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Data-driven battery aging diagnostics and prognostics

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To my family.

Abstract

Lithium-ion (Li-ion) batteries play a pivotal role in transforming the transportation sector from heavily relying on fossil fuels to a low-carbon solution. But, as an electrochemical device, a battery will inevitably undergo irreversible degradation over time. Therefore, accurate and reliable aging diagnostics and prognostics become indispensable for safe and efficient battery usage. However, diverse aging mechanisms, stochastic usage patterns, and cell-to-cell variations impose significant challenges. As the importance of vehicle operating data is becoming more apparent, an increasing number of automotive companies are collecting battery field data.

In this thesis, a series of machine learning (ML) frameworks, using both field data collected during vehicle operation and laboratory cycling data, for battery aging diagnostics and prognostics is developed. Among these, a data-driven multi-model fusion method is proposed to accurately and robustly estimate battery capacity under real-world arbitrary usage profiles. Additionally, a battery aging prediction framework is developed based on a combination of offline global models created using different ML methods applying histogram operational data and cell individualized models that are online adapted. Finally, the thesis presents an early-life prediction pipeline leveraging time-series and histogram data, showing that these two feature sources are effectively interchangeable and complementary. These algorithms are extensively evaluated with various data sources of different battery kinds. The evaluation results indicate that the developed methods are accurate and robust, and more importantly, they are applicable to the harsh conditions encountered in real-world vehicle operations.

Keywords: Lithium-ion batteries, battery management system, state of health, remaining useful life, machine learning.

List of Publications

This thesis is based on the following publications:

[A] **Yizhou Zhang**, Torsten Wik, John Bergström, Changfu Zou, "State of health estimation for lithium-ion batteries under arbitrary usage using data-driven multi-model fusion". Submitted for publication in IEEE transactions on transportation electrification.

[B] **Yizhou Zhang**, Torsten Wik, John Bergström, Michael Pecht, Changfu Zou, "A machine learning-based framework for online prediction of battery ageing trajectory and lifetime using histogram data". Published in Journal of Power Sources, Apr. 526, p. 231110, Apr. 2022.

[C] **Yizhou Zhang**, Torsten Wik, Yicun Huang, John Bergström, Changfu Zou, "Data-driven battery life prediction considering both onsite measurement and usage information". Accepted by IFAC World Congress 2023.

Other publications by the author, not included in this thesis, are:

[D] Practical battery State of Health estimation using data-driven multi-model fusion, "**Yizhou Zhang**, Torsten Wik, John Bergström, Changfu Zou". Accepted by IFAC World Congress 2023.

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Acronyms

GHG:	Greenhouse gas
Li-ion:	Lithium-ion
BMS:	Battery management system
SoH:	State of health
RUL:	Remaining useful life
EV:	Electric vehicle
NMC:	Nickel-manganese-cobalt
SEI:	Solid electrolyte interphase
SoC:	State of charge
SoP:	State of power
SoE:	State of energy
BOL:	Beginning of life
EOL:	End of life
ML:	Machine learning
BRR:	Bayesian ridge regression
SVR:	Support vector regression
RFR:	Random forest regression
GPR:	Gaussian process regression
NN:	Neural network
RBF:	Radial basis function
ReLU:	Rectified linear unit function

ECM:	Equivalent circuit model
EM:	Electrochemical model
LSTM:	Long short-term memory
RPT:	Reference performance test

Contents

Part I

Overview

CHAPTER 1

Introduction

1.1 Introduction

Transformative changes in the transport sector are crucial for meeting climate mitigation targets [1]. As of 2019, 23% of the direct CO_2 emissions originate from the transport sector, among which 69% comes from road vehicles, as shown in Fig. 1.1, and are still growing steadily. Electromobility

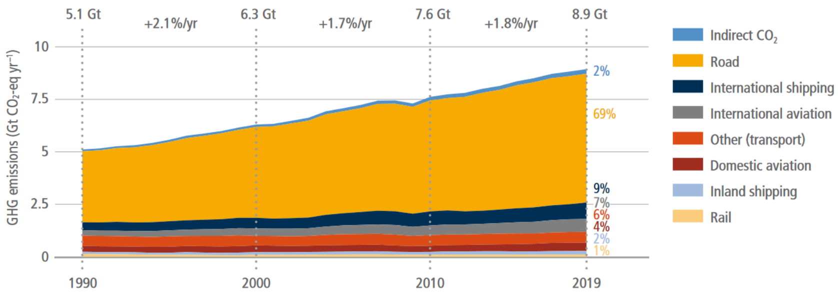

Figure 1.1: Transport global greenhouse gas (GHG) emissions trends [1].

powered with low-carbon electricity is one of the most significant contributors to reducing transportation GHG [2], [3]. Therefore, the electrification transformation of the automotive industry is indispensable. On the other hand, increasingly complex in-vehicle software functionality, software as a service business strategy, and software-defined vehicle concepts have profoundly impacted traditional automotive manufacturers [4]. The car industry is undergoing an unprecedented change from manufacturing mechanical-centric products to providing a software-oriented mobility service.

Lithium-ion (Li-ion) batteries, due to their high energy density, low cost, and relatively long lifetime, play a pivotal role in the powertrain electrification transition [5], [6]. Additionally, a battery management system (BMS), with accurate and reliable estimation and control algorithms, becomes crucial to ensure the safe and efficient use of the battery [7]. Fortunately, with ever-increasing attention on the software functionality of the vehicle, running advanced algorithms based on model-based or data-driven methods has become feasible and preferable for automotive companies [8].

As an electrochemical device, the aging characteristics of Li-ion batteries are highly complex and nonlinear [9]. The degradation path may be affected by not only the intrinsic factors, e.g., pack design and manufacture variability, but also extrinsic factors such as temperature and usage profiles [10]. Therefore, accurate and reliable aging state estimation and prediction are challenging, especially when conducted outside of the laboratory [11], [12]. With increasing awareness of “data is the new money” and regulatory pressure from the governments, more and more automakers are deliberately saving their vehicle’s operational usage data, including battery usage data, either on-board or in a cloud system [13]. However, massive amounts of data saving and transmission may consequently add more cost to the already narrow profit margin that most car markers have. Hence, how to intelligently save the right and suitable data to perform the necessary tasks becomes a natural question. Moreover, data collected in real-world applications unnecessarily have certain shortcomings, such as interrupted measurements and noisy signals. Field data, therefore, do not have the same quality as the data collected in a lab setting. This is why the robustness of the designed battery aging diagnostics or prognostics algorithms is crucial to overcome the data quality issues faced using real-world data. Last but not least, rigid vehicle development, comprehensive verification processes, and rigorous homologation requirements often require extensive tests to be conducted before the launching of new vehicles. For batteries, it can be an extensive cycling campaign for up to several years under different packaging levels, e.g., cell, module, and pack. Such a large amount of lab data can be of great use. Thus, how to systematically leverage the lab data with real-world field data to improve the overall algorithm performance becomes necessary.

