Modelling, State Estimation & Optimal Charging Control for A Lithium-Ion Battery

Changfu Zou

Submitted in total fulfilment of the requirements of the degree of

Doctor of Philosophy

Department of Mechanical Engineering
THE UNIVERSITY OF MELBOURNE

November 2016

Produced on archival quality paper.
Copyright © 2016 Changfu Zou

All rights reserved. No part of the publication may be reproduced in any form by print, photoprint, microfilm or any other means without written permission from the author.
LITHIUM-ION (Li-ion) batteries are well-suited chemistry technology for a myriad of applications such as portable electronic devices, electrified vehicles and energy storage in power systems. To satisfy consumers’ demanding requirements, the batteries are desirable to have high energy/power density, long life cycles and fast charge capability but provide these at affordable costs with safe and reliable operations. However, the current batteries are limited in one or more of these characteristics. This thesis leverages physics-based models and model-based estimation/control techniques to improve battery performances.

The first phase of this project focuses on battery model simplification. A physics-based Li-ion battery model capturing the coupled electrochemical, thermal, electrical and ageing dynamics is first developed. Through identification of disperse time-scales inherent in the initial model, a framework for PDE battery model simplification is proposed by using singular perturbations and averaging theory. The novelty arises from the systematic procedure for model simplification through which the underpinning assumptions can be explicitly stated. As a consequence, libraries of simplified battery models with interconnections are obtained and applicable for a range of applications. Simulations are conducted to validate the low order models against the initial model. The capability of the proposed modelling framework is demonstrated on a model-based evaluation procedure for charging strategies.

With appropriate battery models obtained, model-based algorithms are developed to estimate the battery state-of-charge (SOC) and state-of-health (SOH). First, a nonlinear es-
timator synthesised from an extended Kalman filter (EKF) and a reduced-order battery model from the proposed model simplification framework is designed to simultaneously estimate the SOC and SOH. Second, a novel multi-time-scale estimation algorithm is proposed for a class of singularly perturbed systems and is pertinently applied to estimate battery SOC and SOH in different time-scales. This reduces computational burden and avoids potential issues such as ill-conditioned gains and convergence. Illustrative results show that the multi-time-scale observer is effective in estimating both the SOC and SOH despite a range of common errors due to modelling, initialisation and measurements.

Finally, by utilising a computationally efficient model from the above model simplification framework and the developed multi-time-scale estimator, a novel model predictive control (MPC) based charging algorithm is proposed for a Li-ion battery to balance the competing objectives, i.e. battery’s health and charging time. Specifically, the charging strategy is formulated as a linear-time-varying (LTV) MPC problem for tracking SOC and SOH reference signals, in which constraints on the input current and battery internal states are explicitly considered. The capability of the proposed charging strategy is demonstrated via high fidelity simulations. As compared to its industry benchmark, i.e. the CCCV approaches, the MPC-based charging is found to have the ability to provide longer battery lifetime yet requiring less charging time. Furthermore, the proposed algorithm offers battery users the freedom to specify battery lifetime or charging time based on their requirements.

The developed modelling, estimation and control algorithms are very general so as to be applicable to different types of Li-ion batteries and cell chemistries.
Declaration

This is to certify that

1. the thesis comprises only my original work towards the PhD,
2. due acknowledgement has been made in the text to all other material used,
3. the thesis is less than 100,000 words in length, exclusive of tables, maps, bibliographies and appendices.

Signed,

Changfu Zou

November 2016
Acknowledgements

First and foremost, I would like to express sincere gratitude to my supervisors, Prof. Chris Manzie and Prof. Dragan Nešić, for their astute mentorship, thoughtful guidance, and unwavering support. Chris’s ability to approach research problems in a systematic way, find technical weaknesses, and provide constructive suggestions are uncanny. I am extremely thankful to him for teaching me the skills to conduct research. Moreover, his rigorous work attitude in revising paper drafts and presentation slides and great patience in advising students have been a great influence and a role model to me. Dragan’s outstanding knowledge in singularly perturbed systems and pertinent suggestions were really helpful to my Ph.D project. Meanwhile, his passion for research as well as fitness and cooking was inspirational. It has been a great honour to work with them.

Furthermore, I would like to thank my committee members, Dr. Rohan Shekhar and Dr. Julian de Hoog, for their insightful comments and encouragement, but also for the hard questions which incented me to widen my research. My sincere thanks also go to Dr. Abhijit G. Kallapur and Prof. Sohel Anwar, for their generous collaboration and assistance. The work of battery state estimation was collaborated with Abhijit. Without his guidance, I could not understand and implement estimation algorithms so smoothly and quickly.

I am grateful to colleagues in the research group for their friendship and enabling a great working environment. Particularly, I want to thank Will, Rohan, Tim, Kuan, and Gokul for all the support and interesting discussions, and thank Vincent, Yaqi, Chenyang, Kaixiang and Raymond for playing together and sharing the happiness and experience. I wish
all of you good luck in studies and work with a brilliant future.

Many thanks go to Prof. Scott Moura for offering me the great opportunity to visit University of California, Berkeley and his valuable advice, job recommendation and hospitality. With great research enthusiasm and excellent interpersonal skills and team management, he is my idol for academic/industrial success. I would like to give my acknowledgement to Satadru and Xiaosong in the team for their valuable advice and great friendship.

I have no words to show my deepest gratitude for my parents and sisters who have persistently cared about my study and life and inspired me whenever there is a difficulty. I am highly obliged to my wife for her love and support despite my heavily loaded and unbalanced routine. My heart resonates with her pleasant smile and I deeply appreciate the enjoyable life brought about by her optimistic philosophy and cheerful character.

Finally, I appreciate the excellent thesis template developed by John Papandriopoulos.
Contents

1 Introduction ................................................. 1
    1.1 Thesis Layout ......................................... 4
    1.2 Notational Conventions .............................. 6

2 Literature Review ....................................... 11
    2.1 Li-Ion Battery Background & Fundamentals .......... 11
        2.1.1 The Development of Li-Ion Batteries ............ 11
        2.1.2 Battery Cell Structure & Working Principle ....... 12
    2.2 Battery Cell Models .................................. 14
        2.2.1 First Principle Models ........................... 14
        2.2.2 Simplified Models: Physics-Based ................. 16
        2.2.3 Simplified Models: Equivalent Circuit Based .... 19
        2.2.4 Summary ......................................... 21
    2.3 Battery Estimation Techniques ........................ 21
        2.3.1 State-of-Charge Estimation ...................... 21
        2.3.2 State-of-Health and Charge Estimation ......... 24
    2.4 Battery Charging Strategies .......................... 27
<table>
<thead>
<tr>
<th>Section</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.4.1</td>
<td>Model-Free Charging Strategies</td>
<td>27</td>
</tr>
<tr>
<td>2.4.2</td>
<td>Model-Based Charging Strategies</td>
<td>29</td>
</tr>
<tr>
<td>2.5</td>
<td>Research Aims</td>
<td>31</td>
</tr>
<tr>
<td>3</td>
<td>Battery Modelling</td>
<td>33</td>
</tr>
<tr>
<td>3.1</td>
<td>Full-Order Battery Model</td>
<td>33</td>
</tr>
<tr>
<td>3.2</td>
<td>Model Reformulation in Hilbert Space</td>
<td>41</td>
</tr>
<tr>
<td>3.3</td>
<td>PDE Model Simplification Via Singular Perturbation Techniques</td>
<td>45</td>
</tr>
<tr>
<td>3.3.1</td>
<td>Theory For Model Simplification I: Models $\Sigma_f^2$ And $\Sigma_s^2$</td>
<td>46</td>
</tr>
<tr>
<td>3.3.2</td>
<td>Application To Battery Models: Stage I</td>
<td>50</td>
</tr>
<tr>
<td>3.3.3</td>
<td>Theory For Model Simplification II: Models $\Sigma_m^3$ And $\Sigma_s^3$</td>
<td>51</td>
</tr>
<tr>
<td>3.3.4</td>
<td>Application To Battery Models: Stage II</td>
<td>55</td>
</tr>
<tr>
<td>3.3.5</td>
<td>Validation of PDE model simplification</td>
<td>57</td>
</tr>
<tr>
<td>3.4</td>
<td>Model Simplification Via Spatial Dimension Reduction</td>
<td>66</td>
</tr>
<tr>
<td>3.5</td>
<td>Model Simplification Via Spatial Discretisation</td>
<td>68</td>
</tr>
<tr>
<td>3.6</td>
<td>Further Model Simplifications</td>
<td>74</td>
</tr>
<tr>
<td>3.7</td>
<td>Conclusion</td>
<td>79</td>
</tr>
<tr>
<td>4</td>
<td>Charging Strategy Evaluation</td>
<td>81</td>
</tr>
<tr>
<td>4.1</td>
<td>Introduction</td>
<td>81</td>
</tr>
<tr>
<td>4.2</td>
<td>Charging Strategy Evaluation Procedure</td>
<td>82</td>
</tr>
<tr>
<td>4.3</td>
<td>Simulation Results</td>
<td>84</td>
</tr>
<tr>
<td>4.4</td>
<td>Conclusion</td>
<td>87</td>
</tr>
<tr>
<td>5</td>
<td>Battery State Estimation</td>
<td>89</td>
</tr>
</tbody>
</table>
5.1 SOC and SOH Estimator: One Time-Scale ........................................... 89
  5.1.1 Battery Model for State Estimation .............................................. 90
  5.1.2 Battery Estimator Design ............................................................ 91
5.2 Estimation Result I ................................................................. 92
5.3 SOC and SOH Estimator: Multi-Time-Scale ........................................ 95
  5.3.1 Multi-Time-Scale Observer - Theory Development .......................... 95
  5.3.2 Multi-Time-Scale Observer - Application To Li-Ion Battery ............. 102
5.4 Estimation Result II ................................................................. 109
  5.4.1 Estimation Results in Fast Time-Scale (SOC) ................................. 109
  5.4.2 Estimation Results in Two Time-Scales (SOC + SOH) ....................... 109
5.5 Conclusion .............................................................................. 111

6 Model Predictive Control for Battery Optimal Charging .......................... 117
  6.1 Introduction ........................................................................... 118
  6.2 Optimal Charging Strategy ......................................................... 119
    6.2.1 Control-Oriented Battery Model .............................................. 119
    6.2.2 General MPC Tracking Problem Formulation ............................... 120
    6.2.3 Optimal Battery Charging Problem Formulation ............................ 122
    6.2.4 State Estimation ................................................................. 126
    6.2.5 Overall Problem Formulation .................................................. 127
  6.3 Simulation Results ................................................................. 128
    6.3.1 Simulation Model ................................................................. 128
    6.3.2 Explicit Problem Formulation .................................................. 129
    6.3.3 Simulation results for the proposed strategy ............................... 133
7 Conclusion and Future Work 139

7.1 Summary of Contributions 139

7.1.1 Systematic Battery Model Simplification 139

7.1.2 Model-Based Charging Strategy Evaluation Procedure 140

7.1.3 Multi-Time-Scale Estimation Algorithm and SOC/SOH Estimator 140

7.1.4 MPC-Based Battery Charging Strategy 140

7.2 Future Work 141

7.2.1 Stability Analysis for Battery Models 141

7.2.2 Stability & Robustness of Charging Strategy Control 141

7.2.3 Multi-Time-Scale Estimation Algorithm Extension 142

7.2.4 Development of High-fidelity SOH Models 143

7.2.5 Battery Model Parameter Identification 143

7.2.6 Experimental Validation 144

7.2.7 Extension of The Proposed Algorithms to Battery Packs 144

Bibliography 145
Chapter 1
Introduction

With the rapid increase of internal combustion engine (ICE) vehicles over the course of the 20th century, the effect of tailpipe emissions on air pollution has become a critical international concern [1, 2]. To alleviate this problem, increasingly stringent standards on vehicle emissions and fuel economy are placed, spurring the development of clean and cost-efficient automotive technologies [3]. With zero local emissions and high energy efficiency, electric vehicles (EVs) lie at the heart of future sustainable transport systems. The uptake of new EV registrations is fast-evolving. Until the year of 2015, more than one million EVs have been sold in the world [4]. However, compared with about one billion passenger cars on the road, the amount of EVs is still negligibly small. Table 1.1 shows the EV stock in 2015 and its targets to achieve in 2020 for 14 countries [4]. This again conveys that the EV share in the global market is extremely low, i.e. 3%. From the perspective of consumers, the main challenges for mass production of EVs include their high upfront cost and long charging time.

The battery system used for EV energy storage determines the charging time and is also a key component of the vehicle cost [5]. Thus, to realise an EV paradigm shift, it is necessary to investigate the battery technologies. Compared to lead-acid, nickel-cadmium and nickel-metal-hydride batteries, lithium-ion (Li-ion) batteries have high energy and power density, long lifetime, low self-discharge and no memory effect, and are thus recognised as well-suited chemistry technologies for automotive applications [6, 7]. However, the Li-ion batteries are expensive and their lifetime is greatly dependent on the applied charging strategies [8]. The inappropriately designed charging schemes, e.g. overcharging and
Table 1.1: Electric vehicle stock in 2015 and targets to 2020 based on 14 country’s commitments [4].

<table>
<thead>
<tr>
<th>Countries</th>
<th>2015 EV stock (thousand)</th>
<th>2020 EV stock target (million)</th>
<th>EV share in the total 2020 stock</th>
</tr>
</thead>
<tbody>
<tr>
<td>Austria</td>
<td>5.3</td>
<td>0.2</td>
<td>4%</td>
</tr>
<tr>
<td>China</td>
<td>312.3</td>
<td>4.6</td>
<td>3%</td>
</tr>
<tr>
<td>Denmark</td>
<td>8.1</td>
<td>0.2</td>
<td>9%</td>
</tr>
<tr>
<td>France</td>
<td>54.3</td>
<td>2.0</td>
<td>6%</td>
</tr>
<tr>
<td>Germany</td>
<td>49.2</td>
<td>1.0</td>
<td>2%</td>
</tr>
<tr>
<td>India</td>
<td>6.0</td>
<td>0.3</td>
<td>1%</td>
</tr>
<tr>
<td>Ireland</td>
<td>2.0</td>
<td>0.1</td>
<td>3%</td>
</tr>
<tr>
<td>Japan</td>
<td>126.4</td>
<td>1.0</td>
<td>2%</td>
</tr>
<tr>
<td>Netherlands</td>
<td>87.5</td>
<td>0.3</td>
<td>4%</td>
</tr>
<tr>
<td>Portugal</td>
<td>2.0</td>
<td>0.2</td>
<td>5%</td>
</tr>
<tr>
<td>South Korea</td>
<td>4.3</td>
<td>0.2</td>
<td>1%</td>
</tr>
<tr>
<td>Spain</td>
<td>6.0</td>
<td>0.2</td>
<td>1%</td>
</tr>
<tr>
<td>United Kingdom</td>
<td>49.7</td>
<td>1.6</td>
<td>5%</td>
</tr>
<tr>
<td>United States</td>
<td>101.0</td>
<td>1.2</td>
<td>2%</td>
</tr>
<tr>
<td>Total of all above markets</td>
<td>814.1</td>
<td>12.9</td>
<td>3%</td>
</tr>
</tbody>
</table>

overheating, can accelerate battery capacity fade or even lead to safety issues. Currently, the charging strategies are often conservatively designed to protect battery health but leading to long charging times. Furthermore, without accurate information about their internal dynamics, the batteries are now commonly underutilised with 20–50% energy capacity wasted [9]. Therefore, it is beneficial to develop advanced battery management algorithms, enabling desirable dynamic performance in a safe and reliable operating manner.

The system of a battery pack is commonly composed of two parts, namely, battery cells and battery management system (BMS). The battery cells are typically connected in series and/or in parallel to offer proper power and voltage to the motors. The BMS is responsible for charge and discharge management to ensure safety, reliability and dynamic performance [10, 11]. Specifically, these may involve capacity estimation, fast charging control, thermal management and cell balancing. With an efficient balancing module as described in [12] [13], the cells in a battery pack can potentially have the same operating behaviour. In light of this, it is of utmost importance to understand the characteristics of a single cell. The techniques and algorithms developed for a cell may be then extended
for battery packs to be achieved the best performance.

An efficient tool to investigate a battery cell is its mathematical models. However, it is technically challenging to develop appropriate models for applications such as simulations of battery behaviour and model-based controls. The system models need to contain electrochemical dynamics and reproduce battery behaviours such as the current-voltage response and state-of-charge (SOC) change [3, 14, 15]. With this information, one is able to predict vehicle driving range and to generate terminal conditions for battery charge and discharge operations. Aside from this, the temperature is an important state to include because its strong impact on electrochemical dynamics, e.g. temperature variations will incur change of the diffusion and conductivity coefficients [16, 17]. To predict battery’s lifetime and develop health-conscious charging strategies, the state-of-health (SOH) has to be modelled [8, 18]. The concatenation of all these dynamics is mathematically governed by a set of coupled nonlinear partial different equations (PDEs) [3, 8, 16]. This is computationally intractable for most model-based applications. Consequently, the model development often involves order reductions. Ideally, the derived models should be sufficiently simple but still capture the key system characteristics.

With reliable battery models obtained, one of the potential uses is to develop charging strategy evaluation algorithm. Currently, the constant-current constant-voltage protocol is widely used for battery charging [19, 20]. However, this approach may cause unduly battery SOH degradation or need substantial time to finish the charging task [19]. This motivates the development of improved charging strategies. Traditionally, a statistically significant set of experiments is conducted to explore and evaluate the newly proposed methods, e.g. in [21–23]. This will be greatly limited by extensive experimental time and cost. It would be desirable to develop a model-based algorithm for efficient evaluation of charging strategies.

For monitoring and control purpose, the battery state information is required during operations. Particularly, accurate knowledge of the SOC and SOH is critical for safe and efficient utilisation of the battery cell. While the SOC in an electrode is a function of its corresponding solid-phase Li-ion concentration [24], SOH is related to the aged capacity
For the EV battery system, usually, only the current, voltage and temperature measurements are available. In contrast, the internal states such as solid-phase Li-ion concentrations, electrolyte concentrations and aged capacity are impractical to measure outside of specialised laboratory environment [26, 27]. This necessitates the development of estimation techniques, and model-based approaches utilising the models proposed above may be applicable.

Ultimately, the fast and safe charging strategies can be designed for a Li-ion battery. For CCCV or other similar charging protocols such as constant-current constant power (CCCP) and multiple stage CC (MCC) [19, 22], a common feature is that the charging profiles are determined by some pre-defined current and voltage limits irrespective of battery in-situ physical and chemical characteristics. These model-free approaches are *ad-hoc* and their solutions are often conservative [3, 28]. Model predictive control (MPC) is a model-based control paradigm gaining increased interest in the last 30 years. Its performance in constraint and nonlinearity handling and optimal/sub-optimal control makes it an advanced control technique with significant and widespread impact on industrial process controls [29, 30]. With the computational power fast moving forward, MPC has been introduced in many applications, e.g. vehicle steering system control [31], and may be useful to solve the battery charge control problem.

The overarching goal of this thesis is to propose a systematic model simplification framework for a Li-ion battery and to develop model-based SOC/SOH estimation and optimal charging strategy. The following section summarises the work conducted to achieve this objective.

### 1.1 Thesis Layout

The remainder of this thesis is organised as follows. In Chapter 2, a critical review of battery modelling, estimation algorithms and charging strategies in the literature is presented. The initial battery model and its reduced order alternatives are first discussed, where attention is paid to the potential contribution of a systematic model simplification...
tation framework. The prevalent battery SOC/SOH estimation approaches and charging strategies are then introduced. This motivates possible contributions to be made in battery physical model-based estimator design and close-loop control. Subsequently, the research aims to be achieved in this thesis are outlined.

In Chapter 3, a physics-based high-fidelity Li-ion battery model capturing the electrochemical, thermal, electrical and ageing dynamics is proposed. Starting from this initial model, a framework for PDE model simplification is developed, resulting in families of simplified models that correspond to different applications. To demonstrate the capability of the proposed framework, in Chapter 4, an evaluation procedure based on low order models is proposed to quickly evaluate charging strategies regarding their impact on battery SOH and lifetime.

Chapter 5 presents estimator design for battery SOC and SOH based on reduced-order models from Chapter 3. In the first part, the SOC and SOH are estimated in the same time-scale by using an extended Kalman filter (EKF). To avoid potential issues in estimator design for singularly perturbed systems, the second part of this chapter proposes a multi-time-scale estimation algorithm for a class of systems with coupled fast and slow dynamics. This algorithm is applied to the battery problem for SOC and SOH estimation. The effectiveness of the proposed estimation algorithm is demonstrated by deploying EKF and unscented Kalman filter (UKF).

In Chapter 6, the proposed model-based optimal charging strategy is introduced. The fast charging problem is formulated as an MPC tracking problem to track given SOC and SOH reference signals. The prediction model taken from Chapter 3 and the state estimator from Chapter 5 are incorporated in the problem formulation. The constraints on system input and states for SOH protection are explicitly considered. The comparisons with the benchmark battery charging strategies including CCCV charging are performed. It is found that significant improvements in both the charging time and battery health can be achieved by the proposed strategy.

Finally, Chapter 7 summarises the contributions of this thesis and highlights potential future research opportunities.
1.2 Notational Conventions

To facilitate the description of dynamic models, estimators and controllers for a Li-ion battery in the subsequent chapters, common nomenclature is provided in the following.

**List of symbols**

\[ A \quad \text{Equivalent cross-sectional area of the battery cell} \]
\[ a \quad \text{Interfacial area at the surface of solid particles} \]
\[ C_s \quad \text{Li-ion concentration in solid particles} \]
\[ C_{ss} \quad \text{Li-ion concentration at the surface of solid particles} \]
\[ C_{loss} \quad \text{Li-ion concentration lost due to side reactions} \]
\[ C_{smax} \quad \text{Maximum possible solid-phase Li-ion concentration} \]
\[ C_e \quad \text{Li-ion concentration in the electrolyte} \]
\[ c_T \quad \text{Specific heat capacity} \]
\[ D_s \quad \text{Effective electric diffusion coefficient} \]
\[ D_e \quad \text{Effective ionic diffusion coefficient} \]
\[ F \quad \text{Faraday’s constant} \]
\[ h_T \quad \text{Heat transfer coefficient between the cell and ambient} \]
\[ I \quad \text{Applied current density at terminal} \]
\[ i_c \quad \text{Local current in the electrolyte} \]
\[ i_0 \quad \text{Exchange current density of the normal reaction} \]
\[ i_{0sr} \quad \text{Exchange current density of the side reaction} \]
\[ J \quad \text{Total Li-ion flux at the surface of solid particles} \]
\[ J_I \quad \text{Li-ion flux of the normal intercalation} \]
\[ J_{sr} \quad \text{Li-ion flux of the side reaction} \]
\[ L \quad \text{Total length of the battery cell} \]
\[ L^- \quad \text{The length of the negative electrode} \]
\[ L_{sep} \quad \text{The length of the separator} \]
1.2 Notational Conventions

$L^+$ The length of the positive electrode

$M_f$ Average molecular weight of SEI film

$Q_{sr}$ Capacity fade

$Q_{\text{max}}$ Maximum capacity of the battery

$R$ Universal gas constant

$R_f$ SEI film resistance

$R_p$ The length of the negative electrodes of the solid particles

$r$ Distance along the radius direction of solid particles

$T$ Temperature

$T_s$ Temperature at the surface of a battery cell

$T_f$ Ambient temperature

$U$ Open circuit potential of the normal reaction

$U_{sr}$ Open circuit potential of the side reaction

$V$ Terminal voltage

$x$ Distance along battery thickness direction

$\alpha_n$ Anodic charge-transfer coefficient

$\alpha_p$ Cathodic charge-transfer coefficient

$\alpha_{sr}$ Charge-transfer coefficient of side reaction

$\Delta_S$ Entropy change

$\eta$ Overpotential of the normal reaction

$\eta_{sr}$ Overpotential of the side reaction

$\gamma$ Transference number of the anion

$\kappa$ Effective ionic conductivity in the electrolyte

$\lambda_T$ Heat conductivity

$\mu_s$ Volume fraction of solid particles in the electrode

$\mu_e$ Volume fraction of electrolyte in the electrode/separator

$\Phi_s$ Electric potential in solid particles

$\Phi_e$ Electric potential in the electrolyte

$\rho$ Mass density of battery cell
\( \rho_f \) \hspace{1cm} \text{Average density of SEI film}

\( \frac{\partial U}{\partial T} \) \hspace{1cm} \text{Entropy change}

\( \sigma \) \hspace{1cm} \text{Effective electronic conductivity in the electrode}

\( \sigma_f \) \hspace{1cm} \text{Effective conductivity of SEI film}
1.2 Notational Conventions

List of acronyms

<table>
<thead>
<tr>
<th>Acronym</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>BMS</td>
<td>Battery management system</td>
</tr>
<tr>
<td>CCCV</td>
<td>Constant-current constant-voltage</td>
</tr>
<tr>
<td>C-rate</td>
<td>A normalised metric indicating the input current utilised for operations in amperes relative to battery maximum rated capacity in ampere-hours</td>
</tr>
<tr>
<td>ECM</td>
<td>Equivalent circuit model</td>
</tr>
<tr>
<td>EKF</td>
<td>Extended Kalman filter</td>
</tr>
<tr>
<td>EV</td>
<td>Electric vehicle</td>
</tr>
<tr>
<td>KF</td>
<td>Kalman filter</td>
</tr>
<tr>
<td>Li-ion</td>
<td>Lithium-ion</td>
</tr>
<tr>
<td>LTV</td>
<td>Linear time varying</td>
</tr>
<tr>
<td>MPC</td>
<td>Model predictive control</td>
</tr>
<tr>
<td>ODE</td>
<td>Ordinary differential equation</td>
</tr>
<tr>
<td>PDE</td>
<td>Partial differential equation</td>
</tr>
<tr>
<td>SMO</td>
<td>Sliding mode observer</td>
</tr>
<tr>
<td>SOC</td>
<td>State-of-charge</td>
</tr>
<tr>
<td>SOH</td>
<td>State-of-health</td>
</tr>
<tr>
<td>SPM</td>
<td>Single particle model</td>
</tr>
<tr>
<td>UDDS</td>
<td>Urban dynamometer driving schedule</td>
</tr>
<tr>
<td>UKF</td>
<td>Unscented Kalman filter</td>
</tr>
</tbody>
</table>
Chapter 2
Literature Review

MODEL development and model-based applications including estimator and controller design for a Li-ion battery underpin the aims of this project. This chapter provides an overview of relevant background literature in these areas. The background and fundamentals of a Li-ion battery such as its internal structure and working principles are first reviewed, followed by an introduction to battery first principle models and various model simplifications. Next, the prevalent state estimation algorithms for battery SOC and SOH are discussed. Then, the control algorithms for battery charging strategy development are provided, where the emphasis is placed on model-based control methodology that is used in this thesis to solve battery optimal charging problem. Finally, research aims to be achieved in this thesis are proposed based on research gaps identified from the literature.

2.1 Li-Ion Battery Background & Fundamentals

In the first part of literature review, the Li-ion battery background and fundamentals with respect to its historical development, cell internal structure and electrochemical principle are presented.

2.1.1 The Development of Li-Ion Batteries

The key motivation to develop Li metal-based anodes for a battery is that Li is the most electropositive and the lightest metal [32]. As a result, Li-ion batteries are favourable in
energy density, power density and coulombic efficiency as compared to their counterparts, namely, lead-acid, nickel-cadmium and nickel-metal-hydride batteries. The discovery of intercalation compounds is a significant breakthrough in the development of Li-ion battery technologies, providing potentials to develop high-energy rechargeable cells [7]. On such a basis, crucial advances have been achieved by using metal oxides in intercalation materials that are able to supply high capacities and voltages [33]. In this regard, families of compounds including Li$_x$Mn$_2$O$_4$, Li$_x$CoO$_2$, and Li$_x$PFeO$_4$, were proposed in the work such as [34]. A comparison study on electrode materials was provided in [6]. In contrast to Li-ion batteries with the liquid electrolyte, the solid polymer electrolyte was born to enhance battery operating safety [35]. The polymer technology offers shape versatility, flexibility and lightness in battery designs. Interested readers are referred to battery historical perspective, technology tendency and challenges summarised by Armand and Tarascon [7].

2.1.2 Battery Cell Structure & Working Principle

Before providing an overview of Li-ion battery cell mathematical models in the next section, a brief introduction to its internal structure and working principle is presented. An overview of cell geometry, materials, manufacturing and electrochemical reactions can also be found in the textbook on advanced batteries [36].

As illustrated in Fig. 2.1(a)-(b), a prismatic or cylindric Li-ion battery is composed of a number of electrodes, separators and current collectors. A minimum representative element containing battery physical components and the electrochemical process is given in Fig. 2.1(c). The active materials in the positive and negative electrodes are porous, permitting the lithium ions to be intercalated or de-intercalated. Typically, the negative electrode active material includes graphite. The positive electrode active material is made of one or several metal oxides. The chemical reactions occurred in these electrodes can be exemplified by a Li$_x$C$_6$/Li$_y$XO$_2$ battery with X as the metal part in the following form:

$$\text{Li}_x\text{C}_6 \xrightarrow{\text{discharge}} \text{Li}_0\text{C}_6 + x\text{Li}^+ + xe^-$$  \hspace{1cm} (2.1a)
The positive and negative electrodes are electronically insulated by a porous separator which allows the ions to pass through it but not the electrons. The electrons generated in one electrode by (2.1a) transfer to the other electrode through its adjacent current collector and the external circuit, producing electric currents. The electrodes and separator are all immersed in the liquid or gel electrolyte that is a medium for ions to diffuse.

During sustained charge or discharge operations, the lithium ion diffusion phenomenon incurs, resulting in the concentration gradient across the cell. This concentration gradient is the key idea for energy storage in a battery cell as the free energy of lithium ions in the negative electrode is much higher than that of the positive electrode. The free energy for a given material in the electrode is related to its electric potential. The terminal voltage

\[
\text{Li}_y\text{XO}_2 + y\text{Li}^+ + ye^- \xrightarrow{\text{charge}} \text{Li}_y\text{XO}_2
\]
is defined as the difference in the terminal electric potentials of the positive and negative electrodes. When there is no current flowing into the cell, the terminal voltage is referred to the open circuit potential. The amount of electrical energy of a battery is a function of the terminal voltage and battery capacity. Similar descriptions of cell structure and intercalation-based working principle were also given in [3, 28].

