

4th Conference on Production Systems and Logistics

Approach For Data-Based Optimization In Cell Finishing of Battery Production

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Abstract

Due to the global warming, a significant reduction in the emission of greenhouse gases is necessary. One part of the solution is the electrification of today's transportation and traffic sector. An essential component of today's electric vehicles is the lithium-ion battery (LIB), which is largely responsible for their range, performance and cost. In order to increase the use of such climate-friendly technologies, it is therefore essential to reduce the production costs of LIBs. With a duration of up to three weeks, the process steps of formation and aging are particularly capital-intensive and have high demands on storage capacities. Formation and aging therefore account for up to 30% of the manufacturing costs for battery cells. During formation, the solid electrolyte interphase (SEI) is formed, which has a major influence on the quality and lifetime of the LIB, among other things. In order to reduce production costs and simultaneously increase battery cell quality, it is therefore necessary to optimize the formation and aging process. Because of the complexity and the interdependency of these processes towards previous process parameters the application of machine learning algorithm is predestined to optimize these process steps. For this purpose, a general approach for the application of a machine learning algorithm in the formation and aging are first analysed and relevant parameters from the literature as well as reasonable assumptions about the structure are derived. Based on these requirements and boundary conditions a machine learning algorithm structure will be developed to optimize the cell finishing process in the battery cell production.

Keywords

Battery Cell Production; Data-based Optimization; Cell Finishing; Electromobility; Industry 4.0;

1. Introduction and motivation

The conversion of the energy supply to renewable energies and the increasing demand for electric vehicles are leading to a rapidly growing need for high-performance storage technologies. In this context, the lithiumion battery (LIB) has established itself as a key technology in recent years. So far, the higher manufacturing costs, lower energy and power density, and safety concerns compared to the internal combustion engine limit the widespread use of this technology. The battery accounts for the largest share of the total costs of an electric vehicle, which in turn is partially related to the battery cell production [1]. Optimizing the battery cell production process plays a key role in reducing costs since it is related to almost 20 % of the total production costs [2,3]. Manufacturers are currently attempting to reduce production costs through economies of scale, automation and digitization of production. One of the highest costs shares is created in the cell finishing with 25-30 % of the total production costs [4,5]. The process in the cell finishing are the wetting, formation, degassing and End-of-Line (EoL)-Test as shown in Figure 1.





Figure 1: Battery cell production process for a pouch cell

The wetting process is a storage phase in which the optimal wetting degree with the infiltrated electrolyte should be achieved. This can take several hours. The formation process is defined as the first charging and discharging cycle(s). This process step is characterized by very long process times up to several hours due to the formation process of the solid electrolyte interface (SEI) layer on the anode surface during the first cycles [4,6,7]. To remove the gases that have been produced in the formation, cells are degassed. Followed by a long aging step (part of the EoL-Test) which takes for larger cells up to 3 weeks and is mainly there for the quality control of the production process and especially the formation process [6]. There the self-discharging of the cells is measured. Due to other internal balancing reactions (after formation) to attain an electrochemical equilibrium, the self-discharge can only be measured correctly after a few weeks.

The SEI is an essential component of the LIB including the impact on its initial capacity loss, self-discharge characteristics, rate capability, and safety [8]. The initial SEI creation and its growth are dependent on multiple factors e.g. anode material, electrolyte, and previous process parameters [8]. Beside those factors there are also different factors in the formation process that influence the SEI layer, e.g. the formation protocol, temperature, pressure, wetting degree [9,6]. Therefore, process control and optimization are difficult to conduct due to the high complexity, strong dependency of previous production parameters and sensitivity to the process. There is no standardization regarding an optimal formation protocol. Every cell manufacturer is setting up a functional protocol and process order to fit their individual requirements. To reduce the time identifying a suitable cell finishing protocol or to be able to predict the quality earlier in the production process, research approaches are invested regarding the application of machine learning algorithm to solve those challenges. [10]

2. Technical background

A few machine learning applications and approaches do exist that deal with process optimization for a complex process system with interlinked product-quality relation in the battery cell production. These approaches mainly focus on other production processes than cell finishing. THIEDE ET AL. (2020) [11] have investigated the influence of process parameters on energy consumption of the coating plant in electrode production without considering the effect on the cell quality. CUNHA ET AL. (2020) [12] investigated the relationships between final electrode properties and production parameters of the slurry. In this study several trends between the electrode properties and the independent variables could be found. DUQUESNOY ET AL. (2020) [13] designed a hybrid model based on experiments and physics-based models together with Machine Learning (ML) approaches to determine the influence of production parameters on final electrode properties.

