



## AN ESTIMATION OF LITHIUM-ION BATTERY STATE OF HEALTH – AGEING-INCLUDED MODELLING AND EXPERIMENTAL STUDIES

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### Summary

In this paper we present a study on Li-Ion Battery (LIB) modelling, including battery ageing profile, as a ground for development of state of health (SoH) prediction and monitoring system. An influence of LIB ageing was included by the mean of ageing module, based on voltage, resistance and capacity characteristics, resulting from preliminary experimental measurements. Our model, based on equivalent circuit model (ECM) and parameter estimation procedure, was implemented in MATLAB software and then validated. The simulation results are used for battery SoH estimation, which may be use in a future to optimize the conditions of LIB cycling and therefore extend the battery lifetime.

Keywords: Li – Ion battery, equivalent circuit model, state of health, ageing model

### ESTYMACJA STANU BATERII LITOWO – JONOWYCH – MODELOWANIE STARZENIA I BADANIA EKSPERYMENTALNE

#### Streszczenie

Niniejszy artykuł przedstawia badania nad stworzeniem modelu baterii litowo – jonowej, uwzględniającego procesy starzenia baterii, jako podstawę do budowy systemu monitorowania stanu baterii. Wpływ procesów starzenia został uwzględniony za pomocą specjalnego modułu, zdefiniowanego przy pomocy parametrów (tj. charakterystyki napięciowe, impedancyjne, pojemnościowe) uzyskanych na drodze pomiarów eksperymentalnych. Opracowany model, oparty na równoważnym dwugałęziowym modelu elektrycznym RC, został zaimplementowany w środowisku MATLAB, a następnie poddany walidacji. Wyniki przeprowadzonych symulacji wykorzystano do opracowania algorytmu estymacji stanu baterii, który w przyszłości może posłużyć do optymalizacji warunków pracy ogniw litowo – jonowych.

Słowa kluczowe: baterie Li – Ion, model ECM, monitorowanie stanu baterii, starzenie

## 1. INTRODUCTION

One of the most demanding world energy challenge is focused on increasing production of power, derived from sustainable energy sources. Using an ambient energy sources, instead of a burning fuels, seems to be a smart idea, which may help produce cost-effective and environmentally-friendly devices. Unfortunately, since the abundance of sustainable energy sources is fluctuating through a day, an application of this sources requires an accompanying solutions for energy capturing and storage [1]. For now, the most common solution involves an application of Lithium-Ion batteries (LIBs), used for a broad range of electronic products for new energy technology purposes (i.e. electric vehicles). High energy and power density, low self-discharge rate, relative long lifetime and safety, caused an enhanced interest of the companies, which willingly taking the advantages of this attractive energy solution. Taking

together, it seems that energy storage systems, based on LIBs, established themselves as a leading candidate for the next generation of automotive, aerospace and other energy demanding applications.

Unfortunately, the LIB during its performance is subjected to degradation process. Moreover, the rate of degradation depends on operating environment factors, therefore it may be different for the same cells working under vary conditions. In general, the LIB ageing is caused by the layer of solid electrolyte intercalation interphase (SEI), which overgrowth leads to significant reduction in LIB performance or may even cause a battery failure. From this point of view, there is an undeniable need to estimate the state of health (SoH) of battery, in order to predict the number of cycles left to the end of its lifetime.

In the recent years, an extensive studies have been carried out to find a satisfactory way of battery SoH estimation. Among many reported studies, the most promising one is concern with the theory of

crack propagation on the battery electrodes, which may reflect in the nonlinear relationship between cycling stress and temperature [2]. Regardless of the research methodology, the general idea of SoH is based on comparison of results of LIB ageing model simulations with experimental measurements. If the real-time measured data are convergent with simulation results, the battery health may be assessed as normal. If not, it means that some failures occurred. An applied model should also include all electrical parameters of the circuit load and other parameters i.e. representing charging and discharging characteristic.