2.2 Battery Cell Models

Model-based approaches have achieved considerable success in a myriad of applications, e.g. hybrid electric vehicles [38] and power systems [39], and can be potentially beneficial to the advanced management of Li-ion batteries. The first step for this realisation is to develop appropriate battery models. Various models exist in the literature describing battery dynamics. This section discusses the first principle battery models and their low order alternatives. The open research question inducing the first contribution of this thesis, namely systematic battery model simplification, is identified in the end.

2.2.1 First Principle Models

*Electrochemical Model.* The first principle electrochemical model of a Li-ion battery was initially proposed by Doyle, Fuller, and Newman [14], with use of the porous electrode theory and concentrated solution theory. In this macro-homogeneous model, the ion concentration diffusion dynamics, intercalation kinetics and electrochemical potentials are taken into account and governed by a set of PDEs. Meanwhile, the micro-structural morphology in the electrodes is ignored to simplify the model structure. Critical reviews of this model from control perspectives can be found in [3, 28].

Here the emphasis is given to assumptions made in the development of this model, for further understanding the modelling principles. First, the dynamics over battery length ($y$) and height ($z$) directions are assumed to be negligibly small due to their length scales are several orders larger than the remaining coordinate ($x$). Then, each agglomeration
of the electrode active materials is considered as a spherical solid particle with a constant radius. Based on these two assumptions, a model over two spatial dimensions, namely the thickness direction and particle radius direction, is obtained, and thus called as “Pseudo 2D (P2D) model”. Additionally, it is supposed that the porosity and volume of each domain are constant and no ion diffusion exists between adjacent particles. The implications and remarks of these assumptions can also be found in [40]. The P2D model has been experimentally validated in capturing the electrochemical dynamics [3, 15], and forms a basis in the domain of battery modelling and simulations, with numerous examples presented in the literature [41, 42].

**Thermal Model.** Temperature is an important state in modelling battery dynamics because of its significant effects on parameters such as diffusion and conductivity coefficients as well as on the current-voltage response and battery degradation [43, 44]. Specifically, on the one hand, a high temperature will accelerate battery self-discharge and ageing process. In the worse case, it can cause exothermal reactions, leading to battery damage and safety issues [45]. On the other hand, a low temperature will limit the available discharge energy and increase energy loss stemmed from its high internal resistance [46, 47]. Therefore, an appropriate operating temperature range is essential to ensure battery safety, reliability and dynamic performance.

In this regard, the thermal dynamics have been formulated and added to the above isothermal P2D model for prediction of the coupled thermal and electrical behaviours [17]. This model was extended to incorporate temperature-dependent parameters such as diffusion coefficients and conductivity [48, 49]. The fidelity of this model has been validated against experimental results, e.g. in [27, 44], and thus often serves as a tool to investigate battery behaviours.

**SOH Model.** The SOH dynamics directly relate to battery lifetime and thus need to be considered in battery advanced management [50, 51]. The side reactions occur in the battery cell and induce the capacity fade and internal resistance. Battery degradation mechanisms are complex, widely-varying and dependent on the operating environments [52]. Interested readers should refer to the review papers on battery ageing mechanisms such
as [25, 53]. However, the identification and quantification of all existing ageing mechanisms are technically challenging, and the development of an accurate generic SOH model continues to be an extremely active area of research.

Considerable efforts have been dedicated to investigating battery degradation phenomena. As per [54], the electrolyte oxidation has been recognised as the dominant ageing mechanism in Li-ion batteries. A solid-electrolyte interface (SEI) film will form and grow as a result of the electrolyte oxidation reaction. Along with the growth of the thickness of an SEI film, the internal resistance will increase and the active Li-ions will decrease, resulting in the ageing of battery capacity and power. This reaction has been found to mainly take place at the surface of graphite particles during charge operations [55, 56]. By supposing such a side reaction is irreversible and concentration independent, the first principle model to describe SEI-film dynamics was formulated in [8]. This model has been widely used in the literature to analyse, optimise and control battery degradation process, e.g. [57–59].

**Summary.** The first principle sub-models for electrochemical, thermal, and ageing phenomena have been reviewed, respectively. However, an individual battery model capturing all these dynamics and their coupling relationships has not been proposed. In addition, the concatenation of these PDE models is subject to a prohibitively complicated structure and is thus computationally expensive. For most of model-based applications, simplifications for the full-order model have to be conducted.

### 2.2.2 Simplified Models: Physics-Based

Partial simplifications were previously conducted on the electrochemical model, e.g. in [27, 60–65], the temperature model in [45], and the SOH model in [58, 59]. This subsection reviews reduced order physics-based battery models derived from the above first principle models.

**2D Discretisation.** Considerable research work has been conducted to simplify the PDE based battery models. A straightforward method to do this is using spatial discretisation. The finite difference method was utilised to discretise state variables of the P2D
model over two spatial dimensions in [66, 67]. As a consequence, a large number of differential-algebraic equations (DAEs) are obtained in the system leading to significant computational overhead. By applying a semi-separable matrix technique, the obtained DAE system can be recast into a string interconnected linear system in [68]. However, as observed therein, appreciable modelling errors can be potentially induced for SOC prediction at a 1C operating rate.

**Analytical Approaches for Spatial Dimension Reduction.** Instead of solving the PDEs via 2D discretisation, spatial dimension reduction has been performed by using analytical approaches. Duhamel’s superposition theory has been utilised to eliminate the coordinate along particle radius direction [15], but the obtained expression contains infinite series that are not straightforward to implement in practice. In this regard, an approximate analytic solution of the spherical diffusion equations was presented with a finite number of terms included in the solution [69]. Motivated by the approximate analytical solution, parabolic polynomials have been used to approximate the spatially distributed dynamics [42]. To improve the model fidelity, higher order polynomial approximations were carried out for the solid-phase ion concentrations in [62, 70] and for electrolyte concentrations and ionic potentials in [71]. Another analytical approach, Padé approximation, may also be useful for PDE reduction. As per [61], the battery concentration dynamics can be well captured by Padé approximation when choosing proper states. Here, attention should be given that this approximation does not allow reconstruction of the states.

**Other techniques for PDE reductions.** Additionally, approaches such as proper orthogonal decomposition (POD), residual grouping and Galerkin’s approximation have been used in the PDE model reductions. POD has been shown to be accurate in modelling battery across 1D, but the reduced order model derived can only be generated based on a large amount of a priori information [72]. Similarly, for operations at specific frequency range, the residual grouping can be efficient to reduce the initial model. Galerkin’s approximation was employed to convert PDE based ion diffusion and potential change in the electrolyte to DAEs [73]. To do so, considerable efforts need to be spent on calculation of the residual, inner product and initial conditions.
For further information on PDE reduction techniques, six methods including integral method approximation, Ritz method, finite difference, finite element method and so on were compared in solving the electrolyte diffusion equations \cite{74}. Comparison studies for several electrochemical model reduction techniques can also be found in \cite{60,75}.

**Single Particle Model and Its Alternatives.** Amongst the reduced models obtained by using the above reduction techniques, the single particle model (SPM) is well-known and widely used for model-based applications. Its underlying assumptions, capability and several modifications are introduced here.

It may be literately understood that in the SPM each electrode is assumed to be one lumped solid particle, and the electrolyte dynamics are ignored, as illustrated in Fig. 2.2 \cite{42,76}. This assumption dramatically reduces mathematical complexity, albeit this comes at the expense of model accuracy. In fact, based on experimental results, this model is able to reproduce the battery current-voltage response up to a 1C operating rate, but large errors may be caused at high C-rates \cite{3}. In light of this, a number of extended SPM models have been developed for certain battery applications, e.g. a temperature-dependent SPM in \cite{77}, an SPM with electrolyte states in \cite{78}, and an SPM model with the ageing dynamics in \cite{79}. To enable model-based observer designs, a simplified SPM that neglects the cathode dynamics was considered in \cite{80}.
2.2 Battery Cell Models

Equivalent circuit models (ECMs) are currently extensively employed in battery management systems. In contrast to physics-based models, the mathematical structure of ECM models is simplified by ignoring battery internal ion diffusion and electrochemical dynamics [81, 82]. Instead, the electric/thermal dynamics are fitted from experimental data to an ideal voltage source, several resistors, capacitors, and/or inductors, as given in Figure 2.3. These models with constant parameters have simple mathematical structures and are easy to parameterise, but can suffer from a low accuracy [83, 84].

To address the existing model fidelity issue, time-varying parameters have been considered, e.g. in [85]. Specifically, the values of resistors and capacitors are functions of the input current, SOC and temperature. Comparison studies with respect to the accuracy and complexity of different ECM models can be found in [86, 87]. Another comparisons regarding the accuracy of ECM models and their counterparts, physics-based models,
were performed in [88, 89]. It turns out that the circuit models have limited accuracy, particularly at an extended operating range. For enhanced model fidelity in some states, multiple resistor-capacitor (RC) circuit networks have been proposed in the circuit [90], as depicted in Fig.2.4. However, the benefits of using an ECM will be lost, as the models including a number of states and state/input-dependent parameters can become overly complicated. Moreover, to fit the time-varying parameters, the model calibration has to be conducted prudently and with a large number of experiments.

Recently, fractional-order ECMs have been proposed to predict battery dynamic behaviours. Compared with integer-order models, the argument is that fractional-order models are more objective and original to reveal the system physical nature [91, 92]. In other words, an integer-order model can be regarded as a special case of a fractional-order model. This approach is applied to model the battery dynamics, where some constant phase elements (CPE) are considered instead of the ideal capacitors and ideal resistors. As observed from [93–96], for Li-ion battery modelling, a higher accuracy can be achieved by fractional-order ECMs in comparison to their integer-order counterparts. However, the fractional-order calculus is infinitely dimensional. For the implementation, proper numerical approximations need to be done, which often lead to highly nonlinear alternative models [97]. This may complicate the real-time system identification to update battery parameters along with battery degradation.

Aside from potential issues in accuracy and parameterization, ECMs have the following two pitfalls for their use in battery management. The ion concentrations and aged capacity are critical information for calculation of the battery SOC and SOH [27]. However, as the internal Li-ion behaviours are not explicitly considered, insights into battery internal information such as ion concentrations, over-potentials and aged capacity cannot be derived [3, 9]. This will lead to heuristic results when developing techniques and algorithms based on these circuit models. Furthermore, there is no physical meaning behind model states and parameters so that physical constraints on the states cannot be imposed. In view of the presented disadvantages, the class of ECMs is not considered in this project which pursues systematic modelling, state estimation and control for fast charging applications.
2.3 Battery Estimation Techniques

2.3.1 State-of-Charge Estimation

The methods existed for the SOC estimation can be divided into three categories based on whether they use battery models and the type of battery models. In the following, we...
introduce these SOC estimators and remark their advantages and disadvantages.

**Model-Free Estimators.** Amongst the model-free or black-box approaches for SOC estimation, the coulomb counting method, also known as the current integral method, is now commonly used for battery management. This algorithm can be literally interpreted in the formulation [11]

\[
SOC(t) = SOC(t_0) - \frac{1}{C_n} \int_{t_0}^{t} \eta I(\tau) d\tau, \tag{2.2a}
\]

\[
V(t_0) = U_{ocv}(SOC(t_0)), \text{ at the battery steady-state} \tag{2.2b}
\]

where the input current, \( I \), is the system input and defined as positive for discharge operations and negative for charge. The terminal voltage, \( V \), is the system output. Both the input and terminal voltage are measured from sensors, \( t_0 \) is the initial time, \( C_n \) is the battery nominal capacity, and \( \eta \) is the coulombic efficiency and is usually treated as a constant in the current integral method. The open-circuit voltage, \( U_{ocv}(\cdot) \), is a static nonlinear function of \( SOC \).

The open-loop estimation algorithm in (2.2) is easy to implement, but may suffer from low accuracy and cannot reject disturbances and noises. There are four major reasons that potentially lead to problems for the SOC indication. First, the noises of current and voltage sensors are persistent and unknown. Second, the initial SOC is often obtained by measuring the terminal voltage at the battery steady-state, and then calculating \( SOC(t_0) \) based on a static map, \( SOC \rightarrow U_{ocv}(SOC) \) [102]. This can suffer from the imperfect information of the static map and polluted measurement of \( U_{ocv}(SOC(t_0)) \). Note that it can take up to several hours for a battery to recover its steady-state, otherwise the measured voltage will be different to \( U_{ocv}(SOC(t_0)) \) [103]. Third, the coulombic efficiency is not a constant and can change with battery states and operating conditions such as SOC, temperature and input current [104]. Whereas the open-loop estimator is naturally unable to compensate for the measurement noises and parameter uncertainties. The estimation results may be acceptable for a short term, but the accumulated error can be significant over continuous operations, particularly in some harsh conditions such as large operating rates and low SOC level [105, 106]. Lastly, the nominal capacity will slowly vary with
battery SOH change. This necessitates the recalibration of $C_n$ when appreciable battery degradation incurs.

Other model-free SOC estimation approaches include the neural network and fuzzy logic algorithms. For the neural network method, it needs a huge number of training data, and meanwhile this method can be very computationally expensive [107]. Similarly, the fuzzy logic approach requires considerable battery \textit{a priori} information. These approaches are not detailed here, but interested readers are referred to survey papers, e.g. [11].

\textit{Model-Based Estimators: ECMs or Physics-Based Models.} To address the noises and parameter uncertainties inherently exist in the system, model-based close-loop SOC estimation algorithms have attracted considerable research efforts. By synthesising different battery models and estimation techniques, a wave of research work has been spawned to realise real-time SOC estimation. The obtained approaches can be generally characterised by their accuracy, computation time and robustness.

A large body of related research work focuses on circuit model-based estimator designs. An extended Kalman filter (EKF) synthesised from a resistance-capacitor (RC) battery model was used to estimate the states for a Li-ion battery in [108]. By using a second-order RC battery model, the similar work was conducted in [109]. The key idea behind a Kalman filter is to minimise the mean-square error in state estimation, and it has been proven to be an optimal estimator for linear systems with noisy measurements [110]. The EKF is the nonlinear version of the Kalman filter to deal with model nonlinearities. Based on the class of circuit models as described in Section 2.2.3, the second-order EKF, robust EKF, adaptive EKF, unscented Kalman filters (UKF), and particle filters (PF) were respectively proposed for battery state estimation in the literature, e.g. [105, 111, 112]. Further information on Kalman filter and its various forms can be found in [113]. Other estimation techniques for battery parameters and states such as sliding mode observer (SMO), Luenberger observer and moving horizon estimation (MHO) were provided in [114–116].

It is worth noting that the internal models are important for these SOC estimators. As stated earlier, the ECMs are relatively simple. As a result, the estimation algorithms de-
developed from these models are usually superior in computational efficiency as compared
to physical model-based estimators. However, the problematic accuracy of the ECMs
may impact the estimator’s capability, resulting in unreliable estimate values. This can
further lead to suboptimal solutions when using these estimators for state-feedback con-
trols [3].

In this regard, the white-box or physical model-based approaches have been considered
for SOC estimation. Difficulties for this work attribute to the complicated initial battery
model structure involving the infinitely dimensional state variables over multiple spatial
dimensions. Consequently, the estimation algorithms were usually proposed based on
reduced order physical models. For instance, the SPM model was used for battery state
estimation and was showed to be effective under some operating conditions [24, 117]. By
using the same model but robust nonlinear observers, SOC estimation techniques were
further presented in [118]. It has been demonstrated therein that the noises and distur-
bances can be effectively rejected by the designed estimator at moderate current rates. A
PDE observer was initially proposed for battery state and parameter estimation in [80],
where a reduced SPM model was adopted. To enhance the estimator performance, the
extended SPM models where the electrolyte states and/or the temperature states are in-
corporated have been considered for battery state estimation [77, 78]. Other SOC estima-
tors based on physics-based battery models can be found in the work such as [66, 119].

In general, the mentioned physical model-based approaches are useful to estimate the
SOC for specific applications in some time interval. However, the estimation perfor-
mance is likely to degrade with the battery age. Therefore, for accurate SOC estimation
over the battery lifetime, the state of health dynamic information needs to be taken into
account.

2.3.2 State-of-Health and Charge Estimation

In addition to the state-of-charge, the state-of-health (SOH) is also important for battery
management. Particularly, the SOH, as a measure of the degree of battery’s degrada-
tion, can be used for fault diagnostics, lifetime prediction and health-conscious charge
and discharge control. Several parameters and states, such as the SEI film resistance, capacity, total number of active Li-ions, and diffusion coefficients, are related to the SOH [120]. However, all these quantities are impractical to measure during battery operations. Therefore, to acquire their in-situ information, it is necessary to develop SOH estimation algorithms. An excellent review on health estimation of a Li-ion battery can be found in [18].

**SOH Estimators.** One of the key challenges for SOH estimation is the paucity of control-oriented battery health models that can accurately capture the ageing dynamics. Given the SOH is related to some battery internal parameters and states, different SOH observers exist in the literature. By resorting to the ECM model-based approaches, parameter estimation techniques including impedance-based KF [121], EKF [108], recursive least squares [122] and moving-horizon parameter estimation [123] have been employed to estimate the SOH. As remarked in [124], another challenge in battery SOH estimation is the development of electrochemistry-based degradation models that can provide physical insights into the ageing behaviours but are sufficiently simple. Subsequently, some physical model-based SOH observers have been proposed. By utilising a reduced SPM model and the measurements of current and terminal voltage, nonlinear least squares were developed to estimate the diffusion parameter and boundary input coefficient parameter [80]. By using a high order electrochemical model, similar parameter estimation techniques were adopted in [125]. However, the above research work fails to explicitly consider battery degradation dynamics so that it is unable to realise SOH dynamic controls.

In a Li-ion battery system, the state of charge and health are related. Specifically, with battery ages, some of the system parameters will appreciably change leading to increased model mismatch and SOC estimation error. At the same time, the SOC information is useful for SOH estimation. Accordingly, it would be desirable to design an estimator that can accurately estimate both the SOC and SOH.

**SOC-SOH Estimators.** Some attempts have been recently made in the literature for simultaneous estimation of the two states using model-based approaches. This type of work
basically combines the SOC estimation and SOH estimation together and constructs state and parameter estimation problems. For instance, by using a simplified SPM, the adaptive PDE observer and nonlinear least square technique have been employed to estimate SOC and SOH, respectively, in [80]. The state and parameter estimators were formulated using iterated EKF in [126, 127]. Note that the computation power required by the iterated algorithms can be probably expensive, hindering the real-time state estimation. Also, the tuning process for estimator design parameters is non-trivial. An SOC and SOH estimator was proposed using the SMO method and an electrochemical model, where the SOH was inferred from the ion diffusion coefficient and SEI film resistance and the temperature effects were ignored [26].

All the referred SOC-SOH estimators can be useful in a certain time range. However, the SOH time-scale is much slower than that of the SOC. It is, hence, unnecessary to estimate the SOH in the fast time-scale with compromise of computational efficiency. Furthermore, observer design techniques for singularly perturbed systems may lead to ill-conditioned observer gains and potentially undermine the convergence properties [128].

The concept of multi-time-scale estimation was recently introduced into the battery area for SOC and SOH based equivalent circuit battery models, e.g. in [102, 129]. However, the degradation dynamics have not been explicitly considered in the employed battery models. Aside from this, no rigorous analysis on the stability of estimation errors has been provided.

The multi-time-scale estimation theory was developed for a class of linear systems in [130]. The key idea is to design observers through restriction of the process dynamics on the slow manifold and thus taking analytical and computational advantages that the lower-dimensional systems bring. This work was extended for some nonlinear systems by Kazantzis et al. [131]. The rigorous analysis can be derived to address the effect of ignoring the fast dynamics on the convergence properties of reduced order observers. The work of [131] was then generalised on a wider class of singularly perturbed systems and observers in [132]. In which, the stability of observation errors has been analytically characterised, and a semi-globally practical stability was obtained under certain assump-
tions. Nevertheless, the problems solved above are different to the battery state estimation where both the fast and slow states in the singularly perturbed battery systems are required, for SOC and SOH indication, respectively. Furthermore, the battery operations over multiple charging and discharging cycles will lead to oscillating behaviours in the electrochemical dynamics. Therefore, it would be desirable to develop new techniques for addressing the battery SOC and SOH estimation problem.

2.4 Battery Charging Strategies

Battery charging strategies implicate many nuanced considerations and subtleties such as the charge completion time, battery lifetime, economy and dynamic performance. For user convenience, the fast charging that is able to quickly restore one’s access to the electronic devices or electrified vehicle is desired. However, the enhanced charging speed may come at the cost of degrading battery lifetime. This section reviews the available charging strategies developed from model-free approaches first, followed by charging rate optimisation and control algorithms.

2.4.1 Model-Free Charging Strategies

From the perspective of protecting battery state-of-health, the trickle charging is often the first consideration to restore the capacity of a rechargeable battery. Moreover, in this approach, the issues like overheating and overcharging can be easily avoided. Nevertheless, its charging duration is extremely long such that it may work overnight to finish the charging task. For the cases where the time consumption is an important concern, the constant-current (CC) charging strategy with large C-rates may be considered. Accurate state estimation for SOC, temperature and/or terminal voltage will be required to avoid constraint violations. On the other hand, the time saving may come at the expense of severe battery degradation.

With the merits of trickle charging and fast CC charging, the constant-current constant-
voltage (CCCV) protocol is now the industry standard charging method [18]. It can be literally understood that in this method a battery is charged at a constant current until its terminal voltage reaches the upper limit, and then the charging process enters the CV stage. When the current drops below a threshold value, the whole charging operation is finished. In this scheme, the initial current and constraints on the current and voltage are pre-defined. In some sense, the charging profiles are determined offline. To ensure satisfaction of the voltage constraint at the end of CC stage, the initial current is usually confined to a moderate value. Even though a relatively high current is chosen in the beginning, a long time can be taken in the CV stage [133].

To improve the charging performance, various schemes consisting of CC and CV modes have been studied, with the terminology as “CCCV family”. As per [19], the batteries that are close-to-fully discharged can be recharged by very high currents for a short time without appreciably impacting its health. In other words, a high current can be utilised in the initial charging stage to reduce the completion time. Motivated by this, a five-stage CC strategy was proposed in [21], where the battery was charged under the largest current in the first stage and the current sequentially reduces in the subsequent stages. This strategy was found to be able to charge a battery in less time and support more life cycles in comparison to the CCCV approach. Other charging strategies composed of multi-stage CC followed by a CV stage was presented in [22, 134]. Similar work can also be found in [23, 135].

The impulse charging algorithms have also been actively studied. The basic idea of these strategies is to relax the battery for a while after each charging pulse. This can alleviate ion concentration gradients in the course of battery charging. This approach was found to have the ability to reduce the charging time and increase battery lifetime in [83, 136]. Whereas opposite argument was proposed in [137], where the impulse charging was shown to adversely affect the charging performance due to the effect of Ohm’s law on nonlinear impedance.

In summary, some improvements may be achieved by the alternative CCCV approaches or impulse charging relative to the CCCV approach. A review of several model-free
charging strategies algorithms for nickel and Li-ion batteries was also conducted in [20]. However, in these scenarios, the battery is essentially charged with prescriptive profiles without making use of battery internal in-situ information. This class of approaches is often *ad hoc* and suboptimal.

### 2.4.2 Model-Based Charging Strategies

In contrast to model-free charging strategies, model-based approaches have been considered for the development of charging strategies. Generally speaking, these strategies are built upon model-based optimisation techniques and/or control algorithms. Commonly, the objectives in the development of charging strategies include the shortest charging time, the minimum SOH change, and/or the minimum cost. An optimal charging protocol is to determine the currents which are able to charge the battery to a prescriptive state of charge and at the same time satisfy the specified objectives. One may categorise research work in this domain based on the type of battery models used. This section presents and remarks the existing charging strategies and justifies the potential research opportunities.

The first body of research on model-based charging strategies makes use of circuit battery models. Based on an RC circuit battery model, a dual-optimisation problem was formulated to balance the competing objectives, i.e. the charging time and energy loss due to ohmic loss [85]. The pseudo-spectral technique was used to discretise the problem into nonlinear programming (NLP) which can be efficiently solved using sparse NLP tools. By combining an electrical-thermal circuit model and an empirical SOH model, an open-loop optimal control problem was deployed for battery fast and safe charging [138]. To minimise the charging time, energy loss and temperature increase, an optimal control algorithm for battery charging was derived in [139]. However, these open-loop algorithms are essentially unable to deal with the model uncertainties and disturbances.

Model predictive control (MPC) is a model-based real-time control paradigm with guaranteed stability and some inherent robustness [29, 30]. Moreover, it is capable of handling nonlinearity, multi-variables and constraints. This control algorithm is likely useful for
battery charging strategy development, and a few of related papers appeared recently. Within the tracking MPC framework, a new charging strategy was presented to trade-off the temperature increase and charging time [140], where the genetic algorithm (GA) was used as the optimiser and a CCCV charging profile serves as the reference trajectory. By considering the same objective function and reference, a linear MPC problem based on a first order RC circuit model was formulated in [82]. Both of these algorithms have been demonstrated to satisfy constraints and to achieve benefits relative to the CCCV charging. The related optimal charging strategies can also be found in the literature, e.g. [141]. Whereas, in the absence of physical meaning in the states/parameters and being low accurate, the ECM-based prediction models considered will limit these algorithms.

Physics-based models are promising to develop advanced charging algorithms. Based on a PDE battery model that captures the electrochemical-thermal dynamics, a one-step model predictive control (MPC) was presented to minimise battery charging time [142]. Constraints were imposed on temperature and overpotential of the side reaction to alleviate battery degradation. However, the one-step MPC may not have a guarantee of stability and does not address the control robustness directly. Based on a simplified single particle model (SPM), a nonlinear model predictive control problem was formulated for battery fast charging [143]. To overcome the potential computation and non-convex issues, a differential flatness approach was used to reformulate the model. Using a similar model, a reference governor-based approach was considered for battery charging profile management [144]. By extending the SPM model to incorporate electrolyte and thermal dynamics, an open-loop optimal charging control was then formulated in [145]. Other charging strategies developed from physical model-based control can be referred to the work such as [146–148]. In all of the mentioned work, the battery internal electrochemical state information was assumed available. In addition, none of these approaches explicitly consider battery ageing dynamics.

Therefore, practically optimal charging strategies that can optimally balance the charging objectives including the completion time and the battery lifetime have not been developed yet. A potential approach may be to develop model predictive control algorithms with an appropriate physical battery model and a state estimator. This model should cap-
ture the electrochemical, thermal and ageing dynamics. Meanwhile, the estimator provides real-time information for the SOC and SOH. Furthermore, the user’s requirements with respect to the charging time and battery SOH change may vary between different charging operations. It would be desirable to develop adaptive charging strategies that cover this consideration.

### 2.5 Research Aims

Based on the above literature review and open research opportunities identified, the research aims of this thesis are presented in the following.

*To propose a framework of systematic PDE model simplification for a Li-ion battery*

This research aims to develop a systematic approach for simplification of PDE battery models. The disparate time scales inherent in a battery system will be identified, leading to a singularly perturbed mathematical model structure. Whereas there are no available formal analysis tools for reduction of singularly perturbed PDE systems. To solve this problem, the original model will be reformulated in Hilbert space. The model simplifications will be systematically conducted by using a singular perturbation approach and the averaging theory. For the simplified PDE models, further order reductions are pursued to result in ODE systems. The assumptions based on which reduced models are developed will be explicitly stated and justified. The obtained models form a basis for various model-based applications.

*To develop a model-based procedure to quickly evaluate battery charging strategies*

The first application of simplified models from the model simplification framework proposed in *Research Aim 1* is to evaluate charging strategies. For given charging strategies, an evaluation procedure is developed utilising reduced order models instead of the initial
high order model or experimental methods. This procedure is able to quickly simulate battery operations and ascertain their terminal effect on the state-of-health and lifetime.

To design an accurate and robust model-based estimator for state-of-charge and health estimation

Based on a reduced model that captures the electrochemical and ageing dynamics, a non-linear estimator is first designed to estimate the state-of-charge and health simultaneously. Second, the theory of multi-time-scale estimation will be developed for a class of singularly perturbed systems. This enables decomposition of fast and slow states and estimation of them in different time-scales, alleviating the issues such as ill-conditioned gain, convergence and computation burden. This theoretical work will be applied to estimate battery state-of-charge and health. Illustrative results will be provided to demonstrate the accuracy, robustness and computational complexity of the proposed algorithm.

To propose an optimal fast charging strategy using the model resulted from Research Aim 1 and the estimator from Research Aim 3

With the control-oriented battery model and estimator obtained in the previous steps, the optimal fast charging strategy will be finally proposed. This scheme will be formulated as a linear-time-varying (LTV) MPC problem, and look to directly address the constraints on input current and battery internal states. The efficacy of the proposed approach will be demonstrated via comparisons with the existing charging strategies regarding the charging completion time and SOH change. In addition, the proposed strategy aims to serve users a specialised tool to manipulate SOH evolution profiles under certain assumptions.
Battery models are commonly required for applications such as simulation of battery characteristics, evaluation of charging strategies, state estimation and controller design. By applying first principles, dynamics inside a battery can be mathematically expressed as coupled nonlinear PDEs over multiple domains [8, 14, 17]. However, these models have a high order of computational complexity and are thus not suitable for most model-based applications. This chapter focuses on battery model development in three steps. First, a PDE-based high-fidelity model is proposed, capturing the coupled electrochemical, thermal, electrical and ageing dynamics. This initial model is used as a starting point and benchmark for model simplifications. Based on identification of multiple time-scales inherent in a battery system, a framework for PDE battery model simplification based on the singular perturbations and averaging theory is then developed. As a result, libraries of simplified models are obtained, corresponding to a range of applications. Finally, numerical techniques are applied to the simplified PDE models, leading to a set of ODE systems that are suited for online estimator/controller design.

3.1 Full-Order Battery Model

In a Li-ion battery cell as provided in Fig. 3.1, the system dynamics represent the flow of Li-ions from the solid particles to the electrolyte along the radius direction, $r$, and from

---

1A substantial proportion of this chapter has already published as articles in the IEEE Transactions on Control Systems Technology [149] and the 19th IFAC World Congress [40].
the positive electrode (cathode) to the negative electrode (anode) along the thickness direction, \( x \). Such Li-ion diffusion results in electrochemical, thermal, electrical, and ageing phenomena associated with the main intercalation reaction and side reaction.

