

Characterization and Concept Validation of Lithium-Ion Batteries in Automotive Applications by Load Spectrum Analysis

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ABSTRACT: The design of a lithium-ion battery with the aim to meet given system requirements and to consider aspects of cell aging is complex as the correlation between system configuration and the resulting load on cell level is non-trivial. This paper proposes load spectrum analysis as an effective method for characterization and concept validation. Within the framework of this paper, the suitability of the WLTP Class 3 reference cycle for aging characterization is disproven. The effect of different cooling concepts and cell-to-cell variation within a battery system are evaluated with regard to lithium-ion cell aging and maximum cell and system load.

KEY WORDS: lithium-ion battery, aging, load spectrum analysis, system design

1. INTRODUCTION

1.1. Motivation

As part of the powertrain in electric vehicles, battery systems are exposed to a highly dynamic electrical and thermal load profile. The electrical load is determined by the power demand for propulsion and auxiliary users. The thermal load is caused by environmental conditions like ambient temperature and solar irradiation and it is coupled to the electrical load by reversible and irreversible heat generation.

Load profiles on a system level are often used as design criteria for the early concept phase. Either real load profiles from user data are available or standard cycles defined in specification sheets are used for this purpose. In both cases, battery system design variants are generated and an optimum system design is found through comparison. These variants may differ in the applied thermal management system, cell selection or in the chosen control strategy.

The selected variant will further determine how the applied load on system level is transposed to a load profile on cell level. For lithium-ion cells, which are commonly used in electric vehicle traction batteries, the applied load is of essential importance, as it determines their degradation during usage and also non-usage.

Aging-relevant loads include cell temperature, charging and discharging currents, the average state of charge (\bar{SOC}) and the depth of discharge while cycling (ΔDOD) (1, 2). Consequently, the load on cell level is an objective criterion for system selection and concept validation, as it is essential for aging assessment and thus for warranty commitments.

1.2. Aging-Relevant Load Regimes

The aging of lithium-ion cells is commonly classified into calendar aging, when the cell is at rest, and cyclic aging, when the cell is charged or discharged (2, 3). It is widely assumed that calendar aging is ever present, while additional aging effects are superposed when the cell is cycled (2, 4, 5). For these two classes of aging, different physical-chemical mechanisms are predominant. The mechanisms are triggered by distinct so-called stress factors which are present within the cell load profile. The stress factors for calendar aging are state of charge (SOC) and storage temperature, with storage time as base function (2, 4, 6). The applied discharge and charge currents, ΔDOD , \bar{SOC} and temperature affect the cyclic aging as stress factors together with charge throughput as base function (2, 7, 8). The stress factors are ever present within the load profile, as the aging of lithium-ion cells is. However, the

specific magnitude of the stress factors determines to what extent the aging is accelerated or slowed down.

The stress factors influence the aging mechanisms interdependently. For calendar aging, the formation of the solid electrolyte interface (SEI) between the graphitic anode and the electrolyte is widely assumed as a predominant aging mechanism. For this exothermic mechanism, Vetter et al. (6) state that a high temperature in combination with a high storage SOC results in an accelerated aging. For example, this phenomenon is proven by Keil and Warnecke (5, 9). For cyclic aging, a greater variety of aging mechanisms are involved and their dominance is dependent on cell chemistry. For example, lithium plating is enhanced by a combination of a high SOC, a high charging current and a low temperature (10). Aging due to structural changes is caused by mechanical strain within the electrode material mainly at the anode but also at the cathode. It is forced by cycling with a high discharge current, a high Δ SOC (1) and additionally at high temperatures (11, 12). At high temperatures, the transition metal dissolution within the cathode material is also an important issue for aging (13).

Fig. 1 summarizes graphically the discussed findings by literature review with a focus on NMC-based lithium-ion cells. A differentiation is made between aging mechanisms typically associated with calendar (cal) or cyclic (cyc) and whether the mechanism takes place at the anode (a), the cathode (c) or within the electrolyte (e). In Fig. 1, a two-dimensional area describes the aging mechanism as depending on two stress factors, and vice versa three stress factors for a three-dimensional area.