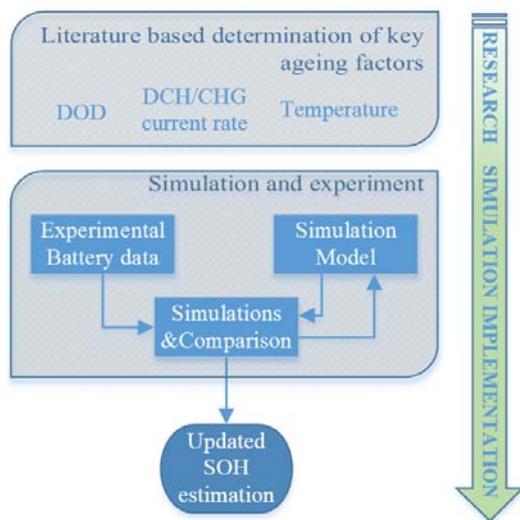


Fig. 1. The sequence of presented study

The aim of our study is to develop a system for LIBs state of health prediction and monitoring, in order to attempt an extension of LIB life, avoid unexpected failures and improve the control of power management systems. Also, the precise definition of ageing model may help to find the most efficient conditions for long-term LIB operation. In this paper we propose a method, where ageing parameters are applied to the equivalent circuit model (ECM) structure. Our simulation model, implemented in a MATLAB software, is based on the 2RC branch ECM and on the parameter estimation procedure (which provides a circuit parameters, allowing to recreate a load dependences on SoC, voltage and temperature) [3]. A developed ECM model is subsequently combined with ageing module. As a response for constantly changing initial ECM parameters, the ageing module is adjusting during the LIB operation, provides an estimation of the left LIB lifetime. The simulation results are compared with experimental measurements and, after model validation, are used for further SoH prediction (fig. 1.)

## 2. THE AGEING OF LI-ION BATTERIES

The ageing of materials and structures, is an extremely important aspect, which should be included in most of the type of condition and health monitoring studies [4–6]. The degradation issue may be considered through a different scale, starting with an atomic scale [7], through a micro [8], up to the macro scale [9]. Regardless the scale of the problem, as well as the materials and methods under investigation, the major idea of all studies is the same – attempt to assess how hard degradation affects on the material properties and, as a result, on the construction/device lifetime. The same problem relates Lithium-Ion Batteries (LIBs) – during LIB performance, each component of the cell is subjected to ageing, therefore the LIB efficiency during its lifetime is decreasing. A possible mechanisms of degradation are different for different LIB components – the current collector is subjected to corrosion, the electrolyte to decomposition, the separator usually melts and the cathode undergoes metal dissolution [10]. However, in general is assumed, that the most significantly differences in ageing mechanisms are noticeable between the anode and the cathode.

The ageing of anodes is usually concerns with the changes in graphite structure (carbon is a commonly choice for LIBs anodes), causing deterioration of the anode properties in time and use. The changes reveal during battery cycling, as well as during storage, and might be assessed by well-defined electrochemical parameters i.e. state/change in state of charge (SoC/ $\Delta$ SoC), state of health (SoH), capacity fade or resistance rise [11]. Except changes in anode structure, the adverse effects of LIB ageing may also concern changes at the Solid-Electrolyte Interphase (SEI) level. In fact, the studies of ageing influence on the SEI layer are one of the most commonly reported studies concerning LIB batteries – the examples may be found in [12–15]. Also, in case of metal-cored electrodes, the LIB ageing effects may result from mechanical disintegration within the electrode – some details about changes in anode volume, reactivity of binder materials or current collector corrosion are reported, respectively, in [16–18]. On the other hand, the ageing of the entire LIB may partly results from cathode ageing. The inseparably conjugated phenomena, such as structural changes of positive active material, dissolution reaction or modification of surface film, have an irreversibly influence on the LIB lifetime. Therefore, a studies in a field of cathode ageing, are mostly focused on cathode structural aspects [19], ageing neutralization [20] or surface electrolyte reactions (including gas generation mechanism) [21], [22].

An additional information about causes, mechanisms and possible effects of LIB ageing are described in great details in overview available in [23]. Also, a comprehensive review on LIB ageing

mechanisms, supplemented by estimations for automotive applications was reported in [24]. Finally, in a reference to the aim of our study, one of the latest extensive review about state of health estimation methods for LIB, may be found in [25].

### 3. PRELIMINARY MEASUREMENTS

#### 3.1. The object of a study

The object of our study were Lithium-Ion batteries (LIBs) from Sony Energy Devices, model US18650V3. This model, due to its high capacity (in relation to its weight) and relatively small dimensions, is commonly used in many applications. The nominal voltage of this battery is estimated at 3.7 V. Other key parameters of the cell are presented in table 1.

