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Quantifying Uncertainties in Reusing Lithium-Ion Batteries from Electric Vehicles

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Abstract

The economic viability of reusing lithium-ion batteries obtained from electric vehicles is highly sensitive to increasing inner resistance and capacity fading of lithium-ion cells. For decision making after removal of the batteries, it is necessary to consider the uncertainties in prediction of the remaining battery lifetime.

The purpose of this article is to provide a way for lifetime prediction by quantifying and considering occurring uncertainties. According to literature, the major risks in lifetime prediction are non-linear change in capacity, increasing cell spreading and exceeding critical limits like deep discharge during battery life. Separate investigation of linear cell aging and these uncertainties, up to battery pack level, helps in identifying correlations between operation conditions and the according failure distribution.

Keywords: Lithium-Ion Battery; Reuse; Lifetime Modelling; Uncertainty Analysis; Cell Failure

1. Introduction

The increasing shift towards electro mobility will lead to numerous used traction batteries in the market. Standridge and Corneal [1] estimate in various scenarios that the number of post-vehicle application battery packs would rise from 1.4 million to 6.8 million by 2035. Lithium-ion batteries used in EVs have a limited lifetime, due to a lack of fulfilling the vehicle requirements after under-running a predefined level of capacity and power.

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The United States Advanced Battery Consortium [2] defines the end-of-life (EOL) of electric vehicle batteries at 80% of the remaining capacity. Nevertheless, the batteries still have significant potential left for alternative uses. Furthermore, in the near future, car manufacturers will offer battery upgrades for existing models to keep sales numbers high. This will lead to a lot of batteries in the market place until 2020, which are only marginally degraded.

To establish an EOL market, it is necessary to define the products for reuse and the associated monetary residual value. For example, in existing after-sales markets for conventional vehicles, the combustion engine, starter motor and generator are established products for remanufacturing [3]. This suggests that an appropriate after-sales market will also be established for EV components. This article focuses on the reuse of the lithium-ion batteries. After removing the batteries from the vehicle, three possible value chains occur. Apart from battery recycling, which is always the final life cycle step, it is possible to remanufacture battery systems and bring specific components like the battery management system (BMS) into the after-sales market. This can reduce the cost for warehousing of spare parts for the OEM. The third value chain is the reuse of battery modules or complete battery packs in so-called Second Life applications with lower performance requirements, like stationary energy storage systems [1, 4]. The reuse of batteries can lead to reduced economic and ecological costs over the entire battery life-cycle. To extend the battery life cycle and enable reusability, it is essential to determine the remaining useful life (RUL). To estimate the RUL, the battery degradation and the uncertainties during degradation like non-linear aging, cell spreading and exceeding of critical limits have to be considered. Current RUL modeling mostly ignores these uncertainties. Thereupon the main purpose of this article is to extend the common RUL estimation with a statistical approach to quantify the uncertainties and estimate the probability of cell failures and cell spreading along battery life.

2. Battery degradation and RUL prediction

To understand RUL modeling, this chapter provides only a brief overview of lithium-ion battery degradation, since published reviews like Vetter et al., Barre et al. and Agubra et al. [5, 6, 7] already give a deeper insight into this broad topic.

The lithium-ion cell is the key component of every lithium-ion battery system. The cells mainly consist of a system of anode, cathode, electrolyte, separator and current collector. Currently in automotive applications the most used cell cathodes are nickel manganese cobalt oxides (NMC), nickel cobalt aluminum oxides (NCA) and partly lithium iron phosphate (LFP). The anode consists usually of graphite [4]. Each material changes and degradation of the cell components due to e.g. vehicle operation affects the cell performance and with it, the ability for reusing the battery system. In general, all cell components are affected by the degradation process, but a few aging processes dominate. The degradation leads to capacity and power fade [8] as well as reduced functionality and reliability. To frame the aging of the lithium-ion cell, it can be separated in calendrical and cycle aging [9], whereby both effects have different major sources. The calendrical aging is typically influenced through stress factors such as temperature and state of charge (SoC) [10]. Very early conducted tests of lithium-ion cells from Wright et al., Bloom et al. and Ramasamy et al. [11, 12, 13] show these dependencies. Due to exponential behavior this is mostly modeled according to the Arrhenius equations [14]. The cycle aging affects the discharging and charging process [9]. This reflects the effect of current rates, temperature and depth of discharge on the battery parameters. To handle the battery degradation and to predict the RUL, battery aging models are applied. Meis et al. [15] summarize and cluster the wide variety of existing battery aging models.