In this section, a high-fidelity battery model that captures all these system dynamic characteristics is formulated. This initial model formulation will be a test bed measuring the accuracy and computational complexity for all later model simplifications. The interconnection of subsystems including the electrochemical (\( \Sigma^c \)), thermal (\( \Sigma^T \)), electrical (\( \Sigma^e \)), and ageing dynamics (\( \Sigma^{SOH} \)) is summarised in Fig. 3.2. In the rest of this section, each of these battery subsystems is elaborated using the notations defined in Section 1.2.

**Dynamics of Mass Diffusion.** Driven by the battery potential difference, the Li-ions diffuse from the centre to the surface of spherical solid particles where while most of them undergo the main intercalation reaction, a small part of them are involved in the side reaction that triggers battery degradation.
During the side reaction of a battery, a passive SEI film forms and grows leading to increased film resistance in the negative electrode, and this is accompanied by consumption of the active Li-ions in the positive electrode [8]. Based on the mass conservation law applied to the Li-ion concentration, the lost number of Li-ions per volume, $C_{\text{loss}}^+$ in the positive electrode is given by

$$C_{\text{loss}}^+ (x, r, t) = \frac{Q_{sr}(x, t)}{Q_{\text{max}}} \bar{C}_s^+ (x, r, t), \quad (3.1)$$

where $\bar{C}_s^+$ is the ideal solid-phase Li-ion concentration in the absence of battery ageing.

In an ideal battery without performance degradation, the mass transport in the solid particles can be described by Fick’s law of diffusion and is given as [3, 15]

$$\frac{\partial \bar{C}_s^+ (x, r, t)}{\partial t} = \frac{D_{\text{eff}, s}^+}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial \bar{C}_s^+ (x, r, t)}{\partial r} \right). \quad (3.2)$$

In existing models of capacity fade, the initial cathode Li-ion concentration is updated in a discontinuous jump at the end of each charging operation to capture the ageing effect. To describe the continuous interaction between Li-ion loss and capacity fade, the concentration dynamics in the positive electrode based on the concentration conservation property is proposed here as

$$\frac{\partial C_s^+ (x, r, t)}{\partial t} = \frac{\partial \bar{C}_s^+ (x, r, t)}{\partial t} - \frac{\partial C_{\text{loss}}^+ (x, r, t)}{\partial t}. \quad (3.3)$$

The combination of (3.1), (3.2), and (3.3) results in a dynamic equation simultaneously describing the Li-ion concentration diffusion and consumption

$$\frac{\partial C_s^+ (x, r, t)}{\partial t} = \frac{\partial \bar{C}_s^+ (x, r, t)}{\partial t} - \frac{Q_{sr}(x, t)}{Q_{\text{max}}} \frac{\partial \bar{C}_s^+ (x, r, t)}{\partial t} - \frac{\bar{C}_s^+ (x, r, t)}{Q_{\text{max}}} \frac{\partial Q_{sr}(x, t)}{\partial t}, \quad (3.4)$$

where the first term of the right-hand side of (3.4) represents the normal Li-ion diffusion, and the last two terms denote Li-ion loss caused by battery side reaction.
Inside the negative electrode, the active Li-ion loss is typically ignored [8, 57], allowing the state behaviour of Li-ion concentration to be presented as (3.2), namely

$$\frac{\partial C_s(x, r, t)}{\partial t} = \frac{D_{\text{eff}}^s}{r^2} \left( r^2 \frac{\partial C_s(x, r, t)}{\partial r} \right).$$ \hspace{1cm} (3.5)

In the electrolyte, the change of Li-ion concentration is related to its gradient-induced diffusive flow and the local electrolyte current and is governed by [3, 14]:

$$\frac{\partial C_j^e(x, t)}{\partial t} = \frac{\partial}{\partial x} \left( \frac{D_{\text{eff}}^{j,e}}{\mu_j^e} \frac{\partial C_j^e(x, t)}{\partial x} \right) + \frac{\gamma_j^i}{F \mu_j^e} \frac{\partial i_j^e(x, t)}{\partial x},$$ \hspace{1cm} (3.6)

where $j \in \{+, -, \text{sep}\}$.

**Dynamics of Electrical States.** The signal transmission of local potentials and currents in the solid particles and electrolyte is very fast. Their steady-state behaviours can be modelled by applying Ohm’s law, modified Ohm’s law, and the law of charge conservation, respectively, as provided in [3]. Here, the dynamic equations that explicitly describe the transient behaviour of the state variables of potentials and currents are proposed in the following

$$\epsilon_{\Phi_s} \frac{\partial \Phi_s^\pm(x, t)}{\partial t} = \frac{\partial \Phi_s^\pm(x, t)}{\partial x} + \frac{I(t) - i_j^\pm(x, t)}{\sigma_{\text{eff}, \pm}},$$ \hspace{1cm} (3.7)

$$\epsilon_{\Phi_j^e} \frac{\partial \Phi_j^e(x, t)}{\partial t} = \frac{\partial \Phi_j^e(x, t)}{\partial x} + \frac{i_j^e(x, t)}{\kappa_{\text{eff}, j}} - \frac{2R\gamma_j^i T_j^i(x, t) \partial \ln C_j^e(x, t)}{F \partial x},$$ \hspace{1cm} (3.8)

$$\epsilon_{i_j^e} \frac{\partial i_j^\pm(x, t)}{\partial t} = \frac{\partial i_j^\pm(x, t)}{\partial x} - Fa^\pm j^\pm(x, t),$$ \hspace{1cm} (3.9)

where $\epsilon_{\Phi_s}, \epsilon_{\Phi_j^e}, \epsilon_{i_j^e}$ are small positive parameters and their reciprocals are respectively related to the transmission speeds of electric potential, ionic potential, and electrolyte current.

Based on Kirchoff’s current law applied in the $x$-direction, the input current through the cross section of a cell is uniform, so that the local solid-phase and electrolyte currents
The total Li-ion flux $J$ consists of the normal intercalation reaction flux, $J_I$, and side reaction flux $J_{sr}$. $J_I$ is governed by the Butler-Volmer kinetic equation and can be expressed as a function of $\Phi_s, \Phi_e, C_s, C_e, T,$ and $R_f$ [150]

$$J_I^\pm (x,t) = \frac{i_0^\pm (x,t)}{F} \left( e^{\frac{n_a F \eta^\pm (x,t)}{RT^\mp(x,t)}} - e^{-\frac{n_a F \eta^\pm (x,t)}{RT^\mp(x,t)}} \right),$$

where the exchange current density, $i_0$, and the overpotential, $\eta$, are described as

$$i_0^\pm (x,t) = K^\pm C_e^\pm (x,t)^{\alpha_n} (C_{\text{max}}^\pm - C_{\text{ss}}^\pm (x,t))^{\alpha_n} C_{ss}^\pm (x,t)^{\alpha_p},$$
\[ \eta^\pm(x,t) = \Phi^\pm_s(x,t) - \Phi^\pm_e(x,t) - U^\pm(x,t) - FR_f(x,t)J^\pm(x,t). \] (3.13)

Here \( k \) is the reaction rate constant, and \( C_{ss} \) is the Li-ion concentration at the surface of these particles, i.e.

\[ C_{ss}^\pm(x,t) := C_s^\pm(x,r = R_p^\pm,t). \] (3.14)

The open circuit potential, \( U \), can be evaluated as a nonlinear function of the surface Li-ion concentration \([15, 41]\).

**Thermal Dynamics.** The heat sources inside a cell include the reaction heat generation, reversible heat generation, and ohmic heat generation. The generated heat is transported through the battery internal conductivity and convection between battery surface and the surrounding environment. To capture temperature distribution and evolution in a Li-ion battery cell, the thermal balance equation is provided as \([16, 17]\)

\[
\rho c_p ^J \frac{\partial T^J(x,t)}{\partial t} = \lambda ^J \frac{\partial^2 T^J(x,t)}{\partial x^2} - (I(t) - i^J_e(x,t)) \frac{\partial \Phi^J_s(x,t)}{\partial x} - i^J_e(x,t) \frac{\partial \Phi^J_e(x,t)}{\partial x} + F a^J f^J(x,t) \eta^J(x,t) + F a^J f^J(x,t) T^J(x,t) \frac{\partial U^J}{\partial T}. \] (3.15)

On the right-hand side of (3.15), the second and third terms represent the reaction heat, and the last two terms are separately the ohmic heat and reversible heat. From the above equation, it follows that the electrical states, \( \Phi_s, \Phi_e, i_e \), are directly involved in heat generation. The elevated temperature, in turn, affects the main intercalation reaction through (3.8) and temperature-dependent parameters including \( D_s^\pm, D_e^\mp, K^- \) and \( K^+ \) via an Arrhenius-like temperature dependency \([17]\). This further accelerates the dynamics of battery ageing via (3.18).

**Dynamics of Battery Ageing.** The main terminal product resulting from the side reaction is a resistive and insoluble SEI film that forms in the graphite anode \([8, 57]\). As battery operating cycles increase, such a film grows and leads to successive capacity fade
and internal resistance rise. 

A lumped capacity fade has been formulated in the initial SOH model of [8]. However, physically, the side reaction, as well as its resulting parasitic flux and SEI film, may exhibit strong spatial variations. To model this process accurately, a spatially distributed aged capacity, \( Q_{sr}(x, t) \), is considered in establishing the full-order battery model. With this in mind, the dynamic equations of capacity fade and internal resistance are proposed in the following form

\[
\frac{\partial Q_{sr}(x, t)}{\partial t} = -F a^- A^- L^- J_{sr}(x, t), \tag{3.16}
\]

\[
\frac{\partial R_f(x, t)}{\partial t} = -\frac{M_f}{\rho_f \sigma_f} J_{sr}(x, t). \tag{3.17}
\]

In (3.16)-(3.17), by assuming the side reaction to be irreversible, the side reaction flux \( J_{sr} \) can be modelled by the Tafel equation [8, 150]

\[
J_{sr}(x, t) = \frac{-i_{0sr}}{F} \exp \left[ \frac{-F \alpha_{sr}(x, t)}{RT^- (x, t)} \right], \tag{3.18}
\]

where the overpotential that determines the rate of side reaction is provided by

\[
\eta_{sr}(x, t) = \Phi^-_x (x, t) - \Phi^-_e (x, t) - U_{sr} - F R_f(x, t) J^- (x, t). \tag{3.19}
\]

**Boundary Conditions.** The complete battery cell over the negative electrode, separator, and positive electrode is linked through the solid-electrolyte interfaces and electrode-separator interfaces. At the centre of solid particles, where \( r=0 \), and the Li-ion diffusion rate is zero due to the symmetry of spherical diffusion. At the surface of solid particles, where \( r=R_p \), the Li-ion concentration flux is governed by the diffusion rate between the solid particles and electrolyte. At the current collectors, namely \( x=0 \) and \( x=L \), the electrolyte currents and electrolyte diffusion rates are all zero, and the thermal transport is determined by Newton’s cooling law. At the electrode-separator interfaces, where \( x=L^- \) and \( x=L^- + L^{sep} \), the electrolyte concentration and temperature are continuous and preserve mass and energy conservation properties. Particularly, in the separator, there
Battery Modelling

is no solid particle as well as solid-phase Li-ions so that for their governing equations $C_s = \Phi_s = Q_{so} = R_f = 0$, and $i_e(x,t) = I(t)$.

**System Outputs.** The system outputs of interest are the terminal voltage, $V(t)$, SOC and SOH. The terminal voltage is presented as the difference between the positive and negative electrical potentials at the terminals, and is a measurable output. The SOC in each electrode is defined as the ratio between the averaged available Li-ion concentration and maximum possible concentration [3]. Specifically, the SOC represents the averaged available charge contained in active materials and is used to evaluate the energy left in a battery. The SOH is quantitatively evaluated by the average capacity fade in the anode electrode and the initial maximum available capacity [25]. With this in mind, these system outputs are formulated as follows

$$V(t) = \Phi^+(L,t) - \Phi^-(0,t),$$  \hspace{1cm} (3.20a)

$$SOC^\pm(t) = \frac{3}{L^+ R^+} \int_0^{L^+} \int_0^{R^+} \frac{C_s^\pm(x_r,t)}{C_{s_{\text{max}}}} \, dr \, dx,$$  \hspace{1cm} (3.20b)

$$SOH(t) = 1 - \int_0^{L^-} \frac{Q_{sr}(x,t)}{Q_{\text{max}}} \, dx.$$  \hspace{1cm} (3.20c)

It is worth mentioning that the outputs in (3.20a)-(3.20c) are finite dimensional whereas the states are all infinite dimensional.

**Summary of Battery Model.** To sum up, the initial high-fidelity battery system (referred to as $\Sigma$) is composed of four subsystems which respectively describe the electrochemical, thermal, electrical, and ageing phenomena. The dynamic characteristics are represented by (3.4)-(3.19), leading to a set of 17 coupled nonlinear PDEs over two spatial dimensions. Besides incorporating all these relevant system dynamics and their coupling relationships into an individual model, three modifications have been proposed. Firstly, the continuous coupling effects between the normal electrochemical reaction and the side reaction with respect to the solid-phase Li-ion concentration have been captured, as provided in (3.4). The dynamic equations of potentials and currents rather than their steady states are explicitly formulated in (3.7)-(3.9). In addition, the spatially distributed capacity fade has been introduced, in (3.16), to reflect the physical nature of battery degrada-
3.2 Model Reformulation in Hilbert Space

In this section, the initial full-order battery model is reformulated in Hilbert space in order to precisely characterise its mathematical structure and to uncover the singular perturbation structure underlying the battery model. The key idea is to present the PDE system in an operator-differential equation form by using function mappings between Euclidean space and Hilbert space.

To do so, the common features of the battery dynamic equations are first extracted. It is defined the state variables in a vector

\[ \bar{x} := [\bar{x}^-, \bar{x}^+, \bar{x}^{\text{sep}}]^T, \]  

where in the negative electrode, separator, and positive electrode, the vectors of state variables are defined as

\[ \bar{x}^- := [C_s^-, C_e^-, \Phi_s^-, \Phi_e^-, i_e^-, T^-, Q_{sr}, R_f]^T, \]  
\[ \bar{x}^{\text{sep}} := [C_s^{\text{sep}}, \Phi_s^{\text{sep}}, T^{\text{sep}}]^T, \]  
\[ \bar{x}^+ := [C_s^+, C_e^+, \Phi_s^+, \Phi_e^+, i_e^+, T^+]^T. \]

The system input, \( u \), is defined as \( u := I(t) \). The domain of definition for all the state variables is \( D(x, r) = \{(x, r) | x \in [0, L], r \in [0, R_p]\} \). \( y_c, y_m \) separately denote the unmeasurable and measured output vectors and are defined as \( y_c := [\text{SOC}^-(t), \text{SOC}^+(t), \text{SOH}(t)]^T \), and \( y_m := V(t) \).

Then, the PDE-based battery system \( \Sigma \) may be rewritten in the general form

\[ \frac{\partial \bar{x}}{\partial t} = F_1 \frac{\partial \bar{x}}{\partial r} + S_1 \frac{\partial \bar{x}}{\partial x} + F_2 \frac{\partial^2 \bar{x}}{\partial r^2} + S_2 \frac{\partial^2 \bar{x}}{\partial x^2} + H(\bar{x}, u), \]  
\[ y_c = g(\bar{x}), \]  
\[ y_m = q(\bar{x}). \]
To do so, specifically, the right-hand side of Eqn (3.2) is first re-written as

\[
\frac{\partial}{\partial r} \left( r^2 \frac{\partial C_s}{\partial r} \right) = 2r \frac{\partial C_i}{\partial r} + r^2 \frac{\partial C_s^2}{\partial r^2}. \tag{3.24}
\]

The above dynamic equations are subject to the boundary conditions:

\[
\begin{align*}
C_1 \mathbf{x}^- |_{x=0} &= 0, \\
C_3 \mathbf{x}^+ |_{x=L} &= 0, \\
C_4 \mathbf{x}^- |_{x=0} &= 0, \\
C_5 \mathbf{x}^+ |_{x=L} &= 0, \\
C_2 \frac{\partial \mathbf{x}^-}{\partial x} |_{x=0} &= D_1, \\
C_4 \frac{\partial \mathbf{x}^+}{\partial x} |_{x=L} &= D_{2u}, \\
C_5 \frac{\partial \mathbf{x}^-}{\partial r} |_{r=0} &= 0, \\
C_6 \frac{\partial \mathbf{x}^+}{\partial r} |_{r=0} &= 0, \\
C_7 \frac{\partial \mathbf{x}^-}{\partial r} |_{r=R_p} &= D_3, \\
C_8 \frac{\partial \mathbf{x}^+}{\partial r} |_{r=R_p} &= D_{4u}, \\
\mathbf{x}^- |_{x=L^-} &= D_5 \mathbf{x}^{sep} |_{x=L^-}, \\
\frac{\partial \mathbf{x}^-}{\partial x} |_{x=L^-} &= D_6 \frac{\partial \mathbf{x}^{sep}}{\partial x} |_{x=L^-}, \\
\mathbf{x}^{sep} |_{x=L^-+L^{sep}} &= D_7 \mathbf{x}^+ |_{x=L^-+L^{sep}}, \\
\frac{\partial \mathbf{x}^{sep}}{\partial x} |_{x=L^-+L^{sep}} &= D_8 \frac{\partial \mathbf{x}^+}{\partial x} |_{x=L^-+L^{sep}}. 
\end{align*}
\tag{3.25}
\]

The initial conditions are given as:

\[
\mathbf{x}(t = t_0, x, r) = \mathbf{x}_0. \tag{3.26}
\]

In (3.23a), \( F_1, F_2, S_1, S_2, \) and \( H \) are all matrix functions and can be decomposed into:

\[
\begin{align*}
F_1 &= \text{diag}(F_1^-, F_1^{sep}, F_1^+), \\
F_2 &= \text{diag}(F_2^-, F_2^{sep}, F_2^+), \\
S_1 &= \text{diag}(S_1^-, S_1^{sep}, S_1^+), \\
S_2 &= \text{diag}(S_2^-, S_2^{sep}, S_2^+), \\
H &= \text{diag}(H^-, H^{sep}, H^+).
\end{align*}
\]

In these matrices, the vector functions can be derived from the battery dynamic equations (3.4)-(3.19), and are further explicitly provided as:

\[
\begin{align*}
F_1^- &= \text{diag}(2D_{\text{eff}}^- / r, 0, 0, 0, 0, 0, 0, 0), \\
F_1^+ &= \text{diag}(2D_{\text{eff}}^+ (1 - Q_{sr} / Q_{\text{max}}), 0, 0, 0, 0, 0), \\
F_2^- &= \text{diag}(D_{\text{eff}}^-, 0, 0, 0, 0, 0, 0, 0).
\end{align*}
\]
3.2 Model Reformulation in Hilbert Space

\[ F_2^+ = \text{diag}(D_{s\text{eff}}^+ (1 - Q_{sr}/Q_{\text{max}}), 0, 0, 0, 0, 0), \]
\[ F_1^{\text{sep}} = F_2^{\text{sep}} = \text{zeros}(3, 3), \]
\[ S_2^- = \text{diag}(0, D_{c\text{eff}}^- / \mu_c^-, 0, 0, 0, \lambda^- / (\rho^- c^-), 0, 0), \]
\[ S_2^+ = \text{diag}(0, D_{c\text{eff}}^+ / \mu_c^+, 0, 0, 0, \lambda^+ / (\rho^+ c^+)), \]
\[ S_2^{\text{sep}} = \text{diag}(D_{c\text{eff}, \text{sep}}^+ / \mu_c^{\text{sep}}, 0, \lambda^{\text{sep}} / (\rho^{\text{sep}} c^{\text{sep}})). \]

\[
S_1^+ = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
S_1^- = \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
H^+ = \begin{pmatrix}
-F a^- A^- L^- J_{sr} C_s^+ Q_{\text{max}}^{-1} \\
0 \\
(1 - i_c^-) (\varepsilon_{\Phi c} \sigma_{\text{eff}}^-)^{-1} \\
i_c^- (\varepsilon_{\Phi c} \kappa_{\text{eff}}^-)^{-1} \\
F a^+ J^+ c_{\text{ie}}^{-1} \\
F a^+ J^+ (\eta^+ + T^+ \frac{\partial U^+}{\partial T}) (\rho^+ c^+)^{-1}
\end{pmatrix}
\]

\[
H^- = \begin{pmatrix}
0 \\
0 \\
(1 - i_c^-) (\varepsilon_{\Phi c} \sigma_{\text{eff}}^-)^{-1} \\
i_c^- (\varepsilon_{\Phi c} \kappa_{\text{eff}}^-)^{-1} \\
F a^- J^+ c_{\text{ie}}^{-1} \\
F a^- J^- (\eta^+ - T^- \frac{\partial U^+}{\partial T}) (\rho^- c^-)^{-1} \\
-F a^- A^- L^- J_{sr} \\
-M f J_{sr} (\rho f \sigma_f)^{-1}
\end{pmatrix}
\]
\[
S_{1}^{\text{sep}} = \begin{pmatrix}
0 & 0 & 0 \\
-\frac{2R v_{\text{sep}} T_{r}}{F C_{e} v_{e} e_{v}} & \frac{1}{v_{e} e_{v}} & 0 \\
0 & -\frac{1}{\sqrt{2} R v_{\text{sep}} C_{e}} & 0
\end{pmatrix}, \quad H_{1}^{\text{sep}} = \begin{pmatrix}
0 & -\frac{\omega}{\sqrt{2} R v_{\text{sep}} C_{e}} & 0
\end{pmatrix}^{T}
\]

In (3.25), the parameter matrices \(C_{1}, \ldots, C_{8}\) and \(D_{1}, \ldots, D_{8}\) are given by:

\[
\begin{align*}
C_{1} &= \text{diag}(0, \ldots, 0, 1, 0, 0, 0), \quad C_{2} = \text{diag}(0, 1, 0, 0, 0, -\lambda^{-}, 0, 0), \\
C_{3} &= \text{diag}(0, \ldots, 0, 1, 0), \quad C_{4} = \text{diag}(0, 1, 0, 0, 0, -\lambda^{+}), \\
C_{5} &= C_{7} = \text{diag}(1, 0, \ldots, 0), \quad C_{6} = C_{8} = \text{diag}(1, 0, \ldots, 0), \\
D_{1} &= [0, \ldots, 0, h \cdot (T_{\text{amb}} - T^{-}), 0, 0]^{T}, \\
D_{2} &= [0, \ldots, 0, h \cdot (-T_{\text{amb}} + T^{+})]^{T}, \\
D_{3} &= [-J^{-} / D_{e}^{\text{eff},-} + 0, \ldots, 0]^{T}, \quad D_{4} = [-J^{+} / D_{e}^{\text{eff},+} + 0, \ldots, 0]^{T}, \\
D_{5} &= D_{7} = \text{diag}(0, 1, 0, 1, 1, 1), \\
D_{6} &= \text{diag}(0, D_{e}^{\text{eff},\text{sep}} / D_{e}^{\text{eff},-} + 0, 0, 0, \lambda^{\text{sep}} / \lambda^{-}), \\
D_{8} &= \text{diag}(0, D_{e}^{\text{eff},+} / D_{e}^{\text{eff},\text{sep}}, 0, 0, 0, \lambda^{+} / \lambda^{\text{sep}}).
\]

To present the PDE system precisely and also simplify the notations, the infinite dimensional system (3.23a)-(3.23c) is reformulated within the Hilbert space \(H(D, \mathbb{R}^{n})\). It is a space of \(n\)-dimensional vector functions defined on a spatial interval and being square integrable [101]. Define the state function \(x\) on \(H\) as:

\[
x(t) = \bar{x}(x, r, t), \quad \forall t > 0, \forall (x, r) \in D(x, r),
\]

the operator \(F\) in \(H(D, \mathbb{R}^{n})\) as:

\[
F_{x} = \frac{\partial \bar{x}}{\partial r} + S_{1} \frac{\partial \bar{x}}{\partial x} + F_{2} \frac{\partial^{2} \bar{x}}{\partial r^{2}} + S_{2} \frac{\partial^{2} \bar{x}}{\partial x^{2}}, \\
\forall x \in D(F) = \{x \in H(D, \mathbb{R}^{n}); \text{Eqs: (3.25)}\},
\]
and the output operators as:

\[ Gx = g, \quad Qx = q. \]

Then, the battery system (3.23a)-(3.26) can be represented compactly in the form

\[ \Sigma^1: \quad \dot{x} = Fx + H(x, u), \quad x(0) = x_0, \]
\[ y_c = Gx, \quad y_m = Qx, \]

where \( H(x(t), u(t)) = H(\bar{x}(x, r, t), u(t)) \) and \( x_0 = \bar{x}_0 \).

### 3.3 PDE Model Simplification Via Singular Perturbation Techniques

The initial physics-based model presented in the previous section is capable of capturing battery system dynamics but is too complex for most model-based applications. In the following, we aim to develop a framework for systematic battery model simplification. Assumptions made on battery physical and chemical properties are gradually stated, justified, and remarked.

The modelling complexity is predominantly attributed to the mathematical representation of nonlinear PDEs and coupled electrical dynamics, mass and thermal diffusion, and battery ageing in different time-scales associated with oscillated trajectories. With this in mind, model simplifications are systematically executed and the exact sequence is schematically interpreted in Fig. 3.3. Firstly, by application of a singular perturbation approach, the fast states are eliminated to derive a simplified (reduced) model \( \Sigma^2 \). In the sequel, based on \( \Sigma^2 \), the singular perturbation approach and averaging theory are appropriately combined to decouple the medium and slow time-scale variables. As a consequence, a boundary layer system \( \Sigma^3_m \) and a reduced system \( \Sigma^3 \) are obtained.
3.3.1 Theory For Model Simplification I: Models $\Sigma_f^2$ And $\Sigma_s^2$

In the first stage, model simplification is motivated by insights into the battery dynamics. The electrical dynamics are much faster than the mass and thermal diffusion process. The difference in the magnitudes stems from the small parameters $\varepsilon_{\Phi s}, \varepsilon_{\Phi e}, \varepsilon_{ie}$ in (3.7)-(3.9). Based on the physical meaning of them, these parameters are assumed to have the same magnitude without loss of generality. Define $\varepsilon_1 := \varepsilon_{\Phi s} = \varepsilon_{\Phi e} = \varepsilon_{ie}$, and $\varepsilon_1$ is a small positive parameter. To investigate the time-scale separation, the system (3.29) can
be written in the standard form of singularly perturbed systems

\[ \Sigma^1 : \]

\[
\begin{align*}
\dot{x}_s &= F_s x_s + H_s(x_s, x_f, u), \quad x_s(0) = x_{s0}, \\
\varepsilon_1 \dot{x}_f &= F_f x_f + H_f(x_s, x_f, u), \quad x_f(0) = x_{f0}, \\
y_c &= G[x_s, x_f]^T, \\
y_m &= Q[x_s, x_f]^T,
\end{align*}
\]

(3.30)

where in the negative electrode, the state function \( x \) can be expressed more explicitly to include \( x^- := [C_s^-, C_e^-, T^-, Q_{sr}, R_f]^T \) and \( x_f^- := [\Phi_s^-, \Phi_e^-, i_e^-]^T \). Similar cases can be done for the positive electrode and the separator. The functions \( H_s, H_f \) are continuous and bounded in their arguments for \((x_s, x_f, u) \in \mathcal{H}_s \times \mathcal{H}_f \times U\), where \( \mathcal{H}_s \) and \( \mathcal{H}_f \) are subspaces of \( \mathcal{H} \).

**Assumption 3.1.** Within battery operating processes, the parameter \( \varepsilon_1 \) involved in electrical dynamics satisfies \( \varepsilon_1 \ll 1 \).

**Justification for Assumption 3.1.** Typically, a charge or discharge operation associated with diffusion process and chemical reaction is completed within several minutes or hours. Meanwhile, the signal transmission speeds of potentials and currents are very large quantities leading to time constants of order microseconds [151]. Thus, in comparison with the electrochemical and thermal states which are nominally of the order 1 in \( \Sigma^1 \), the dynamics of potentials and currents result in a sufficiently small \( \varepsilon_1 \).

**Remark 3.1.** With the parameter \( \varepsilon_1 \) justified to be sufficiently small, the model structure of (3.30) guides the use of singular perturbation approaches for model simplification of the battery system. [100] has some results using a singular perturbation approach on other classes of PDE systems. Then, a similar approach was pursued in [99] for photonics applications. However, without rigorous theoretical justification, this work will investigate the validity of singular perturbation techniques via simulations.

**Assumption 3.2.** In the time-scale \( \tau \) measured from the fast electrical dynamics, the variation of the input current, \( u \), is sufficiently slow.
**Justification for Assumption 3.2.** For practical utilisation of Li-ion batteries, due to physical limitation of battery chargers or power converters, the operating current typically changes in a time period of seconds or longer. Compared to the fast dynamics, the change of $u$ is negligibly slow so that it can be considered as a constant in the fast time-scale.

Based on Assumption 3.1, $\epsilon_1$ is set to zero, leading to a fundamental and abrupt change in the dynamic properties of the fast state $x_f$. The differential equation of $x_f$ in (3.30) is replaced by an algebraic equation

$$0 = F_f x_f + H_f(x_s, x_f, u).$$  \hfill (3.31)

The real root of (3.31) is known to exist in the arguments $\forall t > 0$ and $\forall (x, r) \in D(x, r)$ and is derived as $x_f^* = h(x_s, u)$.

As a result, the fast dynamics for $x_f$ are excluded from the original battery system and instead its quasi-steady state $h(x_s, u)$ is considered. The full-order model $\Sigma^1$ is simplified to the slow (reduced) model with respect to the slow states, $x_s$, and is further presented as $\Sigma^2_s$:

$$\dot{x}_s = F_s x_s + H_s(x_s, h(x_s, u), u), x_s(0) = x_{s0},$$ \hfill (3.32a)

$$y_c = G[x_s, h(x_s, u)]^T,$$ \hfill (3.32b)

$$y_m = Q[x_s, h(x_s, u)]^T.$$ \hfill (3.32c)

**Remark 3.2.** The slow model (3.32) with lumped battery degradation and without consideration of temperature dynamics is typically used as a starting point for model order reduction in the literature such as [59, 61]. A complete battery model is considered in this work to demonstrate the use of singular perturbation approaches for PDE-based battery model simplification. The underlying assumption imposed on the simplified model is explicitly stated and justified.