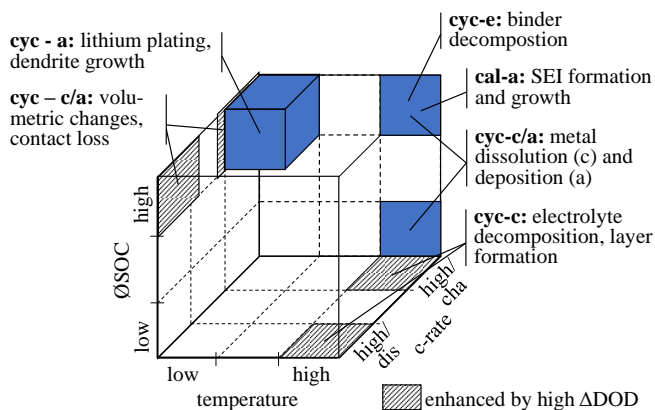


Fig. 1. Aging-relevant load regimes (1, 2, 5, 6, 12–17), calendar (cal), cyclic (cyc), anode (a), cathode (c), electrolyte (e)

1.3. Load Spectrum Analysis

The interdependencies between the different aging mechanisms necessitate extensive testing to characterize cell and system behavior and to verify adequate performance throughout the battery's specified service life. Due to the relevance of the

battery's dynamic behavior, high sample rates are necessary, resulting in large datasets of measuring files if the common standard of logging the measurement data as time series is used. Furthermore, the interaction between system parameters as well as the comparability between aging-relevant cell loads from different tests is limited and thus complicates the data interpretation.

A possible approach to overcome these limitations is the load spectrum analysis. Originally used in mechanical dimensioning, where load-cycles-to-failure are a relevant factor, this method allows the monitoring of frequency and amplitude of system parameters (18). Literature distinguishes between two fundamental approaches of load spectrum analysis: one-parametric and two-parametric methods. The former methods only consider one characteristic of a signal (such as its peaks), allocating its value to predefined classes. The latter methods take two characteristics of a signal into account, which are being evaluated together (such as its peaks and average values) leading to a two dimensional matrix (18). In theory, even higher parametric approaches are possible. These, however, lead to multidimensional data structures, not visually interpretable anymore, but possibly valid for machine learning, pattern recognition algorithms or other strategies of data analysis.

As basic concept of load spectrum analysis, different counting procedures exist to divide the signal values into specified classes. If the time spent in a class is irrelevant, this information can be eliminated. In the case of batteries, where aging mechanisms also depend on the time spent at certain operation conditions, algorithms determining the time spent inside the specified classes are more expedient (19) and are presented in the following. A common counting procedure is Instantaneous Value Counting, evaluating the class of a signal's value at a specified frequency. Other procedures include Residence Time Counting, weighing how long a signal stayed inside a certain class and Rainflow Counting, evaluating the amplitude and time of hysteresis loops in a signal. Related to this method is also the Half Cycle Counting, assessing only half cycles instead of a full hysteresis (18). The suitability of the different methods depends on the nature of the evaluated load cycle and the required information the user aims to extract. Table 1 offers an overview of different algorithms used for battery design and characterization.

In contrast to usual time-based approaches, load spectrum analysis is an event-based strategy. It improves assessment and comparability of occurring stress factors for different load cycles or scenarios. The incentive to apply load spectrum analysis for aging characterization is the assumption that the temporal order of

peak loads can be disregarded for aging assessment (18). For lithium-ion cells, however, a path dependency of aging has been identified (20) which is not representable by load spectrum analysis as information about the sequence of load is eliminated. Since this paper proposes a method for initial considerations within the process of battery system design, path dependency as a long-term effect was neglected. For a holistic investigation of aging, a connection between the enduring load scenarios and the resulting cell aging still has to be established through experiments or findings from battery systems in the field.

