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Simplified Modeling and Characterization of the Internal Impedance of Lithium-Ion Batteries for Automotive Applications

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Abstract

This paper presents a fast and simple approach to model and characterize internal impedance of Li-ion battery. This is an important aspect because, measuring the impedance in a wide frequency range, it is possible to investigate on the modifications of the internal electrochemical cell process and evaluating its State of Health (SOH), that it is an important parameter for the automotive applications. The first part of the paper explains the concept of Electrochemical Impedance Spectroscopy (EIS) and how Li-ion batteries can be represented through electrochemical or empirical models. EIS test consists of the excitation of the battery, or cell, with a sinusoidal current signal, and then measuring the cell voltage, while the frequency response of the system is computed. Then the paper describes a procedure to test the cell and extract the parameters necessary for the model, in particular, the dependence between the parameters of the model and the State of Charge (SOC). Finally the paper shows how the internal impedance of a Li-ion battery is a dynamic parameter that depends on different factors and illustrate how the EIS can be used to obtain an impedance model**.**

Keywords *— Lithium-ion Battery, Battery model, EIS, Internal Impedance, Electric vehicles.*

I. INTRODUCTION

In response to an increasing demand in portable electronics, management and control of energy accumulators (rechargeable batteries) is one of the main themes of research and industrial development for the automotive applications. The most important energy storages are Lithium-ion (Li-ion) batteries, in fact they have high efficiency, high energy density, low self-discharge and relatively long cycle life respect to other technologies. In order to reduce the polluting emissions and the fuel consumption, almost all the automotive sector is interested on the development of this technology. The carmakers are developing and deploying vehicles with an increasing level of electric hybridization, from Hybrid Electric Vehicles (HEVs) [1], to Plug-in Hybrid Electric Vehicles (PHEV) [2] and eventually to pure Electric Vehicles (EVs). High energy and power density, high level of safety and reliability, high life cycle number (which translates in long vehicle mileage) and lowest possible cost are the purpose of research in this area. Thanks to this phenomenon, the mass production of batteries and the study on them is increasing. Lithium-ion batteries have many fields of use and a specific chemistry is more suitable for an application than another depending on their characteristics. An overview and future prospect regarding different Li-ion chemistries, along with their most suitable application, is shown in [3], [4]. Today, Li-Ion batteries have a high cost and are more sensitive to working condition such as their usable capacity [5], temperature [6], power demand and its lifetime due to aging [7].

For these reasons an important purpose is to describe how the battery discharges or degrades during the lifecycle. These effects are associated with changes in the internal impedance of the battery. This impedance appears explicitly in empirical circuit models and it can be identified at different moments of the life cycle, making possible to quantify the changes of the different model parameters. The internal impedance depends on many parameters, the most significant are the State of Charge (SOC), State of Health (SOH) and the temperature.

SOC is the percentage of residual capacity respect to battery nominal capacity and SOH is the percentage of degradation of the battery. The dependence of battery impedance on the SOC and SOH has encouraged many researchers to investigate on this parameter. Due to complex electrochemical structure, battery impedance can take some different effects depending on its different dynamics, as visible in Fig.1. Analyzing the particular shape of this curve, in the appropriate frequency range, SOC and SOH can be evaluated respectively in low frequency [8] and in high frequency [9]. In literature, there are also other methods to evaluate SOC

and SOH [10], [11].

Aim of this paper is create a model that describes the behavior of the internal impedance of a lithium-ion battery and how its parameters depend on the state of charge. It is also possible take into account how the Electrochemical Impedance Spectroscopy (EIS) can be used to analyze the evolution of the different parameters that compose an internal impedance model with an accelerated degradation experiment.

This paper is organized in seven section. Section II provides a literature review to explain the concept of electrochemical impedance spectroscopy. In section III, an impedance model is proposed and its choice is justified. Section IV describes the chosen battery cell and the experimental setup utilized for the characterization tests on the cell itself, which are presented in section V. Section VI illustrates the model parameter extraction procedure and finally, in the section VII shows the conclusions.

