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Direct determination of a single battery internal resistances distribution using a heterogeneous model

Maxime Juston · Nicolas Damay · Christophe Forgez · Stephane Vivier · Karrick Mergo Mbeya · Bogdan Vulturescu · Guy Friedrich

Abstract Lithium-ion batteries are getting larger due to the expansion of transportation and mass storage markets and they can now contain up to thousands of cells. However, a sole damaged cell can significantly impact the whole battery pack efficiency [1]. Thus, the diagnosis of a single cell remains critical for those systems. Many methods exist [2, 3] in which the cell is considered homogeneous. We recently developed a heterogeneous equivalent circuit model that considers a distribution of internal resistances to better represent a real single cell behavior [4,5]. This resistances distribution (RD) may bring valuable information about a single cell internal quality, but only if it is determined with a sufficient accuracy. In this paper, we propose an algorithm that allows a responsive determination of the RD. The results are compared to other determination methods. This resistances distribution (RD), which is determined thanks to the preliminary construction of a homogenous model and a single discharge, is also valid for other operating conditions. This proves the relevance of the determination method and it should now be usable to detect abnormal evolution of the RD during a single cell lifetime. Although this work is developed for a single cell, it can also be used for several cells connected in parallel and may thus be used to detect a damaged cell inside a battery pack.

1 Introduction

Lithium-ion cells are becoming one of the best solutions to store energy in a wide range of applications, in particular among systems where weight or volume are major constraints, such as drones, cars and trains. In the industry, this technology is often not well known and it is to our advantage to develop a simple yet robust model that can be used in a wide range of operating conditions. In order to anticipate the voltage response of a cell to a current profile, model can be used to simulate its behaviour. An option is to build an electrochemical model that takes into account the chemical reactions and other deep level reactions [6–9]. However, the complexity of these model and the number of parameters needed do not suit the expectations of most end users. Another option is to use a mathematical model such as neuronal networks or fuzzy logic [10, 11, 2]. One last option is to use electrical equivalent circuit [12–15]. Although these models are easier to parameterize, their parameters have to depend on current, temperature and State of Charge (SoC) to bring accurate predictions. As a result, those models usually require lookup tables that require many measurements to be completed. These models are useful to predict a cell performance in many operating conditions in order to optimize its sizing, choose an appropriate cooling system or predict its capacity to fulfill a mission.

A previous research [12] led to an homogeneous equivalent circuit of a Ni-mH cell, built from a physical basis. This model only takes into account the main phenomena that occur inside a cell, with the two electrodes behaviours being mixed together. Recently, the usual homogeneous model was extended to a heterogeneous one, the so-called "multi-bunch model" [4]. It aims to model the heterogeneity of the cell through a distribution of one or more parameters. The origin of those distributed parameters can have many sources that will be listed. This model is more accurate than

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an homogeneous one and although it may appear more complicated at first sight, it is faster to characterize than a usual electrical equivalent circuit model. Actually, we found that the addition of the heterogeneous behaviour representation in the model makes the use of SoC-dependant parameters not necessary anymore (except for the open circuit voltage). This allows us to characterize our model around 10 times faster than other usual electrical model that have a precision of 10% in SoC. Once characterized, our model is able to simulate the cell over its whole range of operation. To characterize the heterogeneity of a cell also brings valuable insight into a cell behaviour and internal equivalent properties. We also expect the resistances distribution to change during the battery lifetime due to aging, and one of our goals behind the scope of this paper is to track these changes to identify the mechanisms behind them. Problems related to the determination of the distribution will be presented and our solutions explained.

In this paper, we demonstrate that the measurement of a single discharge, used to build our model, allows us to simulate the cell in a different operating conditions. In section 2 we present the construction of the model, its parameters and variables. Our experimental setup and the used cell are presented in section 3 while section 4 describes the simulation and optimization algorithms. Discussions about the obtained results are in section 5 and last, the conclusion and perspectives of this study are presented in section 6.