Having established the reduced battery system, now we investigate the boundary layer system with respect to the state variables $x_f$. In the fast time-scale $\tau := (t - t_0)/\epsilon_1$, the
battery model of (3.30) can be readily modified to
\[ \frac{d x_s}{d \tau} = e_1 F_s x_f + e_1 H_s(x_s, x_f, u), \] (3.33a)
\[ \frac{d x_f}{d \tau} = F_f x_f + H_f(x_s, x_f, u). \] (3.33b)

By setting \( e_1 = 0 \), the differential equation (3.33a) is degenerated into
\[ \frac{d x_s}{d \tau} = 0 \Rightarrow x_s(\tau) = x_{s0}. \] (3.34)

Through (3.34), the dynamics of the slow state, \( x_s \), are eliminated by freezing its value on \( x_{s0} \). This allows the battery model \( \Sigma^1 \) to be described by less state variables as well as less governing PDEs.

Based on Assumptions 3.1-3.2, there exists \( u(t) = u(t_0 + e_1 \tau) \approx u(t_0) \). In view of this, the boundary layer method [98] can be applied here to investigate the fast dynamics individually in the battery system. Combining this and (3.33b), the fast state, \( x_f \), can be therefore decoupled from the original model and resulted in the following boundary layer (fast) system
\[ \Sigma_f^2: \quad \frac{d x_f}{d \tau} = F_f x_f + H_f(x_{s0}, x_f, u_0), \quad x_f(0) = x_{f0}. \] (3.35)

To shift the quasi-steady state of \( x_f \) to the origin, we define \( z_f = x_f - h(x_{s0}, u_0) \) and (3.35) can be reformulated as
\[ \frac{d z_f}{d \tau} = F_f(z_f + h(x_{s0}, u_0)) + H_f(x_{s0}, z_f + h(x_{s0}, u_0), u_0). \] (3.36)

**Assumption 3.3.** The boundary layer battery system given in (3.36) is uniformly globally exponentially stable (UGES).

**Justification for Assumption 3.3.** As there is no established theory to analytically justify this assumption, it will be tested numerically in Section 3.3.5.

**Remark 3.3.** Assumption 3.3 for the boundary layer system has the similar structure with the classical singular perturbation theory of Tikhonov [98]. When this assumption holds,
the trajectories of the fast state variables $x_f$ in the original battery model $\Sigma^1$ will exponentially converge to its quasi-steady state governed by (3.31).

In this subsection, the electrical and electrochemical-thermal-ageing phenomena have been identified to exhibit two time-scales. A systematic procedure for developing simplified models with interconnections based on clearly specified assumptions has been proposed.

### 3.3.2 Application To Battery Models: Stage I

With simplified models obtained by using a singular perturbation approach in Section 3.3.1, these theoretical results are applied to the Li-ion battery system.

The reduced battery system, $\Sigma^2$, containing electrochemical, thermal and ageing dynamics, is explicitly provided with battery states and parameters:

\[
\frac{\partial C^+}{\partial t} = D_{s}^{\text{eff,}+} \frac{\partial}{\partial r} \left(r^2 \frac{\partial C^+}{\partial r}\right),
\]

\[
\frac{\partial C^-}{\partial t} = D_{s}^{\text{eff,-}} \frac{\partial}{\partial r} \left(r^2 \frac{\partial C^-}{\partial r}\right),
\]

\[
\frac{\partial C}{\partial t} = \frac{\partial}{\partial x} \left(D_{s}^{\text{eff,}j} \frac{\partial C^j}{\partial x}\right) + \frac{\gamma^j}{F_{\mu_e}^j} \frac{\partial i^j}{\partial x},
\]

\[
\rho^j \frac{\partial T^j}{\partial t} = \lambda_T \frac{\partial^2 T^j}{\partial x^2} - \left(I(t) - i^j(x,t)\right) \frac{\partial \Phi^j}{\partial x} - i^j(x,t) \frac{\partial \Phi^j}{\partial x} + F a^j \eta^j(x,t) + F a^j \Phi^j(x,t) \frac{\partial U^j}{\partial T},
\]

\[
\frac{\partial Q_{sr}(x,t)}{\partial t} = - Fa^j I^j(x,t) + Fa^j \Phi^j(x,t) J_{sr}(x,t),
\]

\[
\frac{\partial R_f(x,t)}{\partial t} = - \frac{M_f}{\rho_f \sigma_f} J_{sr}(x,t).
\]

In the above equations, the fast electrical state variables, namely $\Phi^+_s, \Phi_e^j, i^+_e$, converge to their quasi-steady states instantaneously and are governed by

\[
\frac{\partial \Phi^+}{\partial x} = - \frac{I(t) - i^+_e(x,t)}{\sigma^+},
\]
3.3 PDE Model Simplification Via Singular Perturbation Techniques

\[ \frac{\partial \Phi^\pm_e (x,t)}{\partial x} = - \frac{i_e^\pm (x,t)}{\kappa^l} + \frac{2R \gamma^l T^l}{F} \frac{\partial \ln C^\pm_e (x,t)}{\partial x}, \]
\[ \frac{\partial i^\pm_e (x,t)}{\partial x} = F a^\pm f^\pm (x,t). \]

To explicitly reflect the result of model simplification in (3.35), the fast battery system \( \Sigma_2^f \) representing the electrical dynamics is further obtained in the following dynamic equations in the \( \tau \) time-scale

\[ \frac{\partial \Phi^\pm (x,t)}{\partial \tau} = \frac{\partial \Phi^\pm_e (x,t)}{\partial x} + \frac{i_0 - i^\pm_e (x,t)}{\sigma^\pm}, \]
\[ \frac{\partial \Phi^\pm_i (x,t)}{\partial \tau} = \frac{\partial \Phi^\pm_i (x,t)}{\partial x} + \frac{i^\pm_e (x,t)}{\kappa^l}, \]
\[ \frac{\partial i^\pm_e (x,t)}{\partial \tau} = \frac{\partial i^\pm_e (x,t)}{\partial x} - Fa^\pm f^\pm (x,t), \]

where, the Li-ion molar flux \( J \), exchange current density \( i_0 \) and overpotential \( \eta \) take the form

\[ j^\pm (x,t) = \frac{i^\pm_0}{F} \left( e^{\frac{a^e R T^l}{k T_0}} - e^{\frac{-a^e R T^l}{k T_0}} \right), \]
\[ i^\pm_0 = K^\pm C_{e0}^{a^a} (C_{\text{max}}^\pm - C_s^\pm)^{a^a} C_{\text{max}}^{a^p}, \]
\[ \eta^\pm (x,t) = \Phi^\pm_e (x,t) - \Phi^\pm_i (x,t) - U^\pm (C_{\text{max}}^\pm) - F R f_0 j^\pm (x,t). \]

Here, the system input current, \( I \), and the state variables, \( C_s^\pm, C_e^l, T^l, Q_{sr}, R_f \), are all set to their initial values.

3.3.3 Theory For Model Simplification II: Models \( \Sigma^3_m \) And \( \Sigma^3_s \)

Possible model simplification is further investigated in this subsection for the battery model \( \Sigma^2_s \). The normal intercalation reaction and side reaction occur simultaneously during battery operations. However, the normal diffusion involved in mass and thermal phenomena is much faster than the ageing process that is typically quantified in the magnitude of weeks or months. This difference is essentially attributed by the normal intercalation reaction flux and side reaction flux where \( J_I \gg J_{sr} \). Studies on (3.11) and (3.18)
further show that $i_0 > i_{0SR}$, in which $i_{0SR}$ is a positive parameter in the order of $1e-6$, in contrast the value of $i_0$ is as large as 1 [8]. With this in mind, by defining $e_2 := i_{0SR}$, and the system of (3.30) is rewritten as

$$
\dot{x}_{s'} = e_2 \mathcal{F}_{s'} x_{s'} + e_2 H_{s'}(x_{s'}, x_m, h(x_{s'}, x_m, u), u),
$$
$$
x_{s'}(0) = x_{s'0},
$$
$$
\dot{x}_m = \mathcal{F}_m x_m + H_m(x_{s'}, x_m, h(x_{s'}, x_m, u), u),
$$
$$
x_m(0) = x_{m0},
$$
$$
y_e = \mathcal{G}[x_{s'}, x_m, h(x_{s'}, x_m, u)]^T,
$$
$$
y_m = \mathcal{Q}[x_{s'}, x_m, h(x_{s'}, x_m, u)]^T,
$$

where the state functions $x_{s'} := [C_{s,s'}, Q_{sr}, R_f]^T$ and $x_m := [C_{s,m}, C_e, T]^T$. Given the solid-phase Li-ion concentration, $C_{s,s'}$ simultaneously participates in normal diffusion and SEI film growth, it becomes two states after decomposition, i.e. $C_{s,m}$ and $C_{s,s'}$. The functions $H_{s'}, H_m$ are continuous and bounded in their arguments for $(x_{s'}, x_m, u) \in \mathcal{H}_{s'} \times \mathcal{H}_m \times \mathcal{U}$ and $\mathcal{H}_{s'}, \mathcal{H}_m \subseteq \mathcal{H}$.

**Assumption 3.4.** The parameter $e_2$ involved in battery SOH dynamic characteristics satisfies $e_2 \ll 1$.

**Justification for Assumption 3.4.** This can be readily justified using typical Li-ion batteries. During battery charge or discharge process, the electrochemical and thermal states in a cell often experience significant change within a couple of minutes or hours. By comparison, the degree of battery degradation parameterised by $e_2$ is several orders of magnitude slower than the normal intercalation reaction. Thus, in the time-scale measured by the normal intercalation reaction, the parameter $e_2$ as well as the change of battery SOH is negligibly small.

The battery system of $\Sigma^2_s$ has been identified to exhibit in the medium and slow time-scales corresponding to the electrochemical-thermal and ageing phenomena, respectively. The smallness of $e_2$ permits the SOH to work as a small perturbation for the normal lithium-ion intercalation reaction. According to Assumption 3.4, $e_2$ is assumed to be 0, and as a result the battery system (3.32) is simplified to the following boundary layer
system with respect to the medium time-scale state, \( x_m \)

\[
\begin{align*}
\dot{x}_s &= 0, \quad x_s(0) = x_{s0}, \\
\dot{x}_m &= F_m x_m + H_m(x_s, x_m, h(x_{s0}, x_m, u), u), \\
\Sigma^3_m: \quad x_m(0) &= x_{m0}, \\
y_c &= G[x_{s0}, x_m, h(x_{s0}, x_m, u)]^T, \\
y_m &= Q[x_{s0}, x_m, h(x_{s0}, x_m, u)]^T.
\end{align*}
\tag{3.38}
\]

In the battery system (3.38), the differential equation in terms of \( x_{s0} \) from (3.37) is approximated by an algebraic equation through which the slow time-scale state \( x_{s0} \) is fixed to be a constant, i.e. \( x_{s0} = 0 \). After elimination of the dynamics of \( x_{s0} \), the state \( x_m \) is decoupled from the full-order battery system.

To individually investigate the behaviour of the slow state \( x_{s0} \) that is governed by (3.37), further model simplification is needed, as the medium and slow time-scales are still simultaneously involved and leading to undesirable computational complexity. In the usual practice of singular perturbation approaches, the slow system is derived by approximating the fast dynamics with their quasi-steady state values. However, the dynamics in the medium time-scale do not approach a constant steady state, and this requires an alternative approach to analyse them.

From Assumption 3.2 and its justification, the system input \( u(t) \) is known to behave in the medium time-scale, which is the same as the state \( x_m \) does. The consideration of this and smallness of \( \epsilon_2 \) imposed by Assumption 3.4 motivates us to use the averaging theory of [98, 152] to simplify the process of deriving the slow battery model with the state, \( x_{s0} \).

With this in mind, the solution of \( x_{s0} \) in the battery model (3.37) is considered to obtain by averaging the steady-state behaviour of the medium battery system (3.38). Define a static, average mapping \( x_{s0}(t) \rightarrow \mathcal{H}_{av}(x_{s0}(t)) \) as:

\[
\mathcal{H}_{av}(x_{s0}) := \lim_{T_s \to \infty} \frac{1}{T_s} \int_0^{T_s} F_s x_{s0} + H_s(x_{s0}, x_m^*, h(x_{s0}, x_m^*, u^*), u^*) dt.
\tag{3.39}
\]

where \( u^*(t) \) is the specified system input, and \( x_m^*(t) \) is the solution of the battery model.
(3.38) under the specified system input $u^*(t)$. In this general average mapping, it is worth mentioning that $\mathcal{H}_{av}$ is independent of $\varepsilon_2$ and $(x_m(0), u(0)) \in \mathcal{H}_m \times U/\{0\}$.

Therefore, in the slow time-scale $\sigma = \varepsilon_2 t$, the dynamics of $x_{s'}$ from (3.37) are approximated by the following slow (average) battery system

$$\Sigma^3_s : \quad \frac{d x_{s'}}{d \sigma} = \mathcal{H}_{av}(x_{s'}), \quad x_{s'}(0) = x_{s'0}, \quad (3.40a)$$

$$y_c = G[x_{s'}, x_m^*, h(x_{s'}, x_m^*, u^*)]^T. \quad (3.40b)$$

To enable the above utilisation of averaging theory for battery model simplification, an assumption is posed on the medium time-scale dynamics. We define a manifold $\bar{x}_m = h'(x_{s'0}, u)$ to present the quasi-steady state of the medium state variables $x_m$ from battery system of (3.38). Then, $\bar{x}_m$ is the solution of

$$\dot{\bar{x}}_m(t) = F_m \bar{x}_m(t) + H_m(\bar{x}_m(t), u(t)), \quad (3.41)$$

where $u(t) = u(t + kT_c), \forall k \in \{0, 1, \cdots, N\}$. 

**Assumption 3.5.** There exists an integral manifold $z_m := x_m - \bar{x}_m$, where $\bar{x}_m$ is defined in (3.41), and a class-$KL$ function $\beta_h$ such that, for all initial conditions in the domain $\mathcal{D}$, the solutions of $x_m$ in $\Sigma^3_m$ exist and satisfy

$$|z_m| \leq \beta_h(|z_m(0)|, t), \forall t \geq 0. \quad (3.42)$$

**Justification for Assumption 3.5.** It is quantitatively investigated via simulations in Section 3.3.5.

The state of $x_{s'}$ has been decoupled from the original highly nonlinear battery system. To solve the developed mapping $x_{s'} \rightarrow \mathcal{H}_{av}(x_{s'})$, only the behaviour of $x_m$ governed by the battery system $\Sigma^3_m$ is required. Once such a mapping $\mathcal{H}_{av}$ is obtained, the slow battery system in (3.40a)-(3.40b) can be easily implemented.

**Remark 3.4.** To investigate battery SOH characteristics through the slow battery system, the input current $u(t)$ needs to be provided in advance. For any given profile of $u^*(t)$
including periodically or non-periodically time-varying input current, there is a corresponding mapping defined by (3.39) and the averaging theory can be used to simplify the battery model.

### 3.3.4 Application To Battery Models: Stage II

By applying the theoretical results in Subsection 3.3.3 to the Li-ion battery system, the corresponding medium time-scale battery model, $\Sigma^3_{m}$, is provided in complete detail as

\[
\begin{align*}
\frac{\partial C^+_{i,n}(x,r,t)}{\partial t} & = \frac{D^e_{s,+}}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial C^+_{s,m}(x,r,t)}{\partial r} \right), \\
\frac{\partial C^-_{s}(x,r,t)}{\partial t} & = \frac{D^e_{s,-}}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial C^-_{s,m}(x,r,t)}{\partial r} \right), \\
\frac{\partial C^0_{i}(x,t)}{\partial t} & = \frac{\partial}{\partial x} \left( \frac{D^e_{c,i} \partial C^0_{i}(x,t)}{\mu_i^c} \right) + \frac{\eta^i}{F \mu_i^c} \frac{\partial i^i}{\partial x} \\
\rho C^j \frac{\partial T^j(x,t)}{\partial t} & = \lambda^j \frac{\partial^2 T^j(x,t)}{\partial x^2} - (I(t) - i^i(x,t)) \frac{\partial \Phi^j_s(x,t)}{\partial x} - i^i(x,t) \frac{\partial \Phi^j_c(x,t)}{\partial x} + F a \int_T^j \eta^j(x,t) T^j(x,t) \frac{\partial U^j}{\partial T},
\end{align*}
\]

where, $j^j(x,t), i^i_0(x,t), \eta^\pm(x,t)$ are formulated by the following algebraic equations

\[
\begin{align*}
j^j_0(x,t) & = \int_0^j \left( e^{\frac{\alpha_p \eta^\pm(x,t)}{2}} - e^{\frac{-\alpha_p \eta^\pm(x,t)}{2}} \right), \\
\int_0^j \left( e^{\frac{\alpha_p \eta^\pm(x,t)}{2}} - e^{\frac{-\alpha_p \eta^\pm(x,t)}{2}} \right), \\
i^i_0(x,t) & = C^+_{s,s}(x,t) \alpha_n (C^\pm_{s,m}(x,t) - C^\pm_{s,s,n}(x,t)) \alpha_n C^\pm_{s,s}(x,t) \alpha_p \\
\eta^\pm(x,t) & = \Phi^\pm_0(x,t) - \Phi^\pm_0(x,t) - U^\pm(C^\pm_{s,s}(x,t)) + FR_f(x,t_0) j^j_0(x,t).
\end{align*}
\]

In this battery model, the electrochemical-thermal dynamics are described, in which the slow state variables namely $C_{s,s}, Q_{str}, R_f$, are all constant.

After sequential elimination of the electrical, electrochemical, and thermal dynamics, the slow battery model $\Sigma^3_{s}$ describing battery ageing behaviour has been obtained in (3.40a).
and its dynamical equations in the corresponding Euclidian space are provided as

\[
\frac{\partial Q_{sr}(x,t)}{\partial t} = e_2 a^{-} A^{-} L^{-} \lim_{T_s \to \infty} \frac{1}{T_s} \int_{0}^{T_s} e^\phi(x,t) dt,
\]

\[
\frac{\partial R_f(x,t)}{\partial t} = e_2 \frac{M_f}{F_p \sigma_f} \lim_{T_s \to \infty} \frac{1}{T_s} \int_{0}^{T_s} e^\phi(x,t) dt,
\]

\[
\frac{\partial C_{sr}(x,r,t)}{\partial t} = \frac{e_2 a^{-} A^{-} L^{-}}{Q_{\text{max}} T_s} \lim_{T_s \to \infty} \int_{0}^{T_s} C_{sr,m}^+(x,r,t) e^\phi(x,t) dt
\]

\[
+ \frac{D_{\text{eff},+}^2}{r^2 Q_{\text{max}} T_s} \lim_{T_s \to \infty} \int_{0}^{T_s} Q_{sr}(x,t_0) \frac{\partial}{\partial r} \left( r^2 \frac{\partial C_{sr,m}^+(x,r,t)}{\partial r} \right) dt,
\]

where, the function \(\phi(x,t)\) has the form of

\[
\phi(x,t) = -a_{sr} F \eta^*(x,t)
\]

and the overpotential \(\eta^*(x,t)\) is governed by

\[
\eta^*(x,t) = \Phi_s^*(x,t) - \Phi_s^-(x,t) - U_{sr} - FR_f(x,t_0)J_f^*(x,t).
\]

So far, the electrical, electrochemical, thermal, and ageing dynamics within the initial battery model, \(\Sigma^1\), have been separated with the simplified PDE models obtained, i.e. \(\Sigma^2_f\), \(\Sigma^2_s\), \(\Sigma^3_m^\star\), and \(\Sigma^3_s\). The applied Assumptions 3.1-3.5 are similar with the time-scale separation procedure of classical singular perturbation approaches and the averaging theory presented in [98, 152]. Given there is no formal tool to rigorously justify this model simplification framework, numerical solutions will be performed in Section 3.3.5 to validate these simplified models and their associated assumptions.

For model-based estimation and control, the processors in battery management systems and chargers must be able to predict the system behaviours using the internal model in real-time. However, these infinite dimensional models obtained above may be impractical to implement in such processors. In this regard, model order reduction with the aid of numerical techniques is explored. The models, \(\Sigma^2_f, \Sigma^2_s, \Sigma^3_m^\star, \Sigma^3_s\), serve as starting points for further model reduction depending on the characteristics required in the model and its subsequent use.
3.3 PDE Model Simplification Via Singular Perturbation Techniques

In the following sections, it is assumed that the battery state estimation and control over relatively short durations is the principal objective. Hence, the electrochemical-thermal model, $\Sigma_m^3$, is considered as the base model for further reduction. A modelling methodology covering a large range of applications is developed through systematically simplifying the PDE systems. Specifically, finite difference method and polynomial approximations are employed. The effects of grid resolution in discretisation and polynomial order on the complexity and accuracy of reduced order models have been investigated, providing insight into the minimum modelling requirements at different charge rates.

3.3.5 Validation of PDE model simplification

The approximating properties of the simplified models, $\Sigma_f^2$, $\Sigma_m^3$, and $\Sigma_2^3$, obtained in Section 3.1 in comparison to the full-order model as well as their underlying assumptions are investigated.

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Unit</th>
<th>Anode</th>
<th>Separator</th>
<th>Cathode</th>
</tr>
</thead>
<tbody>
<tr>
<td>$brug$</td>
<td>–</td>
<td>4</td>
<td>4</td>
<td>4</td>
</tr>
<tr>
<td>$C_{s_{\text{max}}}$</td>
<td>mol/m$^3$</td>
<td>30555</td>
<td>–</td>
<td>50555</td>
</tr>
<tr>
<td>$C_{s_{0}}$</td>
<td>mol/m$^3$</td>
<td>917</td>
<td>–</td>
<td>48977</td>
</tr>
<tr>
<td>$C_{e_{0}}$</td>
<td>mol/m$^3$</td>
<td>1000</td>
<td>1000</td>
<td>1000</td>
</tr>
<tr>
<td>$c_T$</td>
<td>J/K·kg</td>
<td>1750.3</td>
<td>1329.3</td>
<td>2041.6</td>
</tr>
<tr>
<td>$D_s$</td>
<td>m$^2$/s</td>
<td>$3.9\times10^{-14}$</td>
<td>–</td>
<td>$1.0\times10^{-14}$</td>
</tr>
<tr>
<td>$D_e$</td>
<td>m$^2$/s</td>
<td>$7.5\times10^{-10}$</td>
<td>$7.5\times10^{-10}$</td>
<td>$7.5\times10^{-10}$</td>
</tr>
<tr>
<td>$h_T$</td>
<td>W/(K·m$^2$)</td>
<td>5</td>
<td>–</td>
<td>5</td>
</tr>
<tr>
<td>$i_{\text{bar}}$</td>
<td>A/m$^2$</td>
<td>$1.5\times10^{-6}$</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>$L$</td>
<td>μm</td>
<td>88</td>
<td>20</td>
<td>80</td>
</tr>
<tr>
<td>$k$</td>
<td>Am$^{2.5}$/mol$^{1.5}$</td>
<td>$4.854\times10^{-6}$</td>
<td>–</td>
<td>$2.252\times10^{-6}$</td>
</tr>
<tr>
<td>$R_p$</td>
<td>μm</td>
<td>2</td>
<td>–</td>
<td>2</td>
</tr>
</tbody>
</table>
To validate the established model simplification framework, different Li-ion cell chemistries can be used. Typical battery parameters listed in Table 3.3.5 are adopted from [8, 15, 17, 43] and employed here. These parameters are related to a cylindrical LiCoO$_2$/LiC$_6$ battery cell with 1.8 Ah nominal capacity.

The constant-current constant-voltage (CCCV) protocol is commonly used in current battery management systems and is considered here for battery model simulation. In this method, batteries are charged with an initial constant current for a period until its terminal voltage reaches a predefined threshold, and then the terminal voltage remains at its maximum value until the current drops below a given threshold. The design parameters in CCCV charging including initial current and current/voltage thresholds are respectively set up as 1.5C, 4.2V, and 360mA. “C” is a normalised metric indicating the input current utilised for operations in amperes relative to the battery maximum rated capacity in ampere-hours. As such, 1C implies a battery can be fully charged/discharged in

<table>
<thead>
<tr>
<th>$T_0$</th>
<th>K</th>
<th>298</th>
<th>298</th>
<th>298</th>
</tr>
</thead>
<tbody>
<tr>
<td>$U_{sr}$</td>
<td>V</td>
<td>0.4</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>–</td>
<td>0.5</td>
<td>–</td>
<td>0.5</td>
</tr>
<tr>
<td>$\alpha_{sr}$</td>
<td>–</td>
<td>0.5</td>
<td>–</td>
<td>0.5</td>
</tr>
<tr>
<td>$\partial U / \partial T$</td>
<td>J/mol·K</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>–</td>
<td>0.637</td>
<td>0.637</td>
<td>0.637</td>
</tr>
<tr>
<td>$\lambda_T$</td>
<td>W/K·m</td>
<td>0.809</td>
<td>1.172</td>
<td>0.553</td>
</tr>
<tr>
<td>$\rho$</td>
<td>kg/m$^3$</td>
<td>1233.3</td>
<td>1108.7</td>
<td>1356.7</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>S/m</td>
<td>100</td>
<td>–</td>
<td>100</td>
</tr>
<tr>
<td>$\mu_s$</td>
<td>–</td>
<td>0.49</td>
<td>–</td>
<td>0.59</td>
</tr>
<tr>
<td>$\mu_e$</td>
<td>–</td>
<td>0.485</td>
<td>1</td>
<td>0.385</td>
</tr>
<tr>
<td>$A$</td>
<td>m$^2$</td>
<td>0.0596</td>
<td>0.0596</td>
<td>0.0596</td>
</tr>
<tr>
<td>$F$</td>
<td>C/mol</td>
<td></td>
<td></td>
<td>96487</td>
</tr>
<tr>
<td>$R$</td>
<td>J/mol·k</td>
<td></td>
<td></td>
<td>8.314</td>
</tr>
</tbody>
</table>
one hour. This set of parameters represents one of the fastest charging speeds as well as the crucial operating conditions utilised for most applications in the market including electric vehicles. Following the charging operation, a small constant current of 0.5C is applied for discharging with 3.2V as the terminal condition.

Other charging schemes like the constant-current (CC) in different C-rates, multistage CC [21], and the Urban Dynamometer Driving Schedule (UDDS) charging are also employed and these may warrant higher operating C-rates. To test the model fidelity at high C-rates, the initial UDDS data in [153] are augmented by a factor of three, through which the maximum charging rate has been pushed to 3C. For CC charging, 1C, 2C, 3C, and 5C are respectively considered.

To demonstrate multiple battery charging and discharging cycles through methods of simulation, an assumption regarding the input currents is applied:

**Assumption 3.6.** The input current $u(t)$ for a battery system is known a priori and is periodically time-varying at different operating cycles.

**Remark 3.5.** The validity of Assumption 3.6 is tightly related to the practical utilisation of a Li-ion battery. The charging process at each operating cycle can be controlled with its strategies predetermined in battery chargers. This makes it plausible to assume the system input $u(t)$ is identical for battery charging at different cycles. For discharging cases, typical battery-powered devices are generally repeatedly used for similar tasks like UDDS for electric vehicles. Therefore, this is a good assumption for the purpose of simplifying simulation process.

To solve the PDEs, numerical methods including the finite differencing detailed in [40] is adopted leading to a system consisting of a set of differential-algebraic equations (DAEs). Modelica is an object-oriented language for efficient simulation of physical systems [154]. The non-causal modelling property makes it well suited for solving DAEs [155]. In light of this, simulations for validation of the assumptions and simplified models are implemented in Modelica. In this process, the integrator DASSL associated with Newton’s method is used.

The estimate error in SOC, SOH, or state variables is defined in the following form
\[ \beta(t) := (\omega_r(t) - \omega(t))/\omega_{\text{max}}, \]

where \(\omega_r(t)\) and \(\omega(t)\) represent solutions from the simplified models and benchmark model respectively, and \(\omega_{\text{max}}\) is the maximum value of \(\omega(t)\).

**Justification for Assumption 3.3**

To justify Assumption 3.3 as well as to study trajectories of the fast state variables \(x_f\), the battery model \(\Sigma_f^2\) is employed. In the fast time-scale \(\tau\), the sampling time is chosen as \(1 \times 10^{-8}\) second which ensures the maximum error in state prediction introduced by temporal discretisation is within 0.01%. Based on the theoretical results in (3.31)-(3.36), simulations are implemented where the battery is charged in 1.5C within the considered time interval, and the results are illustrated in Fig. 3.4.

It can be clearly seen that the dynamics of the fast states, \(\Phi_s\), \(\Phi_e\), and \(i_e\), all decay very quickly (within 0.0002s). These states exponentially converge to their steady states in...
the corresponding time-scale. The obtained results illustrate the existence of different

time-scales within the battery internal states and also qualitatively justify Assumption 3.3.

**Justification for Assumption 3.5**

To verify Assumption 3.5 and study characteristics of the state variables $x_m$ individually,
simulations are implemented by using the battery system of (3.38) and its quasi-steady
state governed by (3.41).

The sampling time in studying the medium states corresponding to Li-ion diffusion dy-
Battery Modelling

Figure 3.6: Procedure for battery charging strategy evaluation using the simplified models, $\Sigma^3_m$ and $\Sigma^3_s$.

Dynamics is chosen as 1s which also ensures the maximum error in state prediction introduced by temporal discretisation within 0.01%. To investigate the trajectories of $x_m$ from the model $\Sigma^3_m$ and its quasi-steady state, different thresholds of the terminal voltage during operations can be chosen. Fig. 3.5 provides the simulation results regarding the states, $C^+_{\ell}$ and $C^+_{e}$, for five operating cycles with 4V and 3.6V as the voltage limits.

The medium state variables are shown to almost periodically vary in the course of battery operations. Although large deviations between $x_m$ and $\bar{x}_m$ are detected in the beginning, $x_m - \bar{x}_m$ is asymptotically reducing and approaching to zero at about 7000s. This result echoes our previous analysis on the medium time-scale states used in justification of Assumption 3.1 and Assumption 3.4. Further observation finds that these states converge to their quasi-steady states which are presented as periodically time-varying curves. Therefore, Assumption 3.5 that behind the simplified models, $\Sigma^3_f$ and $\Sigma^3_s$, addressing the stability property of the system $\Sigma^3_m$ is plausible.
Justification for The Simplified models

So far, all the assumptions behind the proposed model simplification framework have been justified. In the following, the closeness of solutions of the simplified models and original model is examined. Given the fast electrical dynamics exhibit in microseconds, the full battery model that includes all the dynamics is involved in numerical stiffness issue and is impractical to simulate. In view of this, to study the behaviours of $x_m$ and $x_s$, the model $\Sigma^2_s$ that well approximates the original model in the slow and medium time-scales is used as a test bed. The sampling time in this process is chosen as 1s, as the same as we did in Justification for Assumption 3.5.