II. ELECTROCHEMICAL IMPEDANCE SPECTROSCOPY

Electrochemical Impedance Spectroscopy (EIS) analyzes the chemical-physical property of electrochemical energy accumulator, in this work Li-Ion batteries. It allows to extract the information necessary to characterize the aging effects [12]. In order to show the results, it is used a Nyquist plot with a negative Y-axis. This method is a good technique capable of identifying the state of the degradation of the cell and it highlights some aging effects that traditional tests do not recognize [13]. This technique is fast, non-destructive and it allows to obtain a typical Nyquist plot shown in Fig.1. The Nyquist plot can be divided in three parts. Low frequencies are related to capacitive effects, in particular, the first semicircle corresponds to the relaxation of charge carriers at the solid-electrolyte interface and, where the graph may show a spike, the semicircle end describes the lithium cation diffusion in the solid-state phase. The other semicircle is dependent to the electrode potential and it can be modeled by a double-layer capacitance and the charge-transfer resistance. On the right side of the plot, the diagonal line with a positive slope is represented by the Warburg impedance. This impedance is associated to the result of the chemical reaction of the solid-state diffusion of the Li+ in the bulk electrode material [14]. The high frequency behavior depends by the geometry of the cell and porosity of the electrode plates. It can be modeled by inductive element and resistor, in particular, it corresponds to the total value of the ohmic resistances. The Nyquist plot can be approximated through different electronic components according to the frequency ranges on which these elements have a major influence, as shown in Fig.2. For instance, high frequencies can be represented through an inductance. Then, the semicircle present on the plot is modeled by a RC-parallel branch. Finally, the Warburg impedance affects the response at low frequencies. The parameters variation respect to cycles life makes more evident the degradation process. This way it is possible to establish a correlation between parameter changes and the SOH of the battery.

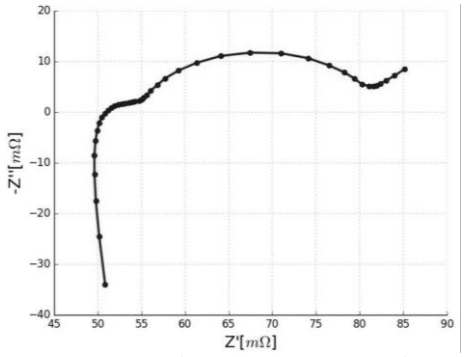

Fig. 1. Typical Nyquist plot of the internal impedance of a Li-ion battery

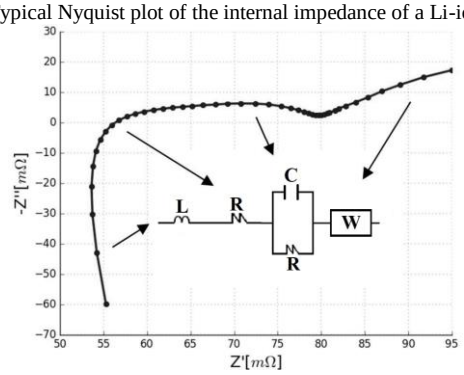

Fig. 2 Reference spectrum of Li-Ion cell in a wide frequency range and battery EIS modeling

III. BATTERY IMPEDANCE MODEL

In order to analyze battery internal processes with different time constants, impedance in a wide frequency range is measured. [15]. These methods are chiefly used to identify the model that describes the short-term behavior of the battery. In general, a sinusoidal signal in current i is applied to the battery and it is measured its voltage response v. The battery is considered as a Linear and Time-Varying (LTI) dynamic system so the impedance Z is computed by the equation:

$$Z = \frac{v(t)}{i(t)} = \frac{Vsin(\omega t+\varphi)}{Isin(\omega t)} \qquad (1)$$

In Eq.1, V is the voltage, I is the current amplitude, ω is the angular frequency, φ is the phase shift.
In the frequency domain, is possible to separate the impedance in a real part $Re(Z) = Re(Z(j\omega))$ and in an imaginary part $Im(Z) = Im(Z(j\omega))$ using the equation:

$$Z(j\omega) = Re(Z) + jIm(Z) = |Z|cos(\varphi) + j|Z|sin(\varphi) \quad (1)$$

Where: $\varphi = \varphi(j\omega)$, $j = \sqrt{-1}$.

