THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

Interpretable Battery Lifetime Prediction Using Early Degradation Data

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To my friends and parents.

Abstract

Battery lifetime prediction using early degradation data is crucial for optimizing the lifecycle management of batteries from cradle to grave, one example is the management of an increasing number of batteries at the end of their first lives at lower economic and technical risk.

In this thesis, we first introduce quantile regression forests (QRF) model to provide both cycle life point prediction and range prediction with uncertainty quantified as the width of the prediction interval. Then two model-agnostic methods are employed to interpret the learned QRF model. Additionally, a machine learning pipeline is proposed to produce the best model among commonly-used machine learning models reported in the battery literature for battery cycle life early prediction. The experimental results illustrate that the QRF model provides the best range prediction performance using a relatively small lab dataset, thanks to its advantage of not assuming any specific distribution of cycle life. Moreover, the two most important input features are identified and their quantitative effect on predicted cycle life is investigated. Furthermore, a generalized capacity knee identification algorithm is developed to identify capacity knee and capacity knee-onset on the capacity fade curve. The proposed knee identification algorithm successfully identifies both the knee and knee-onset on synthetic degradation data as well as experimental degradation data of two chemistry types.

In summary, the learned QRF model can facilitate decision-making under uncertainty by providing more information about cycle life prediction than single point prediction alone, for example, selecting a high-cycle-life fast-charging protocol. The two model-agnostic interpretation methods can be easily applied to other data-driven methods with the aim of identifying important features and revealing the battery degradation process. Lastly, the proposed capacity knee identification algorithm can contribute to a successful second-life battery market from multiple aspects.

Keywords: Lithium-ion battery, lifetime early prediction, uncertainty quantification, interpretable machine learning, capacity knee.

List of Publications

This thesis is based on the following publications:

[A] **Huang Zhang**, Yang Su, Faisal Altaf, Torsten Wik, Sébastien Gros, "Interpretable Battery Cycle Life Range Prediction Using Early Cell Degradation Data". Accepted in IEEE Transactions on Transportation Electrification, Dec. 2022.

[B] **Huang Zhang**, Faisal Altaf, Torsten Wik, Sébastien Gros, "Comparative Analysis of Battery Cycle Life Early Prediction Using Machine Learning Pipeline". Accepted in 22nd IFAC World Congress, Yokohama, Japan, Jul. 2023.

[C] **Huang Zhang**, Faisal Altaf, Torsten Wik, Sébastien Gros, "Battery Capacity Knee Identification Using Unsupervised Time Series Segmentation on Degradation Curvature". Manuscript for submission, May. 2023.

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Acronyms

BESS:	Battery Energy Storage System
BEV:	Battery Electric Vehicle
BMS:	Battery Management System
DEC:	Diethyl Carbonate
DMC:	Dimethyl Carbonate
DoD:	Depth of Discharge
DVA:	Differential Voltage Analysis
EC:	Ethylene Carbonate
ECM:	Equivalent Circuit Model
EIS:	Electrochemical Impedance Spectroscopy
EMC:	Ethyl Methyl Carbonate
EoL:	End of Life
HEV:	Hybrid Electric Vehicle
ICA:	Incremental Capacity Analysis
LAM:	Loss of Active Material
LFP:	Lithium Iron Phosphate
LLI:	Loss of Lithium Inventory
LMO:	Lithium Manganese Oxide
NCA:	Lithium Nickel Cobalt Aluminum Oxide
NMC:	Lithium Nickel Manganese Cobalt Oxide
OCV:	Open Circuit Voltage

PC:	Propylene Carbonate
PHEV:	Plug-in Hybrid Electric Vehicle
QRF:	Quantile Regression Forests
RPT:	Reference Performance Test
SEI:	Solid Electrolyte Interphase
SoC:	State of Charge
SoH:	State of Health
SoP:	State of Power

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Part I

Overview

CHAPTER 1

Introduction

1.1 Motivation

For the purpose of further improving the energy efficiency of road transportation, reducing reliance on fossil fuels, and mitigating carbon emission, the commercialization of xEVs, i.e., all types of battery electric vehicles (BEVs), hybrid electric vehicles (HEVs), and plug-in hybrid electric vehicles (PHEVs), continues accelerating in the global market despite the economic repercussions of the pandemic [1]. Lithium-ion batteries have been the most widely used as the power source in xEVs and stationary energy storage systems, thanks to their key characteristics, such as high power and energy density, rapid response, and long lifetime [2]. However, as a result of complex interactions of underlying physical and chemical degradation mechanisms, their performance (e.g., available energy and available power) degrades over their service lives [3]. In some particular cases, an occurrence of sudden acceleration of capacity fade (called capacity knee) can result in accelerated performance degradation and even safety issues [4]. Therefore, understanding battery degradation processes, and providing lifetime prediction using early degradation data would enable many new possibilities throughout the battery life cycle. Four examples of

such possibilities are rapid validation of new cell manufacturing processes and formation processes [5], early battery replacement planning [6], early battery repurposing planning [7], selection of high-cycle-life fast-charging protocol [8].

However, it is challenging to provide accurate battery lifetime prediction with only limited early degradation data. The main reason is that the degradation process of lithium-ion batteries is highly nonlinear with possibly little capacity fading at early cycles, under the impact of battery design (e.g., battery chemistry and battery geometry), battery manufacturing processes, and operating conditions in applications [5]. The significance and complexity of battery lifetime prediction using early degradation data have made itself an intensive research problem [9] [10] [11] [12] [13] [14] [15] [16] [17] [18] [19].

The battery lifetime prediction methods can be roughly divided into three categories, i.e., model-based methods, data-driven methods, and hybrid methods. Model-based methods typically start with developing a mathematical model (e.g., a physics-based, a semi-empirical, or an empirical model) that captures the battery degradation process, which is then incorporated into a recursive Bayesian filter framework, such as a particle filter [9] [10] [11]. More specifically, the battery model parameters are selected as a state vector being recursively updated with measured capacity data. The future capacity is then predicted and the battery lifetime is obtained when the predicted future capacity reaches a predefined end-of-life (EoL) threshold. Although battery lifetime prediction performance using model-based methods has been successfully demonstrated in the aforementioned studies, the methods still struggle to predict battery lifetime with high accuracy at an early stage, not only because of the limited data available in early life, but also because of the two-stage degradation process separated by the knee.

