# Chapter 1

# Introduction

The matter of sustainable and local emission-free future mobility and industry is strongly present in politics and society and represents one of humankind's most life-threatening challenges. Climate change is a global problem causing the polar ice caps to melt and the sea level to rise. Some areas are experiencing more frequent extreme weather events, such as heavy rainfall, while others are facing longer and more intense heat waves and droughts. The impacts of climate change are wide-ranging and can affect many aspects of our lives. It is a severe threat that needs to be addressed immediately [40, 211]. Human-caused emissions of greenhouse gases are the main contributor to climate change that has occurred since the mid-20$^{\text{th}}$ century [198, 38].

According to Eurostat, the EU's statistical office, the European Union (EU) announced that it was possible to decrease greenhouse gas emissions in various sectors by over 1.5 billion tons of carbon dioxide ($CO_2$) between 1990 and 2020, a decrease of 32%. In stark contrast, emissions from the EU's transportation sector increased by 50 million tons of $CO_2$ in 2020, a 7% increase compared to 1990 [212]. The European Environment Agency's analysis, published on June 1, 2022, showed that the growth in transport volume had caused road transport emissions in Europe to rise over the past two decades. The report revealed that total greenhouse gas emissions from passenger cars and heavy goods vehicles have increased in Europe, despite improvements in combustion engine efficiency and the use of biofuels [6]. In 2018, around 888 million tons of $CO_2$ were released into the atmosphere due to burning fuel for road transportation in the EU. Passenger cars and motorcycles contributed the most with 62% of the total emissions, followed by heavy-duty trucks and buses with 26% and light-duty trucks with 13% [175].

The EU climate law envisions Europe becoming climate-neutral by 2050 and reducing greenhouse gas emissions by at least 55 percent by 2030 compared to 1990 [62]. Electromobility plays the most decisive role in achieving these ambitious climate targets. In this regard, at least 30 million zero-emission electric vehicles are expected to be on EU roads by 2030 [148]. Starting from 2035, all new cars and vans sold in the EU should emit no greenhouse gases [61]. Thus, mobility has to be rethought and reinvented. In the context of this shift, the future automotive sector places significant emphasis on the advancement of energy storage systems, with a particular focus on the development of batteries.

Because of their high energy and power densities, Lithium-Ion-Batteries (LIBs) are essential for developing and operating novel Electric-Vehicles (EVs) [92]. LIBs are distinguished for their comparatively high performance, cyclic stability, relatively low cost and high safety [135]. In addition to advancements in battery cell technology, the Battery-Management-System (BMS) plays a prominent role in EV's battery systems: Not only does it monitor the battery packs used in mobile applications, but it also assures their reliability and optimal performance [52]. The BMS's key objectives for EVs are to increase battery systems' safety and reliability, protect individual cells from damage, improve energy usage efficiency and prolong battery lifetime [85]. Recently, the European Commission has proposed new EU battery regulations to promote sustainability and competitiveness within the battery industry. The proposal includes mandatory requirements for sustainability, safety and labeling for batteries, as well as requirements for managing the End-of-Life (EOL) conditions of batteries. EV batteries' BMS should gather informations about the battery's State-of-Health (SOH) and expected Remaining-Useful-Life (RUL) which should be accessible to the battery owners and independent operators [147].

Thus, in this context of electrifying the future mobility, the significance of onboard battery management and battery state estimation which are the core components of this study becomes notably high.

