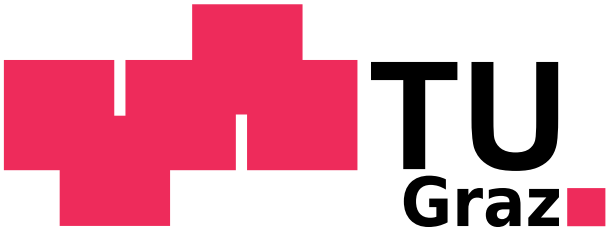

Institute of Materials Physics of the Graz University of Technology:

State-of-the-art lithium-ion cell lifetime modelling approaches

and their applicability in the automotive industry

a Bachelor's Thesis by

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Abstract

Lithium-ion batteries are a very modern power source and their behaviour is very complex, giving rise to a large variety of approaches to modelling their lifetime. Because the technology is so new, it is not yet clear which modelling approaches are suited to which use case. This thesis presents a summary of state-of-the-art modelling approaches and results from research of the past years, and discusses the applicability of individual approaches to various use cases collected from actual automotive clients and people active in the automotive industry, ultimately presenting the recommendations in the form of an easy-to-follow decision tree. This type of work may be very useful for making first decisions when consulting clients on what modelling approaches would best solve their problem.

1 Introduction

Lithium-ion cells are a very popular rechargeable power source, found in widespread applications from handheld electronic devices to battery-powered electric vehicles (EVs). This popularity is founded in its high energy and power densities (as visible in the Ragone plot in Figure 1), and a charge-discharge mechanism that conserves the active materials involved, making them very useful secondary power sources.

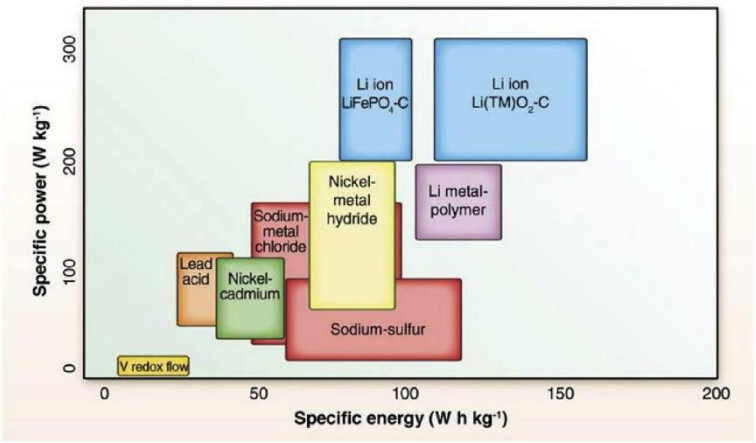

Figure 1 Ragone plot describing specific power and energy for different secondary cells [1]

Lithium-ion cells are constructed in a complex manner, which gives rise to a large amount of aging mechanisms in all parts of the cell [2, 3]. These mechanisms affect the remaining useful lifetime (RUL) of the cell and are dependent on many different factors, such as the ambient and internal cell temperature, the chemical and mechanical properties of the cell's materials, and the operating conditions it experiences during use.

These mechanisms are very complex and lithium-ion batteries are the single most expensive component of battery-powered electric vehicles (making up about a third of total electric vehicle costs [4]). Therefore, over the past 30+ years, research into cell lifetime modelling has been a very active field, especially with a focus on EV cells. In the automotive industry, though, it can be unclear which models are suited to which use case, as these differ with respect to the amount of data and resources available to model users, the stage in the battery's life, the amount of time available for testing, etc.

The aim of this thesis is therefore to establish an overview of the state of the art in battery lifetime modelling, as well as to discuss the applicability of the different models to use cases relevant for members of the automobile industry.

It will achieve this aim by first giving the reader an insight into the composition of commonly used lithium-ion cells in chapter 2, which will also list advantages and disadvantages of various materials.

Behaviours that accelerate aging will be a large focus in this chapter. Chapter 3 will then provide a similar overview of models currently in use and in development, addressing their strengths and limitations. Chapter 4 will then list categories of use cases confronting the automotive industry, discuss the model approaches best suited to them, and then conclude in a use case decision tree that can be used to decide on a suitable model based on the requirements of a user.

2 The lithium-ion cell

The lithium-ion cell consists essentially of six different components: the negative and positive electrodes (referred to henceforth as the anode and cathode, respectively), the electrolyte, the separator, the current collectors, and the cell casing.

While all these materials may undergo aging effects, the electrodes, the separator and the electrolyte have the largest variety of material options which all have their own respective advantages and disadvantages that strongly affect aging mechanisms. They will therefore be discussed in more detail in the following chapter.

The following quantities will be used in describing and comparing electrode materials:

- **Electrode potential** (given in V vs Li/Li^+) describes the average electrode potential measured relative to a pure lithium electrode. The difference between cathode and anode potential gives the cell's open circuit voltage.
- **Specific capacity** (given in mAh/g), which describes the total charge stored in the electrode per unit mass. The lower the discharging current, the slower this capacity is used up.
- **Specific energy** (given in mWh/g), in this thesis calculated by multiplying specific capacity with the electrode potential. The energy, of course, varies from electrode pair to electrode pair.

Another quantity which is important in general when discussing batteries and charge/discharge speeds is the C-rate. If a battery is discharged from a state of full charge at a C-rate of 1C, it means that, regardless of the battery capacity, it should be fully discharged within one hour; if it is discharged at a C-rate of 2C, it should be fully discharged within half an hour. A 15 Ah battery 1C C-rate corresponds to a current of 15 A [5].

2.1 The anode

The most commonly used anode material nowadays is graphite [2], although early research used pure lithium foil anodes, and alternative materials such as lithium titanium oxide (LTO) or silicon are finding use in laboratories as well as, in the case of LTO, in some commercial lithium-ion cells.

Graphite has a very low reduction potential (ranging around 0-0.25 V vs Li/Li^+), which is electrically favourable for an anode, and it has a reasonable specific capacity of 372 mAh/g [6]. During the first few cycles of a cell's lifetime, a passivation layer called the solid-electrolyte interphase (SEI) forms on the anode particles' surface, which increases the cell's internal resistance, decreases the amount of available lithium and causes a rapid decrease in capacity characteristic for the first few cycles of a cell's lifetime

[2]. The fact that graphite particles' volume changes by 13% over the course of a charge-discharge cycle amplifies this, as cracks appear in the SEI layer in which new SEI then forms [2]. SEI layer growth is described by Yuan et al. as one of the four major aging mechanisms in lithium-ion cells [7].

In addition, the graphite's low reduction potential can actually give rise to another problem: under specific conditions such as cooler temperatures and high charging C-rates, the intercalation rate of lithium into the anode is not fast enough to allow all the lithium atoms to neatly fill in, and they instead crowd at the anode surface [6]. Through this effect, lithium plates onto the anode, an effect which can cause dendrites to form as the electric field is stronger at peaks of a material and the lithium is therefore more likely to plate at peaks. These dendrites use up the available lithium in the cell, rapidly reducing its capacity, and can grow long enough that they penetrate the separator, leading to internal short-circuiting and destruction of the cell [2, 6]. Dendrite growth is another of the four major aging mechanisms described by Yuan et al [7].