Tab. 1. US18650V3 key parameters

Product Category	Li-Ion rechargeable battery
Model name	US18650V3
Nominal capacity	2150 mAh
Nominal voltage	3.7 V
Lower/Upper cut-off	2.5 / 4.2 V
Work temperature	-10°C ... 50°C
Materials & Ingredients Information	
Cathode	<ul style="list-style-type: none"> <li>✓ Lithium Nickel Cobalt Manganese Oxides (active material)</li> <li>✓ Polyvinylidene</li> <li>✓ Graphite (active material)</li> </ul>
Anode	<ul style="list-style-type: none"> <li>✓ Graphite</li> <li>✓ Polyvinylidene Fluoride (binder)</li> <li>✓ Organic Solvent (non-aqueous liquid)</li> <li>✓ Lithium Salt</li> </ul>

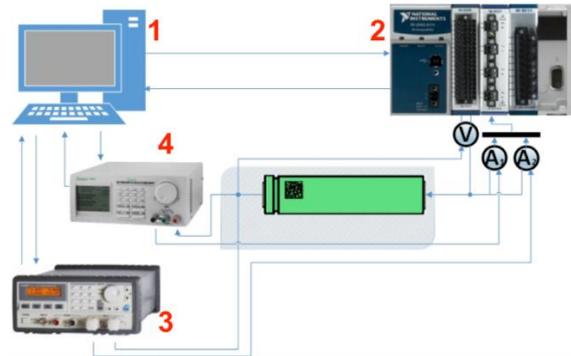
Among materials used in considered cells, an important role plays Lithium Nickel Cobalt Manganese Oxides (NMC). A NMC cells are commonly in battery – they could be found in electric vehicles, e-bikes, various power tools and similar mobile devices. The NMCs have a good performance, excellent specific energy and the lowest self-heating rate from set of well-known battery [26], [27].

#### 3.2. Measurements on LIB performance

Preliminary measurements of battery cycles (charging and discharging) were performed in order to investigate an actual state of batteries. As a result we obtained a voltage, temperature and resistance characteristics, which were used to calculate the ageing factors for development of simulation model. Moreover, the results would help to verify the general correctness of implemented program and find parameters for its further improvements.

#### 3.2.1. Measurement circuit

The scheme of measurement circuit is presented in fig. 2. In order to provide stable and constant temperature of surrounding we applied an environmental chamber. Due to the different nature of the expected temperature fluctuation (the changing temperature of cell as well as the temperature of environment) we applied 2 thermocouples sensors. Applied measurement devices provided appropriate high frequency data acquisition, what improved correctness of measured magnitudes.



1. Computer with battery-testing program in LabView
2. Measurement card cDAQ battery from National Instruments with additional modules
3. Programmable electronic load from Array 3710A
4. Digital switching mode regulated power supply Manson SPD2210

Fig. 2. A measurement circuit – scheme

#### 3.2.2. Measurement algorithm

The measurements were performed on a single cell, at the constant temperature conditions (23°C). First, each lithium-ion battery was treated by nominal magnitudes of voltage and current, provided by cells manufacturer to improve propriety of performed model. Each run included a charging of cell by constant current (CC) – constant voltage (CV) approach and discharging with a 1C rate by constant value, with a 1C rate, down to the cut-off value. During charging state, the cell was powered by a constant value of current, until it achieved a charge voltage equals 4.2 V. Then the current fell, while the voltage was maintained at the same value (4.2V). The cell was fully-charged when cut-off current fell under determined value. The details of charging and discharging dependencies are presented in tab. 2.

Tab. 2. Charging and discharging details

Charge Voltage	4.2 V
Charge Current	2.15 A
Cut-off current	0.1 A
Discharge Current	2.15 A
Cut-off voltage	2.5 V (0% SoC)
Environment temperature	23°C

Since, the process of lithium-ion batteries testing is time-consuming, the progress in measurements for 1C rate was strictly determined by imposed conditions. In order to accelerate the process of cycling, we applied discharging with 2C rate in the same temperature of 23°C. The comparison between cycles with 1C and 2C rate of discharging is presented in fig. 3. At the end, the program of battery cycling was closed in the loop and repeated as many times as it was possible (approx. 1500 cycles).

### 3.2.3. Results

All acquired data were saved, resampled and filtered, in order to calculate an average cycle parameters. Based on that, we performed an average cycle for which we obtained all needed characteristics.