3. Uncertainties during degradation

According to literature [16, 17, 18, 19, 20, 21, 22, 23, 24, 25], the start of non-linear aging, increasing cell parameter spreading and the exceeding of critical limits are the severe sources for uncertainties in lifetime prediction. Fig. 1 illustrates linear and non-linear aging combined with cell spread of cells from a specific battery system along the battery lifetime. In this article, we focus on the relative capacity development, due to the major impact on the residual value of lithium-ion batteries. Furthermore, Schuster et al. [21] illustrate that the development of the resistance is nearly reciprocal to the capacity. Nevertheless, the later proposed method can be applied to other battery parameters like inner resistance and impedance as well.

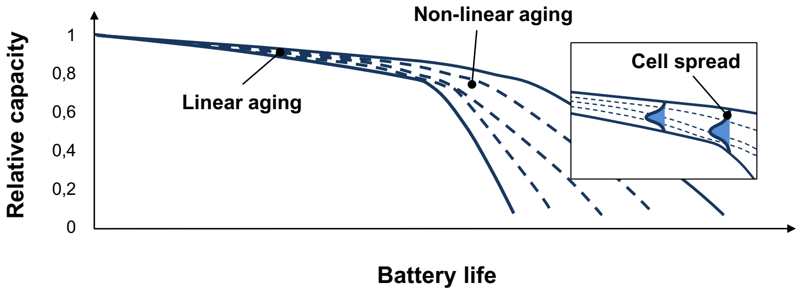

Fig. 1. Impact of uncertainties on linear and non-linear ageing in battery systems

3.1. Non-linear aging

The non-linearity in the aging process leads to high uncertainty in the lifetime prediction and modeling of lithium-ion cells [23, 24, 25]**.** The non-linear aging of lithium-ion cells is a major issue because it is difficult to estimate when the turnaround and accelerated aging occurs. Schuster et al. [21] made a detailed investigation to understand this effect and pointed out that the reason for switching from the linear to nonlinear decrease of capacity can be explained by aging induced lithium plating. It describes the deposition of metallic lithium on the graphite anodes [22]. Initial condition for the formation of lithium plating is, when the graphite potential is reduced below 0 V vs. Li/Li^+ [17]. The lithium plating results in a fast consumption of active lithium, which leads to a sudden drop in capacity. Petzl [16] distinguishes lithium plating in reversible and irreversible deposition of metallic lithium. Bach et al. [18] points out that the plating appears to spread out over the whole graphite anode and quickly consumes the remaining usable lithium. Summarized, after the beginning of non-linear aging initiated through lithium plating, reusing of batteries is not viable anymore. Lithium plating can occur during charging at low temperatures with high current rates and at a high SoC [17]. Schuster et al. [21] shows the facilitation of the turning point at high charging current rates, low temperatures and high depth of discharge (DoD) [21]**.**

To gain insight into the chemical changes which lead to this sudden degradation, Bach et al. [18] applied post-mortem analysis to three different cells, each with different progress of aging. They identified that another important influencing factor for lithium plating are local compression inhomogeneities of the electrodes in the cell housing as well as in battery packs. False battery system design, cell design or production processes lead to inhomogeneous pressure, and these high pressure points turn out to be the initial start for lithium plating. Therefore, both the amount and method of applying pressure is of great importance for maximizing cell performance, lifetime and safety.

3.2. Cell spreading

Apart from the non-linear aging, the cell spreading within the battery system is a further issue. Cell capacities and inner resistance vary within a system. Schuster et al. [20] investigate the cell to cell spread with 484 new and 1908 aged NMC cells. The aged cells came from two identical battery electric vehicles (BEV) and were characterized by the battery parameters capacity and inner resistance. The evaluation of capacity showed a shift from Normal to Weibull distribution with regard to the progress of aging. Furthermore, the number of outliers increases.

In [19], cylindrical cells based on LFP-technology were examined at start of life and end of life conditions. The initial capacity variation of 20000 cells is measured and fit a normal distribution with 1.3% deviation of the initial capacity and 5.8% deviation of the internal resistance. The aged cells show an average capacity loss of 18% with a maximum cell spread between 14% (best cell) and 25% (worst cell). In [26], we accomplished capacity tests for 192 prismatic cells of a Daimler Vito-e-cell BEV and determined a deviation of 6.9% between the capacity of the best and worst cell. The major problem is that cell spreading also occurs within a battery module, which represents the smallest reasonable unit for reuse. Further disassembly to cell level is economically not viable.

