

Electrochemical impedance spectroscopy based estimation of the state of charge of lithium-ion batteries



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ABSTRACT

The increasing market demand for electric vehicles requires a continuous development and improvement of battery systems. A crucial aspect is the application of analytical methods to characterize the battery systems in the most exact way. At the same time these methods should remain as simple as possible. The current analysis procedures use essentially the battery system- and battery cell-characteristic of the voltage curve in relaxation- and operation-time. These approaches alone are not conclusive enough due to the influences of relaxation time and varying load currents.

In recent years, methods based on electrochemical impedance spectroscopy (EIS) have found application for accurate analysis of occurring electrochemical processes and diagnosis of lithium ion batteries.

This publication aims to show how the state of charge can be predicted with the use of EIS and a simplified equivalent circuit for redundant supplementing the OCV-method (open circuit voltage) and the current- counting method. Therefore film batteries with a graphite anode and a $\text{Ni}_{1/3} \text{Mn}_{1/3} \text{Co}_{1/3}$ cathode are charged and discharged according to a specified procedure. The battery is (dis)charged with steps of 10% followed by an EIS measurement. The measured data is used to determine the elements of the simplified equivalent circuit. These elements are analyzed for their different values at various states of charge.

From these measurements can be concluded that EIS can be applied for the estimation of state of charge (in the middle range) as redundant method to other estimation methods. It therefore provides an additionally methods by which statements about the battery conditions can be made.

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1. Introduction

Electrochemical impedance spectroscopy (EIS) has been implemented through several decades to analyze and understand electrochemical processes. This method also has found application in the battery diagnosis of Lead-acid batteries [1], nickel-cadmium batteries [2] and nickel-metal-hydrate batteries [3].

EIS has been applied for some years to decipher the electrochemical processes which take place in a lithium ion battery (LIB). An equivalent circuit model (ECM) can be developed from the EIS data and the knowledge of the chemical and physical processes. The measured data are presented regularly as Nyquist-Plots (NYP). The measured impedance is decomposed with the help of a phase angle in a real part (x axis) and an imaginary part (y axis) which are plotted against each other. The real part is

designated by Z' and the imaginary part as Z'' in the Nyquist representation. Fig. 1 shows a possible equivalent circuit, which is regularly employed [2].

Additionally to the ECM shown in Fig. 1 a wide array of other suggestions exist for the interpretation of the impedance spectra of LIBs [5–8]. In different publications, the graphite electrode was examined through EIS [9–15]. The cell chemistry has a strong effect on the curve of the impedance spectrum. In [4] impedance spectra were recorded of material combination of graphite anodes and various cathode materials. The differences in the impedance spectra can be clearly seen.

Both RC-members of the ECM shown in Fig. 1 thus sum up the processes at the cathode and the anode.

The origin of the measurable capacitances lies in the solid electrolyte interfaces (SEI), which are formed on the anode (especially by graphite) as well as on the cathode. Both SEI are dielectric and function as an isolator. The SEI on the MO_2 -electrode is much thinner, than that on the anode (graphite) and dominates therefore the impedance spectrum. Since both SEI layers have

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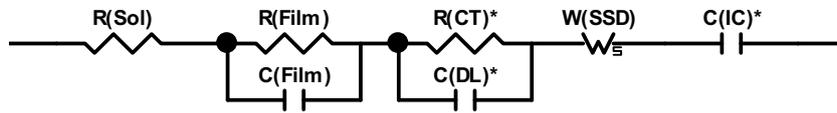


Fig. 1. Typical equivalent circuit to interpret measured spectra of impedance. It comprises a pre-resistance, two (partly up to four) RC members (film diffusion and double layer), a Warburg+ element (solid processes and diffusion) and an interphase element (intercalation). The "*" marks the circuit components that are highly dependent on potential.

similar dielectric constants, it is hard to separate them through EIS, which leads to a blurring of the impedance spectra of both electrodes. Excluded from the SEI formation is a cell structure with LTO (lithium titanate oxid) anodes because no SEI is formed on LTO anodes [33,34].

The resistance connected in parallel to the capacitances is based on the capability of the lithium ions to penetrate this layer, which can be described by a purely ohmic resistance.

Through the specification of an equivalent circuit and different mathematical adaptation methods it is intended to predict different characteristics and conditions of a LIB. This includes particularly the state of charge (SOC) as well as the state of health (SOH) [5,6,16,17,32] and the internal cell temperature [18,19]. These state variables are very hard to predict due to the complex processes in a LIB. Other possibilities to predict the state of charge are: open-circuit voltage measurement [14,19], current-counting method [14,20,21], adaptive systems such as neural networks [14,22–24], Fuzzy-Logic [14] and Kalman filter [14,24–26,30] or hybrid-systems of any methods [14,24]. Additionally, the most methods are based on very complex mathematical models. The equivalent circuit presented in Fig. 1 is too complex to be developed into applications, for example, for battery diagnosis in electric vehicles, and then built into the trip computer of an electric car. Additionally, the analysis of solid diffusion (Warburg, second to last element) and of the intercalation process (ICP, last element) requires work with very low frequencies ($\ll 50$ mHz). The measurement time is therefore too long. This would cause measurement periods of time that are not applicable for the automobile sector.