## 1.1 Motivation and Goals of the Thesis

Accurate SOH estimation of LIBs is crucial for EVs' reliable and safe operation since it influences the steering of the cells' operation limits. Lithium-ion cells degrade over time due to chemical changes resulting from cycling, overcharging and overheating. The manifestation of this degradation can be measured by the cell's SOH which reflects the current condition of a battery compared to its original design specifications at Begin-of-Life (BOL). Based on the SOH estimation, the battery can be operated properly without exceeding the lifetime and safety limits related, e.g., to thermal and plating current limits, voltage and pressure limits caused by cell expansion. Furthermore, the State-of-Charge (SOC) estimation depends on the actual cell capacity, thus, its SOH estimation. As such, accurate estimation of the battery's SOH is crucial to improve its performance, extend its lifespan, and consequently enable the sustainable application of EVs. As a matter of fact, SOH is known to have multiple dimensions that lead to cell individual electrical behavior and degradation. The primary cell degradation mechanisms include the formation, thickness growth and decomposition of Solid-Electrolyte-Interface (SEI) and Cathode-Electrolyte-Interface (CEI). Also to be mentioned are metallic lithium plating on the graphite anode, electrolyte depletion and reaction, active material particle cracking, loss of electrical contact and corrosion of current collectors [60]. For the purpose of battery management and online diagnosis, the degradation mechanisms of a battery can be summarized as either a **Loss-of-Lithium-Inventory (LLI)**, a **Loss-of-Active-Material-at-the-Anode (LAM$_n$)** or a **Loss-of-Active-Material-at-the-Cathode (LAM$_p$)**. The mentioned degradation modes are typically manifested by changes in the battery's electrical performance, particularly in capacity and power fade [83]. A major challenge in the aging estimation of lithium-ion cells is the **inhomogeneous** distribution of cell degradation which represents a leading factor that limits LIBs performance [143]. Therefore, Estimation algorithms that can quantify the inhomogeneity and accurately estimate the SOH even in the presence of inhomogeneous aging are gaining significant importance.

In point of fact, previously developed algorithms for estimating the SOH of batteries in EVs suitable for use in industry and series production are mostly empirical or based on evaluating fully relaxed cell states. These mostly empirical algorithms, such as event-based evaluation of pulses during operation to determine the internal resistance [90], lack a physical model of the battery cells and can be challenging to interpret and parametrize. Estimation methods that rely heavily on relaxed states like capacitance estimation with linear regression [169] have the disadvantage of requiring multiple load cycles followed by long periods of relaxation, making them slow and partially impracticable in automotive applications. These methods may also be unreliable in specific EV applications where the batteries do not have the opportunity to fully relax, such as in taxi or ride-sharing situations. Furthermore, there is still a lack of on-board algorithms for quantifying aging inhomogeneity in lithium-ion cells.

This thesis aims to propose novel algorithms for battery aging estimation that are suitable for on-board applications in an attempt to alleviate the aforementioned problems. The first focus lies in increasing the reliability of cell state estimation methods by introducing an embedded cell model to provide a physically motivated estimation of the cell's internal resistance. The model-based estimation of the cell's internal resistance is supposed to fill the requirements of faster, more accurate and more robust estimation compared with empirical and event-based approaches. Furthermore, developing capacity estimation algorithms based on observing single charging operations and Differential-Voltage-Analysis (DVA) techniques is an integral part of this work. The aim is to enable an accurate estimation of the cell capacities quickly and without the time-consuming consideration of several relaxation phases. A further focus of this thesis lies on increasing the sustainability of electromobility via introducing on-board estimation algorithms, enabling deeper multidimensional insight into the chemical cell degradation. The multidimensional aging estimation considering the cell's LLI, $LAM_n$ and $LAM_p$ can be used for predictive battery operation via early detection of accelerated aging and degradation. Meanwhile, the battery operation strategy can be adaptively adjusted by restricting the cell's current and voltage operation windows to slow battery aging. The multidimensional aging estimation also has great significance for the battery replacement criterion. Extensive multidimensional estimation beyond State-of-Health-Capacity (SOHC) and State-of-Health-Resistance (SOHR) allows fully exploiting the battery lifetime and preventing premature battery replacement with large failure reserves. Thus, the degree of sustainability in electromobility is increased.