The research presented here aims to apply recent data-driven modeling techniques using field data collected during vehicle operation and laboratory cycling data to conduct battery aging diagnostics and prognostics tasks.

1.2 Research questions

- How can field data collected onboard the vehicle be used to estimate battery capacity robustly?
- How to efficiently leverage large fleet data to indicate battery aging status and individually adapt to every vehicle?
- How can time-series and histogram usage data be combined for accurate and robust battery diagnostics and prognostics?

1.3 Contributions

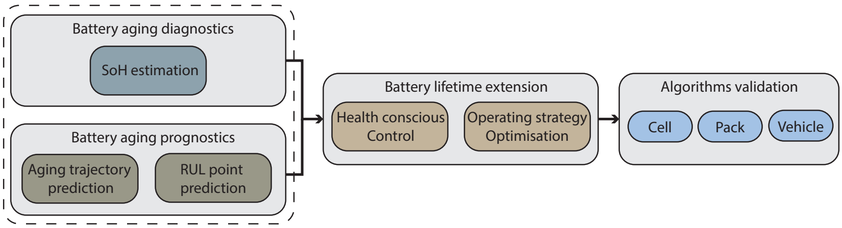

Figure 1.2: The overview of the project work packages, where the part included in the dashed block, is summarized in this thesis.

Fig. 1.2 illustrate the overview of the problems that are investigated or planned to be investigated. The contribution of the part that has been conducted can be summarized as follows, with more details introduced in subsequent chapters.

SoH estimation

Many efforts have previously been made by both academia and industry to improve the estimation of SoH. Still, relatively few methods can practically be applied in real-world applications with good accuracy and robustness. This thesis contributes to this area, as found in Paper A, where a method for a practical battery capacity estimation method under arbitrary usage profiles for automotive applications using data-driven multi-model fusion was developed.

Aging trajectory prediction

Compared to SoH estimation, fewer references can be found in the literature for aging trajectory prediction methods. However, the benefits of an accurate aging trend prediction model are also highly valuable, such as shortening the development cycle, timely preventative maintenance, and lower warranty and insurance costs. One contribution to this can be found in Paper B, where a systematic machine learning framework is proposed using histogram-based features to predict future battery aging trajectories.

Remaining useful life (RUL) prediction

Battery RUL is also a useful and important attribute from both technical and economic perspectives. Conventionally, time-series measurement data are used for conducting such tasks using machine learning methods. However, as demonstrated in Paper C, histogram usage data can achieve similar performance with much less data required. We extend the study of aging trajectory prediction to RUL prediction and conduct a comprehensive comparison of constructing features based on time-series data and usage-related histogram information.

1.4 Thesis outline

Chapter 1 briefly introduces the research project and describes the motivation of the work. Chapter 2 presents the battery system and its important components. They are followed by Chapter 3, which gives an overview of the used machine learning algorithms. Chapter 4 presents the methods for battery aging diagnostics, and Chapter 5 introduces the battery aging prognostics methods. Chapter 6 summarize the included papers. Lastly, Chapter 7 concludes the thesis and also highlights potential future research directions.

CHAPTER 2

Battery systems

Li-ion batteries are widely adopted as power sources for portable electronic devices, interrupted power supplies, and energy storage devices. Furthermore, they have become the dominating battery type for electric vehicles (EVs). This section intends to give a brief introduction to the battery systems with a special focus on the aging perspective of the battery characteristics.

2.1 Working principles

A Li-ion battery consists of a negative electrode, a positive electrode, a separator, an electrolyte, and two current collectors, as illustrated in Fig. 2.1. The negative electrode is often using graphite or, more recently, silicon-added graphite materials, and the positive electrode is a metal oxide, e.g., nickel-manganese-cobalt (NMC). The separator only allows ionic conductivity while being electrically insulated. During discharge, the lithium stored in the negative electrode will be released to the positive side. At the same time, the electrons move in an outer circuit to generate current. During charging, the mechanism is reversed [7], [14].

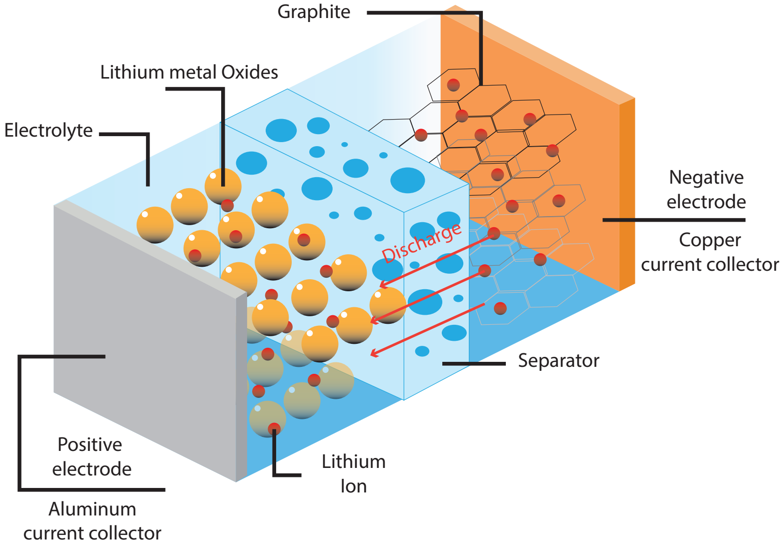

Figure 2.1: The overview of the working principle of the Li-ion battery.

2.2 Battery aging

As introduced in Section 1.1, the performance of the Li-ion batteries degrades over time due to various internal (cell mechanical and electrochemical property change), and external factors (operating conditions) [15]. Fig. 2.2 illustrates some of the common aging mechanisms that can happen within the batteries. Among these, solid electrolyte interphase (SEI) formation and lithium plating are the most severe aging mechanisms argued by many researchers [9], [16], [17].