Further problems can arise at higher frequencies because the induction effects influences the abscissa zero crossing in the Nyquist plot. Whereas lower frequencies do not satisfy the requirements of a short measurement time and contains no

important information for SOC estimation. This work aims to show, that a much simpler equivalent circuit and a relatively small frequency band are sufficient to estimate the SOC of a LIB. Just for the practical implementation is the application of small memory footprint and short measurement time an important indicator for the advantageousness. However, a prerequisite is the permanent detecting of the temperature and the progressive aging because they have the biggest factors influencing the curve of the impedance spectrum, in addition to the SOC. Most of the ECMs investigated by other authors use complex equivalent circuits with Warburg impedances and constant-phase elements [2,13,17,18,27,31]. However, there are other sources which show that the simple thevenin equivalent circuit in Fig. 3 is already showing very good results [35–37]. In a review of different equivalent circuits for state of charge estimation has already been seen that a simple equivalent circuit gives good results. The model parameters are not determined on a measured impedance spectrum. They were identified with a Hybrid Pulse Power Characterization (HPPC) [15]. Another paper compares the equivalent circuit from this paper with an equivalent circuit with more RC elements. The results show that the thevenin equivalent circuit provides a similar good SOC estimation than the other models. The model parameters were determined with an adaptive gain sliding mode observers (AGSMO) for the SOC estimation [28]. In a recent publication there is a SOC-estimation with a dual neural network fusion battery model. For the linear neural network battery model based on the structure of thevenin battery model, the average error of SOC estimation is 1%. The average error is slightly less with more RC-elements. [22]

Fig. 2 shows a typical NYP, which results from a measurement between 100 mHz and 100 kHz, for the batteries used herein.

An approximately half-circular structure results through reduction of the frequency range from 1 Hz to 15,000 Hz. From this structure, a very simple equivalent circuit can be deducted. This circuit is shown in Fig. 3 and is from now on referred as –R-RC-ECM.

Three significant advantages arise through the specification of a simple –R-RC-ECM and through the measurement in a relative small frequency range:

- 1 The measurement can be performed in far less time (=5 s), because frequencies smaller than 1 Hz are omitted.
- 2 In the case of bigger batteries and battery systems, the cables only produce minimal induction effects because the maximal frequency is capped at 15,000 Hz.

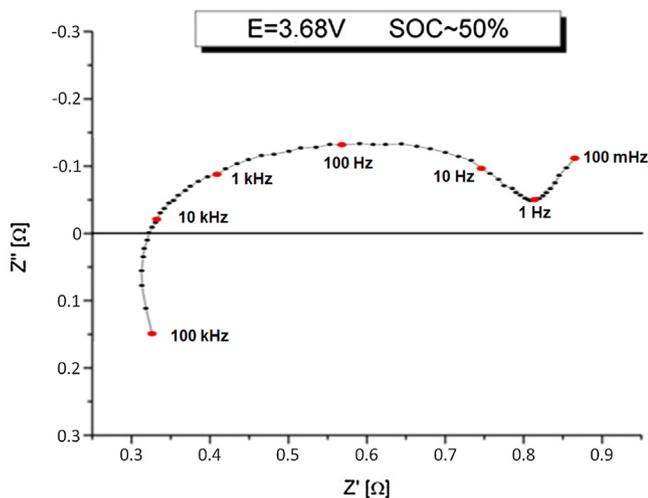


Fig. 2. Nyquist-Plot resulting from an impedance measurement between 100 mHz and 100 kHz of the batteries examined herein. The powers of ten are shown as red areas respectively. The potential E of the film battery shown herein is 3.68 V, which corresponds to a state of charge of 50%. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

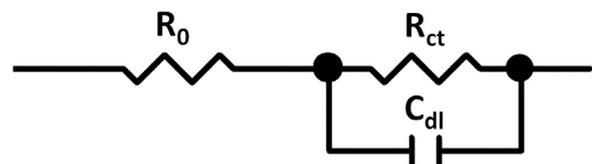


Fig. 3. Most simple equivalent circuit to describe the chemical processes of a lithium ion battery. In the Nyquist-Plot, a simple half-circle results with an additional purely ohmic resistance, that is, a half-circle displaced to the "right".

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