We expect the cell production and the battery system design as the major sources which are responsible for the cell spread. Batches of new cells are unequal due to production inhomogeneities [27]. According to our measurement of over 100 purchased cells it turned out, that cell manufacturers grade the cells and just use the high graded cells for automotive applications. However, there is still a cell spread. In the production process, the electrodes are coated with graphite and lithium metal oxides. During the coating process there are statistical inhomogeneities in porosity and thickness. Furthermore, the wetting of the electrodes with the electrolyte can vary. In addition, there are various other sources for production caused cell spreading and in electric vehicles the battery system designs varies as well. They differ in used cell sizes, connections of cells and e.g. design of cooling system. Due to numerous parallel connections of cells and their parameter spread in capacity and resistance, all the cells do not underlie the same current rate. The thermal isolation and the used cooling system may cause inhomogeneities/gradients of the cell temperatures as well. Fig. 2. Summarizes the three sources production inhomogeneities, load inhomogeneities and false battery system design for cell spreading.

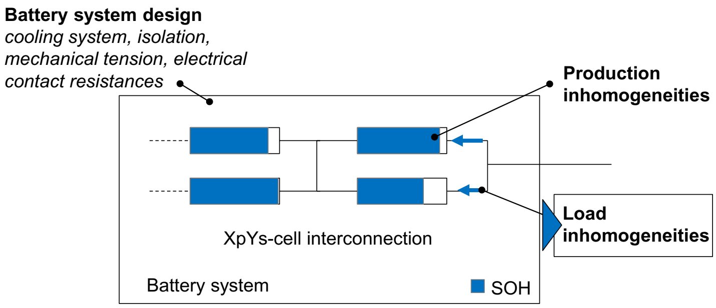

Fig. 2. Sources for cell spreading in battery systems

3.3. Exceeding critical limits

A further source of uncertainty is exceeding operating parameter limits and the consequences like dendrite growth because of deep discharge in combination with low temperatures. These critical conditions should not be reached in operation at any time and have to be prevented by safety devices like the battery management system (BMS). Over- or under voltage can easily be detected and limited by the BMS [21]. Detecting and preventing the operation at too low cell temperature gets more complicated. Therefore, thermal and resulting charge inhomogeneities e.g. in large cells/packs, even if external heating is applied, have to be taken into account [28].

4. Quantifying uncertainties

In the following chapter, ways for the quantification of the above mentioned uncertainties are presented.

4.1. Weibull distribution

To describe the non-linear aging and cell spreading, it is possible to use a translated Weibull distribution. According to [29] the distribution function of the translated Weibull is described by the function

$$F(t) = \begin{cases} 1 - \exp\left[-\left(\frac{(t-to)}{\eta}\right)^{\beta}\right], & to \leq t;\ \beta, \eta > 0 \\ 0, & 0 \leq t < to \end{cases} \quad . \tag{1}$$

where η is the scale parameter, ß the shape parameter, t_0 the location parameter and t the time.

The correlation between distribution function and density function is

$$f(t) = \frac{dF(t)}{dt} \quad . \tag{2}$$

The shape parameter ß defines the distribution of the failure occurrences and can influence the Weibull distribution to approximate different distributions e.g. normal, exponential and Rayleigh. The variation of ß is used in describing the failure rates of products in form of a bathtub curve [29]. It is divided in three sections:

- Early failures with decreasing failure rate: $ß < 1$
- Random failures with constant failure rate: $ß = 1$
- Wear-out failures with increasing failure rate: $ß > 1$

Due to our focus on the turning point from linear to non-linear aging, we investigate the later stage of the battery life which is categorised by wear-out failures. The scale parameter η describes the approximated location of the maximum of the distribution and the characteristic life cycle of the system [29].

The translated Weibull distribution has an additional parameter t_0, compared to the standard Weibull distribution. It shifts the distribution on the time axis. Hereby a distribution without failure occurrence for the time interval $t < t_0$ can be modelled.

4.2. Quantification of non-linear aging

4.2.1. Failure time modelling

In Fig. 3 an exemplary degradation curve for a cell with a theoretically possible failure time is displayed. It is important to understand the impact of different influencing parameters on nonlinear aging. In Fig. 3, qualitative indications of the impact of varying influences are visualised. As mentioned previously, apart from local compression inhomogeneities, the stress factors such as high charging rates especially at low temperatures and high DoD/ΔV facilitate lithium plating [21]. The probability for non-linear aging is increasing and is shifted to an earlier point in time. Specific operations by, for example, more moderate conditions can lead to the opposite effect and delay or withdraw lithium-plating [17]. Petzl [16] points out that adaption of temperature to higher temperature can for example benefit the decrease of deposited metallic lithium.