Silicon has an average anode potential between about 0.3 and 0.5 V [8] and an extraordinarily high theoretical specific capacity of 4212 mAh/g [9], which is very favourable especially in the automotive industry as a smaller amount of anode mass is needed to supply a necessary current. However, it shows volume changes of up to 300-400% during one cycle [2]. As silicon anodes also form SEI layers, this layer cracks during these volume change phases, causing continuous new formation of SEI which therefore continually uses up lithium from the anodes and the electrolyte [2, 9]. This effect is also stronger than it is in graphite anodes because of the more extreme volume changes.

Silicon-carbon-blend (Si-C) anodes have been researched as well. Li et al. have described an anode of Si-C nanoparticles with 84.1 mass percent silicon, which showed a high capacity of 2331 mAh/g at very low charging rates and about 1605 mAh/g at the paper's standard charging rate of 1 A/g, along with better cyclability than regular silicon anodes [10]. However, Si-C anodes still have a volume change problem [2, 10].

LTO's reduction potential at 1.5 V lies higher than that of both silicon-carbon blends and graphite, which is unfavourable as the total battery output voltage is lower, and with 175 mAh/g it also shows a far lower specific capacity than all previously mentioned materials. However, it shows significantly better performance with regards to safety. As its electrode potential is within the electrolyte stability window explained in subchapter 2.3, the amount of SEI this anode forms is much less compared with the other mentioned materials. Its volume also remains virtually unchanged during cycling, meaning that if it forms an SEI, it stays unfractured. This spares the lithium in the cell and allows for a longer lifetime [2].

Pure lithium foil has an extremely high theoretical specific capacity (3860 mAh/g [11]) and specific energy. However, it is no longer used in combination with liquid electrolytes as it is very liable to build dendrites, whose risks were already described earlier. However, solid-state batteries, which have solid electrolytes [11], have the ability to suppress dendrite formation, meaning that pure lithium anodes can potentially be used more widely in the future.

The most important properties of the anode materials mentioned in this chapter are summarized in Table 1.

Table 1 Summary of anode materials with their characteristics, including theoretical capacity values, taken from [6][8][9].

Material	**Electrode potential (vs Li/Li$^+$) / V**	**Specific capacity / mAh/g**	**Safety [9]**
Graphite	0-0.25	372	★★★☆☆☆☆
Silicon	0.3-0.5	4212	★☆☆☆☆☆☆
Si-C	n.a.	1605	★★☆☆☆☆☆
LTO	1.5	175	★★★★★★★
Lithium foil	0	3860	☆☆☆☆☆☆☆

2.2 The cathode

The materials most widely used for EV cathodes as of 2020 were lithium nickel-manganese-cobalt oxides (NMC), whose precedents lithium manganese oxide (LMO) and lithium cobalt oxide (LCO) will also be discussed, and lithium nickel-cobalt-aluminium oxides (NCA). In addition, lithium iron phosphate (LFP) has a market presence due to its favourable stability and cost features [12]. These materials will be presented further in this subchapter.

LMO is relatively safe, low-cost, and non-toxic, and has a high reduction potential of 4.1 V vs Li/Li$^+$. However, manganese is liable to dissolving in the electrolyte and no longer being able to take up lithium, meaning that active material is lost, accelerated by the fact that LMO cathodes experience a 16% volume change during charge-discharge cycles. The dissolved manganese may also move through the separator and adhere to graphite anodes, increasing SEI film growth [2, 9]. In addition, its practical specific capacity of 140 mAh/g [8] and energy density of 410 mWh/g [9] are both relatively low.

LCO, on the other hand, has a similarly high reduction potential of 3.9 V vs Li/Li$^+$ as well as a better practical specific capacity of 148 mAh/g [8]. It also has a very high energy density at 624 mWh/g [9], making it very suitable for storing energy. However, there are several downsides: LCO is not stable in a state of low lithiation, meaning that only about half of the theoretically available lithium in the cathode can practically be used (resulting in the practical capacity value stated above). If the cell is charged beyond the stable SOC limit, the LCO cathode deteriorates rapidly. Additionally, oxygen can be released into the cell and react with other elements, leading to explosion. Cobalt is also highly expensive, very rare, and very toxic [9].

In NMC, the benefits of cobalt and manganese are combined, with the addition of nickel increasing the usable capacity. Cathodes of this material are the current state of the art, making up 79% of EV cathodes [12]. While the specific energy and capacity as well as the reduction potential vary depending on the

relative proportions of the different metals, they generally offer very high energy densities of around 600 mWh/g, high capacity densities of around 160 mAh/g and a reduction potential of around 3.7 V vs Li/Li^+ [9]. However, safety issues still arise from the presence of cobalt and manganese, giving the cell relatively poor cyclability compared to, for example, LFP [2]. It is also only stable up to a certain level of delithiation and degrades rapidly if the cell is charged beyond this level, and the cathodes' nickel content makes them unstable at higher temperatures [2].

NMC's oxidization issues can partially be solved by doping nickel-cobalt cathodes with aluminium resulting in NCA, as aluminium does not dissolve as easily as manganese. These cells have a market share of about 19% and have even higher energy densities and capacity densities than NMC at over 700 mWh/g and around 200 mAh/g, respectively. Their operating voltage is the same as that of NMC at about 3.7 V [9, 12]. Their cyclability, however, is not strongly improved [6].

LFP, on the other hand, has a capacity of 160 mAh/g, an energy density of 410 mWh/g, and its reduction potential is at 3.4 V vs Li/Li^+ [9]. While the energy and operating voltage are unfavourable compared to the various NMC and NCA combinations (accounting for its low EV market share at around 2% [12]), its charge and discharge voltage are both very stable, it is much less toxic due to being cobalt-free, and its material safety is much better; while LFP cathodes also contain oxygen, they are much less likely to release that oxygen due to the strong covalent bonds keeping it contained, which means that LFP as a cathode material is much less likely to cause burns or explosions than lithium metal oxides. In addition, its material costs are much lower than those of NMC/NCA cathodes, a factor which shouldn't be underestimated, as cathode active materials are the most expensive part of a battery cell (making up an estimated 34% of cell costs) [13].

The most important properties of the cathode materials mentioned in this chapter are summarized in Table 2.

Table 2 Summary of cathode materials with their characteristics, including practical capacity values, taken from [6][8][9].

Material	**Electrode potential (vs Li/Li^+) / V**	**Specific capacity / mAh/g**	**Specific energy / mWh/g**	**Safety [9]**
LMO	4.1	140	410	★★★☆
LCO	3.9	148	624	n.a.
NMC	3.7	160	600	★★☆☆
NCA	3.7	200	700	★☆☆☆
LFP	3.4	160	410	★★★★

2.3 The electrolyte

Liquid electrolytes, which are virtually the only ones in use nowadays, consist of a solvent, conducting salts, and potentially additives.

The solvent usually contains a blend of various liquid materials combined to produce a material with favourable characteristics. Most commonly, the blend includes organic diesters, which have a high boiling point and high permittivity; esters, which have a low melting point; and ethers, which have low viscosity. These ensure that the electrolyte is liquid during most battery usage scenarios, allowing for quick material transport.

The fact that the electrolyte consists in large part of organic compounds limits its electrochemical stability window to voltages between 1.0 and 4.5 V [2].

The most common conducting salt is lithium hexafluorophosphate ($LiPF_6$). It is highly soluble, highly conductive, and electrochemically stable. However, it is sensitive towards hydrolysis as higher operating temperatures and easily reacts with trace amounts of water to produce hydrofluoric acid, which further speeds up corrosion and is a highly dangerous substance [9].