To examine the model fidelity under the specified CCCV, a CC or UDDS charging strategy, the following evaluation procedure using the averaged system $\Sigma^3_s$ and the simplified model $\Sigma^3_m$ from the established framework is proposed. Here, degradation to 80% of the initial SOH is used to quantify the battery’s lifetime.

To illustrate the implementation of the simplified models, $\Sigma^3_m$ and $\Sigma^3_s$, for investigation of battery SOH characteristics, a flow chart depicting this sequence is provided in Fig. 3.6.

Fig. 3.7 shows the simulation results under the CCCV charging. It can be seen that a good agreement between the model $\Sigma^3_m$ and the benchmark $\Sigma^2_s$ is obtained for all the medium time-scale states including $C_{\pm}^e$, $C_{\pm}^i$, and $T_{\pm,sep}$. The electrolyte concentrations whose dynamics are often ignored at low charging rates, e.g. [119], are studied. Here it is demonstrated that their changes in both the negative and positive electrodes are as large as 70% and are thus not negligible under the applied charging conditions. Indeed, the developed simplified model $\Sigma^3_m$ accurately captures the electrolyte concentration characteristics. The approximation error in SOC prediction is seen to be less than 0.2%. Further examination of this model under UDDS and CC charging protocols are given in Fig. 3.9-3.8a, where small errors are again observed. At this scale of modelling error and over the time intervals of the driving cycles, the derived medium time-scale model $\Sigma^3_m$ is able to capture the electrochemical-thermal dynamics well.

In some circumstances, such as limited operating range of the cell, further model reduc-
Figure 3.7: Validation of the simplified battery models $\Sigma^3_m$ and $\Sigma^3_s$ in terms of the state variables, $C_s, C_e, T$, and the outputs, SOC and SOH under the CCCV charging test.
3.3 PDE Model Simplification Via Singular Perturbation Techniques

Figure 3.8: Validation for $\Sigma^3_m$ and $\Sigma^3_s$ under the 1, 2, 3, and 5C-rate CC charging schemes. a) the battery is charged to 4.2V. b) the battery is charged with 10% SOC increase and then discharged under 0.5C to 3.2V in each cycle.

The percentage error in SOH prediction under the CCCV charging approach is plotted in Fig. 3.7f. For automotive applications, 20% capacity loss from the initial maximum capacity is often considered as the end of battery’s life (EOL) which corresponds to 223 operating cycles in this case. Although it becomes larger with the increased cycles, the modelling error in SOH at EOL is less than 5%. Such a small magnitude of error tells that the SOH dynamics can be well approximated by the developed slow model $\Sigma^3_s$. Further validations of this model under CC charging in different C-rates are given in Fig. 3.8b. These show the model $\Sigma^3_s$ is able to follow the higher order model $\Sigma^2_s$ over a range of operating rates including the case of 5C. In addition, the ageing indicated by the sim-
The model $\Sigma^3_m$ retains two spatial dimensions, i.e. $x$ and $r$. It is noticed that for each electrode, except Li-ion concentration of solid phase $C_s(x,r,t)$ which involves $r$, the other equations are all with respect to $x$ and $t$. However, the mixed spatial scales result in significant computational overhead, so there is motivation to eliminate the final resolution, $r$, if possible.

An exact analytical solution of (3.4) is provided by [157] as

$$C_s(x,r,t) = C_{s0} - \frac{I(x,t)\rho}{D_{s eff}} \left[ 3\tau + \frac{1}{10} \left( \frac{5r^2}{R_p^2} - 3 \right) \right]$$
3.4 Model Simplification Via Spatial Dimension Reduction

\[
\frac{2J(x, t) \cdot R_P^2}{D_{eff}^s r} \sum_{n=1}^{\infty} \frac{\sin(\lambda_n r / R_p) \exp \left(-\lambda_n^2 \tau\right)}{\lambda_n^2 \sin(\lambda_n)},
\]  

(3.43)

where, \( \tau = D_{s eff}^s / R_P^2 \), \( \lambda_j \) are the positive eigenvalues of \( \lambda_j = \tan(\lambda_j) \), and \( j = 1, 2, \ldots \).

Equation (3.43) involves infinite series and is difficult to be directly used for solving the presented battery model. This motivates to find a possible approximation of \( C_s \), expressed as \( \hat{C}_s \). On the other hand, to capture battery electrochemical behaviours and predict SOC, the concentration states of interest in each solid particle are the surface concentration \( C_{ss}(x, t) \) and average concentration \( \bar{C}_s(x, t) \). With this in mind, an attempt to replace \( C_s(x, r, t) \) by \( C_{ss}(x, t) \) and \( \bar{C}_s(x, t) \) is conducted by assuming:

**Assumption 3.7.** *At a constant charge rate, \( \forall \varepsilon > 0, \exists n(\varepsilon, I) \in \mathbb{Z} \) satisfying*

\[
||C_s(x, r, t) - \hat{C}_{s,n}(x, r, t)||_2 < \varepsilon.
\]  

(3.44)

**Remark 3.6.** A solution for \( \hat{C}_{s,n}(x, r, t) \) was obtained by approximating the solid phase Li-ion concentration with the following polynomial form [70]

\[
\hat{C}_{s,n}(x, r, t) = a_1(x, t) + a_2(x, t) \frac{r^2}{R_P^2} + \cdots + a_n(x, t) \frac{r^{2n}}{R_P^{2n}}.
\]  

(3.45)

By substituting (3.45) into (3.4) and employing the volume-average integration along \( r \), the two spatial dimensional (2D) second-order PDE, (3.4), in each electrode can be approximated by a first-order PDE and an algebraic equation in terms of \( C_{ss}(x, t) \) and \( \bar{C}_s(x, t) \), which are given in the form of

\[
\partial C_s(x, t) / \partial t = \frac{3J(x, t)}{R_P},
\]  

(3.46)

\[
C_{ss}(x, t) = C_{s0} - \frac{J(x, t) \cdot R_P (3\tau + 0.2)}{D_{eff}^s} + \frac{J(x, t) \cdot R_P}{D_{eff}^s} \sum_{n=1}^{\infty} \frac{2 \exp \left(-\lambda_n^2 \tau\right)}{\lambda_n^2}.
\]  

(3.47)

For convenience in the statement, it is defined \( \theta(x, t) := J(x, t)R_P / D_{eff}^s \). In consideration of polynomials in different order in (3.47), a set of approximations for (3.47) can be ob-
tained. For instance, through utilising the second, fourth, and sixth-order polynomials for (3.45), the \( C_{ss}(x,t) \) can be approximated by [70]

\[
C_{ss,1}(x,t) = C_{s0} - \vartheta(x,t) \left( 3\tau + 0.2 \right),
\]

\[ (3.48a) \]

\[
C_{ss,2}(x,t) = C_{s0} - \vartheta(x,t) \left( 3\tau + 0.2 - 2e^{-35\tau}/35 \right),
\]

\[ (3.48b) \]

\[
C_{ss,3}(x,t) = C_{s0} - \vartheta(x,t) \left( 3\tau + 0.2 - 0.1135e^{-100.123\tau} \right)
+ \vartheta(x,t) \left( 0.0864e^{-18.877\tau} \right).
\]

\[ (3.48c) \]

It is worth mentioning that the polynomial order, \( n \) is the tuning parameter for these polynomial approximations to reflect concentration dynamics. The accuracy and application of battery models yielded by different order polynomials, referred to as \( \Sigma^4_m \), will be investigated in the next section.

### 3.5 Model Simplification Via Spatial Discretisation

Since the model \( \Sigma^4_m \) resulted from Section 3.4 is still PDE-based, to obtain ODE-based models, we explore the necessary dimensionality of lumped parameter approximations to the model. A possible approach is to approximate these 1D PDEs by a set of ODEs by using finite difference method.

In this course, each domain \( j \), i.e. the negative electrode, separator, and positive electrode, is discretised into \( N \) segments along \( x \)-direction with the grid resolution obtained as \( \Delta x^j = L^j/N \). The discrete state \( x_N^j(i,r,t) \) is assumed to be uniform within the segment \( i \) and \( i = 1, 2, \cdots, N \). The following assumption underlying the length scale separation within battery state variables is introduced:

**Assumption 3.8.** Given \( \epsilon_1 > 0, \exists N^* \in \mathbb{Z}_+ \), such that \( \forall N \geq N^*, \) the states \( x(x,r,t) \) and \( x_N^j(i,r,t) \) satisfy

\[
\left| \frac{x(x,r,t) - x_N^j(i,r,t)}{x(x,r,t)} \right| < \epsilon_1,
\]
\[ \forall i = 1, \cdots, N, \forall t, \text{ and } \forall x \in [(i - 1)/L, i/L]. \]

**Remark 3.7.** Assumption 3.8 is a standard spatial discretisation requirement which implies better approximations of the PDE-system can be obtained with greater numbers of ODEs. As a result, each state in infinite dimension is approximated by \( N \) states governed by \( N \) ODEs.

Based on Assumption 3.8, the battery model \( \Sigma_n^4 \) can be approximated the following ODE-based model \( \Sigma_n^5 \):

\[
d\bar{C}_s(i) = \frac{3J(i)}{R_p}, \tag{3.49a}
\]

\[
C_{ss,n}(i) := C_s(i, R_p), \tag{3.49b}
\]

\[
\mu_e \frac{dC_e(i)}{dt} = D_{\text{eff}} C_e(i + 1) - 2C_e(i) + C_e(i - 1) + \frac{t_0}{\varepsilon} [i_e(i + 1) - i_e(i)], \tag{3.49c}
\]

\[
\Phi_e(i + 1) - \Phi_e(i) = -\frac{i_e(i)}{\sigma(i)}, \tag{3.49d}
\]

\[
\frac{\Phi_e(i + 1) - \Phi_e(i)}{\Delta x} = -\frac{i_e(i)}{\kappa(i)} + \frac{2RT(i)t_0}{F} \ln C_e(i + 1) - \ln C_e(i), \tag{3.49e}
\]

\[
\frac{i_e(i + 1) - i_e(i)}{\Delta x} = aF_{J(i)}, \tag{3.49f}
\]

\[
J(i) = \frac{i_0(i)}{F} \left[ \exp \left( \frac{\alpha_F \eta(i)}{RT(i)} \right) - \exp \left( -\frac{\alpha_F \eta(i)}{RT(i)} \right) \right], \tag{3.49g}
\]

\[
\rho c_T \frac{dT(i)}{dt} = \lambda_T T(i + 1) - 2T(i) + T(i - 1) + aF_{J(i)} \left( \eta(i) + T(i) \frac{\partial U}{\partial T} \right) - \frac{i_e(i)}{\Delta x} \left( \Phi_e(i + 1) - \Phi_e(i) \right) - \frac{i_s(i)}{\Delta x} \left( \Phi_s(i + 1) - \Phi_s(i) \right). \tag{3.49h}
\]

As a result, the model consisting of \( 8N \) ODEs and \( 11N \) algebraic equations (AEs) associated with \( 19N \) states in the entire battery region is obtained. To investigate the applications of this proposed model at different charge rates, \( N \) is a tuning parameter.

Typical battery parameters like those observed in [15, 17] lead to separation of length scales between some states in (3.49a)-(3.49h). To capture this effect, the \( 19N \) state equations will be considered with different level of discretisation, particularly focusing on the temperature and electric potential. This leads to \( 14N + 2N_T + N_T \) total equations, with now
three discretisation variables to be determined. This model is referred to as $\Sigma_m^5$.

To evaluate the proposed model, $\Sigma_m^5$, over the entire possible operating domain, the inputs are constant currents ranging from 0.5C to 10C. The ambient temperature and initial battery temperature are both set at 25°C. Again, typical Li-ion battery parameters can be used here. In the following simulations, the parameters are taken from [15, 17] and are related to a prismatic LiMnO$_4$/LiC$_6$ battery.

The proposed model (3.49) for a given ($N$, $N_F$, $N_T$ and $n$) set is compared with the initial model $\Sigma_m^3$. During each cycle, fully charged Li-ion cells are discharged from 4.2V to 2.8V using both models. The terminal voltages sampled at 10 Hz, leading to $V_k$ and $\tilde{V}_k$ for the proposed model and PDE model, respectively. The normalised root-mean-square (RMS) error in voltage estimation is consequently defined as

$$\beta := \frac{\sqrt{\sum_{k=1}^{M} (V_k - \tilde{V}_k)^2}}{\sum_{k=1}^{M} \tilde{V}_k} \times 100\%. \quad (3.50)$$

**Minimal States of Electric Potential, $N_F$**

The electric potential gradient in solid particles is examined with a view to determining the minimal $N_F$, at various charge rates. Define the discretised spatial approximation of $\Phi_s$ and the resulting approximation error as:

$$\Phi_s^d(i,t) := \frac{N_F}{T} \int_{i/N_F}^{(i+1)/N_F} \Phi_s(x,t)dx, x \in [i, i + 1] \frac{I}{N_F}, \quad (3.51)$$

$$\Delta \Phi_s(x,t) := \Phi_s(x,t) - \Phi_s^d(i,t), \forall i \in [1, N_F] \cap \mathbb{Z}. \quad (3.52)$$

**Lemma 3.1.** The discretised $\Phi_s^d$ model with $N_F$ satisfies

$$|\Delta \Phi_s(x,t)| \leq \varepsilon_{\Phi_s} \quad (3.53)$$

and $\varepsilon_{\Phi} := I(t)/\sigma N_F$, where $\sigma$ represents the minimum value of $\sigma(T)$ (at initial temperature).
Proof. From equation (3.7), \( \Phi_s \) is monotonic in \( x \), such that the error in any given element \([i, i+1] l / N_{\Phi}\) is

\[
|\Delta \Phi_s(x, t)| \leq |\Phi_s((i+1)l / N_{\Phi}) - \Phi_s(il / N_{\Phi})|. \tag{3.54}
\]

By integration on both sides of (3.7) across one element, we obtain

\[
\int_{il / N_{\Phi}}^{(i+1)l / N_{\Phi}} \frac{\partial \Phi_s(x, t)}{\partial x} \, dx = \int_{il / N_{\Phi}}^{(i+1)l / N_{\Phi}} -\frac{i_s(x, t)}{\sigma(T)} \, dx,
\]

\[\epsilon^{\Phi}_i := |\Phi_s((i+1)l / N_{\Phi}, t) - \Phi_s(il / N_{\Phi}, t)| \leq \int_{il / N_{\Phi}}^{(i+1)l / N_{\Phi}} \frac{i_s(x, t)}{\sigma(T)} \, dx \leq \left| \frac{I(t)}{\sigma N_{\Phi}} \right|. \tag{3.55}\]

So, \( \epsilon^{\Phi} := \max_i \epsilon^{\Phi}_i = I(t)l / (\sigma N_{\Phi}) \).

Remark 3.8. Using typical battery parameters of [15] and considering 10C operation rate, the error in the negative electrode is bounded by \( N_{\Phi} \epsilon^{\Phi}_N \leq 0.541 \text{mV} \), which corresponds to a relative error of 0.06\%. Consequently, the choice \( N_{\Phi}=1 \) seems reasonable. Conversely, in the positive electrode, the reduced conductivity relative to the negative electrode leads to \( N_{\Phi} \epsilon^{\Phi}_P \leq 53.96 \text{mV} \), on a relative error 1.324\%. To ensure high accuracy of the electric potential in this electrode, \( N_{\Phi} = N_{\Phi}^{\eta} \) is chosen to be \( N \).

Remark 3.9. Inaccuracies in \( \Phi_s \) can also potentially impact other states through the coupled dynamics evident in (3.12) and (3.15). However, a first order Taylor series expansion of these equations with perturbed \( \Phi_s \) exhibits only negligible effects on \( \eta \) and \( T \) for realistic parameter values.

Minimal Temperature States, \( N_T \)

The thermal phenomena are heavily coupled with electrochemical reactions, so it is impractical to obtain \( \epsilon_T \) by analytical deduction in a way similar to Section 3.5. In light of this, the temperature distribution along the \( x \) coordinate in an electrode pair can be derived from simulations based on the PDE model. To gain the temperature profile across an entire cell, which consists of \( L/l \) electrode pairs, the heat generation rate is
assumed to be uniform as per [45] and scaled by $q(t)L/l$. The typical heat transfer coefficient ($h$) between the cell and ambient can be 5W/m$^2$·K (normal convection); 30 or 100W/m$^2$·K (forced convection). To explore the extreme temperature difference between battery surface and centre, $h$ is chosen to be 100, the heat transfer coefficient and thermal conductivity are $h=100$W/m$^2$·K, $\lambda_T=0.99$W/m·K. The resulting cell temperature profiles at different current rates are shown in Fig. 3.10.

It is apparent that the peak temperature difference in the cell is bounded by $\Delta T \leq 4$K, which corresponds to a relative error (defined as $\Delta T/\bar{T}$) of 1.24%. Moreover, the lower $h$ and current rate are adopted, the smaller error will be obtained. After also confirming negligible coupling effects in parameters $D_s^{eff}$, $D_e^{eff}$, $\sigma$ and $k$, and states $J$, at this scale of temperature error, the temperature gradient in the cell is evidently negligible. A good approximation can be therefore achieved by the lumped parameter approach with $N_T = 1$ and $T(x,t) = T(t)$. 

Figure 3.10: Temperature profiles in a battery cell at 1, 3, 5 and 10C discharge operations. a) Temperature variation at battery surface (solid) and centre (dashed). b) Temperature distribution across battery thickness direction at the end of discharge operation (2.8V).
Figure 3.11: Comparison of the reduced model (RM) and PDE model in terms of $C_{ss}$, $\bar{C}_s$, $C_e$, and $\eta$ (at current collector) during 0.5C and 10C charge operations. For 0.5C, $n=N=1$; for 10C, $n=3$ and $N=10$.

Table 3.2: Appropriate polynomial profile and grid resolution for different charge rates.

<table>
<thead>
<tr>
<th>Rate</th>
<th>$n$</th>
<th>$N$</th>
<th>$N_T$</th>
<th>$N_\Phi$</th>
<th>$\beta$</th>
<th>$n$</th>
<th>$N$</th>
<th>$N_T$</th>
<th>$N_\Phi$</th>
<th>$\beta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5C</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>0.39</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>0.39</td>
</tr>
<tr>
<td>1C</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>0.87</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>0.87</td>
</tr>
<tr>
<td>2C</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1.97</td>
<td>1</td>
<td>3</td>
<td>1</td>
<td>1</td>
<td>0.73</td>
</tr>
<tr>
<td>3C</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>3.30</td>
<td>2</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>0.83</td>
</tr>
<tr>
<td>4C</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>4.79</td>
<td>2</td>
<td>5</td>
<td>1</td>
<td>1</td>
<td>0.94</td>
</tr>
<tr>
<td>5C</td>
<td>2</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>3.13</td>
<td>3</td>
<td>5</td>
<td>1</td>
<td>1</td>
<td>0.93</td>
</tr>
<tr>
<td>6C</td>
<td>3</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>1.56</td>
<td>3</td>
<td>6</td>
<td>1</td>
<td>1</td>
<td>0.99</td>
</tr>
<tr>
<td>7C</td>
<td>3</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>1.66</td>
<td>3</td>
<td>7</td>
<td>1</td>
<td>1</td>
<td>1.00</td>
</tr>
<tr>
<td>8C</td>
<td>3</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>1.80</td>
<td>3</td>
<td>8</td>
<td>1</td>
<td>1</td>
<td>0.96</td>
</tr>
<tr>
<td>9C</td>
<td>3</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>2.04</td>
<td>3</td>
<td>9</td>
<td>1</td>
<td>1</td>
<td>0.98</td>
</tr>
<tr>
<td>10C</td>
<td>3</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>2.38</td>
<td>3</td>
<td>10</td>
<td>1</td>
<td>1</td>
<td>0.99</td>
</tr>
</tbody>
</table>

Minimal Resolution of Remaining States, $N$

Having reduced the parameters $N_T = N_\Phi = 1$, the modelling complexity is fully stated in terms of $N$ and $n$. These parameters determine the ability of the reduced order model
to reproduce the full PDE model’s outputs to a given accuracy level. Specifying the accuracy levels of interest to be $\beta=5\%$ (for closed loop control) and $\beta=1\%$ (for open loop optimisation), the minimal $N$ and $n$ are ascertained over a range of charge rates and given in Table 3.2.

The accuracy of the single particle model represented by $N=1$ is limited to low charge rates as previously observed in the literature, e.g. [42]. For charge rates $>1C$, to maintain a model accuracy of $1\%$, the increasing of $N$ and $n$ is required. Reducing the accuracy requirement to $5\%$ means the single particle model maintains its ability to $4C$ rates. For high charge/discharge rates, the complexity requirements increase monotonically with the applied current. For fast charge optimisation problems, this potentially means models with up to 10 spatial states and 6th order polynomial approximation must be used.

The significance of this increased modelling request can be expressed in terms of the computational load differences. The setup with $N = 10 \ n = 3$ is 10 times slower than the one with $N = 1$ and $n = 1$, but still 500 times faster as compared to the PDE model.

So far, the output voltage and temperature of the PDE model is well approximated by a library of models consisting of $5N+1$ ODEs and $10N+1$ AEs. As stated earlier, based on the prevalent literature, SOC and SOH are functions of the internal states, $C_{ss}$, $C_{s}$, $C_{e}$, and $\eta$. The capability of the reduced models presented in Table 3.2 to capture these states at different charge rates is examined and shown in Fig. 3.11. It can be seen that close agreement is achieved for the upper and lower bounds of the considered current range. If SOC and SOH are well-captured by these states, then the proposed models are therefore capable of dealing with the constraints on electrochemical model-based fast charging control.

### 3.6 Further Model Simplifications

There exist further possibilities for battery model simplifications based on elimination of some states due to their insensitivity on the model output. Similar to the polynomial approximation in solid-phase concentrations, the initial spatial dependent electrolyte states
3.6 Further Model Simplifications

in (3.6) may also be approximated before the use of spatial discretisation. To realise this, the following assumption is applied:

Assumption 3.9. The spatial and temporal trajectory of electrolyte concentrations, $C_{e}^{\pm,sep}(x,t)$, can be approximated by

$$\dot{C}_e(x,t) = \sum_{i=1}^{n} \theta_i \cdot \phi(t) \cdot \alpha_i(x),$$  \hspace{1cm} (3.56)

such that there exists:

$$|C_e(x,t) - \dot{C}_e(x,t)| < \epsilon_e(n), \forall t \geq 0,$$

where $\alpha_i(\cdot)$ is a basis function, and $\epsilon_e(\cdot)$ is an $L$-function.

Polynomial approximations can be used in (3.56) according to [63]. Specifically, the electrolyte concentrations can be well captured by second-order polynomials at a constant applied current [63]

$$\dot{C}_e(x,t) = (p_1x^2/2 + p_2) \cdot \phi(t) \cdot I(t) + C_{e0},$$  \hspace{1cm} (3.57)

where, $\phi(t)$ can be obtained by substituting (3.57) into (3.6). To capture electrolyte characteristics under time-varying currents as well, this approximation may be extended using

$$\dot{C}_e(x,t) = (p_1x^2/2 + p_2) \cdot \phi(t, I(t)) + C_{e0}.$$  \hspace{1cm} (3.58)

This approach reduces the PDEs, (3.6) to ODEs, whilst remaining the spatial dependency in the electrolyte states. To derive the terminal voltage or the overpotential, the electrolyte concentrations at the boundaries are required. Namely, in this case, further simplification can be conducted by setting $x = 0^\pm$ in (3.58).

If the interested battery operations are at low currents, based on simulation results in Table 3.2, the model $\Sigma_m^5$ with the discretisation element for each electrode $N = 1$ can be chosen as a starting point for further reductions. This model can be regarded as the well-known single particle model (SPM) with both the temperature and electrolyte states.
Assumption 3.10. The temperature at the surface of the battery cell is measurable.

As only a single temperature state is required even at high input currents, measurement of the temperature anywhere on the cell is likely to lead to a reasonable approximation of the internal lumped state. The use of this assumption and the subsequent removal of the temperature states lead to a single particle model with electrolyte diffusion.

Assumption 3.11. The net flow of lithium-ions between the electrolyte and the solid particles over all the domains of a battery cell is negligibly small.

Remark 3.10. This can be justified by comparing the magnitudes of the total ions in the electrolyte, \( n_{Li}^e \), and the total solid-phase ions, \( n_{Li}^s \). Simulations using typical battery parameters listed in Table 3.3.5 show that \( n_{Li}^e < 4\% \cdot n_{Li}^s \).

This allows the use of two conservation equations to solve the electrolyte concentrations and the solid-phase concentrations, separately. As a result, the states in one of the electrodes can be removed from the model.

Based on Assumptions 3.9-3.11, a further reduced battery model, denoted by \( \Sigma_{m}^6 \), can be obtained with its governing equations explicitly given as below

\[
\frac{dC_s(t)}{dt} = -\frac{3}{FL - a - R_p} I(t), \quad (3.59a)
\]
\[
\frac{dq_s(t)}{dt} = -\frac{30D_s}{(R_p)^2} q_s(t) - \frac{22.5}{Fa - L - (R_p)^2} I(t), \quad (3.59b)
\]
\[
\frac{dC_e(t)}{dt} = \frac{D_e}{\mu_e} p_1 \left( C_e(t) - C_{e0}\right) + \frac{\gamma^-}{FL - \mu_e} I(t), \quad (3.59c)
\]

The outputs of interest for the fast dynamic model include the terminal voltage and the SOC and are re-written as

\[
V(t) = \Phi^+(t) - \Phi^-(t) + U^+(C_{s0}(t))
\]
\[-U^-(C_{s0}(t)) + \eta^+(t) - \eta^-(t) - \frac{R_f I(t)}{a - L^-}, \quad (3.60a)
\]
3.6 Further Model Simplifications

\[ \text{SOC}(t) = \frac{C^-_s(t)}{C^-_{\text{smax}}}. \] (3.60b)

To calculate the terminal voltage and SOC, the following algebraic equations are required

\[
\eta^\pm(t) = \pm \frac{2RT}{F} \ln \left( \frac{-I(t) / (2a^\pm K^\pm L^\pm)}{\sqrt{\text{C}^\pm_\text{s}(t)\text{C}^\pm_\text{smax} - \text{C}^\pm_{\text{ss}}(t)^2}} \right),
\] (3.61a)

\[
\Phi^+ (t) - \Phi^- (t) = \frac{2RT}{F} \left( \gamma^+ \ln \text{C}^+_e(t) - \gamma^- \ln \text{C}^-_e(t) \right)
- \left( \frac{L^-}{2K^-} + \frac{L^\text{sep}^-}{K^\text{sep}^-} + \frac{L^+}{2K^+} \right) I(t),
\] (3.61b)

\[
\text{C}^+_s(t) = \frac{L^+ \mu^+_s \text{C}^+_s(t_0) + L^- \mu^-_s \text{C}^-_s(t_0) - L^- \mu^-_s \text{C}^-_s(t)}{L^+ \mu^+_s},
\] (3.61c)

\[
\text{C}^-_s(t_f) = \text{C}^-_s(t_f) - \frac{Q_{\text{sr}}(t_f) - Q_{\text{sr}}(t_0)}{Q_{\text{max}}},
\] (3.61d)

\[
\text{C}^+_e(t) = -\frac{L^- \mu^-_e}{L^+ \mu^+_e} \left( \text{C}^-_e(t) - C_{e0} \right) + C_{e0},
\] (3.61e)

\[
q^+_s(t) = -\frac{(R_p^+)^3 L^- \mu^-_s}{(R_p^-)^3 L^+ \mu^+_s} q^-_s(t),
\] (3.61f)

\[
\text{C}^+_s(t) = \text{C}^+_s(t) + \frac{8R_p^+}{35} q^+_s(t) + \frac{R_p^+}{35F_a^+L^+D^+_s} I(t).
\] (3.61g)

In (3.61), the slow states \(R_f\) and \(Q_{sr}\) appear but are both regarded as constant. \(t_0\) and \(t_f\) are the initial and end time of an operating cycle, respectively.

The reduced ODE model, (3.59)-(3.61), is evaluated against the PDE model \(\Sigma^2_s\). The input current is taken from an urban dynamometer driving schedule (UDDS) driving test. The parameters to be used in simulations are again taken from Table 3.3.5. The modelling error in state or output is defined as

\[ \beta_w(t) = \left| w_r(t) - w(t) \right| / w_{\text{max}}, \]

where \(w_r(t)\) and \(w(t)\) are solutions of the reduced model and the high-fidelity model, respectively, and \(w_{\text{max}}\) is the maximum value of \(w(t)\). The comparison results are illustrated in Fig. 3.12.

The profile of solid-phase Li-ion concentration derived from the reduced model closely matches solution of the initial model. Similarly, a good agreement is achieved in the electrolyte concentration. Accurate computations imply that the SOC will also be modelled accurately. Although the error in the voltage prediction is seen to become larger at higher
charging rates or step changes of the input, the reduced model is a good representation of the initial model with the error in the entire operating cycle bounded by 2%. This level of model mismatch is within an acceptable range for typical observer designs.
3.7 Conclusion

In this chapter, a physics-based model for Li-ion battery that accurately captures the electrochemical, thermal, electrical, and ageing dynamics has been proposed. Starting from this initial system formulation, a novel framework for PDE battery model simplification was developed by judicious use of a singular perturbation approach and the averaging theory. The novelty arises from the systematic simplification procedure through which the underlying assumptions imposed on simplified models can be explicitly stated. The developed model simplification framework including families of battery models is sufficiently general covering different battery types and a range of applications.

Based on a PDE model simplified from the proposed modelling framework, further model reductions were conducted with the aid of spatial dimension reduction, spatial discretisation and state elimination techniques. For a range of operating conditions, the minimum modelling requirements for the proposed models to achieve a given accuracy have been found. These obtained models exhibit significantly reduced computational times relative to the PDE model. Their applications for estimator and controller design will be investigated in the following chapters. The overall model simplification process is summarised in Fig. 3.13.
Figure 3.13: Summary of Li-ion battery model simplification.
Chapter 4

Charging Strategy Evaluation

A safe and fast charging strategy is desired in the utilisation of rechargeable Li-ion batteries. Traditionally, experimental methods are used in exploring and evaluating new strategies, but these require extensive time and cost. This chapter establishes a model-based system for quick and accurate evaluation of charging strategies. Based on the models obtained in the previous chapter, an evaluation procedure synthesised from the averaging theory is proposed. This procedure is implemented on several typical battery charging strategies. The benefits relative to simulations on higher order models are assessed. It has been demonstrated that significant computational savings are possible with the proposed approach.