2 Model and parameter dependency

2.1 Construction of the multibunch model

Our model is based on the assumption that one cell can be divided into elementary volumes that behave homogeneously and have their specific electrical properties. Those volumes are connected in parallel, as showed in Fig. 1.a. We assume that certain volumes, not necessarily spatially bounded, have close electrical properties when compared to each other. These volumes are thus grouped into a "bunch" that is considered homogeneous. By repeating this operation, the cell is discretized into n homogeneous parts having distinct electrical properties, see Fig. 1.b. The origin of those distinct electrical properties can be the position of the tab within the cell [16], the distance of active material to current collector, temperature gradient. The bunches have a local current I_i but the same voltage: V_{cell} . The sum of the local currents I_i is the cell current: I_{cell} . Let n be the number of bunches of our model, sorted in ascending order of resistance value. This idea of variation of electrical parameters within the cell is supported by the recent work of Park et al. [5], who found that a resistance distribution is found by scanning the surface of an active material sample.

Each bunch is modelled using an electrical circuit. The proposed representation comes from the previous article from Damay et al. [17] in which the hypothesis made are the following:

- The double layer capacity can be neglected because of its fast dynamic and the fact that our tests are conducted at constant current during thirty to sixty minutes. We call "high frequency" resistance R_{HF} , the sum of the so called ohmic resistance and the charge transfer one.
- In order to reduce the computation time, the Warburg impedance representing the diffusion phenomenon is modelled by a single RC. More RC circuits can be added to better model the diffusion [12, 13]. The subscript d under those elements stands for diffusion.

Those two hypotheses lead us to the multibunch dynamic model used in this paper, presented in Fig. 1 : one voltage source, one "high frequency" resistance and one parallel RC.

2.2 Parameters values, dependencies and number of bunches

The local voltage source values $U(Q_i)$ varies with the local charge like an increasing and nonlinear function, following the open circuit voltage (OCV) of the cell. The bunch charge is defined by its previous state and the bunch current by

$$Q_{i,t} = Q_{i,t-\Delta t} + \Delta t \cdot I_{i,t-\Delta t} \quad (1)$$

To simplify our model, all the heterogeneity of the electrical properties is concentrated in the values of the $R_{HF,i}$. We expect this parameter to be the most heterogeneous one in a cell due to variations in the impedance of the current collector, electrolyte and charge transfer. This heterogeneity is represented with varying values of R_{HF} from one bunch to another. The determination of the distribution of the R_{HF} is done either by searching a mathematical distribution (a Weibull one), or by searching directly values fitting the experiments. Differences between both methods will be discussed later. Values of the distributed parameters at different temperature and current are computed as follow:

$$R_{HF,i} = R_{HF,i}(50\%SoC, 1C, 25^\circ C) \cdot \beta(I, T) \quad (2)$$

$$\text{where } \beta(I, T) = \frac{R_{HF,meas}(50\%SoC, I, T)}{R_{HF,meas}(50\%SoC, 1C, 25^\circ C)}$$

Because our model does not need a SoC dependency, we determine the electrical parameters values at only one SoC (50%), one current (1C) and temperature (25 °C). All the other values of the parameters are determined by multiplying the measured value with a coefficient that is extracted from dependencies lookup tables. The determination of the lookup tables for dependencies are detailed in the article