This is countered by developing alternative lithium salts based on fluorosulfonyl anions, such as LiFSI and LiTFSI. These have the added advantage of thermal stability and resistance towards hydrolysis, but they are costly to produce.

LiBOB, based on lithium, boron, and oxygen, is an additional conductive salt which is more environmentally friendly than the aforementioned salts, and is thermally stable. However, it is a poor conductor and can develop gas during cell formation, which can be potentially dangerous [9].

In addition, various carbonate additives are being researched as potential SEI stabilizers for silicon-based anodes, such as fluoroethylene carbonate and vinylene carbonate [9, 14].

It should be noted that while liquid electrolytes make up the absolute majority, solid electrolytes have been in research for the past decade and are beginning to see experimental in-traffic use with moderate success [15].

2.4 The separator

Separators should principally allow ionic conduction while being electrically non-conductive, and they should be highly porous with very fine pores to suppress dendrite development, while also being thin enough to not compromise the cell's energy density. They should also be stable, durable, and resistant to deformation.

The most-used separators are dry and wet membrane separators. Additional experimental efforts are being invested in the development of composite nano-fibrous membranes.

Dry membrane separators usually consist of polyethylene (PE) or polypropylene (PP). They are thin (25-40 μm) and have strong tensile strength (which is preferable for wrapped cells). However, they have low porosity, which is unfavourable as it reduces ion conductivity, and they show high shrinkage in response

to high temperatures, and their melting temperature is restricted to that of PP, which is at around 160 °C [9].

Wet membrane separators are usually constructed out of specific PEs. They are even thinner (<25 µm) with homogeneous small pores and high porosity, meaning that they conduct Li^+ better and resist dendrite growth. However, they also experience high shrinkage and have a lower melting point of 135 °C [9].

So-called shutdown separators, created by inserting a PE layer between two PP layers, are used as they irreversibly shut the cell down in response to very high temperatures (135 °C<). The PE layer melts and clogs up the pores within the PP layer while the latter remains mechanically stable to ensure that the cell doesn't short-circuit. The downside is that this effect can only occur if the cell's temperature increases slowly enough that the PE layer has time to melt before the PP layer's melting point at 160 °C is reached [9].

Research is being conducted on nano-fibrous fleeces, whose benefits would be a lack of shrinkage and high mechanical stability, with no additional production difficulties as separators are already difficult to produce due to demand for high homogeneity in their microstructure [9].

2.5 Summary of main aging mechanisms in lithium-ion cells

In conclusion, lithium-ion cell aging results from many different effects. The stability window of liquid electrolytes induces SEI layer formation on the anode, which uses up available lithium, and is worsened if anode materials undergo cyclical volume changes. Additionally, fast charging and cooler temperatures lead to lithium plating and dendrite formation, which can potentially destroy the cell. Cathode materials based on metal oxides react easily to water within the cells, reducing active cathode material and potentially leading to ruptures in the cell. Nickel-rich cathode materials specifically are unstable at higher temperatures. The conducting salts in the electrolyte easily react with water to further corrode manganese cathodes, Additionally, the common separator materials have mechanical stability problems and present upper thermal limits for lithium-ion cells.

3 Approaches to modelling lithium-ion cell aging

Modelling lithium-ion cell aging behaviour allows cell users to make predictions about battery lifetimes and how much RUL the cell has left. A large amount of research is focused on various approaches to modelling lithium-ion cell aging behaviour. While different methods of categorizing and sorting models are found throughout the literature, the one chosen in this thesis is the widely accepted separation into the following three categories: physics-based models, data-driven models, and semi-empirical models. Models referred to as "online" are, in this thesis, models that are able to make use of data from the vehicle they are currently implemented in. Offline models, on the other hand, are models that are either trained using historical data, or that do not use data at all.

The cell's state of health (SOH) is an abstract concept that cannot be measured. Therefore, the parameter modelled is usually the capacity of the cell, as it directly reflects the SOH of the cell and fades over cell lifetime. However, the internal resistance, which grows over cell lifetime, may also be used [16]. The end of life (EOL) of the cell is then defined to be reached when the capacity sinks to a certain level. The usual capacity threshold is at 80% of the initial capacity in the literature [17], although papers that use the publicly available NASA Ames Prognostic Center of Excellence (PCoE) dataset for training and testing go down to 70%, as this is how far the data were collected [18].

In the following chapter, a summary of lithium-ion cell lifetime modelling approaches along with their historical development is presented, with the subchapters grouping the models into the categories mentioned above.

3.1 Physics-based models

Physics-based models (also called physics-of-failure-based models [19], electrochemical models, or simply the model-based approach [16]) aim to replicate the physical and electrochemical processes occurring within the cell with the help of chemical and electrochemical formulae. They require in-depth knowledge of the cell and its materials but are not dependent on large amounts of data.

The most successful and accurate of these models was described and developed in 1993 by Doyle, Fuller and Newman [20]. It models the cells using five partial differential equations describing charge and mass conservation inside the electrodes and electrolyte as well as the transfer between them.

This model, called the Doyle-Fuller-Newman (DFN) model or the pseudo-two-dimensional (P2D) model, laid the groundwork for future electrochemical models, the majority of which built on improving and optimizing the original DFN model, which, while very accurate, was also extremely computationally costly, requiring up to a minute for a single simulation step [20, 21]. The original model also does not account for irreversible long-term chemical degradation [20].

Another popular model which developed from the P2D model is the single particle model (SPM), which is a simplification of the cell that assumes the electrodes to consist of a single particle rather than many small ones [22]. While it is far simpler, faster, and more versatile, its geometric simplifications have to be fine-tuned in cases where the model deviates strongly from reality. This is due to the fact that SPMs

assume that the reaction current is constant along the electrode surface, which is not the case for cells that, for example, are experiencing high discharge currents or have thick electrodes.

A difficulty arising from the nature of physics-based models is that many different coefficients describing material properties of the cell need to be provided as input. Examples of coefficients required by most physics-based models are the transfer number of Li^+ ions, as well as the transfer coefficients of the anodic and cathodic electrochemical reactions [22–24]. While tests can be performed to determine the various constants, these may be destructive to cells and beyond the capacities of model implementers. Methods of treating this problem are described further in subchapter 3.3.

A different problem with electrochemical models is that researchers have only recently begun to address both the problem of SEI growth and the effects of temperature on electrochemical processes more widely [21]. In recent years, research on these problems has led to the development of SPM models with possible online applications that include transfer function modelling of SEI layer growth [25]. Additionally, it led to the development P2D models including factors modelling SEI growth that also address the effect of temperature on SEI growth and on capacity fade. These, however, don't show online viability yet [26, 27].

The fact that SEI layer growth is only starting to see more widespread research now is a problem considering its significance as an aging mechanism discussed in chapter 2.

Dendrite formation, a phenomenon caused by lithium plating, is another significant aging mechanism which has received specific focus in research. Recent efforts within physics-based models have been invested into modelling the anode potential, which, as presented in chapter 2, is a strong indicator for how much lithium plating is occurring. These efforts have so far seen great success and wider implementation in models [28, 29].

3.2 Data-driven models

Data-driven models (also referred to as empirical models [26]), in contrast, use large amounts of data in combination with statistical methods to model cell behaviour. These require very little to no knowledge of the processes occurring inside the cell.

How data are used to predict capacity fade varies; while some models filter data to fit parameters of linear, polynomial, and exponential functions (with the latter being the most common model [30]), others are nonparametric and adapt their complexity to the amount of available data [31, 32].