Instead of developing an explicit mathematical model to capture the battery degradation process in the first place, data-driven methods that are used in some studies directly learn a mapping function from input features extracted from battery early degradation data to the battery lifetime, given a training set of input-output pairs [12] [13] [14]. While in some other studies that also employ data-driven methods [15] [16], the degradation process is firstly divided into a fixed number of time windows, and then a mapping function is learned from input features extracted from time windows to corresponding capacity changes so that the whole future capacity fade trajectory can be forecasted. The battery lifetime is then obtained when the forecasted future capacity

reaches a predefined EoL threshold. Additionally, the uncertainty associated with battery lifetime prediction is quantified as the width of a confidence interval in some of the aforementioned studies, with a Gaussian distribution of battery lifetime assumed. However, the Gaussian distribution assumption of battery lifetime does not necessarily hold in practice [12].

The objective of hybrid methods is to leverage the strengths of several different models to provide superior lifetime prediction performance. Typically with the hybrid data-driven and model-based approach, a degradation model is incorporated into a recursive Bayesian filter framework (e.g., particle filter [17] [18], Kalman filter [19]), in which the data-driven model can be used for estimating the internal states of the battery from measurements [17], predicting the future measurements [17] [19] and as a replacement of a degradation model [18] in the model-based prediction case. The hybrid data-driven and model-based approach may potentially provide better prediction performance but is challenging to use in online applications due to its high computational effort.

Although advanced data-driven methods provide superior lifetime prediction performance, with possibly quantified uncertainty, in spite of minimum knowledge of underlying battery degradation mechanisms, the interpretability of learned data-driven models that are used for battery lifetime prediction is still under-explored in the existing literature. It could, though, facilitate degradation diagnostics and prognostics, and discoveries of new degradation mechanisms and their resulting pathways. Similarly, a generalized battery capacity knee identification method that leverages battery degradation prior knowledge to improve knee identification performance remains underdeveloped.

This thesis work aims to narrow the gaps indicated above by addressing the following research problems:

- battery lifetime prediction using early degradation data.
- battery capacity knee identification.

1.2 Thesis Contributions

This thesis is based on three papers and major contributions are summarized as follows:

- With the advantage of not assuming any specific distribution of cycle life, a quantile regression forests (QRF) model is introduced to provide cycle life range prediction with uncertainty quantified as the width of the prediction interval, in addition to point predictions with high accuracy. The learned QRF model is later used in an example application for selecting the high-cycle-life fast-charging protocol (see Paper A).
- To interpret the learned QRF model for cycle life prediction, two model-agnostic interpretation techniques are employed to first rank individual feature importance and then quantitatively show the marginal effect each feature has on the predicted battery cycle life. The rationalization of the underlying battery degradation process agrees with what has been revealed by these two interpretation techniques (see Paper A).
- To make model selection easier prior to online deployment, a machine learning pipeline is proposed for automating the process of producing the best model among commonly-used machine learning models for both battery cycle life point prediction and range prediction. Simulation results show that quantile regression models are not only capable of providing cycle life point prediction with high accuracy but also cycle life range prediction with high reliability (see Paper B).
- As the concept of the capacity knee is largely related to the degradation rate of the capacity fade, we first use approximated curvature to measure the rate of change of degradation rate in discrete time, and then formulate the knee identification problem as an unsupervised time series segmentation problem given an assumption of three discrete states in the whole degradation process. By adopting the regime extracting algorithm, the locations of the state changes are found as the knee-onset and the knee itself on the capacity fade curve, respectively (See Paper C).

1.3 Thesis Outline

This thesis is divided into two parts, Part I provides motivation, an introduction, methodology, and conclusions; Part II includes papers on which this thesis is built. To be more specific about Part I,

- Chapter 1 motivates research problems that this thesis is addressing and provides background information;
- Chapter 2 reviews rechargeable lithium-ion automotive batteries;
- Chapter 3 reviews existing battery cell degradation diagnostics methods, capacity knee identification methods, and lifetime prediction methods;
- Chapter 4 provides a summary of papers on which this thesis is built;
- Chapter 5 concludes this thesis and recommends future work.

CHAPTER 2

Lithium-ion Batteries

Various types of rechargeable batteries have been used in electric vehicles (EVs). For example, lead-acid batteries are commonly used for starting, lighting, and ignition applications, nickel-metal hydride batteries have been used as the primary energy storage choice for hybrid electric vehicles (HEVs). Today lithium-ion batteries are the exclusive energy sources for the propulsion of plug-in hybrid electric vehicles (PHEVs) and battery electric vehicles (BEVs) [20].

Lithium-ion batteries have been widely used in different types of EVs, thanks to their outstanding characteristics, such as high energy and power density, high Coulombic efficiency (99%) and energy efficiency (up to 95%), as well as a wide range of different power to energy ratios [2]. In this chapter, only lithium-ion batteries used in different types of EVs will be discussed.

2.1 Battery Cell Materials

The lithium-ion battery cell consists of four major components [21]:

- The anode (or negative electrode): the reducing electrode that releases

electrons to the external circuit and undergoes oxidation during the electrochemical reaction (discharge).

- The cathode (or positive electrode): the oxidizing electrode that accepts electrons from the external circuit and undergoes reduction during the electrochemical reaction (discharge).
- The electrolyte: the ionic conductor that provides the medium for the transfer of ions between the negative and positive electrode inside the cell.
- The separator: a porous membrane that is placed between the negative and positive electrode to prevent short circuits.

The electrochemical reaction of a cell during discharge is also illustrated in Fig. 2.1.

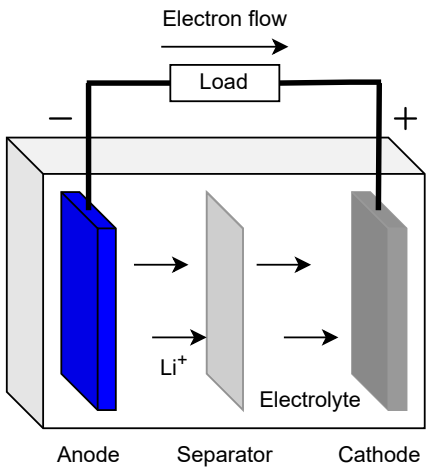

Figure 2.1: Electrochemical reaction of a cell during discharge [21].