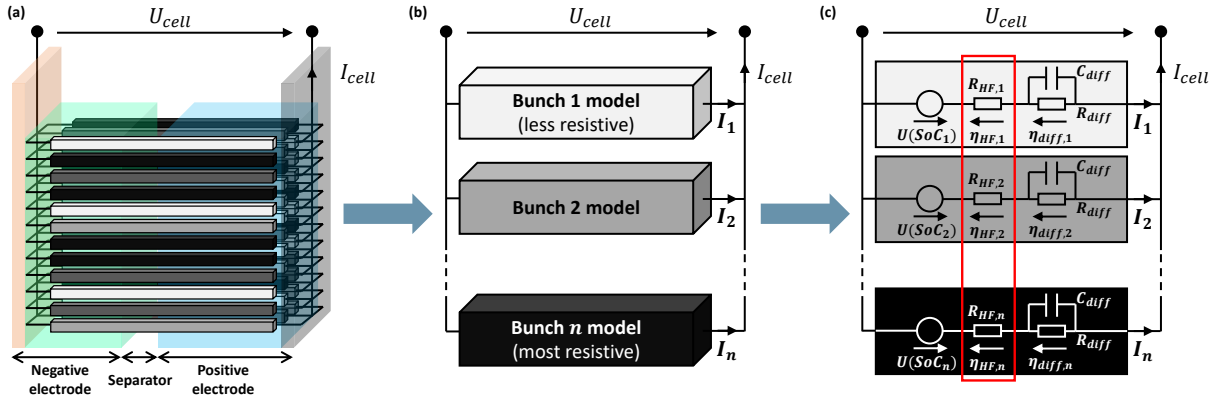


Fig. 1: (a) Division of the cell in a high number of electrical isotropic volumes. With (1) negative current collector, (2) negative active material, (3) electrolyte, (4) positive active material, (5) positive current collector. (b) Model with grouped homogeneous volumes into bunches. (c) Multibunch model with n bunches.

from Damay et al [4]. Conversely to the R_{HF} values, the values of R_d and C_d are the same for each bunch.

A summary of the differences between a usual equivalent electrical model and the multibunch model can be found in Tab. 1 where I refers to the cell current, as opposed to the local current.

Table 1: Differences between a homogeneous equivalent electrical model and the multibunch model

Parameter	Homogeneous	Multibunch by n
OCV	$U(Q_{cell})$	$U(Q_i), i = [1..n]$
R_{HF}	$R_e(SOC, I, T) + R_{ct}(SOC, I, T)$	$R_{HF,i}(I, T)$ and distributed
R_d	$R_d(SOC, I, T)$	$R_d(SOC\ 50\%, I, T) \cdot n$
C_d	$C_d(SOC, I, T)$	$C_d(SOC\ 50\%, I, T) / n$

Regarding the number of bunches, the higher they are, the more accurate the model is. Several number have been tested, and the optimal number found for a single cell seems to be the higher one. Because the computation time is roughly a first order function of the number of bunch, the chosen number here is a compromise between accuracy and computation time. The impact of n on accuracy and computation time can be found in Fig. 2. Other way of searching for the distribution gives similar results. For the following, we choose $n=20$.

3 Cell used and experiments

3.1 Cell characteristic and experimental setup

The cell used for this paper is a 40Ah LiFePO_4 - graphite cell. It is already aged and its actual capacity is around 37.9 Ah. The discharge current limit is 2C, the charge current

limit is 1C, for a voltage varying between 2,5 and 3,7V. Regarding the temperature, the operating range is -15 to 50°C.

As the measured temperature will be used to compute the parameters values of all bunches, the cell needs to be the most homogeneous possible. Thus we chose to insulate the cell with a polyurethane box, as pictured in Fig. 3. This way, we consider the temperature to be the same in the surface or in the hearth of the battery.

The cell was surrounded by at least 8 cm of polyurethane on each side, except on the top where we used glass wool to be able to connect the cell. The connecting cables are also insulated with polyurethane foam. The whole setup was then placed into a climatic chamber to access the initials temperatures and connected to a Bio-Logic system.

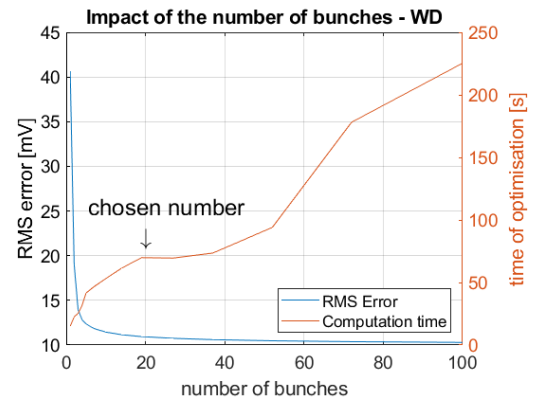


Fig. 2: Influence of the number of bunches to the precision and convergence time for Weibull Distribution.

