in real-world EVs, it has the potential to reduce CO2 emissions by 6.54 g/km, a substantial figure aligning with the global goal of carbon neutrality.

The generated scientific results have been documented and resulted in 4 peerreviewed articles published on top-tier journals, 3 peer-reviewed articles published on leading international conferences, 2 journal articles under review, and 1 European patent application. Additionally, the results have been disseminated through six oral presentations/invited talks and a set of newsletters, e.g., on websites of Chalmers, CEVT, and the Swedish Electromobility Center. The next phase involves employing the developed ageing prediction models for optimised battery lifetime and expanding the fast charging results from simulation and tailored lab-scale coin cells to real-world commercial batteries. Fortunately, we have secured a new research grant, "Datadriven förlängning av livslängden och optimering av prestanda för fordonsbatterisystem" (Project no. P2023-00611), funded by the Swedish Energy Agency through the Vehicle Strategic Research and Innovation Program.

## Publikationslista

Following publications are included in this project and has been extensively discussed above.

- [1] Y. Zhang, T. Wik, J. Bergström, M. Pecht, and C. Zou, "A machine learningbased framework for online prediction of battery ageing trajectory and lifetime using histogram data," J. Power Sources, vol. 526, p. 231110, Apr. 2022. DOI: 10.1016/j.jpowsour.2022.231110.
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Administrativ bilaga

Vetenskapliga artiklar

- Zhang, Y., Wik, T., Bergström, J., & Zou, C. (2023). State of health estimation for lithium-ion batteries under arbitrary usage using data-driven multi-model fusion. *IEEE Transactions on Transportation Electrification*. DOI: 10.1109/TTE.2023.3267124
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