Anode Materials

Currently, state-of-the-art negative electrode materials used in lithium-ion batteries are synthetic and artificial graphites, natural graphites, and amor-

phous carbons [22]. Synthetic and artificial graphites are the most commonly used negative electrode materials in EVs due to their high levels of purity and consistent quality [2]. A small amount of silicon is also added to the anode in some commercial cells in order to increase cell energy further [22]. Lithium titanate is used in some commercial cells, which makes these cells more suitable for power applications, such as in electric buses [22]. Lastly, lithium metal is considered as an ideal anode material for applications, especially in all-solid-state batteries that utilize ceramic or polymetric electrolytes [2].

Cathode Materials

The cathode has been a bottleneck in terms of specific capacity since the commercialization of lithium-ion batteries. The most widely used positive electrode materials in EVs are lithium nickel manganese cobalt oxide (NMC), lithium manganese oxide (LMO), lithium nickel cobalt aluminum oxide (NCA), and lithium iron phosphate (LFP) [2]. Fig. 2.2 illustrates the energy density versus the specific energy of different cell chemistries at the positive electrode. Generally, a higher amount of Ni content implies a higher capacity. Therefore, a commonly employed strategy to maximize the energy content of NMC cathodes is to maximize the Ni content [23]. With outstanding rate capability at an affordable price, LMO is often blended with Ni-rich layered cathodes with the aim of increasing power density and safety [2]. While reducing the LMO content in cathode material blends will improve energy density further [2]. Currently, the state-of-the-art cathode materials are NCA which has the advantage of capacity retention, along with NMC-532 and NMC-622 [2]. Despite the relatively low volumetric capacity of LFP, its robustness offers a promising prospect in heavy-duty vehicle applications like buses and trucks, where its extended cycle life and excellent rate capability become advantageous [2].

Electrolytes

Commercial lithium-ion batteries generally contain electrolytes that are based on lithium hexafluorophosphate ($LiPF_6$) as conducting salt [24]. This conducting salt is dissolved in mixtures of cyclic and linear organic carbonate solvents which typically consist of ethylene carbonate (EC), dimethyl carbonate (DMC), diethyl carbonate (DEC), ethyl methyl carbonate (EMC) and propylene carbonate (PC) [25]. In carbonate-based electrolytes, the primary

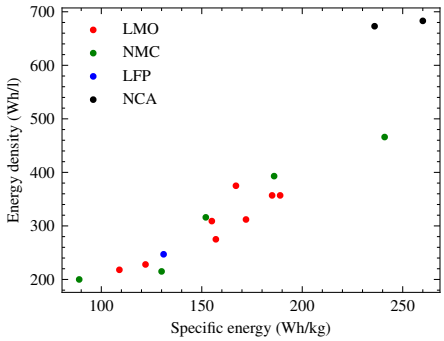

Figure 2.2: Energy density versus specific energy of different cell chemistries at the positive electrode [2].

cost driver is $LiPF_6$ whose price depends on the purity and composition [2].

Separators

When employing a liquid organic electrolyte in a lithium-ion battery cell, it is essential to place a porous membrane as a separator between the positive and negative electrode to avoid any electrical short circuits [26]. Currently, there are four main types of separators that are available, which vary in terms of chemistry and production process, i.e., microporous membranes, non-woven mats, ceramic-coated separators, and solid inorganic, polymeric or hybrid electrolytes [26]. The solid electrolytes that function as both a separator and an electrolyte are considered safer than the conventional setup of a separator and a liquid organic electrolyte [27]. Therefore, solid electrolytes are gaining more and more attention. However, major challenges such as high production costs remain to be overcome by technology breakthroughs [27].

2.2 Battery Cell Characterization Tests

The continuous advances in lithium-ion cell technology (e.g., new electrode material, cell design, and manufacture) motivate the need for non-invasive cell characterization tests in the lab, in which three parameters are typically measured, i.e., capacity, internal resistance/impedance, and open circuit volt-

age (OCV) [28]. These tests can be used for both characterizing initial cell performance and cell-to-cell variations, and tracking the evolution of cell performance (i.e., cell degradation) through reference performance tests (RPTs) [29]. Notably, there are different ways of conducting characterization tests, and consequently, capacity, resistance, and OCV measurements may vary from one to another depending on the specific experimental setup.

Capacity Tests

Depending on ambient temperature and charge-discharge C-rate, the measured capacity of the same cell can be different, which captures different degradation information. Note that C-rate is defined as the current value which discharges a battery from a fully charged state to a fully discharge state in one hour [28]. Lower C-rates ($\leq$ C/10) provide thermodynamic information through loss of lithium inventory (LLI) and loss of active material (LAM), which enables incremental capacity analysis (ICA) [30] and differential voltage analysis (DVA) [31]. Higher C-rates ($\geq$ C/10) provide a combination of both thermodynamic information and kinetic information through impedance growth.

The experimental data obtained from constant-current capacity tests can be used for two purposes, i.e., modeling the battery degradation process using physics-based models, (semi-)empirical models, or machine learning models; understanding the causal relationship between battery usage profile (e.g., static cycling aging, dynamic cycling aging, storage aging) and capacity fade.

Internal Resistance/Impedance Tests

Intrinsically, the power capability of a battery cell is associated with its impedance characteristics. The impedance of a cell determines the voltage response to a given current load, characterized by its amplitude, frequency, and time duration [28]. The resistive part (i.e., the real part of the complex impedance) directly contributes to the dissipative heat generation of a cell, and the resulting cell temperature increase while in use [32]. Moreover, the resistive part consists of pure ohmic resistance, charge transfer resistance, and entropy change [28]. Therefore, the cooling system design mainly depends on the resistive values of cell impedance.

Two well-established cell impedance measurement techniques are introduced

here.

Pulse Power Tests

The pulse power tests were initially proposed by the United States Advanced Battery Consortium (USABC) [33], and are also known as hybrid pulse power characterization (HPPC). The tests measure the voltage response to a square-wave current load that is applied to a cell. The resistance is then obtained as the ratio of the measured voltage response to the applied current, which consists of three parts, i.e., ohmic resistance, charge transfer resistance, and polarization resistance [28]. The experimental data obtained from pulse power tests can be utilized to parameterize equivalent circuit models (ECMs) that are used to estimate the state of power (SoP) [34], and state of charge (SoC) of a cell [35]. Moreover, pulse power tests have also been employed to parameterize cell electro-thermal models [36], and characterize cell degradation through resistance rise [37].