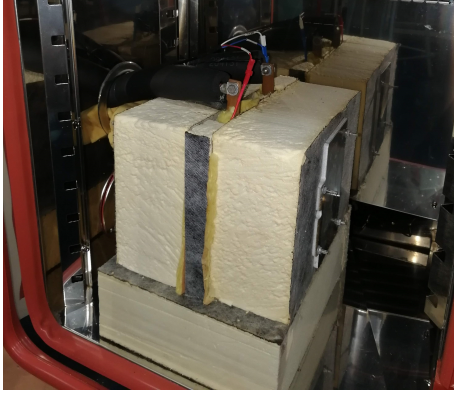


Fig. 3: Experimental setup with the cell and polyurethane box inside the stove. Cable are insulated with polyurethane foam.

3.2 Tests descriptions

The cell was discharged with an initial temperature of 10°C, 25°C and 38°C with a current of 1C and 2C for each temperature. The maximum temperature allowed by the constructor was hit during the 2C discharge with initial temperature of 38 °C, resulting in a half discharge only. The measured temperature was acquired using a thermocouple on the largest side of the cell, within polyurethane setup.

4 Simulation and optimization algorithms

4.1 Simulation of the cell

Once the dependency tables are established, the current and temperature curves acquired and the basic electrical parameters of the cell known, we only need to define the heterogeneity of our model. The flow chart of our coupled simulation-optimization algorithm is represented in Fig. 4.

The cell is modelled by an algorithm that uses as input the measured temperature and current during a discharge, the initial state of the cell and the distribution of the parameter R_{HF} . The simulation algorithm has three steps.

- First the electrical parameters (resistances, capacities, time constants) are updated with regard to current and temperature;
- Then, with regards to the previous local currents and the hypothesis that they remain constant during the time step dt , the local states of charge Q_i and voltages $U(Q_i)$ and the voltage across the RC element are computed;
- Finally the new local currents and cell voltage are computed by solving a linear system using cell current, the locals OCV $U(Q_i)$ and the voltages $U_{Rd,i}$.

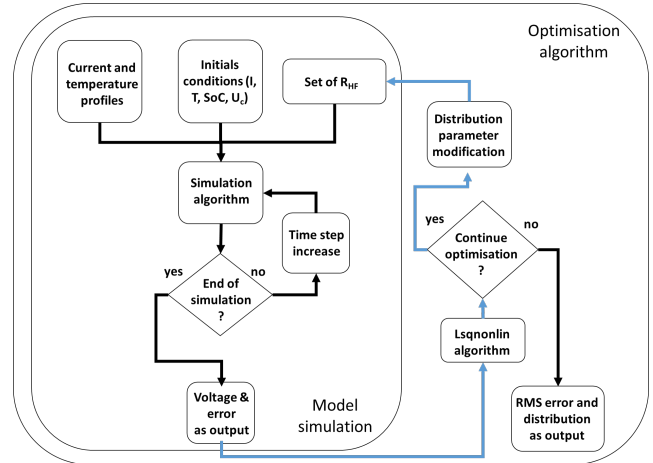


Fig. 4: flow chart of our algorithm

4.2 optimization algorithm and methods

In order to simplify the definition of the optimization function, we chose to use conductances instead of resistances. This is because the equivalent conductance of parallel bunches is simply the sum of the conductances. In the following of this article, we will still use the denomination R_{HF} to avoid excessive vocabulary.

This section is dedicated to the determination of the R_{HF} values, that are distributed among the bunches. To find the best set, we tried three methods:

- an indirect determination using a Weibull distribution (WD);
- a direct determination using a free distribution initialized by a Weibull one (FD-WI);
- a direct determination using a free distribution initialized by a constant one (FD).