A popular and common data-driven group of models are linear autoregressive (AR) models that use root mean square error (RSME) minimization to optimize model parameters. While these are widely used, they have an array of shortcomings. Many modelling approaches are built on optimizing and improving AR models in terms of computational cost and required data.

One aspect of AR models that has been improved upon in the literature is its linearity, which is not completely applicable to the complex lithium-ion cell behaviour. Nonlinear degradation AR models, or ND-AR models, have been developed with nonlinear factors either based on the current cycle number

[33] or the ratio of the current cycle number to the total number of cycles predicted by the AR model [34], both showing improved performance relative to the regular AR model. An additional benefit of the latter is that it provides a prediction of the total number of cycles in a cell's lifetime around the middle of its lifetime (about 50 cycles for LMO batteries and around 280 cycles for LFP batteries) and improves upon this estimate as the cell continues to age [34].

Nonlinear AR models have also been approached from the perspective of artificial neural networks (ANNs) in the form of nonlinear ARs with exogenous input (NARX models), which have been validated for cells with various chemistries [17] and outperformed both a Gaussian Process Regression model (another kind of ANN-based model) and a semi-empirical curve-fitting model in terms of computational efficiency and error relative to data in a study by van Mierlo et al. [35].

Support vector regression (SVR) models, another improvement on AR models in terms of efficiency and applicability to nonlinear trends, evolved from the ANN-based support vector machine. While AR models seek to reduce the error between the model and the data to zero, SVR models reduce it to a value below a defined small error level ε [16, 36], meaning that they are solvable with less computational effort, and can be applied in situations where few data are available [16]. For example, the SVR model used as a comparison model by Zhao et al. [37], used 50% of three different cells' total lifetimes as training data and the rest as testing data with a resulting prediction RSME under 2.5%.

However, both the computation time and the retraining time of an SVR model are long, and they are usually either trained offline and used in that state [16, 37, 38], or adapted versions of the SVR models that update the offline-trained model using online data are implemented [16].

An updated version of SVR models are relevance vector machine (RVM) models, which are more efficient, as they first filter the most relevant data from the entire dataset and are then trained on those. In addition, GPR models, neurofuzzy models, and other approaches have seen application in the past years [16].

A very exciting upcoming area of aging modelling are online vehicular cloud-based models, where models are trained in the cloud and then additionally sent to vehicles [16]. A recent neural-network-based model presented by Li et al. was trained in this method, then trained additionally on the cell's internal data. After only 100 cycles (of a lifetime of around 1500 cycles), the models predicted EOL varied from the actual EOL by a maximum mean error of 4.2%, which is very good, and the model is trained further during the cell's lifetime, improving the prediction [39]. While this modelling technology is very new, Bosch has already presented their cloud services in 2019, meaning that the technology is already in a usable state [40].

As mentioned in the introduction to this chapter, capacity fade and internal resistance rise are the two parameters that are usually used to reflect battery state of health. However, it is difficult and damaging to the cell to obtain these quantities directly. A full charge-discharge cycle is required to directly measure the capacity, which is harmful to the cell and very rarely actually realized during the cell's in-use stage,

and to fully characterize the internal impedance of the cell, electronic impedance spectroscopy (EIS) data has to be collected; this is time-consuming and basically impossible in online applications.

Therefore, many papers on data-driven models have been researching other indicators based either on their own data or on publicly available datasets such as the previously mentioned NASA PCoE dataset, which contains EIS measurements taken after each cell charge-discharge cycle [18]. The hypothetical indicators are then tested with regards to their correlation to the EIS and capacity data, and if necessary, the data are optimized to improve the correlation [37, 41, 42].

Feng et al. used this process to find a promising new indicator in the form of time to voltage saturation (TVS) and time to current saturation (TCS), respectively the time during a constant current-constant voltage (CC-CV) charging cycle until the cell voltage reaches its maximum during charging and the time until the charging current reached its minimum. These data were extracted solely from current, voltage, and time measurements, which are easy to take online [41], and they were used to train a model to a high level of accuracy [42]. Zhao et al. examined the time interval of equal charging voltage difference (TIECVD) and the time interval of equal discharging voltage difference (TIEDVD), respectively the time that passed while charging and discharging between two previously defined voltage levels during CC charging. These were then used to train an SVR model which proved to reduce computational time by an average of about 31%, making it potentially applicable even in online situations (which is good news, considering the previously-mentioned fact that regular SVR models are not well-suited for online use), as well as more accurate for the most part than the standard SVR model [37].

Besides alternative health indicators, recent papers have also focused more on improving the data that cells are tested on to reflect the dynamic reality of the conditions that cells face in daily use. In a two-paper series, Lucu et al. present a cyclical aging model that was trained on one cell with variable temperature conditions and one with variable temperature, depth of discharge, charging and discharging C-rates, and average state of charge. They also present a pure calendar aging model trained on two cells that underwent dynamic temperature and state of charge conditions. While these are very low amounts of data, the model responded very well and the papers highlight the necessity for training data that reflect real-world applications better [31, 32].

In addition, the NARX model by Khaleghi et al. performed very well even when only supplied partial training curves, which is also important for future online applications [17].

Signal processing methods for optimizing data have been mentioned already and have been in development and in use for several years now as well, such as the discrete wavelet transform (DWT) [43], which takes into account the fact that aged cells fluctuate more [16], and the Box-Cox method, which was used by Zhou et al. [41] to improve the linearity of the correlation between their parameter and the capacity.

3.3 Semi-empirical models

Semi-empirical models, also called hybrid models in the literature [3], combine the data-driven approach with the physical-model-based approach in various ways. These encompass equivalent circuit models (ECMs), models that parametrize electrochemical equations using online and offline data, and models that parametrize curves that are not based on physical processes but on data fitting.

ECMs are an example of a frequently-used semi-empirical model, implemented often in the battery management system (BMS), the part of a battery that is responsible for managing the battery with the help of online data collection and modelling [24]. ECMs can accurately replicate battery cycle behaviour using an equivalent RC circuit whose resistance and capacitance values are determined through fits of either historical cell data or online driving data. Additionally, they have been used in order to model cell thermodynamics as the effects of both capacitive and resistive circuit elements are well-explored and well-described phenomena [23].

Another area that can be described via semi-empirical modelling was briefly touched upon in subchapter 3.1; as researchers are working on reducing the computational complexity of physics-based models as well as improve their quality, models have been developed whose parameters are updated using online data.

For example, Zhang et al. presented the SPM model with SEI layer growth which was mentioned in subchapter 3.1, which periodically updates its parameters using online data during the cell lifetime [25]. A P2D model presented in a paper by Ramadesigan et al parametrized electrochemical equations using estimates from past charge-discharge cycles, and was also therefore able to model how these parameters changed over the cell's lifetime, giving estimates at cycle numbers 25, 50, 100, and 200 [44].

Another interesting recent study by Li et al. analyzed the sensitivity of their NMC-graphite-cell-based reduced-order electrochemical model outputs (cathode bulk and surface state of charge (SOC), terminal voltage, and anode potential) towards changes in 26 physical model parameters during different SOC phases and at different charging C-rates. The terminal voltage was identified as sensitive towards 14 out of 26 parameters, meaning that the parameters could be easily and accurately identified using "fast data-based methods" [29], reducing costs and testing time and allowing parameters to be obtained independently from cell manufacturers and without destroying a cell. The other 12 parameters were further investigated and the paper concluded that "the inaccurate identification of these parameters has little influence on the operation of BMS functionality" [29].