Electrochemical Impedance Spectroscopy Tests

Electrochemical impedance spectroscopy (EIS) tests were introduced to investigate the electrochemical behavior of a cell over a wide range of frequencies [28]. In EIS tests, a small amplitude sinusoidal potential as input stimulus is applied to an electrochemical cell. As a result, the current response in a linear or pseudo-linear system is a sinusoid with the same frequency but with a different amplitude and a phase shift relative to the input. The current response and the input voltage are then used to calculate the impedance of the cell in the frequency domain. The impedance spectrum of a cell is typically represented by a Nyquist plot that consists of the real and imaginary parts of the impedance. The experimental data obtained from EIS tests can be used to characterize the electrochemical dynamics of a cell [38], estimate cell temperature [39], parameterize ECMs [40], and identify cell degradation mechanisms [41] [42].

OCV Tests

OCV tests measure the equilibrium voltage of a cell as a function of the SoC. At the cell level, the OCV curve is defined by OCV curves of two electrodes, the loading ratio between two electrodes, and an SoC offset between two

electrodes [43], while at the electrode level, the OCV curve is defined as the potential difference between the electrode and the reference [44]. An example of the impact of changes of loading ratio and SoC offset on capacity loss is given in Ref. [28].

To understand the OCV curves, the definition of SoC is of equal importance. However, the definition of SoC in the literature varies from one to another. There are mainly four different SoC definitions in the literature, which are listed as follows:

- USABC definition: The ratio of the Ampere hours remaining in a cell at a given rate to the rated capacity under the same specified conditions [33].

- Thermodynamic definition: The ratio of the remaining intercalation sites for lithium ions to the total number of available intercalation sites [45].

- Low-rate definition: The ratio of the remaining exchangeable lithium ions to the maximum number of exchangeable lithium ions at a low rate (e.g., C/25) for a given potential window. [46].

The low-rate definition is generally applicable to half cells, full cells, and battery packs.

Two commonly used OCV measurement techniques are introduced here.

Galvanostatic Intermittent Titration Technique

One test procedure is proposed by USABC, i.e., after resting for one hour, the cell is discharged at 10% SoC increments and the voltage is recorded [47]. More accurate measurements of OCV can be achieved by reducing the SoC increments to less than 10% and increasing rest periods to longer than one hour [28]. However, to avoid prohibitively long test time, there is a trade-off between the accuracy of OCV measurements and the cost of longer rest periods and shorter SoC increments. The OCV curves obtained from GITT tests can be used to characterize cell performance (e.g., rate capabilities of a cell [48]), study OCV hysteresis of different chemistries [49], parameterize OCV models [44] and ECMs [50], and identify degradation mechanisms [51].

Pseudo-OCV Tests

Depending on the required accuracy of OCV measurements, the long test time of a GITT test may hinder its wide adoption in battery studies. In this regard, pseudo-OCV provides an alternative solution to obtain OCV curves with significantly less time, i.e., cycling a cell at a low charge and discharge rate (typically $\leq$ C/25) and then averaging the charge and discharge curve in order to address the cell hysteresis issue [30] [52]. The reason for the low current rate is to reduce the kinetic effects, electrode polarization, and heat generation due to ohmic resistance. The OCV curves obtained from pseudo-OCV tests can be used to identify and quantify degradation modes after taking their derivatives (e.g., incremental capacity analysis [30], differential voltage analysis [31]), improve model-based voltage estimation accuracy by including hysteresis effects [53], and estimate SoC [54].

2.3 Battery Cell Degradation Mechanisms and Modes

As a result of an intricate interplay of various physical and chemical degradation mechanisms, the performance of lithium-ion battery cells degrades, for example, cell capacity fade and cell resistance/impedance rise. Inside the cell, degradation mechanisms occur at different components, i.e., the anode, the cathode, the electrolyte, the separator, and the current collectors [55] [3]. However, considering the influence of the electrolyte and its own degradation mainly occurs in interaction with the electrodes, the degradation mechanisms of the cell are therefore discussed at the anode and cathode separately.

Degradation Mechanisms at the Anode

Graphite, an allotrope of carbon, is the primary material used for anodes in lithium-ion battery cells (see Subsection 2.1). As a result, degradation mechanisms at the graphite anode have been better studied than those at the cathode in the literature. However, it is generally difficult to generalize those degradation mechanisms that have been reported in the literature as each lithium-ion battery cell has its own cell design (e.g., chemistry and geometry) [43] and manufacture [56], which intrinsically has an impact on the

cell degradation. Moreover, most of the literature focuses on the whole cells without investigating the anode and cathode respectively. Therefore, only the dominant degradation mechanisms at the anode are listed as follows [55]:

- solid electrolyte interphase (SEI) growth;
- SEI decomposition;
- electrolyte decomposition;
- binder decomposition;
- graphite exfoliation;
- lithium plating/dendrite formation;
- loss of electric contact;
- electrode particle cracking;
- corrosion of current collector.

Degradation Mechanisms at the Cathode

Similarly, cathode materials (see Subsection 2.1) have a significant impact on the performance of lithium-ion battery cells. In the literature, lithium manganese oxides with spinel structure and lithium nickel cobalt mixed oxides with layered structures have been intensively studied. The dominant degradation mechanisms at the cathode of these materials are listed as follows [55]:

- electrolyte decomposition;
- binder decomposition;
- loss of electric contact;
- corrosion of current collector;
- structural disorder;
- electrode particle cracking;
- transition metal dissolution/dendrite formation.

The aforementioned dominant degradation mechanisms that occur at the anode or the cathode are illustrated in Fig. 2.3. Moreover, these degradation mechanisms are clustered into three degradation modes, which have unique and measurable effects on the OCV of lithium-ion cells and electrodes [3]. The three degradation modes are as follows:

- loss of lithium inventory;
- loss of active material of the negative electrode ($\mathrm{LAM_{NE}}$);
- loss of active material of the positive electrode ($\mathrm{LAM_{PE}}$).