Table 1. Utilization of load spectrum analysis in design and characterization of battery systems.

| | Approach and evaluated parameters |
|------|---|
| (21) | Instantaneous Value Counting (C-rate, SOC, T) Rainflow Counting (Δ DOD, \emptyset SOC) Total charge throughput (C-rate) |
| (22) | Range Pair Mean Counting (SOC) Half Cycle Counting (C-rate, Δ DOD, \emptyset SOC) Total charge throughput (C-rate) |
| (23) | Instantaneous Value Counting (SOC, T) Rainflow Counting (SOC) |
| (24) | Rainflow Counting (C-rate, Δ DOD, SOC, T) |

2. METHOD

The objective of the presented work was to develop an effective method for the characterization of loads that affect battery systems in automotive applications. This includes a reduction of complexity for data analysis and for the evaluation of the corresponding effects on lithium-ion cell aging.

To meet this objective, a generic analytic framework was developed which is able to process given design criteria as input and allows aging characterization by load spectrum analysis upon given output measures (Fig. 2).

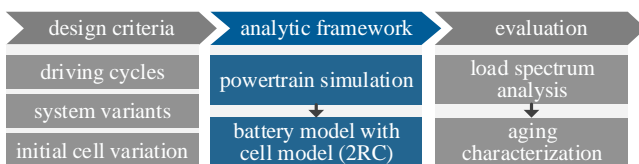


Fig. 2. Simulation framework

The analytic framework consists of a powertrain simulation presented in (25) to calculate the longitudinal dynamics of a conventional electric car. With this simulation model and based on a given velocity profile of a driving cycle, the resulting load

demand on the battery system level is calculated. The load demand on the system level is the input for an electric-thermal battery model. The battery model is capable of displaying different system design variants: cooling concepts, wiring concepts and cell-to-cell variations within the battery system. Further, the battery model allows the conversion of load demand on the battery system level to the corresponding load demand on the cell level and the derivation of parameter variations (current and temperature) for individual cells within the battery system. The battery model comprises a 2RC cell model which was parametrized with a Panasonic NCR18650PF cell and allows the determination of the cell's voltage response which is dependent on cell temperature, current magnitude and cell SOC (26). Eventually, current, temperature, charge throughput and SOC for individual cells are available as framework output for further investigation by load spectrum analysis. Within the framework, Instantaneous Value Counting was applied as counting procedure.

3. RESULTS

3.1 Aging Characterization on Cell Level

As a first example for aging characterization with load spectrum analysis, the test cycle WLTP Class 3 and real driving profiles from fleet tests are compared. WLTP Class 3 was used as it is widely accepted for the comparison of consumption ratios and emission values for different vehicles. So, the question stands, whether the WLTP Class 3 is also appropriate for the comparison of electric vehicle performance in terms of battery aging. As reference for realistic battery loads, about 12,800 real driving cycles from fleet tests are used, as they represent the driving behavior of more than 100 test persons over a period of more than 12 months, while using different types of electric and conventional vehicles (27). For simulation, cell-to-cell variation and battery cooling was neglected at first. Further, the SOC at cycle start is adjusted to each driving profile individually for simulation, according to a study that proves drivers of electric vehicles do not start all journeys with a fully charged battery but recharge the vehicle battery when the SOC reaches 32 % on average (28). Similarly, the cell temperature at cycle and simulation start is adjusted according to the known driving cycle date and to the appropriate monthly average temperature in Germany. Consequently, SOC and cell temperature at simulation start varies for the different real driving cycles. During the subsequent simulation run, the SOC evolves in accordance to the cycle power demand. The cell temperature develops as a result of reversible