Using a WD was already done in a previous article [4]. In this article we propose on the one hand an optimization of the three Weibull parameters of the indirect determination and on the other hand the possibility to directly find the R_{HF} values that were previously extracted from the WD. While the Weibull distribution is very adaptable mathematically speaking, it remains possible to miss an outsider in the resistances values. Using a free distribution allows us to be able to find those points, while increasing the number of parameters to the number of bunches $n=20$. In order to decrease the time needed, we imposed an ordered solution, meaning that an initial conductance G_1 is searched for and then the difference between G_n and G_{n+1} (equation 3)

$$\forall i, j \in [2, n], \quad G_i = G_1 + \sum_{j=2}^i \Delta G_j \quad (3)$$

subject to $\forall i \in [2, n], \quad 0 \leq \Delta G_{min} \leq \Delta G \leq \Delta G_{max}$

The objective function for a free distribution is then:

$$\min_{G_i} f_e(t, I, T, G_i) = [U_{sim}(t, I, T, G_i) - U_{meas}(t)] \quad (4)$$

where f_e is the error vector for a given discharge.

5 Results and discussions

5.1 Quality of the solution

The aim of the optimization operation is to find a suitable set of conductances to model the cell. However, we also want to obtain a distribution that is physically meaningful. We found that a free distribution (FD) may lead to a less physical solution. An example of this can be found in Fig. 5 where the free set gives a distribution with a few equal subsequent values and one greater value at the end. This is not in accordance with our hypothesis of a regular distribution of conductances. A non-regular evolution is possible but is considered less likely because it doesn't suit our hypothesis of a continuum of electrical properties inside the cell. A free distribution initialized by a Weibull (FD-WI) distribution brings a better regularity among the set of resistances, as expected from the model construction. This accordance to our hypothesis can hardly be differentiated by a computer, thus this way of searching for conductances is the one we selected for future works. Moreover, a free set of resistances is a good way to detect damaged cell with a different signature within a larger pack. It allows our algorithm to find a set that includes outsider values that would not fit in a usual distribution. Thus this way of determining a set of conductances seems to be the most suitable method.

5.2 Model Robustness

By optimizing the parameter of a Weibull distribution for a single discharge, we obtained RMS error between 9.7 and 14.5 mV for each discharge. Despite the good results, we could not find a link between the multiple sets of optimised parameters of the Weibull law that would suggest a unique distribution to model this cell.

Our model aims to model the heterogeneity of the cell. The most heterogeneous discharge should be a low temperature - high current one. Thus we make the hypothesis that optimizing our resistance set over this discharge increases the distribution overall robustness. To verify that point, we simulated the farthest discharge in term of conditions, namely the hottest discharge with a low current, with our low temperature - high current set. The results are presented in Fig. 6

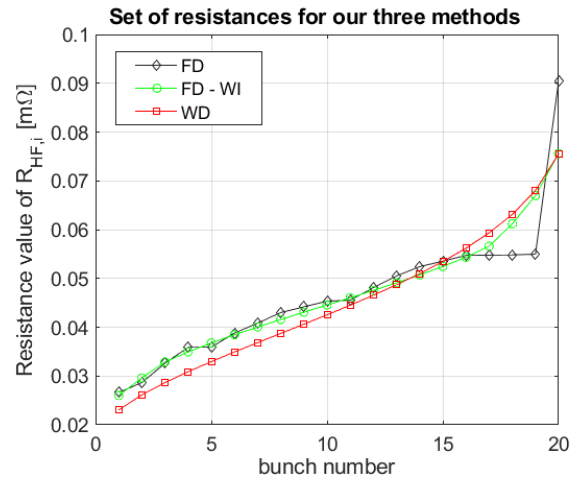


Fig. 5: Difference in resistance values without and with a Weibull initialization for the free set of resistances and for the Weibull set at 1C and 25°C