The most valuable results come from current characteristic of average cycle. Based on discharging stage in cycle of lithium-ion battery, it is possible to calculate the noticeable magnitude of battery output, which has great impact on cell performance. This parameter is called the capacitance and may be calculated from current line by reading further samples and adding them together. In addition, the response of tested cell on determined environment conditions and charge/discharge approach, gave us the voltage characteristic. Based on the drop of voltage we were able to calculate the internal cell resistance, which directly describes the energy wasted in form of heat generated and emitting during process. The dependency between determined current and battery response during one absolute cycle is presented below. The measurements on the single cell also caused its temperature changes, what may be observed in fig. 4. During discharging the temperature was rising rapidly and the emitted heat reached the peak (approx. 42°C) when the voltage achieved cut-off value (0% SoC). A measured voltage equal to 2.5 V triggered a 120 seconds of stabilization step and after which the charging stage is coming. Thus, an initial magnitude of voltage, measured after stabilization, is equal to 3.2 V.

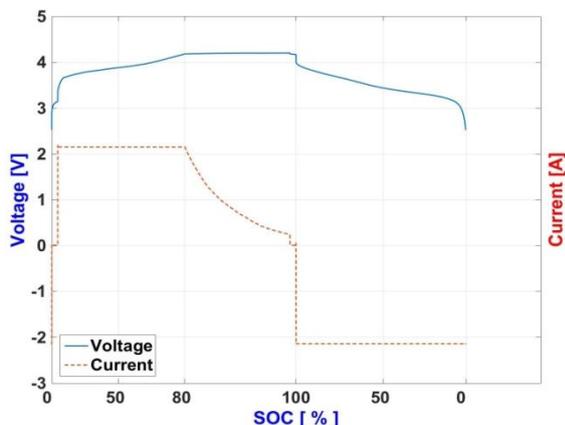


Fig. 3. Voltage characteristic for 1C charge and 2 C discharge

The battery temperature have not hesitate so rapidly as the voltage, and it was estimated on 40°C. Then, the temperature fell down, despite the fact of applying a constant current (when charging with high power). The temperature was established when the constant voltage stage came and the current started to fall. This, according to Joule Law, was the reason of lower heat emittance. Based on temperature changes it is easy to predict a value of an energy wasted (emitted as a heat) during cycle. The similarity of temperature characteristics for several different runs (fig. 4), clearly shows that conditions of the battery performance were approximately the same. Moreover, if the cell demonstrates comparable response for each cycle, it may be considered as a validation of this factor for other defined conditions.

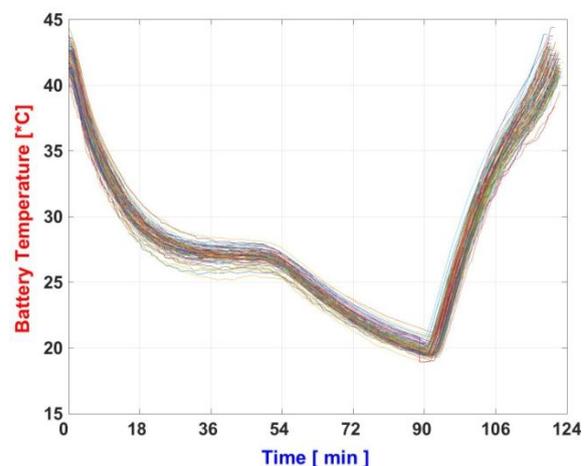


Fig. 4. Battery temperature characteristics

Based on the simulations of the battery lifetime, assuming 1C-rate charge and 2C-rate discharge, the degradation factor was calculated in common of capacitance fall (assuming starting point as a 100% SoC). For the first few hundred cycles, the drop in degradation factor seemed to be unnoticeable, thus the value of SoH parameter was close to a 100%. A 5% fall of SoH value was noted around 1000<sup>th</sup> cycle. After that, the rate of SoH drop was accelerated, reaching a 80% of SoH about 1500<sup>th</sup> cycle. However, due to the smooth character of the curve presented in fig. 5, there was no specific number of cycles which might be considered as a breakthrough in SoH decrease.