Additionally, Lamorgese et al. state that it is "critical to provide estimates" of the two SEI-relevant parameters relevant to their electrochemical model [26].

As described in subchapter 3.2 for data-driven models, semi-empirical models can also be parametrized using alternative health indicators that improve their online usability. To get the values of the resistive and capacitive elements of their fractional-order ECM, for example, Guha et al. reconstructed the EIS data solely from the input charging current and the output terminal voltage [45].

3.4 Summary of lithium-ion cell lifetime modelling approaches

In summary, physics-based models have the ability to specifically address physical phenomena such as lithium plating and SEI formation in their mathematical formulae. However, they are generally computationally costly, which is a known fact and research is dedicated to improving this. Simplifications to these costly models may need fine-tuning with regards to parameters to better reflect the real cell's behaviour, leading to efforts to link electrochemical models to offline and online data in order to parametrize them better. This form of semi-empirical model is also popular as it can update parameters of electrochemical formulae during the cell's lifetime, which results in more accurate models.

Other important semi-empirical models are ECMs, which can use both offline and online data when calculating resistances and capacitances. These can also reconstruct cells' EIS spectra, which is very useful as the collection of EIS data is time-consuming.

Finally, data-driven models exist in a variety of forms. Statistical and ANN models have been improving strongly and now, with vehicular-cloud-based technology, are even able to predict EOL at high accuracy after very little online and offline training. While these are often parametrized with IR and capacity data, alternative health indicators have been a popular research topic, both for data for semi-empirical models and for data-driven models. In addition, data reflecting the conditions of real-life cells, which include partial charges and discharges as well as dynamic environmental circumstances, are slowly being used to train more and more models. In conclusion, data-driven models are more easily optimizable than physics-based ones, and they can be trained on data from environmental conditions that lie beyond the scope of simplified physics-based models. However, they cannot take specific electrochemical facts into account.

4. Automotive industry use cases and the decision tree

Use cases, in this thesis, represent questions and problems that members of the automotive industry have that can be solved using specific models. The use cases examined in the following chapter have been extracted from thirty questions posed by clients to AVL representatives between January and September of 2021, as well as participant FAQs from three AVL webinars on lithium-ion battery simulation that took place in a period from September 2019 to May 2021 [46–48]. For reasons of clarity, these will be grouped according to the battery lifetime stage they are most relevant to.

The lifetime of batteries will be examined in the following three different stages: Their development phase, their in-use phase, and their second life.

Questions concerning the development stage mostly focus on resources required to build a battery and to supply the model with data. The subchapter on development stage use cases will discuss how the model that the battery will later use during its in-use stage can be chosen so that specific resource groups are spared during the development stage.

During the in-use stage, client questions mostly concern risks that may end battery lifetime prematurely. These risks can either reduce the battery capacity too rapidly, meaning that the capacity-defined EOL is reached earlier than guaranteed by battery producers, or they pose security risks in the form of sudden mechanical and chemical failures. As the former is the more common subject of client questions, and as the focus of this thesis is on the question of lifetime maximization rather than security maximization, the subchapter on the in-use stage is focused on capacity-defined EOL and how models can be used to best delay the EOL.

The second life, and the applicability of a battery for second life, is most closely tied to the remaining value and RUL of a battery. Therefore, the subchapter on second life is most focused on early accurate prediction of the RUL, which enables early prediction of whether a cell is viable for second-life applications.

The use cases and questions that clients typically have are summarized into the following key questions:

1. How can the resources needed to model battery lifetime be reduced?
2. How can the lifetime of a lithium-ion battery be maximized?
3. How can the RUL of the battery be estimated?

The following subchapters will answer these questions using the knowledge presented in chapter 3 as well as some new sources, and these answers will be summarized in subchapter

4.1 The development stage

The resources currently needed to model a cell are:

- Data (both data quality and quantity)
- Cells (for conducting lifetime tests)
- Human labour (expertise and work hours required for conducting tests)

- Time (for lifetime tests as well as computation time)
- Energy

4.1.1 Data quality

Data, which are relevant for semi-empirical and data-driven models, is currently needed both in high quality and in high quantity. While a certain level of data quality is needed for the model to be reliable, signal processing methods were discussed in subchapter 3.2, where the DWT method was presented, which could feasibly work with fluctuating data to relatively good results [43]. However, it follows the dynamic stress testing current schedule, which requires a battery to be tested under dynamic current conditions from 90% SOC to 20% SOC, which is time-consuming and rarely realisable under in-use conditions. This means that low data quality can only be accepted if clients are ready to invest the time required to obtain the data amount to compensate for noisy data; Otherwise, the way that data are measured must be improved to reduce the noise.

On the other hand, of course offline physics-based models can be used if the necessary physical parameters are available. However, if the modelled cells are innovative (with novel chemistries, geometry, etc), it may be necessary to take some cells apart to parametrize them, which may be difficult if qualitative data measurement is not possible. Of course, it also means that cells and human labour are necessary to compensate for the inability to measure qualitative data.

4.1.2 Data quantity

As discussed in subchapter 3.2, the amount of data required in advance to supply a model has been decreasing with the ever-growing focus on online cell modelling. Models parametrized solely using online data are ideal as they do not require additional datasets or cell testing. Of course, the better and more representative the data, the less data is required for testing, meaning that data quality and quantity should not be low at the same time. On the other hand, if neither data nor online connections are available, a reliable physics-based model that can be used offline may be used. However, as mentioned in subchapter 3.1 and in the previous section, these require cell testing in the laboratory to obtain the physical parameters in cases where these parameters cannot be extracted from cycling data, meaning that a larger amount of human labour is required.

Of course, the need for external sets of historical data can also be eliminated through the use of vehicular cloud-based modelling, if the technology is available, as batteries are trained on each other's past in-use data.

4.1.3 Cells

Situations may arise where available datasets do not accurately describe the cells that should be modelled, such as when the cell chemistry of the present cells differs too much from the chemistry of the cells whose data was collected or the conditions the cells were tested under do not resemble those the cells

will meet in their lifetimes. In these cases, producers must collect their own data, primarily through lifetime testing.

Of course, in the same way the amount of data can be minimized, the amount of cells needed for testing can be minimized either if a physics-based model which can reliably be used offline is implemented, or if a mostly online data-driven model is used. The former, however, is only possible if the parameters needed for the physics-based model can be sufficiently estimated without necessary data, which may be difficult for cells with novel chemistries and geometries.

If the parameters cannot be estimated accurately enough, semi-empirical models that parametrize electrochemical equations using cycling data are a valid solution for minimizing the amount of cells used for testing in the development stage.

Additionally, the design-of-experiment phase could include computational methods to help decide how many cells should be tested under which conditions, as done in the paper by Prochazka et al., reducing the 1024 potential test points down to 46 required ones to characterize the cells [49]. Such work helps minimize the amount of redundancies; this effort is further discussed below in section 4.1.6.

4.1.4 Human labour

The amount of human labour is decreased in the same way the amount of cells used and the amount of test cases considered is decreased: Through viable offline models (as long as the parameters do not have to be determined through laboratory work), through electrochemical models with online parametrization, and through purely online-data-driven models. Of course, here as well, vehicular cloud-based models are optimal as they require neither laboratory cycling of cells, nor parameters determined through cell deconstruction. Laboratory work can also be minimized if data does not have to be pre-processed very much prior to training and modelling, such as the online NARX model presented by Khaleghi et al. that only used charging voltage rather than a refined health indicator such as TIEDVD for modelling [17].