4.1 Introduction

To avoid high cost in maintenance or replacement, batteries have to be operated in an appropriate manner that does not rapidly degrade their SOH. The SOH degradation is highly dependent on the charging strategies used [142]. Aside from battery’s lifetime, its ability to supply requested power demand and absorb currents requested for fast charging is also very important. Developing an optimal charging strategy to address these concerns is thus necessary.

Research on battery charging strategies has experienced considerable growth with a mag-
Charging Strategy Evaluation

In the literature, e.g., [19, 23, 142], trickle charging, whereby a small constant current is chosen during the entire process, was used to restore battery charge without unduly impacting SOH. To achieve accelerated charging rates, the constant-current constant-voltage (CCCV) protocol [18] is commonly used in current battery management systems. However, this method is recognised to be suboptimal for both charging efficiency and battery health [3, 22]. A boost charging scheme is suggested by [19] and demonstrates that very high current rates can be used for batteries at low state-of-charge levels for a short time. Motivated by this approach, multi-stage constant current algorithms with piece-wise decreased charging rates are proposed in [21]. Through intensive experiments, these algorithms have been shown to offer advantages relative to the CCCV method. Other improved charging strategies, such as modified CCCV, combinations of several CC and CV, and optimal control-oriented methods, are presented in [23, 135, 142].

To demonstrate the advantages of new methods, experiments are typically designed and conducted. These require a battery screening test, preprocessing, a statistically significant set of charge and discharge operations, and state characterisation. These methods, however, are costly and time-consuming. Moreover, charging performance relates to various and coupled in-situ dynamics of the battery internal states like Li-ion concentration and local electrolyte current [18], and is difficult to accurately analyse in an experimental manner with the available input-output data. In this regard, a model-based simulation procedure for quickly and accurately evaluating a given charging strategy is proposed in this chapter.

4.2 Charging Strategy Evaluation Procedure

The battery models to be used for charging strategy evaluation is taken from Chapter 3. Specifically, a medium time-scale model, $\Sigma_{m}$, and a slow time-scale model, $\Sigma_{s}$, that capture the electrochemical-thermal dynamics and the degradation phenomenon, respec-
4.2 Charging Strategy Evaluation Procedure

...are considered. The dynamic equations of these two models are recapped here:

\[
\begin{align*}
\Sigma_m^3: & \quad \dot{x}_m = F_m x_m + H_m(x_s^0, x_m, h(x_s^0, x_m, u), x_m(0) = x_m(0), \quad (4.1) \\
\Sigma_s^3: & \quad \dot{x}_s = \epsilon_2 H_{av}(x_s^0), \quad x_s(0) = x_s(0), \quad (4.2)
\end{align*}
\]

where \( H_{av}(x_s) \) has the definition of

\[
H_{av}(x_s) := \frac{1}{T_s} \int_0^{T_s} F_s x_s + H_s(x_s^0, x_s^0, h(x_s^0, x_s^0, u^*), u^*) dt. \quad (4.3)
\]

Basically, these models in (4.1)-(4.3) have various applications including: assessment on the effects of different charging strategies by means of simulation instead of the original model or experiments; model-based estimation, control, and optimisation for battery dynamic performance and lifetime. In this chapter, we aim to evaluate different charging strategies. For any given charging strategy, we can sequentially run (4.1) and (4.2) to derive the state, \( x_s \), as well as the SOH.

**Assumption 4.1.** There exists a constant \( u_{av} \) such that the input current satisfies:

\[
\frac{1}{T_c} \int_{t+(k+1)T_c}^{t+(k+1)T_c} u(\tau) d\tau = u_{av}, \forall t \in [0, T_c], \forall k = 0, 1 \ldots N,
\]

where \( T_c \) is the time period for a complete cycle composed of charging, discharging, and relax modes, and \( N \) is the cycle number representing battery’s lifetime.

**Remark 4.1.** The above assumption implies the periodic input currents are used for battery successive operating cycles. Its validity is tightly related to the practical utilisation of a Li-ion battery. The charging process at each operating cycle can be controlled with its strategies predetermined in battery chargers. This makes it plausible to assume the system inputs for battery charging at different cycles are identical. For discharging cases, typical battery-powered devices are generally repeatedly used for similar tasks like UDDS for electric vehicles. Therefore, it is a reasonable assumption simplifying the simulation process.

Charging strategies are defined in terms of current, \( u(t) \), and terminal voltage, \( V(t) \). For
example, the CCCV strategy specifies a current level for a period until $V(t)=V_{\text{max}}$, and remains the maximum voltage until $u(t)=I_{\text{min}}$. The combination of current and voltage levels and their thresholds defines the charging strategy, $u$. To analyse and compare various charging strategies, the following evaluation procedure using the averaged system $\Sigma_3^s$ and the simplified model $\Sigma_3^m$ from the established framework is proposed. Here, degradation to 80% of the initial SOH is used to quantify battery’s lifetime.

Procedure 4.1.

1. Specify a battery charging strategy, $u$, initial conditions of the states, $[x_{s0}, x_{m0}]$, and the cycle number $k=1$.
2. Run the battery model (4.1) under the specified charging strategy.
3. Calculate the static mapping $x_s \rightarrow H_{av}(x_s)$ using (4.3). For arbitrary initial conditions, a sufficiently large value of $T_s$ can be chosen to approach the infinite time interval. If the initial values $x_{s0}$ and $x_{m0}$ are within the limit cycles of the steady state of the $x_{m}$, then in practice, $T_s$ can be set to $T_c$.
4. Run the averaged battery system (4.2) and record $x_s$ at the time $t = kT_c$.
5. Calculate SOH using (3.20c).
6. If SOH > 80%, then increment $k$, update $x_{s0}$ and go to Step 2, else go to Step 7.
7. Output the battery lifetime, $k$.

Through the proposed procedure, the SOH evolution of a battery under a given charging strategy can be derived. The performance of different charging strategies can be evaluated based on the life cycles they can support.

### 4.3 Simulation Results

In this section, simulation results are presented to demonstrate the efficacy of the proposed evaluation procedure for different charging strategies. The simulation environment and parameters are the same as the previous chapter.

In addition to a CCCV charging and a multistage constant-current (MCC) charging pro-
Table 4.1: Comparison of computational time for 400 cycles associated with different battery models.

<table>
<thead>
<tr>
<th>Models</th>
<th>Sampling time (s)</th>
<th>Simulation time (mins)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Sigma_c^2$</td>
<td>1</td>
<td>171</td>
</tr>
<tr>
<td>$\Sigma_c^3 + \Sigma_{inf}$</td>
<td>1</td>
<td>120</td>
</tr>
<tr>
<td>$\Sigma_c^4$</td>
<td>1000</td>
<td>$1.8 \times 10^{-4}$</td>
</tr>
</tbody>
</table>

posed by [21, 22], an alternative MCC charging strategy is proposed for the purpose of comparison. Relative to its counterparts, the proposed strategy has higher current rate and voltage limit in the initial stage. Specifically, the voltage is allowed to slightly exceed 4.2V which is often considered as the voltage safety level in model-free or ECM-based strategies. This is enabled by two facts observed from previous work. First, according to [19], the effect on battery’s health is inappreciable when the voltage is over 4.2V at low SOC levels. Second, with battery internal information and physical constraints, the voltage limit may be relaxed as per [142]. While the proposed strategy is ad hoc in terms of the determination of C-rate, voltage limit, and duration during each stage, it is useful to demonstrate the potential differences in battery states that may be observed from the simplified models.

The integrator DASSL with flexible integration stepsize is chosen for these simulations. The integration tolerance is setup as 0.0001. The spatial discretisation method presented in Section 3.5 is utilised in solving the PDE system with each electrode and separator discretised into 15 elements. To ensure the maximum error in state prediction introduced by temporal discretisation within 0.01%, the sampling time is chosen as 1 second for the models that contain the medium states while as 1000 seconds for the averaged battery system. In the simulation of $\Sigma_c^3$, the mapping $x_s \rightarrow H_{av}(x_s)$ is assumed to be already available. The CPU-time for integration is recorded for the assessment of computational efficiency.

The estimate error in SOH prediction is defined as $\beta := |x_r - x| / x \times 100\%$, where $x_r$ and $x$ are respectively the solutions of the tested model and the benchmark.

The CCCV, multistage CC, and the proposed charging method are simulated and comprehensively compared with respect to their required charging times and terminal effects.
Figure 4.1: Comparisons of the CCCV, multistage constant current charging, and proposed charging method in terms of the input current, the terminal voltage, and their terminal effect on battery SOH.
on battery SOH, as provided in Fig. 4.1. The accuracy of the simplified model from the established framework is first examined. Given the fast electrical dynamics exhibit in microseconds, the full model that includes all the dynamics is impractical to simulate. The model $\Sigma^2_s$ that well approximates the original model in the slow and medium time scales is used as a test bed. From Fig. 4.1(e), it can be seen the percentage deviations in SOH prediction by using the proposed procedure are less than 5% for all the considered charging strategies. These results illustrate the obtained simplified models are capable of accurately capturing system dynamics, and the Procedure 1 is reliable in evaluating charging strategies.

Table 4.1 compares the computational times for simulating 400 cycles associated with different battery models, where all the simulations are conducted on a desktop computer with 3.4GHz processor and 8GB RAM. It is found the time of 29.8% can be saved by using $\Sigma^3_m$ and $\Sigma^3_s$ as proposed in Procedure 1 rather than the high order model $\Sigma^2_s$.

For SOH prediction, if the static mapping as shown in Fig. 4.1(c) is available a priori, simulations using $\Sigma^3_s$ become extremely fast. Thus, this proposed evaluation system based on simplified models is able to significantly improve the computational efficiency.

In addition, simulation results of Fig. 4.1 show the multistage CC can support 60% more cycles and save 17.6% charging time relative to the CCCV. This is consistent with the experimental outputs observed from [21]. Meanwhile, the proposed method can transfer similar capacity to the battery but achieves 13% more lifetime and 11.6% less charging time compared to the multistage CC. This may be useful information for developing optimal charging strategies.

## 4.4 Conclusion

In this chapter, by using low order models simplified from the initial 17 nonlinear coupled PDE system, a model-based evaluation procedure for battery charging strategies is proposed. This procedure was found to offer a substantial advantage in terms of computational efficiency with a reasonable loss in the accuracy of model predictions. Further-
more, this proposed evaluation procedure is sufficiently general such that other accurate battery ageing models developed in the future can be potentially incorporated to achieve a higher accuracy for SOH prediction.
Chapter 5
Battery State Estimation

ACCURATE knowledge of state-of-charge (SOC) and state-of-health (SOH) is critical for safe, reliable and efficient utilisation of lithium-ion batteries. Many state estimators found in the literature resort to equivalent circuit models. However, these models are often not accurate during an extended range of operations and thus can be problematic for SOC monitoring. Furthermore, they do not explicitly address SOH dynamics. To avoid these issues, two approaches based on physics-based models are proposed in this chapter for estimation of the SOC and SOH. First, based on an ODE model developed before, a nonlinear estimator is proposed to estimate the SOC and SOH simultaneously. Second, a multi-time-scale estimation algorithm is proposed for a class of nonlinear singularly perturbed systems. After justification of the underlying assumptions, this algorithm is applied to the battery system for SOC estimation in the fast time-scale and SOH estimation in the slow time-scale.

5.1 SOC and SOH Estimator: One Time-Scale

There is an ever-growing trend towards electrifying the powertrain in automotive industry to address the increasingly stringent standards on tailpipe emission and fuel economy. However, electric vehicles typically suffer from high relative costs, range anxiety and long charging time [5], which are all related to the battery system. Although Li-ion

\footnote{The first part of this chapter has already been presented and published at the 54th IEEE Conference on Decision and Control [153], and the second part has been published as an article in the Journal of Power Sources [159].}
batteries have been recognised as a suitable cell chemistry technology for vehicle applications, the properties such as energy/power density, ability to sustain fast “refuelling”, longevity, and safety are still clearly inferior to their counterpart, the internal combustion engines [6]. Accordingly, for a great adoption of electric vehicles, it is important to conduct advanced battery management.

The state information of SOC and SOH is crucial for battery management. As known from the definitions in (3.20) that SOC is a function of solid-phase ion concentrations, and SOH is a function of capacity fade. However, the ion concentrations and capacity fade inside a battery cell are difficult to measure during battery operations. This motivates the design of estimators for battery internal states, enabling real-time monitoring and control of the SOC and SOH.

In this section, based on a reduced physics-based battery model, an extended Kalman filter (EKF) is proposed for simultaneous SOC and SOH estimation. The estimator’s performance including accuracy, convergence speed and robustness is examined via simulations.

5.1.1 Battery Model for State Estimation

The battery model used here is adopted from the model simplification framework proposed in Chapter 3. Starting from the model $\Sigma^2_s$ that captures the electrochemical and ageing dynamics, reductions can be systematically conducted using singular perturbations, spatial dimension reduction and spatial discretisation as per Sections 3.2-3.5. As a consequence, a reduced order model is derived with 9 states in the state vector $x$

$$x := [C_s^-, C_s^+, C_e^-, C_e^+, q_s^-, q_e^+, Q_{sr}, R_f]^T,$$  \hspace{1cm} (5.1)

where the notation of $x$ is abused and different to the one in previous chapters. Assumption 3.10 regarding measurable temperature state has been applied on the battery model, leading to less states in (5.1).

The obtained ODE battery model consists of $9N$ ODEs, and can be further written in a
5.1 SOC and SOH Estimator: One Time-Scale

compact form

\[
\dot{x} = f(x, u), \quad (5.2a)
\]
\[
[y; z] = h(x, u), \quad (5.2b)
\]

where the system input \( u = I \), the measured output \( y := V \), and the unmeasured output vector \( z := [SOC^-, SOC^+, SOH]^T \). The problem here is to estimate \( z \) based on the model (5.2a)-(5.2b), the input current and measurement of the terminal voltage.

To further simplify the course of model-based estimator design, this model (5.2) is discretised on the temporal domain with Euler method leading to

\[
x_k = f_d(x_{k-1}, u_{k-1}), \quad (5.3a)
\]
\[
[y_k; z_k] = h_d(x_k, u_{k-1}), \quad (5.3b)
\]

where \( u(t) = u(k\Delta t) =: u_k, \forall t \in [k\Delta t, (k + 1)\Delta t], k \in \mathbb{N} \), and \( \Delta t \) is the sampling period.

5.1.2 Battery Estimator Design

The developed ODE model is used for estimation of both the SOC and SOH for a Li-ion battery. The challenges in front of the estimator design problem are mainly arisen from two aspects. The battery dynamic and output equations are highly nonlinear. In addition, the battery model is subject to a hybrid mathematical structure given it is different for charging and discharging operations. Particularly, in the initial model, the ageing dynamics are only included in the charging stage but not discharging process.

The Kalman filter is an optimal estimator for linear systems with noisy measurements under certain assumptions [110]. In order to address these difficulties associated with the battery model, an EKF is designed for online SOC and SOH estimation. In the process, for the calculation of Kalman gain and covariance, the battery model (5.3) is successively linearised around its reference trajectory to a linear time varying (LTV) system.

To describe the course of battery SOC and SOH estimator design, \( \hat{x} \) is the estimated state,
\( \hat{y} \) and \( \hat{z} \) consist of the estimated outputs, \( y \) is the measured terminal voltage, \( K \) is the Kalman gain, \( P \) is the estimate covariance, and \( R, Q \) are weighting matrices to be tuned for minimising the mean-square state estimate error.

Based on the developed battery model (5.3), the following EKF formulation is deployed:

\[
\begin{align*}
\dot{x}_{k|k} &= f^d(\dot{x}_{k-1|k-1}, u_{k-1}) + K_k(y_k - \hat{y}_k), \\
[\dot{y}_k; \hat{z}_k] &= h^d(\dot{x}_{k|k-1}, u_{k-1}),
\end{align*}
\]

where, the Kalman gain \( K_k \) is calculated from

\[
K_k = P_{k|k-1}H_k^T(H_kP_{k|k-1}H_k^T + R)^{-1},
\]

and the estimate covariance \( P \) is propagated and updated through (5.6) at each time step

\[
\begin{align*}
P_{k|k-1} &= F_kP_{k-1|k-1}F_k^T + Q, \quad P_{0|0} = P_0, \\
P_{k|k} &= (I - K_kH_k)P_{k|k-1}.
\end{align*}
\]

In (5.6a), \( F_{k-1} \) is the Jacobian matrix derived from the nonlinear function \( f^d(\cdot) \) based on the previously estimate state \( \dot{x}_{k-1|k-1} \), \( H_k \) in (5.6b) is the Jacobian matrix derived from the nonlinear function \( h^d(\cdot) \) based on the current propagated state \( \dot{x}_{k|k-1} \), and \( I \) is the identity matrix.

This proposed algorithm is then implemented for simultaneous SOC and SOH estimation. By using the input signal \( u_{k-1} \) and the previously estimated state \( \dot{x}_{k-1|k-1} \), the state \( \dot{x}_{k|k-1} \) can be predicted based on the nominal battery model. This estimated state is then updated by the voltage measurement \( y_k \) and the calculated Kalman gain \( K_k \). In the case of simulation investigation, the voltage measurement \( y_k \) is obtained from the “measured” plant, the high-fidelity battery model. Finally, according to (3.20b) and (3.20c), \( \hat{z}_{SOC^-}, \hat{z}_{SOC^+}, \hat{z}_{SOH} \) can be calculated using \( \hat{x}_1, \hat{x}_2, \) and \( \hat{x}_8 \), respectively.

### 5.2 Estimation Result I

Simulations are conducted in this section to examine the designed estimator’s capability for SOC and SOH estimation as compared to the true results derived from the high-
5.2 Estimation Result I

The high-fidelity model with noisy input is solved in Modelica associated with Visual C++, providing measured values for the terminal voltage. The EKF based on the derived ODE model is implemented in Matlab.

The battery is charged and discharged at 1C constant current for successive operating cycles. Where, the terminal voltage thresholds for charge and discharge operations are set up as 4.15V and 2.9V, respectively. The time period of each operating cycle is defined as 6800s, consisting of charge, discharge, and relax modes to mimic the real world battery utilisation. Zero input current is used during the relax mode. To test the estimator’s capability of capturing the SOH dynamics which have been justified to behave in a slow time scale, a long operating time of 20 cycles is chosen. All the states are initialised with 10% relative error, and additional white Gaussian noise with a standard deviation of 80mA/m² is added to the system input. The estimate error for SOC/SOH is defined as $e_z(t) := |z_r(t) - z(t)|/z_0$, where $z_r, z$ are separately the solutions of the ODE model and

![Figure 5.1: Evolution of state estimation for the solid-phase Li-concentrations, $C_s^-, C_s^+$, and the capacity fade $Q_{sr}$ over 20 cycles.](image-url)
the PDE model, and the absolute error, $|z_r - z|$, is normalised on its corresponding initial value $z_0 = z(0)$.

The internal states in the battery system are reproduced by the proposed estimator, and estimation results for these critical states, $C_s^-, C_s^+, \text{ and } Q_{sr}$ are shown in Fig. 5.1. It can be seen that the employed EKF is able to quickly compensate the initial deviations added to the states. All these states closely follow its true value during both the charging and discharging operations.

Fig. 5.2 depicts the evolution of estimation errors for SOC/SOH. Although large initial errors are assumed to exist in the SOC (10%) and SOH (0.01%) estimation problem, the state estimator is capable of driving its outputs to converge to the vicinity of the true states. For SOCs in the negative and positive electrodes, their steady-state estimation errors are both less than 2%. At the same time, the SOH is also accurately estimated with the steady-state error bounded by 0.001%.

In addition, steady-state errors and periodic error sequences have been observed from
Fig. 5.2. Indeed, exact convergence to the origin for the estimation errors is not possible due to the persistent noises applied to the system input and measurements and model mismatches resulting from model order reductions and successive linearisation. Furthermore, the periodic nature of the plant input potentially leads to the periodic variation of error dynamics.

Based on the above analysis, the designed estimator for battery’s SOC and SOH estimation is accurate and robust in dealing with the model mismatches, initial errors, and noises.

5.3 SOC and SOH Estimator: Multi-Time-Scale

The SOC and SOH were estimated in the same time-scale in the previous section. However, it is unnecessary to estimate the SOH in the fast time-scale and sacrifice the computational efficiency. Furthermore, the estimator design for nonlinear singularly perturbed systems may cause ill-conditioned gains and convergence issues as per [131].

To address these problems, a multi-time-scale estimation algorithm for a class of nonlinear singularly perturbed systems is proposed in this section. Stability property of the estimation errors is analytically characterised by adopting a deterministic version of EKF. This proposed algorithm is applied to estimate the SOC and SOH for a Li-ion battery. Illustrative results shall be provided to show the effectiveness of the designed battery estimator.

5.3.1 Multi-Time-Scale Observer - Theory Development

This subsection describes the development of a multi-time-scale estimation algorithm. We consider a nonlinear singularly perturbed system with coupled fast and slow states, \( x_f \in X_f \subset \mathbb{R}^{n_f} \) and \( x_s \in X_s \subset \mathbb{R}^{n_s} \), where \( X_f \) and \( X_s \) are bounded sets. \( u \) is the system input and \( u \in U \). Specifically, in this system, while \( y_f \) and \( y_s \) represent the fast and slow measurable system outputs, \( z_f \) and \( z_s \) are the unmeasurable system outputs separately.
in the fast and slow time-scales and belong to the sets of $\mathbb{R}^m_f$ and $\mathbb{R}^m_s$. $e$ is a perturbation parameter and is small and positive. The system governing equations can be formulated as

\begin{align*}
\dot{x}_f &= F_f(x_f, x_s, u, e), \quad (5.7a) \\
\dot{x}_s &= eF_s(x_f, x_s, u, e), \quad (5.7b) \\
y_f &= H_f(x_f, x_s, u, e), \quad (5.7c) \\
z_f &= W_f(x_f, x_s, e), \quad (5.7d) \\
y_s &= H_s(x_f, x_s, e), \quad (5.7e) \\
z_s &= W_s(x_f, x_s, e). \quad (5.7f)
\end{align*}

**Assumption 5.1.** The perturbation parameter $e$ in (5.7b) is sufficiently small such that it satisfies $0 < e \ll 1$.

According to [98], if Assumption 5.1 holds, the slow state $x_s$ can be replaced by an equilibrium state $\bar{x}_s$ and then a boundary layer system can be obtained to approximate the fast dynamics, i.e.:

\begin{align*}
\dot{x}_f(t) &= F_f(x_f(t), \bar{x}_s, u(t), 0), \quad (5.8a) \\
y_f(t) &= H_f(x_f(t), \bar{x}_s, u(t), 0), \quad (5.8b) \\
z_f(t) &= W_f(x_f(t), \bar{x}_s). \quad (5.8c)
\end{align*}

In the following, an assumption on the stability property of the system (5.8) is imposed. In the statement, a class-$K$ function means that a function, $\gamma(\cdot)$, from $\mathbb{R}_{\geq 0}$ to $\mathbb{R}_{\geq 0}$ is continuous, strictly increasing and $\gamma(0) = 0$. A function, $\beta(\cdot, \cdot)$, defined on $\mathbb{R}_{\geq 0} \times \mathbb{R}_{\geq 0}$ taking values in $\mathbb{R}_{\geq 0}$ is said to be a class-$KL$ function if it is continuous, class-$K$ function in its first argument, and decreasing to zero in its second argument.

**Assumption 5.2.** $x_f^e, x_f^r$ are the solutions of (5.7) and (5.8) for a given $u$, respectively. There exists an integral manifold $e_f := x_f^r - x_f^e$, a class-$K$ function $\gamma$, and a class-$KL$ function $\beta$ such
that, for all initial conditions in the domain $\mathcal{X}$, there exists

$$|e_f(t)| \leq \beta(e_f(0), t) + \gamma(e), \quad \forall t \geq 0.$$ 

Given $u$ in the system (5.8) is time-varying and oscillating, the effect of the fast dynamics on the slow states may be determined predominately by the average of the steady-state behaviour of the fast system (5.8). In particular, this applies when there exists a well-defined average according to the next definition that is a special case of Definition 5.1 from [152].

**Definition 5.1.** The continuous and bounded function $F_s(x_f, x_s, u, e)$ with its arguments $x_f, x_s$ in bounded sets $\mathcal{X}_f, \mathcal{X}_s$, and piecewise differentiable input $u(t) \in \mathbb{U}$, is said to all have a well-defined average, $F_s$, if $\forall \rho > 0$, $\exists T^* > 0$ and $\exists e^* > 0$ such that $\forall T \geq T^*, \forall e \in (0, e^*)$ there exists a measurable function $e : \mathbb{R}_{\geq 0} \to \mathbb{R}^m$ satisfying

$$\|e(t)\|_{\infty} \leq \|u(t)\|_{\infty},$$

such that the following inequality holds

$$\left| \frac{1}{T} \int_0^T [F_s(x_f(t), x_s, u(t), e) - F_s(x_s, e(t))] dt \right| \leq \rho e.$$  

(5.10)

The role of $e(t)$ is in *Definition 5.1* allows an ensemble of solutions for the slow dynamics, if there exist, corresponding to multiple steady-state solutions of the boundary layer system [152].

**Assumption 5.3.** For each $(u, x_{f0}, x_{s0}) \in \mathbb{U} \times \mathcal{X}_f \times \mathcal{X}_s$, the functions, $F_s(x_f, x_s, u, e)$, $W_s(x_f, x_s, e)$, and $W_s(x_f, x_s, e)$ from (5.7) have a well-defined average.

When *Assumptions 5.1* and 5.3 are valid, the general average of $F_s(x_f, x_s, u, e)$ is formulated in the following

$$F_s(x_s, e) := \frac{1}{T} \int_0^T F_s(x_f^*(t), x_s, u(t), 0) dt.$$  

(5.11)
where $e(t)$ is some measure of $u(t)$ satisfying (5.9), and $x_f^* = x_f^*(x_{f0}, u(t))$ is the solution of (5.8) by a specified input $u(t)$ defined over the interval $[0, T]$. Similarly, $W_s(\cdot)$, $H_s(\cdot)$ in the output equations can be averaged by setting $e$ in (5.10) to be zero. For a given $u(t)$, the obtained average, $W_s(\cdot), H_s(\cdot)$, are formulated by

$$W_s(x_s) := \frac{1}{T} \int_0^T W_s(x_f(t), x_s, 0) dt,$$

$$H_s(x_s) := \frac{1}{T} \int_0^T H_s(x_f(t), x_s, 0) dt. \tag{5.13}$$

**Remark 5.1.** The accuracy of approximation in (5.11) and (5.12) follow directly from Taylor series expansion of $F_s(x_s, e)$ and $W_s(x_f, x_s)$.

Based on the average defined above, the reduced (average) system can be obtained, approximating the slow dynamics of the initial system (5.7b) in the time scale $\tau = et$ as

$$\frac{dx_s}{d\tau} = F_s(x_s, e), \tag{5.14a}$$

$$z_s = W_s(x_s). \tag{5.14b}$$

**Assumption 5.4.** The system (5.14) has a unique solution $x_s^* (\tau) \in S$, for $\tau \in [\tau_0, \tau_1]$, where $S$ is a compact subset of $X_s$.

**Remark 5.2.** Assumptions 5.2 and 5.4 are standard in the singular perturbation literature as the model simplification due to time-scale separation can only be justified if the boundary layer dynamics are uniformly asymptotically stable and the solution of the reduced system uniquely exists. These two assumptions hold in many practical systems, including ODE representations of lithium-ion batteries as will be discussed in Section 5.3.2.

To estimate $z_f(t)$ and $z_s(\tau)$, the multi-time-scale estimator structure is proposed in the following. To facilitate the statement, $\hat{x}_f(t)$ and $\hat{x}_s(\tau)$ denote the estimates of $x_f(t)$ and $x_s(\tau)$, and $K_f(t), K_s(\tau)$ are the estimator gains. By using the nominal battery models obtained in (5.8) and (5.14), the dynamic equations for the proposed estimation algorithm
are provided

\[
\frac{d\hat{x}_f}{dt} = F_f(\hat{x}_f, \bar{x}_s, u) + K_f(y_f - \hat{y}_f), \tag{5.15a}
\]
\[
\hat{y}_f = H_f(\hat{x}_f, \bar{x}_s, u), \tag{5.15b}
\]
\[
\hat{z}_f = W_f(\hat{x}_f, \bar{x}_s), \tag{5.15c}
\]
\[
\frac{d\hat{x}_s}{d\tau} = F_s(\hat{x}_s, \bar{x}_s, u) + K_s(y_s - \hat{y}_s), \tag{5.15d}
\]
\[
\hat{y}_s = H_s(\hat{x}_s), \tag{5.15e}
\]
\[
\hat{z}_s = W_s(\hat{x}_s). \tag{5.15f}
\]

Note that the multi-time-scale nature of the problem is maintained through \(\bar{x}_s\) in (5.15a)-(5.15c) requiring \(\hat{x}_s\) in (5.15d)-(5.15f).