The SEI layer is a passivation layer growing on the surface of active electrode materials. The SEI can form on both positive and negative electrodes, but comparatively, the negative side is dominant. When the voltage is outside the liquid electrolyte electrochemical stability voltage window, the liquid electrolyte will react with lithium ions and electrons within the electrodes to form a solid electrolyte [16], [18]. The SEI is formed in the first cycle when the cells are being charged, causing the cell to lose roughly 10% capacity. But the formed SEI layer limits further electrolyte reactions [19]. During the batter us-

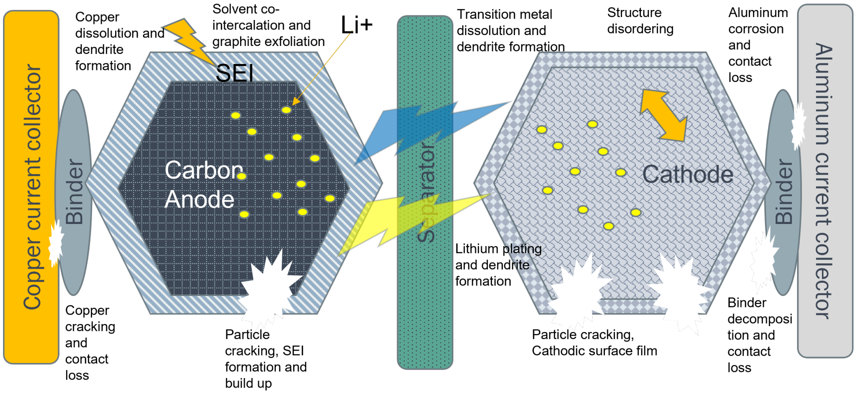

Figure 2.2: Different aging mechanisms that can potentially happen during the usage of the battery. Figure modified from [10].

age phase, the SEI layer will gradually increase. The electrolyte can continue to react with, e.g., newly exposed electrode area after cracking, plated lithium, or other transition metal, contributing to the SEI layer's further growth [20]. The continuous formation of the SEI causes the battery to lose capacity and increase in resistance irreversibly, consequently leading to further degradation of the battery [21]. The growth rate of the SEI layer is highly related to the battery operating conditions [22]. High storing or cycling temperature leads to a high diffusion rate and hence, a potentially higher SEI growth rate. Similarly, a higher current causes more particle cracking, therefore contributing to further SEI formation [23], [24].

Another major aging mechanism is lithium plating, where the lithium ions, instead of intercalating into negative electrodes, are deposited on the surface of the negative electrode, causing battery capacity retention and potentially also safety issues if the lithium penetrates the separator [25]–[27]. Lithium plating is highly related to the cycling conditions of the battery, especially during the charging process. The onset of the lithium plating can be attributed to several factors, such as high charging current or low temperature causing high electrolyte potential, which favors the side reaction instead of the normal intercalation process [28].

Apart from the above two major aging mechanisms, many other aging processes could be happening simultaneously. For instance, electrode structure change, particle fracture, and electrolyte drying. Moreover, all the aging mechanisms may interplay with each other to further complicate the already tricky battery degradation problem [9], [17], [29].

2.3 Battery states

Accurate estimation or prediction of the battery's internal states is essential to ensure safe and optimal battery usage. Fig. 2.3 illustrates the information flow of the battery's internal states.

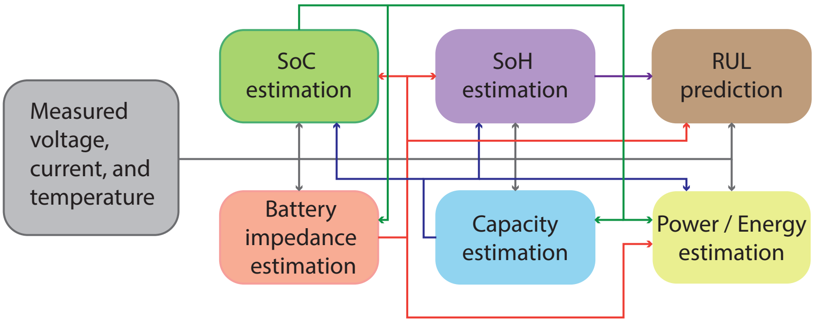

Figure 2.3: Information flow of different battery internal states.

The state of charge represents the ratio between the currently available battery capacity and the capacity when the battery is fully charged and serves as a fuel gauge for electric vehicles. In the academic community, many efforts have been devoted to this area in the past. Methods such as the Kalman filter, incremental capacity analysis, and machine learning have been used to estimate the state of charge (SoC) value. Some of the methods have been adopted by the industry and deployed to commercial products as well. For a detailed review of different SoC estimation methods, readers are referred to [30]–[32].

Battery impedance is an essential input for estimating many other key internal states, e.g., SoC, state of power (SoP), and SoH. The value of the impedance will change depending on the aging state of the battery and also the operating conditions (current, SoC, and temperature). Some of the existing methods to estimate impedance are using electrochemical impedance spectroscopy or applying an equivalent circuit model using a Kalman filter, or adopting an electrochemical model using an observer [32], [33].

The battery capacity represents the available charge and discharge capability at the current stage. Due to degradation, the capacity will usually decrease over time. The retention of the capacity value leads to shortening the vehicle's available driving range and affecting vehicle performance. Therefore, accurately estimating battery capacity is important. Noteworthy, both impedance and capacity can be used to indicate battery SoH, which can be defined as:

$$\mathrm{SoH}_R = \frac{R}{R_{nom}} \times 100\%, \tag{2.1}$$

$$\mathrm{SoH}_C = \frac{C}{C_{nom}} \times 100\%, \tag{2.2}$$

where R and C represent the current battery impedance and capacity, respectively, and R_{nom} and C_{nom} are the nominal battery impedance and capacity at the beginning of life (BOL) [34].

As the battery degrades over time, knowing how long / how many cycles the battery can still be used until its end of life (EOL) is of course, important. Such an attribute is often called the RUL of the battery. Predictive maintenance, maximizing the residual value of the battery, and mitigating warranty claims all rely on an accurate and reliable RUL prediction [11], [35].

Battery state of power/energy (SoP/SoE) is two states that will impact the drivability and longevity of the vehicles and need to be estimated all the time in the car [32]. SoP predicts the battery's maximum available charge and discharge power without triggering premature aging or violating safety boundaries. And SoE indicates the available energy stored in the battery.