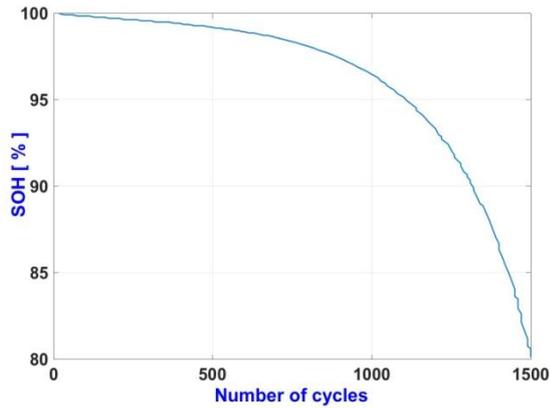


Fig. 5. Decreasing trend of SoH, as a response to LIB capacity fall

An internal resistance directly defines the efficiency of a single cell, which internal parameter allows for current flow. The fact is also, that a lower internal resistance results in a higher applicable C-rate of battery discharge, which has a great impact on LIB efficiency. Figures 6 presents a voltage characteristics of pulse discharge, with a 1C and 2C rate respectively. Test runs of pulse discharge (from 100% - 0% SoC) were performed in room temperature (23°C) with load time equals 180s and stabilization time about 21 minutes. Following this scheme, a discharging could be performed exactly with 20 pulses (for 1C rate) and 10 pulses (for 2C rate).

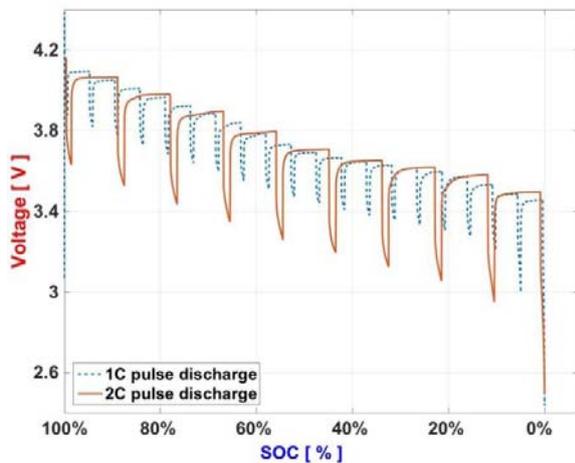


Fig. 6. Voltage characteristic during pulse discharge by 1C/2C rate

It is worth to mention, that temporary increases in voltage, observed in fig. 6, are resulting only from stabilization of the cell. The speed of voltage increases, occurring after load stages, directly defines both: the resistance and parameters of equivalent circuit model (ECM). Moreover, the speed of stabilization characterizes individually each type of cell and precisely defines its performance, understood as its capability to return to initial conditions.

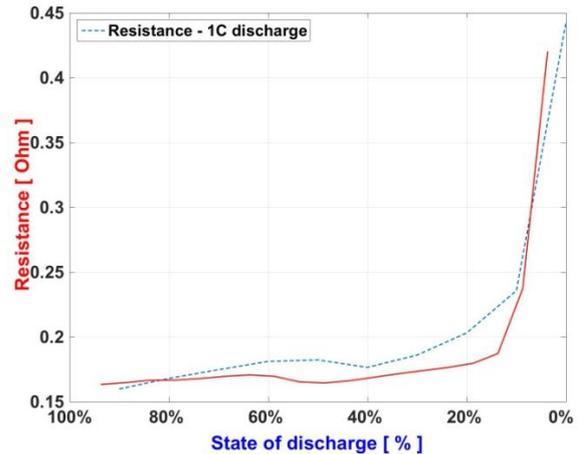


Fig. 7. Resistance characteristic during pulse discharge by 1C/2C rate

The pulse discharge approach allows to determine some parameters for a single cell. For example, based on the ratio of the first voltage value before loading to the last value during pulse discharge, it is possible to calculate an internal resistance during this short period of time. A resistance characteristics, corresponding with voltage characteristics from fig. 6, are presented in fig. 7. The comparison between resistance characteristics (fig. 7) shows that higher current causes an earlier rise of resistance. In the end (10% SoC) of both, 1C and 2C rate cases, a resistance rapidly increases, what also causes a higher voltage drop and the temperature rise. Therefore, discharging battery down to 0% of SoC is unprofitable – it generates unwanted heat and also accelerates an ageing.

#### 4. A LITHIUM-ION BATTERY MODELLING

The major problem with existing battery models is, due to imposed simplifications, they do not provide a prediction of battery cycle life. On the other hand, a battery cycle life has strongly nonlinear dependency with the depth of discharge (DOD) parameter, which may be easily determined by empirical battery testing [2]. Because in already reported models, there is no leading nonlinear mechanism for this damage parameter (as a function of DOD), the resulting loss of capacity is assumed as a linear function of charge throughput.