The estimation errors are defined as

\[
\tilde{x}_f(t) := x_f(t) - \hat{x}_f(t), \tag{5.16a}
\]
\[
\tilde{x}_s(\tau) := x_s(\tau) - \hat{x}_s(\tau). \tag{5.16b}
\]

The estimation error dynamics for the fast and slow state variables can be formulated based on (5.7)-(5.16)

\[
\frac{d\tilde{x}_f}{dt} = F_f(x_f, x_s, u, e) - F_f(\hat{x}_f, \hat{x}_s, u_f) - K_f(y_f - \hat{y}_f) = F_f(x_f, x_s, u) - F_f(\hat{x}_f, \hat{x}_s, u_f) + \zeta_f - K_f(y_f - \hat{y}_f) = [A_f - K_fC_f]\tilde{x}_f + \zeta_f + \Delta_f(\hat{x}_f, x_f, u), \tag{5.17a}
\]
\[
\frac{d\tilde{x}_s}{d\tau} = F_s(x_f, x_s, u, e) - F_s(\hat{x}_s) - K_s(y_s - \hat{y}_s) = F_s(x_s, e) - F_s(\hat{x}_s) + \zeta_s - K_s(y_s - \hat{y}_s) = [A_s - K_sC_s]\tilde{x}_s + \zeta_s + \Delta_s(\hat{x}_s, x_s, u), \tag{5.17b}
\]

where \(\zeta_f, \zeta_s\) represent the model mismatch between the initial model (5.7) and its sim-
plified counterparts (5.8) and (5.14). Additionally,

\[
A_f(t) \triangleq \frac{\partial F_f}{\partial x_f}(\hat{x}_f, \bar{x}_s, u_f), \quad (5.18a)
\]

\[
C_f(t) \triangleq \frac{\partial H_f}{\partial x_f}(\hat{x}_f, \bar{x}_s, u_f), \quad (5.18b)
\]

\[
A_s(\tau) \triangleq \frac{\partial F_s}{\partial x_s}(\hat{x}_s, e), \quad C_s(\tau) \triangleq \frac{\partial H_s}{\partial x_s}(\hat{x}_s, e), \quad (5.18c)
\]

\[
\Delta_f := F_f(x_f, \bar{x}_s, u) - F_f(\hat{x}_f, \bar{x}_s, u_f) - A_f\hat{x}_f
\]

\[
- K_f[H_f(x_f, u_f) - H_f(\hat{x}_f, u_f) - C_f\hat{x}_f], \quad (5.18d)
\]

\[
\Delta_s := F_s(x_s, e) - F_s(\hat{x}_s, e) - A_s\hat{x}_s
\]

\[
- K_s[H_s(x_s) - H_s(\hat{x}_s) - C_s\hat{x}_s]. \quad (5.18e)
\]

**Assumption 5.5.** The functions, \( F_f, F_s, H_f, H_s, W_f, W_s \), from (5.8)-(5.14) are Lipschitz continuous in their arguments. Furthermore, \( x_f, x_s \) and \( u \) is bounded.

**Assumption 5.6.** For the Jacobian matrices, \( A_f, A_s, C_f, C_s \), from (5.18a)-(5.18c), the pairs \( (A_f, C_f) \) and \( (A_s, C_s) \) are both uniformly observable.

**Assumption 5.7.** There exists an EKF gain trajectory, \( K(t) \), so that in the system (5.17), \( \hat{x}_f \) and \( \bar{x}_s \) are semi-globally practically stable. Namely, there exist positive constants \( c_f, c_s, k_f, k_s, \lambda_f, \lambda_s \) and functions \( \gamma_f(\cdot), \gamma_s(\cdot) \in \mathcal{K} \) such that \( \forall t > t_0 > 0 \)

\[
\|\hat{x}_f(0)\| \leq c_f, \quad \|\bar{x}_s(0)\| \leq c_s
\]

\[
\Rightarrow \|\hat{x}_f(t)\| \leq k_f e^{-\lambda_f(t-t_0)} + \gamma_f(\|\hat{x}_f(t)\|_{\text{max}}),
\]

\[
\|\bar{x}_s(\tau)\| \leq k_s e^{-\lambda_s(\tau-t_0)} + \gamma_s(\|\bar{x}_s(\tau)\|_{\text{max}}).
\]

**Remark 5.3.** Assumption 5.7 may be potentially relaxed to a more general case which covers other nonlinear estimation algorithms in addition to EKF. In this case, the method of [132], where only the slow states of singularly perturbed systems are of interest and estimated, can be possibly extended for estimator design around the boundary-layer model and slow model separately for the slow and fast states.
In the following proposition, the main theoretical result of this paper is presented. The errors in estimation of $z_f$ and $z_s$ are defined as $\tilde{z}_f(t) := z_f(t) - \hat{z}_f(t)$, and $\tilde{z}_s(\tau) := z_s(\tau) - \hat{z}_s(\tau)$.

**Proposition 5.1.** Let Assumptions 5.1-5.7 all hold. Consider the observer (5.15) applied to estimate $z_f$ and $z_s$ in (5.7), if there exist positive constants $d_f, d_s, KL$ functions $b_f, b_s$, and $K$ functions $g_f, g_s$ such that for all $t \in [t_0, t_1], \tau \in [\tau_0, \tau_1], \|\tilde{z}_f(0)\| \leq d_f$ and $\|\tilde{z}_s(0)\| \leq d_s$, there exist

\[ |\tilde{z}_f(t)| < \beta (|z_{f0} - \hat{z}_{f0}|, t) + \gamma_f(e), \]  
(5.19a)

\[ |\tilde{z}_s(\tau)| < \beta (|z_{s0} - \hat{z}_{s0}|, \tau) + \gamma_s(e), \]  
(5.19b)

where $\beta$ is a class-$KL$ function, $\gamma_f > 0, \gamma_s > 0$ and are class-$K$ functions, $t \in [t_0, t_1]$ and $\tau \in [\tau_0, \tau_1]$.

**Sketch of proof.** Based on Assumption 5.5, the inequality relationship in (5.19a) can be re-written as

\[ |\tilde{z}_f(t)| = |W_f (x_f(t), x_s(t)) - W_f (\hat{x}_f(t), \hat{x}_s)| \leq L_1 |x_f(t) - \hat{x}_f(t)| + L_2 |x_s(t) - \hat{x}_s| \]
\[ < \beta (|z_{f0} - \hat{z}_{f0}|, t) + \gamma_f(e). \]  
(5.20)

Based on Assumptions 5.4-5.7, and following the procedure of Lemma 11.2 in [160], the $|x_f(t) - \hat{x}_f(t)|$ in (5.20) can be readily shown to be semi-globally practically stable and the final error ball is determined by $\|\tilde{z}_f\|_{\text{max}}$. In consideration of 5.1-5.2 and Tikhonov’s theorem [161], it can be obtained that $|x_s(t) - \hat{x}_s| = O(e), \forall t \in [t_b, t_1]$. Therefore, (5.19a) has been proved. The similar work can be conducted to prove (5.19b).

**Remark 5.4.** Proposition 5.3.1 implies the mismatch between the initial system (5.7) and the decoupled systems, (5.8) and (5.14), can be compensated by the designed estimator. That is, the errors in estimation of $z_f$ and $z_s$ converge in an asymptotic manner to some bounded sets, although the design process of the multi-time-scale observer is simplified relative to conventional estimation approaches. More importantly, the time scales of the convergence are different for the fast and slow error dynamics.
5.3.2 Multi-Time-Scale Observer - Application To Li-Ion Battery

As stated earlier, the system dynamics for a Li-ion battery include electrochemical, thermal, electrical, and ageing behaviours and are originally governed by a set of coupled nonlinear PDEs. Time-scale separation techniques may be applied to this battery model for system simplification. Given Assumptions 5.1-5.4 assumptions on the battery system have been justified in Chapter 3, a singular perturbation approach and the averaging theory can be systematically applied to the initial model. As a consequence, a fast model that captures electrochemical-thermal dynamics and a slow model governing ageing process can be derived. These two PDE-based models elaborated in Chapter 3 are used for further model reductions to estimate battery SOC and SOH.

Battery Models in Two Time-Scales. The generic multi-time-scale observer design algorithm in the previous section will now be formulated with battery models approximating the PDE models. The first step is to define the fast state vector as 
\[ x_f = \begin{bmatrix} C_s(t) \\ q_s(t) \\ C_e(t) \end{bmatrix} \]

The applied current density, \( I(t) \), is taken as the system input. It is known that these states are sufficient to capture the fast dynamics under Assumptions 3.7-3.11. Based on the reduced order model, (3.59), obtained in Section 3.6, the resulting fast battery dynamics representing \( F_f(\cdot) \) in (5.8) are explicitly given by

\[
F_f(x_f, u) = \begin{bmatrix}
\frac{3}{FL} - a R_p^{-1} I(t) \\
- \frac{30 D_e}{(R_p^2) q_s(t)} - \frac{22.5}{Fa - L - (R_p^2)} I(t) \\
\frac{D_e}{\mu_c} p_1 \left( C_e^{-1}(t) - C_{e0} \right) + \frac{\gamma}{FL - \mu_c} I(t)
\end{bmatrix}
\]  

(5.21)

The slow state vector is defined as \( x_s = [Q_{sr}(t), R_f(t)]^T \). These state dynamics, \( F_s(\cdot) \), presented in (5.7b) is derived from the model \( \Sigma_3^3 \) and is formulated as

\[
F_s(x_f, x_s, u, \varepsilon) = \begin{bmatrix}
a^{-1} A^{-1} i_0 \exp \left( \frac{-F \delta_{sr} \eta_{sr}(t)}{RT} \right) \\
\frac{M_j i_0}{F \rho_j^2 \sigma_f} \exp \left( \frac{-F \delta_{sr} \eta_{sr}(t)}{RT} \right)
\end{bmatrix}
\]  

(5.22)
The notation and explicit form of output equations are the same as in (3.60)-(3.61). To permit close-loop estimator design for the SOH during cycling operations, in this work it is considered to measure the resistance, \( R_f \), at the end of each operating cycle via electrochemical impedance spectroscopy (EIS). Details for the EIS technique can be referred to [162].

**Multi-Time-Scale Battery Observer.** The proposed multi-time-scale algorithm is now applied to estimate SOC and SOH of a Li-ion battery. The parameters for simulations are taken from [8, 149]. Based on the obtained models, which are denoted by \( \Sigma_f' \) and \( \Sigma_s' \), the observer with fast and slow dynamics is designed in this subsection. This algorithm for state estimation is illustrated in Fig. 5.6.

The system model (5.14) requires evaluation of the averaged function \( F_s(x_s, e) \), which implicitly assumes same charging strategy is used in each cycle. In this work, the averaged function \( F_s(x_s, e) \) is approximated using a two-step process.

The first step is to generate the mapping \( F_s(x_s, SOC, u) \), where \( u, SOC \) are specified at a series of constant values. The battery is charged to a fixed SOC value and then experiences a discharge operation to the initial SOC. This is followed by a relaxed period to recover the equilibrium state. This whole process is defined as a “cycle” with the period of \( t_f \). To enable the same capacity is delivered in each cycle, no any voltage constraint is enforced in the course of operations. When the system input is specified at a constant value \( u \) at the time interval \([0, t_f]\) and the slow states are fixed at their equilibrium state, i.e. constant values in this case, the fast battery model can be run to reproduce the solution of the fast states, \( x_f^*(t) \). The trajectory of the fast states can be then substituted into (5.11) to derive the function, \( F_s(\cdot) \), according to

\[
F_s(x_s, SOC', u') = \left[ \frac{a^- A^- i_0}{t_f} \int_0^{t_f} \exp\left( -\frac{F_s(x_s', SOC', u')}{RT} \right) dt \right] \frac{M_f i_0}{F \rho_f \sigma f t_f} \int_0^{t_f} \exp\left( -\frac{F_s(x_s', SOC', u')}{RT} \right) dt \right]
\]  

(5.23)
Figure 5.3: The map of battery degradation rates at different charging C-rates, capacity fade and SOC. a) the $Q_{sr}/Q_{\max}$ is fixed at 0; b) the SOC change in each cycle is fixed at 0.45–0.55. Unit of $\hat{F}_s$: 1/sec.

and the overpotential, $\eta_{sr}$, from (3.19) is re-formulated by setting $\epsilon = 0$ and $u(t) = u^i$ as

$$\eta_{sr}(t) = \Phi_{s}^{-}(t) - \Phi_{s}^{-}(t) - U_{sr} - \frac{R_f(t_0)u^i}{a^2 L}. \quad (5.24)$$

SOC$^i$ in the function $F_s(\cdot)$ can be understood in some sense as an average measure of the fast states under the current $u^i$. In other words, there exists a solution: $SOC = SOC(x_{f0}, \bar{x}_s, u, t)$. By sequentially performing this process with different $u^i$ over multiple cycles and multiple depths of charge, the map, $F_s(x_s, SOC, u)$, can be generated. Fig. 5.3(a)-(b) illustrates this 3D map by respectively fixing the SOC and $x_s$. It can be clearly seen that $F_s(\cdot)$ greatly increases at larger values of SOC or $u$. Whilst the impact of $x_s$ is negligibly small, indi-
cating that the degradation rate is (almost) independent of the battery’s SOH. It is worth noting that this map can be generated offline, and is expected to be used for online state estimation.

Step 2 in the approximation of $F_s(x_s, e)$ for general $u(t)$ and SOC($t$) is to use a piecewise constant current in (5.23) and the summation of $F_s(SOC^i, u^i)$ along the trajectory:

$$\hat{F}_s(x_s, e) := \frac{1}{N} \sum_{i=1}^{N} F_s(SOC^i, u^i) \cdot \Delta t$$

(5.25)

where $\Delta t$ is the sampling time, and $u^i$ is a constant value approximating $u(t)$ over the time period of $[i, i + 1] \Delta t$.

**Assumption 5.8.** For any given charging strategy, $u(t)$, and $x_s \in X_s$, $\hat{F}_s(x_s, e)$ in (5.25) and $F_s(x_s, e)$ are sufficiently close.

Justification for Assumption 5.8: This is examined in terms of several typically used charging strategies, including the constant-current constant-voltage (CCCV) charging and multi-stage-CC (MCC) charging. For the MCC algorithm, the battery is charged under a large constant-current in the beginning, and is then charged by lower constant-currents for further several stages. For each charging strategy, the battery is started from the same initial states. The simulation results for both the charging strategies are given in Fig. 5.4. It is found that the profiles of $\hat{F}_s$ closely follow its counterpart $F_s$, while the errors over 200 cycles in prediction of battery SOH are within 1%. At such a scale of errors, it is reasonable to use the approximation in (5.25). Furthermore, for battery state estimation, the existing errors are supposed to be compensated by the designed robust estimator.

In addition to the justification of Assumptions 5.1-5.4 shown before, the validity of other assumptions underpinning Proposition 1 is examined on the developed battery models.

Justification for Assumption 5.5: It can be easily founded that in the reduced models, $\Sigma_f$ and $\Sigma_s$, the functions, $F_f$, $F_s$, $H_f$, $W_f$, $W_s$, are Lipschitz continuous. The requirement on bounded states and input also follows the nature of practical battery charge and discharge operations.
Figure 5.4: The quasi-steady-state map, \( F_s(x_s,e) \), and its approximation, \( \hat{F}_s(x_s,e) \), for different charging strategies. The left-hand-side shows the MCC charging results, while the right-hand-side presents the CCCV charging results. \( F_s \) is drawn in blue solid line, and \( \hat{F}_s \) is represented by the red dot-dashed line.

Justification for Assumption 5.6: The local observability can be examined by calculating the corresponding observability matrix \( O = [C;CA;\cdots;CA^{n-1}] \) for both the slow and fast models. A comprehensive introduction or the observability matrix calculation and analysis refers to [64, 163]. By means of numerical calculation, the rank of observability matrices for both the battery models during operations can be obtained and are shown in Fig. 5.5. Given the full rank is obtained over the whole estimation process, the two reduced models are thus fully observable in a linear sense.

Proposition 1 relies on the use of an EKF, as depicted in Assumption 5.7. However, as noted
in Remark 5.2, it may be possible to replace the EKF with other estimators. Here both the EKF and a UKF alternative are considered, although the latter is not currently shown to satisfy Proposition 1.

Justification for Assumption 5.7: The EKF design method can be implemented based around the models \( \Sigma_f \) and \( \Sigma_s \). Within the framework of continuous-time EKF, the gains, \( K_f(t) \) and \( K_s(\tau) \), are calculated from

\[
K_f = P_f C_f^T (C_f P_f C_f^T + R_f)^{-1}, \tag{5.26a}
\]
\[
K_s = P_s C_s^T (C_s P_s C_s^T + R_s)^{-1}. \tag{5.26b}
\]

The covariances in (5.26), \( P_f(t) \) and \( P_s(\tau) \), are propagated and updated through (5.27)

\[
\dot{P}_f = A_f P_f + P_f A_f^T + Q_f - P_f C_f^T R_f^{-1} C_f P_f, \tag{5.27a}
\]
\[
P_f(0) = P_{f0}, \tag{5.27b}
\]
\[
\dot{P}_s = A_s P_s + P_s A_s^T + Q_s - P_s C_s^T R_s^{-1} C_s P_s, \tag{5.27c}
\]
\[ P_f(0) = P_{d0}, \]  

(5.27d)

where, \( A_f, A_s, C_f, C_s \) are the Jacobian matrices defined in (5.18a)-(5.18c), \( Q \) represents covariances of the process noises, and \( R \) denotes covariances of the measurement noise.

Unlike the stochastic sampling technique used in the EKF, the unscented transform is used to pick a minimal set of sigma points around the mean. These sigma points are directly propagated through the nonlinear models of \( \Sigma_f \) and \( \Sigma_s \). The mean and covariance of the estimate are then recovered from the sigma points. The mathematical equations for UKF propagation and update are not repeated here but the readers are referred to literature such as [164].

When all the assumptions are satisfied, we are allowed to design an appropriate observer for SOC and SOH estimation using the derived battery models and the methodology of multi-time-scale observer design. With the state \( \hat{x}_f \) and \( \hat{x}_s \) obtained from the observers, \( \hat{z}_{SOC} \) and \( \hat{z}_{SOH} \) can be calculated according to their output functions. In this proposed estimation algorithm, the design parameters, including \( (P_{f0}, Q_f, R_f) \) and \( (P_{s0}, Q_s, R_s) \), can be tuned separately in different time-scales without risk of ill-conditional gains and
5.4 Estimation Result II

To demonstrate the proposed multi-time-scale estimation methodology, simulations are conducted on a lithium-ion battery. The estimated states are validated against the results from the original high-fidelity battery model, $\Sigma^2$, described in Chapter 3.

5.4.1 Estimation Results in Fast Time-Scale (SOC)

To test the proposed estimation algorithm in terms of the fast dynamics, the UDDS current shown in Fig. 3.12 is employed as the system input. To check the robustness of the proposed estimator, additional white Gaussian noise with a standard deviation of 5mV is added to the voltage measurement. In addition, the fast time-scale states are initialised with 50% initial errors.

Fig. 5.7 depicts the estimation results regarding the terminal voltage and SOC by using EKF and UKF. It can be seen that the estimated states using both the EKF and UKF converge to the true values rapidly from the imposed initial error. Furthermore, the steady-state errors are less than 1% for the terminal voltage and less than 3% for the SOC.

In this case, the UKF has similar accuracy to the EKF. Meanwhile, the UKF exhibits slightly faster convergence in this simulation, but it comes at increased computational expense.

5.4.2 Estimation Results in Two Time-Scales (SOC + SOH)

A multi-stage CC charging protocol is now considered to investigate the multi-time-scale estimation of SOC and SOH. The battery is charged with multi-stage constant current, where the current is 2.5C, 1.5C, 1C, and 0.5C, respectively, as shown for one cycle in Fig. 5.8. Each time the terminal voltage reaches the maximum safe voltage level of 4.2V,
the current is switched to a lower current. A relaxed period is maintained when the battery is fully charged. The discharging operation occurs at 1C until the terminal voltage drops to 3.3V, followed by a final relax period. In this simulation, all the slow states are initialised with 10% initial errors, while the fast time-scale states are initialised with 20% errors.

Simulations for the designed estimator over 450 cycles are conducted under the specified charging protocol. Fig. 5.9 illustrates the estimation result for SOC and SOH, where the SOC trajectory of its first 40 cycles is provided for visualisation. By deliberately using relatively high charging rates, one observation from this figure is that the battery degrades to its end of life, i.e. 80% of the initial SOH, in these operating cycles. Regarding the estimation performance, the designed EKF is found to quickly reject the initial error at the fast states within the first 0.1 cycles, and the slow state initial errors are rejected within 10 cycles.

All the estimated states converge to the vicinity of their true values. The steady-state estimation errors in the SOC and SOH are bounded by 3%. These results verify Assumption 5.7 and demonstrate the effectiveness of the proposed multi-time-scale estimation algorithm.

For comparison, the proposed multi-time-scale estimator is also implemented with a UKF. It shows similar results as the EKF in the estimation accuracy but faster convergence can be potentially obtained, albeit again at increased computation cost. This indicates there is merit in extending Assumption 5.7 to consider wider classes of estimation algorithms.

The computational efficiency of the proposed multi-time-scale estimator is quantitatively evaluated, where the estimator designed in Section 5.1 is used as a benchmark. The tic and toc functions in MATLAB work together to measure the elapsed time. For both the estimators, the algorithm EKF is used, and the system input and simulation setup are identically specified. The comparison result of simulating 40 operating cycles is shown in Table 5.1. Clearly, the multi-time-scale estimator can be implemented much faster than its counterpart. More accurately, 95% of simulation time can be saved through time-scale
Table 5.1: Computational time comparison between the multi-time-scale and one-time-scale estimators for simulating 40 operating cycles.

<table>
<thead>
<tr>
<th>Estimators</th>
<th>Computation time (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>One-time-scale estimator</td>
<td>1150</td>
</tr>
<tr>
<td>Multi-time-scale estimator</td>
<td>55.7</td>
</tr>
</tbody>
</table>

separation. In contrast, a large amount of time is required by the one-time-scale estimator to monitor the SOH in the fast time-scale and to calculate a set of matrices online whose size remarkably increases when considering the slow dynamics.

Based on this analysis, the designed nonlinear robust estimator for SOC and SOH within the multi-time-scale estimation framework is effective in dealing with model mismatch, initial errors and noise. Meanwhile, steady-state errors have been observed in the estimation results, this is consistent with theoretical analysis. That is, exact convergence to the origin for the estimation errors is not possible due to the presence of uncertainties. Furthermore, significantly computational time-saving can be achieved by the proposed approach relative to the one where the SOC and SOH are estimated in the same time-scale.

In this work, the accuracy of simplified models has been validated against the initial battery model. Although a dominant ageing mechanism namely SEI film based ageing has been considered in this model, various factors may contribute to battery SOH change. The identification and quantification of all ageing mechanisms are still an ongoing work. It is important for readers to note that a different SOH model can be easily fitted into the proposed methodology of multi-time-scale estimation.

5.5 Conclusion

An EKF-based estimation algorithm synthesised from a reduced-order battery model that describes the electrochemical and ageing dynamics has been proposed for state estimation. Illustrative results showed that this designed estimator is effective in estimation of the SOC and SOH.
In view of potential issues in singularly perturbed systems such as heavy computational burden and ill-conditioned estimator gains, a multi-time-scale estimation technique has been developed. Assumptions imposed on the internal models and the estimation error dynamic systems were explicitly stated. It has been analytically proved that under these assumptions, an appropriately designed estimator is able to compensate the uncertainties including modelling errors brought about by time-scale separation. The proposed estimation algorithm was applied to a Li-ion battery. It has been demonstrated capable of accurate estimation of SOC and SOH in different time-scales despite a range of common errors. Furthermore, the established methodology for state estimation of singularly perturbed systems is very general so that it can be applied to other battery chemistries where proper models exist.
Figure 5.7: Estimation results of the terminal voltage and SOC under validated against the true values. (a) The terminal voltage. (b) The relative absolute error in voltage estimation. (c) The ion concentration in the anode particle. (d) The relative absolute error in SOC estimation.
Figure 5.8: The current and voltage profiles in the 4-stage CC charging protocol.
Figure 5.9: Estimation results of the SOC and SOH in multi-time-scales. (a) The true SOC (black solid line) and its estimated values from the EKF (red dot-dashed line) and UKF (blue dotted line). (d). The relative absolute error in SOC estimation. (b) and (c) are separately the zoom-in figures of (a) and (d) in their first cycle. (e) and (f) are the SOH profiles and respective estimation errors.
Chapter 6

Model Predictive Control for Battery Optimal Charging

1

THE charging time and lifetime are important performances for lithium-ion (Li-ion) batteries, but are often competing objectives for charging management. Model-based charging controls are challenging due to the complicated battery system structure that is composed of nonlinear partial differential equations (PDEs) and involved in multiple time-scales. This chapter proposes a new methodology for battery charging control, enabling an optimal trade-off between the charging time and battery state-of-health (SOH). Using recently developed model reduction approaches, a physics-based low-order battery model is first proposed and used to formulate a model-based charging strategy. The optimal fast charging problem is formulated in the framework of tracking model predictive control (MPC). This directly considers the tracking performance for provided SOC and SOH references, and explicitly addresses constraints imposed on input current and battery internal state. The capability of this proposed charging strategy is demonstrated via simulations to be effective in tracking the desirable SOH trajectories. By comparing to the constant-current constant-voltage (CCCV) charging protocol, the MPC-based charging appears promising in terms of both the charging time and SOH. In addition, this obtained charging strategy is practical for real-time implementation.

1A substantial proportion of this chapter has been submitted for publication in the IEEE/ASME Transactions on Mechatronics [165].
6.1 Introduction

For the electric vehicles, the high upfront cost, largely contributed by its Li-ion battery system, and the long charging time are unavoidable problems [5]. Although the battery price is predicted to decrease in the future [4], an immediate solution for cost-saving is to increase battery’s lifetime. The battery degradation is mainly dependent on charging strategies used [8] and in general, the lifetime is a competing factor to the charging time. To charge the battery as fast as possible but without unduly compromising its lifetime, optimal fast charging strategies are needed.

The model-free approaches such as constant-current constant-voltage (CCCV) and multi-stage CC in [19, 22] are ad hoc and often conservative with respect to the charging time and battery lifetime. The main reason is that charging profiles in the model-free approaches are determined by some pre-defined current and voltage limits irrespective of battery in-situ physical and chemical characteristics [3]. This motivates the development of model-based algorithms for battery charging. The existing technical challenges to realise this arise from the complexity of battery dynamic system and unmeasurable internal states. In view of this, it would be desirable to fuse model-based charging strategies with model simplifications and state estimation.

Recently, some attempts have been made to use physics-based models and MPC for charging strategy development. A one-step MPC was presented for battery optimal charging based on an electrochemical-thermal model [142]. Based on a simplified single particle model (SPM), a nonlinear model predictive control problem was formulated for battery fast charging [143, 166]. To overcome the potential computation and non-convex issues therein, a differential flatness approach was used to reformulate the model. Other charging strategies developed from physical model-based control can be referred to the work, e.g. [145–148]. However, none of these charging algorithms have explicitly considered battery ageing dynamics. Furthermore, the battery internal electrochemical states were assumed available in these control algorithms.

To address the issues discussed above, a novel and practical charging strategy based on
6.2 Optimal Charging Strategy

In this section, we describe the formulation of model predictive control for optimal charging of a Li-ion battery. The first step in the proposed approach is the specification of the reduced order model to be used in the controller.

6.2.1 Control-Oriented Battery Model

A Li-ion battery containing coupled electrical, electrochemical, thermal and ageing dynamics via 17 PDEs is used as the starting point for control-oriented modelling [149]. Model reductions can be systematically conducted in a two-step process. First, since the electrochemical-thermal dynamics are much faster than the degradation process but significantly slower than the electrical dynamics, time-scale separation techniques are used to decouple subsystems. This results in libraries of simplified PDE battery models approximating the initial model.

The second step is to reduce the PDEs to computationally tractable ODE systems using standard methods. This includes polynomial approximations and finite differencing separately for spatial dimension reduction and discretisation. The resulting models arising from these simplifications are detailed in Chapter 3, and have been proposed for use in
state estimation in Chapter 5.

Within the reduced order model, the following state vector $x := [C_s, q_s, C_e, T, Q_{sr}]^T$ is defined with input, $u$, representing the applied current. The output vector of interest is defined as $z := [SOC, SOH, T, V]^T$. Note that state-of-charge and health are defined as

$$SOC(t) = \frac{C_s(t) - C_{s,0\%}}{C_{s,100\%} - C_{s,0\%}}$$

$$SOH(t) = 1 - \frac{Q_{sr}(t)}{Q_{\text{max}}},$$

These two states are not directly measurable, and must be estimated online for battery monitoring and state-feedback controls.

Consequently, the control-oriented battery model is summarised in the form of

$$\dot{x}(t) = f(x(t), u(t))$$

$$z(t) = g(x(t), u(t)) := \begin{bmatrix} C_1x(t) + c_1 \\ C_2x(t) + c_2 \\ C_3x(t) \\ h(x(t), u(t)) \end{bmatrix}$$

where $f, g, h$ are nonlinear functions, $C_1, C_2, C_3$ are constant matrices, and $c_1, c_2$ are constants. A full description of the model structure (6.2) is provided in [159].

We now develop a model-based control approach utilising the reduced order model, beginning with a short overview of the general formulation of the MPC tracking problem.

### 6.2.2 General MPC Tracking Problem Formulation

The formulation of a general MPC tracking problem from [167, 168] is presented in this subsection, before practical modifications to this approach are proposed in the subsequent subsection.

For a given exogenous reference signal, $y'$, suppose an admissible steady-state, $(x', u')$,
can be generated for the state and input. The decision variable, $u(k)$, is the sequence of the predicted inputs over the control horizon namely $u(k) = \{u(k), \cdots, u(k + N - 1)\}$. The notation $\| \cdot \|^2$ is interpreted as the squared weighted 2-norm, i.e. $\|x\|^2_Q = x^TQx$. The open-loop optimisation problem solved at each time step $k$ with $x_k$ as a state measurement is given by

$$
\begin{align*}
\mathbf{u}^*(k) &= \arg\min_{\mathbf{u}(k)} \sum_{i=0}^{N-1} \|x(i) - x'(k + i)\|^2_Q + \|u(i) - u'(k + i)\|^2_R \\
&\quad + \|x(N) - x'(k + N)\|^2_P \\
&\text{subject to } \forall i \in \{0, \cdots, N - 1\} \\
x(i + 1) &= Ax(i) + Bu(i) \tag{6.3b} \\
x(0) &= x_k \tag{6.3c} \\
u(i) &\in \mathcal{U} \subset \mathbb{R}^m \tag{6.3d} \\
x(i) &\in \mathcal{X} \subset \mathbb{R}^n \tag{6.3e} \\
x(N) &\in \mathcal{X}_f \subset \mathbb{R}^n \tag{6.3f}
\end{align*}
$$

The following paragraphs highlight the constituent parts of the algorithm.

**Cost function.** In (6.3a), on the right-hand side, the first two terms consist of the stage cost, penalising the deviation between the system input/state and their corresponding future target trajectories. The stage cost is formulated to invoke desired dynamic behaviour throughout the prediction horizon, $N$, by balancing tracking error against the control effort required to achieve it through appropriate selection of the matrices $Q$ and $R$. The third term is a terminal cost, used to enforce convergence towards the desired operating condition, and weighted through the matrix $P$. As discussed in [30], the three weighting matrices $P$, $Q$ and $R$ must be positive semi-definite to ensure stability.