2.4 Battery management system (BMS)

Typically, all the battery's internal states mentioned in the previous section need to be calculated in a microcontroller onboard a vehicle. Such a microcontroller is often regarded as the master of the BMS. Apart from conducting such core estimation work, the BMS will also continuously measure cell voltage, current, and temperature. Furthermore, many protection functions, such

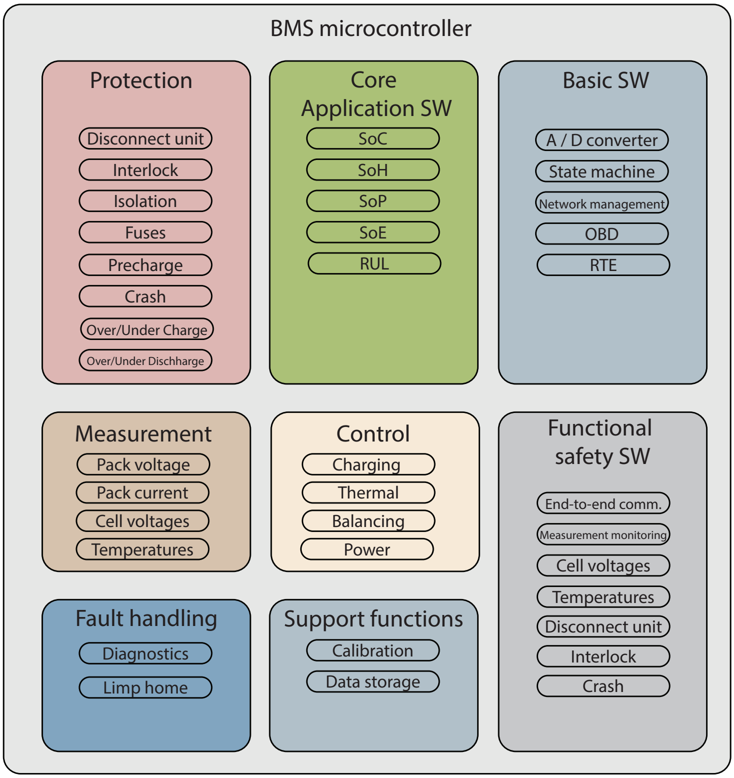

Figure 2.4: Overview of the functions of a typical BMS.

as battery disconnect unit control, isolation check, and interlock check, will also run on the BMS to ensure safe operation and avoid hazardous situations [36]–[38]. Fig.2.4 shows a typical BMS function diagram. Traditionally, low-end microcontrollers are of first choice in the automotive industry due to cost reasons. That entails limited computational power and memory space. Thus, advanced algorithms with heavy computational requirements may become prohibited. Fortunately, with ever-increasing telecommunications techniques and data storage capabilities, next-generation cloud-based BMS is becoming feasible and potentially economical [8], [39], [40]. Fig. 2.5 illustrates the structure of a cloud-based BMS. Many of the advanced but data-heavy

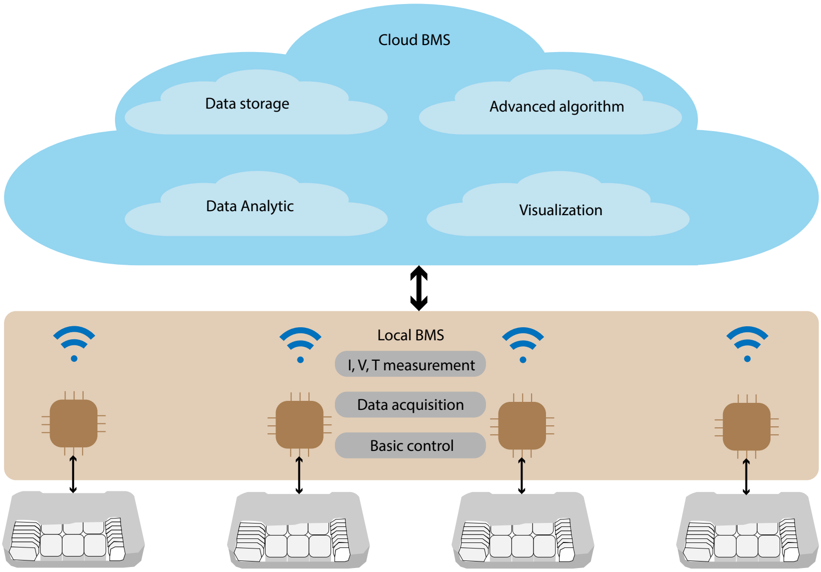

Figure 2.5: Illustration of a cloud-based BMS.

or computational-heavy algorithms can be run in the cloud and only send the final result to the vehicle. Meanwhile, more and more companies realize the importance of vehicle data. Massive amounts of operating information can now be logged and saved in a data center. To take full advantage of such an asset, state-of-art data-driven methods need to be applied to better manifest the battery's internal state and future trends.

CHAPTER 3

Machine learning (ML)

ML is a set of methods using data to conduct learning, reasoning, and execution [41]. Through model training, ML algorithms build a model based on sampled data and then make predictions or actions using the trained model. In general, ML can be divided into three categories: supervised learning, unsupervised learning, and reinforcement learning. Among these, supervised learning refers to the case when the model is trained from labeled input-output data. In other words, the results (output) are first defined by the domain experts, and the goal of the model is to mimic the labeling done by the experts. Within the supervised learning framework, there are two main categories of problems, one being regression and the other being classification [42], [43]. The regression problem is intended to estimate the relationship between a desired output and one or more input features. In contrast, classification is the problem of identifying which categories an observation belongs to.

In this thesis, battery aging diagnostics and prognostics problems are in focus, and usually, such problems are formulated as supervised learning problems. If not stated otherwise, all the ML algorithms we refer to in later chapters long to the regression category.

3.1 ML algorithms

Among the large pool of ML algorithms, some are particularly interesting for solving battery aging diagnostics and prognostics tasks due to their characteristics. Here, several typical such algorithms, applied later in the appended papers, are briefly introduced.

Bayesian ridge regression (BRR)

Fitting a straight line or a hyperplane to data is probably the most straightforward way to form a linear model. The simple and neat model construction makes the computation extremely simple and the model output easy to interpret. The contracted model, usually called linear regression, and for a line, the model has the form $y = \theta^T \cdot x_p + c$, where θ and c are the model parameters that need to be found, and x_p is the training data. Applying a Bayesian approach to a linear regression model leads to the so-called BRR. Instead of treating the coefficient θ as a single variable, BRR assumes θ as a spherical Gaussian distribution, defined as $P(\theta) = \mathcal{N}(\theta; 0, \Sigma_0)$, having zero mean and a covariance Σ_0. To simplify the calculation, Σ_0 is chosen to be $\mathcal{I}\alpha$, where $\mathcal{I}$ is the identity matrix and α is a positive hyperparameter [44], [45]. For a detailed treatment of the algorithm, we refer to [46]. As with all linear models, the drawback is when the linear assumption does not hold. As a consequence, the model's performance will suffer when treating complex and nonlinear systems.