$Z = Z(j\omega)$ is the absolute value of the impedance, expressed by:

$$|Z| = |Z(j\omega)| = \sqrt{Re(Z)^2 + Im(Z)^2} \qquad (2)$$

The mathematical battery model should be defined from the real and imaginary values of impedance displayed on the Nyquist plot, as shown in Fig.2. This plot can be divided in four sections, which describe internal processes at different time constants. There are many models to describe the behavior of a battery [16] but it is needed to use a particular passive elements of an Equivalent Electrical Circuit (EEC) to simulate different four sections (as shown in Fig.2). The inductance (L) describe the inductive behavior due to the metal elements of the cell and cables. The resistance (R_{ohm}) is the sum of the resistance of current collectors, electrodes, electrolyte and separators [9]. The Warburg Impedance **(W)** considers the diffusion of Li-Ion in the porous active material of the electrodes The RC group is used to model the semicircle present in Fig. 2. This semicircle is dependent to the electrode potential and is modeled by a charge-transfer resistance (R_1) and a double-layer capacitance (CPE_1). This parameter is represented by a Constant Phase Element (CPE), a non-ideal capacity represented by an irrational transfer function [17]. In the case of cell EIS, it is needed to simulate the same Nyquist plot using not simple capacity, but the Constant Phase Elements (CPEs). Indeed, the impedance curve is not represented by a circle arcs, which should describe an ideal ohmic-capacitive behavior, but it is really represented by an ellipse arcs. More details about CPE and Warburg elements are shown in [9] and in [17].

The impedance of a CPE can be expressed in fractional calculus with the equation:

$$Z_{CPE}(s) = \frac{1}{fs^a} \qquad (4)$$

Where: Z_{CPE} is the impedance of the CPE; s is the Laplace operator; f is the fractional coefficient; and a is the fractional order, $a \in \Re$, $0 \leq a \leq 1$. It should be noted that the CPE represents a resistance when a = 0 and represents a capacitance when a = 1.

In the purpose model the Warburg Impedance are neglected because the behavior of the battery at low frequency is not investigated. The final simplified model is shown in Fig 3. The SOC dependency of the model is implemented in look-up table form, thus a running SOC calculator block is needed inside the model. An algorithm integrates the instantaneous battery load current and computes the SOC through its definition:

$$SOC(t) = SOC_0 - 100\frac{\int_0^t I(t)dt}{Q\ 3600} \qquad (5)$$

where: SOC_0 is the initial state of charge, $I(t)$ the instantaneous load current in A and Q is the battery cell capacity in Ah.

The SOC information thus exits the calculator block and is fed into the model: the whole model is easily implemented in MATLAB/Simulink environment.

TABLE I. BATTERY DATASHEET PARAMETERS

Property	Value
Nominal capacity	2.5 Ah
Nominal voltage	3.3 V
Max. voltage	3.6 V
Min. voltage	2.5 V
Max. discharge current	70 A (21-C)
Max. charge current	10 A (4-C)
Operating temperature	-30 °C to 55°C
Cycle life	> 1000 cycles

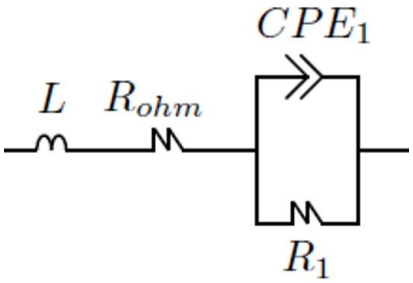

Fig. 3 Internal Impedance model

IV. Experimental Setup

The characterization tests are performed on a specific $LiFePO_4$ cell from A123, the ANR26650M1-B cylindrical model: its main parameters are reported in Table 1.

The experimental setup is shown in Fig. 4 and is made up by a MaterialMates Instruments MM540 potentiostat, connected to the battery cells under test through power wires and directly communicating with a computer through USB interface. The tester has one channels available, capable of withstanding 12 V and carrying 30 A either in charge than in discharge. The MaterialMates Instruments MM540 can be programmed through a software interface to charge/discharge the connected battery cells with user-defined dynamical current or voltage profiles. The instrument measures current and voltage and calculates the real and imaginary values of impedance.