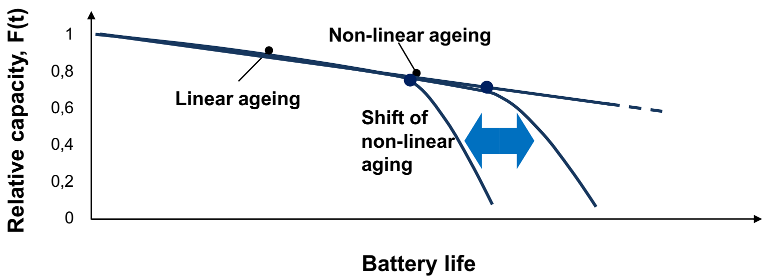

Fig. 3. Adaption of failure time

For the implementation of our approach, it is important to derive correlations between the onset point for non-linear aging and the stress factors like current rate, temperature and DoD. Therefore, it is necessary to gather empirical data of the specific cell. Examples are the publications of Schuster et al. [21, 30], where they investigated high-energy lithium-ion cells of the type Molicel IHR18650A by E-One Moli Energy Corp. with a nominal capacity of 1.95 Ah. The results showed the dependency between non-linear aging and current rate (charge/discharge), temperature and DoD. Based on these empirical data points our used parameters can be fitted, considering the stress factors and for example occurring events: Event I: high charging rate, Event II: high charging rates and low temperatures as well as Event III: high charging rates and high DoD. All are beneficial for lithium plating. Thus the impact of the stress factors on the onset of non-linear aging can be first analyzed and then discretized in shift to earlier or later time per e.g. cycle. This shift can be weighted according to the impact.

Apart from this way of quantifying the impact of stress factors on the time point of non-linear aging, Petzl [16] developed a linear regression model, though just for low temperatures, which models the correlation between stress factors and the occurring mass/area increase of irreversible and reversible lithium plating. Again, for parameterization cell testing data sets are necessary.

To analyse the load curves and identify critical degradation, event counting methods can be applied. In each load profile, critical and beneficial signal ranges are identified and counted, which influence lithium plating and therefore the begin of non-linear aging. It can be useful to create a transition matrix, as it is usually applied on analyzing load cycles in dynamic fatigue analyzes. Transition matrices are a useful tool for visualizing signal patterns, and to identify critical loads [29]. Every incidence, when one of the signals or the combination enters its critical signal range, the time step and magnitude of the signal is recorded. Since the events mentioned in

Fig. 4 are most favorable for the formation of lithium plating, it is necessary to analyze the C-rate and the combinations of high charging rates with low temperatures as well as high charging rates and high DoD [21].

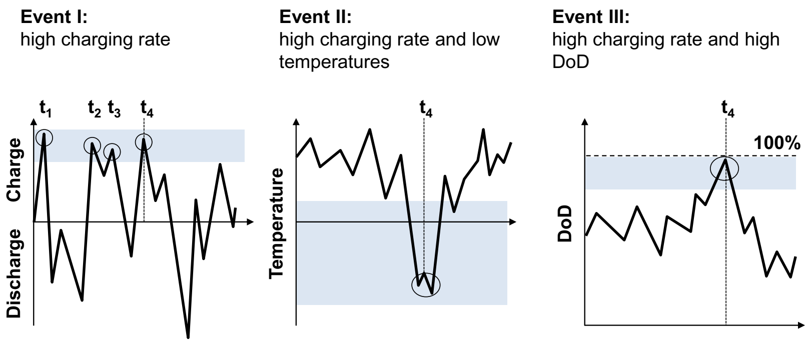

Fig. 4. Counting crucial events for lithium-plating

4.2.2. Simplified event counting

For the parameterization of the failure time models, it is essential to gather the cell data in time-consuming tests or during online monitoring in vehicle operation. A more simplified way for the first quantification of the risk of non-linear aging is just counting events like described in 4.2.1. For each load curve only critical events are counted. Afterwards it is possible to compare the batteries amongst each other and state the number of events. Furthermore, it allows counting the events for exceeding the critical limits.

4.3. Model-based approach

Finally in Fig. 5 we suggest a combination of the proposed ways with a performance based aging model. In [14] a holistic aging model is presented which is combined with an electric-thermal model and an impedance based electric model. We combined this model with our failure time model and integrated it into our battery system model to enable a variation of cell input parameters and finally quantify the overall cell spread. The data of the production induced initial cell spread is used as input data for a parametric equivalent model of a battery pack. Each cell is initially modeled with slightly different parameters as an equivalent circuit, later the parameter set gets updated by the aging model. The cells therefore experience different stresses (current, temperature) which in turn lead to different aging progresses of each cell in the system. The two mechanisms (compensating currents within parallel interconnections as well as current variations due to parameter variations which are superimposed) may lead to a self-enhancing aging progress. The resulting load spread after a passed time step is used as input data for the aging model until the end of the load history. Parallel, the output stress factors from the electric-thermal model can be used as input parameters for the counting and failure time model. This holistic model approach can simulate the capacity development of each cell in the battery system over the battery life, the according failure time and number of critical events.