Support vector regression (SVR)

SVR is a kernel-based algorithm utilizing a nonlinear mapping function $\Phi(\cdot)$ to transform the data from low-dimensional space x_p to a high-dimensional space $\Phi(x_p)$. Such that the estimated function is linear in $\Phi(x_p)$, $f(x_p) = w^T \Phi(x_p) + b$, where both w and b are the model parameters that need to be found. Similar to linear regression in the form, but can be solved as a quadratic programming problem. The optimization problem can be formulated as follows:

$$\begin{aligned} \min_{w,b,\xi,\xi^*} \quad & \frac{1}{2}\|w\|^2 + C\sum_{p}^{\mathcal{P}}(\xi_p + \xi_p^*) \\ \text{subject to} \quad & \begin{cases} y_p - w^T\Phi(x_p) - b \leq \epsilon + \xi_p \\ w^T\Phi(x_p) + b - y_p \leq \epsilon + \xi_p^* \\ \xi_p, \xi_p^* \geq 0 \end{cases} \end{aligned} \tag{3.1}$$

where ξ_p and ξ_p^* are positive slack variables, C is a regularisation coefficient, and ϵ is the error tolerance coefficient. Both C and ϵ are hyperparameters to be determined using cross-validation. To avoid explicitly computing $\Phi(x_p)$, the kernel trick can be used [47]. Intuitively, SVR constructs a tube around a hyperplane with the aim of letting as many training points as possible fall into the tube and, at the same time, the hyperplane is kept as flat as possible, as depicted in Fig. 3.1.

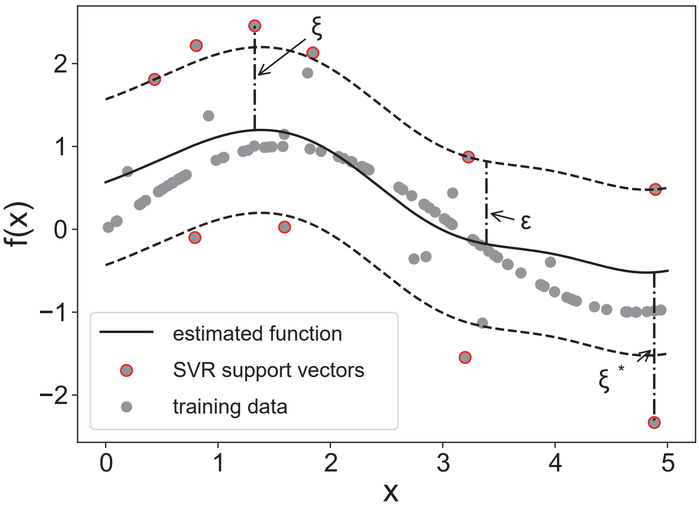

Figure 3.1: Illustration of SVR with one single feature.

Random forest regression (RFR)

RFR is an ensemble supervised learning method leveraging two core techniques called “bootstrap” and data aggregation. At the sampling collection stage, every individual tree randomly selects a subset of the training data and feature to conduct prediction, and this process is called “bootstrap”. After each tree has its own prediction result, RFR aggregate all the results by taking an average [48]. The total number of decision trees and the maximum depth of each individual tree are two important hyperparameters to make a trade-off between model performance, computational resources, and training time. The overall structure of the RFR algorithms is illustrated in Fig. 3.2. Comparatively, RFR has a better bias-variance trade-off than the single decision tree model. Moreover, the prediction results from RFR models are generally easier to interpret compared to other complex nonlinear ML algorithms.

Gaussian process regression (GPR)

GPR is a non-parametric supervised learning algorithm which means that a finite set of parameters cannot represent the model, which is rather given in a function format $f(\cdot)$ [49]. Often the function is assumed to be distributed according to a Gaussian process and represented as

$$f(x) \sim \mathrm{GP}\left(m(x), \kappa(x, x')\right), \tag{3.2}$$

where x and x' are two arbitrary data samples, $m(x)$ represents the mean value of $f(x)$, and $\kappa(x, x')$ is the covariance of $f(\cdot)$ between the points x and x'. Notably, both the mean and the covariance functions can incorporate prior knowledge about the estimated function $f(x)$. The posterior probability of the predicted output $f(x^*)$ at any point x^* in the test set can be calculated by calculating the probability of conditioning f on the complete training dataset $(\mathbf{X}, f(\mathbf{X}))$, where $\mathbf{X}$ is a vector of all the $\mathcal{P}$ training samples, i.e., $\mathbf{X} = [x_1, \cdots, x_{\mathcal{P}}]$. The predicted output probability distribution can be

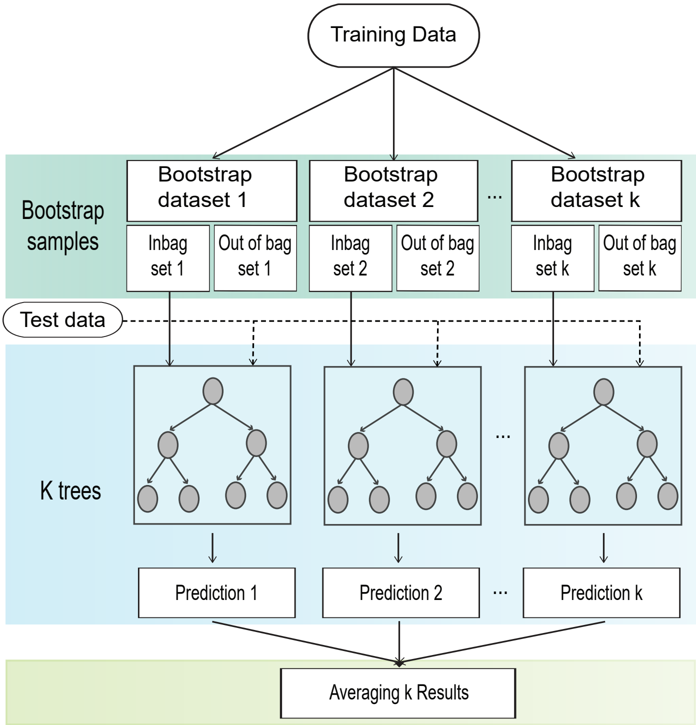

Figure 3.2: Illustration of RFR algorithm.