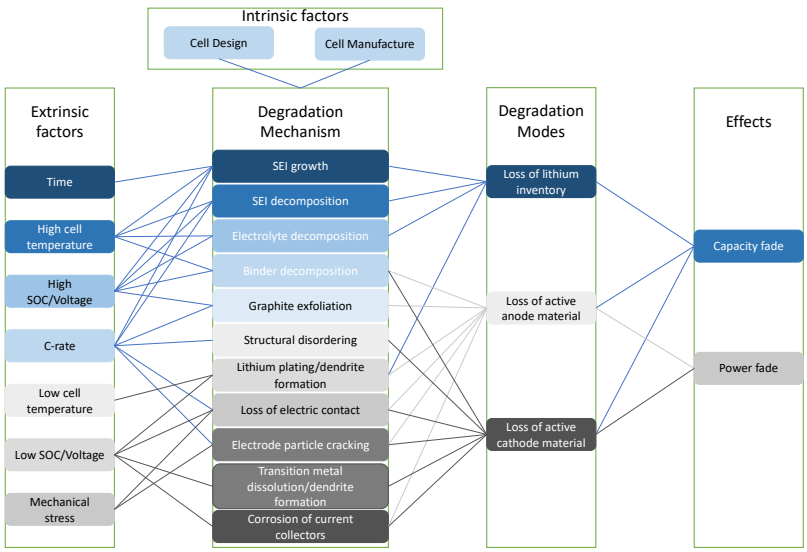

Figure 2.3: Cause and effect of dominant degradation mechanisms and their associated degradation modes [3] [55] [43].

CHAPTER 3

Methods

In this chapter, battery cell degradation diagnosis methods, capacity knee identification methods, and lifetime prediction methods are reviewed.

3.1 Battery Cell Degradation Diagnostics

The goal of battery cell degradation diagnostics is to identify and quantify degradation modes in a lithium-ion battery cell. Currently, diagnostic methods can be divided into three categories, i.e., post-mortem analysis, model-based analysis, and curve-based analysis [57].

Post-mortem Analysis

Post-mortem analysis involves safely disassembling aged battery cells in a well-controlled clean environment and then carefully examining each of their components through material analysis, in order to identify and then quantify dominant degradation mechanisms [58] [59] [60]. According to the physico-chemical aspect, the post-mortem analysis can be further divided into three subcategories,

Morphology Analysis

The morphology analysis is to examine the morphology of the electrode surface. Depending on different resolution requirements, optical microscopy [61], scanning electron microscopy [62], and transmission electron microscopy [63] are commonly used in morphology analysis.

Composition Analysis

The composition analysis is to examine the element composition of active materials and their concentration distribution on the electrode surface and at different depths. Techniques such as energy dispersive X-ray spectroscopy [59], X-ray photoelectron spectroscopy [64], inductively coupled plasma- atomic emission spectroscopy [65] have been reported to be used in the composition analysis.

Structure Analysis

The structure analysis is to examine the crystal structure on the surface, for which X-ray diffraction is commonly used [63].

Model-based Analysis

The model-based diagnostic methods mainly involve electrochemical models derived from first principles using porous electrode theory (e.g., the Doyle-Fuller-Newman (DFN) model [66]) and equivalent circuit models [67].

Electrochemical Models

Some of the electrochemical model parameters are important health indicators, which are closely related to the degradation of lithium-ion battery cells [68]. Therefore, identifying these aging parameters in electrochemical models and then correlating them with underlying degradation mechanisms and modes would facilitate battery degradation diagnosis. Some aging parameters in electrochemical models that have been identified in the literature are the volume fraction of active material at the anode [69], SEI resistance [69], the resistance of deposit layer [69], the diffusion coefficient of electrolyte [69], the diffusion coefficient of the solid phase [68], the electrochemical reaction rate

constant [68], the cathode particle surface area [70], stoichiometry limits [70], and porosities of the cathode, separator, and anode [70].

Equivalent Circuit Models

The experimental data obtained from electrochemical impedance spectroscopy (EIS) tests contains rich information about cell degradation caused by internal resistance/impedance (see Subsubsection 2.2) and can therefore be used to identify cell degradation mechanisms [41] [42].

Typically, the impedance spectrum of a cell is represented by a Nyquist plot, which is often modeled by an equivalent circuit model (ECM). As a commonly-used model, the adapted Randles-equivalent circuit model (AR-ECM) is illustrated in Fig. 3.1, in which changes of four resistances (ohmic R_{ohm}, SEI R_{SEI}, charge-transfer R_{ct}, and Warburg R_w) are tracked to identify and quantify degradation mechanisms, i.e., ohmic resistance increase (ORI), loss of lithium inventory (LLI), and loss of active material (LAM) [41].

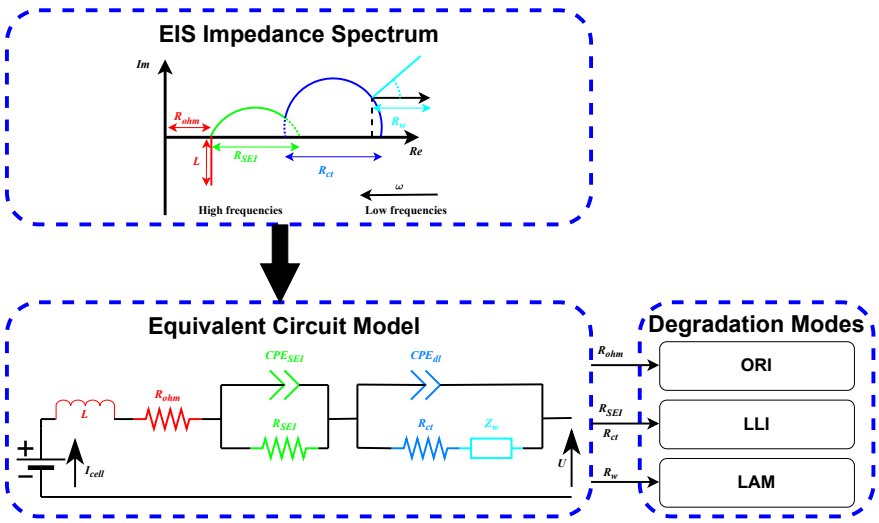

Figure 3.1: Using EIS impedance spectrum and ECM to identify degradation mechanisms [41].

Curve-based Analysis

As it is illustrated in Fig. 3.2, most physics-based models generally capture the most dominant degradation mechanisms at the micro-scale [71] or even nano-scale [72]. However, previous studies have illustrated that meso-scale and macro-scale inhomogeneities in the structure of the electrodes caused by cell manufacture can have a significant impact on safety (e.g., thermal runaway [73]) and cell degradation in the long term (e.g., capacity knee occurrence [56]), which may not be captured by bottom-up physics-based models.