In regard of quality prediction, TURETSKYY ET AL. (2021) [14] developed a model for deriving necessary interim product features (IPFs) to achieve desired final product properties (FPPs). A total of 191 battery cells from the entire process chain except formation and aging were used as data for the predictive model. SCHNELL ET AL. (2019) [15] used and compared ML models to predict cell quality and identify quality parameters. STOCK ET AL. (2022) [16] analysed two algorithms regarding the ability to predict quality and cluster cells based on the results already in the cell production process.

As previously stated, the cell finishing, especially the formation and aging, have only been insufficiently researched in terms of their potential in machine learning based optimization. One challenge in the databased optimization is the high dependency of the cell finishing on multiple factors and properties. To reduce the complexity of this, three main clusters have been identified and are introduced in the following.

2.1 Material and design properties

In SHIN ET AL. (2020) [17] differences between the SEI formation, compositions and challenges in a graphite vs. Silicon containing Si/graphite anode are analysed. It was shown that the SEI and the optimal formation parameters are highly dependent on the used anode material and their properties. In Li et al. (2017) [18] the impact of different electrolyte additives on the formation and the SEI quality have been studied. It was shown that the additives not only have an impact on the SEI layer composition but also should be considered for the development of the formation parameters. In GÜNTER ET AL. (2020) [19] the impact of the cell format on the electrolyte filling and wetting process have been analysed. The results are stressing out that the electrolyte filling and wetting process as a preparation for the formation process has to be designed based on the individual cell design and format. Since the structural design and later the electrochemical reactions are dependent on the used materials, format and design, the cell finishing parameters and the detection of the quality has to be adapted to every material combination in the cell.

2.2 Process parameters before cell finishing

In LIU ET AL. (2017) [20] the porosity and film thickness of both electrodes are related to the achievable specific energy density and the capacity loss in the formation. The results show that, in general, cells with increased porosity have also a higher capacity. Increasing the porosity can improve the conductivity and diffusivity of lithium ions through the electrode. However, an optimum porosity cannot be derived because the formation of the solid electrolyte interphase also varies with varying porosity within the electrode. GÜNTER ET AL. (2019) [21] investigate the relationship between the amount of electrolyte and the maximum current rate during formation. The results show that there is no change in the increase in current rate above a ratio of 1.2 between electrolyte quantity and pore volume. Overall, it can be said that previous process parameters, e.g. the electrode manufacturing steps and the electrolyte filling, have an impact on the optimal process parameters.

2.3 Process parameters in the cell finishing

In the approach of XU ET AL. (2019) [22], an optimal multistage charging protocol for lithium-ion batteries is developed for an LFP (Lithium iron phosphate) cell using an electrochemical and thermal model. In the model, the relationships of the capacity drop due to the increase in solid electrolyte interphase (SEI) are to be minimized, the SEI potential maximized to reduce lithium plating, and the temperature rise reduced to avoid thermal runaway. The model result shows that the optimized charging current profile varies with state of charge (SOC) and cycle number. In the approach of DREES ET AL. (2021) [23] an electrode equivalent circuit model is introduced to reduce the process time of the formation time. The model optimises the charging profile of the cell by predicting the electrode voltages. In HEIMES ET AL. (2022) [6] it was shown that pressure and temperature during the formation can decrease the formation time. Different approaches already exist to develop an optimal process parameter setting in the formation, which however are limited

to the local optimization and cannot or only to limited extent integrate previous process parameters and properties in their optimization.

2.4 Research gap

There is a significant gap in terms of an universal approach for the identification of the ideal process parameter in the cell finishing or for the early quality prediction in the cell finishing due to various reaction taking place after the formation. In the next chapter, a framework is presented to describe an approach how to develop a data-based optimisation model in the cell finishing.

3. Approach and methodology

The standard process for the application of Machine Learning is the Cross Industry Standard Process for Data Mining (CRISP-DM). This approach contains six steps: Business understanding, data understanding, data preparation, modelling, evaluation and deployment. The approach was chosen because the step monitoring is not included in the CRISP-DM approach and also not considered in this paper instead e.g. the CRISP-ML or the approach of AMERSHI ET AL. (2019) [24]. [25],[26]

3.1 Business and data understanding

The first two steps, business understanding and data understanding, are closely linked and are therefore combined in the CRISP-DM process. Here, economic or research-oriented goals are derived and translated into machine learning-specific goals. In the cell finishing there are two main challenges in terms of production optimization:

- Standardized parameter definition for an optimal cell finishing process with minimum process times while maintaining or even increasing the cell quality.
- Quality prediction before or directly after the formation process for a reduction or even elimination of the EoL-Test, especially the aging process.