## 1.2 Contributions

To provide a clear and organized overview of the contributions made in this thesis, the published scientific articles produced by the candidate as the main contributing author are listed below. Content from these publications is used for this dissertation. Text elements, figures, pictures and tables from these publications are not explicitly cited within the thesis, as far as the author of this work created them himself. These articles include:

### Peer-Reviewed Journal Publications (2)

1. Y. Bensaad, F. Friedrichs, J. Sieg, J. Bähr, A. Fill, K.P. Birke "Multidimensional Estimation of Inhomogeneous Lithium-Ion Cell Aging via Modal Differential Voltage Analysis" Journal of Energy Storage, 2023. DOI: 10.1016/j.est.2023.107108

2. Y. Bensaad, F. Friedrichs, T. Baumhöfer, M. Eswein, J. Bähr, A. Fill, K.P. Birke "Embedded Real-Time Fractional-Order Equivalent Circuit Model for Internal Resistance Estimation of Lithium-Ion Cells" Journal of Energy Storage, 2023. DOI: 10.1016/j.est.2023.107516

## Peer-Reviewed Conference Proceedings (1)

1. Y. Bensaad, F. Friedrichs, T. Baumhöfer, J. Bähr, A. Fill, K.P. Birke "High-Precision On-Board Capacity Estimation of Lithium-Ion Cells using a Fractional-Order Cell Model and Singular Value Decomposition" The 2023 IEEE Vehicle Power and Propulsion (IEEE VPPC 2023).

Furthermore, the following patents have also been developed in the scope of this work and are pending to the **German Patent and Trade Mark Office**:

## Patents (3)

1. Y. Bensaad, F. Friedrichs, T. Baumhöfer, J. Bähr "Verfahren zur modellbasierten Abschätzung der Impedanz einer galvanischen Zelle einer Sekundärbatterie und dessen Verwendung sowie Batteriezellenüberwachungsvorrichtung und Fahrzeug" DE Application No. 102022004803.5

2. Y. Bensaad, F. Friedrichs, J. Sieg, J. Bähr "Verfahren zur Schätzung eines Gesundheitszustands einer Batteriezelle eines elektrischen Energiespeichers, Computerprogrammprodukt sowie elektronische Recheneinrichtung" DE Application No. 102022002866.2

3. Y. Bensaad, F. Friedrichs, J. Bähr "Verfahren zum Schätzen eines aktuellen Ladezustands zumindest einer Batteriezelle eines elektrischen Energiespeichers, Computerprogrammprodukt sowie Batteriemanagementsystem" DE Application No. 102023000898.2

In the following, an overview of the structure of this thesis is presented. **Chapter 2** gives a detailed literature review of the fundamental principles of lithium-ion cell technology. The theoretical foundation discusses the principles behind the design and functionality of lithium-ion cells. Questions regarding cell degradation mechanisms over aging and how they can be characterized are discussed, and state-of-the-art battery modeling and SOH estimation techniques are presented.

The full aging estimation algorithm described in this thesis is built according to Fig. 1.1 based on two basic parts which include the electrical cell model described in Chapter 3 and the DVA described in Chapter 4. The final result consists of merging both parts, as described in Chapter 5.

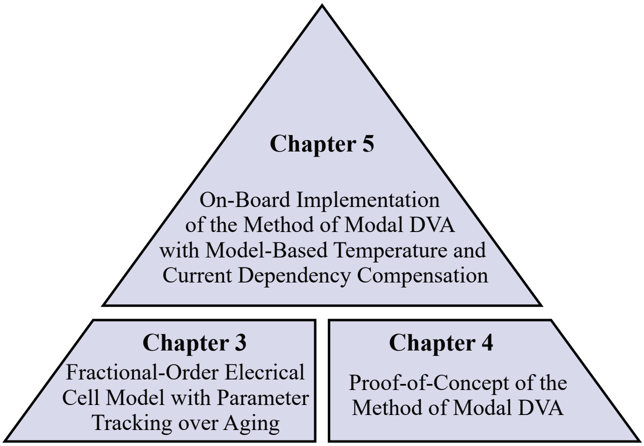


FIGURE 1.1: Schematic representation of the thematic structure of the thesis.