4.1.5 Time

The amount of time needed for testing, similarly to human labour, can be reduced by using models that are trained on unrefined data. Additionally, Gu et al. describe a method of accelerated life testing that, instead of focusing on reducing the amount of cells required for testing, reduces the amount of cycles cells are tested for, which reduces the total time required (of course, under the assumption that multiple cells can be cycled in parallel) [50].

In addition, time can be saved by implementing vehicular cloud-based models as well, provided that a finished model is suitable for this case. If not, whether the development of such a model would take longer than cell lifetime testing to supply a finished model with data must be determined.

4.1.6 Energy

Energy, which here represents the resources (time, electricity, computation space, etc) that are wasted during inefficient processes, should also be taken into consideration when training models. The

following example illustrates why it is treated as a separate resource here: No matter how efficient lifetime tests are, they still need time, and the above section on reducing the time needed was about how the lifetime tests can be optimized in a way that saves time, but potentially increases the amount of other resources needed. On the other hand, if time is simply wasted due to an inefficient testing protocol, this is wasted energy that can be optimized without a necessary need for other resources.

In order to minimize the amount of wasted energy, regardless of which specific data-driven or semi-empirical model is chosen, the training protocols must be chosen in a way to minimize redundancies. Some papers, such as the two-paper series by Lucu et al. [31, 32], specifically used different training profiles with increasing amounts of data and conditions in which data were extracted (different temperatures, etc) and presented the profile with the optimal ratio of training data to prediction accuracy. These also reduce the amount of cells and data used.

As mentioned above in the part on cells as a resource, optimal design-of-experiment (DoE) using statistical modelling techniques significantly helps reduce energy waste as well [49].

4.1.7 Graphic summary of development stage

The six sections in this subchapter are summarized in a graphical format in Figure 2 which will then form a branch of the final decision tree at the end of this chapter.

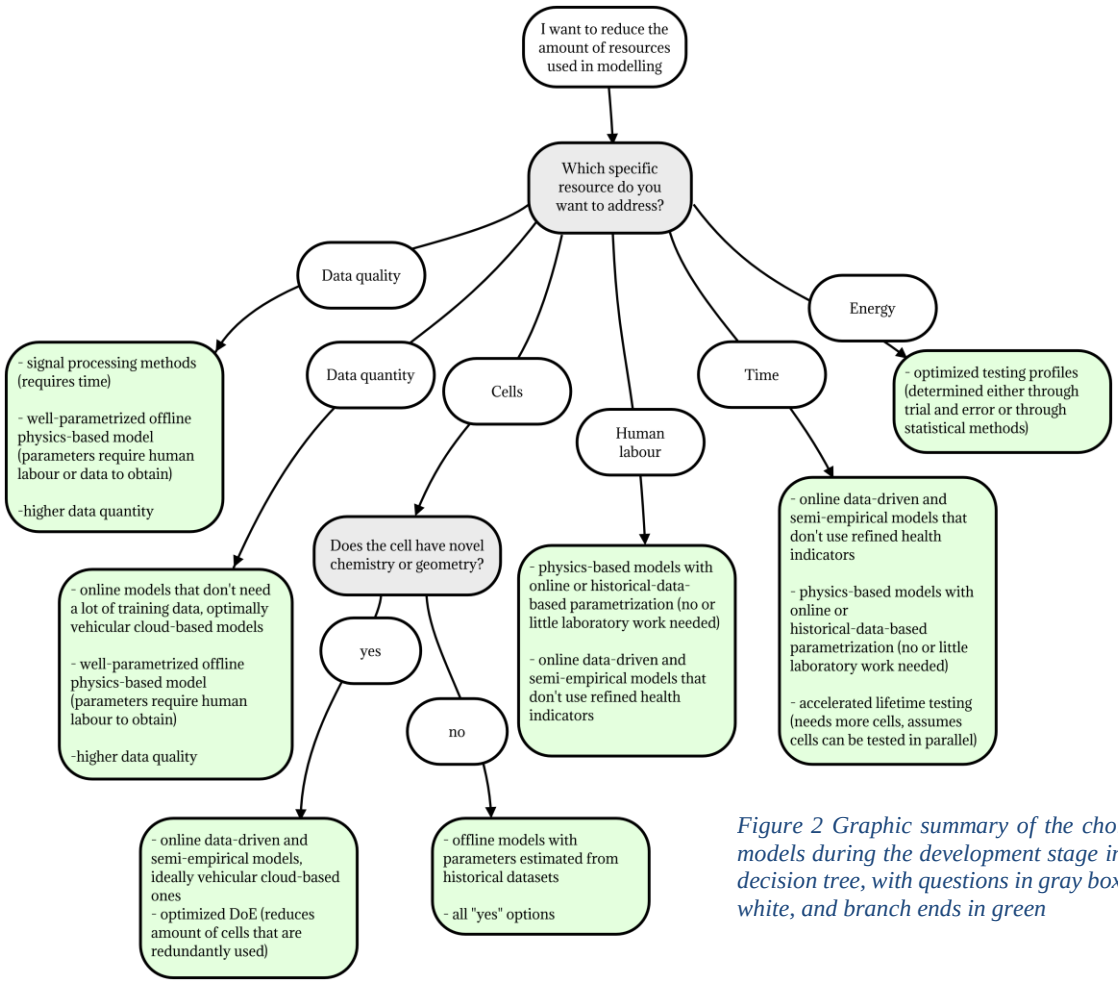

Figure 2 Graphic summary of the choice of suitable models during the development stage in the form of a decision tree, with questions in gray boxes, answers in white, and branch ends in green

4.2 The in-use stage

The use cases concerning the in-use stage mostly deal with how risks to cell lifetime duration can be minimized. The models recommended in these cases should be able to identify behaviour that damages cell capacity as early as possible to alert drivers to the damage this behaviour is doing, or to use the information to regulate battery behaviour.

Offline models are not regarded as suitable for this subset of use cases because the results are not accurate, as they do not fully reflect the circumstances of the battery that should be modelled. In a 2018 paper, Wang et al. compare the modelling performance of a BMS-suitable ECM model whose parameters are determined using offline data to the same model parametrized using online data. They conclude that the offline-trained model both takes longer to calculate parameters and provides less accurate results, as its data has a mean error ten times higher than that of the online-trained model [50]. This supports the conclusion that online-trained models are more suitable for predicting remaining useful lifetime.

Therefore, the models recommended should be online models, specifically those that can work with dynamic data, such as the models by Lucu et al. [31, 32], and those that can make lifetime predictions even with partial charging curves such as the NARX model presented by Khaleghi et al. [17]. These are very recent state-of-the-art models and the topic of dynamic testing data will hopefully be subject of further research in the future.

Other models that work well are electrochemical models whose parameters are estimated with online cycling data and periodically updated to improve modelling accuracy, such as the model by Ramadesigan et al. (theoretically capable of providing parameter estimates every 25 cycles) [44] and the one by Zhang et al., which updates its parameters every 20 cycles [25]. These updates are good for lifetime modelling as they respond to the dynamic conditions that EVs experience and reflect the effect these conditions have on the cell's state of health.

Of course, as electrochemical models are more complex to develop, this is more practicable if the chemical behaviour of the cell is not very different from the behaviour that electrochemical models can predict. On the topic of LFP cathodes, for example, Jokar et al. mentioned that there "is a lack of reliable [electrochemical] models" for simulating their behaviour [21]. In these cases, the recommended models should be data-driven.