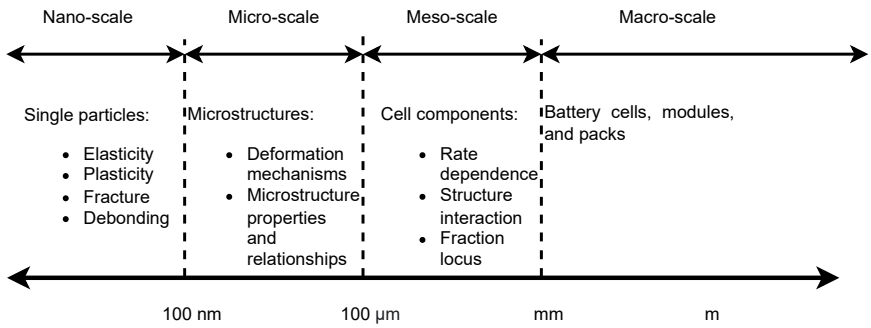

Figure 3.2: Mechanical properties of lithium-ion batteries at different length scales [74].

Curve-based analysis methods that utilize measurements from cell characterization tests (see Section 2.2) provide an alternative solution to cell degradation diagnosis. Examples of utilizing different measurements from cell characterization tests are:

- EIS impedance spectrum [42];
- discharge voltage curve [12];
- pseudo open circuit voltage (OCV) [3];
- derivatives of OCV or cell capacity, such as incremental capacity analysis (ICA) [30] and differential voltage analysis (DVA) [31].

3.2 Capacity Knee Identification Methods

As a result of a complex interplay of various physical and chemical degradation mechanisms (see Section 2.3), the performance of lithium-ion battery cells degrades over their lives, for example, capacity fade and resistance/impedance rise. In some cases, sudden acceleration of capacity fade (so-called capacity knee) is observed to occur, which results in accelerated performance degradation and even safety issues of a cell [4]. Therefore, avoiding or at least delaying the occurrence of the knee is essential to guarantee a long battery lifetime within safety constraints.

The IEEE Standard 485$^{\text{TM}}$-2020 defines the capacity knee as the transition when "the capacity slowly declines throughout most of the battery's life, but begins to decrease rapidly in the latter stages" [75]. However, this definition only qualitatively describes what a capacity knee is, and therefore, is not applicable to knee identification. A mathematical definition of curvature is provided in Ref. [76], in which the knee is defined as the point of maximum curvature where a continuous function deviates the most from a straight line. For a continuous function f, the curvature $\kappa(x)$ of f at any point, is defined as

$$\kappa(x) = \frac{f''(x)}{(1 + f'(x)^2)^{1.5}}. \tag{3.1}$$

The curvature ((3.1)) at a point x can be either positive, negative, or 0, depending on the second derivative of the function f.

Although a knee can be mathematically well defined for continuous functions, it is challenging to identify the knee using Eqn (3.1) in practice. The reasons are as follows:

- Realistic battery capacity data is obtained as discrete data, either from direct measurements in the lab (see Subsection 2.2) or from real-time estimations in the field. Moreover, battery capacity data that is estimated in the field is also expected to be noisy due to highly varying battery usage profiles and fluctuating environmental conditions, and sometimes unevenly sampled if capacity data is only obtained from periodic diagnostic cycles [77].

- The curvature calculation using Eqn (3.1) requires both the first and second derivatives. Numerical differentiation amplifies the noise in the capacity data, which is especially problematic in the field where capacity

data may be noisier than in the lab. Interpolating or extrapolating capacity data due to uneven sampling may introduce additional errors that are subsequently amplified by numerical differentiation as well.

Therefore, it is not possible to identify the knee as the point of maximum curvature using Eqn (3.1) in practice.

To date, only few studies in the battery literature have attempted to identify the knee on the capacity fade curve, both in offline scenarios [76] [78] [79] [16], and online scenarios [76] [80]. In offline scenarios, the knee is identified given the complete capacity fade data of a cell with knee occurrence; while in online scenarios, the knee has to be identified on the fly whenever new capacity fade data is available during battery usage. A curvature-inspired algorithm is proposed in Ref. [76] for both online and offline knee identification, which is applicable to a wide range of systems including battery capacity knee identification. In Ref. [78], an empirical model that characterizes the capacity fade curve with consideration of knee point occurrence is first proposed. After fitting the empirical model with experimental data, the knee is identified offline as the intersection of two tangent lines at two points, i.e., the points with minimum and maximum absolute slope, respectively. In Ref. [80], a strip-shaped safety zone is first learned from experimental data (the height of IC curve versus cycle number), and the knee can be identified online as the last cycle of four consecutive cycles beyond the safety zone using the quantile regression method. In a more recent work [79], by directly fitting the Bacon and Watts model [81] to the complete capacity fade data, the knee is identified offline as the intersection of two straight lines. Finally, in Ref. [16] the capacity fade gradients at the early and late life are first fitted using linear regression, and their angle bisector is then calculated. The knee is identified offline as the intersection of their angle bisector and the complete capacity fade curve.

3.3 Lifetime Prediction Methods

Historically, battery lifetime prediction has been restricted to using relatively small lab data under well-controlled operating conditions. This section first reviews lifetime prediction methods using lab data and then discusses the challenges of lifetime prediction methods using field data, and potential second-life applications.

Lifetime Prediction Methods Using Lab Data

In the context of an electric vehicle, the reduction of cell capacity can be translated into the reduction of maximum driving distance, while the impedance characteristics of a cell have a direct impact on the power capability. Therefore, cell capacity and cell resistance are commonly used for defining the state-of-health (SoH) of a lithium-ion cell. In this thesis, cell capacity as the primary SoH metric is investigated.

The battery degradation models in lifetime prediction methods using lab data can be roughly divided into four categories, i.e., physics-based models, semi-empirical models, empirical models, and machine learning models.

Physics-based Models

Broadly defined, physics-based models include electrochemical models derived from first principles using porous electrode theory (e.g., the Doyle-Fuller-Newman (DFN) model [66], single particle model (SPM) [82]), and equivalent circuit models (ECMs) [67].

The electrochemical models may be coupled with multiple mechanical or chemical degradation models that capture underlying degradation mechanisms, such as the SEI growth model [83] [84], lithium plating model [83] [85] [86] [87], and particle cracking [83] [71] [88]. After model parameterization and validation with experimental data of a specific cell, these models could be used to forecast the future capacity fade trajectory via model simulation under certain operating conditions [89] [90]. Then the battery lifetime is predicted as the time when the forecasted capacity reaches a predefined EoL threshold.