**Chapter 3** introduces a new on-board capable method for model-based estimation of the internal resistance of lithium-ion cells. The method relates to a fractional-order electrical **Equivalent-Circuit-Model (ECM)** consisting of an ohmic resistance, ZARC and Warburg elements. The mentioned ZARC elements are crucial for accurately modeling the behavior of systems that exhibit non-ideal capacitive behavior and will be introduced in Sec. 2.3.1. A parametrization algorithm is presented based on a BOL model parameterization using Electrochemical-Impedance-Spectroscopy (EIS), time domain measurements with aged cells and a nonlinear optimization routine. The on-board internal resistance estimation approach using filtering algorithms and feedback control during dynamical driving is discussed. Furthermore, this section outlines the embedded real-time implementation for a state-of-the-art BMS, combining float- and fixed-point arithmetic. Finally, the validation tests to assess the convergence and error of the SOHR estimation are addressed.

**Chapter 4** presents a novel method for **Differential-Voltage-Analysis (DVA)** inspired by data-driven **modal reduction** to quantify the cell's multidimensional aging and degradation inhomogeneity. The proposed algorithm encrypts offline findings from measured Differential-Voltage (DV) curves and electrochemical simulations in low-rank patterns, suitable for the on-board application. The parametrization and validation of the algorithm are performed by exploiting an already existing dataset of eight automotive pouch cells with graphite anode and

NMC cathode that were cycled for accelerated aging with two different load profiles, yielding homogeneous and inhomogeneous cell aging. The multidimensional aging estimation presented in this section includes the cell's SOHC, LLI, $LAM_n$ and $LAM_p$. Furthermore, the estimation of cell aging inhomogeneity using a pattern recognition approach is introduced.

**Chapter 5** combines the electrical ECM described in Chapter 3 and the DVA method in Chapter 4 to give a robust SOHC estimator based on recursive estimation techniques and observing the Alternating-Current (AC) charging behavior. This Chapter looks at the applicability of the presented **DVA method under real-life conditions** with a state-of-the-art EV. The process of data acquisition using in-vehicle data communication protocols with the EV in a climate chamber is described. Furthermore, the validation results of the whole algorithm are presented. First, the temperature dependency of the DV curves and the extent to which it can be normalized using the cell model are presented. Then, the SOHC estimation accuracy is assessed for training and testing data, different temperatures and charging start and end SOCs.

In summary, the overall algorithm provides two aging estimates that complement each other and take into account the reduction in range as well as the efficiency of an EV. These are the SOHC and the SOHR as shown in Fig. 1.2.

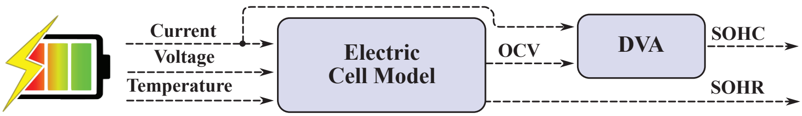


FIGURE 1.2: Schematic representation of the full algorithm presented in the thesis.

# Chapter 2

# Theoretical Foundation

This Chapter introduces the theoretical basis for this thesis by discussing the fundamental design principles, functionality and degradation of lithium-ion cells. The basic functionality and packaging shapes of these cells are first presented. The mechanisms of cell degradation during aging are then discussed in depth. In addition, cell modeling and characterization in the time and frequency domains are addressed. Finally, state-of-the-art approaches for estimating cells' SOH are outlined.

## 2.1 Lithium-Ion Cell Fundamentals

Lithium-ion cells are a common type of secondary cells. The first market-ready lithium-ion cell was introduced by Sony Energytec Inc. in 1991 and quickly became popular due to its high energy density which made it suitable for portable devices such as smartphones, tablets and laptops [107]. In light of electric mobility, lithium-ion cells are considered to be the key technology for sufficiently long driving ranges with EVs and, thus, the path toward local emission-free mobility.