represented as:

$$p\left(f(x^*)|x^*, \mathbf{X}, f(\mathbf{X})\right) \sim \mathcal{N}\left(f(x^*); m^*, \Sigma^*\right) \tag{3.3}$$

$$m^* = \mathcal{K}(\mathbf{X}, x^*)^T \mathcal{K}(\mathbf{X}, x^*)^{-1} f(\mathbf{X}) \tag{3.4}$$

$$\Sigma^* = \mathcal{K}(\mathbf{X}, x^*) - \mathcal{K}(\mathbf{X}, x^*)^T \mathcal{K}(\mathbf{X}, x^*)^{-1} \mathcal{K}(\mathbf{X}, x^*) \tag{3.5}$$

where $\mathcal{K}$ is the covariance kernel matrix having κ as elements. Usually, the kernel function $\kappa(x, x')$ can be seen as a hyperparameter that can be tuned during training using cross-validation. Typical kernel functions are radial basis function (RBF), Matérn kernel, and rational quadratic kernel [49]. Fig. 3.3 illustrates a typical GPR algorithm. Due to its Bayesian origin, GPR can naturally propagate its prediction uncertainty, which is seen as a merit for

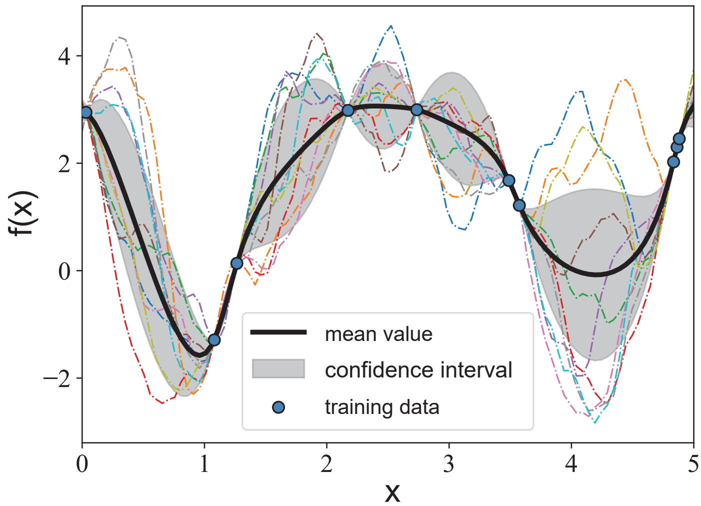

Figure 3.3: Illustration of GPR algorithm with one single feature.

many applications. However, when the data amount increases, the necessity of a matrix inversion during posterior probability population, as given in (3.5), becomes computationally expensive.

Neural network (NN)

Conceptually, the invention of the NN was inspired by the human brain's cognition process, where the goal is to learn from data and find the best model to map from input to output. NN usually consists of several layers, with each layer made of multiple units, which can also be referred to as neurons. The mathematical form for an arbitrary neuron $j \in \{1, \cdots, J_l\}$ in a hidden layer $l \in \{0, 1, \cdots, L\}$ can be represented as

$$z_{j,l} = \sum_{j=1}^{l_J} w_{j,l} h_{j,l-1} + b_{j,l} \tag{3.6}$$

$$h_{j,l} = a_l(z_{j,l}) \tag{3.7}$$

where w, b, and a are the weight of the unit, the bias factor, and the activation function, respectively. Depending on the preference of the out, the activation function can choose accordingly, or it can also be selected through cross-validation. Typical activation functions are the rectified linear unit (ReLU) function, the *tanh* function, and the *sigmoid* function. Then a loss function is chosen to measure the difference between the model's output and the ground truth. With all the weights and biases as decision variables, gradient descent-based algorithms are often adopted to solve the optimization problem [43]. The typical NN architecture and its mathematical representation are shown in Fig. 3.4.

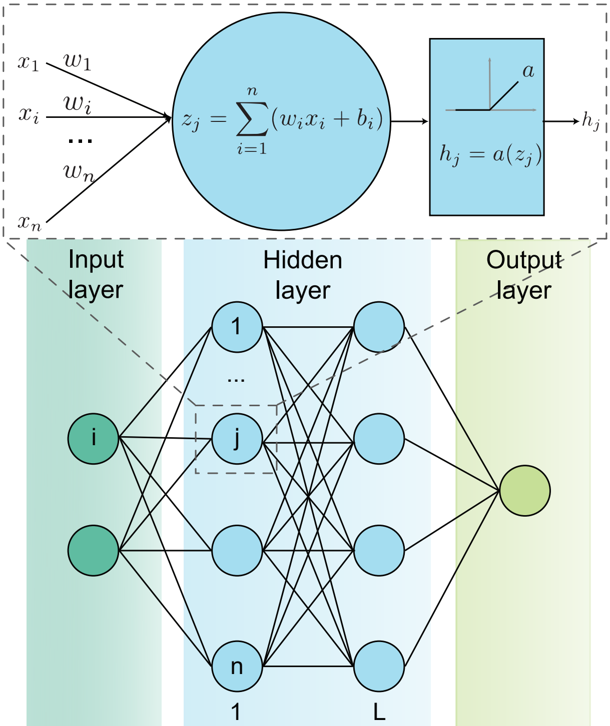

Figure 3.4: Illustration of NN.

The remarkable ability to detect patterns and identify trends from complex and nonlinear systems with complicated or imprecise data has made NN an attractive method for many applications, such as image recognition and natural language processing. Furthermore, the capability of handling large datasets and the flexibility to accommodate parallel computation make NN a preferred choice in many real-world applications. On the other hand, NN also has its own pitfalls, e.g., a high required tuning effort, risk of overfitting, and relatively long training time.

CHAPTER 4

Battery aging diagnostics

During normal charging and discharging operation, batteries undergo the main intended electrochemical reactions but also several complex and interacting side reactions, which induce the degradation of the battery over time as introduced in Chapter 3. To ensure safe and optimal usage of the battery systems, accurate and reliable aging diagnostics are essential. Often, battery aging is referred to as SoH estimation [50]. It is worth mentioning that the battery SoH can be defined in various ways, e.g., resistance growth or capacity decrease. In this thesis, the definition of the SoH is adopted as the ratio between the current available capacity and the nominal capacity at the BOL.

In recent years, many efforts have been made by both academia and industry to estimate battery capacity, as reviewed in [33], [51]–[55]. Generally, these methods can broadly be divided into the empirical method [56]–[60], model-based methods [61]–[64], and data-driven methods [65]–[67]. Based on the extensive laboratory cycling data, the empirical model build up a mathematical relationship between the cell capacity and some common battery degradation features, e.g., Ah counting, equivalent cycle number, or elaborated time. The simple and niche formulation made such methods attractive in the early days. However, compared to cycling cells in a laboratory setting in a well-controlled environment, cells used in real-world applications undergo much more dynamic and ever-changing operating conditions. Therefore, the empirical model fitted with lab data fails to extrapolate to the battery operated in the field [12].