Both approaches could reduce the cell cost about 7 % for the optimal process parameters and 4 % for the quality prediction [4]. In Figure 2 the basic concept of the two mentioned use cases in the cell finishing are presented. In general there are roughly around 2,100 product process relation in the battery production [27].



Figure 2: Overview of the main optimization use cases in the cell finishing

But the measurable process, material and intermediate product feature do not have the same impact on the cell finishing. The mixing, coating, drying and calendaring are the main steps to create the electrode structure

and are therefore critical for the reaction mechanism in the battery cell, e.g. diffusion processes and de/intercalation processes [8],[20]. The electrolyte filling has also a high impact on the wetting time and formation results [21],[19]. The vacuum drying, stacking and contacting process have a medium impact on the cell finishing. The slitting and cutting processes only have a limited effect on the cell finishing. Therefore, the data points for the use case has to be selected.

3.2 Data preparation

The business and data understanding are followed by data preparation. In this process, data is selected, cleaned and standardized. Data that does not meet the required quality are removed during this process. Also, only input variables that have an influence on the modelling are selected. In the cleaning process, missing values are determined by, for example, interpolation or the average, and distortions are removed. In the so-called feature engineering, new variables (features) can be derived from the already existing ones. Features are divided into predictors and output factors. While predictors provide the input for the machine learning algorithms, the output is predicted by the algorithm.

3.3 Modelling, evaluation and deployment

Most machine learning algorithms can process a high number of predictors. However, most algorithms are single output algorithm and can only predict one output. For both approaches in the cell finishing, the process optimization and the quality prediction, algorithm which predict multiple targets simultaneously are necessary. There are different approaches for multi target algorithm as shown in Figure 3. In general, the approaches can be clustered into two groups: Problem transformation and algorithm adaption. The approach of the problem transformation is to reduce the problem into single output problems and solve them individually. In the algorithm adaption, single output algorithms are adapted and modified to solve a multi output problem. Those kinds of algorithm are more difficult to develop but can also consider interdependencies between the output variables. [28]



Figure 3: Overview of multi-output regression methods [28]

For the optimal process parameter development, there is a strong interdependency between the output parameter. Therefore, the algorithm adaption approach is favoured. For the quality prediction both approaches could be used. Based on the selected use case an algorithm has to be selected, modified and trained. The resulting model is assessed in the subsequent evaluation and compared with the defined success criteria. If the model does not meet the criteria, the previous steps are run through again. In machine learning, this usually involves going back as far as the modelling stage, and more rarely also the data preparation stage. If the algorithm covers the defined target, then it is transferred into practice.

4. Case study: Cell finishing

Business and data understanding: To address one of the two main challenges in the cell finishing EL-cells in a three-electrode setup were manufactured in the pilot line at the PEM. The cell configuration is listed in Table 1. The goal of the case study was to predict the cell quality based on variable process parameters. Due to the limited amount of data, the number of variables has been primarily reduced to four variables in the cell finishing. The electrode manufacturing and the selected variables for this test series have been the electrolyte amount, wetting time, formation temperature and C-rate during the charging of the cell. The formation protocol was a constant current-charging phase until 4.2 V and a constant charging-constant and voltage-discharging until 3.0 V. In total 57 data points have been generated with four variable production parameters. The range of the varied process parameters have been listed in Table 2.

	Positive electrode	Negative electrode	
Active material	NMC 622	SMG-A5	
Areal capacity	$2.37 \text{ mAh/cm}^2 \qquad 2.70 \text{ mAh/cm}^2$		
Loading density	14.4 mg/cm ²	8.8 mg/cm ²	
Thickness	117 μm	133 µm	
Current collector material	Aluminium	Copper	
Separator	20 µm Celgard 2320		
Electrolyte	1.0M LiPF6 in EC:DMC (1:1)		

Table 1: Cell configuration of the manufactured cells

Table 2: Overview of the process parameter variation range

Process	Electrolyte	Wetting temperature	Formation	C-Rate during the
parameter	amount [µl]	[°C]	temperature [°C]	charging in formation [C]
Range	50 - 100	20 - 40	20 - 50	1/20 - 1

For the quality parameter that should be predicted with the machine learning algorithm, the quality parameter of the SEI layer that develop during the first cycles are an important factor. The quality of the SEI layer can not be measured only by one quality parameter. It is related to numerous factors, e.g. initial capacity loss, self-discharge characteristics, cycle life [8]. To reduce the complexity for this study, the output variable of the algorithm is reduced to the capacity after the first full cycle.