For the EIS test, the instrument has a dedicated Frequency Response Analyzer (FRA) that produces the excitation signals.

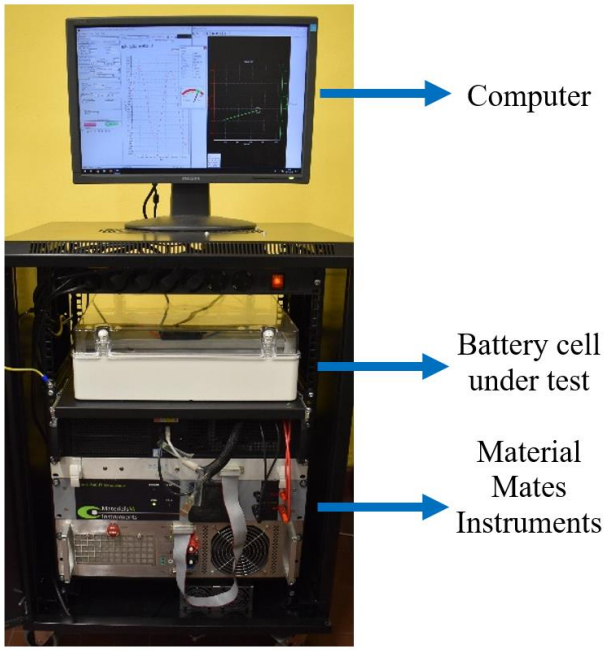

Fig. 4 Laboratory EIS test setup

V. Characterization Procedure

The battery cells are characterized by a set of subsequent tests, which aims to collect the necessary data to parametrize the presented impedance model. The nominal current (1C) of the cell is used as a reference current.

A. *Preconditioning test*

The preconditioning test consists of three charge/discharge cycles at 0.5C (i.e. 1.25 A) carried out at ambient temperature. A 90-minute rest is undertaken after either every charge or discharge. The test ends after roughly 24 hours.

B. *Capacity test*

The capacity of the cell is necessary to define the state of charge variable (Q), as shown in (5). A nominal capacity value is

always given by the manufacturer in the battery cell datasheet (i.e. TABLE I), but, since it represents a underestimation, a test should be performed to evaluate a more realistic value.

Battery capacity normally depends on the discharge current at which the test is performed, following Peukert's law:

$$Q_{1C} = I^k t \qquad (6)$$

Where: Q_{1C} is the capacity of the cell measured during a 1C discharge, I is the discharge current, t is the discharge time required and k is the Peukert's coefficient (≥ 1). The total charge that can be obtained from the cell is not significantly dependent on the current value because for the lithium-ion batteries, Peukert's coefficient is very near to 1 [18].

This test is performed by discharging the battery with a constant current equal to C/5. A low current is chosen to avoid that the lower discharging threshold voltage to be reached prematurely. In this paper a unique value of capacitance extracted at C/5 has been considered, until the lower voltage threshold is reached (i.e. 2,5 V). A capacity value is calculated approximately 2650 mAh, as shown in Fig. 5.

C. EIS Test

For the EIS test, first the cell is carried to the desired state of charge then a sinusoidal signal in current is applied. The offset of the signal is equal to 0,5 A in discharge, the maximum peak is 1A and the frequency range is from 5 Hz to 1000 Hz. The duration time is chosen in order to have a SOC variation very little, in particular the discharge cell is less than 1%. The test is repeated every 10% of SOC.

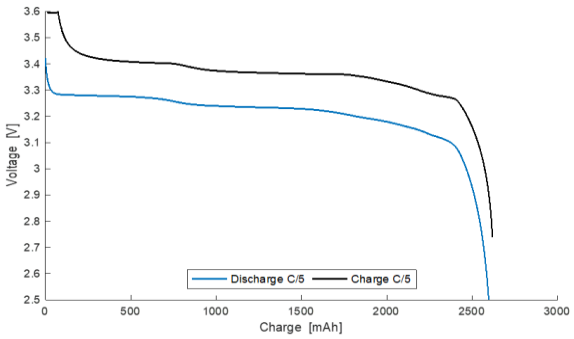

Fig. 5 Battery capacity test

VI. PARAMETER EXTRACTION

The model parameter extraction process is based on the Nyquist plot. First of all, the value of R_{ohm} is approximated with the intersection between impedance curve and real axis. The experimental values are reported in Fig.6.