Compared to the empirical models with poor generalization, model-based methods are capable of continuously updating internal parameters based on the measurements and show better performance for specific applications [35]. Usually, model-based methods can be further divided into the ones using equivalent circuit models (ECMs) and the ones using electrochemical models (EMs). ECMs are constructed using only electric circuitry components. The simple and straightforward implementation requirement has made them attractive for battery aging diagnostics. However, the performance may degrade over time when batteries deployed to the application encounter rough and complicated operating conditions, e.g., EVs. Additionally, the model accuracy may decrease over time due to a lack of regular RPT tests to recalibrate models [55]. EMs are derived from the porous electrode and liquid concentrated solution theory, which captures the battery's internal characteristics in detail [68], [69]. To model the aging behaviors, several aging models, for example, SEI [70], [71], lithium plating [26], and particle cracking models [72] are coupled to the original EM. However, the high computational request for running such models, and difficulties in parameterization, have made such methods hard to use in online applications [73].

In contrast to building an aging model from the first principle, data-driven methods are the mechanism-agnostic ways to capture the aging state by exploring only the measured signals from the battery operation. Due to recent digitalization trend and the introduction of new technologies, such as digital twin, battery intelligent management system [39], [74], [75], data-driven methods have drawn much interest in solving battery diagnostics problems. Both feature-based ML algorithms and end-to-end deep learning methods have been proposed to estimate the capacity of the batteries [51]–[53], [65]–[67]. For the feature-based methods, the designers manually select the relevant features to indicate the aging states from the measured raw data or estimated states from the BMS using domain knowledge [76], [77]. Some of the common features are extracted from measured voltage, current, temperature, or accumulated time. On the other hand, filtered or calculated signals have also been explored to estimate battery SoH, such as incremental capacity (IC), differential voltage

(DV), and differential temperature (DT). For example, Deng et al. [78] constructed features from the discharge capacity and voltage curve under pulse discharging conditions and showed a clear correlation between the features and the cell capacity. Sui et al. [79] used the fuzzy entropy of the voltage response after the pulse excitation to successfully estimate the battery SoH. The measurement signal during the charging relaxation period can also manifest the aging state, which has been investigated by Zhu et al. [80]. Differential measured signal to highlight the material phase change process has been widely used as a battery diagnostics tool [81]–[85]. Hence, the rich aging-related information made them good candidates as input features for machine learning algorithms. She et al. [86] constructed the IC-related feature from real-world bus operating data to indicate the aging status of the battery. Wang et al. [87], and Li et al. [88] estimate the battery SOH using features extracted from the DT curve. In contrast to manually constructing features, adopting deep learning algorithms can potentially skip the feature engineering steps and use only raw measurement data as input. For example, Li et al. [89] used voltage signals under repetitive CC-CV charging cycles to estimate cell capacity using Long short-term memory (LSTM) NN. Tagade et al. [90] adopted the raw measurement data from the discharging part to train a deep GPR algorithm to estimate battery capacity.

Even though most of the methods mentioned in the previous paragraph demonstrate good estimation results, the practical applicability of these methods on real-world applications, especially automotive applications, is not thoroughly investigated [12], [91], [92]. Compared to cycling cells in the laboratory, cells deployed in the real-world experience much more diverse cycling conditions and operating environments. Additionally, noisy measurements and varying sampling rates also impose additional difficulties for the diagnostic algorithm to function well in practice. To tackle these challenges, a charging scenario-based feature extraction strategy is introduced in Paper A to ensure SoH estimation under arbitrary operating conditions and a Kalman filter-based online model fusion algorithm to enhance the estimation accuracy and robustness.

CHAPTER 5

Battery aging prognostics

Battery aging diagnostic algorithms intend to probe the current aging state. In contrast, battery aging prognostic algorithms try to predict how long the batteries can still be used until their EOL, as depicted in Fig. 5.1. Accurate

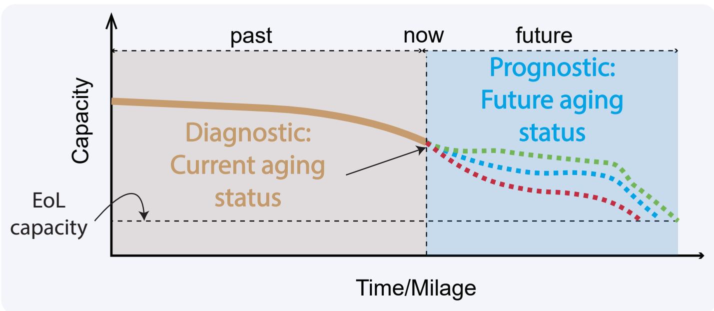

Figure 5.1: Illustration of battery aging diagnostics and prognostics.

aging prognostics are of great importance for predictive maintenance, health-conscious usage, and evaluation of residual value. According to the prediction target, the prognostic method can be categorized as future aging trajectory prediction and RUL point prediction [50]. The main difference is that the aging trajectory prediction aims to extract information about different aging states by predicting the aging states at selected points in time until the battery reaches its EOL. While RUL point prediction only focuses on how long the battery can still be used.

Due to similar characteristics, methods applied to battery aging diagnostics can often be adapted to conduct battery aging prognostics as well. Therefore, methods such as empirical methods and model-based methods can also be used for battery prognostic tasks as reviewed in [35], [52], [55]. However, the shortcomings of these methods for battery aging diagnostics apply also to prognostics, which will limit their effectiveness in a real-world deployment. Comparatively, data-driven methods have a better fidelity and complexity trade-off and may thus suit better for such a task.

For aging trajectory prediction, a growing number of data-driven methods have been developed. For instance, Li et al. [93] developed a recurrent NN-based sequence-to-sequence model using the historic capacity information to predict the future capacity trajectory. Richardson et al. [94] applied a GPR to successfully predict both the short-term and long-term capacity trajectory and quantify the estimation confidence interval.