Data preparation:

Outliers were identified and removed. For the data preparation, non-numerical attributes are converted to numerical attributes. Based on the assumption that the data are approximately Gaussian distributed, the dataset was converted into a standard normal distribution with mean value of 0 and standard deviation of 1. The quality target is transformed by Min-Max normalization to a range between 0-1. After the data preparation there were 18 observations with four features and one quality target.

Modelling, evaluation and deployment:

For the modelling the support vector regression (SVR) algorithm was selected since this algorithm also showed good results for small samples sizes compared to other algorithms [29]. The aim of SVR is the identification of a ε -intensive function. In this context, ε refers to a threshold, and the data points that fall within the ε band are considered to be accurately predicted, while those outside the ε band are not included

in the fitting of the regression. The support vectors are data points with residuals greater than the threshold, and they determine the regression line. The function for the linear SVR model is given in equation 1. [30]

$$f(\mathbf{x}, \mathbf{w}) = \sum_{j=1}^{m} w_j g_j(\mathbf{x}) + b \tag{1}$$

x: multi-dimensional input data points

- w: weights for each transformation
- b: constant term

 $g_j(\cdot)$: a set of non-linear transformation

The loss function $L(y, f(\mathbf{x}, w))$, which quantifies the error cost and is 0 for all points inside of the ε -intensive function band, is defined as seen in equation 2 [30]:

$$L(y, f(\mathbf{x}, w)) = \begin{cases} 0 & 0, \text{ if } |y - f(\mathbf{x}, w)| \le \varepsilon, \\ |y - f(\mathbf{x}, w)| - \varepsilon & \text{ otherwise} \end{cases}$$
(2)

For the training of the model 13 data points are used and five data points are used for the evaluation. This data set is split into a training set (80 % of the data) and a test set (20 % of the data). After a first cross validation hyper-tuning of specific parameters are conducted with the grid search approach. For the SVR there are five parameters C, ε and three kernel parameters [27]. The values after the parameter tuning has been set to: C = 1.707 and ε = 0.001, kernel type = radial basis function, gamma = 1 / (n_{features} * X_{var}). After the training the model was evaluated with five new data points. The results are shown in Figure 4.



Figure 4: Prediction result of cell capacity after formation (mAh) with support vector regression

The results show that the prediction based on the SVR model is always predicting a lower capacity. The strong unilateral deviation can be a sign of overfitting of the model due to insufficient data base. This applies when all data points in the training set are used as support vectors. Therefore, the model actually overfits and has a poor prediction accuracy with new data sets. Afterwards a sensitivity analysis on the impact of the varied parameter on the cell quality is conducted. It was shown that the current rate in formation charging identified has the highest impact followed by the wetting temperature. The formation temperature has a much smaller impact than the other two factors.

5. Summary, discussion and outlook

In this paper two general use cases for the application of machine learning algorithm in the cell finishing has been introduced, the optimal process parameter selection and the quality prediction. The influencing parameter has been clustered into three groups: material and design parameters, process parameters before the cell finishing and process parameters within the cell finishing. For the modelling selection and development, the challenges within the cell finishing has been analysed and the multi-output regression algorithms has been introduced as a solution for both use cases.

On a data set of manufactured 3-electrode cells the quality prediction approach has been applied using a SVR model. The focus of the varied parameters was on the process parameters within the cell finishing. The following three process parameters have been varied: the electrolyte amount, the wetting temperature, the C-Rate and the formation temperature. As the output (quality prediction parameter) the capacity of the cells after three cycle have been selected. After data pre-processing and model development as well as training the results showed that the model prediction shows in four out of five data points a strong unilateral deviation. That is probably due to the small data base. The model tends towards overfitting. A sensitivity analysis showed that wetting temperature and C-rate during the formation have a higher impact on the quality than the formation temperature. Based on the results the next steps are (i) expand the data base to reduce prediction errors, (ii) modify the model to a multi-output regression model and (iii) compare and benchmarking the model towards other algorithm approaches, e.g. KNN, decisions trees.

Acknowledgements

This work was supported by the project "FormEL" (03XP0296C) as part of the competence cluster "ProZell" by the Federal Ministry of Education and Research in Germany (BMBF). Only the authors are responsible for the content of this contribution. Additional gratitude belongs to the project partners from the elenia Institute for High Voltage Technology and Power Systems of the Technical University of Braunschweig, the Münster Electrochemical Energy Technology (MEET) of the University Münster, , the Bavarian Center for Battery Technology (BayBatt) of the University of Bayreuth, the Institute for Electrical Energy Storage (EES) of the Technical University of Munich and the Chair of Production Engineering of E-Mobility Components (PEM) of the RWTH Aachen. Further support in the experiment conduction, algorithm development and visualization were provided by Haoyu Wang and Markus Wassermann.

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Biography

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