In order to prepare an accurate prediction of battery degradation, it is desired to apply electrochemical models. In our studies, electrochemical nonlinearities has been recognized as a set of functions, depending on the battery usage profile. The parameters value for ageing function were taken from a preliminary measurements (section 3) performed in a various conditions as well as from the studies reported in [28]. Referring to model reported in paper [17], we combined two aspects of Li-Ion cell modelling and simulation – battery ageing and the prediction of battery behavior in working system. The former aspect was considered through

a electrochemical model, the latter through an equivalent circuit model (ECM). A possible inputs and outputs for our model are summarized in a table 3.

Tab. 3. Possible inputs and output of applied model

Input type	Input parameters	Output parameters	Usage
Measured signal	Load (A) Time (s) Cycles done (default 0)	Voltage response (V) SoC (%) SoH(%) SoH (cycles left)	Verification of ECM and ageing model
	Usage DOD (%) Charge current(A) Discharge current(A)		
Constant signal		No. of equivalent cycles Temperature change °C	Checking how certain parameter affects on battery lifecycle

#### 4.1. An ECM-based model

A program, developed in MATLAB® environment, was capable to estimate SoC and SoH while reading in the real-time discharge and charge battery parameters and use test battery profile simulation as well. Data could be provided from the preliminary measurements (section 3) or from already reported practical applications. The damage parameters were applied to the empirical equivalent circuit model (ECM) structure based on approach developed by Huirra et. al [3]. Ageing parameters were derived from review article, available in [28], and then fine-tuned to suit measured data from multiple cycles.

Thermal model [26] was used to model the battery temperature. The cooling of the cell was assumed through the convection and the heating came from internal resistance. Created model of a single cell was multiplied and connected in order to achieve a battery pack characteristic. Figure 8 presents a general structure of developed MATLAB Battery Model.

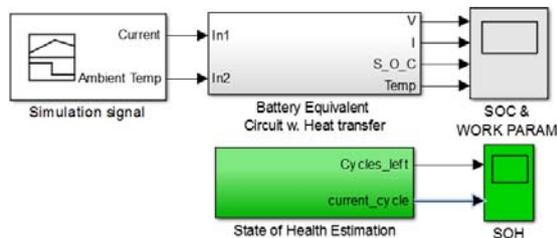


Fig. 8. Scheme of implemented battery model

Simulation model for estimation of SoC of a lithium-ion cell was implemented in MATLAB as a Simulink ECM block diagram. A scheme of applied equivalent circuit model with two RC branches is presented in fig. 9. ECM was combined with ageing module, which during battery lifetime, was able to

adjust model parameters ( $R_1$ ,  $R_2$ ,  $R_3$ ,  $C_1$ ,  $C_2$ ). The initial values of ECM elements were based on model developed by Huirra et al. [3] and during simulation were constantly altered. The parameters fine-tuning used an estimation procedure ([3], [29]), which allowed to simulate load dependencies of ECM components (implemented as lookup tables) on battery SoC and on battery voltage. Using this approach, we were able to include a needed compensation, resulting from an adverse changes in battery capacity.

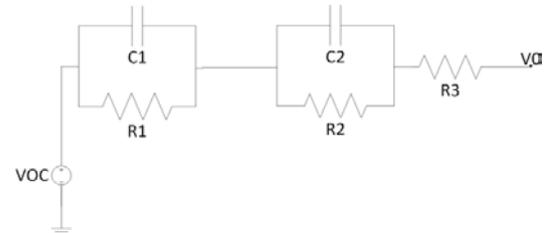


Fig. 9. Equivalent circuit model (ECM) diagram

#### 4.2. Model validation - simulation results

A validation of the ECM model and degradation prediction algorithm, was performed by the mean of series of simulations. The general information flow during validation is presented in fig. 10.

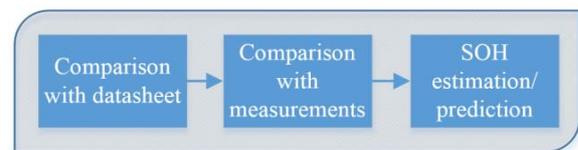


Fig. 10. The sequence of simulation

In order to check and adjust model parameters, each simulation was performed with a conjunction with an appropriate experimental test. The proper model adjustment was necessary to prepare satisfying characteristics of charge/discharge before simulation of battery degradation.