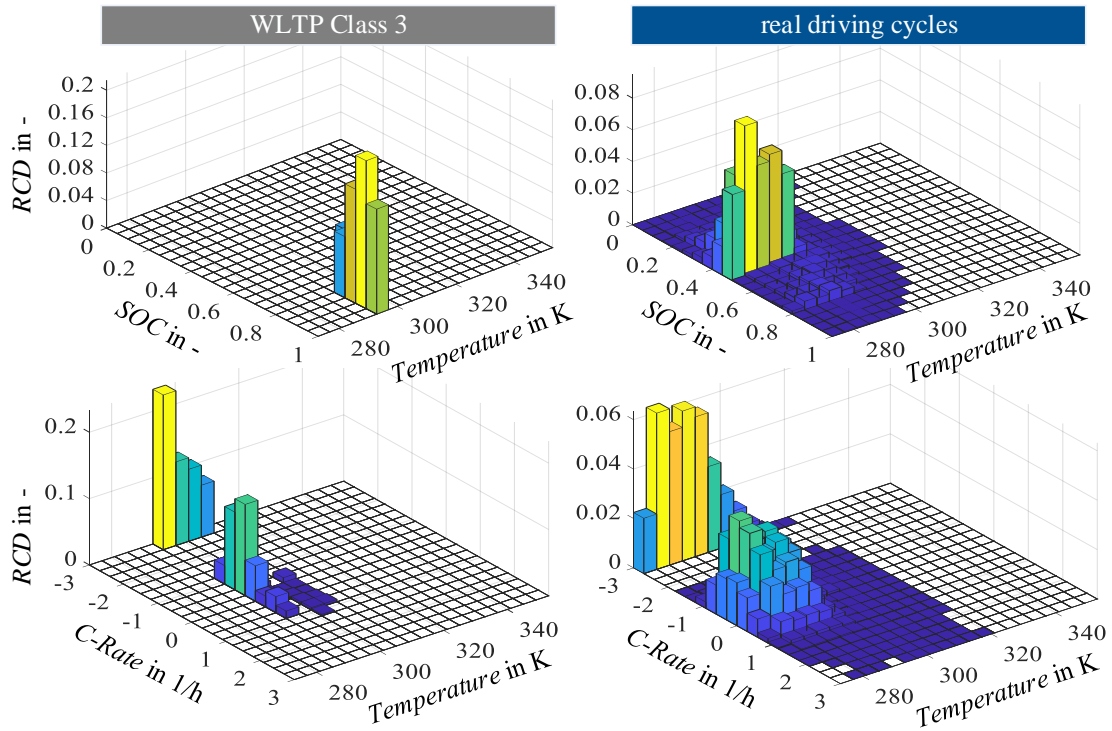


Fig. 3. Comparison between load spectra of WLTP Class 3 standardized cycle vs. real driving cycles

and irreversible heat generation as well as heat exchange with the environment. The results of simulation and load spectrum analysis are shown in Fig. 3 in which the stress factors temperature, SOC and C-rate are analyzed for aging characterization. For the C-rate, a negative value describes a discharge load, while a positive value describes a charge load due to regenerative braking. The relative charge throughput (RCD) is entered on the z-axis as base function of cyclic aging (5).

The results show that the WLTP Class 3 occupies load regimes with peak frequencies positioned differently and also a smaller frequency variation in comparison to real driving cycles. So, for the WLTP Class 3, moderate temperatures in combination with high SOC values and high discharging currents are present. In comparison, low temperature values are also combined with high charge and discharge currents, as well as low SOC values for real driving cycles. Additionally, the loads due to real driving cycles exhibit a distribution with a wider spread. These findings show that the WLTP Class 3 does not cover the full range of aging-relevant regimes, which are occupied in the load spectrum of real driving cycles. With respect to the discussed findings in section 1.2, the WLTP Class 3 will not cause the same aging for lithium-ion cells as the cells will exhibit under real usage. These results show that the WLTP Class 3 is not an appropriate standard test cycle for giving a reference of battery aging due to real driving loads.

3.2 Aging Characterization on System Level

Further examples for the utilization of load spectrum analysis are shown on system level. To fulfill the power and range demand in electric vehicles a parallel connection of lithium-ion cells may be necessary. Lithium-ion cells, as chemical systems with a complex manufacturing process, are prone to variance in production. Hence, cell parameters, such as internal resistance and capacity, show fluctuations, mostly regarded and measured as a standard distribution (20, 29, 30). These parameter variations lead to compensating currents and furthermore complex non-linear relations between system load and cell load (31). Consequently, cells connected in parallel behave and age non-uniformly in a battery system within a magnitude that cannot be neglected (30). Further influences, directly derivable from the theoretical assessment in the previous sections, include the design and control strategy of a thermal management system as well as the electric control strategy of the pack itself, including peak power control, voltage limitation and power degradation at high temperatures, low SOC or other aging-relevant regimes.