### 2.1.1 Operational Principles and Functionality

A lithium-ion cell, like other galvanic cells, consists of an **electrolyte**, a **separator** and two electrodes (**anode** and **cathode**) [86]. The purpose of the battery cell is to transform chemical energy into electricity through oxidation and reduction reactions which occur simultaneously but at different electrodes. When the redox reaction cannot be reversed, it is called a primary cell, and when it can be reversed, it is referred to as a secondary cell. Fig. 2.1 shows the operating principle of a lithium-ion cell during the discharging process. During discharging, the positively charged lithium-ions $Li^{+}$ are deintercalated from the negative electrode's active material, called the anode, by releasing electrons. The lithium-ions dissolved in the electrolyte migrate through the separator to the cathode. Then, they are intercalated into the active material of the cathode. The intercalation consists of the detachment of the solvate membrane, the penetration into the active material and the acquisition of an electron from the host structure. Finally, the lithium atoms must be transported from the surface into the material to balance the lithium concentration gradients within the electrode. Since the porous separator between the electrode layers is electrically insulating and allows only the solvated lithium-ions to pass through, the released electrons are transported via the external electrical circuit. Thus, an electrical load introduced in the external circuit can convert the supplied electrical energy from the battery cell into other forms of energy. During charging, the movement of lithium-ions is reversed, and they move from the cathode through the electrolyte and separator to the anode [146, 107]. In the following, the components of a cell are presented in more detail.

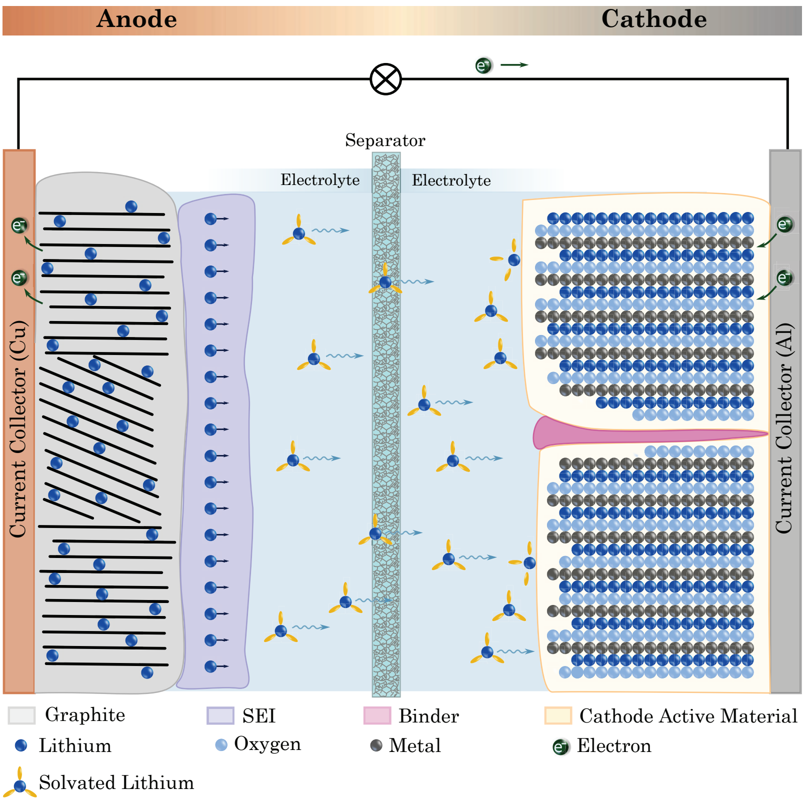


FIGURE 2.1: Schematic representation of the function of a lithium-ion cell based on the example of a discharge process (simplified abstraction).