ECMs are lumped-element models with fewer parameters than those in electrochemical models, with the aim of capturing the electrical behavior of battery cells [91]. To achieve lifetime prediction, observers are first designed to estimate the internal states (e.g., state-of-charge (SoC) and internal resistance) of ECMs. Then the estimated internal states are used to identify an empirical health degradation model, which is lastly used to predict battery lifetime [92] [93] [94].

(Semi-)empirical Models

To achieve onboard battery lifetime prediction in a battery management system (BMS), empirical or semi-empirical models may be the most commonly used model type to capture the direct relationship between the operating conditions and the battery SoH with affordable computational cost. It is common in the literature that only cycling aging as the function of cycle number, or equivalent full cycle number, or Ah-throughput, is considered in empirical models, such as polynomial [95], exponential [9], logarithmic [10], and hybrid [96] models, while semi-empirical models often consider both calendar aging and cycling aging with square-root-of-time dependency due to SEI growth [97] and Arrhenius temperature dependency [98]. To develop a (semi-)empirical model for a specific type of lithium-ion cell, relevant stress factors need to be first identified for both calendar aging and cycling aging, for example, storage temperature [99] [100] [101], storage voltage [99], and storage SoC [100] [101] in calendar aging; charge and discharge C-rate [102] [100], average voltage [99], average SoC [100] [101], depth-of-discharge (DoD) [99] [102] [101], and ambient temperature [102] [100] [101] in cycling aging.

After parameterizing and validating these (semi-)empirical models with experimental data of a specific cell, they are incorporated into a recursive Bayesian filter framework, such as a particle filter [9] [10] [11]. The model parameters are then recursively updated with onboard measured capacity data. Lastly, the battery lifetime is predicted by identifying the point at which the predicted capacity reaches a predefined EoL threshold.

Machine Learning Models

Generally, machine learning models that have been used for battery lifetime prediction in the literature can be either non-probabilistic or probabilistic. Non-probabilistic machine learning models include autoregression based models [103] [104], elastic net [12], support vector regression [105] [106], random forest regression [107], gradient boosting regression tree [108], long short-term memory [109] [110], and recurrent neural network [111]. Probabilistic machine learning models include Gaussian process regression [15], relevance vector machine [112] [113], quantile regression forest [114], and Bayesian Neural Network [115].

Different from the aforementioned explicit health degradation models that

capture the battery degradation process in the first place, some studies use machine learning models to learn a mapping function directly from input features, extracted from early degradation data to the battery lifetime, given a training set of input-output pairs [12] [13] [14]. While in some other studies, the degradation process is firstly divided into a fixed number of time windows, and then a mapping function is learned from input features extracted from usage patterns (e.g., the time spent within certain voltage, current, and temperature ranges) to capacity changes in the corresponding time windows [15] [16]. In this way, the whole future capacity fade trajectory can be forecasted, and the battery lifetime is then predicted as the time when the forecasted capacity reaches a predefined EoL threshold.

Lifetime Prediction Methods Using Field Data

In contrast to high-quality lab data under well-controlled operating conditions, battery lifetime prediction using field data that contains realistic battery usage profiles in first-life in-vehicle applications and second-life stationary applications faces several issues:

- The field data is expected to be noisy due to highly varying battery usage profiles and fluctuating environmental conditions, sometimes missing due to long time parking, and even corrupted because of faulty hardware or software.

- Unlike lab data, which is usually measured at the cell level, field data is measured at multi-levels, and therefore contains heterogeneity information within a module or a pack. It is therefore challenging for a pre-estimated model using lab data at the cell level to make accurate lifetime predictions using field data at multi-levels.

- To track the evolution of cell degradation throughout its life in the lab, regular reference performance tests (RPTs) are usually conducted, in which three parameters are typically measured, i.e., capacity, internal resistance/impedance, and OCV (see Section 2.2). However, the aforementioned ground truth is lacking in the field, which is required to verify lifetime prediction methods and validate lifetime prediction performance.

In comparison to a large number of studies about battery lifetime prediction using lab data, existing studies that achieve battery lifetime prediction using field data are scarce due to the lack of publicly available battery data in the field that includes battery usage data and health data. Nevertheless, machine learning models with input features that are independent of battery chemistry, usage, or history (e.g., from partial SoC windows [116] [16], or (partial) charging phase [117] [118] [119] [120] [121], or histogram data [15] [16] [122]) have shown great promise for battery lifetime prediction using field data. Alternatively, it is hypothesized that the hybrid data-driven and physics-based approach can potentially lead to performance improvement for lifetime prediction by leveraging the advantages of both. Parameterizing physics-based models requires a relatively small amount of data, which can be used to generate synthetic data for machine learning models. Health-related model parameters can be inferred online and provide physics-informed input features to machine learning models. In contrast, machine learning models are relatively easy to implement and can discover degradation mechanisms and their resulting pathways [114]. Therefore, machine learning models can be advantageous in enhancing and complementing the capability of physics-based models [123]. In this regard, some exemplary architectures include physics-informed neural networks [124], neural ordinary differential equations [125], and universal differential equations [126]. The development of these hybrid architectures is expected to accelerate thanks to their key advantage of extrapolating outside the training data much more accurately than machine learning models.

Lifetime Prediction Methods at Repurposing

With the accelerated uptake of xEVs, i.e., all types of pure electric vehicles (BEVs), hybrid electric vehicles (HEVs), and plug-in hybrid electric vehicles (PHEVs), in the global market despite the economic repercussions of the pandemic [127], a growing number of retired xEV batteries need to be handled properly. Instead of being recycled, an ideal scenario would be that a fraction of retired xEV batteries can be repurposed to less-demanding second-life applications in stationary battery energy storage systems (BESSs). At the same time, some revenue could be fed back to the xEV manufacturers, which may reduce xEV prices, and thereby make xEVs more competitive [128].

As one of the technical barriers of repurposing, battery lifetime prediction

for its intended second-life application faces two major issues:

- Although the Global Battery Alliance has taken the initiative to enable battery data sharing using the battery passport [129], historical battery data that may contain information on its degradation pathway caused by first-life usage may still be unavailable or limited due to, for example, proprietary reason. Without additional battery characterization tests (see Section 2.2), battery lifetime prediction for its intended second-life application may therefore be highly uncertain.
- The operating conditions for batteries will most likely differ significantly between their first lives in xEVs and their second lives in stationary BESSs. Representative load profiles may need to be extracted from different types of second-life applications and then used in lab tests to characterize second-life battery degradation in each type of second-life application.

CHAPTER 4

Summary of Included Papers

This chapter provides a summary of the included papers.