Considering these aspects, load spectrum analysis proves to be an appropriate instrument, when conducted on a system level and further when considering single cell behavior. To show the suitability of the approach, a simulative investigation of several generic automotive battery packs was conducted using the analytic framework presented in section 2.

As in reality, the exact composition of cells with their respective parameters in parallel connection is random. This fact has to be taken into account in simulation. To gain statistically relevant results, a Monte-Carlo Simulation was performed. For the investigation of battery design safety issues, an analysis of worst-case scenarios has to be conducted as well. This, however, is beyond the scope of this paper and therefore is omitted from the results. The goal of this paper is to show the plausibility of utilizing load spectrum analysis for battery system design. Therefore, two representative use cases are discussed in the following. Both examples use the analytic simulation framework presented in a prior part of this paper.

The first example shows the behavior of 20 Panasonic NCR18650PF cells in a parallel connection (20p1s) during fast charging with constant current (80 % SOC in 30 minutes). The maximum allowed C-Rate was defined as 1.5 1/h to strike a balance between a short charging time and excessive aging of the cells. It is assumed that the cells' internal resistances and capacities have an aforementioned standard deviation of $\sigma_{\text{resistance}}=1.1 \cdot 10^{-2} \Omega$ and $\sigma_{\text{capacity}}=3.5 \cdot 10^{-3} \Omega$, respectively. In a second simulation, the standard deviation of the cells' parameters was doubled relatively to the reference. The resulting loads, represented as the C-rate on the cell level, were evaluated with a discrete resolution of 0.1 1/h and are depicted in Fig. 4.

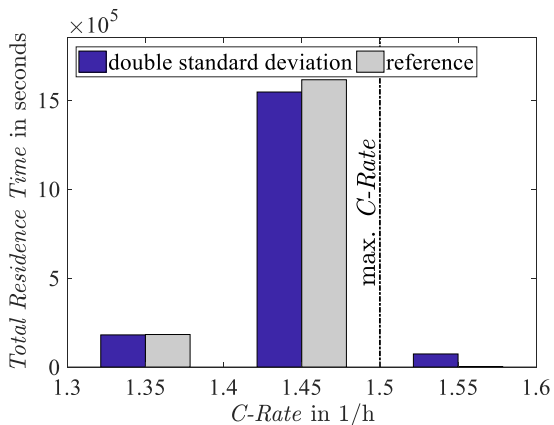


Fig. 4. Fast charging (0.8 SOC in 30 min) of a 20p1s connection of Panasonic NCR18650PF cells.

Reference: $\sigma_{\text{resistance}}=1.1 \cdot 10^{-2}$, $\sigma_{\text{capacity}}=3.5 \cdot 10^{-3}$.

The results of the reference system show a maximum cell load of 1.45 1/h for most of the cells, with another small peak at 1.35 1/h. The system with double standard deviation, however, develops another load level at 1.55 1/h and so exceeds the defined maximum C-rate and overloads the cells. While it is unknown from the load spectrum how the currents – or the time in overload – are distributed between the cells, load spectrum analysis allows for

identifying misuse or overtaxing of cells as well as the impact of different cell parameters or their standard deviation, respectively. To safely operate the system with double standard deviation, the charging current has to be limited. It therefore becomes apparent that statistical fluctuations of the cell properties directly influence the maximum charging current of a parallel connection.

Another use-case of load spectrum analysis is examined in a second example by analyzing the influence of different cooling configurations on battery system temperature. Therefore, a 72p28s battery system with standardly distributed internal resistances and capacities is simulated. The current is chosen to correspond to a constant travel of 120 km/h of a Smart Fortwo car retrofitted to electric drive (25). The cooling of the battery pack is modeled in a simplified way with no thermal interaction between the cells with a constant ambient temperature of $T_{\infty}=30 \text{ }^{\circ}\text{C}$ for all cells. Different cooling capacities are taken into account by variation of the heat transfer coefficient α between the cells and their environment. The first configuration is defined by $\alpha_1=50 \text{ W/m}^2\text{K}$, the second configuration possesses better cooling characteristics – achievable, e.g., by increasing the volume flow rate in a fluid cooled battery pack – of $\alpha_2=65 \text{ W/m}^2\text{K}$ which are opposed in Fig. 5.