To validate the parameters of ECM, we run a set of simulations of discharge employing different load values. The results are shown in figure 11. We also performed a simulations of discharge for various temperatures, in order to provide a data for heat transfer and voltage/capacity temperature influence validation. A discharge voltage characteristics for three different environment temperatures are presented in fig. 12. In both case, all obtained results stay in agreement with discharge load/temperature characteristic data from battery datasheet. Therefore we concluded, that our model correctly reproduces the behavior of real battery cell and might be used for future comparison with experimental data.

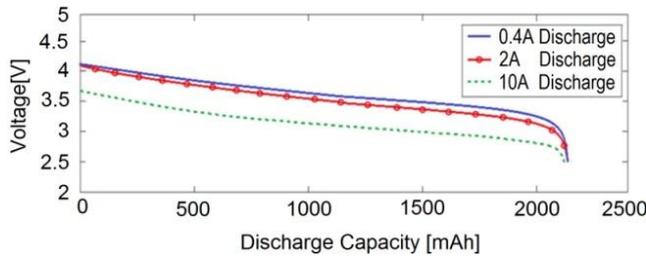


Fig. 11. Load dependency on LIB discharge curves

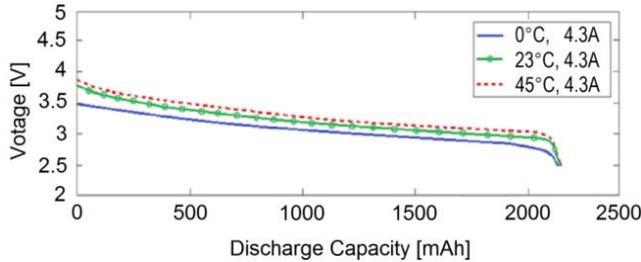


Fig. 12. Temperature dependency on LIB discharge curves

Due to the ECM validity, we were able to initiate the state of health (SoH) estimation algorithm. The results of multiple simulations were prepared for further comparison with the long-run experimental tests on real LIBs. In case of detection of any difference in future, the changes will be thoroughly examined and the code for LIB ageing will be improved.

## 5. STATE OF HEALTH MONITORING (SOH)

The idea of assessment of SoH is based on model-assisted approach to diagnostics [30]. According to this idea, the simulation results are compared with experimental results at each cycle of battery charging/discharging, and the correlation between model and experiments is tested. If the correlation is relatively small, the model is updated at each cycle. Updated model is used for further prediction of remaining battery life. Additionally, the difference between experiments and simulations may be analyzed and thus help with SoH assessment.

As long as the current SoH of the Lithium-Ion battery can be estimated with methods based only on present data, the accurate lifetime estimation relies on the precision of the ageing model. The main factors, like operation temperature, depth of discharge (DoD) or current rate [31] which influence battery ageing (and cause a fade of capacity), should therefore be deeply discussed and included into a battery model.

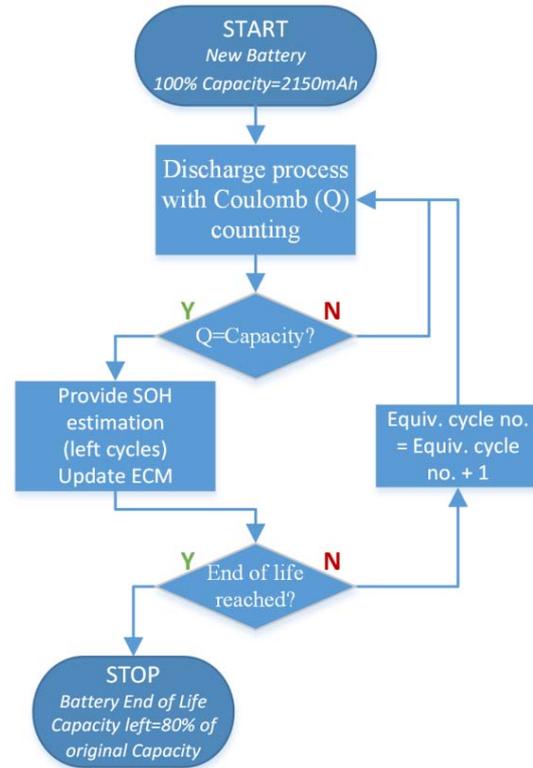


Fig. 13. A block diagram for SoH estimation algorithm

### 5.1. SoH estimation – algorithm

SoH parameters estimation is a subsystem, using ageing function able to estimate a number of full cycles (in 20°C) which left, until the battery reaches assumed level of original capacity (usually 80%). Our SoH simulation model includes so-called *cycle equivalent counter (Coulomb counting)* which allows to consider one battery cycle as a sum of consecutive partial discharges, performed until the loss of total charge ( $Q$ ) is equal to the battery capacity (fig. 13). When this condition is satisfied, the equivalent cycle number increases and mean values of discharge current, as well as a mean temperature during equivalent cycle, is calculated. The results are then compared with battery look-up tables and also with ageing parameters based on experimental measurements. Therefore, after each full cycle, the SoH (number of left cycles) may be estimated and provided together with the correction of ECM parameters. A block diagram, for SoH estimation algorithm described above, is presented in fig. 14