If no online data are available, the solution cannot be presented by a model. Rather, on-board generic recommendations on and self-regulation of driving and charging behaviour would have to suffice, rather than specific recommendations provided in response to the driver's driving data.

This subchapter is summarized graphically in Figure 3, another branch of the final decision tree. Its main decision focuses on whether or not online data is available. If it is, the model that can be implemented depends on the capabilities of the battery's BMS.

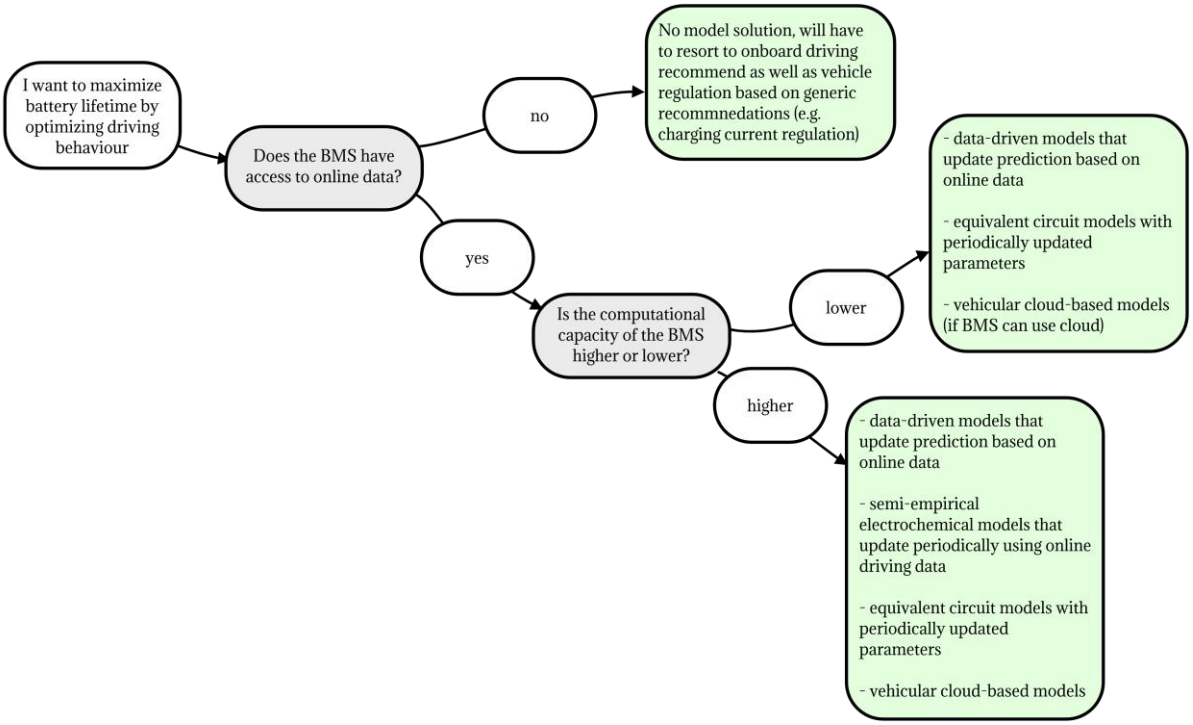

Figure 3 Graphic summary of the choice of a suitable model in the in-use stage in the form of a decision tree, with questions in gray boxes, answers in white, and branch ends in green

4.3 Second life

Principally, the remaining value is directly correlated to RUL. It is also assumed in this chapter that remaining value assumptions are most useful if they are provided as early as possible in a cell's life, so that plans for the cell's second life may be made at early points in its lifetime.

Therefore, models that are only trained online may not be ideal, depending on how many cycles are required to train the model up to an acceptable prediction error level. On the other hand, reliable physics-based models should theoretically be able to provide a relatively accurate estimate even prior to the battery's implementation in a vehicle.

However, the fact that it is not data-driven means that human labour would be required to determine the parameters in advance. Also, even if the non-data-driven model is accurate, the fact that it isn't updated with online data makes the estimated EOL vulnerable to unpredictable errors caused by how the cell is used, an aspect discussed in the previous subchapter.

Therefore, there must be a trade-off between how early the EOL must be known and how much the estimate may be influenced by how it is used. Specifically with data-driven models such as the SVR model by Zhao et al. [37], predictions could be made at about the 50% mark of total battery life. Similarly, the ND-AR model by Guo et al. preliminarily provided an EOL estimated by the autoregressive model around the middle of the battery's lifetime and refined it along its life using the

nonlinear degradation factor. Therefore, at least a rough estimate of the battery's RUL is available for a large portion of the battery's life.

Alternatively, if the technology is available, vehicular cloud-based models such as the ANN model described in subchapter 3.2 may be used, as their accuracy is astonishing even at early stages (around 7% of a cell's lifetime data used for training) and Li et al. specifically note that the model was developed for second-life-relevant use as well [39]. This, of course, improves the trade-off between how early the EOL must be predicted and how much the prediction may be influenced throughout its lifetime, as this model also improves its prediction during the cell's lifetime.

Figure 4 contains the final branch of the decision tree, again mainly differentiating between online and offline models.

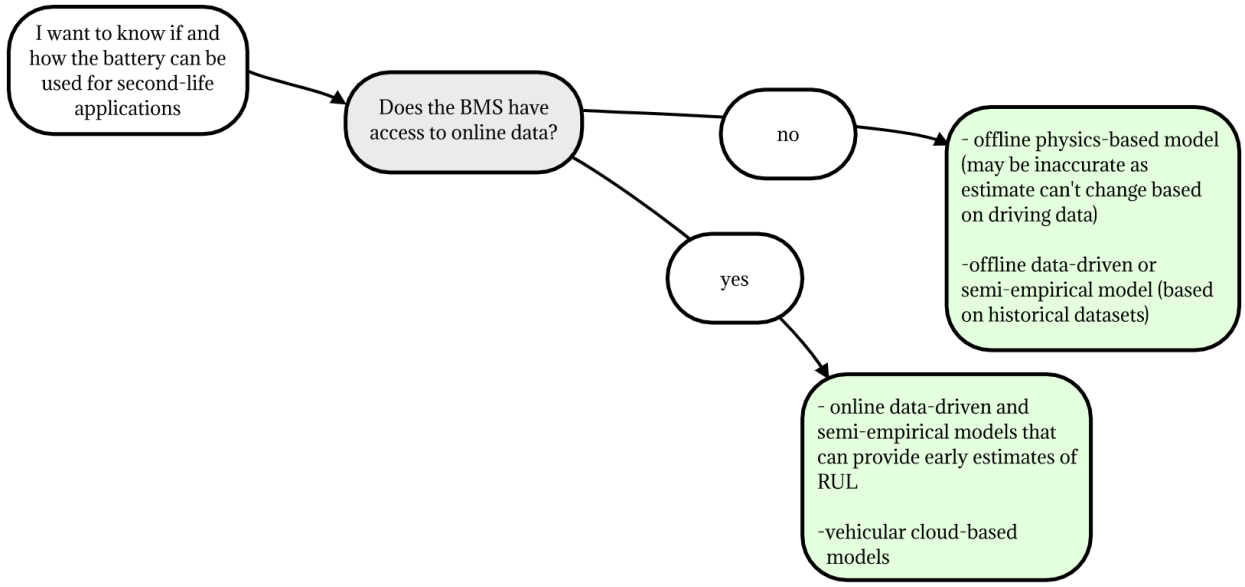

Figure 4 Graphic summary of choice of a suitable model in the second life stage in the form of a decision tree, with questions in gray boxes, answers in white, and branch ends in green

4.4 The use case-model decision tree and a use case example

The three decision tree branches developed in the previous three subchapters can now be combined in order to create the final decision tree in Figure 5, starting with the question of what broad problem a model should solve, and ending with the various models applicable to the use case.