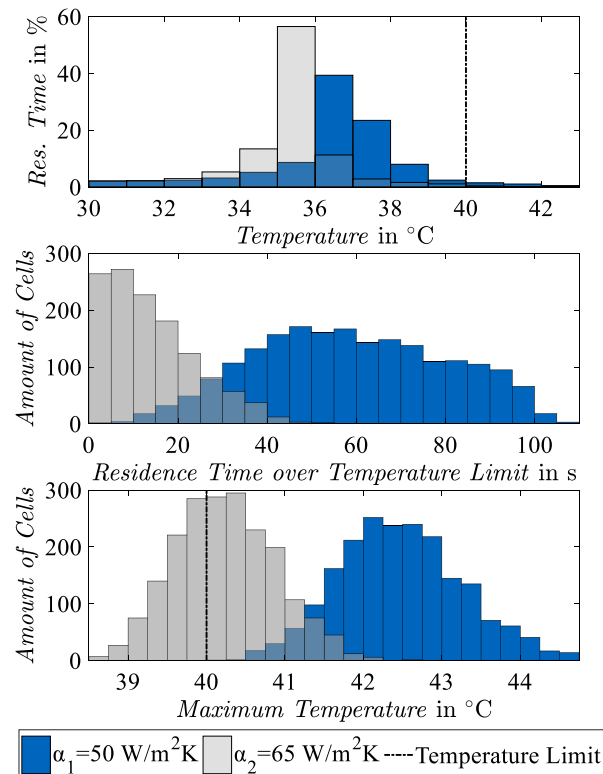


Fig. 5. Comparison of 72p28s battery packs with different cooling characteristics during constant travel and $T_{\infty}=30 \text{ }^{\circ}\text{C}$.

During simulation, three load spectra are generated. The top spectra track the percental residence time at the specified

temperature levels. The dashed line in the graph shows a maximum desired temperature of 40 °C for the lithium-ion cells. The load spectra shown in the middle visualize the total time the individual cells spend above the specified temperature limit of 40 °C. The lower spectra show the cell temperatures achieved in maximum. The evaluation of load spectra shows that the average temperatures in the pack with better cooling, represented by the case with $\alpha_2=65$ W/m²K, are lower and the cells spend less time above the defined temperature limit, respectively. The evaluation states that the given system requirements with regard to continuous power and thermal properties cannot be fulfilled by the second cooling option with $\alpha_1=50$ W/m²K. Furthermore, the graph's columns follow a normal distribution, which is also expected, since the cells' heat generation depends on the – normally distributed – internal resistances.

4. CONCLUSION

The discussed results show that load spectrum analysis is an effective method for load characterization and aging assessment of lithium-ion cells in the early design phase. It helps to compare different system concepts and therefore is suitable for initial system selection. When using load spectrum analysis, the applied counting procedure needs to be selected carefully as procedures that work with averaging processes may smooth the underlying loads. Especially in the context of cell aging, occurring peak loads are essential and need to be considered for system design. Therefore, Instantaneous Value Counting is a suitable procedure for the application of aging characterization and was used in the shown analyses.

When using load spectrum analysis for aging characterization of lithium-ion cells, the loss of information about temporal sequence within the load profile need to be considered. Consequently, path dependency as aging influence cannot be investigated. For the final selection of system design, additional methods which include the temporal order of load need to be applied.

CONTRIBUTIONS

T. Gewalt and C. Reiter were mainly responsible for the concept of this paper, contributed equally to the paper and share the first authorship. M. Baumann proposed the concept of load spectrum analysis for aging characterization. He and X. Lin developed the tool for the load spectrum analysis in a former non-published project. M. Baumann conducted the cell measurements and parameterization of the cell model. T. Krahl evaluated suitability of load spectrum analysis for battery system design. A. Hahn

provided important input for simulation assumptions. M. Lienkamp made an essential contribution to the conceptual planning of the research project. He revised the paper critically for important intellectual content. M. Lienkamp gave final approval for the version to be published and agrees to all aspects of the work. He accepts responsibility for the overall integrity of the paper.

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