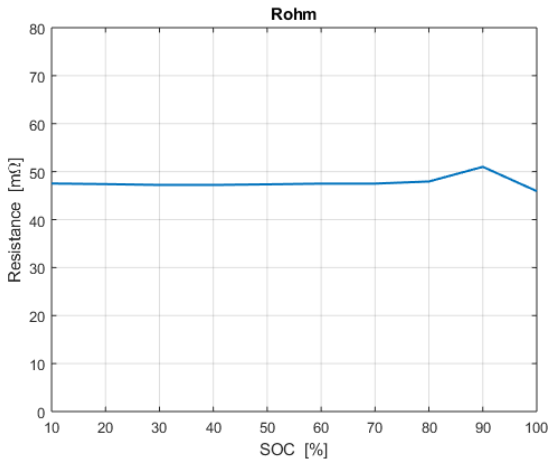

Fig. 6 Extracted impedance model parameter, Ohmic Resistance.

Subsequently, the other three parameters are calculated by the simplex algorithm [19]. The input of the algorithm are the experimental impedance measurements (frequency, the impedance real part and imaginary part), and a circuit model. The algorithm

allows to calculate the parameters of the circuit model that provide the impedance function experimental, using the fminsearch algorithm bounded (fminsearchbnd) of Optimization Toolbox of MATLAB. The fminsearch algorithm allows to find a local minimum of an unconstrained non-linear problem, fminsearchbnd instead has the advantage of solving problems of the same type with constraints. An example of best fit as shown in Fig.7.

Fig. 8 shows the experimental values of the inductance while the parameters of the RC group are reported in Fig.9 and in Fig.10. In particular, Fig.9 shows the values of R1 and in Fig.10 are shown respectively, the fractional coefficient and the fractional order of constant phase element.

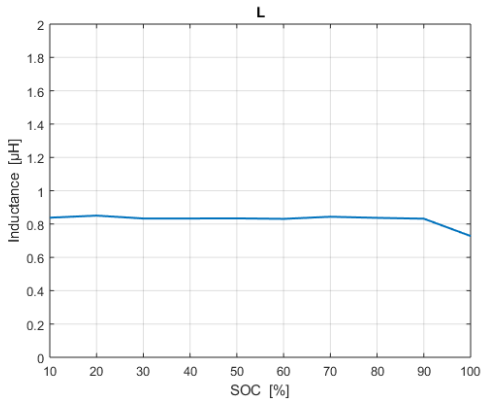

Fig. 7 Measured and fitted waveforms.

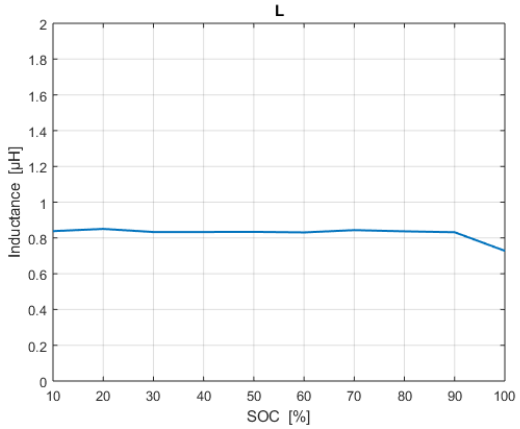

Fig. 8 Extracted impedance model parameter, Inductance.