**Horizon length.** Lengthening the prediction horizon, $N$, typically increases the guaranteed domain of attraction about the desired equilibrium point, albeit at a computational cost.

**Prediction model.** (6.3b) represents the nominal system dynamic model through which the future states can be propagated for prediction. $x(i), u(i)$ are separately the predicted state
and input $i$ time step into the future from the current time $k$. In (6.3c), $x(0)$ is initialised using the current state.

**Constraints.** (6.3d)-(6.3e) present the constraints applied to the system input and state. Note that output constraints may be mapped to state constraints. The constraint sets $X$ and $U$ are convex and closed subset of $\mathbb{R}^n$ and $\mathbb{R}^m$, respectively. In (6.3f), a terminal constraint is used to provide a stability guarantee. In the presence of plant-model mismatch and other disturbances, the constrained optimisation problem (6.3) may become infeasible. It is possible to alleviate this issue by softening some or all of the constraints at the potential cost of constraint adherence.

**Implementation of the controller.** At each time step $k$, the optimal input sequence, $u^*(k)$, is obtained by solving the optimisation problem (6.3). Only the first input, $u^*(k)$, is applied to the system, while the remaining inputs namely $\{u^*(k+1), \cdots, u^*(k+N-1)\}$ are discarded. This receding horizon implementation ensures continual feedback in the close-loop system, thereby providing some inherent robustness in this approach [29].

### 6.2.3 Optimal Battery Charging Problem Formulation

The optimal charging control problem is now formulated by employing the battery model developed in Section 6.2.1 and modifying the general MPC algorithm presented in Section 6.2.2. The first stage in the problem definition is the specification of reference trajectories for the state of charge and health.

**SOC and SOH references.** To ensure reachability of the reference trajectories the following assumption is placed on both reference specifications:

**Assumption 6.1.** The change rates of SOC and SOH reference signals with respect to the time satisfy the condition

\[
0 \leq \left( \frac{\partial \text{SOC}^r}{\partial t} \right)_{\text{min}} \leq \frac{\partial \text{SOC}^r}{\partial t} \leq \left( \frac{\partial \text{SOC}^r}{\partial t} \right)_{\text{max}} \leq \left( \frac{\partial \text{SOH}^r}{\partial t} \right)_{\text{min}} \leq \frac{\partial \text{SOH}^r}{\partial t} \leq \left( \frac{\partial \text{SOH}^r}{\partial t} \right)_{\text{max}} < 0
\]
This assumption avoids nonsensical references being provided to the controller, such as attempting to achieve fast charging with zero state of health degradation within the cell. In Section 6.3, the state of charge reference will be determined through simulation (without consideration of state of health), whilst the state of health reference over a long time scale will be user specified, but satisfying (6.5).

**Battery model.** This step describes the development of an appropriate battery model to be used in (6.3b). To further reduced the computation complexity of model-based control in processors of battery management systems, the nonlinear battery model (6.2) is successively linearised based on the reference trajectory and previous system input, resulting in

$$x(k + i + 1) = A_{k+i}x(k + i) + B_{k+i}u(k + i) + d_{k+i}$$

(6.6)

where $A_{k+i}, B_{k+i}, d_{k+i}$ are defined as

$$A_{k+i} = \frac{\partial f(x(k+i), u(k-1))}{\partial x}$$

(6.7a)

$$B_{k+i} = \frac{\partial f(x(k+i), u(k-1))}{\partial u}$$

(6.7b)

$$d_{k+i} = f(x(k+i), u(k-1)) - A_{k+i}x(k + i) - B_{k+i}u(k - 1)$$

(6.7c)

It is noted that in (6.6), $N$ linearisations are required along the prediction horizon to solve the optimisation problem. This may pose computational issues, particularly for a large value of $N$. To alleviate the computational burden caused by successively calculating the linearisation matrices, the linearised model is assumed to be time-invariant over the horizon. Explicitly, the following further approximation is employed in developing the nominal plant model:

$$A_{k+i} \approx A_k, B_{k+i} \approx B_k$$

(6.8)

$$d_{k+i} \approx d_k, \forall i \in \{0, \cdots, N - 1\}$$

(6.9)
The system outputs of interest for battery charging control include the SOC and SOH degradation with the definition of

\[ y(i) := [\text{SOC}(i), \Delta \text{SOH}(i)]^T \]  (6.10a)

\[ \Delta \text{SOH}(i) := \text{SOH}(i) - \text{SOH}(i - 1) \]  (6.10b)

By combining (6.2b) and (6.10), the output equation for the nominal model can be rewritten as

\[ y(i) = Cx(i) := [C_1 x(i), C_2 x(i)]^T \]  (6.11)

The fidelity of the obtained battery model via (6.6)-(6.11) will be examined in Section 6.3.1.

Cost function. Using the reachability condition of the reference signals (Assumption 1), it follows from [168] that the MPC cost function can be reformulated as an output tracking problem. Furthermore, given the state of charge is simply the integral of the system input (the charging current) and the (6.4) has been applied, then the input weight vector can be set as \( R := 0 \). This leaves the stage cost as containing two competing objectives of maximising charge rate whilst simultaneously minimising change in state of health. The balance between these two factors may be captured by the use of a weight, \( w \), in the output penalty matrix:

\[
Q := \begin{bmatrix}
1 & 0 \\
0 & w
\end{bmatrix}
\]  (6.12)

The following assumption is now placed on \( w \):

**Assumption 6.2.** There exists an optimal \( w^* \) such that minimising the stage cost over the horizon using the \( P, Q, \) and \( R \) matrices defined above achieves the permitted state of health degradation specified by the reference trajectory, \( \text{SOH}^* \).

In general, the optimal value \( w^* \) is not known a priori, and must be obtained using some form of online algorithms. The online update law for \( w \) can be given in the following
6.2 Optimal Charging Strategy

general form, with the error feedback taken as the SOH tracking error in the slow time scale, \( j \), i.e.:

\[
w_{j+1} = W (w_j, SOH'(j) - SOH(j))
\]  \( (6.13) \)

where \( W(\cdot) \) is a monotonic function ensuring \( w \) converges \( w^* \). If \( w \) is linearly related to \((SOH' - SOH)\), then the integral control can be employed here, of which the convergence properties are well established. Similar approaches have been proposed in other application areas such as hybrid vehicle powertrain control [169].

The matrix \( P \) is calculated from the following discrete Lyapunov equation

\[
(A_k + B_k K)^T P_k (A_k + B_k K) - P_k = -(Q + K^T R K)
\]  \( (6.14) \)

where \( K \) is a stabilizing control gain such that \((A_k + B_k K)\) is Schur. Since \( A_k \) and \( B_k \) are time varying, this makes \( P_k \) time varying, unlike in [167].

*Input and State constraints.* Input constraints are specified for charging (defined as a negative input current) up to some physically allowable limit, i.e.:

\[-I_{\text{max}} \leq u(i) \leq 0\]  \( (6.15) \)

The only constraints that need to be applied to the battery internal states reflect the need to avoid permanent damage to the battery chemistry. Whilst there is no universally accepted model to capture all phenomena such as solid-electrolyte film growth, lithium plating and electrolyte degradation, it is known that maintaining certain key states within acceptable ranges can avoid these problems. The following state constraints are considered in the charging problem:

\[
C_{ss,\text{min}} \leq C_{ss}^i (i) \leq C_{ss,\text{max}} \quad (6.16a)
\]

\[
C_{e,\text{min}} \leq C_{e}^i (i) \leq C_{e,\text{max}} \quad (6.16b)
\]

\[
T(i) \leq T_{\text{max}} \quad (6.16c)
\]
or in the compact form for $i = \{1, \cdots, N - 1\}$:

$$Mx(i) \leq c_0$$  \hspace{1cm} (6.17)

Note that the terminal set constraint (6.3f) can potentially also be represented in this compact form. In alternative linear-time-varying (LTV)-MPC implementations, such as [31], the terminal set is replaced with point constraint, $X_f = 0$, in order to demonstrate provable asymptotic stability. The inherent stability of the battery dynamics (i.e. SOC and SOH are naturally restricted to the range of $[0, 1]$) potentially alleviates the need to consider this set constraint, whilst guaranteeing overall system stability. Consequently, the terminal set constraint is relaxed in this case, with stability checked only via case studies in Section 6.3.

Given the uncertainty of the impact associated with explicit violation of these constraints, it is proposed to implement them as soft constraints by introducing a vector of slack variables, $s$. This will alleviate any infeasibility issues in the solution of the optimisation problem, at the cost of increasing the number of decision variables in the optimisation problem.

Under this relaxation, an additional term $s^T Y s$ is added to the stage cost, and the state constraints are replaced by:

$$Mx(i) \leq c_0 + s$$  \hspace{1cm} (6.18)
$$s \geq 0$$  \hspace{1cm} (6.19)

### 6.2.4 State Estimation

The current battery state, $x(0)$, is required to initialise the prediction model for solving the optimisation problem. A model-based estimation algorithm developed in [159] can be adopted here to estimate the required states, $\hat{x}$, and the output, $\hat{y}$. Based on the model
(6.2), the estimator is formulated in the following

\[ \hat{x}(k+1) = f(\hat{x}(k), u(k)) + L_k \cdot [z(k+1) - \hat{z}(k+1)] \]  
\[ \hat{z}(k+1) = g(f(\hat{x}(k), u(k)), u(k)) \]  
\[ \hat{y}(k+1) = C\hat{x}(k+1) \]

The details including calculation of the estimator gain \( L_k \) can be found in [159]. As shown therein, this estimator is effective in obtaining SOC and SOH despite a range of common errors due to model order reduction, linearisation and measurement noise.

6.2.5 Overall Problem Formulation

Based on the analysis in Sections 2.1-2.4, the overall problem formulation for battery health-conscious fast charging is summarised as

\[ u^*(k) = \arg \min_{u(k)} \sum_{i=0}^{N-1} \| \hat{y}(i) - y'(k+i) \|_Q^2 + \| s \|_Y^2 + \| \hat{y}(N) - y'(k+N) \|_P^2 \]  
subject to \( \forall i \in \{0, \cdots, N-1\} \)

\[ \hat{x}(i+1) = A_k \hat{x}(i) + B_k u(i) + \hat{d}_k \]
\[ \hat{y}(i) = C\hat{x}(i) \]
\[ \hat{x}(0) = x_k \]
\[ -I_{max} \leq u(i) \leq 0 \]
\[ M\hat{x}(i) \leq c_0 + s \]
\[ s \geq 0 \]

Furthermore, this proposed control algorithm is illustrated in Fig. 6.1. For given references, \( SOC^r \) and \( SOH^r \), and state information from the estimator, \( \hat{x}(k) \), the designed MPC controller calculates the optimal input current, \( u(k) \), at each time step \( k \). The inherent robustness of receding horizon control will reject small disturbances arising from the
output estimation, $\hat{y}$, rather than measurements.

### 6.3 Simulation Results

The proposed charging strategy is implemented in this section through simulations on a high fidelity battery model described by 10 coupled PDE equations across the electro-chemical, thermal and ageing domains [149]. The implementation of the PDEs utilises the high-resolution simulator described in [78, 170], with appropriate augmentation for the battery ageing states. The model is parameterised using Li-ion parameters taken from [8, 171], and the simulations are carried out using MATLAB R2012b and YALMIP [172].

#### 6.3.1 Simulation Model

To evaluate the accuracy of the reduced order model relative to the original PDE system, comprehensive simulations are conducted over a range of four constant current tests and the Urban Dynamometer Driving Schedule (UDDS). The root-mean-square error (RMSE) in terminal voltage is used to infer the model accuracy over the five tests as shown in Table 6.1.

Whilst the prediction errors increase at higher charging rates, the RMS error remains below 35mV (corresponding to about 1% error on a normalised scale) in all tests indicating good agreement between the models. For illustration, the trajectories of the two models
6.3 Simulation Results

Figure 6.2: Validation of the proposed model under the UDDS test.

Table 6.1: Accuracy of the reduced ODE model relative to the initial PDE model

<table>
<thead>
<tr>
<th>C-rate</th>
<th>0.5C</th>
<th>1C</th>
<th>2C</th>
<th>4C</th>
<th>UDDS</th>
</tr>
</thead>
<tbody>
<tr>
<td>RMSE (mV)</td>
<td>11.2</td>
<td>17.9</td>
<td>24.3</td>
<td>35.4</td>
<td>15.1</td>
</tr>
</tbody>
</table>

run over the UDDS cycle in open loop are provided in Fig. 6.2.

6.3.2 Explicit Problem Formulation

In this section, the remaining parameters required in the problem formulation are obtained. These include the additional weighting matrices, constraints and reference trajectories.

To generate a battery SOC reference, the fastest possible charging strategy is considered. This is determined by ignoring any SOH penalty, i.e. setting \( w = 0 \) in (6.12). In this case, the weighting matrix penalising non-zero slack variables is chosen as \( Y = 10^5 \times I \). The prediction horizon is considered to be \( N = 10 \).
The next step is to determine the stabilising control gain, $K$, for the system which must satisfy the condition

$$|\lambda(A_k + B_k \bar{K})| < 1 \quad (6.22)$$

where $\lambda(X)$ is taken to be the maximum eigenvalue of $X$. This could be potentially achieved by solving an optimisation problem for all $A_k$ and $B_k$ of the form:

$$\min_{\bar{K}} |\lambda(A_k + B_k \bar{K})| \quad (6.23)$$

This would result in a single $\bar{K}$ that can be used in (6.14). Alternatively, it may be possible to identify $\bar{K}$ satisfying (6.22) using a heuristic approach. The latter approach leads to the selection $\bar{K} = -[1.22 \times 10^{-5}, 7.63 \times 10^{-6}, -7.30 \times 10^{-12}, 7.34 \times 10^{-10}, 1.35 \times 10^{-21}]$, which was found to satisfy (6.22) for all $A_k, B_k$ in the five tests described in Section 6.3.1.

Once $K$ is established, the solution for $P_k$ may be obtained from (6.14) at each iteration.

The setup for (6.16) will depend on battery cells and their responding applications. For the sake of demonstrating the proposed charging strategy, we choose the upper and lower bounds for the surface concentration as $C_{ss,max} = 0.95C_{s,max}$ and $C_{ss,min} = 0.05C_{s,max}$, where the parameter $C_{s,max}$ represents the maximum possible solid-phase Li-ion concentration. The aggressive cases namely $C_{ss,max} = C_{s,max}$ and $C_{ss,min} = 0$ are not used here to protect the battery in consideration of state errors due to modelling, estimation and disturbance. Similarly, $C_{e,min} = 0.1C_{e0}$ and $C_{e,max} = 2C_{e0}$ are set up for the electrolyte state, where the parameter $C_{e0}$ is the quasi-steady-state electrolyte concentration. The maximum temperature is $T_{max} = 40^\circ$. This is a reasonable value to prevent over-heating. For the input constraint, $I_{max} = 10C$, corresponding to 6 minutes to fully charge a battery.

The resulting fastest possible charging trajectory is illustrated in Fig. 6.3. As might reasonably be expected, the largest charging rates are present when no state constraints are active. Furthermore, there does not appear to be any significant constraint violations due to the large penalty matrix $Y$. Subsequently, the developed trajectories are chosen as reasonable for adoption as $y^*_1$ in (6.21a) when implementing the reference tracking MPC.
Figure 6.3: Battery SOC reference generated by setting $w = 0$. (a) Input current. (b) State of charge. (c) Normalised surface concentration. (d) Normalised electrolyte concentration. (e) Temperature.

The reference for state of health degradation, $y^r_2$, is taken to be specified by the user as satisfying Assumption 6.1, with different choices to be investigated in Subsection 6.3.3.
Assumption 6.2 requires an optimal $w^*$ exists to deliver the desired balance between fast charging and state of health degradation, whilst this optimal value can potentially be determined through selection of an iterative law (6.13).

To first test the validity of Assumption 6.2, a value for $w$ is chosen from the set \{0.001, 0.01, 0.1, 1, 10, 100, 1000\} and simulation over one charging event is performed with the control strategy (6.21). The charging completion times and SOH evolution profiles derived from the obtained charging strategies are illustrated in Fig. 6.4. It can be seen that as $w$ increases from 0.001 to 1000, the change in state of health is diminished at the expense of charging time. Whilst this trend is expected, thus validating Assumption 6.2, the nonlinear change in $w$ to achieve a linear change in SOH is potentially problematic.

To deal with this circumstance, the update of $w_j$ must be carefully chosen to ensure the required range of possible values can be covered. Consequently, the following two-step
update is proposed, and necessitates the introduction of a new intermediate variable, $a_j$:

$$a_{j+1} = a_j + K_I \cdot (SOH'(j) - SOH(j))$$

(6.24a)

$$w_{j+1} = 10^{q_{j+1}}$$

(6.24b)

where, the integral control gain is chosen as $K_I = 250$, and $a_0 = 1.2$.

### 6.3.3 Simulation results for the proposed strategy

The proposed strategy (6.21) with the specific aspects introduced in Subsections 6.3.1-6.3.2 can be now implemented.

The computational requirement of the proposed charging strategy is first studied. The sampling time for the fast electrochemical and thermal dynamics is chosen to be 1 second. The simulations are conducted on a desk computer with 3.4GHz processor and Gurobi is used as the optimizer. The time solving the optimisation problem (6.21) is less than 10 milliseconds. This means that the computation time is not an issue for real-time implementation of the proposed charging strategy.

As an initial comparison, the proposed method with the fixed weight $w = 1$ is compared to the well-known CCCV strategies [19]. The first CCCV charging profile, denoted CCCV1, is specified to have the current limit of 5C and voltage limit of 4.2V. Comparing the proposed MPC strategy and CCCV1 over a single charging cycle is displayed in Fig. 6.5.

It can be seen that the CCCV1 (green dot-dash lines in Fig. 6.5) takes similar charging time as the MPC based approach to accomplish the same charging task, but leads to 47% greater decrease in battery state of health. The improved relative performance of the proposed strategy can be attributed to the incorporation of a state of health model within the approach. This allows a more aggressive charging approach when it will not unduly impact SOH. The decrease in voltage under the MPC approach at 950 seconds is due to the related decrease in charging current, as might intuitively be expected.
Figure 6.5: Comparison of the proposed charging strategy with the industry standard charging methods, CCCV1 and CCCV2.

Given the voltage limit in a CCCV charging will impact the degradation rate during operations [19], a second CCCV charging profile, denoted CCCV2, with the voltage limit of 4.15V is considered for further comparison. The charging results arising from the use of CCCV2 are also provided in Fig. 6.5 (red dash lines). These depict a reduction in state of health degradation as might reasonably be expected from the reduced voltage limit. However, the charging time is no longer equivalent to the proposed MPC strategy,
6.4 Conclusion

reinforcing the benefits of utilising model-based information in the charging approach.

To demonstrate the ability of the proposed approach to track a reference SOH trajectory over a large number of cycles, the adaptive weight law (6.24) is now activated and simulations are performed over a longer time period. In the first instances, the battery lifetime (taken to be when the SOH degrades to 80% of its maximum value) is taken to be 200 and 400 cycles respectively, with constant associated degradation rates. The results are depicted in Fig. 6.6.

It may be observed that the SOH in both cases tracks the reference trajectories with only a slight oscillation about zero tracking error. If desired, further tuning of the algorithm (6.24) and the gain $K_I$ may lead to reductions in these fluctuations, although they are not too severe in this case. The effect on charging time is also presented in Fig. 6.6(b), where it can be observed that the effect of doubling time almost three times as long.

To further demonstrate the utility of the proposed approach, an additional scenario is now considered whereby the battery is initially allowed to be charged quite aggressively, over a period of 100 cycles, before switching to a more conservative charging mode. The results depicted in Fig. 6.7 demonstrate successful tracking in this scenario, which is not as easily handled by existing CCCV approaches. Furthermore, it highlights the potential for “second-life” uses of batteries enabled by appropriate control actions.

6.4 Conclusion

In this chapter, a new charging algorithm has been proposed for a Li-ion battery to balance the competing objectives of battery lifetime and charging time. Under the proposed algorithm, the user can specify the balance through a desired SOH evolution profile.

The novelty of this work arises from model-based control algorithm for battery fast charging. Specifically, an optimal charging control problem has been formulated for SOC and SOH reference tracking based on the MPC algorithm, which makes use of an internal battery model and explicitly handles operating constraints.
Figure 6.6: Tracking performance of two given battery SOH evolution references and their corresponding charging times.

The capability of the proposed approach has been demonstrated by incorporating a multi-time-scale estimation algorithm in high fidelity simulations. Significant improvement was observed in terms of charging time and state of health preservation relative to the industry standard charging algorithms.
Figure 6.7: Tracking performance of the third given SOH reference and its corresponding charging time.
Chapter 7
Conclusion

This thesis has presented new techniques and algorithms for applications of a Li-ion battery including modelling, charging strategy evaluation, state estimation and control. This chapter summarises the contributions that have been made to the literature and reveals the potential directions for future research.

7.1 Summary of Contributions

Motivated by the requirements of Li-ion batteries for further mobile electrification, this thesis pursued to develop optimal solutions for advanced battery management. The research aims established in Chapter 2 have been addressed with four distinct and original contributions. These contributions include a framework for systematic PDE battery model simplification, an evaluation procedure for charging strategies, an estimation algorithm for battery SOC and SOH, and an optimal fast charging strategy.

7.1.1 Systematic Battery Model Simplification

Starting from an initial PDE-based model for a Li-ion battery that captures coupled electrochemical, thermal, electrical and ageing dynamics, a novel framework for model simplification was developed using singular perturbation techniques. The novelty arises from the systematic procedure for model simplification through which the assumptions underpinning simplified models can be explicitly stated. Numerical approaches were
Conclusion and Future Work

Further used to identify the minimal modelling requirements for specific accuracy and operating conditions. Simulation results have demonstrated that the simplified models are capable of efficiently predicting battery characteristics with significantly reduced computational time relative to the original model. The developed model simplification framework is sufficiently general to cater for a variety of battery types and a range of applications (Chapter 3).

7.1.2 Model-Based Charging Strategy Evaluation Procedure

An evaluation procedure has been proposed for battery charging strategies utilising simplified models instead of their high-order counterpart. A substantial advantage on computational efficiency was obtained whilst maintaining reasonable accuracy. Furthermore, the proposed model-based evaluation procedure is generic so that it is capable of SOH prediction when other battery ageing models are considered (Chapter 4).

7.1.3 Multi-Time-Scale Estimation Algorithm and SOC/SOH Estimator

A novel multi-time-scale estimation algorithm was proposed for a class of singularly perturbed systems. It has been proven that under certain assumptions, an appropriately designed estimator is able to compensate for the modelling errors brought about by time-scale separation of the battery coupled dynamics. This proposed algorithm has been applied to a Li-ion battery for the SOC and SOH estimation. Based on the low order physics-based battery models, the capability of this algorithm has been demonstrated using both EKF and UKF. Simulation results have shown that the designed observers are effective in both the fast and slow state estimation. This avoids potential issues in state estimation for singularly perturbed systems including intensive computational requirement, ill-conditioned gain and convergence problem (Chapter 5).

7.1.4 MPC-Based Battery Charging Strategy

An MPC-based charging strategy has been proposed for a Li-ion battery to balance two competing objectives, i.e. battery lifetime and charging time. Specifically, an optimal
charging control problem has been formulated for SOC and SOH reference tracking in the framework of LTV-MPC, for the first time. The model adopted from Chapter 3 was used to predict battery dynamics. The operating constraints imposed on system input and states were explicitly handled. Under the proposed algorithm, the users can flexibly specify the balance between charging time and battery lifetime. The capability of the proposed approach has been demonstrated by incorporating a multi-time-scale estimation algorithm in high fidelity simulations. Significant improvement was observed in terms of the charging time and the state of health preservation relative to the industry standard charging algorithms (Chapter 6).

7.2 Future Work

In this section, potential future research opportunities stemmed from the contributions of this thesis are discussed.

7.2.1 Stability Analysis for Battery Models

In Chapter 3, Assumptions 3.3 and 3.5 in relation to the stability of the boundary layer battery models have been justified via numerical simulations. As remarked, there is no established theory to address the stability properties for such coupled nonlinear PDE systems. For specific classes of PDE systems, Christofides [173] has presented some results in singular perturbation approaches for model simplifications. It may be worth exploring its extension or generalisation and potential applications to the battery systems. The development of formal tools for analytical justification of these assumptions will enable the model simplification framework to be rigorous, and thus enhances the theoretical basis of this work and similar applications.

7.2.2 Stability & Robustness of Charging Strategy Control

In the proposed MPC-based battery charging algorithm, the stability has not been rigorously characterised. This yields research opportunities for the future work. In alternative
MPC implementations, such as [31], the terminal set was replaced with point constraint in order to demonstrate provable asymptotic stability. However, in the SOC and SOH tracking problem, the pre-defined references are likely to be unreachable such that the point terminal constraint may not be reasonably imposed. In this regard, the dynamic trajectory planning and tracking MPC have been united in [168, 174] to guarantee that the closed-loop system asymptotically converges to the optimal reachable periodic trajectory. This approach can be probably modified for battery controls where the trajectories may be nonperiodic.

Additionally, in this thesis, the robustness has not been explicitly addressed but is avoided by softening the state constraints. For some applications where hard constraints have to be imposed for safety, robust MPC techniques will be required to deal with the model-plant mismatches, estimation errors, and noises. A variety of robustness techniques exist such as open or closed loop min-max MPC [175] and constraint tightening [176]. These approaches may be applicable to battery control problems.

### 7.2.3 Multi-Time-Scale Estimation Algorithm Extension

The multi-time-scale estimation algorithm in Chapter 5 assumed that a deterministic EKF is utilised. As illustrated in Figs. 5.7-5.9, the estimation algorithm with a UKF alternative can offer better performance in the convergence, albeit this came at increased computation cost. This reveals the potential to relax the applied assumption for a more general multi-time-scale estimator with a wide class of estimation algorithms covered. To fulfil this objective, the method described in [132], where only the slow states of singularly perturbed systems were of interest and estimated, can be possibly extended for state estimation in both the slow and fast time-scales.

Aside from this, some of the battery parameters, such as diffusion coefficients and reaction rate constants, may change with the age of the battery in practice [80, 177]. For accurate SOC and SOH estimation over the lifetime of a battery, it is important to investigate these characteristics. Possible approaches for this problem include joint estimator designs for both the parameters and states.
7.2 Future Work

7.2.4 Development of High-fidelity SOH Models

The SOH battery model used in this thesis was originally derived from a dominant ageing mechanism, namely a resistive film is generated and consumes cyclable lithium during battery operations [8]. This model assumes an irreversible solvent reduction reaction at the anode solid-electrolyte interface (SEI) during the charging process only. However, in addition to SEI film, various other factors contribute to battery degradation such as lithium plating, electrolyte decomposition and particle fracture. The identification and quantification of all ageing mechanisms are still an ongoing work. Developing an accurate yet general battery ageing model would be important for applications including accurate lifetime prediction and optimal fast charging control. In addition to the physics-based approaches, experimental data-driven models may also be useful. Some work has been conducted in [52, 178, 179], where the impacting factors of battery ageing such as temperature, charging rate and charging depth are sequentially fixed at constant values, allowing the investigation of the effect of these factors individually. Once a more accurate SOH model is available, it can be fitted into the proposed methodologies of battery state estimation and optimal charging control.

7.2.5 Battery Model Parameter Identification

Identification of the parameters of battery models is a hot research topic, which allows real world validation and implementation of the developed algorithms. This main challenge for identification of physics-based battery models, particularly the initial PDE model, attributes to the lacking of identifiability in some parameters [180, 181]. Therefore, sensitivity analysis over a set of operating conditions may be desired for identification of parameters from nonlinear battery models. Fisher information has been demonstrated as an effective tool to derive and rank parameter sensitivities with excellent examples in [182, 183], and may be applicable to the models presented in Chapter 3. With the knowledge of sensitivity of each parameter, identification may be performed, and possible approaches include gradient methods and global optimisation algorithms such as
genetic algorithm (GA) and particle swarm optimisation (PSO) [184].

### 7.2.6 Experimental Validation

The developed algorithms and techniques in this thesis were validated by means of simulations. The simulation models enable viewing of the internal battery states, which is not possible with real cells though. This forms a critical part of the analysis. However, it is agreed that experiments are the only real validation of any proposed approach and thus should be pursued in the future. Upon successful validation of simulation results presented in this work, the SOC/SOH estimator and fast charging strategy can be implemented as a part of battery management systems.

### 7.2.7 Extension of The Proposed Algorithms to Battery Packs

This project focuses on modelling and model-based applications of an individual cell. However, electric vehicles, energy storage systems in smart grids and similar devices rely on battery packs to achieve the required voltage and power. The cells in a pack or between packs can be different in state-of-charge, temperature distribution, operating conditions, and/or chemistry components. In this circumstance, prior to utilisation of our developed techniques, the development of cell balance systems, using approaches such as [12, 13], would be desirable. Alternatively, the centralised algorithms may be fused into some distributed estimation and control structures, e.g. [185], and realise the management of battery packs.


[49] S. Anwar, C. Zou, and C. Manzie, “Distributed thermal-electrochemical modeling of a lithium-ion battery to study the effect of high charging rates,” in *IFAC World Congress*, 2014, pp. 6258–6263.


[85] X. Hu, S. Li, H. Peng, and F. Sun, “Charging time and loss optimization for linmc
and LiFePO$_4$ batteries based on equivalent circuit models,” *J. Power Sources*, vol. 239, pp. 449–457, 2013.


[87] H. He, R. Xiong, H. Guo, and S. Li, “Comparison study on the battery models used for the energy management of batteries in electric vehicles,” *Energy Conversion and Management*, vol. 64, no. 4, pp. 113–121, 2012.


[94] J. Xu, C. C. Mi, B. Cao, and J. Cao, “A new method to estimate the state of charge of lithium-ion batteries based on the battery impedance model,” *J. power sources*, vol. 233, pp. 277–284, 2013.


[133] P. Liu, J. Wang, J. Hicks-Garner, E. Sherman, S. Soukiazian, M. Verbrugge, H. Tataria, J. Musser, and P. Finamore, “Aging mechanisms of LiFePO₄ batteries