4.1 Paper A

Huang Zhang, Yang Su, Faisal Altaf, Torsten Wik, Sébastien Gros
Interpretable Battery Cycle Life Range Prediction Using Early Cell Degradation Data
Accepted in IEEE Transactions on Transportation Electrification, Dec. 2022 .

Battery lifetime prediction using early degradation data has many applications throughout the battery product life cycle. To address this research problem, the quantile regression forests (QRF) model is introduced in this paper to provide cycle life range prediction with uncertainty quantified as the width of the prediction interval, in addition to point predictions with high accuracy. The prediction performance of the QRF model is demonstrated on a publicly available battery dataset under realistic fast-charging protocols. Using two model-agnostic interpretation techniques, the two most important

input features are identified and their effect on predicted battery cycle life is quantitatively investigated. An important advantage of this method compared to others is that no assumptions on the statistical distribution have to be made, which otherwise easily corrupts uncertainty estimates.

4.2 Paper B

Huang Zhang, Faisal Altaf, Torsten Wik, Sébastien Gros
Comparative Analysis of Battery Cycle Life Early Prediction Using Machine Learning Pipeline
Accepted in 22nd IFAC World Congress, Yokohama, Japan, Jul. 2023 .

Lithium-ion battery system is one of the most critical but expensive components for both electric vehicles and stationary energy storage applications. In this paper, to produce the best model for both battery cycle life point prediction and range prediction (i.e., confidence intervals or prediction intervals), a pipeline-based approach is proposed, in which a full 33-feature set is generated manually based on battery degradation knowledge, and then used to learn the best model among five machine learning (ML) models that have been reported in the battery lifetime prediction literature, and two quantile regression models for battery cycle life prediction. The calibration and sharpness property of battery cycle life range prediction is properly evaluated by their coverage probability and width respectively. The experimental results show that the gradient boosting regression tree model provides the best point prediction performance, while the quantile regression forest model provides the best range prediction performance with both full 33-feature set and the MIT 6-feature set extracted by Severson et al. [12].

4.3 Paper C

Huang Zhang, Faisal Altaf, Torsten Wik, Sébastien Gros
Battery Capacity Knee Identification Using Unsupervised Time Series Segmentation on Degradation Curvature
Manuscript for submission, May. 2023 .

Battery capacity knee occurrence can have a significant impact on safety and profitability in battery usage. To address potential concerns regarding

possible capacity knee occurrence, a generalized capacity knee identification algorithm is proposed in this paper. With one experimental battery dataset of NMC cells, it is demonstrated that our proposed capacity knee identification algorithm successfully identified the knee while the state-of-the-art algorithm fails to identify it. The method also provides an estimate of the knee-onset, which can give an early warning of accelerated degradation and thus have significant economic impact in early battery replacement planning, and also battery repurposing to second-life applications. Lastly, the capacity knee-onsets and capacity knees that are identified using our proposed algorithm on lab data can also be used to systematically evaluate the knee prediction performance of both model-based methods and data-driven methods.

CHAPTER 5

Concluding Remarks and Future Work

Lithium-ion battery lifetime prediction using early degradation data offers many new possibilities throughout the battery lifecycle. However, it is challenging to provide accurate battery lifetime prediction with only small amounts of degradation data at early cycles due to the complex degradation process with the possible accelerated degradation rate at late cycles. At the same time, battery capacity knee occurrence can have a significant impact on safety and profitability in battery usage. In this thesis, we address the problem of battery lifetime prediction using early degradation data, the quantile regression forests (QRF) model is introduced for battery cycle life prediction with uncertainty quantified. Additionally, two model-agnostic interpretation techniques are employed to interpret the learned QRF model for cycle life prediction. Lastly, a machine learning pipeline is proposed for automating the process of producing the best model among commonly-used machine learning models for battery cycle life prediction. We also propose a generalized capacity knee identification algorithm that leverages battery degradation prior knowledge to address concerns arising from possible capacity knee occurrence in battery usage.

The prediction performance of the QRF model is demonstrated on a pub-

licly available battery dataset under realistic fast-charging protocols. In addition to battery cycle life point prediction with high accuracy, the QRF model also provides range prediction with uncertainty quantified as the width of the prediction interval, without assuming any specific distribution of battery cycle life. The learned QRF model is then used in an example application for selecting the high-cycle-life fast-charging protocol. Using model-agnostics interpretation techniques, the two most important input features are identified and their effects on predicted battery cycle life are quantitatively investigated. It is worth mentioning that these model-agnostic interpretation techniques can be easily used for other advanced machine learning models with the aim of finding important features and revealing battery degradation mechanisms. Further comparative analysis of battery lifetime prediction using the proposed machine learning pipeline shows that quantile regression models, such as QRF, are capable of providing both cycle life point prediction with high accuracy and cycle life range prediction with high reliability. The proposed capacity knee identification algorithm is benchmarked to the state-of-the-art knee identification algorithm on both experimental degradation data of both LFP and NMC cells, and synthetic degradation data. The capacity knee-onsets identified using our proposed algorithm can give a much earlier warning of accelerated degradation, which can have significant economic value in early battery classification, early battery replacement planning, and even early battery repurposing to second-life applications.

Finally, there are several improvements that can be made in future work. Firstly, the battery dataset used in this work is relatively small with cells tested at an ambient temperature of 30 ℃. A larger battery dataset with cells tested at ambient temperatures other than 30 ℃ is desired for validating the consistent prediction performance of the QRF model and effectiveness of the two interpretation techniques. Secondly, calendar aging during any dedicated resting period for a long time (e.g., during storage) is not considered in this work. Its impact on the prediction performance of the QRF model also needs to be investigated. Thirdly, it would be interesting to compare the prediction performance of a hybrid model that claims to leverage the strength of both physics-based degradation models and data-driven models with that of the QRF model only, as in this work. Fourthly, based on a relatively small lab dataset under well-controlled operating conditions, a full 33-feature set was manually extracted in this work. However, this feature set may be difficult

to extract from the field data due to the fact that battery usage in real-world applications has less controlled conditions, less accurate sensors, and possible data corruption. A new feature set that enables battery lifetime prediction in the field as a function of a realistic battery usage profile will be needed. Lastly, the effectiveness of our proposed capacity knee identification algorithm has been successfully validated on experimental datasets of two chemistry types under a well-controlled lab environment. It is recommended that battery degradation data obtained from dynamic cycling tests under realistic driving profiles should also be explored.

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