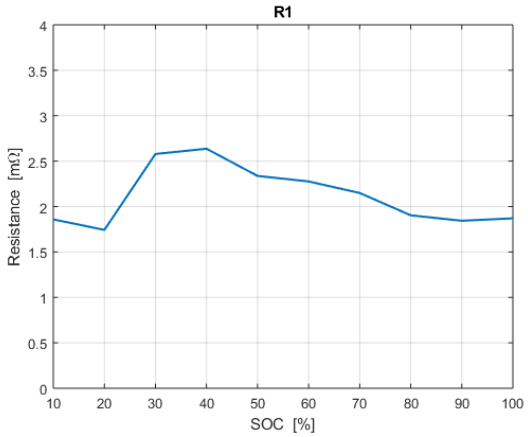

Fig. 9 Extracted impedance model parameter, Resistance.

VII. Conclusions

A battery impedance modeling approach, mostly suited for automotive applications, has been presented in this paper.

The model is composed by four parameters. The ohmic resistance and the inductance remain almost constant for all the tests, consequently it can be stated that there is not dependence on the state of charge for these parameters. Instead, the other parameters depend on the soc. In particular, both the resistance and the fractional coefficient increase when the state of charge decreases but have the minimum value than the SOC is 20%. This value corresponds approximately to the change of concavity of the polarization curve of the battery, as shown in Fig.5. It could be a direct correlation between this aspect and the impedance, it will be verified in the future.

The frictional order is congruent with the theoretical value in fact, it is always between 0 and 1. The Constant Phase Element is more similar at a capacitance when the cell is complete charge and this aspect is confirmed by the value closest to 1 that it is 100% of the SoC

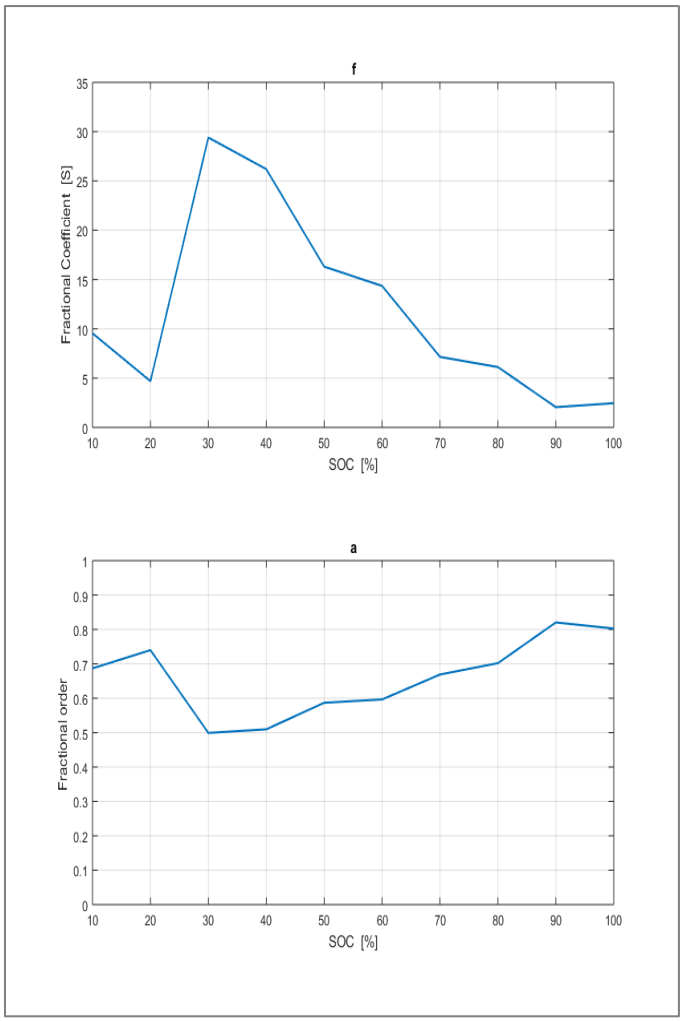

Fig. 10 Extracted impedance model parameter, Constant Phase Element. (a) is the frictional coefficient and (b) is the frictional order.

The next step of the authors will be used the Electrochemical Impedance Spectroscopy to analyze the evolution of the different parameters that compose this model with an accelerated degradation experiment.

Since no tests have been done to evaluate the battery impedance dependency on temperature and current rate, these aspects will be also object of future work from the authors, in order to extend the model validity to a broader set of operating conditions.

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