However, these methods often require repeatable cycling profiles, which are not comparable with the random usage pattern encountered in real-world applications. Additionally, extensively using time-series-based measurement as feature input may require large data storage space in the cloud, e.g., one vehicle with several complex cycles can easily reach gigabyte data size [11], [12]. Therefore, from both economic and energy-saving points of view, they impose challenges. Several pilot works explored the usage of histogram-like data to formulate feature input to minimize the input data size. Richardson et al. [95] adopted capacity throughput and time duration as feature inputs, while Greenbank et al. [96] mainly used calendar time and time spent in specific voltage windows as inputs. However, all data used in these works are still laboratory repetitive cycling data; thus, the practical capability of such a model may be limited. In Paper B, a machine learning framework is proposed leveraging histogram-based usage data to formulate features to predict the future aging trajectory of the battery. Additionally, to tackle the problem of cell-to-cell variations, an online adaptive correction model is also developed.

Similar to aging trajectory prediction, data-driven methods can also be applied to predict the RUL point prediction. Severson et al. [97] innovatively discovered a feature based on the variance of the time series voltage signal and used an elastic net model to successfully predict the RUL of the battery only using the first 100 cycles information. Jones et al. [98] applied electrochemical impedance spectroscopy measurements as feature input and trained probabilistic ML models to predict the future discharge capacity. However, to the best of the author's knowledge, there is no work using usage-related histogram information to forecast battery lifetime and examine its relationship with the methods using time-series measurements. In Paper C, a comprehensive comparison study was conducted to investigate the prediction performance between using time series measurement data and adopting histogram usage information.

CHAPTER 6

Summary of included papers

This chapter provides a summary of the included papers.

6.1 Paper A

Yizhou Zhang, Torsten Wik, John Bergström, Changfu Zou
State of health estimation for lithium-ion batteries under arbitrary usage using data-driven multi-model fusion
Submitted for publication in IEEE transactions on transportation electrification .

This paper investigates the problem of real-world battery aging diagnostics. More specifically, estimating the battery state of health under the condition of dynamic vehicle operating conditions, stochastic user behaviors, and cell-to-cell variations. First, all possible operating conditions are categorized into six scenarios which are complementary to each other. Second, a comprehensive feature pool is extracted for each scenario to indicate the battery aging state. Then four machine learning algorithms are applied, of which two are probabilistic-based, and the other two are frequentist-based to estimate the aging state individually using time-series data. A histogram-based online adaptive model is trained to predict the one-step-ahead capacity of the battery. In the end, a Kalman filter is applied to systematically fuse all estimation and prediction results to increase accuracy and robustness.

6.2 Paper B

Yizhou Zhang, Torsten Wik, John Bergström, Michael Pecht, Changfu Zou
A machine learning-based framework for online prediction of battery ageing trajectory and lifetime using histogram data
Published in Journal of Power Sources,
vol. 526, p. 231110, Apr. 2022.
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This paper developed a data-driven method to systematically predict the future aging trajectory for batteries used in real-world applications. The wide range of operating conditions, highly nonlinear aging trends, and cell-to-cell variations posed challenges for battery health prediction. To tackle this, a histogram-based usage-related feature pool was first constructed regardless of raw data format and usage profiles. The proposed prediction framework has been trained and tested both on laboratory cycling data and real-world vehicle fleet data, all showing promising results. Moreover, an online adaptation algorithm was developed to further reduce the errors by up to 13%.

6.3 Paper C

Yizhou Zhang, Torsten Wik, Yicun Huang, John Bergström, Changfu Zou
Data-driven battery life prediction considering both onsite measurement and usage information
Accepted by IFAC World Congress 2023 .

This work developed a data-driven battery early-life prediction algorithm using both time-series, measurement-related features, and usage-related features. A comprehensive comparison study using these two different feature sources is also conducted. The comparison indicates these two feature sources are, in principle, complementary to each other. Therefore, if possible, combining these two feature sources can increase the prediction accuracy and enhance the robustness. Additionally, four commonly used machine learning algorithms are applied to compare the effectiveness and performance of these algorithms on battery early-life predictions problem. In the end, batteries with different cell chemistry are used to verify the applicability and generality of the developed methods.

CHAPTER 7

Concluding remarks and future work

Batteries play a pivotal role in renewable energy integration, electric vehicle roll-out, and stable electric grid operation, thereby significantly contributing to mitigating the climate change issue on our planet. Safe and efficient use of batteries requires detailed information regarding their current aging status, future aging trend, and RUL. Hence, accurate and robust battery aging diagnostics and prognostics are indispensable. This thesis developed a series of methods applying machine learning algorithms on laboratory cycling data and real-world field data to tackle battery aging estimation and prediction problems. The developed methods' effectiveness and generality are demonstrated with cells of different chemistry. Additionally, numerical estimation and prediction error results also show the advantages and benefits of the proposed methods.

Although many efforts have been made in the battery aging area, including this thesis, many research questions remain unanswered. Some research prospects are highlighted here for followers to build more fruitful results.

Undoubtedly, applying field data for battery aging diagnostics and prognostics tasks is beneficial and necessary. However, as often, opportunities come together with challenges. One of the biggest issues of leveraging field data is the lack of ground truth validation data. Since the battery used in real-world application rarely have a controlled environment to run a thorough reference performance test (RPT) to record the true aging state or EOL condition, the proposed estimation or prediction model may find it difficult to evaluate their performance. Quick RPT tests or on-purpose RPT tests, which can easily integrate into day-to-day usage (during vehicle charging), can potentially be a way forward to solve such issues. Furthermore, the data usually does not contain cells that are cycled to their EOL, which is often the case for automotive applications. It may intrinsically cause a data imbalance issue, hindering the model's fidelity and applicability. This thesis presented an online battery adaptation method, but the model cannot update its structure or parameters based on the new coming data. Methods that can continuously learn from the incoming new data or models that can adapt on the fly are promising directions for further research.

Another interesting area is how to combine physics-based models together with data-driven models. Physics-based models can detail the individual aging mechanism but are difficult to parameterize and compute. On the other hand, the data-driven model serves as a black box model, so the prediction result is hard to interpret. Therefore, developing a physics-embedded data-driven model that can bring advantages from both ends should potentially achieve better performance.

The overview of the project scope is shown in Fig. 1.2 of Section 1.1. This thesis only explored the battery aging diagnostics and prognostics part, whereas the strategy for extending the lifetime of the battery has not been covered. Hence, a natural continuation of the work is to make the utmost of the aging estimation and prediction information to optimize the battery's future usage so that the battery's lifetime can be extended. Some early attempts have been made in this area, such as using pulse charging [8], [99], lithium plating free fast charging [100], or dynamic reconfiguration of the battery [101]. However, the practicability and the feasibility of such methods applied to the end products still need much more investigation to meet the harsh operating conditions that are encountered in real-world applications.

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