These failure time points can be used to fit the statistical distribution e.g. Weibull, which represents the failure probability of the battery system. Finally, this information represents the lifetime prognosis with considering the previously mentioned uncertainties.

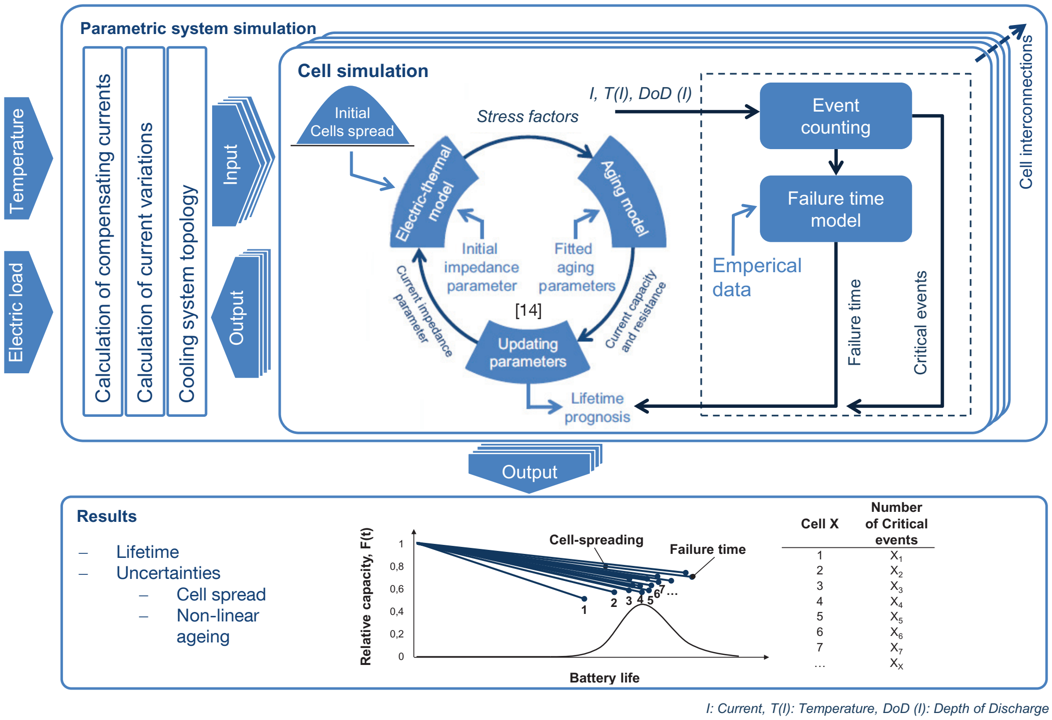

Fig. 5. Model-based approach

5. Conclusion and outlook

Cell failure and with aging increasing cell spread are highly sensitive for reusing lithium-ion batteries and thereby crucial for the sustainability of electric vehicles. To evaluate the reasonable value chains after the vehicle use and the possible economic benefit, it is required to consider the uncertainties. This article first summarizes the battery degradation and RUL modelling. Furthermore, the risks in lifetime prediction of lithium-ion cells and the sources for failure are discussed. Thereupon ways for the quantification of this risk along battery life are provided.

Although the focus of the methods in this paper are on simulation based analysis they generally can also be extended to real world automotive applications. Recording the vehicle battery's usage history is fundamental therefore and can be achieved by a server-coupled battery management system (sBMS) approach comprising the usage history in a so called Battery Pass [26]. Thereby parameters like individual cell voltages, temperatures and battery pack current are measured and characteristic state parameters like SOC are calculated by the BMS within the vehicle. This data is pre-filtered and periodically transmitted to a server platform. Via client applications the measurement data can be processed to determine the actual parameters of each parallel cell group within the battery pack and to get the data for the parametric equivalent circuit model of a battery pack. With this method it is possible to keep track of the degradation process of electric vehicles' batteries online as well as to predict its future development and the future statistical distributions within the pack.

Our results are the starting point for the quantification of risk in reusing lithium-ion batteries. This article should give a basis for discussion, how the uncertainties in RUL prediction of lithium-ion batteries can be handled and quantified. Apart from that, it helps to model the uncertainty and later to investigate the sensitivity for the economic outcome of remanufacturing or Second Life of used batteries.

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