### 5.2. SoH estimation – results

The results of SoH estimation provide a number of left full cycles, determined after each full discharge. A minimum SoC and temperature range, occurring during each iteration, is used to compute ageing and capacity loss. Simulation model uses a look-up tables to store ageing factors, as well as a function derived from experimental measurements,

for calculation of SoH changes. In order to provide a practical guidance for a LIBs users, we also create a spatial graph of usable energy. A 3D surface, presented in fig. 14, shows conjugated temperature and DoD dependence on total useful battery capacity.

According to our experimental measurements, the best working condition for tested LIB cell are: the temperature range between 25 – 40°C and minimum value of SoC between 30 – 60%.

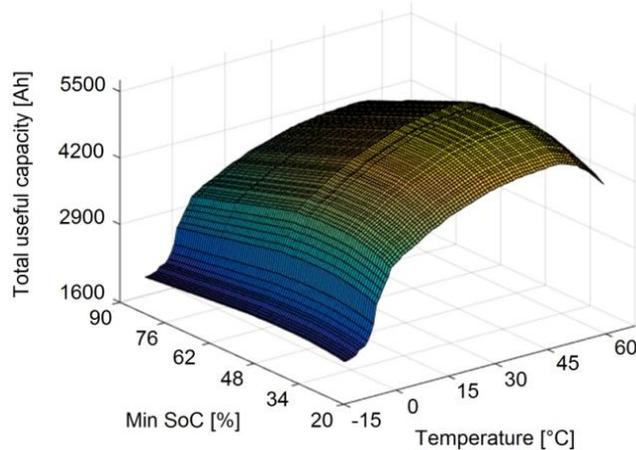


Fig. 14. DoD and temperature dependence on total useful capacity of LIB

An influence of current load dependency on ageing is assumed as a linear function, defined on the 1C and 2C-rate measurements (section 3), where 1C charge/discharge run was taken as a reference point. Constructed function (fig. 15) was added to the SoH estimation function. For future purposes, more accurate and representative function, should be based on the wider range of current load dependencies.

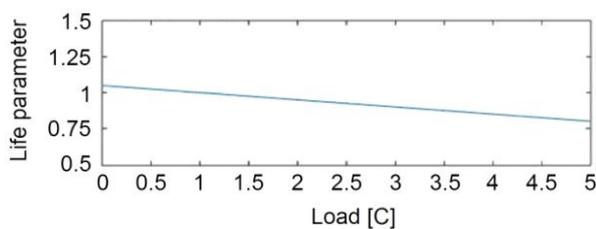


Fig. 15. Linear function of load dependence on LIB ageing

## 6. CONCLUSIONS

The health monitoring is constantly evolving branch of modern engineering. Today, there is plenty of research, using health monitoring approach, in order to develop the real time assessment of state of vary objects – starting from small mechanical components (i.e. bearings), ending with large scales constructions (such as bridges or buildings). The health monitoring may also be

applied to estimate the state of single energy devices (batteries) or even entire power management system.

In our study, we applied the state of health monitoring, to develop a method for prediction of a lifetime for lithium-ion battery (LIB). For this purpose, we developed a computational model of LIB, based on the 2RC branch of equivalent circuit model (ECM). The ECM model was extended with thermal influence module and ageing function, including adverse effects of LIB degradation. The definition of ageing function was supported by experimental measurements, providing all needed parameters and characteristics. However, such of determination of ageing influence, may be not satisfied the needed accuracy for results of simulation, and thus may mislead the validation. Therefore, the accuracy resulting from our model, may vary, when input parameters are going beyond a standard usage range. It also remains an open research question: how properly define a LIB model including all internal (material, structural etc.) and external (temperature, input/output parameters) influences, with required level of accuracy? Nevertheless, we believe that our model, as a conjugation of existing models with preliminary measurements, is able to give a fuller description of LIB performance and form a solid ground for future development of more comprehensive state of health monitoring solutions.

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