The following example will be used to illustrate how the decision tree may be used in practice. A producer has a large dataset collected from development tests, and could feasibly supply a model with these data. They are implementing the developed battery with a BMS that cannot access online data. In addition, the producer has concerns that, if driver behaviour is too reckless, the battery will not keep as long as the warranty claims.

Therefore, they follow the "maximize battery lifetime by optimizing driving behaviour" track, then answer "no" to the question of whether they can access online data. They end at the node that states that no specific modelling solution exists to their problem under their circumstances. This can either motivate

them to implement online data access in their BMS or use their offline model to estimate the cell's second-life viability.

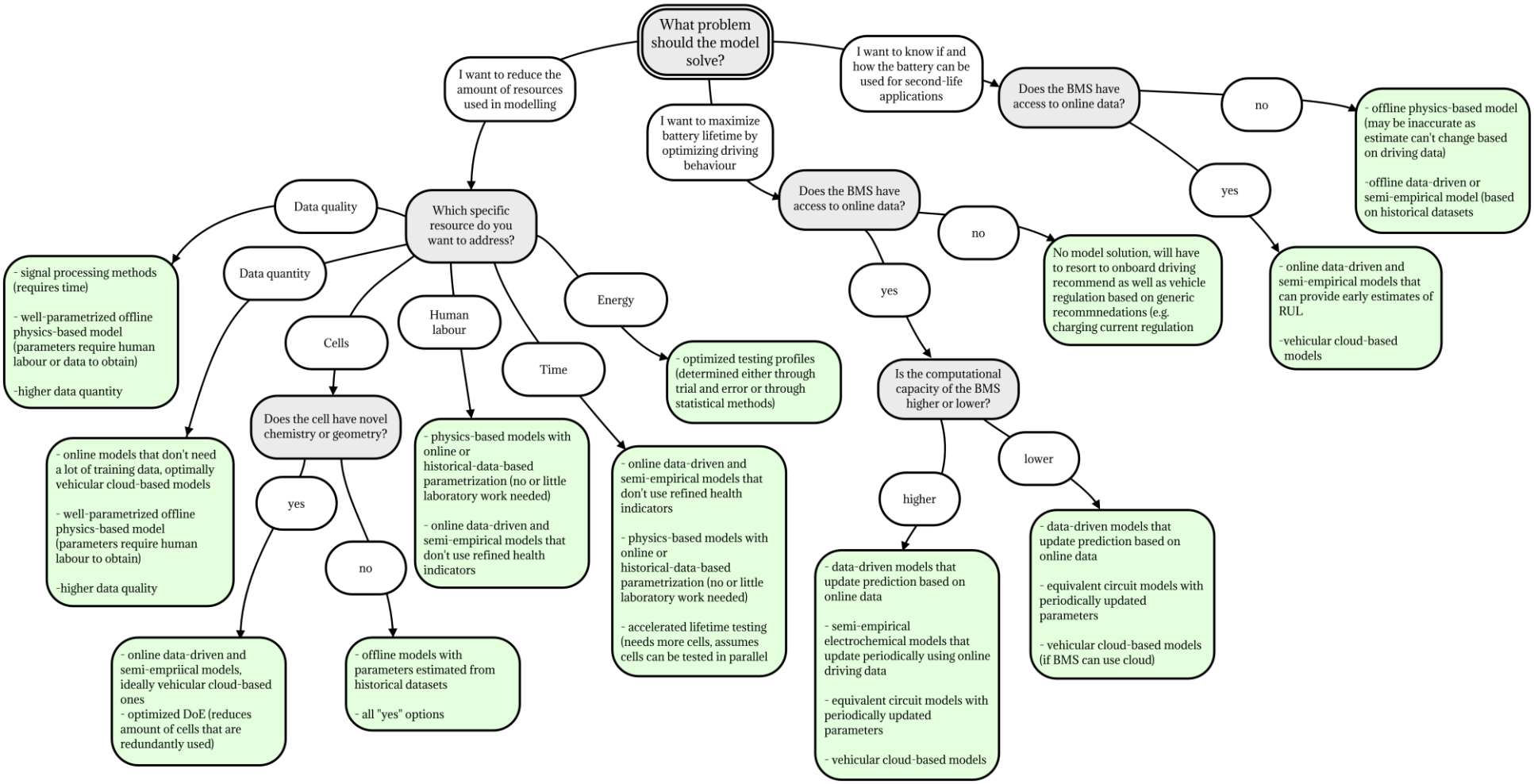

Figure 5 Final decision tree formed out of conclusions in chapter 4, with questions in gray boxes, answers in white, and branch ends in green

5. Conclusion and outlook

In conclusion, there is a large variety in battery modelling technologies and approaches, each with their respective advantages and disadvantages that make them very applicable in certain use cases and less applicable in others. This thesis serves to demystify the choice of model in use cases specific to the automotive industry.

It is an interesting time in the field of lithium-ion battery modelling as there is currently an increase in models trained on dynamic data, which is sure to impact the accuracy of models in the future as electric vehicles become more widespread and by default must be usable under diverse usage scenarios and climate conditions found globally. In addition, the combination of this dynamic data with the upcoming vehicular cloud-based technology can allow for fleets of vehicles with novel chemistries to be trained on each other's data and provide earlier and more accurate EOL estimates than ever before.

In addition, the European Union is in the process of implementing a new set of regulations and requirements for batteries that will certainly shape the market within the European Union. These set minimum requirements for the durability of batteries, a required minimum level of recycled material in cells (specifically with focusses on critical raw materials such as cobalt or natural graphite), and minimization of the carbon footprint of cell production, among numerous proposals[51].These regulations will certainly shift modelling focus to include second-life use in the modelling perspective, which up until now has been topic of few papers.

Considering these developments, the state of the art of modelling technology is expected to evolve further over the coming years, allowing for very interesting expansions on the research presented in this thesis.

6. Glossary

ANN ..... artificial neural network

AR ..... autoregressive

CC-CV ..... constant-current constant-voltage charging mode

DoE ..... design of experiment

EIS ..... electronic impedance spectroscopy

EOL ..... end of life

EV ..... electric vehicle

LCO ..... lithium cobalt oxide

LFP ..... lithium iron phosphate

LMO ..... lithium manganese oxide

NARX ..... nonlinear autoregressive with exogenous input

NCA ..... lithium nickel-cobalt-aluminium oxide

ND-AR ..... nonlinear degradation autoregressive

NMC ..... lithium nickel-manganese-cobalt oxide

RMSE ..... root mean square error

RUL ..... remaining useful lifetime

RVM ..... relevance vector machine

SEI ..... solid-electrolyte interphase

SOC ..... state of charge

SOH ..... state of health

SVR ..... selection vector relevance

TCS ..... time to current saturation

TIECVD ..... time interval of equal charging voltage difference

TIEDVD ..... time interval of equal discharging voltage difference

TVS ..... time to voltage saturation

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