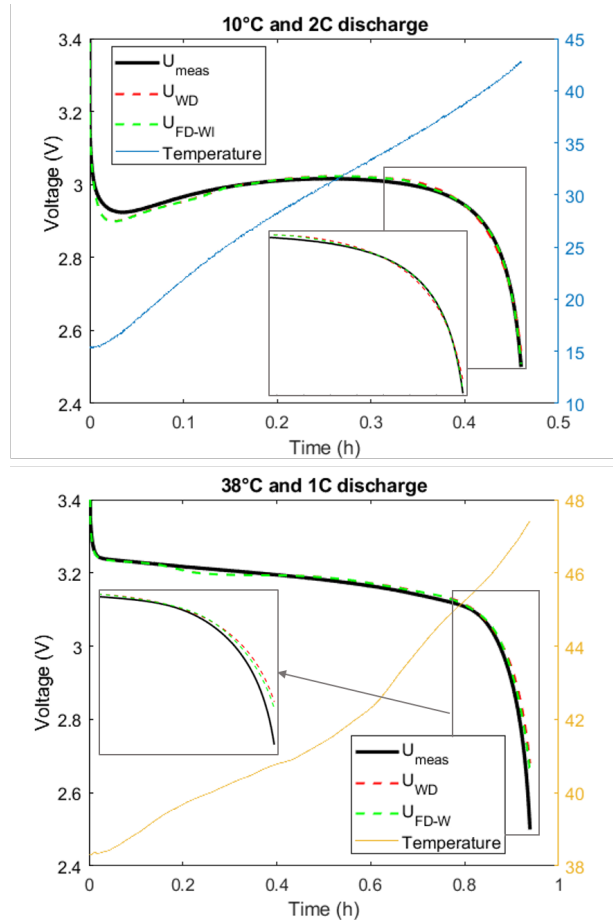


Fig. 6: a). Optimization over a low temperature discharge with RMS error of 14.46 mV (Weibull) and 13.21 mV (free) and b). validation over a high temperature one with RMS error of 23.65 mV (Weibull) and 19.40 mV (free)

We then tried to let the optimization function set the R_{HF} values freely, again for the low temperature - high current discharge. As for the WD, we present in Fig. 6 the simulation over the high temperature discharge and the low temperature one.

The simulation error over discharges is smaller when a FD-WI is used. Even if the difference is small, the possibility not to follow any law seems very interesting for us in order to better diagnose the degradation of the cell, as will be explained in section 6.

6 Conclusions and perspectives

In this paper we presented an electrical equivalent model to model the heterogeneity of a commercial $LiFePO_4$ - graphite cell. This model is based on a physical approach that takes into account various phenomena. The construction is based on the hypotheses that one cell can be divided into parts that have the same electrical properties, that we group into several bunches. They differ from another thanks to a set of "high frequency" resistances that regroup the charge transfer, the current collector, the electrolyte and the contact resistances.

The proposed model has been experimentally validated, and we showed that we were able to define a set of resistances that can model the cell in another operating point. Our hypothesis that a measured discharge at high current and low temperature allows us to simulate at best other operating condition was verified in our simulation and comparison to measurements. By finding a Weibull distribution and then exploring values around it, we are able to reduce the computation time and to find a more physical solution

Regarding the set of R_{HF} values, we expect to follow it through the cell life in order to obtain a non-invasive tool to characterize the degradation of the cell. If a part of the cell experience severe degradation, the shape of the set of conductances will be modified and we expect the new shape not to follow a natural distribution law. This would allow to smartly define periods of maintenance for the cells or change the cells usage to prevent them from severe degradations. This justify the choice of a free distribution, initialized by a Weibull one, that may detect abnormal variations of the resistance set.

We plan to apply this simulation method to other technologies of cells to further verify that we are able to simulate them over the whole operating range. In order to further increase the precision of our model, a more developed bunch model could be considered if the computational time remain acceptable.

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